

Theoretical Study of Electron Dynamics in Multi-Orbital Antiferromagnetic Metals

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Abstract

Chromium (Cr) is known to be a typical antiferromagnetic metal characterized by an incommensurate spin-density-wave (SDW) ground state due to the Fermi-surface nesting. The magnetic and electronic properties of Cr have widely been investigated for a long time. Such studies of $3d$ transition-metal compounds have been renewed by a recent discovery of iron-based superconductors, and newly focused on the aspect of the multi-orbital nature of $3d$ atoms. In the field of iron-based superconductors, it is now recognized that the orbital degrees of freedom play a crucial role not only in the mechanism of superconductivity but also in the understanding of antiferromagnetic metallic phase adjacent to the superconducting phase. Therefore, it is important to include all of $3d$ orbitals in theoretical works of transition-metal compounds. Physical properties of the typical transition-metal compound, chromium, must be accordingly reinvestigated from this viewpoint. In this thesis, we theoretically study the electron dynamics in multi-orbital antiferromagnetic metals, especially focusing on iron arsenide and Cr. Our aim of studying both materials is to deepen our understanding of the physics of antiferromagnetic transition-metal compounds characterized by multi-orbital nature.

The parent compound of iron-arsenide superconductors exhibits an antiferromagnetism with stripe-type order below Néel temperature. The band structure of the stripe phase is characterized by Dirac-type dispersions, which are induced by the folded paramagnetic dispersion with multi-orbital nature. In the stripe phase, the resistivity shows anisotropy in the conducting Fe-As plane. The experimental data suggest that impurities play a crucial role in the anisotropy. We start with a five- $3d$ -orbital Hubbard model, and use a mean-field approximation. Then, we examine the anisotropy of resistivity by applying the memory-function approach treating isotropic nonmagnetic impurities. The resistivity obtained by the memory-function approach yields a proper anisotropic behavior near the undoped region: The resistivity in the antiferromagnetically ordered direction is first smaller than that in the ferromagnetically ordered direction, and the anisotropy reverses as holes are introduced. The origin of the anisotropy can be understood from the interplay of impurity scattering and the character of Fermi surfaces including the Dirac-type one.

Motivated by the crucial role of orbitals in the iron-arsenide antiferromagnetic metallic phase, we reinvestigate electron dynamics of the typical itinerant SDW metal, Cr. We again use multi-band Hubbard model composed of $3d$ and $4s$ orbitals. After the SDW mean-field approximation, we obtain the dynamical spin susceptibilities and L_3 -edge resonant inelastic x-ray scattering (RIXS) spectra by employing random phase approximation. In our calculation, we assume the perfect commensurate SDW state. We find a collective spin-wave excitation undamped up to ~ 0.6 eV. Above the energy, excitations overlap individual particle-hole excitations as expected. In RIXS spectra, particle-hole excitations with various orbital channels show a large spectral weight, masking the spectra of the spin collective mode. However, it may be possible to detect the spin-wave excitation in RIXS experiments in the future if resolution is high enough.

The above two topics in this thesis provide the evidence of crucial roles of $3d$ orbitals in the antiferromagnetic metal of $3d$ transition-metal compounds. This contributes to the deeper understanding of the physics of multi-orbital antiferromagnetic metal.

Publication List

This thesis is based on the following original papers.

Published papers

1. “Spin dynamics and resonant inelastic x-ray scattering in chromium with commensurate spin-density wave order”: Chapter 4
K. Sugimoto, Z. Li, E. Kaneshita, K. Tsutsui, and T. Tohyama, Phys. Rev. B **87**, 134418 (2013).
2. “Memory function approach to in-plane anisotropic resistivity in the antiferromagnetic phase of iron arsenide superconductors”: Chapter 3
K. Sugimoto, P. Prelovšek, E. Kaneshita, and T. Tohyama, Phys. Rev. B **90**, 125157 (2014).

Related paper

3. “Optical Conductivity of Antiferromagnetic Metallic Chromium: Mean-field Calculation for the Multi-orbital Hubbard Model”
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Chapter 1

INTRODUCTION

After the discovery of iron-based superconductors and succeeding intensive study of related materials, it is recognized that the interplay of spin and orbital degrees of freedom is crucial for understanding the physical properties of transition metal compounds. These degrees of freedom are expected to relate to the pairing mechanism in superconducting phase of iron-based superconductors. The itinerant antiferromagnetic phase is adjacent to the superconducting phase of iron arsenide. Since the antiferromagnetism comes from the nesting of Fermi surface involving multiple $3d$ orbitals, the investigation of the antiferromagnetic phase is suitable for understanding a role of spins and orbitals. In addition, to deepen our knowledge, we also focus on a typical material with itinerant spin-density wave (SDW) state, chromium (Cr).

In this chapter, we first introduce itinerant SDW in Cr in Sec. 1.1. Iron-based superconductors are introduced in Sec. 1.2. The anisotropy of resistivity in iron arsenide is introduced in Sec. 1.3. In Sec. 1.4, we introduce the collective mode in Cr, and show the possibility of its observation by resonant inelastic x-ray scattering. Finally, we give the purpose of this thesis in Sec. 1.5.

1.1 Spin-Density Wave in Chromium

Antiferromagnetism in Cr was first observed by Shull and Wilkinson with use of neutron diffraction [1]. After several years, Corliss *et al.* noticed that the antiferromagnetism in Cr is not simple [2]: While the spin is aligned alternately up and down in short-range scheme, the spin moment is modulated in long-range scheme. The modulation wave of spin moment is referred to as SDW. In 1962, Overhauser explained that a good nesting of Fermi surfaces, i.e., a coincidence of shapes of an electron-like Fermi surface (an electron pocket) and a hole-like Fermi surface (a hole pocket), leads to the SDW order [3]. The wave vector of the SDW is determined from a vector directing from an electron pocket to a hole pocket in wave-vector space, which is referred to as a nesting vector. In this theory, energy bands are folded with regard to the nesting vector, and energy-band gaps (referred to as SDW gaps) open at crossing points of the folded bands. The theoretical treatment of opening gaps and stabilization of the SDW state is very similar to those in a superconducting state [4].

The nesting condition is written as

$$\varepsilon_e(\mathbf{k}) - \varepsilon_F = -(\varepsilon_h(\mathbf{k} + \mathbf{Q}) - \varepsilon_F), \quad (1.1)$$

where ε_F is the Fermi energy, $\varepsilon_e(\mathbf{k})$ and $\varepsilon_h(\mathbf{k})$ are the energy dispersions of the electron pocket and the hole pocket, respectively, and $\mathbf{k}$ is the wave vector near the Fermi-wave vectors. $\mathbf{Q}$ is a nesting vector, which is almost the same as an SDW vector. A difference between the wave length of SDW and the periodicity of the lattice leads to

an incommensurate SDW.

In the system that has only well nested electron and hole pockets, the stabilization of the SDW state leads to a full gap in its band structure, implying that the system becomes insulator [5]. However, if there are other Fermi surfaces not contributing to the nesting condition, the system is still metallic after the SDW transition. Cr belongs to such itinerant SDW materials, and its physical properties have been well established theoretically and experimentally [6, 7, 8]. There are also many other materials with itinerant SDW state [9]. Iron-based superconductors discussed below also have the itinerant SDW state.

The crystal structure of Cr is a body-centered cubic (BCC) lattice, and the spin configuration is antiferromagnetic, where the spin at the center of the BCC lattice directs opposite to the spins at the corners of the lattice. There are many first-principles band-structure calculations of paramagnetic Cr, elucidating the nesting of its Fermi surfaces leading to the SDW order [10, 11, 12, 13, 14]. Figure 1.1(a) shows the Fermi surfaces of Cr in the paramagnetic phase. They are composed of an octahedral electron pocket at the Γ point [the wave vector $\mathbf{k} = (0, 0, 0)$] and an octahedral hole pocket at the H point [$\mathbf{k} = (2\pi/a, 0, 0)$], which are almost the same shape. However, a slight difference of the size of these octahedrons leads to the stabilization of the incommensurate SDW state. The nesting vector is $\mathbf{Q} = \frac{2\pi}{a}(1 - \delta, 0, 0)$, where a is a lattice constant of BCC structure and $\delta = 0.048$ - 0.05 [15, 16, 17]. Note that we can adjust the Fermi energy to make $\mathbf{Q}$ in Eq. (1.1) commensurate ($\delta = 0$) by doping aliovalent atoms. When the Mn (Fe) concentration x in $\text{Cr}_{1-x}\text{Mn}_x$ ($\text{Cr}_{1-x}\text{Fe}_x$) exceeds 0.003 (0.02), a commensurate SDW state is stabilized [7].

It is well known that a longitudinal SDW (L-SDW) phase appears below 122 K and a transvers SDW (T-SDW) phase does between 122 K and 311 K [6]. Figures 1.1(b) and (c) schematically show the T-SDW and L-SDW, respectively.

1.2 Iron-Based Superconductors

1.2.1 Overview

After the discovery of iron-based superconductors $\text{LaFePO}_{1-x}\text{F}_x$ in 2006 [18] and $\text{LaFeAsO}_{1-x}\text{F}_x$ in 2008 [19], many researchers in condensed matter physics began to study them and their related materials. This is not only because the compounds with iron, conventionally considered to be unsuitable for a component of a superconductor, show superconductivity, but also because they have prominently high superconducting transition temperature $T_c = 26$ K. The details have been described by review articles [20, 21, 22, 23, 24] or books [25, 26, 27]. Review articles focusing on superconductivity [28], magnetism [29], and optical conductivity [30] are also available.

One of the most noticeable features of iron-based superconductors is that they are quasi-two dimensional system. The primitive unit cell of LaFeAsO is shown in Fig. 1.2. It has iron-pnictogen (pnictogen is a chemical element of group 15 in the periodic table, e.g., As, P, etc.) layers, and Fe atoms form square lattices in the layers. Other ions out of these quasi-two dimensional structures scarcely contribute to the transport of electrons. The low dimensionality enhances electron correlation, which is considered to be the origin of high-temperature superconducting transition.

Several other classes have been found in the surveys of iron-based superconductors. Rotter *et al.* discovered $T_c = 38$ K in $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ [31] soon after the discovery of iron-based superconductors. $A\text{Fe}_2\text{As}_2$, where A is alkaline earth metals ($A = \text{Ba}, \text{Sr}, \text{Ca}$), is referred to as 122-type compounds, while LaFeAsO is referred to as 1111-

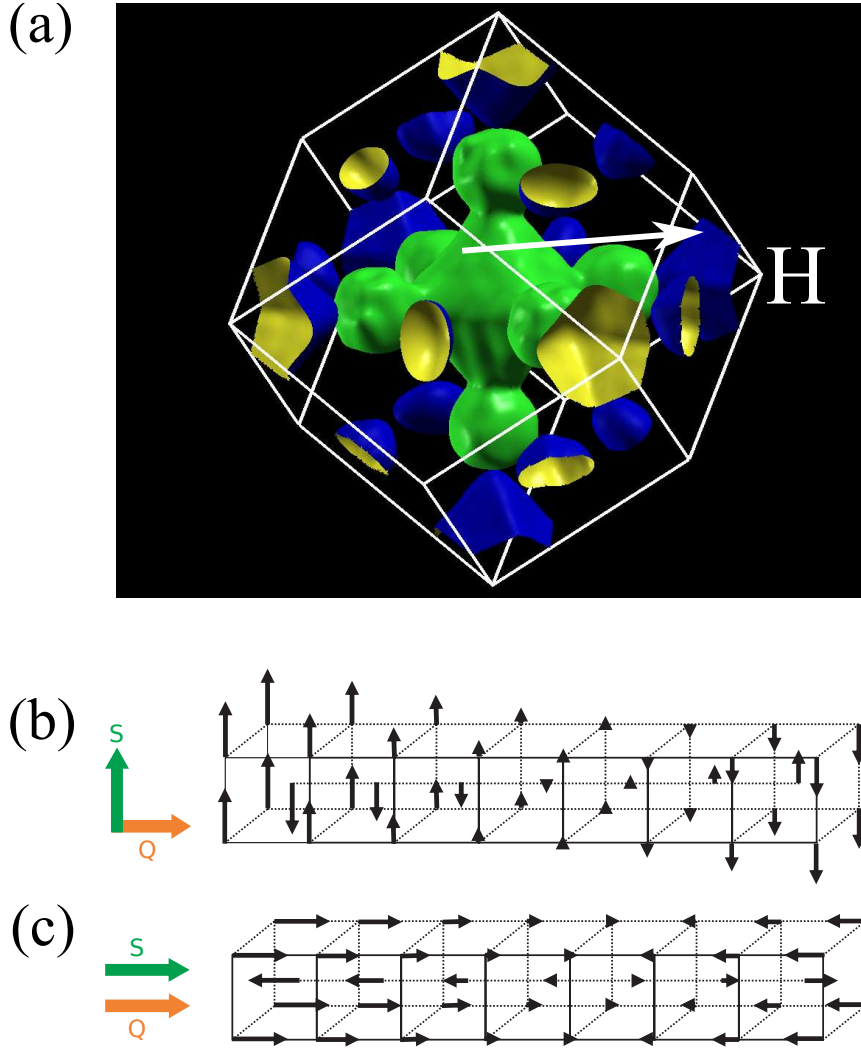


Fig. 1.1 (a) Fermi surfaces of Cr in the first Brillouin zone. Green surfaces are hole pocket, while blue ones are electron pockets. The quite well nesting of hole pockets at Γ point and electron pockets at H points causes the SDW in Cr. An white arrow is a nesting vector. (b) A schematic diagram of T-SDW, where the spin polarization $\mathbf{S}$ is perpendicular to the SDW vector $\mathbf{Q}$. (c) A schematic diagram of L-SDW, where the spin polarization is parallel to the SDW vector.

type compounds. Since large single-crystal samples (several milli-meters order) are available in the 122-type compounds, the systems are the comparably well investigated experimentally and theoretically among iron-based superconductors. In this thesis, we investigate the anisotropy of resistivity in BaFe_2As_2 and carrier-doped one.

Another noticeable feature of iron-based superconductors is that their parent compounds have an itinerant antiferromagnetic phase. The Néel temperature is suppressed and the superconducting phase appears with doping isovalent or aliovalent ions. Figure 1.3 shows the typical phase diagram of iron-based superconductors. The relation of magnetism and superconductivity implies that the high-temperature superconductivity originates from the magnetic fluctuations.

The iron-based superconductors have a structural phase transition slightly above the Néel temperature, where the structure changes from the tetragonal one to the

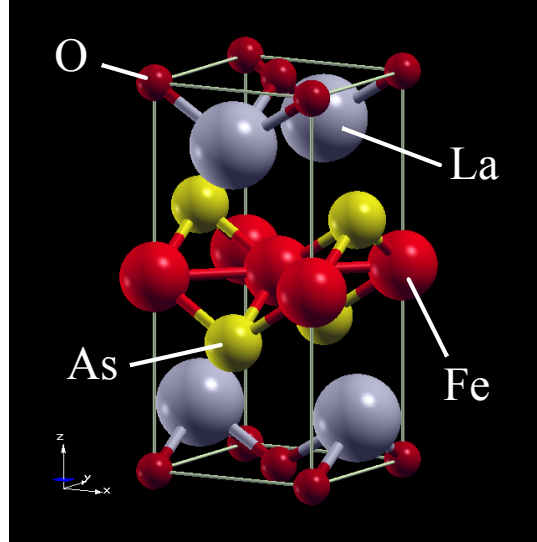


Fig. 1.2 The primitive unit cell of LaFeAsO , which is composed of FeAs layer and intercalation ions, La and O .

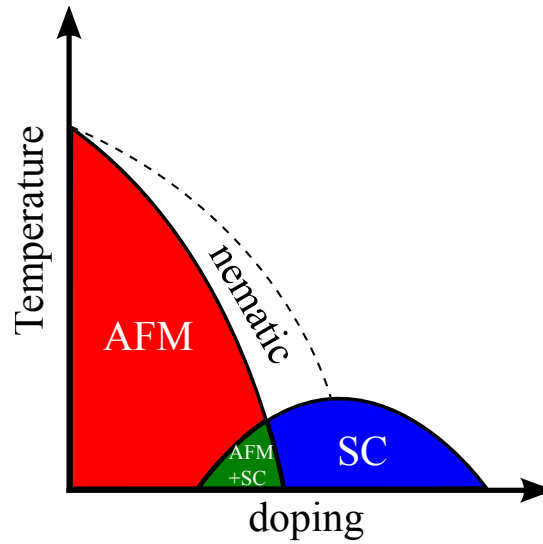


Fig. 1.3 The typical phase diagram of iron-based superconductors. 122-type compounds have the coexistence phase of antiferromagnetism (AFM) and superconductivity (SC), while 1111-type compounds do not. The nematic phase emerges through the change from four-fold symmetry to two-fold symmetry by structural phase transition.

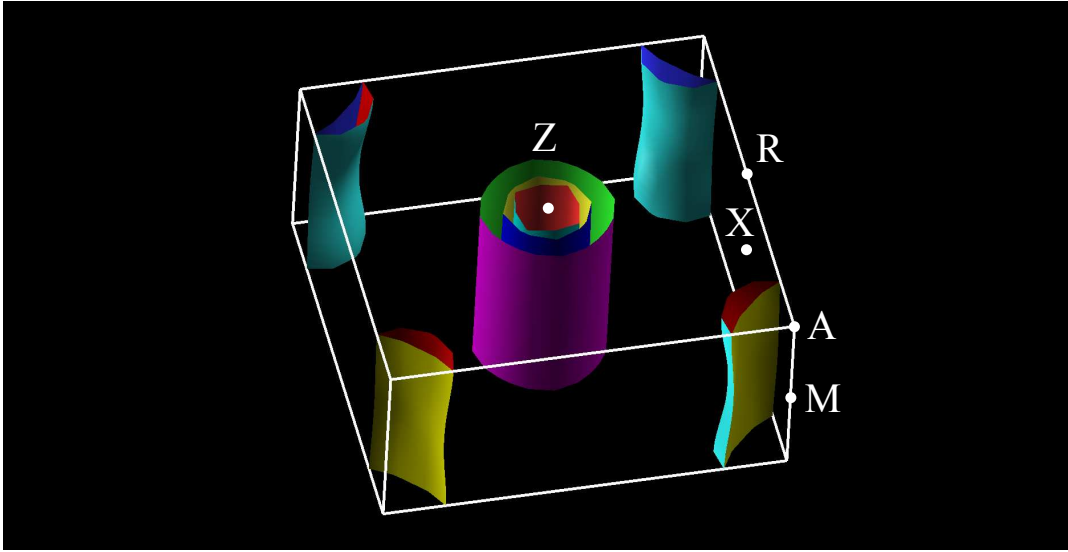


Fig. 1.4 Fermi surface of LaFeAsO in the first Brillouin zone. Fermi surfaces at the center of the Brillouin zone are hole pockets, while ones at the corners are electron pockets.

orthorhombic one with lowering temperature. This means that the Fe squared lattices are distorted to rectangle lattices. The phase below the structural transition temperature is referred to as a nematic phase.

1.2.2 Fermi surface and antiferromagnetism

Reflecting the small crystal-field energy splitting of each orbital in iron-based superconductors, multiple Fermi surfaces with multiple $3d$ -orbital components appear in the Brillouin zone. Figure 1.4 shows the Fermi surface of LaFeAsO obtained from a first-principles calculation. There are three hole pockets around Γ point and two electron pockets around M points, which are consistent with the measurement of angle-resolved photo emission spectroscopy (ARPES) [32]. Thanks to the quasi-two dimensionality, the Fermi surfaces are almost cylindrical. Fermi surfaces in other iron-based superconductors are almost same as those in LaFeAsO. This is due to the fact that characters of FeAs planes dominantly appears in the Fermi surface.

The good nesting of hole pockets and electron pocket is confirmed by the emergence of the antiferromagnetic phase. Because of its nesting vector, the configuration of the antiferromagnetic order is stripe [33]. The stripe antiferromagnetic order was at first predicted by theory and the measurement of optical conductivity [33], and sooner confirmed experimentally by neutron scattering for 1111-type compounds [34, 35] and 122-type compounds [36]. The configuration of the spin alignment is shown in Fig. 1.5. We define a -axis as an antiferromagnetically ordered direction, and b -axis as a ferromagnetically ordered direction. The four-fold symmetry of the crystal is broken to the two-fold symmetry by the structural phase transition above the Néel temperature, and the symmetry of the spin alignment is also the two-fold symmetry.

The band structure in the antiferromagnetic phase is basically obtained by reconstruction of paramagnetic band structure through a folding of the electron band and an opening of the SDW gaps. Figure 1.6 shows the electron band structure near the Fermi energy in 122-type compounds in the antiferromagnetic phase [37]. It is

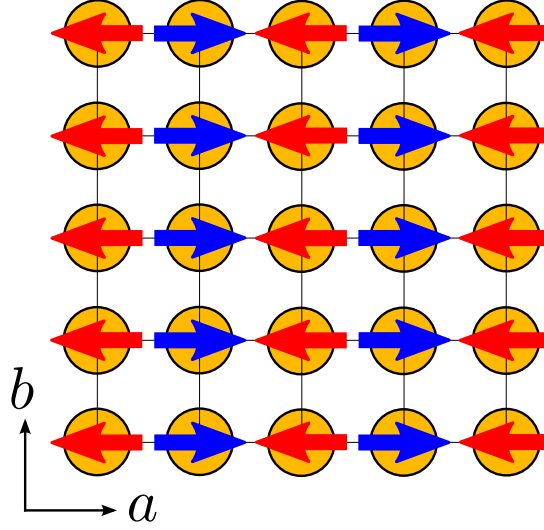


Fig. 1.5 Spin configuration of iron arsenide in antiferromagnetic phase. The spin aligns antiferromagnetically in one direction, and ferromagnetically in another direction. a -axis is along the antiferromagnetic-ordering direction, and b -axis is along the ferromagnetic-ordering direction.

interesting that Dirac-type linear dispersions (Dirac cone) whose point nodes exist very near the Fermi energy appear. The transport coefficients of iron arsenide in the antiferromagnetic phase are mainly attributed to the Dirac cones [37]. The existence of Dirac cones in the antiferromagnetic phase is predicted by SDW mean-field analysis [38, 39], and confirmed by a measurement of ARPES [40].

1.2.3 Superconducting pairing

It was suggested, soon after the discovery of iron-based superconductors, that the superconducting pairing in iron-based superconductors is mediated by a spin fluctuation in analogy to cuprates [41, 42]. This is guaranteed by a large spin fluctuation in the superconducting phase observed as a large peak structure in neutron-scattering experiments [43]. The superconducting pairing mediated by spin fluctuation is so-called $s_{\pm}$ -wave superconductivity. This is because the sign of superconducting order parameters at hole pockets are different from ones at electron pockets if we assume the spin-fluctuation mediated superconducting pairing mechanism.

However, the $s_{\pm}$ -wave superconductivity is weak against disorder, in contradiction to experiments that the superconducting phase is robust against doped impurities [44, 45, 46] or defects made by irradiation of α -particle [47] or proton beam [48]. Onari and Kontani suggested that the superconducting pairing in iron pnictide is mediated orbital fluctuation that superconducting phase is robust against the disorders, which is referred to as s_{++} -wave superconductivity due to the sign of the order parameters in the Fermi surfaces [49]. The s_{++} -wave mechanism can also explain the peak structure appearing in the spectrum of neutron scattering. The mechanism of the superconductivity in iron-based superconductors is still controversial.

Anyway, it is no doubt that both the spin and the orbitals of $3d$ electrons play an important role in iron-based superconductors. We can expect that understanding the orbital character in the antiferromagnetic phase leads to useful hints of the pairing mechanism.

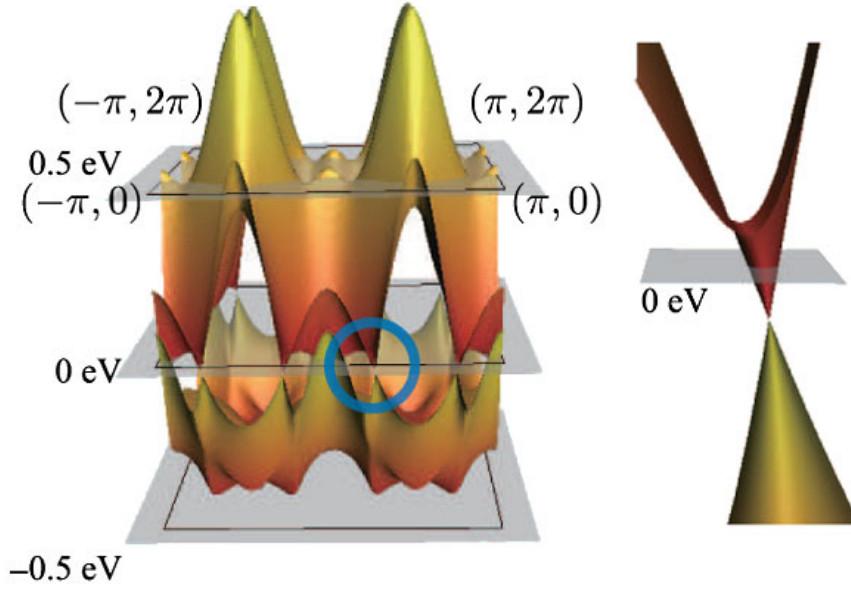


Fig. 1.6 Left panel: The band structure near the Fermi energy in the SDW state of 122-type iron-based superconductors. Right panel: The magnification of the band structure near the Dirac point indicated by the circle in the left panel. Reprinted with permission from T. Morinari *et al.* [37]. Copyright 2010 American Physical Society.

1.3 Anisotropy of Resistivity in Iron Arsenides

The anisotropy in the FeAs plane occurs through the breakdown of the four-fold symmetry by the tetragonal-to-orthorhombic structural phase transition and stripe-antiferromagnetic phase transition. The anisotropy also appears above the structural phase transition due to a nematic fluctuation. A nematic state with two-fold symmetry distinguishes two Fe-Fe directions. The nematic character is first observed by measurements of anisotropy of resistivity with uniaxial pressure detwinning the polycrystalline sample [50]. The nematic order is proved to be intrinsic by measuring the magnetic torque [51]. This implies that the anisotropy of resistivity is ascribed from not uniaxial pressure but the nematic order. The signature of the nematic order appears not only the anisotropy of resistivity [50, 52, 53, 54, 55, 56], but also the anisotropy of optical spectra [57, 58, 59]. In addition, the anisotropic state directly observed by scanning tunneling spectroscopy [60, 61] and NMR [62]. Furthermore, the orbital ordering in nematic phase is clearly shown by ARPES [63].

The origin of the anisotropy appearing in optical conductivity can be explained from the band structure of iron arsenide [64, 65]. Since only direct optical transitions dominantly contribute to its spectra, the orbitals of an initial state and a final state in an optical transition play important roles in the anisotropy of optical conductivity [65]. However, the situation is different with regard to the anisotropy of resistivity. In calculations of resistivity, we should determine what scatters electrons in iron arsenide.

The anisotropy of resistivity is measured above the structural transition temperature T_S and the antiferromagnetic transition temperature T_N . One of the possible origins of the anisotropy above the transition temperatures has been suggested based on spin fluctuation [66]. In this assumption, the spin fluctuations scatter electrons

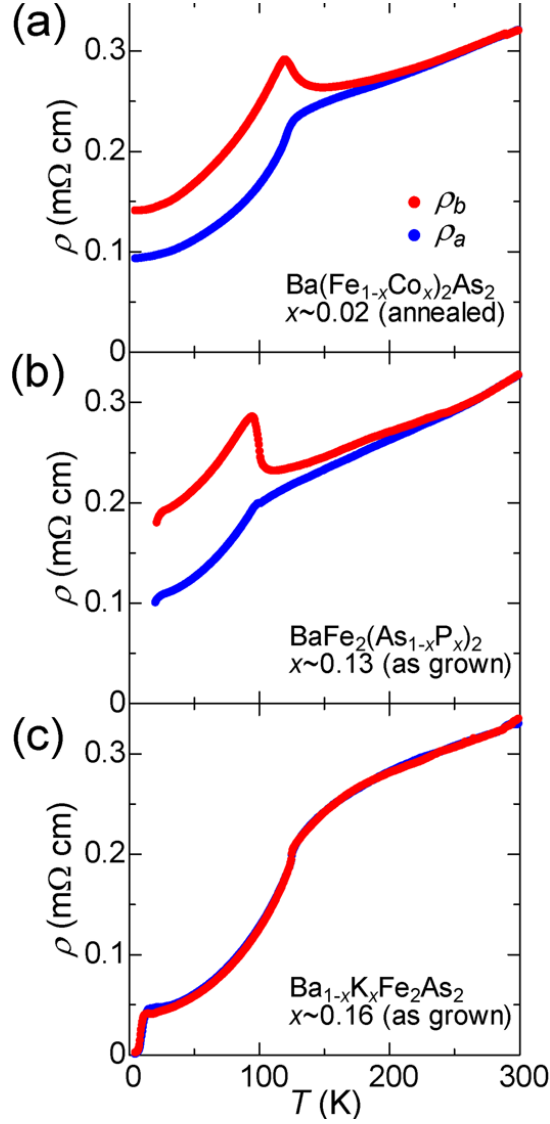


Fig. 1.7 Experimentally observed temperature dependence of anisotropy of resistivity in (a) $\text{Ba}(\text{Fe}_{0.98}\text{Co}_{0.02})_2\text{As}_2$, (b) $\text{BaFe}_2(\text{As}_{0.87}\text{P}_{0.13})_2$, and (c) $\text{Ba}_{0.84}\text{K}_{0.16}\text{Fe}_2\text{As}_2$. The red curve and the blue curve indicate the resistivity along a - or b -axis, respectively. Reprinted with permission from S. Ishida *et al.* [70]. Copyright 2013 American Chemical Society.

develops just above the transition temperatures, which is considered as the origin of the resistivity. Hot spots ascribed to the elliptic electron pockets and circular hole pockets lead to the anisotropy of charge transport. Another possible origin is orbital ordering [67, 68] that may contribute to the pairing of electrons in superconducting phase. Although a prediction of the conductivity derived from Drude weight is inconsistent with experiments [69], the resistivity including impurity scattering reproduces the anisotropy observed in experiments [68].

Since the antiferromagnetic phase has electronic states different from that of the paramagnetic phase, the origin might be distinguished from those in the paramagnetic phase. Resistivity measurements for detwinned samples of Co-doped BaFe_2As_2 [50, 56] have clearly shown that in-plane anisotropy is strongly enhanced below T_N . In

the antiferromagnetic phase of parent compound BaFe_2As_2 , the anisotropy almost disappears with annealing the sample [53], while it remains finite in Co-doped and P-doped samples [56] as shown in Fig. 1.7 (a) and (b), respectively. This result suggests that the anisotropy of resistivity should be induced by impurity scattering of Co atoms substituted for Fe atoms. In hole-doped compounds $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$, the anisotropy disappears [70] and reverses [71] as shown in Fig. 1.7 (c). This reversing of the anisotropy has been proposed theoretically by taking into account spin fluctuation in the nematic state of paramagnetic phase [66, 71]. However, this theory cannot be applied to the antiferromagnetic phase. It is still unclear whether the effect of impurity is crucial in accounting for the anisotropy [72] or the Drude weight defined without impurities is enough for this purpose [73, 74, 75].

Reflecting the importance of understanding the nematic phase in the study of iron-based superconductors, there are several reviews focusing on this phase. The progress of investigation on electronic anisotropy is summarized by Fisher *et al.* [76]. The origin of nematic phase is discussed in a review article written by Fernandes *et al.* [77].

1.4 Resonant Inelastic X-ray Scattering: a newly appearing probe of collective mode

Taking the opportunity of the discovery of iron-based superconductors, the spin dynamics of itinerant antiferromagnetic systems with multiple $3d$ orbitals has attracted much attentions again. The aspect of $3d$ -orbital degrees of freedom is also newly focused on. Although Cr as representative material of itinerant SDW has been investigated for long time, it is important to reinvestigate at Cr again from the view of orbitals.

The spin dynamics of the incommensurate Cr has been investigated by the measurements of neutron scattering, optical conductivity, and NMR until now [6, 7]. Especially, neutron scattering is considered as the most powerful tool for the investigation of spin excitations. The incommensurate spin excitations in the low-energy region (<100 meV) have been clarified experimentally [8] and theoretically [78, 79]. However, the spin dynamics in the high-energy region, where the interplay of collective spin-wave excitation and individual particle-hole excitation is expected to occur, has rarely been examined except for the data up to 550 meV with a large error bar [80].

Another candidate for observation of collective mode is resonant inelastic x-ray scattering (RIXS) [81]. The RIXS spectrum is theoretically written as the summation of spin and charge susceptibilities reflecting these collective modes. Moreover, the coefficients in the summation depend on the orbitals of excited electrons, which allow us to observe the orbital degrees of freedom. However, the observation of collective mode in RIXS requires large x-ray flux, whose magnitude was hard to achieve ten or more years ago.

Recent developments of x-ray synchrotron radiation facility have solved these problems [81]. Accompanying with the developments, the resolution of x-ray scattering spectrum is in the order of several hundreds meV. We can now regard RIXS as a complement of neutron-scattering experiments for detecting spin excitations.

The theoretical calculation of spin susceptibilities [82] and RIXS spectra [83] in antiferromagnetic phase of iron arsenide has already been carried out. The calculated RIXS spectra illustrate the relationship between orbital excitations and spin excitations. The RIXS is thus a powerful tool for detecting the orbital physics.

1.5 Purpose of This Thesis

The purpose of this thesis is to elucidate roles of orbitals and spins in transition metals and their compounds. The metals with an SDW state are suitable for this purpose. Thus, we focus on iron arsenide and Cr. Iron arsenide is one of the high-temperature superconductors, and the understanding of its physical properties is highly desired. Cr is a representative material of an itinerant SDW. The basic knowledge of this material is well established, but multi-orbital nature is still not known well. These two materials are the best choice for the study of multi-orbital antiferromagnetic metals.

We first introduce the model and methods used in this thesis in Chap. 2. We derive the multi-orbital Hubbard model in Sec. 2.1, which is the basis of our study. The antiferromagnetic state is obtained by a mean-field theory of the Hubbard model. Next, we derive the formula of resistivity (Sec. 2.2) and RIXS spectra (Sec. 2.3) to be used in iron arsenide and Cr, respectively.

In Chap. 3, we theoretically examine the resistivity in the antiferromagnetic phase of iron arsenide at zero temperature. The Fermi surfaces in the AFM state, including Dirac pockets [38, 37], are distributed near the Γ point in a doping-dependent manner. The anisotropy of resistivity obtained by a memory-function approach [84] taking into account impurity scattering is consistent with the experiments near the undoped region, where the resistivity in the antiferromagnetically ordered direction is smaller than that in the ferromagnetically ordered direction. We find that the anisotropy of the scattering rate is independent of hole concentration, once the two electron pockets above and below the Γ -point hole pocket disappear. On the other hand, the anisotropy of the Drude weight monotonically decreases reflecting the distortion of the Dirac Fermi surface, and this eventually leads to the reverse of the anisotropy of resistivity. The reverse has been observed in experiments [71]. These results indicate that the origin of the anisotropy is attributed to the interplay of impurity scattering and anisotropic electronic states, both of which are strongly influenced by the topology of the Fermi surface.

For understanding the dynamical properties of multi-orbital systems, we investigate Cr with a commensurate SDW state by using a multi-band Hubbard model composed of $3d$ and $4s$ orbitals in Chap. 4. After a self-consistent calculation based on an SDW mean-field approximation, we obtain the dynamical spin susceptibility by employing random-phase approximation. We find a collective spin-wave excitation undamped up to ~ 0.6 eV. Above the energy, the excitation overlaps individual particle-hole excitations, leading to broad spectral weight. From the results of the longitudinal spin susceptibility, we find that its spectral weight is mainly distributed above the energy of the spin-wave mode. We expect that these features may be observed in inelastic neutron scattering experiments in the near future.

RIXS tuned for the L edge of transition metal has been recognized as a powerful tool to investigate not only charge and orbital ($d-d$) excitations but also spin excitation in the energy and momentum spaces. By using a fast-collision approximation for the RIXS process, we calculate Cr L -edge RIXS intensity and compare the spectrum with inelastic neutron scattering spectrum. We find large spectral weight coming from particle-hole excitations with various orbital channels. This eventually makes the collective spin-wave excitation less visible. This is a contrast to cuprates, where the spin-wave excitation has been clearly observed in the Cu L -edge RIXS [85, 86]. However, since there are remnants of the spin-wave excitation, it may be possible to

observe the excitation in RIXS in the near future.

Chapter 2

MODEL AND METHODS

In iron arsenide and chromium (Cr), which we are treating in this thesis, their Fermi surface is composed of $3d$ -orbital electrons ($4s$ -orbital electrons is hybridized in the band structure near the Fermi surface in Cr). In order to investigate the electron dynamics of these materials, we use a multi-orbital Hubbard model. The characters of d orbitals are included in the kinetic terms as hopping integrals. On-site Coulomb interactions between $3d$ -orbital electrons are also orbital-dependent. However, since they exhibit a bit complicated form, we assume that on-site Coulomb interactions are independent of a kind of orbitals.

The ground state of these materials is a spin-density-wave (SDW) state. This state is obtained from a self-consistent calculation based on a mean-field approximation applying to the multi-orbital Hubbard model. The process of obtaining the SDW order parameter is very similar to the process of obtaining a superconducting order parameter from Bogoliubov-de Gennes equations. After obtaining the order parameter, we mainly derive two physical quantities; resistivity and resonant inelastic x-ray scattering (RIXS) spectrum.

The resistivity is calculated from a memory-function approach. This approach is applied to the calculation of resistivity with dilute impurity scattering. The original work of memory function is applied to single-band electron gas. We improve the memory-function formalism to be capable of applying to multi-orbital systems. This result is applied to the calculation of the anisotropy of resistivity in iron arsenide in Chap. 3.

The RIXS spectrum is calculated from charge and spin susceptibilities. The susceptibilities are obtained from random phase approximation (RPA). Thanks to recent developments of energy resolution, RIXS becomes a new tool for detecting charge and spin excitations. We apply the formula of the RIXS spectrum to Cr in Chap. 4.

2.1 Multi-Orbital Hubbard Model in Antiferromagnetic Phase

In this section, we introduce a $3d$ -orbital Hubbard model, which contains multi-orbital characters. The Hamiltonian of this model consists of kinetic energy terms including hopping integrals and on-site Coulomb interaction terms. To obtain the SDW ground state, we make use of a mean-field approximation to the on-site interaction terms. The order parameter in the SDW state is obtained by numerical self-consistent calculations. We also derive Green's function of the multi-orbital Hamiltonian.

2.1.1 3d-orbital Hamiltonian

We consider a system where one unit cell contains one 3d-transition metal atom. The lattice spacing is set to be unit length. The 3d-orbital Hamiltonian with on-site Coulomb interaction is written as $H_{3d} = H_0 + H_{\text{int}}$ [87], where

$$H_0 = \sum_{i,j} \sum_{\sigma,\mu,\nu} \{t(\Delta_{i,j}; \mu, \nu) + \varepsilon_\mu \delta_{\mu,\nu}\} c_{i\mu\sigma}^\dagger c_{j\nu\sigma} \quad (2.1)$$

and

$$H_{\text{int}} = \frac{1}{2} \left\{ \sum_{i,\mu,\sigma} U_\mu n_{i\mu\sigma} n_{i\mu,-\sigma} + \sum_{i,\mu \neq \nu,\sigma} U_{\mu,\nu} n_{i\mu\sigma} n_{i\nu,-\sigma} + \sum_{i,\mu \neq \nu,\sigma} (U_{\mu,\nu} - J_{\mu,\nu}) n_{i\mu\sigma} n_{i\nu\sigma} \right. \\ \left. - \sum_{i,\mu \neq \nu,\sigma} J_{\mu,\nu} c_{i\mu\sigma}^\dagger c_{i\mu,-\sigma} c_{i\nu,-\sigma}^\dagger c_{i\nu\sigma} + \sum_{i,\mu \neq \nu,\sigma} J'_{\mu,\nu} c_{i\mu\sigma}^\dagger c_{i\mu,-\sigma}^\dagger c_{i\nu,-\sigma} c_{i\nu\sigma} \right\} \quad (2.2)$$

with $n_{i\mu\sigma} = c_{i\mu\sigma}^\dagger c_{i\mu\sigma}$, where $c_{i\mu\sigma}$ ($c_{i\mu\sigma}^\dagger$) is an annihilation (creation) operator of an electron at site i with spin σ and orbital μ . They satisfy the following anti-commutation relations

$$\{c_{i\mu\sigma}, c_{j\nu\sigma'}\} = \{c_{i\mu\sigma}^\dagger, c_{j\nu\sigma'}^\dagger\} = 0 \quad (2.3)$$

$$\{c_{i\mu\sigma}, c_{j\nu\sigma'}^\dagger\} = \delta_{i,j} \delta_{\mu,\nu} \delta_{\sigma,\sigma'}. \quad (2.4)$$

$t(\Delta_{i,j}; \mu, \nu)$ is a hopping integral between site i with orbital μ and site j with orbital ν , and ε_μ is on-site energy of orbital μ . U_μ , $U_{\mu,\nu}$, $J_{\mu,\nu}$, and $J'_{\mu,\nu}$ are called inter-orbital, intra-orbital, Hund's coupling, and pair hopping interactions, respectively. We assume that $J'_{\mu,\nu} = J_{\mu,\nu}$ and they are independent of the orbital index; $U_\mu = U$, $U_{\mu,\nu} = U'$, and $J_{\mu,\nu} = J$. Imposed rotational symmetry $U' = U - 2J$, Eq. (2.2) becomes

$$H_{\text{int}} = \frac{U}{2} \sum_{i,\mu,\sigma} n_{i\mu\sigma} n_{i\mu,-\sigma} + \frac{U-2J}{2} \sum_{i,\mu \neq \nu,\sigma} n_{i\mu\sigma} n_{i\nu,-\sigma} + \frac{U-3J}{2} \sum_{i,\mu \neq \nu,\sigma} n_{i\mu\sigma} n_{i\nu\sigma} \\ - \frac{J}{2} \sum_{i,\mu \neq \nu,\sigma} c_{i\mu\sigma}^\dagger c_{i\mu,-\sigma} c_{i\nu,-\sigma}^\dagger c_{i\nu\sigma} + \frac{J}{2} \sum_{i,\mu \neq \nu,\sigma} c_{i\mu\sigma}^\dagger c_{i\nu\sigma} c_{i\mu,-\sigma}^\dagger c_{i\nu,-\sigma}. \quad (2.5)$$

The Coulomb term H_U proportional to U , reads

$$H_U = \frac{U}{2} \sum_{i,\mu,\sigma} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\mu,-\sigma}^\dagger c_{i\mu,-\sigma} \\ + \frac{U}{2} \sum_{i,\mu \neq \nu,\sigma} \left\{ c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu,-\sigma}^\dagger c_{i\nu,-\sigma} + c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma}^\dagger c_{i\nu\sigma} \right\} \\ = \frac{U}{2} \sum_{i,\mu,\nu,\sigma,\sigma'} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} (1 - \delta_{\mu,\nu} \delta_{\sigma,\sigma'}), \quad (2.6)$$

and the exchange term H_J proportional to J reads

$$H_J = -J \sum_{i,\mu \neq \nu,\sigma} \left\{ c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu,-\sigma}^\dagger c_{i\nu,-\sigma} + \frac{3}{2} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma}^\dagger c_{i\nu\sigma} \right\}$$

$$\begin{aligned}
& + \frac{1}{2} c_{i\mu\sigma}^\dagger c_{i\mu,-\sigma} c_{i\nu,-\sigma}^\dagger c_{i\nu\sigma} - \frac{1}{2} c_{i\mu\sigma}^\dagger c_{i\nu\sigma} c_{i\mu,-\sigma}^\dagger c_{i\nu,-\sigma} \Big\} \\
& = -\frac{J}{2} \sum_{i,\mu \neq \nu, \sigma, \sigma'} \left\{ c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} (2 + \delta_{\sigma, \sigma'}) \right. \\
& \quad \left. - \left(c_{i\mu\sigma}^\dagger c_{i\nu\sigma} c_{i\nu\sigma'}^\dagger c_{i\mu\sigma'} + c_{i\mu\sigma}^\dagger c_{i\nu\sigma} c_{i\mu\sigma'}^\dagger c_{i\nu\sigma'} \right) (1 - \delta_{\sigma, \sigma'}) \right\}. \quad (2.7)
\end{aligned}$$

2.1.2 Mean-field approximation

Generally speaking, it is difficult to solve a many-body problem. In order to convert it to a one-body problem, we apply the mean-field approximation to the interaction Hamiltonians. For H_U ,

$$\begin{aligned}
& c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \\
& \simeq \left\{ \left\langle c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \right\rangle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} + c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \left\langle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \right\rangle \left\langle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle \right\} \\
& - \delta_{\sigma, \sigma'} \left\{ \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle c_{i\nu\sigma}^\dagger c_{i\mu\sigma} + c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \left\langle c_{i\nu\sigma}^\dagger c_{i\mu\sigma} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle \left\langle c_{i\nu\sigma}^\dagger c_{i\mu\sigma} \right\rangle \right\} \quad (2.8)
\end{aligned}$$

because the Wick's theorem says

$$\begin{aligned}
& \left\langle c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle \\
& = \left\langle c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \right\rangle \left\langle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma'} \right\rangle \left\langle c_{i\nu\sigma'}^\dagger c_{i\mu\sigma} \right\rangle \\
& = \left\langle \left\{ \left\langle c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \right\rangle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} + c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \left\langle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \right\rangle \left\langle c_{i\nu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle \right\} \right. \\
& \quad \left. - \delta_{\sigma, \sigma'} \left\{ \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle c_{i\nu\sigma}^\dagger c_{i\mu\sigma} + c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \left\langle c_{i\nu\sigma}^\dagger c_{i\mu\sigma} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle \left\langle c_{i\nu\sigma}^\dagger c_{i\mu\sigma} \right\rangle \right\} \right\rangle. \quad (2.9)
\end{aligned}$$

We also obtain, for H_J ,

$$\begin{aligned}
& c_{i\mu\sigma}^\dagger c_{i\nu\sigma} c_{i\nu\sigma'}^\dagger c_{i\mu\sigma'} (1 - \delta_{\sigma, \sigma'}) \\
& \simeq \left\{ \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle c_{i\nu\sigma'}^\dagger c_{i\mu\sigma'} + c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \left\langle c_{i\nu\sigma'}^\dagger c_{i\mu\sigma'} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle \left\langle c_{i\nu\sigma'}^\dagger c_{i\mu\sigma'} \right\rangle \right\} \\
& \quad \times (1 - \delta_{\sigma, \sigma'}) \quad (2.10)
\end{aligned}$$

and

$$\begin{aligned}
& c_{i\mu\sigma}^\dagger c_{i\nu\sigma} c_{i\mu\sigma'}^\dagger c_{i\nu\sigma'} (1 - \delta_{\sigma, \sigma'}) \\
& \simeq \left\{ \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle c_{i\mu\sigma'}^\dagger c_{i\nu\sigma'} + c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \left\langle c_{i\mu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle - \left\langle c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \right\rangle \left\langle c_{i\mu\sigma'}^\dagger c_{i\nu\sigma'} \right\rangle \right\} \\
& \quad \times (1 - \delta_{\sigma, \sigma'}). \quad (2.11)
\end{aligned}$$

With wave vector $\mathbf{k}$ in a periodic boundary condition, we can write the annihilation and creation operators as

$$\begin{aligned}
c_{i\nu\sigma} &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}_i} c_{\mathbf{k}\nu\sigma} \\
c_{i\mu\sigma}^\dagger &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{r}_i} c_{\mathbf{k}\mu\sigma}^\dagger, \quad (2.12)
\end{aligned}$$

respectively, with the number of lattice point N . We define

$$n_{i\mu\nu\sigma} = c_{i\mu\sigma}^\dagger c_{i\nu\sigma} = \sum_{\mathbf{q}} n_{\mathbf{q}\mu\nu\sigma} e^{i\mathbf{q}\cdot\mathbf{r}} \quad (2.13)$$

$$n_{\mathbf{q}\mu\nu\sigma} = \frac{1}{N} \sum_{\mathbf{k}} c_{\mathbf{k}\mu\sigma}^\dagger c_{\mathbf{k}+\mathbf{q},\nu\sigma}. \quad (2.14)$$

The mean-field 3d-orbital Hamiltonian is $H_{3d}^{\text{MF}} = H_0 + H_U^{\text{MF}} + H_J^{\text{MF}}$, where

$$H_0 = \sum_{\mathbf{k},\sigma} \sum_{\mu,\nu} \sum_{\Delta} t(\Delta; \mu, \nu) e^{-i\mathbf{k}\cdot\Delta} c_{\mathbf{k}\mu\sigma}^\dagger c_{\mathbf{k}\nu\sigma}, \quad (2.15)$$

$$H_U^{\text{MF}} = U \sum_{\mathbf{k},\sigma} \sum_{\mu,\nu} \sum_{\mathbf{q}} \left\{ \left(\sum_{\nu',\sigma'} \langle n_{\mathbf{q}\nu'\nu'\sigma'} \rangle - \langle n_{\mathbf{q}\mu\mu\sigma} \rangle \right) \delta_{\mu,\nu} - \langle n_{\mathbf{q}\nu\mu\sigma} \rangle (1 - \delta_{\mu,\nu}) \right\} \\ \times c_{\mathbf{k}+\mathbf{q},\mu\sigma}^\dagger c_{\mathbf{k}\nu\sigma} + C_U, \quad (2.16)$$

and

$$H_J^{\text{MF}} = J \sum_{\mathbf{k},\sigma} \sum_{\mu,\nu} \sum_{\mathbf{q}} \left[\left\{ -3 \left(\sum_{\nu'} \langle n_{\mathbf{q},\nu',\nu',\sigma} \rangle - \langle n_{\mathbf{q}\mu\mu\sigma} \rangle \right) - 2 \left(\sum_{\nu'} \langle n_{\mathbf{q}\nu'\nu',-\sigma} \rangle - \langle n_{\mathbf{q}\mu\mu,-\sigma} \rangle \right) \right\} \delta_{\mu,\nu} \right. \\ \left. + \{ 3 \langle n_{\mathbf{q}\nu\mu\sigma} \rangle + \langle n_{\mathbf{q}\nu\mu,-\sigma} \rangle + \langle n_{\mathbf{q}\mu\nu,-\sigma} \rangle \} (1 - \delta_{\mu,\nu}) \right] c_{\mathbf{k}+\mathbf{q},\mu\sigma}^\dagger c_{\mathbf{k}\nu\sigma} + C_J. \quad (2.17)$$

Here, C_U and C_J are constant terms. To confirm whether the considered state is the ground state, we should estimate the total energy containing these constant terms.

2.1.3 Spin-density wave

The Brillouin zone with an SDW is folded to $1/N_{\text{SDW}}$ as compared with the original paramagnetic Brillouin zone, where N_{SDW} is a period of the SDW. $\langle n_{\mathbf{q}\mu\nu\sigma} \rangle$ appearing in Eqs. (2.16) and (2.17) is non-zero if $\mathbf{q} = l\mathbf{Q}$ ($l = 0, \dots, N_{\text{SDW}} - 1$), where $\mathbf{Q}$ is an SDW wave vector [88]. The summation of $\mathbf{k}$ becomes

$$\mathbf{k} \rightarrow \mathbf{k}_0 + m\mathbf{Q}, \quad \sum_{\mathbf{k}} \rightarrow \sum_{\mathbf{k}_0} \sum_{m=0}^{N_{\text{SDW}}-1}, \quad (2.18)$$

where the summation of $\mathbf{k}_0$ is over the $1/N_{\text{SDW}}$ folded Brillouin zone. The Hamiltonians in Eqs. (2.15), (2.16), and (2.17) with the SDW are written as

$$H_0 = \sum_{\mathbf{k}_0,\sigma} \sum_{\mu,\nu} \sum_m c_{\mathbf{k}_0+m\mathbf{Q},\mu\sigma}^\dagger \left\{ \sum_{\Delta} t(\Delta; \mu, \nu) e^{-i(\mathbf{k}_0+m\mathbf{Q})\cdot\Delta} \right\} c_{\mathbf{k}_0+m\mathbf{Q},\nu\sigma}, \quad (2.19)$$

$$H_U^{\text{MF}} = U \sum_{\mathbf{k}_0,\sigma} \sum_{\mu,\nu} \sum_{m,m'} c_{\mathbf{k}_0+m'\mathbf{Q},\mu\sigma}^\dagger \left\{ \left(\sum_{\nu',\sigma'} \langle n_{(m'-m)\mathbf{Q},\nu'\nu'\sigma'} \rangle \right. \right.$$

$$- \langle n_{(m'-m)\mathbf{Q},\mu\mu\sigma} \rangle \Big) \delta_{\mu,\nu} - \langle n_{(m'-m)\mathbf{Q},\nu\mu\sigma} \rangle (1 - \delta_{\mu,\nu}) \Big\} c_{\mathbf{k}_0+m\mathbf{Q},\nu\sigma}, \quad (2.20)$$

and

$$\begin{aligned} H_J^{\text{MF}} = & J \sum_{\mathbf{k},\sigma} \sum_{\mu,\nu} \sum_{m,m'} c_{\mathbf{k}_0+m'\mathbf{Q},\mu,\sigma}^\dagger \\ & \times \left[\left\{ -3 \left(\langle n_{(m'-m)\mathbf{Q},\sigma} \rangle - \langle n_{(m'-m)\mathbf{Q},\mu\sigma} \rangle \right) - 2 \left(\langle n_{(m'-m)\mathbf{Q},-\sigma} \rangle - \langle n_{(m'-m)\mathbf{Q},\mu,-\sigma} \rangle \right) \right\} \right. \\ & \times \delta_{\mu,\nu} + \left. \left(3 \langle n_{(m'-m)\mathbf{Q},\nu\mu\sigma} \rangle + \langle n_{(m'-m)\mathbf{Q},\nu\mu,-\sigma} \rangle + \langle n_{(m'-m)\mathbf{Q},\mu\nu,-\sigma} \rangle \right) (1 - \delta_{\mu,\nu}) \right] \\ & \times c_{\mathbf{k}_0+m\mathbf{Q},\nu\sigma}, \quad (2.21) \end{aligned}$$

respectively. Here, we define $n_{(m'-m)\mathbf{Q},\mu\sigma} = n_{(m'-m)\mathbf{Q},\mu\mu\sigma}$ and $n_{(m'-m)\mathbf{Q},\sigma} = \sum_{\nu'} n_{(m'-m)\mathbf{Q},\nu'\sigma}$. The mean-field 3d-orbitals Hamiltonian with SDW can also be written as

$$H_{3d}^{\text{MF}} = \sum_{\mathbf{k}_0,\sigma} \sum_{\mu,\nu} \sum_{m,m'} c_{\mathbf{k}_0+m'\mathbf{Q},\mu\sigma}^\dagger (H_{\mathbf{k}_0,\sigma}^{\text{MF}})_{\mu,m';\nu,m} c_{\mathbf{k}_0+m\mathbf{Q},\nu\sigma}, \quad (2.22)$$

where $(H_{\mathbf{k}_0,\sigma}^{\text{MF}})_{\mu,m';\nu,m}$ is the matrix element of the mean-field Hamiltonian.

By Bogoliubov transformation, we obtain its wave function and energy. The operator of an electron transforms as

$$c_{\mathbf{k}_0+m\mathbf{Q},\mu\sigma} = \sum_{\epsilon} \psi_{\mu,m;\epsilon}(\mathbf{k}_0, \sigma) \gamma_{\mathbf{k}_0,\epsilon,\sigma} \quad (2.23)$$

with wave function $\psi_{\mu,m;\epsilon}(\mathbf{k}_0, \sigma)$ and band quasi-particle operator $\gamma_{\mathbf{k}_0,\epsilon,\sigma}$, where ϵ is the band index. $\gamma_{\mathbf{k}_0,\epsilon,\sigma}$ satisfies an anti-commutation relationship of ordinary fermions. The number of band index is equal to the product of N_{SDW} and the number of orbitals. The Hamiltonian becomes

$$H_{3d}^{\text{MF}} = \sum_{\mathbf{k}_0,\sigma} \sum_{\epsilon} E_{\mathbf{k}_0,\epsilon,\sigma} \gamma_{\mathbf{k}_0,\epsilon,\sigma}^\dagger \gamma_{\mathbf{k}_0,\epsilon,\sigma}, \quad (2.24)$$

where $E_{\mathbf{k}_0,\epsilon,\sigma}$ is the energy of the band quasi-particle. Note that Bogoliubov transformation is a unitary transformation, which satisfies

$$\sum_{\epsilon} \psi_{\mu,m;\epsilon}(\mathbf{k}_0, \sigma) \psi_{\mu',m';\epsilon}^*(\mathbf{k}_0, \sigma) = \delta_{\mu,\mu'} \delta_{m,m'}. \quad (2.25)$$

$\langle n_{\mathbf{q}\mu\nu\sigma} \rangle$ is written with band quasi-particle operators as

$$\begin{aligned} \langle n_{l\mathbf{Q},\mu\nu\sigma} \rangle &= \frac{1}{N} \sum_{\mathbf{k}} \langle c_{\mathbf{k}\mu\sigma}^\dagger c_{\mathbf{k}+l\mathbf{Q},\nu\sigma} \rangle = \frac{1}{N} \sum_{\mathbf{k}_0,m} \langle c_{\mathbf{k}_0+m\mathbf{Q},\mu\sigma}^\dagger c_{\mathbf{k}_0+(m+l)\mathbf{Q},\nu\sigma} \rangle \\ &= \frac{1}{N} \sum_{\mathbf{k}_0,m} \sum_{\epsilon} \psi_{\mu,m;\epsilon}^*(\mathbf{k}_0, \sigma) \psi_{\nu,m+l;\epsilon}(\mathbf{k}_0, \sigma) f(E_{\mathbf{k}_0,\epsilon,\sigma}), \end{aligned} \quad (2.26)$$

and this equation implies $\langle n_{-l\mathbf{Q},\mu\nu\sigma} \rangle = \langle n_{l\mathbf{Q},\nu\mu\sigma} \rangle^*$. In Eq. (2.26), we use the relation

$$\langle \gamma_{\mathbf{k}_0,\epsilon,\sigma}^\dagger \gamma_{\mathbf{k}_0,\epsilon',\sigma} \rangle = f(E_{\mathbf{k}_0,\epsilon,\sigma}) \delta_{\epsilon,\epsilon'}, \quad (2.27)$$

where $f(E - E_c) = 1/(e^{(E-E_c)/k_B T} + 1)$ is a Fermi distribution function with chemical potential E_c . We can obtain a self-consistent result of $\langle n_{q\mu\nu\sigma} \rangle$ by diagonalizing Eq. (2.22) and calculating Eq. (2.26), repeatedly.

2.1.4 Multi-orbital Green's function

With use of the band-quasiparticle annihilation operators $\gamma_\epsilon(\mathbf{k}_0, \sigma)$, the electron annihilation operator $c_{i\mu\sigma}$ becomes

$$\begin{aligned} c_{i\mu\sigma} &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k}_0, m} e^{i(\mathbf{k}_0 + m\mathbf{Q}) \cdot \mathbf{r}_i} c_{\mathbf{k}_0 + m\mathbf{Q}, \mu\sigma} \\ &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k}_0, m} \sum_{\epsilon} e^{i(\mathbf{k}_0 + m\mathbf{Q}) \cdot \mathbf{r}_i} \psi_{\mu, m; \epsilon}(\mathbf{k}_0, \sigma) \gamma_{\mathbf{k}_0, \epsilon, \sigma}. \end{aligned} \quad (2.28)$$

Temperature Green's function of band quasiparticles is given as

$$\begin{aligned} \mathcal{G}_{\epsilon, \epsilon', \sigma}^{\text{MF}}(\mathbf{k}_0, \tau; \mathbf{k}'_0, \tau') &= -\langle T \gamma_{\mathbf{k}_0, \epsilon, \sigma}(\tau) \gamma_{\mathbf{k}'_0, \epsilon', \sigma}^\dagger(\tau') \rangle \\ &= \frac{1}{\hbar\beta} \sum_{\omega_n} \mathcal{G}_{\epsilon, \sigma}^{\text{MF}}(\mathbf{k}_0, i\omega_n) e^{-i\omega_n(\tau - \tau')} \delta_{\epsilon, \epsilon'} \delta_{\mathbf{k}_0, \mathbf{k}'_0}, \end{aligned} \quad (2.29)$$

where τ is an imaginary time and

$$\mathcal{G}_{\epsilon, \sigma}^{\text{MF}}(\mathbf{k}_0, i\omega_n) = \frac{1}{i\omega_n - (E_{\mathbf{k}_0, \sigma, \epsilon} - E_c)/\hbar} \quad (2.30)$$

$$\omega_n = \frac{(2n+1)\pi}{\hbar\beta}, \quad n = 0, \pm 1, \pm 2, \dots \quad (2.31)$$

Since the temperature Green's function depends on $\tau - \tau'$, it can be written as

$$\mathcal{G}_{\epsilon, \epsilon', \sigma}^{\text{MF}}(\mathbf{k}_0, \tau; \mathbf{k}'_0, \tau') = \mathcal{G}_{\epsilon, \sigma}^{\text{MF}}(\mathbf{k}_0, \tau - \tau') \delta_{\epsilon, \epsilon'} \delta_{\mathbf{k}_0, \mathbf{k}'_0}. \quad (2.32)$$

Thus, temperature Green's function of single electron is

$$\begin{aligned} \mathcal{G}_{\mu, \nu, \sigma}(\mathbf{r}_i, \tau; \mathbf{r}_j, \tau') &= -\langle T c_{i\mu\sigma}(\tau) c_{j\nu\sigma}^\dagger(\tau') \rangle \\ &= \frac{1}{N} \sum_{\mathbf{k}_0, \mathbf{k}'_0} \sum_{m, m'} \sum_{\epsilon, \epsilon'} \frac{1}{\hbar\beta} \sum_{\omega_n} \mathcal{G}_{\epsilon, \sigma}^{\text{MF}}(\mathbf{k}_0, i\omega_n) e^{-i\omega_n(\tau - \tau')} \delta_{\epsilon, \epsilon'} \delta_{\mathbf{k}_0, \mathbf{k}'_0} \\ &\quad \times \psi_{\mu, m; \epsilon}(\mathbf{k}_0, \sigma) \psi_{\nu, m'; \epsilon'}^*(\mathbf{k}'_0, \sigma) e^{i(\mathbf{k}_0 + m\mathbf{Q}) \cdot \mathbf{r}_i} e^{-i(\mathbf{k}'_0 + m'\mathbf{Q}) \cdot \mathbf{r}_j} \\ &= \frac{1}{N} \sum_{\mathbf{k}_0} \sum_{m, m'} \sum_{\epsilon} \frac{1}{\hbar\beta} \sum_{\omega_n} \frac{\psi_{\mu, m; \epsilon}(\mathbf{k}_0, \sigma) \psi_{\nu, m'; \epsilon}^*(\mathbf{k}_0, \sigma)}{i\omega_n - (E_{\mathbf{k}_0, \sigma, \epsilon} - E_c)/\hbar} \\ &\quad \times e^{i(\mathbf{k}_0 + m\mathbf{Q}) \cdot \mathbf{r}_i - i(\mathbf{k}_0 + m'\mathbf{Q}) \cdot \mathbf{r}_j} e^{-i\omega_n(\tau - \tau')}. \end{aligned} \quad (2.33)$$

2.2 Memory-Function Formalism

The memory-function approach for calculating the conductivity in homogeneous free electron gas with dilute impurity is introduced by Götze and Wölfle [84]. We expand the memory-function formalization from the case of free electrons to the case of a multi-orbital system.

2.2.1 Conductivity by Kubo formula

The retarded correlation function of operators P and Q , or the response P from external field Q in linear response theory, is written as

$$\chi_{PQ}(z) = \langle\langle P; Q \rangle\rangle_z = -i \int_0^\infty dt e^{izt} \langle [P(t), Q] \rangle, \quad (2.34)$$

where $P(t)$ is the Heisenberg representation of P . The equation of motion on $P(t)$ is $\frac{\partial P(t)}{\partial t} = \frac{1}{i\hbar} [P(t), H^{\text{total}}]$, where H^{total} is a full Hamiltonian of considering system. Integrated by parts, Eq. (2.34) becomes

$$\begin{aligned} z \langle\langle P; Q \rangle\rangle_z &= \langle [P, Q] \rangle + \frac{1}{\hbar} \langle\langle [P, H^{\text{total}}]; Q \rangle\rangle_z \\ &= \langle [P, Q] \rangle - \frac{1}{\hbar} \langle\langle P; [Q, H^{\text{total}}] \rangle\rangle_z. \end{aligned} \quad (2.35)$$

According to Kubo formula, optical conductivity is written as [89]

$$\sigma_{\alpha\beta}(z) = \frac{i}{\hbar z N} \{ \chi_{\alpha\beta}(z) - \hbar \langle \tau_{\alpha\beta} \rangle \}, \quad (2.36)$$

where $z = \omega + i\eta$ with an infinitesimal constant η and $\chi_{\alpha\beta}(z) = \langle\langle j^{(\alpha)}; j^{(\beta)} \rangle\rangle_z$ is the current-current correlation function with α component of the current operator $j^{(\alpha)}$.

To define the current operator corresponding to the Hamiltonian in Sec. 2.1, an external field is introduced as Peierls phase in creation and annihilation operators;

$$c_{i\mu\sigma}^\dagger \rightarrow e^{-i\frac{e}{\hbar c} \mathbf{A} \cdot \mathbf{r}_i} c_{i\mu\sigma}^\dagger, \quad c_{j\nu\sigma} \rightarrow e^{i\frac{e}{\hbar c} \mathbf{A} \cdot \mathbf{r}_j} c_{j\nu\sigma}, \quad (2.37)$$

where $\mathbf{A}$ is a vector potential. The Hamiltonian is expanded in terms of $\mathbf{A}$ as

$$H_{3d}(\mathbf{A}) = H_{3d}(0) - \frac{1}{c} \mathbf{j} \cdot \mathbf{A} - \frac{1}{2c^2} \mathbf{A} \cdot \tau \mathbf{A} + O(A^3), \quad (2.38)$$

where $\mathbf{j}$ is a current operator and τ is a stress tensor. In the case of the mean-field Hamiltonian, i.e., $H_{3d}(0) = H_{3d}^{\text{MF}}$, the current operator is given by

$$\mathbf{j} = -c \left. \frac{\partial H_{3d}^{\text{MF}}}{\partial \mathbf{A}} \right|_{\mathbf{A}=0} = \sum_{\mathbf{k}_0, \sigma} \sum_{\epsilon, \epsilon'} \mathbf{J}_{\epsilon, \epsilon'}(\mathbf{k}_0, \sigma) \gamma_{\mathbf{k}_0, \epsilon, \sigma}^\dagger \gamma_{\mathbf{k}_0, \epsilon', \sigma} \quad (2.39)$$

with

$$\begin{aligned} \mathbf{J}_{\epsilon, \epsilon'}(\mathbf{k}_0, \sigma) &= \frac{i}{N} \frac{e}{\hbar} \sum_m \sum_{i, j} \sum_{\mu, \nu} \Delta_{i, j} t(\Delta_{i, j}; \mu, \nu) \\ &\quad \times e^{-i(\mathbf{k}_0 + m\mathbf{Q}) \cdot \Delta_{i, j}} \psi_{\mu, m; \epsilon}^*(\mathbf{k}_0, \sigma) \psi_{\nu, m; \epsilon'}(\mathbf{k}_0, \sigma), \end{aligned} \quad (2.40)$$

and the stress tensor is given by

$$\tau_{\alpha\beta} = \sum_{\mathbf{k}_0, \sigma} \sum_{\epsilon, \epsilon'} \tau_{\epsilon, \epsilon'}^{(\alpha, \beta)}(\mathbf{k}_0, \sigma) \gamma_{\mathbf{k}_0, \epsilon, \sigma}^\dagger \gamma_{\mathbf{k}_0, \epsilon', \sigma} \quad (2.41)$$

with

$$\begin{aligned} \tau_{\epsilon, \epsilon'}^{(\alpha, \beta)}(\mathbf{k}_0, \sigma) &= \frac{1}{N} \left(\frac{e}{\hbar} \right)^2 \sum_m \sum_{i,j} \sum_{\mu, \nu} \Delta_{i,j}^{(\alpha)} \Delta_{i,j}^{(\beta)} t(\Delta_{i,j}; \mu, \nu) \\ &\quad \times e^{-i(\mathbf{k}_0 + m\mathbf{Q}) \cdot \Delta_{i,j}} \psi_{\mu, m; \epsilon}^*(\mathbf{k}_0, \sigma) \psi_{\nu, m; \epsilon'}(\mathbf{k}_0, \sigma), \end{aligned} \quad (2.42)$$

where $\Delta_{i,j}^{(\alpha)}$ is the α component of $\Delta_{i,j}$. If we consider the Hamiltonian H_{3d}^{MF} , the correlation function is written as

$$\chi_{\alpha\alpha}(z) = \sum_{\mathbf{k}_0, \sigma} \sum_{\epsilon, \epsilon'} \frac{f(E_{\mathbf{k}_0, \epsilon, \sigma}) - f(E_{\mathbf{k}_0, \epsilon', \sigma})}{(E_{\mathbf{k}_0, \epsilon, \sigma} - E_{\mathbf{k}_0, \epsilon', \sigma})/\hbar + z} \left| J_{\epsilon, \epsilon'}^{(\alpha)}(\mathbf{k}_0, \sigma) \right|^2. \quad (2.43)$$

2.2.2 Memory function and resistivity

In order to take the effect of nonmagnetic impurity into account, we introduce an impurity at site l inducing a local potential I_{imp} that acts equally on all of local orbitals and conserves the spins and the orbitals in the scattering processes. The Hamiltonian is written as

$$\begin{aligned} H' &= I_{\text{imp}} \sum_l \sum_{\sigma} \sum_{\alpha} c_{l\alpha\sigma}^{\dagger} c_{l\alpha\sigma} \\ &= \frac{1}{N} \sum_{\mathbf{k}_0, \mathbf{k}'_0, \sigma} \sum_{\epsilon, \epsilon'} \sum_l I_{\epsilon, \epsilon'}^l(\mathbf{k}_0, \mathbf{k}'_0) \gamma_{\mathbf{k}_0, \epsilon, \sigma}^{\dagger} \gamma_{\mathbf{k}'_0, \epsilon', \sigma} \end{aligned} \quad (2.44)$$

with

$$I_{\epsilon, \epsilon'}^l(\mathbf{k}_0, \mathbf{k}'_0) = I_{\text{imp}} \sum_{\mu, m, m'} e^{-i(\mathbf{k}_0 - \mathbf{k}'_0 + (m - m')\mathbf{Q}) \cdot \mathbf{r}_l} \psi_{\mu, m; \epsilon}^*(\mathbf{k}_0, \sigma) \psi_{\mu, m'; \epsilon'}(\mathbf{k}'_0, \sigma). \quad (2.45)$$

The total Hamiltonian of our system is thus $H = H^{\text{total}} = H_{3d}^{\text{MF}} + H'$.

In order to calculate resistivity, we make use of a memory function. The memory function in a system with impurities represents a relaxation due to the impurities and its imaginary part corresponds to the scattering rate of the system [84]. Therefore, the memory function should vanish if there is no impurity. This condition will be satisfied if the memory function can be expanded in terms of the impurity concentration c , starting from the first order of c [$O(c)$] [84]. Since we are interested in resistivity, we have to consider the expansion in the limit of zero frequency, i.e., $\omega \rightarrow 0$. Based on these considerations, we introduce a memory function for multi-orbital systems, defined by

$$M_{\alpha\beta}(z) \equiv - \frac{iz \text{Im} \chi_{\alpha\beta}(z)}{i \text{Im} \chi_{\alpha\beta}(z) + 2N D_{\alpha\beta}(z)}, \quad (2.46)$$

where

$$D_{\alpha\beta}(z) = \frac{1}{2N} (\text{Re} \chi_{\alpha\beta}(z) - \hbar \langle \tau_{\alpha\beta} \rangle). \quad (2.47)$$

$D_{\alpha\alpha}(z = 0)$ corresponds to charge stiffness or Drude weight. The real part of the optical conductivity is

$$\text{Re} \sigma_{\alpha\beta}(z) = \frac{2D_{\alpha\beta}(z)}{\hbar} \frac{\text{Im} M_{\alpha\beta}(z)}{\{\omega + \text{Re} M_{\alpha\beta}(z)\}^2 + \{\text{Im} M_{\alpha\beta}(z)\}^2}. \quad (2.48)$$

We note that Eq. (2.48) leads to a Drude formula with scattering rate $\text{Im} M_{\alpha\beta}$, if $D_{\alpha\beta}(z)$ is independent of z .

$M_{\alpha\beta}(z)$ given by Eq. (2.46) is slightly different from that for a single-orbital system: $i \text{Im } \chi_{\alpha\beta}$ in the numerator of Eq. (2.46) corresponds to $\text{Re } \chi_{\alpha\beta} + i \text{Im } \chi_{\alpha\beta}$ in Eq. (21) of Ref. [84]. It is important to notice that $\text{Re } \chi_{\alpha\beta}$ remains finite even if $c = 0$. This is due to the presence of interband transition in multi-orbital systems. Therefore, removing $\text{Re } \chi_{\alpha\beta}$ in our definition is necessary for satisfying the condition that the memory function should be expanded starting from $O(c)$.

When $\alpha = \beta$, we obtain from Eq. (2.35) that

$$\begin{aligned} z\chi_{\alpha\alpha}(z) &= \frac{1}{\hbar} \left\langle \left\langle A^{(\alpha)}; j^{(\alpha)} \right\rangle \right\rangle_z \\ &= \frac{1}{\hbar z} \left(\left\langle \left[A^{(\alpha)}, j^{(\alpha)} \right] \right\rangle - \frac{1}{\hbar} \left\langle \left\langle A^{(\alpha)}; A^{(\alpha)} \right\rangle \right\rangle_z \right), \end{aligned} \quad (2.49)$$

where $A^{(\alpha)} = [j^{(\alpha)}, H^{\text{total}}]$. Apparently, $\langle [A^{(\alpha)}, j^{(\alpha)}] \rangle = \frac{1}{\hbar} \left\langle \left\langle A^{(\alpha)}; A^{(\alpha)} \right\rangle \right\rangle_{z=0}$. Thus, we obtain

$$z\chi_{\alpha\alpha}(z) = -\frac{1}{\hbar^2 z} \left\{ \left\langle \left\langle A^{(\alpha)}; A^{(\alpha)} \right\rangle \right\rangle_z - \left\langle \left\langle A^{(\alpha)}; A^{(\alpha)} \right\rangle \right\rangle_{z=0} \right\}. \quad (2.50)$$

The commutator $A^{(\alpha)}$ is explicitly written as

$$A^{(\alpha)} = \sum_{\mathbf{k}_0, \mathbf{k}'_0} \sum_{\sigma} \sum_{\epsilon, \epsilon'} \sum_l A_{\epsilon, \epsilon'}^{(\alpha), l}(\mathbf{k}_0, \mathbf{k}'_0, \sigma) \gamma_{\epsilon}^{\dagger}(\mathbf{k}_0, \sigma) \gamma_{\epsilon'}(\mathbf{k}'_0, \sigma) \quad (2.51)$$

with

$$\begin{aligned} A_{\epsilon, \epsilon'}^{(\alpha), l}(\mathbf{k}_0, \mathbf{k}'_0, \sigma) &= \{E_{\mathbf{k}_0, \epsilon', \sigma} - E_{\mathbf{k}_0, \epsilon, \sigma}\} J_{\epsilon, \epsilon'}^{(\alpha)}(\mathbf{k}_0, \sigma) \delta_{\mathbf{k}_0, \mathbf{k}'_0} \\ &+ \frac{1}{N} \left\{ J_{\epsilon, \epsilon}^{(\alpha)}(\mathbf{k}_0, \sigma) I_{\epsilon', \epsilon'}^l(\mathbf{k}_0, \mathbf{k}'_0) - I_{\epsilon, \epsilon'}^l(\mathbf{k}_0, \mathbf{k}'_0) J_{\epsilon', \epsilon'}^{(\alpha)}(\mathbf{k}'_0, \sigma) \right\}. \end{aligned} \quad (2.52)$$

Note that we ignore the inter-band matrix elements of the current operator taking the view that the elastic scattering is prohibited in potential impurity scattering. Regarding H' as a perturbation, the correlation function is

$$\left\langle \left\langle A^{(\alpha)}; A^{(\alpha)} \right\rangle \right\rangle_z = \sum_{\mathbf{k}_0, \mathbf{k}'_0} \sum_{\sigma} \sum_{\epsilon, \epsilon'} \left| \sum_l A_{\epsilon, \epsilon'}^{(\alpha), l}(\mathbf{k}_0, \mathbf{k}'_0, \sigma) \right|^2 \frac{f(E_{\mathbf{k}'_0, \epsilon', \sigma}) - f(E_{\mathbf{k}_0, \epsilon, \sigma})}{z - (E_{\mathbf{k}'_0, \epsilon', \sigma} - E_{\mathbf{k}_0, \epsilon, \sigma})/\hbar} \quad (2.53)$$

within Born approximation. We assume $z \text{Im } \chi_{\alpha\alpha}(z) = \text{Im } \{z\chi_{\alpha\alpha}(z)\}$ since $z = \omega + i\eta$, where η is an infinitesimal value. This means

$$\begin{aligned} z \text{Im } \chi_{\alpha\alpha}(z) &= -\frac{1}{\hbar^2} \text{Im} \left[\frac{\left\langle \left\langle A^{(\alpha)}; A^{(\alpha)} \right\rangle \right\rangle_z - \left\langle \left\langle A_{\alpha}; A_{\alpha} \right\rangle \right\rangle_0}{z} \right] \\ &= \frac{\pi}{\hbar\omega} \sum_{\mathbf{k}_0, \mathbf{k}'_0} \sum_{\sigma} \sum_{\epsilon, \epsilon'} \left| \sum_l A_{\epsilon, \epsilon'}^{(\alpha), l} \right|^2 \{f(E_{\mathbf{k}'_0, \epsilon', \sigma}) - f(E_{\mathbf{k}_0, \epsilon, \sigma})\} \\ &\quad \times \delta(\hbar\omega - (E_{\mathbf{k}'_0, \epsilon', \sigma} - E_{\mathbf{k}_0, \epsilon, \sigma})/\hbar). \end{aligned} \quad (2.54)$$

In the limit of $\omega \rightarrow 0$, the right-hand side of Eq. (2.54) becomes

$$\pi \sum_{\mathbf{k}_0, \mathbf{k}'_0} \sum_{\sigma} \sum_{\epsilon, \epsilon'} \left| \sum_l A_{\epsilon, \epsilon'}^{(\alpha), l} \right|^2 \delta(E_{\mathbf{k}'_0, \epsilon', \sigma} - E_F) \delta(E_{\mathbf{k}'_0, \epsilon', \sigma} - E_{\mathbf{k}_0, \epsilon, \sigma}). \quad (2.55)$$

Note that $\frac{\partial f(E_{\mathbf{k}_0, \epsilon, \sigma})}{\partial E_{\mathbf{k}_0, \epsilon, \sigma}} = -\delta(E_{\mathbf{k}_0, \epsilon, \sigma} - E_F)$ at absolute zero temperature. The δ -function in Eq. (2.55) implies that the first term in Eq. (2.52) does not contribute to the final result. In the dilute impurity concentration, we can approximate

$$\begin{aligned} \left| \sum_l A_{\epsilon, \epsilon'}^{(\alpha), l} \right|^2 &= \sum_l \left| A_{\epsilon, \epsilon'}^{(\alpha), l} \right|^2 + \sum_{l \neq l'} \left| A_{\epsilon, \epsilon'}^{(\alpha), l} A_{\epsilon, \epsilon'}^{(\alpha), l'} \right| \\ &\simeq N_i \sum_l \left| A_{\epsilon, \epsilon'}^{(\alpha), l=0} \right|^2, \end{aligned} \quad (2.56)$$

where $N_i = cN$ is the number of the impurity sites. The second term is neglected because it is $O(c^2)$.

We find that the contribution from $[j^{(\alpha)}, H_{3d}^{\text{MF}}]$ in Eq. (2.52) vanishes in the limit of $z \rightarrow 0$. This guarantees that, in the limit of $z \rightarrow 0$, $M_{\alpha\alpha}(z)$ given by Eq. (2.46) can be expanded starting from $O(c)$. In the dilute limit of c , i.e., $O(c)$, we obtain from Eq. (2.54)

$$\begin{aligned} M_{\alpha\alpha}(0) &= -\frac{i}{2ND_{\alpha\alpha}(0)} \lim_{z \rightarrow 0} z \text{Im} \chi_{\alpha\alpha}(z) + O(c^2) \\ &\simeq \sum_{\mathbf{k}_0} M_{\alpha\alpha}(\mathbf{k}_0, 0), \end{aligned} \quad (2.57)$$

where

$$\begin{aligned} \text{Im} M_{\alpha\alpha}(\mathbf{k}_0, 0) &= \frac{\pi c}{2D_{\alpha\alpha}(0)} \sum_{\mathbf{k}'_0} \sum_{\epsilon, \epsilon'} \sum_{\sigma} \left| A_{\epsilon, \epsilon'}^{(\alpha), l=0}(\mathbf{k}_0, \mathbf{k}'_0, \sigma) \right|^2 \\ &\quad \times \delta(E_{\mathbf{k}_0, \epsilon, \sigma} - E_F) \delta(E_{\mathbf{k}_0, \epsilon, \sigma} - E_{\mathbf{k}'_0, \epsilon', \sigma}). \end{aligned} \quad (2.58)$$

With use of Eq. (2.57), where $\text{Re} M_{\alpha\alpha}(0) = 0$, the resistivity is written as

$$\rho_{\alpha} = \frac{1}{\text{Re} \sigma_{\alpha\alpha}(0)} = \frac{\hbar \text{Im} M_{\alpha\alpha}(0)}{2D_{\alpha\alpha}(0)}. \quad (2.59)$$

The Drude weight (2.47) is evaluated from Eqs. (2.41) and (2.43).

2.3 Dynamical Susceptibilities and Resonant Inelastic X-ray Scattering Spectra

Dynamical charge and spin susceptibilities represent the response of charge and spin densities from the oscillated external fields. From the linear response theory, the dynamical charge and spin susceptibilities are related to the correlation functions of charge density and spin operators, respectively. In conventional way, charge susceptibility is obtained from measurements of permittivity, and spin susceptibility is done from the spectra of neutron scattering. RIXS is considered as a new tool for detecting these excitations. The RIXS spectrum is written as the superposition of the dynamical spin and charge susceptibilities; investigating the dynamical susceptibilities through the RIXS spectrum allows us to know the electronic dynamics of the system both in the charge and the spin channels.

In this section, we derive the dynamical susceptibilities within RPA, and RIXS spectra within fast-collision approximation.

2.3.1 Dynamical charge and spin susceptibilities

An external electric field $\mathbf{E}^{\text{ex}}$ and an external magnetic field $\mathbf{H}^{\text{ex}}$ couple to the degree of freedom on charge and spin, respectively. The deformation of Hamiltonian is

$$\mathcal{H}^{\text{ex}}(t') = \sum_j \{n_j(t') |\mathbf{E}^{\text{ex}}(\mathbf{r}'_j, t')| + \mathbf{s}_j(t') \cdot \mathbf{H}^{\text{ex}}(\mathbf{r}'_j, t')\}, \quad (2.60)$$

where $n_j(t')$ and $\mathbf{s}_j(t')$ are charge density operator and spin operator at site j (position $\mathbf{r}_j$) and time t' , respectively. The charge and spin operators are defined as

$$n_i = n_{i,\uparrow} + n_{i,\downarrow} \quad (2.61)$$

and

$$s_{i,x} = \frac{1}{2} (s_{i,+} + s_{i,-}), \quad s_{i,y} = \frac{1}{2i} (s_{i,+} - s_{i,-}), \quad s_{i,z} = \frac{1}{2} (n_{i,\uparrow} - n_{i,\downarrow}), \quad (2.62)$$

respectively, where

$$n_{i,\sigma} = \sum_{\mu} c_{i,\mu,\sigma}^{\dagger} c_{i,\mu,\sigma}, \quad s_{i,+} = c_{i,\mu,\uparrow}^{\dagger} c_{i,\mu,\downarrow}, \quad s_{i,-} = c_{i,\mu,\downarrow}^{\dagger} c_{i,\mu,\uparrow}. \quad (2.63)$$

According to the linear response theory, the deviation of charge density and spin from their average value by external field is [88]

$$\begin{pmatrix} \delta \langle n_i(t) \rangle_{\text{ex}} \\ \delta \langle s_{i,x}(t) \rangle_{\text{ex}} \\ \delta \langle s_{i,y}(t) \rangle_{\text{ex}} \\ \delta \langle s_{i,z}(t) \rangle_{\text{ex}} \end{pmatrix} = \int_{-\infty}^{\infty} dt' \sum_j \frac{1}{i\hbar} \left\langle \begin{pmatrix} [n, n] & [n, s_x] & [n, s_y] & [n, s_z] \\ [s_x, n] & [s_x, s_x] & [s_x, s_y] & [s_x, s_z] \\ [s_y, n] & [s_y, s_x] & [s_y, s_y] & [s_y, s_z] \\ [s_z, n] & [s_z, s_x] & [s_z, s_y] & [s_z, s_z] \end{pmatrix} \right\rangle \begin{pmatrix} E^{\text{ex}}(\mathbf{r}_j, t') \\ H_x^{\text{ex}}(\mathbf{r}_j, t') \\ H_y^{\text{ex}}(\mathbf{r}_j, t') \\ H_z^{\text{ex}}(\mathbf{r}_j, t') \end{pmatrix}, \quad (2.64)$$

where we omit i and t from the left operators in the commutators and j and t' from the right ones.

The retarded correlation function is defined as

$$D_{\kappa \nu}^{Rss'}(\mathbf{r}_i, t; \mathbf{r}_j, t') = -i \left\langle \left[c_{i,\kappa,\sigma_1}^{\dagger}(t) c_{i,\lambda,\sigma_2}(t), c_{j,\mu,\sigma_3}^{\dagger}(t') c_{j,\nu,\sigma_4}(t') \right] \right\rangle \theta(t - t'), \quad (2.65)$$

where s (s') denotes a spin pair $\sigma_1\sigma_2$ ($\sigma_3\sigma_4$): s takes $\uparrow, \downarrow, +$, and $-$ corresponding to $\uparrow\uparrow, \downarrow\downarrow, \uparrow\downarrow$, and $\downarrow\uparrow$, respectively. Corresponding temperature correlation function depending on an imaginary time variables τ is

$$\mathcal{D}_{\kappa \nu}^{ss'}(\mathbf{r}_i, \tau; \mathbf{r}_j, \tau') = - \left\langle T_{\tau} c_{i,\kappa,\sigma_1}^{\dagger}(\tau) c_{i,\lambda,\sigma_2}(\tau) c_{j,\mu,\sigma_3}^{\dagger}(\tau') c_{j,\nu,\sigma_4}(\tau') \right\rangle, \quad (2.66)$$

where the symbol T_{τ} orders the operators according to their values of τ with the smallest at the right. T_{τ} also includes the signature factor $(-1)^P$, where P is the number of permutations of fermion operators.

2.3.2 Random phase approximation

Susceptibility is defined as

$$\chi_{\kappa\mu\nu}^{ss'}(\mathbf{r}_i, t; \mathbf{r}_j, t') = -\frac{1}{\hbar} D_{\kappa\mu\nu}^{Rss'}(\mathbf{r}_i, t; \mathbf{r}_j, t'). \quad (2.67)$$

We abbreviate $\chi(\mathbf{r}_i, t; \mathbf{r}_j, t')$ as $\chi(\mathbf{r}_i, \mathbf{r}_j, t - t')$ since it depends on $t - t'$. Its Fourier coefficient is

$$\begin{aligned} \chi_{\kappa\mu\nu}^{ss'}(\mathbf{q}, \mathbf{q}', \omega) &= \frac{1}{N} \sum_{i,j} \int_{-\infty}^{\infty} dt e^{-i\mathbf{q}\cdot\mathbf{r}_i + i\mathbf{q}'\cdot\mathbf{r}_j} e^{i\omega t} \chi_{\kappa\mu\nu}^{ss'}(\mathbf{r}_i, \mathbf{r}_j, t) \\ &= \frac{i}{N\hbar} \sum_{\mathbf{k}', \mathbf{k}''} \int_0^{\infty} dt e^{i\omega t} \left\langle \left[c_{\mathbf{k}', \kappa, \sigma_1}^\dagger(t) c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2}(t), c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} \right] \right\rangle \end{aligned} \quad (2.68)$$

The Fourier component of transverse spin susceptibility $\chi^{+-}(\mathbf{r}_i, \mathbf{r}_j, t) = \langle [s_{i,+}(t), s_{j,-}] \rangle / \hbar$, longitudinal spin susceptibility $\chi^z(\mathbf{r}_i, \mathbf{r}_j, t) = \langle [s_{i,z}(t), s_{j,z}] \rangle / \hbar$, and charge susceptibility $\chi^n(\mathbf{r}_i, \mathbf{r}_j, t) = \langle [n_i(t), n_j] \rangle / \hbar$ are written as

$$\chi^{+-}(\mathbf{q}, \omega) = \sum_{\kappa, \mu} \chi_{\kappa\mu}^{+-}(\mathbf{q}, \mathbf{q}, \omega) \quad (2.69)$$

$$\chi^z(\mathbf{q}, \omega) = \frac{1}{4} \sum_{\kappa, \mu} \sum_{\sigma=\uparrow, \downarrow} \left\{ \chi_{\kappa\mu}^{ss}(\mathbf{q}, \mathbf{q}, \omega) - \chi_{\kappa\mu}^{s\bar{s}}(\mathbf{q}, \mathbf{q}, \omega) \right\} \quad (2.70)$$

and

$$\chi^n(\mathbf{q}, \omega) = \frac{1}{4} \sum_{\kappa, \mu} \sum_{\sigma=\uparrow, \downarrow} \left\{ \chi_{\kappa\mu}^{ss}(\mathbf{q}, \mathbf{q}, \omega) + \chi_{\kappa\mu}^{s\bar{s}}(\mathbf{q}, \mathbf{q}, \omega) \right\}, \quad (2.71)$$

respectively, where $\bar{s}$ denotes $\downarrow$ ($\uparrow$) for $s = \uparrow$ ($\downarrow$). Here, we consider only $\mathbf{q} = \mathbf{q}'$ term. This is because $\mathbf{q}$ and $\mathbf{q}'$ in Eq. (2.68) are restricted to the form $\mathbf{k} + l\mathbf{Q}$ and $\mathbf{k} + l'\mathbf{Q}$, respectively, where l and l' are integer.

According to perturbation theory, if Hamiltonian is written as $H = H_{3d}^{\text{MF}} + H_{\text{int}}$, the average value of an operator $A(\tau)$ written as Heisenberg representation is

$$\langle A(\tau) \rangle = \sum_{n=0}^{\infty} \left(-\frac{1}{\hbar} \right)^n \frac{1}{n!} \int_0^{\beta\hbar} d\tau_1 \cdots \int_0^{\beta\hbar} d\tau_n \langle T_\tau \{ H_{\text{int}}(\tau_1) \cdots H_{\text{int}}(\tau_n) A(\tau) \} \rangle_c, \quad (2.72)$$

where the symbol c at the right parentheses means that only connected Feynman diagrams remain in the decomposition of Wick's theorem and averages are taken with non-perturbation Hamiltonian H_{3d}^{MF} . Note that the average on left hand side is taken with total Hamiltonian H . Though, of course, H_{3d}^{MF} contains the interaction Hamiltonian in mean-field approximation level, we again add the interaction Hamiltonian to treat the electron correlation in the SDW state.

Within RPA, the dynamical susceptibilities are connected to bare susceptibilities χ_0 through the relation

$$\begin{pmatrix} \chi^{+-} \\ \chi^{\uparrow\uparrow} \\ \chi^{\downarrow\uparrow} \end{pmatrix} = \begin{pmatrix} \chi_0^{+-} \\ \chi_0^{\uparrow\uparrow} \\ 0 \end{pmatrix} + \begin{pmatrix} \chi_0^{+-} V^{-+} & 0 & 0 \\ 0 & \chi_0^{\uparrow\uparrow} V^{\uparrow\uparrow} & \chi_0^{\uparrow\uparrow} V^{\uparrow\downarrow} \\ 0 & \chi_0^{\downarrow\uparrow} V^{\downarrow\uparrow} & \chi_0^{\downarrow\uparrow} V^{\downarrow\downarrow} \end{pmatrix} \begin{pmatrix} \chi^{+-} \\ \chi^{\uparrow\uparrow} \\ \chi^{\downarrow\uparrow} \end{pmatrix}. \quad (2.73)$$

Table. 2.1 The non-zero elements of $V^{ss'}$, where $s = (\sigma_1, \sigma_2)$ and $s' = (\sigma'_1, \sigma'_2)$. Each of the four subscripts takes one of the $3d$ orbitals.

	$\sigma_1 = \sigma_2 = \sigma'_1 = \sigma'_2$	$\sigma_1 = \sigma_2 \neq \sigma'_1 = \sigma'_2$	$\sigma_1 = \sigma'_2 \neq \sigma_2 = \sigma'_1$
$\mu = \nu = \kappa = \lambda$	–	$-U$	U
$\mu = \kappa \neq \nu = \lambda$	–	$-J$	J
$\mu = \nu \neq \kappa = \lambda$	$-U + 3J$	$-U + 2J$	J
$\mu = \lambda \neq \nu = \kappa$	$U - 3J$	$-J$	$U - 2J$

The product of susceptibility χ and interaction V is taken as a matrix product represented in the orbital basis such as

$$[\chi_0^{ss_1} V^{s_1 s_2} \chi^{s_2 s'}]_{\lambda \mu \kappa \nu}(\mathbf{r}_i, \mathbf{r}_j, t) = \sum_{\kappa', \lambda', \mu', \nu', l} \chi_{\lambda \mu \kappa \nu'}^{ss_1}(\mathbf{r}_i, \mathbf{r}_l, t) V_{\mu' \lambda' \kappa' \nu}^{s_1 s_2} \chi_{\lambda' \mu \kappa' \nu}^{s_2 s'}(\mathbf{r}_l, \mathbf{r}_j, t), \quad (2.74)$$

and their Fourier components are

$$\begin{aligned} & [\chi_0^{ss_1} V^{s_1 s_2} \chi^{s_2 s'}]_{\lambda \mu \kappa \nu}(\mathbf{q}, \mathbf{q}, \omega) \\ &= \sum_{\kappa', \lambda', \mu', \nu', m} \chi_{\lambda \mu \kappa \nu'}^{ss_1}(\mathbf{q}, \mathbf{q} + m\mathbf{Q}, \omega) V_{\mu' \lambda' \kappa' \nu}^{s_1 s_2} \chi_{\lambda' \mu \kappa' \nu}^{s_2 s'}(\mathbf{q} + m\mathbf{Q}, \mathbf{q}, \omega). \end{aligned} \quad (2.75)$$

The nonzero elements of the interaction matrix $V_{\mu \lambda \nu \kappa}^{ss'}$ are shown in Table 2.1.

2.3.3 Bare susceptibility

The non-perturbation temperature correlation function $\mathcal{D}_0$ is

$$\begin{aligned} \mathcal{D}_{0 \lambda \mu \kappa \nu}^{ss'}(\mathbf{r}_i, \tau; \mathbf{r}_j, \tau') &= -\left\langle T_\tau c_{i, \kappa, \sigma_1}^\dagger(\tau) c_{i, \lambda, \sigma_2}(\tau) c_{j, \mu, \sigma_3}^\dagger(\tau') c_{j, \nu, \sigma_4}(\tau') \right\rangle_0 \\ &= \left\langle T_\tau \left[c_{i, \lambda, \sigma_2}(\tau) c_{j, \mu, \sigma_3}^\dagger(\tau') \right] \right\rangle_0 \left\langle T_\tau \left[c_{j, \nu, \sigma_4}(\tau') c_{i, \kappa, \sigma_1}^\dagger(\tau) \right] \right\rangle_0. \end{aligned} \quad (2.76)$$

We neglect the same-time correlation function in Eq. (2.76). We set $\sigma_2 = \sigma_3 = \sigma$ and $\sigma_1 = \sigma_4 = \sigma'$. Since $\mathcal{D}_0(\mathbf{r}_i, \tau; \mathbf{r}_j, \tau')$ depends on $\tau - \tau'$, we set $\tau - \tau'$ as τ , and

$$\mathcal{D}_{0 \lambda \mu \kappa \nu}^{ss'}(\mathbf{r}_i, \mathbf{r}_j, \tau) = \mathcal{G}_{\lambda, \mu, \sigma}(\mathbf{r}_i, \mathbf{r}_j, \tau) \mathcal{G}_{\nu, \kappa, \sigma'}(\mathbf{r}_j, \mathbf{r}_i, -\tau). \quad (2.77)$$

Its Fourier component is

$$\begin{aligned} \mathcal{D}_{0 \lambda \mu \kappa \nu}^{ss'}(\mathbf{k}' + \frac{\mathbf{q}}{2}, \mathbf{k}' - \frac{\mathbf{q}}{2}, i\Omega_n) &= \sum_{\mathbf{r}_i, \mathbf{r}_j} \int_0^{\beta\hbar} d\tau e^{-i(\mathbf{k}' + \frac{\mathbf{q}}{2}) \cdot \mathbf{r}_i + i(\mathbf{k}' - \frac{\mathbf{q}}{2}) \cdot \mathbf{r}_j} e^{i\Omega_n \tau} \mathcal{D}_{0 \lambda \mu \kappa \nu}^{ss'}(\mathbf{r}_i, \mathbf{r}_j, \tau) \\ &= \frac{1}{N\beta\hbar} \sum_{\mathbf{k}_2, \mathbf{q}_2} \sum_{\omega_n} \mathcal{G}_{\lambda, \mu, \sigma}(\mathbf{k}' + \mathbf{k}_2 + \frac{\mathbf{q} - \mathbf{q}_2}{2}, \mathbf{k}' + \mathbf{k}_2 - \frac{\mathbf{q} - \mathbf{q}_2}{2}, i\omega_n + i\Omega_n) \\ &\quad \times \mathcal{G}_{\nu, \kappa, \sigma'}(\mathbf{k}_2 + \frac{\mathbf{q}_2}{2}, \mathbf{k}_2 - \frac{\mathbf{q}_2}{2}, i\omega_n), \end{aligned} \quad (2.78)$$

where $\omega_n = (2n+1)\pi/\beta\hbar$ and $\Omega_n = 2n\pi/\beta\hbar$. The Fourier component of temperature Green's function (2.33) is written as

$$\mathcal{G}_{\lambda, \mu, \sigma}(\mathbf{r}_i, \mathbf{r}_j, \tau) = \frac{1}{N\beta\hbar} \sum_{\mathbf{k}, \mathbf{q}} \sum_{\omega_n} e^{i(\mathbf{k} + \frac{\mathbf{q}}{2}) \cdot \mathbf{r}_i - i(\mathbf{k} - \frac{\mathbf{q}}{2}) \cdot \mathbf{r}_j} e^{-i\omega_n \tau} \mathcal{G}_{\lambda, \mu, \sigma}(\mathbf{k} + \frac{\mathbf{q}}{2}, \mathbf{k} - \frac{\mathbf{q}}{2}, i\omega_n). \quad (2.79)$$

and the bare susceptibility is defined as

$$\chi_{0\lambda\mu\kappa\nu}^{ss'}(\mathbf{k}' + \frac{\mathbf{q}}{2}, \mathbf{k}' - \frac{\mathbf{q}}{2}, i\Omega_n) = -\frac{1}{\hbar} \mathcal{D}_{0\lambda\mu\kappa\nu}^{ss'}(\mathbf{k}' + \frac{\mathbf{q}}{2}, \mathbf{k}' - \frac{\mathbf{q}}{2}, i\Omega_n). \quad (2.80)$$

We set

$$\mathbf{k}' = \mathbf{k} + \frac{l_1 + l_2}{2}\mathbf{Q}, \quad \mathbf{q} = (l_1 - l_2)\mathbf{Q}, \quad \mathbf{k}_2 = \mathbf{p} + \frac{n}{2}\mathbf{Q}, \quad \mathbf{q}_2 = n\mathbf{Q} \quad (2.81)$$

$$\sum_{\mathbf{k}_2} \rightarrow \sum_{\mathbf{p}}, \quad \sum_{\mathbf{q}_2} \rightarrow \sum_n, \quad (2.82)$$

where l_1 and l_2 are integer. With use of Eqs. (2.33) and (2.79), we obtain

$$\begin{aligned} & \chi_{0\lambda\mu\kappa\nu}^{ss'}(\mathbf{k} + l_1\mathbf{Q}, \mathbf{k} + l_2\mathbf{Q}, i\Omega_n) \\ &= -\frac{1}{N\beta} \sum_{\omega_n} \sum_{\mathbf{p}, n} \sum_{\mathbf{k}_0, \epsilon} \sum_{m_1, m_2} \frac{\psi_{\lambda, m_1; \epsilon}(\mathbf{k}_0, \sigma) \psi_{\mu, m_2; \epsilon}^*(\mathbf{k}_0, \sigma)}{i\hbar\omega_n + i\hbar\Omega_n - E_{\mathbf{k}_0, \epsilon, \sigma}} \\ & \quad \times \delta_{\mathbf{k}_0 + m_1\mathbf{Q}, \mathbf{k} + \mathbf{p} + l_1\mathbf{Q}} \delta_{\mathbf{k}_0 + m_2\mathbf{Q}, \mathbf{k} + \mathbf{p} + (l_2 + n)\mathbf{Q}} \\ & \quad \times \sum_{\mathbf{k}'_0, \epsilon'} \sum_{m'_1, m'_2} \frac{\psi_{\nu, m'_1; \epsilon'}(\mathbf{k}'_0, \sigma') \psi_{\kappa, m'_2; \epsilon'}^*(\mathbf{k}'_0, \sigma')}{i\hbar\omega_n - E_{\mathbf{k}'_0, \epsilon', \sigma'}} \delta_{\mathbf{k}'_0 + m'_1\mathbf{Q}, \mathbf{p} + n\mathbf{Q}} \delta_{\mathbf{k}'_0 + m'_2\mathbf{Q}, \mathbf{p}}. \end{aligned} \quad (2.83)$$

Figure 2.1 shows the diagram of the bare susceptibility (2.83). The summation over Matsubara frequency is

$$\frac{1}{\beta} \sum_{\omega_n} \frac{1}{i\hbar\omega_n + i\hbar\Omega_n - E} \cdot \frac{1}{i\hbar\omega_n - E'} = \frac{f(E) - f(E')}{E - E' - i\hbar\Omega_n}. \quad (2.84)$$

We set

$$\mathbf{p} = \mathbf{p}_0 + m\mathbf{Q}, \quad \sum_{\mathbf{p}} = \sum_{\mathbf{p}_0} \sum_m \quad (2.85)$$

and obtain

$$\begin{aligned} & \chi_{0\lambda\mu\kappa\nu}^{ss'}(\mathbf{k} + l_1\mathbf{Q}, \mathbf{k} + l_2\mathbf{Q}, i\Omega_n) \\ &= -\frac{1}{N_k} \sum_{\mathbf{p}_0} \sum_{m, n} \sum_{\mathbf{k}_0, \epsilon} \sum_{m_1, m_2} \sum_{\mathbf{k}'_0, \epsilon'} \sum_{m'_1, m'_2} \frac{f(E_{\mathbf{k}_0, \epsilon, \sigma}) - f(E_{\mathbf{k}'_0, \epsilon', \sigma'})}{E_{\mathbf{k}_0, \epsilon, \sigma} - E_{\mathbf{k}'_0, \epsilon', \sigma'} - i\hbar\Omega_n} \\ & \quad \times \psi_{\lambda, m_1; \epsilon}(\mathbf{k}_0, \sigma) \psi_{\mu, m_2; \epsilon}^*(\mathbf{k}_0, \sigma) \delta_{\mathbf{k}_0 + m_1\mathbf{Q}, \mathbf{k} + \mathbf{p}_0 + (m + l_1)\mathbf{Q}} \delta_{\mathbf{k}_0 + m_2\mathbf{Q}, \mathbf{k} + \mathbf{p}_0 + (m + n + l_2)\mathbf{Q}} \\ & \quad \times \psi_{\nu, m'_1; \epsilon'}(\mathbf{k}'_0, \sigma') \psi_{\kappa, m'_2; \epsilon'}^*(\mathbf{k}'_0, \sigma') \delta_{\mathbf{k}'_0 + m'_1\mathbf{Q}, \mathbf{p}_0 + (m + n)\mathbf{Q}} \delta_{\mathbf{k}'_0 + m'_2\mathbf{Q}, \mathbf{p}_0 + m\mathbf{Q}}. \end{aligned} \quad (2.86)$$

This equation implies that

$$\mathbf{k}_0 = \mathbf{k} + \mathbf{p}_0, \quad m_1 = m + l_1, \quad m_2 = m + n + l_2 \quad (2.87)$$

$$\mathbf{k}'_0 = \mathbf{p}_0, \quad m'_1 = m + n, \quad m'_2 = m. \quad (2.88)$$

As a result, we obtain the representation of the bare susceptibility as

$$\begin{aligned} & \chi_{0\lambda\mu\kappa\nu}^{ss'}(\mathbf{k} + l_1\mathbf{Q}, \mathbf{k} + l_2\mathbf{Q}, i\Omega_n \rightarrow \omega + i\eta) \\ &= -\frac{1}{N} \sum_{\mathbf{p}_0} \sum_{m, n} \sum_{\epsilon, \epsilon'} \frac{f(E_{\mathbf{p}_0 + \mathbf{k}, \epsilon, \sigma}) - f(E_{\mathbf{p}_0, \epsilon', \sigma'})}{E_{\mathbf{p}_0 + \mathbf{k}, \epsilon, \sigma} - E_{\mathbf{p}_0, \epsilon', \sigma'} - \hbar(\omega + i\eta)} \\ & \quad \times \psi_{\lambda, m + l_1; \epsilon}(\mathbf{p}_0 + \mathbf{k}, \sigma) \psi_{\mu, m + n + l_2; \epsilon}^*(\mathbf{p}_0 + \mathbf{k}, \sigma) \times \psi_{\nu, m + n; \epsilon'}(\mathbf{p}_0, \sigma') \psi_{\kappa, m; \epsilon'}^*(\mathbf{p}_0, \sigma'). \end{aligned} \quad (2.89)$$

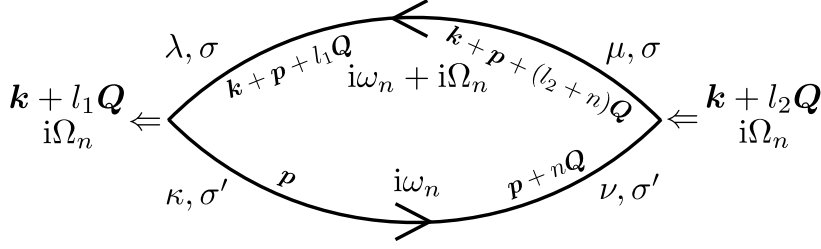


Fig. 2.1 The diagram of bare susceptibility in Eq. (2.83). The solid lines of the bubble indicate temperature Green's functions of single electron. The upper line is a particle line with frequency $i\omega_n + i\Omega_n$ and spin σ , and lower line is a hole line with frequency $i\omega_n$ and spin σ' . Intrinsic degrees of freedom $\mathbf{p}$, $n\mathbf{Q}$, and $i\omega_n$ are summed out.

2.3.4 Dynamical susceptibility in the ground state

We define the eigenstate $|m\rangle$ of Hamiltonian H with eigenvalue E_m , which forms a complete set $1 = \sum_m |m\rangle\langle m|$. The statistical average is written as $\langle \dots \rangle = \frac{1}{Z} \sum_m e^{-\beta E_m} \langle m | \dots | m \rangle$, where $Z = \sum_m e^{-\beta E_m}$ is a distribution function. The dynamical susceptibility (2.68) becomes

$$\begin{aligned} \chi_{\kappa \nu}^{ss'}(\mathbf{q}, \mathbf{q}', \omega) &= \frac{i}{N\hbar} \sum_{\mathbf{k}', \mathbf{k}''} \int_0^\infty dt e^{i\omega t - \eta t} \\ &\times \frac{1}{Z} \left\{ \sum_{m,n} e^{-\beta E_n} \langle n | c_{\mathbf{k}', \kappa, \sigma_1}^\dagger(t) c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2}(t) | m \rangle \langle m | c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} | n \rangle \right. \\ &\left. - \sum_{m,n} e^{-\beta E_m} \langle m | c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} | n \rangle \langle n | c_{\mathbf{k}', \kappa, \sigma_1}^\dagger(t) c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2}(t) | m \rangle \right\}, \quad (2.90) \end{aligned}$$

where η is a convergence factor. With use of the Heisenberg representation $c_{\mathbf{k}, \mu, \sigma}(t) = e^{iHt/\hbar} c_{\mathbf{k}, \mu, \sigma} e^{-iHt/\hbar}$, we obtain

$$\begin{aligned} \chi_{\kappa \nu}^{ss'}(\mathbf{q}, \mathbf{q}', \omega) &= -\frac{1}{N} \sum_{\mathbf{k}', \mathbf{k}''} \sum_{m,n} \frac{1}{\hbar\omega + E_n - E_m + i\hbar\eta} \frac{1}{Z} e^{-\beta E_n} (1 - e^{\beta(E_n - E_m)}) \\ &\times \langle n | c_{\mathbf{k}', \kappa, \sigma_1}^\dagger c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2} | m \rangle \langle m | c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} | n \rangle. \quad (2.91) \end{aligned}$$

At low temperature limit, it is enough for $e^{-\beta E_n}$ to consider only the ground state energy $E_n = E_g$. In this limit, the distribution function $Z = e^{-\beta E_g}$. Thus

$$\begin{aligned} \lim_{T \rightarrow 0} \chi_{\kappa \nu}^{ss'}(\mathbf{q}, \mathbf{q}', \omega) &= -\frac{1}{N} \sum_{\mathbf{k}', \mathbf{k}''} \sum_m \frac{1}{\hbar\omega + E_g - E_m + i\hbar\eta} (1 - e^{\beta(E_g - E_m)}) \\ &\times \langle g | c_{\mathbf{k}', \kappa, \sigma_1}^\dagger c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2} | m \rangle \langle m | c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} | g \rangle. \quad (2.92) \end{aligned}$$

With use of $\lim_{T \rightarrow 0} e^{\beta(E_g - E_m)} = \delta_{m,g}$,

$$\lim_{T \rightarrow 0} \chi_{\kappa \nu}^{ss'}(\mathbf{q}, \mathbf{q}', \omega)$$

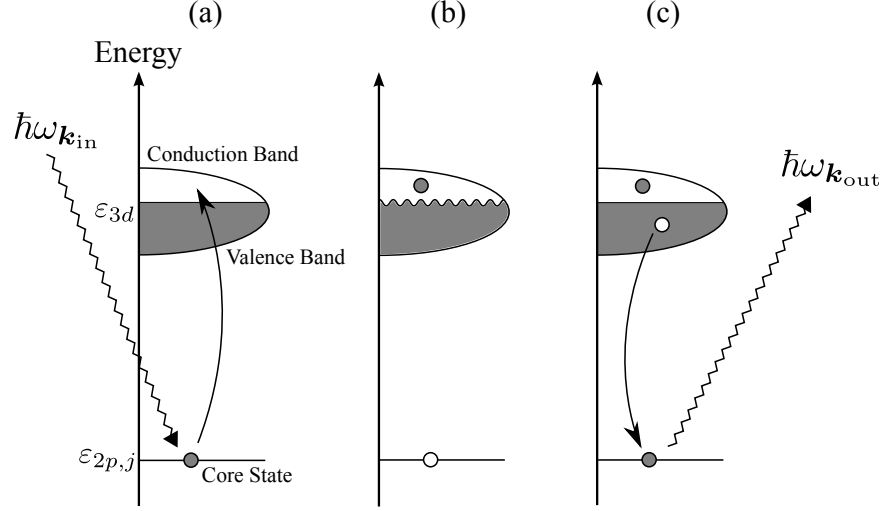


Fig. 2.2 The schematically illustrated process of RIXS. (a) A photon with energy $\hbar\omega_{\mathbf{k}_{in}}$ excites an electron at the core state with energy $\varepsilon_{2p,j}$ to conduction band. ε_{3d} is the average energy of conduction and valence band of $3d$ and $4s$ orbitals. (b) The excited electron interacts with the electrons in valence band. In fast-collision approximation, this process is neglected due to the very short time in this process compared to the relaxation time. (c) The electron in valence band falls into the core-hole state, and the photon is emitted with energy $\hbar\omega_{\mathbf{k}_{out}}$.

$$\begin{aligned}
&= -\frac{1}{N} \sum_{\mathbf{k}', \mathbf{k}''} \langle g | c_{\mathbf{k}', \kappa, \sigma_1}^\dagger c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2} \frac{1}{\hbar\omega + E_g - H + i\hbar\eta} c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} | g \rangle \\
&\quad + \frac{1}{N} \frac{1}{\hbar\omega + i\hbar\eta} \sum_{\mathbf{k}', \mathbf{k}''} \langle g | c_{\mathbf{k}', \kappa, \sigma_1}^\dagger c_{\mathbf{k}'+\mathbf{q}, \lambda, \sigma_2} | g \rangle \langle g | c_{\mathbf{k}''+\mathbf{q}', \mu, \sigma_3}^\dagger c_{\mathbf{k}'', \nu, \sigma_4} | g \rangle. \quad (2.93)
\end{aligned}$$

The second term in Eq. (2.93) is omitted at finite ω .

2.4 Resonant Inelastic X-ray Scattering

The L -edge RIXS in $3d$ transition-metal compounds consists of two processes: The x-ray absorption and emission by way of an intermediate state with core holes, as schematically illustrated in Fig. 2.2. The x-ray absorption in Cr is accompanied by the creation of a $2p$ -core hole and an electron in $3d$ and $4s$ orbitals. The $2p$ -core-hole state has a hybridized spin with the orbital angular momentum due to spin-orbit coupling. When the electron relaxed through the x-ray emission is not the excited electron from the $2p$ -core state, the resulting orbital occupation of $3d$ and $4s$ electrons varies from the initial state. Note that the spin of the relaxed electron may be either up or down, since the spin of the core hole is hybridized. The final state may thus involves a spin flip. This fact means that we can investigate spin excitations in addition to charge and orbital ones through the L -edge RIXS.

Many review articles on RIXS have been published until now. The RIXS study in early days are summarized by Kotani and Shin [90] and Kotani [91]. The recent progress on RIXS is comprehensively reviewed by Ament *et al.* [92]. The review focusing on high-temperature superconductors are given by Ishii *et al.* [81] and Dean [93].

2.4.1 Kramers-Heisenberg equation

Under the electromagnetic field, the kinetic term of Hamiltonian is written as

$$H = \sum_{j=1}^{N_e} \frac{1}{2m} \left(\mathbf{p}_j - \frac{e}{c} \mathbf{A}(\mathbf{r}_j) \right)^2 = H_0 + H' \quad (2.94)$$

$$H_0 = \sum_{j=1}^{N_e} \frac{\mathbf{p}_j^2}{2m}, \quad H' = \sum_{j=1}^{N_e} \left(-\frac{e}{mc} \mathbf{A}(\mathbf{r}_j) \cdot \mathbf{p}_j + \frac{e^2}{2mc^2} \mathbf{A}(\mathbf{r}_j)^2 \right) \quad (2.95)$$

where the summation j runs over all N_e electrons of the system, $\mathbf{p}_j$ is the momentum operator of the electron, and $\mathbf{A}(\mathbf{r}_j)$ is the vector potential at j -th electron's position $\mathbf{r}_j$. Hereafter, we neglect the second order of the vector potential in H' , and we use $\mathbf{p} = -i\hbar\nabla$ and the Coulomb gauge $\nabla \cdot \mathbf{A} = 0$. In this Hamiltonian (2.94), we ignore a Zeeman splitting term and a spin-orbit interaction term since they can be assumed very small compared to the kinetic term [92]. The vector potential can be written with second quantization form as

$$\mathbf{A}(\mathbf{r}) = \sum_{\mathbf{k}, \epsilon} \sqrt{\frac{2\pi c^2 \hbar}{2\mathcal{V} \epsilon_0 \omega_{\mathbf{k}}}} \left(\epsilon a_{\mathbf{k}, \epsilon} e^{i\mathbf{k} \cdot \mathbf{r}} + \epsilon^* a_{\mathbf{k}, \epsilon}^\dagger e^{-i\mathbf{k} \cdot \mathbf{r}} \right) \quad (2.96)$$

where $\mathcal{V}$ is the volume of the system, $a_{\mathbf{k}, \epsilon}$ ($a_{\mathbf{k}, \epsilon}^\dagger$) is an annihilation (creation) operator of the photon with wave vector $\mathbf{k}$ and polarization ϵ .

In RIXS process, an incident photon with wave vector $\mathbf{k}_{\text{in}}$ and polarization ϵ_{in} goes into the initial ground state with energy E_g , and the core-electron excites to conduction band, which makes a core-hole state and has the energy E_n . After some interaction with electrons in valence band and excited electron during a few period, an electron in valence band falls off to the core-hole state, emitting the photon with wave vector $\mathbf{k}_{\text{out}}$ and polarization ϵ_{out} . We define the initial state, intermediate state, and final state of this Hamiltonian in RIXS process as $|g; \mathbf{k}_{\text{in}}, \epsilon_{\text{in}}\rangle$, $|n\rangle$, and $|f; \mathbf{k}_{\text{out}}, \epsilon_{\text{out}}\rangle$, respectively. The corresponding energies are $E_g + \hbar\omega_{\mathbf{k}_{\text{in}}}$, E_n , and $E_f + \hbar\omega_{\mathbf{k}_{\text{out}}}$, respectively, where $\omega_{\mathbf{k}} = c|\mathbf{k}|$ is the energy dispersion relation of the photon. Fermi's golden rule to the second order gives the transition rate w of this process as

$$w = \frac{2\pi}{\hbar} \delta(E_g - E_f + \omega) \sum_f \left| \langle f | H' | g \rangle + \sum_n \frac{\langle f | H' | n \rangle \langle n | H' | g \rangle}{(E_g + \hbar\omega_{\mathbf{k}_{\text{in}}}) - E_n} \right|, \quad (2.97)$$

where $\omega = \omega_{\mathbf{k}_{\text{in}}} - \omega_{\mathbf{k}_{\text{out}}}$. Since we are focusing on the resonant x-ray scattering where $E_g + \hbar\omega_{\mathbf{k}_{\text{in}}} \simeq E_n$, the second term in Eq. (2.97) is dominant. The second term is called resonant term. Note that the first term is dominant in non-resonant x-ray scattering spectroscopy.

In the first order of vector potential magnitude, the resonant term becomes

$$\begin{aligned} & \sum_n \frac{\langle f | H' | n \rangle \langle n | H' | g \rangle}{(E_g + \hbar\omega_{\mathbf{k}_{\text{in}}}) - E_n} \\ & \simeq \frac{e^2 \hbar}{2m^2 \mathcal{V} \epsilon_0 \sqrt{\omega_{\mathbf{k}_{\text{in}}} \omega_{\mathbf{k}_{\text{out}}}}} \sum_n \sum_{i,j=1}^{N_e} \frac{\langle f | e^{-i\mathbf{k}_{\text{out}} \cdot \mathbf{r}_i} \epsilon_{\text{out}}^* \cdot \mathbf{p}_i | n \rangle \langle n | e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}_j} \epsilon_{\text{in}} \cdot \mathbf{p}_j | g \rangle}{E_g + \hbar\omega_{\mathbf{k}_{\text{in}}} - E_n + i\Gamma_n}, \end{aligned} \quad (2.98)$$

where Γ_n is a lifetime broadening introduced for intermediate states containing the interaction with the excited electron and the valence band electrons. The double-differential cross section $d^2\sigma$ is obtained by multiplying the transition ratio w by the density of photon states in the solid angle and energy $\mathcal{V} |\mathbf{k}_{\text{out}}|^2 d|\mathbf{k}_{\text{out}}| d\Omega / (2\pi)^3$ and dividing the incident flux $c/\mathcal{V}$ [94]. Thus the intensity of RIXS spectra $I_{\text{RIXS}}(\omega, \mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{out}}) = d^2\sigma/d(\hbar\omega_{\mathbf{k}_{\text{out}}})d\Omega$ is

$$I_{\text{RIXS}}(\omega, \mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{out}}) = r_e^2 m^2 \omega_{\mathbf{k}_{\text{out}}}^3 \omega_{\mathbf{k}_{\text{in}}} \sum_f |\mathcal{F}_{fg}(\mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{out}})|^2 \delta(E_g - E_f + \hbar\omega) \quad (2.99)$$

where $r_e = e^2/mc^2$ is the classical electron radius, and

$$\mathcal{F}_{fg}(\mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{out}}) = \sum_n \frac{\langle f | \mathcal{D}_{\mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{out}}}^\dagger | n \rangle \langle n | \mathcal{D}_{\mathbf{k}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{in}}} | g \rangle}{E_g + \hbar\omega_{\mathbf{k}_{\text{in}}} - E_n + i\Gamma_n} \quad (2.100)$$

with dipole transition operator

$$\mathcal{D}_{\mathbf{k}, \boldsymbol{\varepsilon}} = \frac{1}{im\omega_{\mathbf{k}}} \sum_{i=1}^{N_e} e^{i\mathbf{k} \cdot \mathbf{r}_i} \boldsymbol{\varepsilon} \cdot \mathbf{p}_i. \quad (2.101)$$

Equation (2.99) is referred to as Kramers-Heisenberg equation for RIXS. It is generally used to calculate the RIXS cross section.

2.4.2 Dipole transition operators

Let us consider a direct L -edge RIXS, which is the transition of an electron from core band of $2p$ orbital to conduction band of $3d$ (and $4s$ for Cr) orbital induced by an incident photon. We assume that the state of $2p$ orbital with total angular momentum j and its z -component j_z enters to the state in $3d$ orbital conduction band with orbital α , and spin σ . The states in core band and in conduction band are written as $|2p; j, j_z\rangle = \psi_{2p, j, j_z}(\mathbf{r} - \mathbf{R}_i)$ and $|3d; \alpha, \sigma\rangle = \psi_{3d, \alpha, \sigma}(\mathbf{r} - \mathbf{R}_i)$, respectively. Here, $\mathbf{R}_i$ is the position of atom to which the i -th electron is bounded. The second quantization form, the transition dipole operator at $\mathbf{R}_i$ is

$$\mathcal{D}_{\mathbf{R}_i, \mathbf{k}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{in}}} = \sum_{\alpha, \sigma} \sum_{j_z} \langle 3d; \alpha, \sigma | \frac{1}{im\omega_{\mathbf{k}_{\text{in}}}} e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}_i} \boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{p}_i | 2p; j, j_z \rangle d_{i, \alpha, \sigma}^\dagger p_{i, j, j_z}, \quad (2.102)$$

where $d_{i, \alpha, \sigma}^\dagger$ is a creation operator of the $3d$ electron at $\mathbf{R}_i$ with orbital α and spin σ , and p_{i, j, j_z} is an annihilation operator of the $2p$ electron at $\mathbf{R}_i$ with total angular momentum j and its z -component j_z . We can select the total angular momentum j by adjusting the incident x-ray energy to resonant energy $\epsilon_j^{(L \text{ edge})} = E_n - E_g$, where E_n is the intermediate state with $2p$ core-hole state whose total angular momentum is j . X-ray exciting $j = 3/2$ electron in $2p$ orbital is referred to as L_3 edge, and one exciting $j = 1/2$ is as L_2 edge. The matrix element of dipole transition in the atom at $\mathbf{R}_i$ is written as

$$\begin{aligned} & \langle 3d; \alpha, \sigma | \frac{1}{im\omega_{\mathbf{k}_{\text{in}}}} e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}_i} \boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{p}_i | 2p; j, j_z \rangle \\ &= \frac{1}{im\omega_{\mathbf{k}_{\text{in}}}} \int d\mathbf{r}_i \psi_{3d, \alpha, \sigma}(\mathbf{r}_i - \mathbf{R}_i) (e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}_i} \boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{p}_i) \psi_{2p, j, j_z}(\mathbf{r}_i - \mathbf{R}_i) \end{aligned}$$

$$= \frac{e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{R}_i}}{im\omega_{\mathbf{k}_{\text{in}}}} \int d\mathbf{r}' \psi_{3d,\alpha,\sigma}(\mathbf{r}') \left(e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}'} \boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{p}_i \right) \psi_{2p,j,j_z}(\mathbf{r}'), \quad (2.103)$$

where $\mathbf{r}' = \mathbf{r}_i - \mathbf{R}_i$. In the dipole limit, we can assume that $e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}'} \simeq 1$ ($e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}_i} \simeq e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{R}_i}$). The momentum operator $\mathbf{p}$ satisfies $\mathbf{p}_i = m \frac{d\mathbf{r}_i}{dt} = m \frac{d\mathbf{r}'}{dt}$. With use of the Heisenberg equation $\frac{d\mathbf{r}'}{dt} = \frac{i}{\hbar} \left[\sum_{j=1}^{N_e} \frac{\mathbf{p}_j^2}{2m}, \mathbf{r}' \right]$ and the canonical commutation relationship $[\mathbf{r}_i, \mathbf{p}_j] = i\hbar\delta_{i,j}$ ($[\mathbf{p}_i^2, \mathbf{r}_j] = \frac{2\hbar}{i}\mathbf{p}_i$), the matrix element of dipole transition operator in absorption step is

$$\begin{aligned} & \langle 3d; \alpha, \sigma | \frac{1}{im\omega_{\mathbf{k}_{\text{in}}}} e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{r}_i} \boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{p}_i | 2p; j, j_z \rangle \\ & \simeq \frac{e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{R}_i}}{im\omega_{\mathbf{k}}} \int d\mathbf{r}' \psi_{3d,\alpha,\sigma}(\mathbf{r}') (\boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{p}_i) \psi_{2p,j,j_z}(\mathbf{r}'), \\ & \simeq \frac{e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{R}_i}}{\hbar\omega_{\mathbf{k}_{\text{in}}}} (\epsilon_{3d} - \epsilon_{2p,j}) \int d\mathbf{r}_i \psi_{3d,\alpha,\sigma}(\mathbf{r}') (\boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{r}') \psi_{2p,j,j_z}(\mathbf{r}') \\ & \simeq e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{R}_i} \int d\mathbf{r}' \psi_{3d,\alpha,\sigma}(\mathbf{r}') (\boldsymbol{\varepsilon}_{\text{in}} \cdot \mathbf{r}') \psi_{2p,j,j_z}(\mathbf{r}') = e^{i\mathbf{k}_{\text{in}} \cdot \mathbf{R}_i} c_{\alpha,\sigma}^{j,j_z}(\boldsymbol{\varepsilon}_{\text{in}}), \end{aligned} \quad (2.104)$$

where ϵ_{3d} is the average single-electron energy of the 3d electrons, $\epsilon_{2p,j}$ is one of the 2p electrons with total angular momentum j , and

$$c_{\alpha,\sigma}^{j,j_z}(\boldsymbol{\varepsilon}) = \int d\mathbf{r}' \psi_{3d,\alpha,\sigma}(\mathbf{r}') (\boldsymbol{\varepsilon} \cdot \mathbf{r}') \psi_{2p,j,j_z}(\mathbf{r}') \quad (2.105)$$

which is evaluated by using atomic orbital wave function. We use the resonant condition $\epsilon_{3d} - \epsilon_{2p,j} \simeq \epsilon_j^{(L \text{ edge})} \simeq \hbar\omega_{\mathbf{k}_{\text{in}}}$. The 3d-electron creation operator and the 2p-electron annihilation operator are written as $d_{i,\alpha,\sigma}^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_i} d_{\mathbf{k},\alpha,\sigma}^\dagger$ and $p_{i,j,j_z} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_i} p_{\mathbf{k},j,j_z}$ in Fourier series, respectively. The Fourier series of the dipole transition operator is

$$\mathcal{D}_{\mathbf{k}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{in}}} = \sum_{\mathbf{k}} \sum_{\alpha,\sigma} \sum_{j,j_z} c_{\alpha,\sigma}^{j,j_z}(\boldsymbol{\varepsilon}_{\text{in}}) d_{\mathbf{k}+\mathbf{k}_{\text{in}},\alpha,\sigma}^\dagger p_{\mathbf{k},j,j_z}. \quad (2.106)$$

In the emitting process, the valence band electron falls into the core-hole state which is made by an incident photon. We obtain the dipole transition operator in the same manner, which is

$$\mathcal{D}_{\mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{out}}}^\dagger = \sum_{\mathbf{k}} \sum_{\alpha,\sigma} \sum_{j,j_z} c_{\alpha,\sigma}^{j,j_z}(\boldsymbol{\varepsilon}_{\text{out}})^* p_{\mathbf{k},\alpha,\sigma}^\dagger d_{\mathbf{k}+\mathbf{k}_{\text{out}},\alpha,\sigma}. \quad (2.107)$$

Derivation of $c_{\alpha,\sigma}^{j,j_z}(\boldsymbol{\varepsilon})$

We make use of the atomic orbital wave function for the calculation of $c_{\alpha,\sigma}^{j,j_z}(\boldsymbol{\varepsilon})$. The wave function $\psi_{nlm}(\mathbf{r} = (r, \theta, \phi))\chi_\sigma$, which has principal quantum number n , azimuthal quantum number l , magnetic quantum number m , and spin quantum number σ , is written as

$$\psi_{nlm}(\mathbf{r})\chi_\sigma = R_{nl}(r)Y_{lm}(\theta, \phi)\chi_\sigma \longrightarrow |n, l, m\rangle|\sigma\rangle, \quad (2.108)$$

where R_{nl} , Y_{lm} , χ_σ are radial wave function, spherical harmonics, and spin function, respectively. 2p electrons have $n = 2$, $l = 1$, and 3d electrons have $n = 3$ and

$l = 2$. $|2p; j, j_z\rangle$ and $|3d; \alpha, \sigma\rangle = |3d; \alpha\rangle|\sigma\rangle$ can be obtained from the superposition of $|n, l, m\rangle|\sigma\rangle$, e.g.,

$$|n, j = l \pm \frac{1}{2}, j_z\rangle = \sqrt{\frac{l + \frac{1}{2} \mp j_z}{2l + 1}} |n, l, m = j_z + \frac{1}{2}\rangle |\downarrow\rangle \\ \pm \sqrt{\frac{l + \frac{1}{2} \pm j_z}{2l + 1}} |n, l, m = j_z - \frac{1}{2}\rangle |\uparrow\rangle, \quad (2.109)$$

which $|2p; j, j_z\rangle = |n = 2, j = 1 \pm \frac{1}{2}, j_z\rangle$, and

$$|3d, m = 2\rangle = i \frac{1}{\sqrt{2}} |xy\rangle + \frac{1}{\sqrt{2}} |x^2 - y^2\rangle, \quad |3d, m = -2\rangle = -i \frac{1}{\sqrt{2}} |xy\rangle + \frac{1}{\sqrt{2}} |x^2 - y^2\rangle \quad (2.110)$$

$$|3d, m = 1\rangle = -i \frac{1}{\sqrt{2}} |yz\rangle - \frac{1}{\sqrt{2}} |zx\rangle, \quad |3d, m = -1\rangle = -i \frac{1}{\sqrt{2}} |yz\rangle + \frac{1}{\sqrt{2}} |zx\rangle \quad (2.111)$$

$$|3d, m = 0\rangle = \frac{1}{\sqrt{2}} |3z^2 - r^2\rangle, \quad (2.112)$$

where $|3d, m\rangle = |n = 3, l = 2, m\rangle$ and $|\alpha\rangle = |3d; \alpha\rangle$ with atomic real 3d orbital α . Note that here the spin quantization axis is assumed along z direction, which requires the rotation of spin quantization axis if it is another direction.

We define that the angle between $\boldsymbol{\varepsilon}$ and z -axis is θ and the axis between x -axis and the vector projected $\boldsymbol{\varepsilon}$ to xy plane is ϕ . When $\mathbf{r}$ directs to z -axis,

$$\boldsymbol{\varepsilon} \cdot \mathbf{r} = r (\varepsilon_x \sin \theta \cos \phi + \varepsilon_y \sin \theta \sin \phi + \varepsilon_z \cos \theta) \quad (2.113)$$

$$= r \left(\frac{\varepsilon_x - i\varepsilon_y}{2} \sin \theta e^{i\phi} + \frac{\varepsilon_x + i\varepsilon_y}{2} \sin \theta e^{-i\phi} + \varepsilon_z \cos \theta \right). \quad (2.114)$$

The arbitrary of the coordinates is finally integrated out. With use of the identities of spherical harmonics

$$Y_{1,\pm 1}(\theta, \phi) = \mp \sqrt{\frac{3}{8\pi}} \sin \theta e^{\pm i\phi}, \quad Y_{1,0} = \sqrt{\frac{3}{4\pi}} \cos \theta, \quad (2.115)$$

we obtain

$$\boldsymbol{\varepsilon} \cdot \mathbf{r} = r \sqrt{\frac{4\pi}{3}} \left\{ \frac{-\varepsilon_x + i\varepsilon_y}{\sqrt{2}} Y_{1,1}(\theta, \phi) + \frac{\varepsilon_x + i\varepsilon_y}{\sqrt{2}} Y_{1,-1}(\theta, \phi) + \varepsilon_z Y_{1,0}(\theta, \phi) \right\}. \quad (2.116)$$

The matrix element of dipole operator is

$$\langle n', l', m' | \boldsymbol{\varepsilon} \cdot \mathbf{r} | n, l, m \rangle \\ = \sqrt{\frac{4\pi}{3}} \int dr r^2 R_{n'l'}^* r R_{nl} \left\{ \frac{-\varepsilon_x + i\varepsilon_y}{\sqrt{2}} \int d\Omega Y_{l',m'}^* Y_{1,1} Y_{l,m} \right. \\ \left. + \frac{\varepsilon_x + i\varepsilon_y}{\sqrt{2}} \int d\Omega Y_{l',m'}^* Y_{1,-1} Y_{l,m} + \varepsilon_z \int d\Omega Y_{l',m'}^* Y_{1,0} Y_{l,m} \right\} \quad (2.117)$$

where $d\Omega = \sin \theta d\theta d\phi$. We define Gaunt coefficient;

$$c^{l_1}(l', m'; l_2, m_2) = \int d\Omega Y_{l',m'}^* Y_{l_1,m_1} Y_{l_2,m_2}. \quad (2.118)$$

It can be analytically calculated as

$$\begin{aligned} c^{l_1}(l', m'; l_2, m_2) \\ = (-1)^{m'} \sqrt{\frac{(2l_1+1)(2l_2+1)(2l'+1)}{4\pi}} \begin{pmatrix} l_1 & l_2 & l' \\ m_1 & m_2 & -m' \end{pmatrix} \begin{pmatrix} l_1 & l_2 & l' \\ 0 & 0 & 0 \end{pmatrix}, \end{aligned} \quad (2.119)$$

where

$$\begin{aligned} \begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix} &= \delta_{m_1+m_2+m_3,0} (-1)^{l_1+m_2-m_3} \\ &\times \sqrt{\frac{(l_1+l_2-l_3)!(l_1-l_2+l_3)!(-l_1+l_2+l_3)!(l_3+m_3)!(l_3-m_3)!}{(l_1+l_2+l_3+1)!(l_1+m_1)!(l_1-m_1)!(l_2+m_2)!(l_2-m_2)!}} \\ &\times \sum_{\lambda} (-1)^{\lambda} \frac{(l_1-m_1+\lambda)!(l_2+l_3+m_1-\lambda)!}{\lambda!(-l_1+l_2+l_3-\lambda)!(l_1-l_2+m_3+\lambda)!(l_3-m_3-\lambda)!} \end{aligned} \quad (2.120)$$

is referred to as Wigner's 3- j . λ is an integer satisfying $\max(0, l_2 - l_1 - m_3) \leq \lambda \leq \min(l_3 - m_3, l_2 - l_1 + l_3)$. Thus, the matrix element of dipole operator is written as

$$\begin{aligned} \langle n', l', m' | \boldsymbol{\varepsilon} \cdot \mathbf{r} | n, l, m \rangle &= \sqrt{\frac{4\pi}{3}} \langle r \rangle_{n,l}^{n',l'} \left\{ \frac{-\varepsilon_x + i\varepsilon_y}{\sqrt{2}} c^1(l', m+1; l, m) \delta_{m', m+1} \right. \\ &\quad \left. + \frac{\varepsilon_x + i\varepsilon_y}{\sqrt{2}} c^1(l', m-1; l, m) \delta_{m', m-1} + \varepsilon_z c^1(l', m; l, m) \delta_{m', m} \right\}, \end{aligned} \quad (2.121)$$

where $\langle r \rangle_{n,l}^{n',l'} = \int dr r^2 R_{n'l'}^* r R_{nl}$.

2.4.3 RIXS spectra

Equation (2.100) is expanded to

$$\begin{aligned} \mathcal{F}_{fg}(\mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{out}}) &= \sum_n \langle f | \mathcal{D}_{\mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{out}}}^\dagger | n \rangle \frac{1}{\hbar\omega_{\mathbf{k}_{\text{in}}} - (\epsilon_{3d} - \epsilon_{2p,j}) - i\Gamma_n} \\ &\times \sum_{l=0}^{\infty} \left\{ \frac{(E_n - E_g) - (\epsilon_{3d} - \epsilon_{2p,j})}{\hbar\omega_{\mathbf{k}_{\text{in}}} - (\epsilon_{3d} - \epsilon_{2p,j}) - i\Gamma_n} \right\}^l \langle n | \mathcal{D}_{\mathbf{k}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{in}}} | g \rangle. \end{aligned} \quad (2.122)$$

We assume that Γ_n is independent of n ($\Gamma_n \simeq \Gamma$). When the core-hole life time is short enough, i.e., Γ is large as compared with characteristic energy scale of $E_n - E_g$, we can approximate that $l=0$ term in the right hand side of Eq. (2.122) is dominant. Equation (2.122) is approximated as

$$\mathcal{F}_{fg}(\mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{out}}) \simeq \frac{1}{\hbar\omega_{\mathbf{k}_{\text{in}}} - (\epsilon_{3d} - \epsilon_{2p,j}) - i\Gamma} \langle f | \mathcal{D}_{\mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{out}}}^\dagger \mathcal{D}_{\mathbf{k}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{in}}} | g \rangle, \quad (2.123)$$

which is referred to as a fast-collision approximation. With use of Eq. (2.106) and (2.107),

$$\begin{aligned} &\langle f | \mathcal{D}_{\mathbf{k}_{\text{out}}, \boldsymbol{\varepsilon}_{\text{out}}}^\dagger \mathcal{D}_{\mathbf{k}_{\text{in}}, \boldsymbol{\varepsilon}_{\text{in}}} | g \rangle \\ &= \sum_{\mathbf{k}, \mathbf{k}'} \sum_{\alpha, \alpha'} \sum_{\sigma, \sigma'} \sum_{j_z, j'_z} c_{\alpha', \sigma'}^{j', j'_z}(\boldsymbol{\varepsilon}_{\text{out}})^* c_{\alpha, \sigma}^{j, j_z}(\boldsymbol{\varepsilon}_{\text{in}}) \langle f | d_{\mathbf{k}' + \mathbf{k}_{\text{out}}, \alpha', \sigma'}^\dagger d_{\mathbf{k} + \mathbf{k}_{\text{in}}, \alpha, \sigma}^\dagger p_{\mathbf{k}', j, j'_z}^\dagger p_{\mathbf{k}, j, j_z} | g \rangle \end{aligned}$$

$$= - \sum_{\mathbf{k}} \sum_{\alpha, \alpha'} \sum_{\sigma, \sigma'} \sum_{j_z} c_{\alpha, \sigma}^{j, j_z}(\epsilon_{\text{in}}) c_{\alpha', \sigma'}^{j, j_z}(\epsilon_{\text{out}})^* \langle f | d_{\mathbf{k}+\mathbf{k}_{\text{in}}, \alpha, \sigma}^\dagger d_{\mathbf{k}+\mathbf{k}_{\text{out}}, \alpha', \sigma'} | g \rangle, \quad (2.124)$$

where we use the identity $p_{\mathbf{k}', j, j_z}^\dagger | g \rangle = 0$ implying that core $2p$ states are filled with electrons in the ground state.

With use of the identity $\delta(E_g - E_f + \hbar\omega) = -\frac{1}{\pi} \text{Im} \frac{1}{E_g - E_f + \hbar\omega + i\eta}$, the RIXS spectra (2.99) is

$$\begin{aligned} & I_{\text{RIXS}}(\omega, \mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \epsilon_{\text{in}}, \epsilon_{\text{out}}) \\ &= - \frac{r_e^2 m^2 \omega_{\mathbf{k}_{\text{out}}}^3 \omega_{\mathbf{k}_{\text{in}}}}{\pi} \text{Im} \sum_f |\mathcal{F}_{fg}(\mathbf{k}_{\text{in}}, \mathbf{k}_{\text{out}}, \epsilon_{\text{in}}, \epsilon_{\text{out}})|^2 \frac{1}{E_g - E_f + \hbar\omega + i\eta} \\ &= - \frac{r_e^2 m^2 \omega_{\mathbf{k}_{\text{out}}}^3 \omega_{\mathbf{k}_{\text{in}}}}{\pi} \frac{1}{\{\hbar\omega_{\mathbf{k}_{\text{in}}} - (\epsilon_{3d} - \epsilon_{2p, j})\}^2 + \Gamma^2} \\ &\quad \times \text{Im} \left[\sum_{\mathbf{k}, \mathbf{k}'} \sum_{\kappa, \lambda, \mu, \nu} \sum_{\sigma_1, \sigma_2, \sigma_3, \sigma_4} \sum_{j_z} c_{\kappa, \sigma_1}^{j, j_z}(\epsilon_{\text{out}}) c_{\lambda, \sigma_2}^{j, j_z}(\epsilon_{\text{in}})^* \sum_{j'_z} c_{\mu, \sigma_3}^{j, j'_z}(\epsilon_{\text{in}}) c_{\nu, \sigma_4}^{j, j'_z}(\epsilon_{\text{out}})^* \right. \\ &\quad \left. \times \langle g | d_{\mathbf{k}+\mathbf{k}_{\text{out}}, \kappa, \sigma_1}^\dagger d_{\mathbf{k}+\mathbf{k}_{\text{in}}, \lambda, \sigma_2} \frac{1}{E_g - H + \hbar\omega + i\eta} d_{\mathbf{k}'+\mathbf{k}_{\text{in}}, \mu, \sigma_3}^\dagger d_{\mathbf{k}'+\mathbf{k}_{\text{out}}, \nu, \sigma_4} | g \rangle \right]. \quad (2.125) \end{aligned}$$

$\omega_{\mathbf{k}_{\text{in}}}$ and $\omega_{\mathbf{k}_{\text{out}}}$ can be approximated to be constant. Thus, the RIXS spectrum depends on $\mathbf{q} = \mathbf{k}_{\text{in}} - \mathbf{k}_{\text{out}}$, and it becomes

$$\begin{aligned} & I_{\text{RIXS}}(\omega, \mathbf{q}, \epsilon_{\text{in}}, \epsilon_{\text{out}}) \\ &\propto -\text{Im} \left[\sum_{\mathbf{k}, \mathbf{k}'} \sum_{\kappa, \lambda, \mu, \nu} \sum_{\sigma_1, \sigma_2, \sigma_3, \sigma_4} \sum_{j_z} c_{\kappa, \sigma_1}^{j, j_z}(\epsilon_{\text{out}}) c_{\lambda, \sigma_2}^{j, j_z}(\epsilon_{\text{in}})^* \sum_{j'_z} c_{\mu, \sigma_3}^{j, j'_z}(\epsilon_{\text{in}}) c_{\nu, \sigma_4}^{j, j'_z}(\epsilon_{\text{out}})^* \right. \\ &\quad \left. \times \langle g | d_{\mathbf{k}, \kappa, \sigma_1}^\dagger d_{\mathbf{k}+\mathbf{q}, \lambda, \sigma_2} \frac{1}{E_g - H + \hbar\omega + i\eta} d_{\mathbf{k}'+\mathbf{q}, \mu, \sigma_3}^\dagger d_{\mathbf{k}', \nu, \sigma_4} | g \rangle \right]. \quad (2.126) \end{aligned}$$

Thus, the RIXS spectra is the superposition of dynamical susceptibilities (2.93), which is

$$\begin{aligned} & I_{\text{RIXS}}(\omega, \mathbf{q}, \epsilon_{\text{in}}, \epsilon_{\text{out}}) \\ &\propto \text{Im} \left[\sum_{\kappa, \lambda, \mu, \nu} \sum_{\sigma_1, \sigma_2, \sigma_3, \sigma_4} \sum_{j_z} c_{\kappa, \sigma_1}^{j, j_z}(\epsilon_{\text{out}}) c_{\lambda, \sigma_2}^{j, j_z}(\epsilon_{\text{in}})^* \sum_{j'_z} c_{\mu, \sigma_3}^{j, j'_z}(\epsilon_{\text{in}}) c_{\nu, \sigma_4}^{j, j'_z}(\epsilon_{\text{out}})^* \right. \\ &\quad \left. \times \chi_{\lambda \mu}^{ss'}(\mathbf{q}, \mathbf{q}, \omega) \right]. \quad (2.127) \end{aligned}$$

This is the final result of RIXS spectrum. We should calculate only the set of $c_{\alpha, \sigma}^{j, j_z}(\epsilon)$ and the dynamical susceptibilities $\chi_{\lambda \mu}^{ss'}(\mathbf{q}, \mathbf{q}, \omega)$ to obtain the RIXS spectra in the fast-collision approximation.

Chapter 3

IN-PLANE ANISOTROPY OF RESISTIVITY IN ANTIFERROMAGNETIC IRON ARSENIDE

Below the Néel temperature, iron arsenide leads to stripe-ordered antiferromagnetism, breaking their four-fold symmetry. The in-plane anisotropy due to the breakdown of the symmetry is observed by the measurement of resistivity. The origin of the anisotropy of resistivity is controversial with regard to either the spin fluctuation and orbital fluctuation. Both fluctuations are considered as the source of pairing mechanism in iron-arsenide superconductors. The previous theoretical studies of the anisotropy introduced in Chap. 1 are related to the anisotropy above the Néel temperature. In contrast to the studies, we first obtain antiferromagnetic phase as the ground state of iron-pnictides from multi-orbital Hubbard model with a mean field approximation from the procedure introduced in Sec. 2.1. The resistivity is then driven from memory-function formalism discussed in Sec. 2.2. We show that the anisotropy of resistivity in iron arsenides with antiferromagnetic phase comes from the reconstructed Fermi surface due to the anisotropic antiferromagnetic ordering.

In this chapter, we introduce a model Hamiltonian obtained from first-principles calculations in Sec. 3.1. Our result of anisotropy of resistivity is shown in Sec. 3.2. We also discuss the anisotropy of resistivity in paramagnetic phase in Sec. 3.3 from the view point of orbital ordering.

3.1 Model Hamiltonian

In the study of iron-based superconductors, a simplified model Hamiltonian that reproduces the band structure and Fermi surface obtained from first-principles calculation is usually used. The most simplified model is a two-orbital Hamiltonian containing only d_{yz} and d_{zx} orbitals [95, 96]. The model reflects the fact that these two orbitals are dominant on Fermi surface. However, as shown before, d_{xy} orbital is also present on Fermi surface. To treat this, a three-orbital model [97] is sometimes used.

To accurately treat the five-orbital character in iron-based superconductors, a five-orbital model that includes all five d orbitals is usually used. This model is often determined to reproduce the electronic structure of LaFeAsO [42, 98, 99, 100]. This

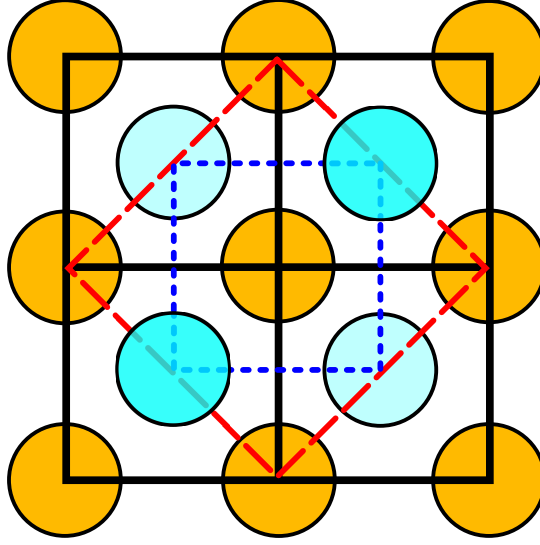


Fig. 3.1 The schematically illustrated unit cell of LaFeAsO. The orange circles at the corners of the squares represent Fe atoms, and the light blue circles middle of them represent As atoms. Red dashed lines indicate the original unit cell, while blue dotted lines indicate reduced unit cell. As atoms locate alternately above and below Fe plane (See Fig. 1.2).

model is in two dimensions, reflecting quasi-two dimensionality appearing in the cylindrical structure of the Fermi surface in LaFeAsO. Since As atoms locate alternately above and below the Fe plane as shown in Fig. 3.1, the original unit cell contains two Fe atoms, which leads to ten-orbital model. In order to avoid the intricacy, the model is downfolded to reduced unit cell containing one Fe atom. The parameters of, for instance, BaFe₂As₂ taking into account the three dimensionality are given by Graser *et al.* [101]. The comprehensive sets of parameters are given by Miyake *et al.* [102].

We show another hopping integrals given by Kuroki *et al.* [42] in Tab. 3.1. The Hamiltonian is given in Eq. (2.1). Figure 3.2 shows the Fermi surface in the reduced Brillouin zone corresponding to the reduced unit cell with this parameters. We make use of the parameters given by Kuroki *et al.* [42] in following calculations. Although tis parameters are obtained from the band structure of LaFeAsO, the band structure is similar to that of BaFe₂As₂. Orbitals dominantly contributing to the Fermi surface are t_{2g} orbitals: hole pockets consist of d_{yz} and d_{zx} orbitals, while electron pockets consist of d_{xy} and d_{yz} orbitals.

3.2 Anisotropy of Resistivity in Antiferromagnetic Phase

3.2.1 Parameters and Fermi surfaces

In addition to the kinetic term Hamiltonian (2.1) with parameters in Tab. 3.1, the interaction Hamiltonian (2.2) is introduced in order to obtain the antiferromagnetic state. We set $U = 1.2$ eV and $J = 0.22$ eV to yield a magnetic moment $m = 0.8\mu_B$ (μ_B is the Bohr magneton) at $n = 6.0$, where n is the electron density and

$$m = \sum_{\mu} \langle n_{\mathbf{Q},\mu,\uparrow} - n_{\mathbf{Q},\mu,\downarrow} \rangle \mu_B. \quad (3.1)$$

(μ, ν) \ $[\Delta x, \Delta y]$	[1,0]	[1,1]	[2,0]	[2,1]	[2,2]	σ_y	I	σ_d
(1,1)	-0.7		-0.4	0.2	-0.1	+	+	+
(1,2)		-1.1			-0.2	-	-	+(1,3)
(1,3)	1.1	-1.1			-0.2	+	-	+(1,2)
(1,4)	-3.0			-0.2		+	+	-
(1,5)		-1.7			0.1	-	+	+
(2,2)	-0.8	2.4	0.2	-0.4	0.4	+	+	+(3,3)
(2,3)		0.9		-0.2	0.4	-	+	+
(2,4)		1.0		0.2		-	-	-(3,4)
(2,5)	-2.4	-0.1		-0.3		+	-	+(3,5)
(3,3)	-3.4	2.4	-0.2	0.1	0.4	+	+	+(2,2)
(3,4)	3.5	-1.0		0.2		+	-	-(2,4)
(3,5)		-0.1				-	-	+(2,5)
(4,4)	3.1	-0.7	-0.2			+	+	+
(4,5)				0.1		-	+	-
(5,5)	1.6	1.2	-0.3	-0.3	-0.3	+	+	+

Table. 3.1 Hopping integrals $t(\Delta x, \Delta y; \mu, \nu)$ in units of 0.1eV. $[\Delta x, \Delta y]$ denotes the in-plane hopping vector, and (μ, ν) the orbitals. σ_y , I , and σ_d corresponds to $t(\Delta x, -\Delta y; \mu, \nu)$, $t(-\Delta x, -\Delta y; \mu, \nu)$, $t(\Delta y, \Delta x; \mu, \nu)$, respectively, where ‘ $\pm$ ’ and ‘ $\pm(\mu', \nu')$ ’ in the row of (μ, ν) mean that the corresponding hopping is equal to $\pm t(\Delta x, \Delta y; \mu, \nu)$ and $\pm t(\Delta x, \Delta y; \mu', \nu')$, respectively. This table, combined with the relation $t(\Delta x, \Delta y; \mu, \nu) = t(-\Delta x, -\Delta y; \nu, \mu)$, gives all the in-plane hoppings ≥ 0.01 eV up to fifth neighbors. The correspondence of orbitals and numbers are 1: $d_{3z^2-r^2}$, 2: d_{xz} , 3: d_{yz} , 4: $d_{x^2-y^2}$, and 5: d_{xy} , where x , y , and z refer to those for reduced unit cell. The on-site energies for the five orbitals are $(\varepsilon_1, \varepsilon_2, \varepsilon_3, \varepsilon_4, \varepsilon_5) = (10.75, 10.96, 10.96, 10.62, 11.12)$ eV. The table is obtained from parameters given by Kuroki *et al.* [42]

This value is chosen to be close to the measured m of BaFe_2As_2 [36]. The calculated value of m linearly decreases with increasing n [37]. This is because the nesting condition between the Fermi surfaces at the Y and M points in the paramagnetic phase becomes worse. This will lead to an incommensurate antiferromagnetic state and thus our assumption of the commensurate antiferromagnetic state may fail. Actually, m at the $(\pi, 0)$ -ordered state in our calculation shows a discontinuous change when n exceeds 6.03.

The Fermi surfaces at $n = 5.90$, 5.95, and 6.0 are shown in Figs. 3.3(a), 3.3(b), and 3.3(c), respectively. At $n = 6.0$, there are five pockets: A hole pocket at the Γ point, two pockets (Dirac pockets) coming from the Dirac-type linear dispersions on the left and right sides of the hole pocket, and two electron pockets above and below the hole pocket. At $n = 5.98$, the Dirac points meet the Fermi level [37]. At $n = 5.95$, the electron pockets disappear, but the Dirac pockets remain [Fig. 3.3(b)]. With further reducing n , i.e., increasing hole concentration, the Dirac pockets grow as shown in Fig. 3.3(a), since the Dirac points are above the Fermi level.

3.2.2 Anisotropy of resistivity

Since Co is substituted for Fe in electron-doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, Co atoms act as impurities. In hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, K does not directly affect Fe sites, but may give an additional potential through electrostatic interaction. Figure 3.4 shows the

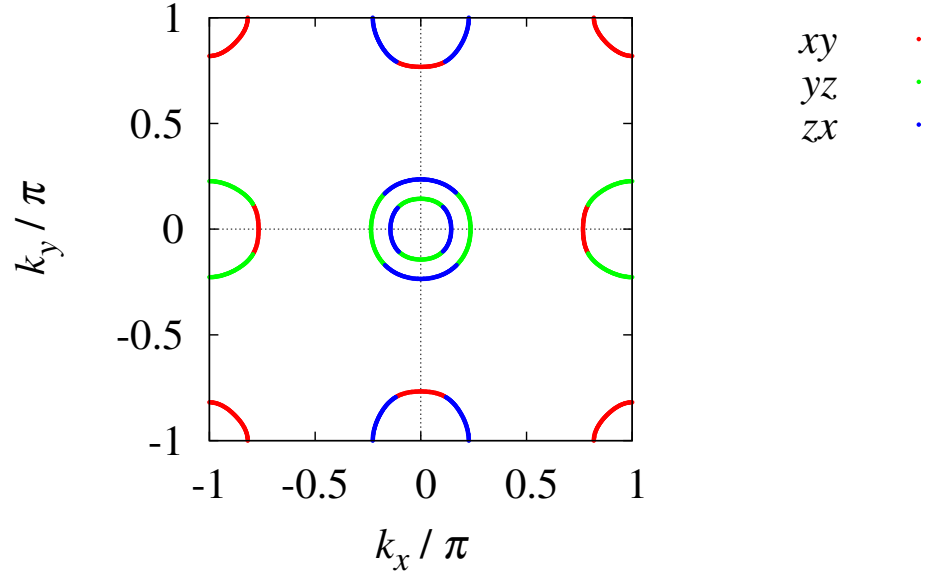


Fig. 3.2 Fermi surface of LaFeAsO for reduced Brillouin zone. Color indicates the dominant orbital on the Fermi surface. Red is for d_{xy} orbital, green is for d_{yz} orbital, and blue is for d_{zx} orbital.

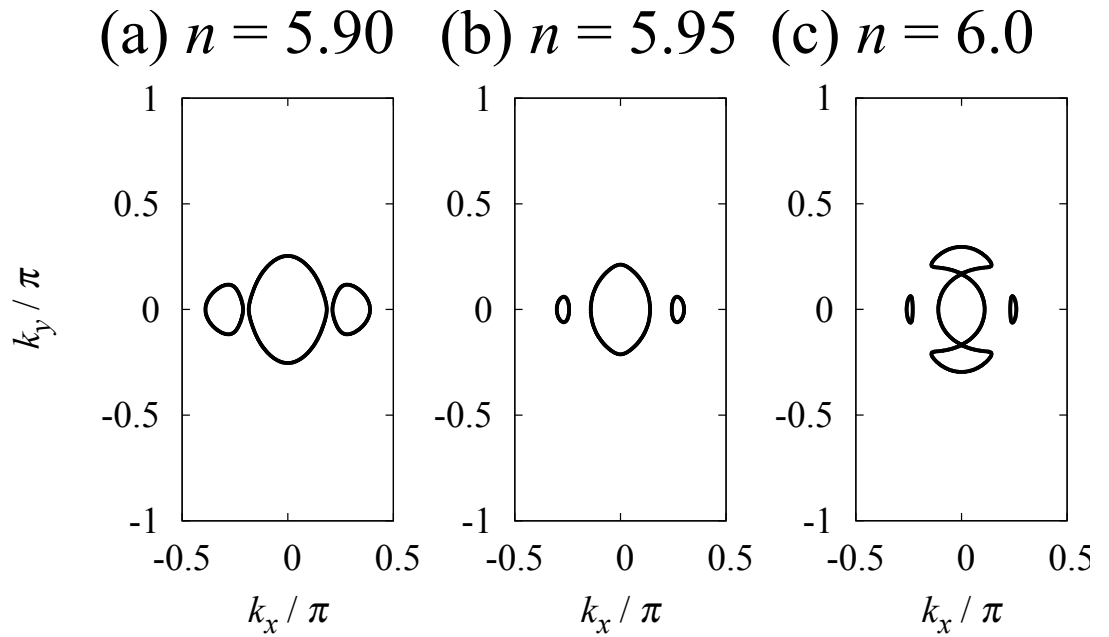


Fig. 3.3 The Fermi surfaces at (a) $n = 5.90$, (b) $n = 5.95$, and (c) $n = 6.0$ in the stripe-type AFM Brillouin zone.

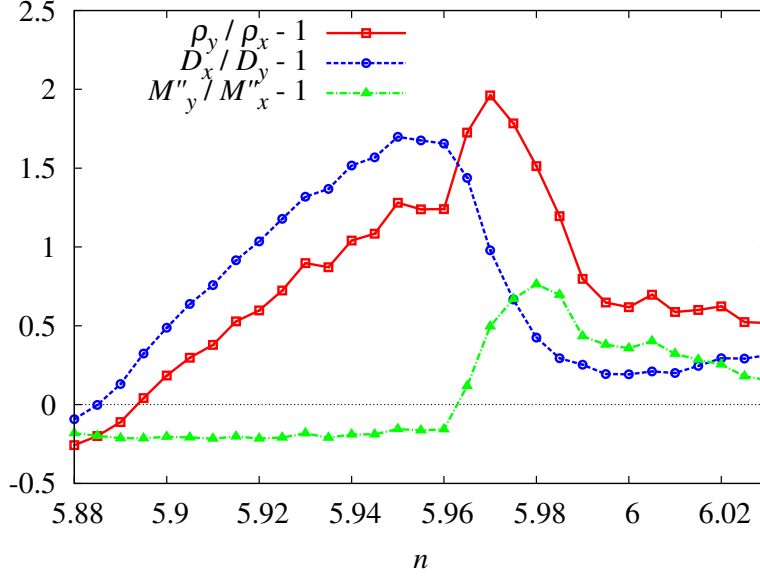


Fig. 3.4 The electron density n dependence of anisotropy of resistivity $\rho_y/\rho_x - 1$ (red squares), Drude weight $D_x/D_y - 1$ (blue circles), and memory function $M''_y/M''_x - 1$ (green triangles). Here, $D_{x(y)} = D_{xx(yy)}(0)$ and $M''_{x(y)} = \text{Im } M_{xx(yy)}(0)$. The impurity is ignored in the calculations of the Drude weight.

anisotropy of resistivity given by $\rho_y/\rho_x - 1$. We find that $\rho_y > \rho_x$ near $n = 6.0$, which agrees with the experiments in electron-doped $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ [50, 56]. The ratio of anisotropy shows a maximum value of 1.9 at $n = 5.97$ and decreases with increasing n . This doping dependence is qualitatively consistent with experimental observations, although the maximum of the ratio appears above $n = 6.0$ in the experiments [56, 71]. Here, we emphasize that, around undoping ($n = 6.0$) region, including impurity scattering makes the in-plane anisotropy of resistivity (2.59) much larger than that of the Drude weight (2.47) calculated by using Eq. (2.41) and (2.43). Note that the impurity is ignored in the calculations of the Drude weight. We thus can say that the anisotropy of the resistivity is enhanced by the effect of impurity scattering described by the memory function (2.57).

Below $n = 5.96$, the anisotropy of Drude weight monotonically decreases while the anisotropy of memory function, i.e., the anisotropy of scattering rate, is almost constant with a negative sign, opposite to that near $n = 6.0$. In this region, the two electron pockets at $n = 6.0$ disappear, and the Fermi surface has two Dirac pockets and one hole pocket as shown in Fig. 3.3. This topology of the Fermi surface is unchanged down to $n = 5.88$. Therefore, we may say that the anisotropy of scattering rate depends only on the topology of the Fermi surface, while the anisotropy of Drude weight depends on the details of the Fermi surface such as the curvature of Dirac pockets.

The combination of the Drude weight and the scattering rate leads to the monotonic decrease of the anisotropy of resistivity below $n = 5.96$. The anisotropy is finally reversed in the hole-doped side of $n < 5.89$, i.e., $\rho_y < \rho_x$. The reverse of anisotropy is also consistent with the doping dependence of anisotropy in hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, [71] although the observed carrier concentration where the anisotropy is reversed ($x \sim 0.2$) quantitatively disagrees with our results. The agree-

ment with the experiments will be improved by tuning the hopping parameters and by being beyond the dilute limit for large c and the Born approximation. Furthermore, the commensurability that we have assumed may be getting worse away from $n = 6$ as mentioned above, leading to a possible modification of the present doping dependence of anisotropy through the change of Fermi surface topology. We believe that such factors will give a better agreement with experiments, though there is no specific calculation. These remain as a future problem.

In order to understand the origin of the anisotropy in the scattering rate near $n = 6.0$ and its doping independent behavior, we plot $\text{Im } M_{\alpha\alpha}(\mathbf{k}_0, 0)/(cI_{\text{imp}}^2)$ [see Eq. (2.57)] on the Fermi surfaces in Figs. 3.5(a), 3.5(c) and Figs. 3.5(b), 3.5(d) for $\alpha = x$ and $\alpha = y$, respectively. Figure 3.5(a) shows that the scattering rate for $\alpha = x$ mainly comes from the Dirac pockets. On the other hand, Fig. 3.5(b) shows that the electron pockets significantly contribute to the scattering rate for $\alpha = y$. In other words, electrons are mainly scattered at the Dirac pockets (electron pockets) when current flows along the x (y) direction. At $n = 6.0$, we find that the integrated scattering rate over the Fermi surfaces is larger for $\alpha = y$ than for $\alpha = x$.

Since the electron pockets controls the scattering rate for $\alpha = y$, we can expect the reverse of the anisotropy if the electron pockets disappear [see Figs. 3.3(a) and 3.3(b)]. This is actually the case below $n = 5.96$ [see Figs. 3.5(c) and 3.5(d)]. These results indicate that both including the impurity scattering and treating the Fermi-surface evolutions are essential for a proper understanding of the origin of the in-plane anisotropy of resistivity.

3.3 Anisotropy of Resistivity in Paramagnetic Phase

In the light of our findings in the antiferromagnetic phase, we discuss the anisotropy in the paramagnetic phase. Above the Néel temperature T_N and structural transition temperature T_S , the anisotropy may be related to the nematic state observed experimentally [51]. A possible mechanism of the nematicity is the orbital ordering and fluctuation. Angle resolved photo-emission spectroscopy measurements have clearly shown the splitting of d_{xz} and d_{yz} orbitals ($\varepsilon_{yz} > \varepsilon_{xz}$) in the nematic state [63]. The splitting is an evidence of the orbital ordering. Based on this observation, we introduce an energy level splitting between ε_{yz} and ε_{xz} in H_0 (2.1) and calculate the Drude weight in the paramagnetic state at $n = 6$. We find that the anisotropy of the Drude weight is opposite to the observed anisotropy of the resistivity, as shown in the $\varepsilon_{yz} > \varepsilon_{xz}$ region of Fig. 3.6.

Then, according to the case in the antiferromagnetic phase, we introduce impurities and apply the memory-function approach, resulting in the anisotropy consistent with experiments, i.e., $\rho_y > \rho_x$ at $\varepsilon_{yz} > \varepsilon_{xz}$ as shown in Fig. 3.6. The interplay of impurity scattering and anisotropic Fermi surfaces due to the orbital ordering has also theoretically been reported by a T -matrix formalism [68]. Therefore, we suggest that the orbital ordering is one of the possible origins of the anisotropy of resistivity above T_S .

3.4 Summary

We have investigated the in-plane anisotropy of resistivity with multi-orbital Hubbard model in the antiferromagnetic phase. The resistivity has been obtained from the memory-function approach. Both the Drude weight and the scattering rate tend to make resistivity along the ferromagnetic direction larger than that of antiferro-

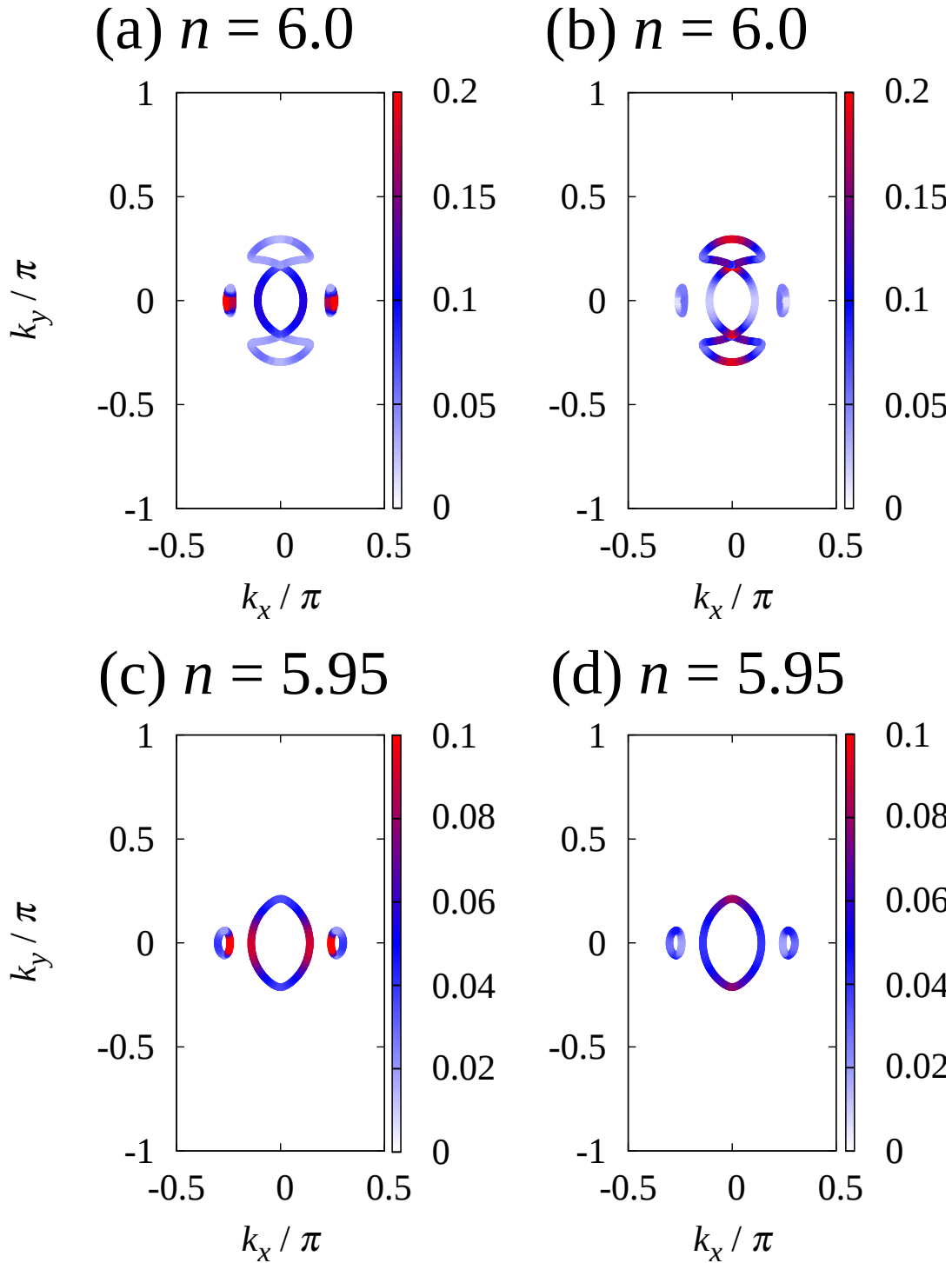


Fig. 3.5 The intensity of $\text{Im } M_{\alpha\alpha}(\mathbf{k}_0, 0)/(cI_{\text{imp}}^2)$. (a) $\alpha = x$ and $n = 6.0$. (b) $\alpha = y$ and $n = 6.0$. (c) and (d): The same as (a) and (b), respectively, but $n = 5.95$.

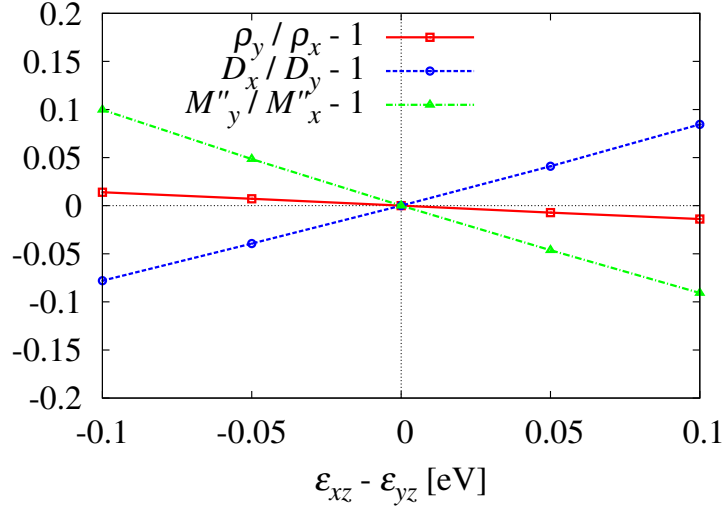


Fig. 3.6 Anisotropy vs energy separation $d_{xz} - d_{yz}$. Plotted are three quantities indicating anisotropy with respect to the energy separation: the anisotropy of resistivity $\rho_y/\rho_x - 1$ (red squares), Drude weight $D_x/D_y - 1$ (blue circles), and memory function $M''_y/M''_x - 1$ (green triangles). Here, $D_{x(y)} = D_{xx(yy)}(0)$ and $M''_{x(y)} = \text{Im } M_{xx(yy)}(0)$.

magnetic direction in parent compounds, which is consistent with in-plane anisotropy in experiments. When holes are introduced, the anisotropy of the scattering rate reverses and becomes independent of carrier concentration, which is related to the disappearance of electron pockets. On the other hand, the Drude weight monotonically decreases as a consequence of deformation of the Dirac pocket with hole doping. This eventually leads to the reverse of anisotropy of resistivity, which is also consistent with experiments. These results indicate that the origin of the anisotropy is attributed to the interplay of impurity scattering and anisotropic electronic states, both of which are strongly influenced by the topology of Fermi surface. In addition, we have successfully obtained the anisotropy of resistivity consistent with experiments. These results suggest the importance of impurity scattering in the anisotropy of resistivity of iron arsenide.

Chapter 4

SPIN AND CHARGE EXCITATIONS IN ANTIFERROMAGNETIC CHROMIUM

A representative material of itinerant antiferromagnet is chromium (Cr). After the discovery of spin-density wave (SDW) order in Cr, many investigations have been carried out. Among them, neutron scattering experiment is the most suitable for the study of SDW ground state and its magnetic excitations.

Recent development of synchrotron radiation equipment has proved that resonant inelastic x-ray scattering (RIXS) is a new tool to detect magnetic excitations. Needless to say, we observe not only spin excitation but also charge excitation, since inelastic x-ray scattering directly excites charge degree of freedom. Our calculations suggest that measurements of RIXS can detect the magnetic and charge excitations in Cr. We can also detect the orbital character of materials from the polarization of the x-ray as shown in this chapter.

We introduce electronic states of Cr in Sec. 4.1. The antiferromagnetic state of Cr is obtained from the procedure introduced in Sec. 2.1, and the excitation properties of the state are discussed in the following sections. Optical conductivity of Cr is calculated in Sec. 4.2. A measurement of optical conductivity is often used to see a gap opening at Fermi energy, i.e., the evidence of an SDW ordering. In Sec. 4.3, we calculate the dynamical susceptibilities of spin and charge. The calculated dynamical spin susceptibility is a prediction to high-energy inelastic neutron scattering in Cr. We calculate the RIXS spectra in Sec. 4.4 from the dynamical susceptibilities obtained in Sec. 4.3.

4.1 Electronic Structure of Chromium

From the theoretical viewpoint, it is necessary to include all of the $3d$ orbitals in a model of Cr for the precise description of both the collective and individual excitations, as proven in the study of the antiferromagnetic phase of iron arsenides [82, 83]. For the incommensurate SDW state, it is not easy to perform band-structure calculations including all of the orbitals, since we need to use a large unit cell. In fact, commensurability with the vector $(2\pi/a, 0, 0)$ has been assumed in the previous first-principles calculations [103, 104, 105, 106]. In model calculations for the commensurate SDW state, two-band models have been used, and the detailed band structures have been ignored [107]. Therefore, it is important to perform the study of spin dynamics of Cr by using a precise band structure, even though the commensurate SDW state is

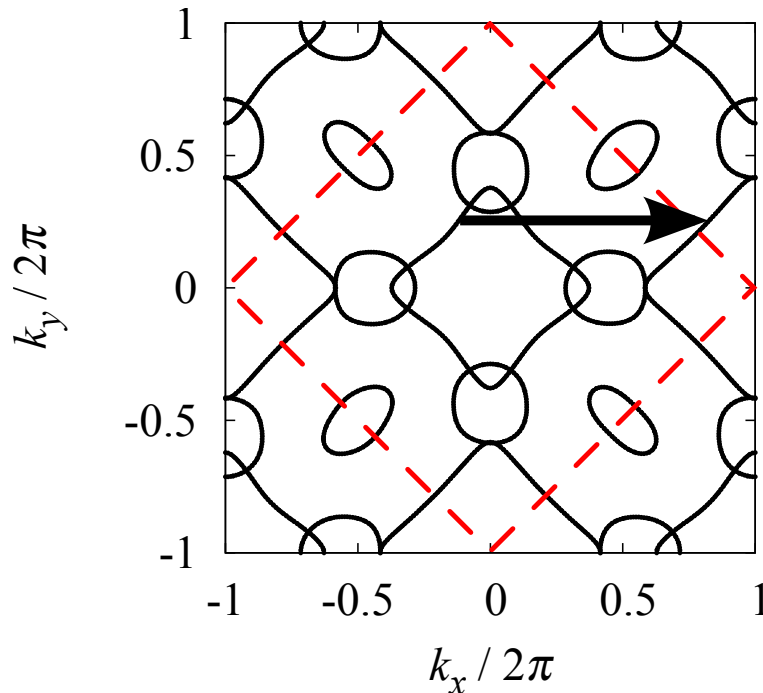


Fig. 4.1 Fermi surface of paramagnetic Cr at $k_z = 0$. Red dashed lines indicate the Brillouin zone, and the arrow indicates the nesting vector. The lattice constant a is set to be unity.

assumed. The assumption will not affect the high-energy spin dynamics, since the incommensurability seen in pure Cr is determined by low-energy band structure near the Fermi level.

The hopping integrals of Cr are given by Harrison [108] in terms of the Slater-Koster parameters [109]. However, we derive the hopping integrals by ourselves since the shape of its Fermi surface given by Harrison is inaccurate.

The values of hopping integrals are obtained from the paramagnetic phase with the lattice constant $a = 2.883 \text{ \AA}$ of body-centered cubic (bcc) lattice. First, the first-principles band structure is calculated by PWscf [110], where the projector augmented wave pseudopotentials with Cr.pw91-sp-van.UPF is used. Then, we calculate the values of hopping integrals in Eq. (2.1) by Wannier90 [111], inputting the PWscf data. We take $8 \times 8 \times 8$ points in the first Brillouin zone of the bcc lattice and set the orbitals to be five $3d$ orbitals and one $4s$ orbital.

Figure 4.1 shows the Fermi surface of paramagnetic Cr at $k_z = 0$. The tetrahedrons (drawn as square in Fig. 4.1) at Γ point $(k_x, k_y, k_z) = (0, 0, 0)$, which is a hole pocket, and that at H point $(2\pi, 0, 0)$, which is an electron pocket, are almost the same size. Thus, it is clear that the Cr has a good nesting vector. The small difference of these tetrahedrons leads to the incommensurate SDW. This Fermi surface well reproduces the previous results obtained from other first-principles calculations [10, 11, 12, 13, 14].

We assume that the SDW in Cr is a perfect commensurate state in spite of its incommensurate state confirmed experimentally since it is hard to work out numerically with the condition of the incommensurate state. This assumption is expected hardly to affect our conclusions.

We self-consistently solve mean-field equations to determine the order parameters as introduced in Sec. 2.1. From the self-consistent solution, we obtain $E_{\mathbf{k}_0, \sigma, \epsilon}$ and

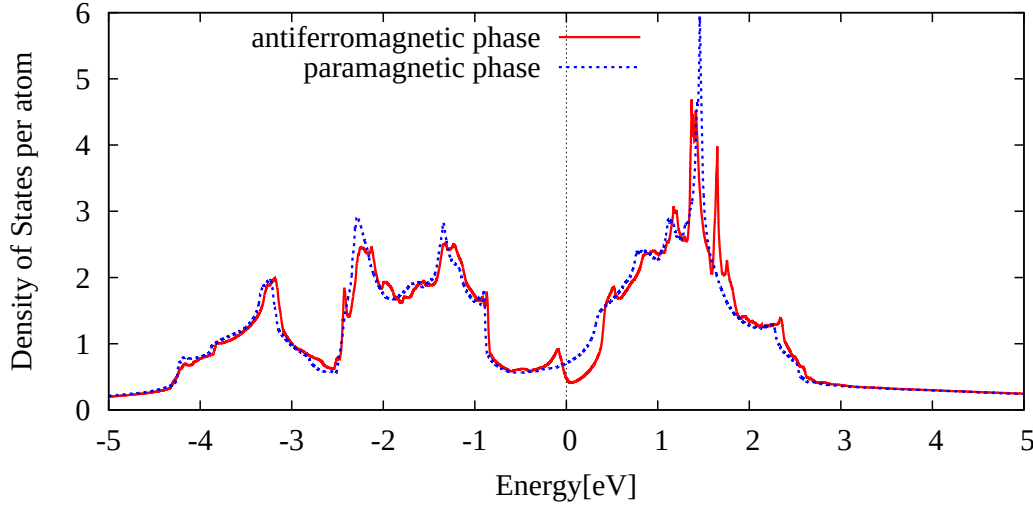


Fig. 4.2 The density of states in the antiferromagnetic phase and paramagnetic phase of Cr. Zero energy corresponds to the Fermi energy.

$\psi_{\mu,m;\epsilon}(\mathbf{k}_0, \sigma)$. In the self-consistent calculation, we use $50 \times 50 \times 50$ meshes in the first BZ of the bcc lattice. The chemical potential is determined so that the number of electrons should be 6.0 per site, i.e., the number of the valence electrons in Cr.

The values of U and J in Eq. (2.2) are unknown. Considering that the magnetic moment defined in Eq. (3.1) is sensitive to the choice of U and J , we determine their value so that the calculated magnetic moment should be close to the observed one, $0.6 \mu_B$, in commensurate Cr [7]. In our calculation, we neglect the on-site interaction of $4s$ orbital. We obtain $U = 2.5$ eV and $J = 0.1U$. Note that there are several possible values of U and J , but the conclusions presented in this chapter are independent of the choice of U and J . We find that the calculated magnetic moment, $0.6 \mu_B$, is composed of $0.48 \mu_B$ in t_{2g} orbitals and $0.12 \mu_B$ in e_g orbitals.

The electronic state in the antiferromagnetic state calculated by our method are consistent with previous results of the band structure [103], the Fermi surface [104], and the density of states [105, 106]. Figure 4.2 shows the density of states in the antiferromagnetic phase and paramagnetic phase of Cr derived by tetrahedron technique [112]. The SDW gap opens at the Fermi energy. Figure 4.3 shows the Fermi surface in the antiferromagnetic phase at $k_z = 0$. The antiferromagnetic ordering reconstructs the original Fermi surface shown in Fig. 4.1.

4.2 Optical Conductivity

Figure 4.4 shows the optical conductivity driven from Kubo formula (2.36) in the paramagnetic and the antiferromagnetic phases of Cr. We treat the direct interband transition. The Drude component is not included in our calculation. The optical conductivity in the paramagnetic phase is broad, from 0 eV to 2 eV, and has no characteristic structure. On the other hand, a remarkable peak appears in the antiferromagnetic phase around 0.55 eV. The peak is derived from the dip in the density of states for antiferromagnetic Cr.

Such a peak in the antiferromagnetic phase has also been shown in experiment [113, 114]. However, the peak position in the experiment of pure Cr appears around 0.1 eV, which is much smaller than our calculated result. This is because we treat the ordered

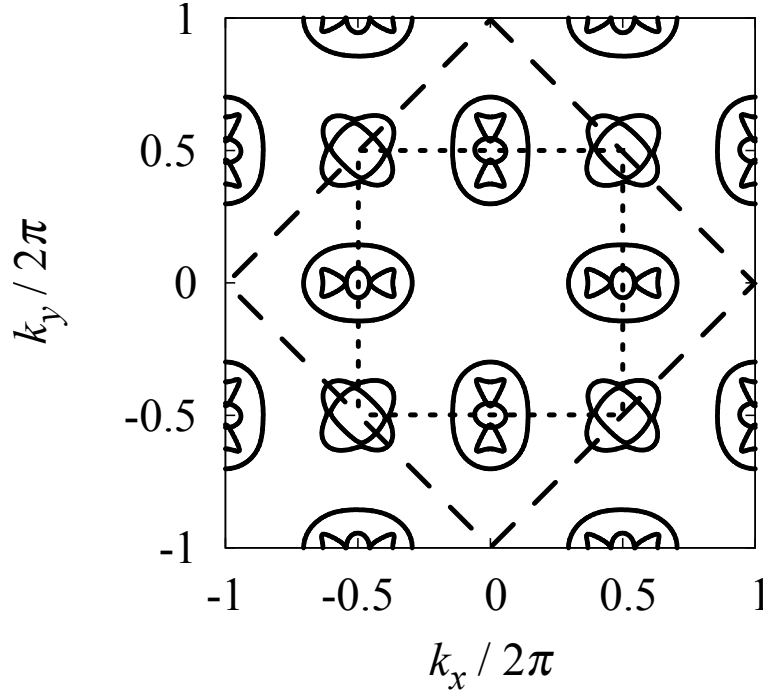


Fig. 4.3 Fermi surface of the commensurate antiferromagnetic Cr at $k_z = 0$. Dashed lines indicate the Brillouin zone ascribed to the original unit cell, while dotted lines indicate reduced Brillouin zone originating from magnetic unit cell. The lattice constant a is set to be unity.

state of chromium as a commensurate antiferromagnetic state, and calculate the band structures in the mean-field approximation, both of which neglect the effect of band renormalization due to correlation. These treatments may lead to a larger band gap or to a shift of the peak position. Thus, we should see the data of commensurate Cr alloy, even though our hopping integrals are obtained for pure Cr.

The energy is roughly consistent with an observed peak position (~ 0.45 eV) of the optical conductivity for commensurate Cr alloys [113, 114], although the calculated one shows a slightly larger value. The 0.1 eV overestimate of the peak position in our theory as compared with the experimental value may come from neglecting the effect of band renormalization due to correlation. This amounts to a factor of $3/4$ for the band renormalization: The energy scale in our theory should be multiplied by $3/4$ to make a comparison with experiment. The same factor is obtained from the comparison of the peak positions of the optical conductivity in the paramagnetic phase between theory (1.2 eV) shown in Fig. 4.4 and experiment (0.9 eV) [115].

4.3 Spin and Charge Excitations in Commensurate Spin-Density Wave State

Figure 4.5(a) shows the imaginary part of the transverse spin susceptibility, $\text{Im} \chi^{+-}(\mathbf{q}, \omega)$ given in Eq. (2.69), in the commensurate SDW state of Cr from $\mathbf{q} = (0, 0, 0)$ to $(2\pi, 0, 0)$. Here, the lattice constant a is taken to be a unit length. We note that this $\mathbf{q}$ range corresponds to a range from $\mathbf{q} = 2\pi(-1, 1, 0)$ to $2\pi(0, 1, 0)$. In Fig. 4.5(a), there appears dispersion with strong intensity starting from the magnetic

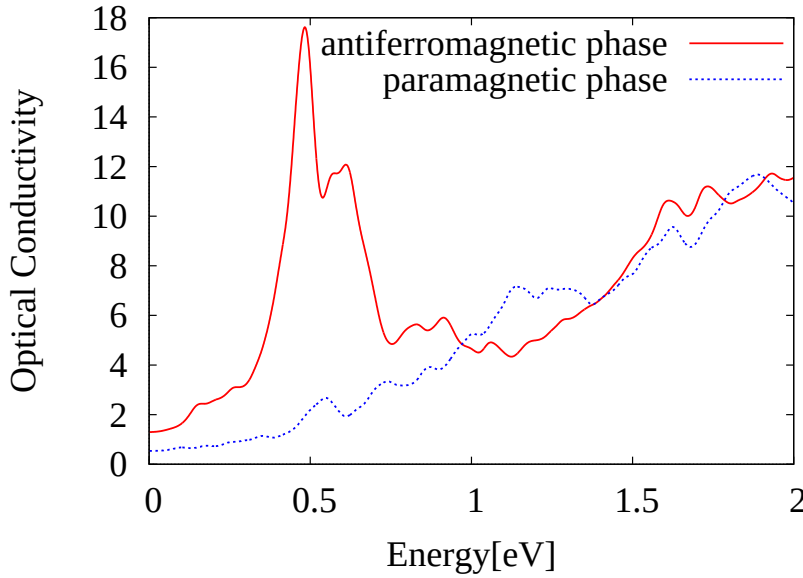


Fig. 4.4 Interband contribution to the real part of the optical conductivity in antiferromagnetism (red solid line) and paramagnetism (blue dotted line). The conductivity is calculated along the x direction. The Drude component is not included.

zone center $\mathbf{q} = (2\pi, 0, 0)$, corresponding to a collective spin-wave excitation. The collective excitation exhibits a prominent weight up to around 0.6 eV, but gradually loses its sharpness and weight above 0.6 eV. A possible cause of this damping is individual particle-hole excitations. To investigate this, we plot the particle-hole excitation spectrum, i.e., the imaginary part of the bare transverse spin susceptibility, $\text{Im}\chi_0^{+-}(\mathbf{q}, \omega)$, in Fig. 4.5(c). We find that the collective excitation gradually penetrates into the particle-hole continuum around 0.6 eV. Therefore, it is clear that the damping is caused by the particle-hole excitations. A similar origin of damping has been discussed for iron arsenides based on the same calculation scheme [82].

The spectral weight distribution and dispersive feature in Fig. 4.5(c) come from the band structure in the commensurate SDW phase. Therefore, the threshold energy (~ 0.5 eV) of particle-hole excitation at $\mathbf{q} = (0, 0, 0)$ may be seen through the optical process. In fact, the calculated peak of the optical conductivity induced by the commensurate SDW order discussed in Sec. 4.2 is located at 0.55 eV, similar to the threshold energy.

The longitudinal spin excitation $\text{Im}\chi^z(\mathbf{q}, \omega)$ given in Eq. (2.70) is shown in Fig. 4.5(b). The strong spectral weight is located above 0.4 eV near $\mathbf{q} = (2\pi, 0, 0)$. We find that the main part of the weight is distributed above the collective spin-wave energy shown in Fig. 4.5(a). This means that, if the experimental setup of inelastic neutron scattering is properly chosen to detect both the transverse and longitudinal excitations, a broad and less dispersive spectral distribution is expected near the magnetic zone center up to 1 eV. By taking into account that the magnetic component parallel to the scattering vector does not contribute to the scattering, such kind of setup may be achieved for both a longitudinal SDW (L-SDW) phase below 122 K and a transvers SDW (T-SDW) phase between 122 K and 311 K in Cr [6], assuming that the high-energy excitations in the incommensurate phases are similar to those in the commensurate phase: The momentum transfer $\mathbf{q}$ for L-SDW should be set to

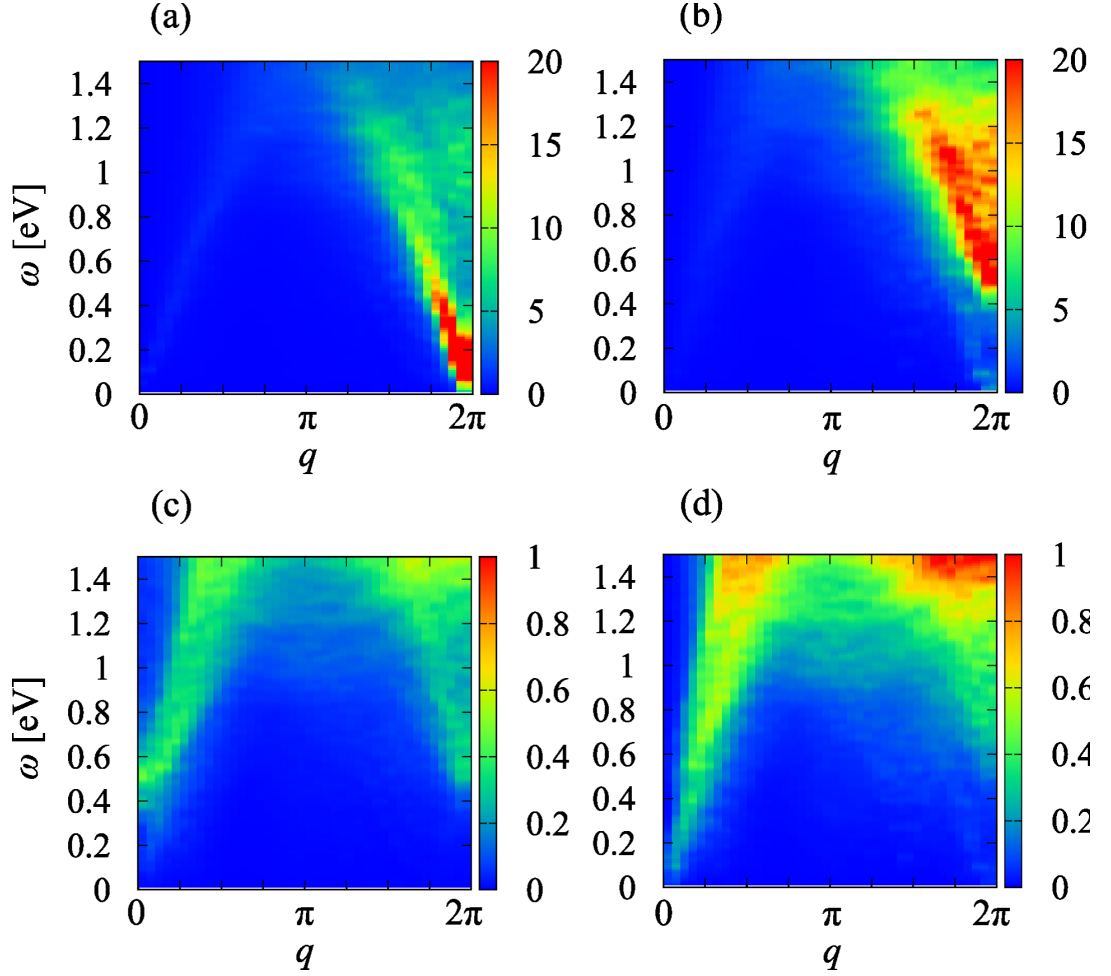


Fig. 4.5 Contour plot of the imaginary part of spin susceptibility in the commensurate SDW state along the $(q, 0, 0)$ direction. (a) $\text{Im}\chi^{+-}$ (transverse) and (b) $\text{Im}\chi^z$ (longitudinal). (c) $\text{Im}\chi_0^{+-}$ (transverse) and (d) $\text{Im}\chi_0^z$ (longitudinal) for the bare susceptibility. The magnitude of intensity shown by the color bar in (a) is limited up to 20, but the maximum intensity is 460 at around $q = 2\pi$ and $\omega \sim 0.1$ eV. We use $40 \times 20 \times 20$ meshes in the first BZ of the bcc lattice.

near $(0, 2\pi, 0)$, while $\mathbf{q}$ for T-SDW should be set to near $(2\pi, 0, 0)$ or $(0, 2\pi, 0)$. The experimental confirmation of these features is highly desired.

The difference of the spectral properties between the transverse and longitudinal excitations naturally comes from the difference of the bare spin susceptibility as shown in Figs. 4.5(c) and 4.5(d): The spectral weight of $\text{Im}\chi_0^z$ [Fig. 4.5(d)] near $\mathbf{q} = (2\pi, 0, 0)$ is distributed higher in energy than that of $\text{Im}\chi_0^{+-}$ [Fig. 4.5(c)].

To see the detailed structures of gapless excitations, we investigate the spectra around the Γ point, where the spectral intensity is moderately weak enough to see the whole structure clearly. We also note that small $\mathbf{q}$ region contributes to L -edge RIXS spectra as discussed in Sec. 4.4. The imaginary part of χ^{+-} , χ^z , and χ^n near the Γ point are shown in Figs. 4.6(a), 4.6(b), and 4.6(c), respectively. χ^n is given in Eq. (2.71). The intensity of the transverse mode, $\text{Im}\chi^{+-}$, is larger than that of the longitudinal mode, $\text{Im}\chi^z$, below 0.5 eV. The velocity of the transverse mode v_s is estimated to be 1.8 eV $\text{\AA}$. This value is consistent with a theoretically estimated value based on a band gap (~ 1.5 eV $\text{\AA}$) [79, 107]. We notice that v_s is smaller

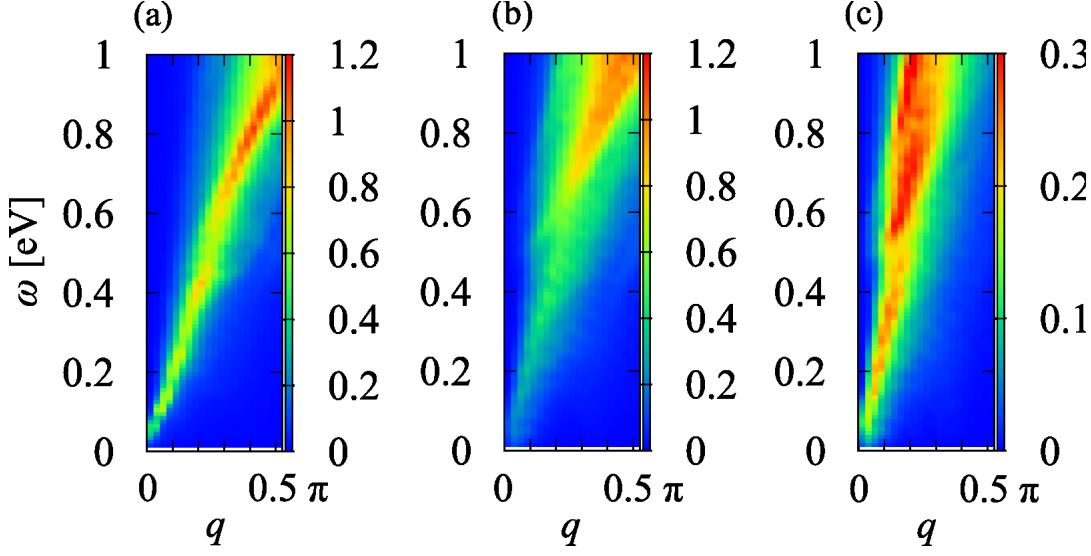


Fig. 4.6 Contour plot of the imaginary part of dynamical susceptibility near the BZ center. (a) Transverse spin mode, $\text{Im}\chi^{+-}$, (b) longitudinal spin mode, $\text{Im}\chi^z$, and (c) charge mode, $\text{Im}\chi^n$ along the $(q, 0, 0)$ direction. The magnitude of intensity is shown by the color bar on the right of each panel. We use $80 \times 20 \times 20$ meshes in the first BZ of the bcc lattice.

than that of the charge mode. This is expected from the large U limit, where the exchange interaction determining the spin-wave velocity, $t^2/(U - J)$, is smaller than the hopping integral t which determines the energy scale of charge motion.

4.4 Resonant Inelastic X-ray Scattering spectra

4.4.1 Scattering geometry

The geometry for our RIXS calculation is illustrated in Fig. 4.7. The angle between the momenta of incoming ($\mathbf{k}_{\text{in}}$) and outgoing ($\mathbf{k}_{\text{out}}$) x-ray is α , and the norms of these momenta are almost the same. In this geometry, the momentum transfer $\mathbf{q} = \mathbf{k}_{\text{in}} - \mathbf{k}_{\text{out}}$ is on the x -axis, i.e., $\mathbf{q} = (q, 0, 0)$. We change $\mathbf{q}$ by varying the angle α . The incoming beam is either π -polarized (parallel to the scattering plane: $\boldsymbol{\pi}_{\text{in}}$) or σ -polarized (perpendicular to the scattering plane: $\boldsymbol{\sigma}_{\text{in}}$). The outgoing X-ray is chosen to be a summation of both polarizations $\boldsymbol{\pi}_{\text{out}}$ and $\boldsymbol{\sigma}_{\text{out}}$, taking into account existing experimental conditions. The spin is assumed to direct parallel to the z -axis, which is perpendicular to the ordering vector $\mathbf{Q}$. This geometry corresponds to one of two spin polarizations in the T-SDW phase between 122 K and 311 K in the incommensurate chromium [6]. We note that, since a characteristic x-ray energy for Cr L_3 absorption is 572 eV, the momentum space where Cr L_3 -edge RIXS can access is limited to $q \lesssim 0.53\pi$.

4.4.2 Calculated RIXS spectra

The calculated RIXS spectra (2.127) for the commensurate SDW state are plotted in Fig. 4.8 for both the π and σ polarizations along the $(q, 0, 0)$ direction. The spectra are symmetric with respect to $q = 0$. This is in contrast with the case of iron arsenides [83], where asymmetric spectral distribution appears. Such a contrasting

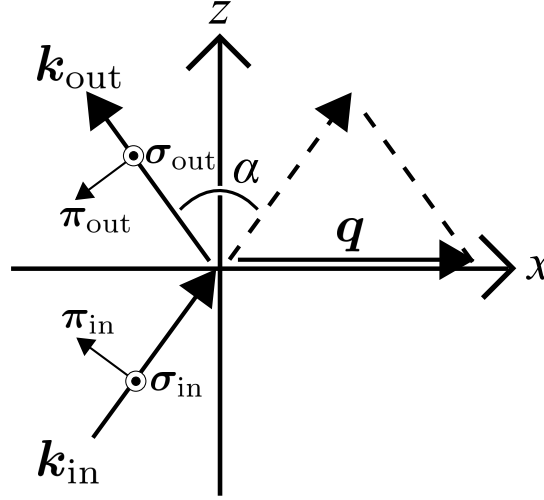


Fig. 4.7 The geometry of the scattering process used in the RIXS calculation. $\mathbf{q} = \mathbf{k}_{\text{in}} - \mathbf{k}_{\text{out}}$. The scattering plane is the xz -plane, and $\mathbf{q} = (q, 0, 0)$. We change q by varying the angle α between $\mathbf{k}_{\text{in}}$ and $\mathbf{k}_{\text{out}}$. σ_{in} (σ_{out}) and π_{in} (π_{out}) denote σ - and π -polarization vectors, respectively, of incoming (outgoing) x-ray.

behavior is originated from the difference of geometry for the scattering process: The angle between $\mathbf{k}_{\text{in}}$ and $\mathbf{k}_{\text{out}}$ in iron arsenides is fixed unlike the geometry in Fig. 4.7. The high-energy part of the spectra above 1 eV is shown in Figs. 4.8(a) and 4.8(b). The change of incident-photon polarization induces the shift of the intense spectral cloud at high energy ~ 2 eV from $q = \pm 0.34\pi$ for the π polarization to $\pm 0.5\pi$ for the σ polarization in the present setup of the scattering geometry.

The intensity of the spectra increases with increasing energy and shows a broad maximum at $\omega \sim 2$ eV. With further increasing energy, the intensity decreases. This behavior is common to other types of scattering geometry. The main contribution to the broad peak comes from interband excitations between different orbitals as discussed below. The broad peak at $\omega \sim 2$ eV is expected to appear in RIXS experiments for Cr.

The low-energy region below $\omega = 1$ eV is shown in Figs. 4.8(c) and 4.8(d). We note that the elastic line at $\mathbf{q} = (0, 0, 0)$ is omitted from the figures. According to the dynamical spin susceptibility shown in Fig. 4.6, we expect low-energy spin and charge excitations appearing from $\omega = 0$ eV at $\mathbf{q} = (0, 0, 0)$. However, the presence of high-intensity excitations in the high energy makes the low-energy spectra less visible in Figs. 4.8(c) and 4.8(d). The present scale of intensity in Figs. 4.8(c) and 4.8(d) makes the excitations less visible because of the presence of high intensity in the high-energy excitations. However, the low-energy excitations certainly exist even though their intensity is small.

In order to make clear the presence of the low-energy excitations indicated by Fig. 4.6, we show the line shape of the RIXS spectra for both polarizations in Fig. 4.9. We find dispersive low-energy excitations, starting from near $\omega = 0$ eV and $\mathbf{q} = (0, 0, 0)$, with the same energy scale as the spin and charge modes shown in Fig. 4.6, although it is difficult to identify each mode.

One may have a question why the spin and charge modes are less clear in the RIXS spectra. In order to resolve this question, we should notice a difference in the suffixes of the dynamical susceptibility between RIXS in Eq. (2.127) and, for example, the transverse spin mode in Eq. (2.69). In Eq. (2.69), only $\kappa = \lambda$ and $\mu = \nu$, i.e.,

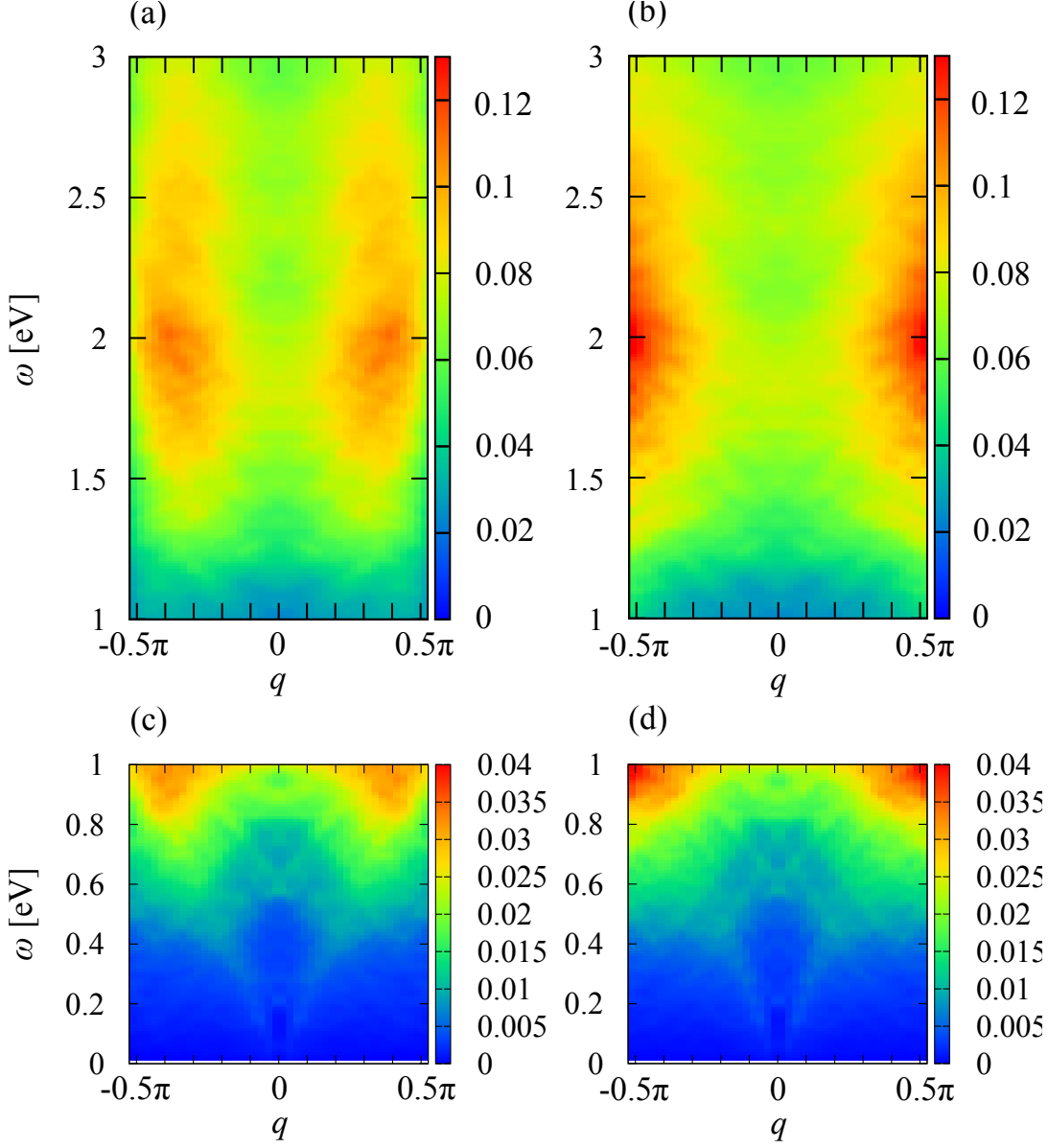


Fig. 4.8 Contour plot of the calculated RIXS spectra of chromium for the commensurate SDW state along the $(q, 0, 0)$ direction. High-energy region ($1 \text{ eV} \leq \omega \leq 3 \text{ eV}$): (a) π polarization and (b) σ polarization. Low-energy region ($0 \text{ eV} \leq \omega \leq 1 \text{ eV}$): (c) π polarization and (d) σ polarization.

excitations within the same orbital, are taken from the dynamical susceptibility in Eq. (2.68), while in Eq. (2.127) κ , λ , μ , and ν are independently taken. In order to see the effect of the difference, we calculate the RIXS spectra only taking $\kappa = \lambda$ and $\mu = \nu$ in Eq. (2.127) and showing the results in Fig. 4.10. We find that the spectral weight near 1 eV is dramatically suppressed and the low-energy modes become clear in intensity. This means that the RIXS intensity near 1 eV in Fig. 4.8 is dominated by excitations among different orbitals, i.e., off-diagonal orbital excitations. Thus we conclude that the low-energy collective modes such as spin-wave excitation are overwhelmed by the off-diagonal orbital excitations. We note that this does not happen in the case of a single-band Hubbard model describing cuprate compounds.

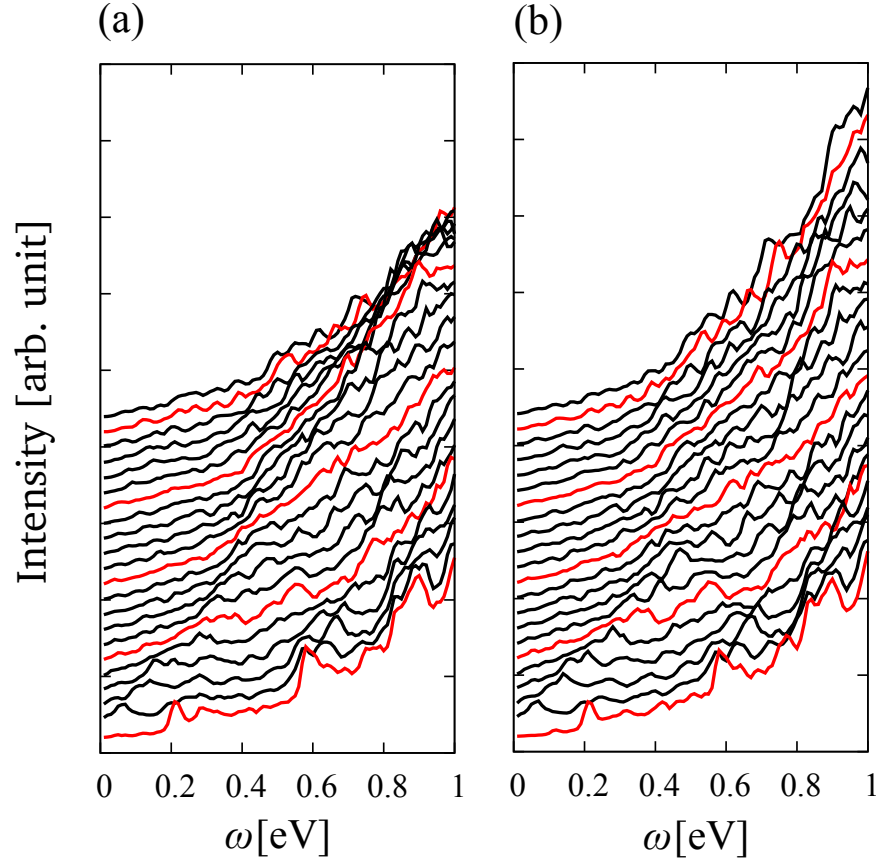


Fig. 4.9 The line shape of RIXS spectra of chromium in the commensurate SDW state for (a) π - and (b) σ -polarization. Each line is plotted for momentum transfer between $(0,0,0)$ - $(21\pi/35, 0, 0)$ from bottom to top with the increments of $\pi/35$.

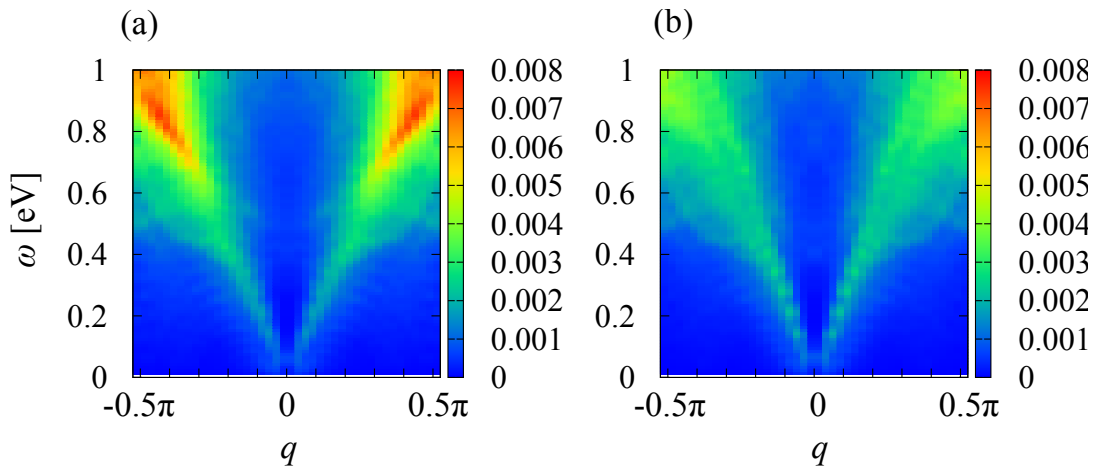


Fig. 4.10 Contour plot of the RIXS spectra obtained by restricting the orbital indices in the dynamical susceptibility to only $\kappa = \lambda$ and $\mu = \nu$ in Eq. (2.68). (a) the π polarization and (b) the σ polarization along the $(q, 0, 0)$ direction.

In order to make clear the possibility of observing the spin-wave excitation, we comment on the contributions from other excitations. The energy scale of the spin-wave excitation that we want to detect is around 0.1-0.4 eV, which is higher than that of phonons [116, 117]. Even though multi-phonon excitations can be seen in RIXS [118, 117, 119], the phonons do not overwhelm the spin excitations because of weak electron-phonon coupling expected in Cr. On the other hand, elastic scattering may overlap with the spin-wave excitations. However, if the energy resolution of RIXS is improved, the overlap becomes small and the spin-wave excitations may become detectable, although the intensity of the spin-wave excitations is weak due to the off-diagonal orbital contributions as discussed above.

Finally, we comment on a peak structure around 0.6 eV near $q = 0$ in Fig. 4.10(a). Since the energy position is almost equal to a peak seen in the optical conductivity in Sec. 4.2, we judge the peak to be due to the SDW-gap formation. We also expect that this structure may be detected by RIXS with high resolution.

4.5 Summary

We have investigated the spin dynamics and RIXS for Cr with the commensurate SDW state by using a multi-band Hubbard model composed of $3d$ and $4s$ orbitals. We have employed a self-consistent calculation of the ground state based on the SDW mean-field approximation. On top of the ground state, we have calculated the dynamical spin susceptibility within RPA. By evaluating the hopping integrals from the down-folding procedure of first-principles band calculation, the electronic states in Cr are properly described by our model. The electronic states in the commensurate SDW phase obtained by the mean-field theory are also consistent with those from the previous first-principles band structure calculations. Therefore, the calculated dynamical susceptibilities reflect realistic particle-hole excitations. It is well-known that RPA cannot describe the band renormalization effect due to correlation. The factor of the band renormalization is estimated to be roughly $3/4$, by comparing the peak positions of the optical conductivity between theory and experiment.

We have found that a collective spin-wave mode appears in the spin-transvers excitation spectrum. The collective mode does not damp up to ~ 0.6 eV. Above the energy, the excitation overlaps individual particle-hole excitations, leading to broad spectral weight. We expect that this feature appears in inelastic neutron scattering experiments for Cr, even though Cr shows incommensurate spin excitations below 0.1 eV. When the contribution from the longitudinal spin excitation to inelastic neutron scattering increases, the spectral weight above the spin-wave mode is expected to be enhanced. This may also be detected by future experiments.

RIXS tuned for the L -edge is a powerful tool to investigate not only the charge and orbital excitations but also the spin excitation in the energy and momentum spaces. By using a fast-collision approximation for the RIXS process, we have calculated Cr L_3 -edge RIXS intensity. We have found large spectral weight coming from interband excitations between different orbitals. This eventually masks collective spin-wave excitations within the same orbital, weakening the intensity of them. Weak as the intensity is, it may be possible to detect them if the experimental resolution of RIXS is high enough.

Finally, we would like to refer that experiments of RIXS on Cr is now being carried out at SPring-8 by Harada and Hiraka, based on our prediction [120].

Chapter 5

CONCLUSIONS

Pure chromium (Cr) is a typical material of the metal with spin-density-wave (SDW) ground state. The SDW is caused by the nesting of its Fermi surfaces. Its magnetic and electronic properties have been well investigated experimentally and theoretically for a long time. However, taking the opportunity of the discovery of iron-based superconductors, whose parent compounds have SDW ground state, multi-orbital nature in $3d$ -transition metals is newly focused on. With regard to the origin of the superconductivity in iron-based superconductors, it is now considered that the orbital degrees of freedom in $3d$ -transition metals play important roles in their physical properties. We have tried to shed light on the physics, especially electron dynamics, of $3d$ -transition metals with multi-orbital nature in the SDW state.

In order to understand the electron dynamics of multi-orbital antiferromagnetic metals, we have investigated iron arsenide and Cr. For iron arsenide, we have elucidated the anisotropy of resistivity in stripe-ordered antiferromagnetic phase. For Cr, we have calculated the dynamical susceptibilities and resonant inelastic x-ray scattering (RIXS) spectra. To obtain the SDW ground state of these materials, we have made use of multi-orbital Hubbard models with a mean-field approximation. These results are summarized below for completion of this thesis.

First, we have investigated the in-plane anisotropy of resistivity with a multiorbital Hubbard model in the stripe-ordered antiferromagnetic phase. The resistivity has been obtained from a memory-function approach. Both the Drude weight and the scattering rate tend to make resistivity along the ferromagnetic direction larger than that of antiferromagnetic direction in parent compound, which is consistent with in-plane anisotropy in experiments. When holes are introduced, the anisotropy of the scattering rate reverses and becomes independent of carrier concentration, which is related to the disappearance of electron pockets. On the other hand, the Drude weight monotonically decreases as a consequence of deformation of the Dirac pocket with hole doping. This eventually leads to the reverse of anisotropy of resistivity, which is also consistent with experiments. These results indicate that the origin of the anisotropy is attributed to the interplay of impurity scattering and anisotropic electronic states, both of which are strongly influenced by the topology of Fermi surface. In addition, we have successfully obtained the anisotropy of resistivity consistent with experiments. These results suggest the importance of impurity scattering in the anisotropy of resistivity of iron arsenide.

Second, we have investigated the spin dynamics and RIXS spectra for Cr with the commensurate spin-density wave (SDW) state. By evaluating the hopping integrals from the down-folding procedure of first-principles band calculation, the electronic states in Cr are properly described by our model, which is confirmed by calculation of optical conductivity. We have calculated the dynamical spin susceptibility within

random-phase approximation. We have calculated the dynamical spin susceptibility within random-phase approximation. We have obtained a collective spin-wave mode appearing in the spin-transvers excitation spectra. The collective mode does not damp up to ~ 0.6 eV. Above the energy, the excitation overlaps individual particle-hole excitations, leading to broad spectral weight. We expect that this feature appears in inelastic neutron scattering experiments for Cr, even though Cr shows an incommensurate spin excitations below 0.1 eV. When the contribution from the longitudinal spin excitation to inelastic neutron scattering increases, the spectral weight above the spin-wave mode is expected to be enhanced. This may also be detected by future experiments. By using a fast-collision approximation for the RIXS process, we have calculated Cr L_3 -edge RIXS intensity. We have found large spectral weight coming from interband excitations between different orbitals. This eventually masks collective spin-wave excitations within the same orbital. Even though the excitations are weak in intensity, it may be possible to detect them if the experimental resolution of RIXS is high enough.

Our results promise the importance of multi-orbital properties to research of condensed matter physics. The study of multi-orbital physics in correlated electron systems should be pushed forward strongly.

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