Central Peak in the Tunneling Model and its Extended Model

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Linear coupling of soft modes to thermal diffusion mode is one of the plausible origins of central peak observed experimentally in the structural phase transitions. In fact, in the low temperature phase of the structural phase transition or the pyroelectric crystals where the spontaneous polarization exists, the soft mode with zero-wave vector is easily shown to be coupled to the thermal diffusion mode and the intensity ratio of the central peak and the soft mode is given by the similar relation as Landau-Placzek formula in liquid-gas transition;

$$\frac{I\ (\mathrm{central})}{I\ (\mathrm{soft})} = \frac{C_E - C_X}{C_X} = \frac{X_T - X_S}{X_S} \tag{1}$$

Here C_X and C_E are the specific heat at constant order parameter and at constanct conjugate field, X_T and X_S are corresponding isothermal and adiabatic suspectibility, respectively. On the other hand, above the transition point, the soft mode with zero-wave vector cannot be coupled to the thermal diffusion mode while the soft mode with nonzero wave-vector can be coupled, but the intensity of the resultant central peak is too weak for small wave vector.

In this report the hydrodynamic approach due to Mori is applied to investigate the effect of this coupling to the extended tunneling model and it is shown that in piezoelectric crystals a shrap central peak is possible to observe even above the transition point since this coupling strength is enhanced on account of critical fluctuations near the transition point. Model hamiltonian is

$$H = H_P + H_L + H_{P-L}$$

$$\equiv -2\Omega S_0^z - \frac{1}{2N}\sum_k J_k S_k^x S_{-k}^x + \frac{1}{2}\sum_k (P_k P_{-k} + \omega_k^2 Q_k Q_{-k})$$

$$+\frac{1}{\sqrt{N}}\sum_{k}F_k Q_k S^x_{-k} + \frac{1}{2N}\sum_{p,k}V(p,k)\,Q_p Q_k S^x_{-p-k}$$

$$+\frac{1}{2N}\sum_{p,k}W(p,k)\,S^x_p S^x_k Q_{-p-k}. \tag{2}$$

Macroscopic dynamical variables are chosen to be

$$\{\widetilde{A}^n_q\} \equiv \widetilde{S}^x_q,\ \widetilde{S}^y_q,\ \widetilde{Q}_q,\ \widetilde{P}_q,\ \widetilde{H}_q\ . \tag{3}$$

These variables are constructed from original ones $\{A^n_q\}$ by means of orthonormalization of Schmidt;

$$\widetilde{S}^x_q = S^x_q/\sqrt{\chi^{xx}},\qquad \widetilde{S}^y_q = S^y_q/\sqrt{\chi^{yy}},$$

$$\widetilde{P}_q = P_q/\sqrt{\chi^{pp}},\qquad \widetilde{Q}_q = \frac{1}{\sqrt{n_Q}}\left\{Q_q - \frac{\chi^{Qx}}{\chi^{xx}}S^x_q\right\},$$

$$\widetilde{H}_q = \frac{1}{\sqrt{n_H}}\left\{H_q - \frac{\chi^{Hx}}{\chi^{xx}}S^x_q - \frac{1}{n_Q}\left(\chi^{HQ} - \frac{\chi^{Hx}\chi^{xQ}}{\chi^{xx}}\right)\left(Q_q - \frac{\chi^{Qx}}{\chi^{xx}}S^x_q\right)\right\}. \tag{4}$$

Here X^{nm} are susceptibilities. Equations of motion for these variables are

$$\frac{1}{dt}\widetilde{A}^n_q = -i\,C^{nm}(q)\,\widetilde{A}^m_q + R^n_q\ (\text{random force}). \tag{5}$$

with the use of those coefficients $C^{nm}_{(q)}$ and the damping function, we can obtain the response function in the form;

$$\chi^{xx}(q,\omega) = \frac{\omega_s(q)^2\,\chi^{xx}(q,0)}{\underbrace{\omega_s(q)^2-\omega^2-i\omega\Gamma_s(q,\omega)}_{(s)} - \underbrace{\frac{|C^{yQ}|^2(\omega^2+i\omega\Gamma_a(q,\omega))}{\omega_a^2-\omega^2-i\omega\Gamma_a(q,\omega)}}_{(a)} - \underbrace{\frac{|C^{yH}|^2 i\omega}{\Gamma_H - i\omega}}_{(t)}}. \tag{6}$$

Here $\omega_s(q)$ is the soft mode frequency which is easily evaluated as

$$\omega_s(q) \equiv C^{xy} = \sqrt{\frac{2\Omega\langle S^z\rangle N}{\chi^{xx}}}\ \xrightarrow{q\to 0}\ \omega_s(0) \propto |T-T_c|^{1/2}. \tag{7}$$

We see in this equation that the soft mode (s) is coupled to the acoustic mode (a) and thermal diffusion mode (t). Our main concern is the coupling coefficient $|C^{yH}|^2$ whose expression is obtained as

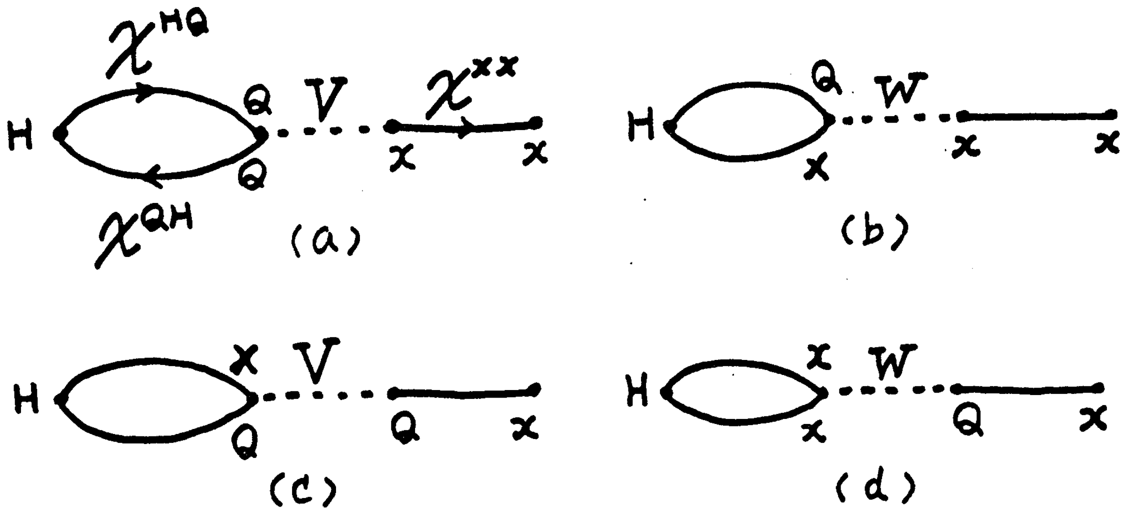
$$C^{yH}(q) = \frac{-i}{\sqrt{n_H\chi^{yy}}}\left[\frac{N\langle S^z\rangle}{\chi^{xx}}\left\{\chi^{Hx} - \frac{1}{n_Q}\left(\chi^{HQ} - \frac{\chi^{Hx}\chi^{xQ}}{\chi^{xx}}\right)\right\}\right.$$

$$\left. -\frac{1}{2N}\sum_p (J_{p+q}-J_p)\langle S^z_{-p}S^x_p\rangle + \frac{1}{\sqrt{N}}\sum_p (F_{p+q}-F_p)\langle Q_p S^z_{-p}\rangle\right], \tag{8}$$

which is shown to reduce in the lowest order in q to the following form;

$$C^{yH}(q) \simeq \frac{i}{\sqrt{CT}} \frac{\sqrt{2\Omega < S_0^z >}}{\chi^{xx}(q)} \chi^{Hx}(q) , \tag{9}$$

where C is the specific heat. Lowest contribution of $X^{HX}_{(q)}$ with respect to the interactions V and W are given by the following diagrams.



Among these diagrams, the first one (a) gives the most divergent contribution to $X^{HX}_{(q)}$ in piezoelectric case, the expression of which is

$$\chi^{Hx} \simeq \chi^{HL^x} \simeq T\, \chi^{xx}(q) \triangle(0)^4 \sum_k \frac{V(k, q-k)}{\omega_k \omega_{q-k}\, \omega_s(k)^2 \omega_s(q+k)^2} \tag{10}$$

In the vicinity of T_c and for small values of q, we can estimate the Eq. (10) as

$$\chi^{HL^x} \simeq \frac{B_0 q}{\sqrt{2\Omega < S_0^z >}} (\chi^{xx}(q))^{3/2}, \tag{11}$$

where B_0 is a constant. Using the relations

$$\chi^{xx}(q) \propto \frac{1}{\omega_s(q)^2}, \quad \omega_s(q)^2 \approx k_0 \tau + k_1 q^2 \text{ and } \tau \equiv \frac{T - T_c}{T_c} ,$$

we get

$$|C^{Hy}|^2 \begin{cases} \propto q^2/\tau & \text{for } \tau \gg \tau_q \equiv \dfrac{k_1}{k_0} q^2 \\ \simeq \dfrac{B_0 2\Omega < S^z >}{C T k_1} & \text{for } \tau \ll \tau_q \end{cases} \tag{12}$$

For the latter case, the coupling in the vicinity of the transition point tends to a constant which is independent of both small parameters q and τ. Thus, the central peak may be expected to be observable in this region.

The integrated intensity of the central peak is given by

$$I_r = \frac{\pi k T \,|C^{Hy}(q)|^2}{\{\omega_s(q)^2 + |C^{Hy}(q)|^2\}\,\omega_s(q)^2}$$

$$\propto \begin{cases} \dfrac{q^2}{|T - T_c + \triangle T_r(q)| \cdot |T - T_c|^2} & \text{for } \tau \gg \tau_q \\ \dfrac{1}{q^2\,|q^2 + (k_0/k_1)\triangle T_r(0)|} & \text{for } \tau \ll \tau_q \end{cases} \tag{13}$$

with $\Delta Tr(q) \equiv |C^{Hy}(q)|^2/k_0$. On the other hand, in the non-piezoelectric case, i.e., in the absence of the piezoelectric coupling Fq, $C^{Hy}(q)$ is easily shown to have the following form

$$C^{Hy}(q) \simeq (A_0 - A_1\sqrt{\tau})\,q^2 \longrightarrow |C^{Hy}(q)|^2 \propto q^4 \tag{14}$$

Then in this case central peak is impossible to observe for small value of q.

When the dipole interaction is taken into account, its asymetric nature leads to the logarithmic divergence in the piezo-electric case;

$$|C^{Hy}(q)|^2 \propto q^2 \,\ln^2 (\chi^{xx}(q)) \tag{15}$$

In the present study our calculations have been confined to the lowest order in V and W. When we proceed perturbation calculations further, the second order calculation of the selfenergy leads to the velocity change ΔV of the acoustic mode:

$$-\triangle v \propto \tau^{-1/2} \qquad \text{for } \tau \gg \tau_q$$

This shows that this simple perturbation breaks down in the very vicinity of transition point where the velocity of the acoustic mode becomes negative. Then the dynamical scaling approach is appropriate in this region.

We have shown that the coupling coefficient $|C^{Hy}(q)|^2$ is enhanced near and above T_c due to effects of critical fluctuations. Consequently, the coupling becomes independent of q for $\tau \ll \tau_q \equiv (k_1/k_0)\,q^2$ in the piezoelectric case; This would lead to the observable central peak in some cases.

It should be noted that, for zone boundary soft modes such as in $S_rT_iO_3$ the central peak should results from another reason because the concept of the hydrodynamic mode becomes meaningless for such large momentum. In such a case non-linear coupling to hydrodynamic modes or non-linear effects of critical fluctuations, or effects of crystal imperfections would lead to the central peak as have been discussed by several authors.

The detailed calcuations are now in progress and those accounts will be published elsewhere.

Lattice Dynamical Theory of Sinusoidal Antiferroelectricity in Thiourea

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It has been known through an analysis of the observed satellites in X-ray scattering that thiourea has sinusoidal antiferroelectric polarization along c-axis in the phases II–IV. It is also suggested that this sinusoidal structure is caused by the condensation of a soft mode with a special wave length and polarization.

A model is proposed to interpret the existence of such a soft mode. Dipolar and qudrupolar interaction between thiourea molecules are taken into consideration together with simplified short range repulsive interactions and the dynamical matrix for molecular rotational vibration is set up and dispersion relations are derived. It is shown that softening of a mode with a sinusoidal polarization is expected to exist and possible temperature effect may account for the observed behaviour in thiourea.

The detailed calculations will be published in Prog. Theor. Phys..