Inelastic x-ray scattering study of plasmons in liquid alkali metals

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Overview of thesis:
Inelastic x-ray scattering study of plasmons in liquid alkali metals

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In this work, we study plasmon behaviors in liquid metals. Inelastic x-ray scattering (IXS) techniques are applied to liquid alkali metals, and the plasmon excitation energies (dispersion relations) and the line widths are determined as a function of the momentum transfer $q$. It is found for the first time that the $q$ dependences of these properties in the liquid state are different from those in the solid state. We discuss this difference, considering the structural variation upon melting. In particular, we derive a formula for evaluating the plasmon line width in liquid metals at $q=0$, and reproduce the experimental results. This study provides useful information on the relation between electronic state and the disordered ionic structure in liquid metals. The details of this work are as follows.

【Background】
The relation between the electronic states and the ionic structure in liquid metals is less clearly understood than that in the crystalline systems, because of the structural disorder of ions in the liquid state. Behaviors of plasmon, a collective excitation of electrons, provide useful information on how the valence electrons are influenced by the ions in liquid metals because the excitation energy and the line width of plasmon strongly reflect the electron-ion interaction. The aim of this study is to clarify how the plasmon behavior is modified by the structural variation upon melting.

【Experimental】
IXS experiments are performed for liquid Rb and Cs near the melting point. For Rb, the experiments are also done in the solid state. A sample cell made of sapphire, which is designed for x-ray scattering experiments, is used to contain the liquid sample. The IXS measurements are carried out at the Taiwan inelastic x-ray scattering beamline BL12XU of SPring-8. The range of $q$ is from $0.18 – 0.90 \, \text{Å}^{-1}$ for solid Rb, from $0.18 – 0.80 \, \text{Å}^{-1}$ for liquid Rb, and from $0.32 – 0.55 \, \text{Å}^{-1}$ for liquid Cs.

【Formulation for the plasmon line width in liquid metals】
We derive a formula for evaluating the plasmon line width at $q = 0$, $\Delta E_{1/2}(0)$. 
The formula is based on the nearly free electron model and is written as,

\[
\Delta E_{1/2}(0) = \frac{\hbar \omega_p}{3n_{\text{ion}}(\hbar \omega_p / E_F)^4} \int \frac{d^3q'}{(2\pi)^3} \left( \frac{q'}{k_F} \right)^4 \left| \frac{v_{\text{ion}}(q')}{E_F} \right|^2 S(q') \text{Im} \varepsilon_{\text{EG}}(q', \omega_p),
\]

where \( \omega_p \) is the plasma frequency, \( n_{\text{ion}} \) is the ionic density, \( E_F \) is the Fermi energy, \( k_F \) is the Fermi wave number, \( v_{\text{ion}} \) is the screened pseudopotential, \( S(q) \) is the liquid structure factor, and \( \varepsilon_{\text{EG}} \) is the dielectric function of the electron gas. We apply this formula to liquid alkali metals and find that the line widths of liquid alkali metals are narrower than those of solid alkali metals. We show that this narrowing originates from the vanishing of crystalline structure upon melting.

**[Plasmons in liquid Rb and Cs]**

The plasmon dispersion relations of solid and liquid Rb, and of liquid Cs are derived from the IXS spectra. We find that the plasmon dispersions of liquid Rb and Cs are different from those of solid Rb and Cs. For solid Cs, the previously reported plasmon dispersion is used for comparison.

The plasmon dispersion is defined as \( \varepsilon (q, \omega) = 0 \) (\( \varepsilon \) is the dielectric function), thus the variation in the plasmon dispersion upon melting can be studied from the difference of \( \varepsilon \) between the solid and liquid phases. Since \( \varepsilon \) reflects the interband transitions, the electronic density of states (DOS) is useful information. With the aid of the theoretical DOS in the solid and the liquid state, we qualitatively explain the variation in the plasmon dispersion upon melting, and it is suggested that the effect of ions on the dispersion relation is smaller in the liquid state than in the solid state.

The line widths of solid and liquid Rb are also derived from the IXS spectra, and it is found that the line width becomes narrower upon melting. This narrowing is quantitatively reproduced by the formula described above, from which we interpret that the effect of the ions on the plasmon damping in Rb becomes less important with the structural variation upon melting.

**[Conclusion]**

The observed variations in plasmon excitation energy and the line width suggest that the effect of ions decreases upon melting. This study, especially the formula of \( \Delta E_{1/2}(0) \) for liquid metals, provides valuable insight into how the electronic states are related to the ionic structure in liquid metals. This insight will be useful to clarify the electronic states in various disordered systems, such as expanded fluid metals and metallic glasses.