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Dynamics of Jahn-Teller Phase Transitions

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The dynamic behavior of a system undergoing a cooperative Jahn-Teller structural transition is investigated. Coupling to both acoustic and optic modes are considered. Particular emphasis is placed on the low-frequency response. In addition to the peaks in the dynamic structure factor corresponding to the soft acoustic mode, a central component is obtained, the width of which goes to zero as the transition temperature is approached. Unlike other transitions for which a central component has been observed, the width of the central peak for Jahn-Teller transitions such as that in $TmVO_4$ may be sufficiently wide to allow a direct observation of the narrowing by neutron scattering or other techniques.

I. INTRODUCTION

The cooperative Jahn-Teller phase transitions[1-3] are characterized by a soft acoustic mode. The low-frequency elastic constant of this mode vanishes at the transition point.[2] The soft elastic constant is strongly frequency dependent. Very different results are obtained depending on whether $\omega\tau \gg 1$ or $\omega\tau \ll 1$, where τ is the relaxation time of the electronic levels.[2] The phonons modulate the electron populations of the electronic levels. The difference between the high- and low-frequency response results because at low frequencies the modulations in the electron populations caused by the phonons have time to decay within each period of the wave, whereas at high frequency there is not sufficient time for this to occur. This relaxation process gives rise to a peak at $\omega = 0$ in the dynamic structure factor. The width of the peak is determined by the relaxation τ and vanishes at the transition point. In this paper the dynamic susceptibility is calculated using an equation of motion method. The relaxation time is introduced by adding Bloch-like relaxation terms to the equations of motion for the pseudospin operators describing the doubly degenerate electronic level. The elastic constants for $\omega\tau \ll 1$ are equal to those calculated thermodynamically. The results in both limits $\omega\tau \gg 1$ and $\omega\tau \ll 1$ agree with results previously obtained.[2]

Central peaks have recently been observed by neutron scattering in other materials undergoing structural transitions such as Nb_3Sn and $SrTiO_3$.[4-6] For those transitions the mechanism giving rise to the central peak is, however, quite different. In Nb_3Sn the central peak has been interpreted as due to third-order anharmonic phonon interactions,[5] whereas the mechanism which gives rise to the central peak in $SrTiO_3$ is not yet established.

As a particular example, the calculations will be applied to $TmVO_4$ which has a non-Kramers doubly degenerate crystal-field-split ground-state electronic level well separated from the higher-lying electronic levels.[7] $TmVO_4$ undergoes a second-order (or very nearly second-order) transition at 2.1 °K. In $TmVO_4$ the doubly degenerate level couples strongly with two different symmetry strains and optic-phonon displacements,[8] and the calculations have been carried out for this case. In appropriate limits the model will describe coupling to nondegenerate strains and optic phonons of a single symmetry, as well as the classic model in which the electronic levels couple to doubly degenerate strain and optic-phonon displacements, as in the case of an octahedrally coordinated Jahn–Teller ion.[1]

II. HAMILTONIAN

The model Hamiltonian will be written in the form,[1-3]

$$H = \sum_{nq}\sum_{\alpha=1,2} [\tfrac{1}{2} P_{n\alpha}(q) P_{n\alpha}(-q) + \tfrac{1}{2}\omega_{n\alpha}^2(q) Q_{n\alpha}(q) Q_{n\alpha}(-q)] + \sum_{nq}\sum_{\alpha=1,2} \xi_{n\alpha}(q) Q_{n\alpha}(q)\sigma_\alpha(-q). \quad (1)$$

The first two terms describe noninteracting phonons, while the last term describes the coupling of these phonons with the doubly degenerate electronic level, where $\sigma_\alpha(q)$ is the lattice Fourier transform of the Pauli operators $\sigma_\alpha(l)$ associated with site l. The sum n runs over all phonon modes of a given irreducible representation specified by α. This symmetry is different for acoustic and optic modes. For optic phonons $\xi_{n\alpha}(0)$ is nonzero whereas for acoustic modes $\xi_{n\alpha}(q)$ is linear in q in the small q limit, and its value is direction dependent. In the case of acoustic phonons it is the accompanying strain field $e_\alpha(q)$ which couples in the same way to the electronic levels and has the same symmetry (at $q=0$) as the optic-phonon normal-mode displacements $Q_{n\alpha}(q)$. The coupling to the acoustic phonons may therefore be written, alternatively,[2]

$$\sum_{\alpha q} \eta_\alpha e_\alpha(q)\sigma_\alpha(-q), \quad (2)$$

where η_α is a constant. The strain $e_\alpha(q)$ is a linear combination of the individual strain components $e_{ij}(q)$.

For later reference it will be convenient to introduce the transformation[3,9,10]

$$\hat{Q}_{n\alpha}(q)=Q_{n\alpha}(q)+\gamma_{n\alpha}(q), \quad \hat{P}_{n\alpha}(q)=P_{n\alpha}(q), \tag{3}$$

where

$$\gamma_{n\alpha}(q)=\xi_{n\alpha}(-q)\,\sigma_\alpha(q)/\omega^2_{n\alpha}(q).$$

The Hamiltonian (1) then takes the form

$$H=\sum_{nq\ \alpha=1,2}\left[\tfrac{1}{2}\hat{P}_{n\alpha}(q)\hat{P}_{n\alpha}(-q)+\tfrac{1}{2}\omega^2_{n\alpha}(q)\hat{Q}_{n\alpha}(q)\hat{Q}_{n\alpha}(-q)\right]-\frac{1}{2}\sum_{q\ \alpha=1,2}J_\alpha(q)\sigma_\alpha(q)\sigma_\alpha(-q), \tag{4}$$

where

$$J_\alpha(q)=\sum_n\frac{\xi_{n\alpha}(q)\xi_{n\alpha}(-q)}{\omega^2_{n\alpha}(q)} . \tag{5}$$

The new Hamiltonian describes displaced oscillators of the same frequency with an additional pseudospin interaction. Introducing lattice Fourier transforms the last term may be written,

$$-\frac{1}{2}\sum_{ll'\ \alpha=1,2}J_\alpha(l-l')\sigma_\alpha(l)\sigma_\alpha(l'). \tag{6}$$

This term describes an effective interaction between the pseudospins at sites l and l'. It is this interaction which gives rise to the cooperative Jahn–Teller transition.

III. EQUATIONS OF MOTION

The Hamiltonians Eqs. (1) and (4) have been written with the high-temperature phase as reference configuration. The distortion from the high-temperature phase is described by nonvanishing thermal expectation values of the operators σ_α and $Q_{n\alpha}$ for one of the two symmetry components, say $\alpha=1$. The thermally averaged quantities will be determined in a mean-field approximation (MFA). The dynamic behavior will be described by equations of motion linearized about the thermally averaged quantities, i.e., by the random-phase approximation (RPA). From the Hamiltonian equation (1) the equations of motion may be written

$$\frac{d}{dt}\sigma_1(q)=2\sum\xi_{n2}(q')Q_{n2}(q')\sigma_3(q-q')-[\sigma_1(q)-\langle\sigma_1\rangle_t]/\tau_1,$$

$$\frac{d}{dt}\sigma_2(q)=-2\sum\xi_{n1}(q')Q_{n1}(q')\sigma_3(q-q')-[\sigma_2(q)-\langle\sigma_2\rangle_t]/\tau_2,$$

$$\frac{d}{dt}\sigma_3(q)=2\sum\xi_{n1}(q')Q_{n1}(q')\sigma_2(q-q')-2\sum\xi_{n2}(q')Q_{n2}(q')\sigma_1(q-q')-[\sigma_3(q)-\langle\sigma_3\rangle_t]/\tau_3, \tag{7}$$

$$\frac{\partial^2}{\partial t^2}Q_{n\alpha}(q)=\omega^2_{n\alpha}(q)Q_{n\alpha}(q)+\xi_{n\alpha}(-q)\sigma_\alpha(q) . \tag{8}$$

In the equations for σ_α we have introduced relaxation terms, where τ_α denotes the relaxation time for the αth spin component to relax to its instantaneous equilibrium value[11] $\langle\sigma_\alpha\rangle_t$ in the presence of the fluctuating phonon field.

We set

$$Q_{n\alpha}(q)=\langle Q_{n\alpha}\rangle+r_{n\alpha}(q),$$
$$\sigma_\alpha(q)=\langle\sigma_\alpha\rangle+\mu_\alpha(q), \tag{9}$$

where $\langle Q_{n\alpha}\rangle=\langle\sigma_\alpha\rangle=0$ for $\alpha=2$. Substituting in Eq. (8) and setting the static parts of the equation of motion equal to zero, we obtain in the case of optic phonons the relationship

$$\xi_{n1}(0)\langle Q_{n1}(0)\rangle=\frac{-|\xi_{n1}(0)|^2}{\omega^2_{n1}(0)}\langle\sigma_1\rangle . \tag{10}$$

For acoustic phonons the limit $q\to 0$ has to be taken along particular directions determined by the symmetry of the strain field to which the electronic levels couple, as specified by Eq. (2). When expressed in terms of the static strain and the constant η_1 introduced in Eq. (2), the corresponding relationship for acoustic phonons takes the form

$$\eta_1\langle e_1\rangle=-(\eta_1^2/c_1 v)\langle\sigma_1\rangle , \tag{11}$$

where v is the volume of the unit cell. The elastic constant c_1 is defined by

$$\omega^2_{a1}(q)=c_1 q^2, \tag{12}$$

where $\omega_{a1}(q)$ is the uncoupled acoustic mode frequency. Thus with the identifications

$$\eta_1\langle e_1\rangle\equiv\xi_{a1}(0)\langle Q_{a1}(0)\rangle, \quad |\xi_{a1}(0)|^2/\omega^2_{a1}(0)\equiv\eta_1^2/c_1 v, \tag{13}$$

where $n=a$ denotes the acoustic mode, the relationship Eq. (10) may be used both for acoustic and optic modes.[12]

According to Eqs. (10) and (11) the strains and the internal displacements are proportional to $\langle\sigma_1\rangle$. In the mean-field approximation $\langle\sigma_1\rangle$ is given by

$$\langle\sigma_1\rangle = \tanh\beta J_1'(0)\,\langle\sigma_1\rangle\,, \tag{14}$$

as follows from Eq. (4). The transition temperature is defined by the limit $\langle\sigma_1\rangle \to 0$, $T \to T_c$, or

$$kT_c = J_1'(0)\,. \tag{15}$$

Because $\sigma_\alpha(l)\sigma_\alpha(l') = 1$ for $l = l'$ and thus adds only a constant to the Hamiltonian, it is important to subtract the term $l = l'$ in the mean field,[3,13] $\sum_{l'} J_1(ll')\langle\sigma_1\rangle$. Thus in Eq. (14) $J_1'(0)$ is given by

$$J_1'(q) = J_1(q) - \frac{1}{N}\sum_q J_1(q)\,, \tag{16}$$

where the last term represents the subtraction of the $l = l'$ term. $J_2'(q)$ which will be introduced below is defined analogously.

Whereas the thermally averaged mean field has a single component, the instantaneous mean field has two components, acting on σ_1 and σ_2, respectively, given by

$$w_\alpha = \left\{-\sum_n \xi_{n\alpha}(q)\,Q_{n\alpha}(q) - [J_\alpha(q) - J_\alpha'(q)]\,\sigma_\alpha(q)\right\}, \quad \alpha = 1, 2. \tag{17}$$

The first term follows directly from Eq. (1). The last two terms subtracts the self-interaction on the same site. Introducing

$$w^2 = w_1^2 + w_2^2 \tag{18}$$

we obtain

$$\begin{aligned}&\langle\sigma_\alpha\rangle_t = (w_\alpha/w)\tanh\beta w\,, \quad \alpha = 1, 2\\&\langle\sigma_3\rangle_t = 0.\end{aligned} \tag{19}$$

We expand the equations of motion in the fluctuating quantities μ_α and $r_{n\alpha}$ and keep only linear terms. When Fourier transformed these equations take the form

$$\begin{aligned}
&\left\{-i\omega + \frac{A_1}{\tau_1}\left[\chi_{11}^{-1}(q) + J_1(q)\right]\right\}\mu_1(q) = -\frac{A_1}{\tau_1}\sum_n \xi_{n1}(q)\,r_{n1}(q)\,,\\
&[\omega_{n1}^2(q) - \omega^2 - i\omega\Gamma_{n1}(q)]\,r_{n1}(q) = -\xi_{n1}(-q)\,\mu_1(q)\,,\\
&\{-i\omega + (A_2/\tau_2)[\chi_{22}^{-1}(q) + J_2(q)]\}\,\mu_2(q) = 2J_1(0)\langle\sigma_1\rangle\,\mu_3(q) - (A_2/\tau_2)\sum_n \xi_{n2}(q)\,r_{n2}(q)\,,\\
&[-i\omega + (1/\tau_3)]\,\mu_3(q) = -2J_1(0)\langle\sigma_1\rangle\,\mu_2(q) - 2\langle\sigma_1\rangle\sum_n \xi_{n2}(q)\,r_{n2}(q)\,,\\
&[\omega_{n2}^2 - \omega^2 - i\omega\Gamma_{n2}(q)]\,r_{n2}(q) = -\xi_{n2}(-q)\,\mu_2(q)\,.
\end{aligned} \tag{20}$$

The first two equations and the last three equations form two separate coupled sets of equations, one for each value of α. In these equations we have introduced the temperature-dependent functions

$$\begin{aligned}&A_1 = \beta(1 - \langle\sigma_1\rangle^2)\,,\\&A_2 = \tanh\beta J_1'(0)\,\langle\sigma_1\rangle / J_1'(0)\langle\sigma_1\rangle\,,\end{aligned} \tag{21}$$

with the limits

$$\begin{aligned}&A_1 = A_2 = \beta, \qquad T > T_c\\&A_2 = 1/J_1'(0), \quad T < T_c\,.\end{aligned} \tag{22}$$

The last identity follows from Eq. (14). We have further introduced

$$\chi_{\alpha\alpha}(q) = A_\alpha / [1 - J_\alpha'(q)A_\alpha], \quad \alpha = 1, 2 \tag{23}$$

where $\chi_{\alpha\alpha}(q)$ are components of the static susceptibility tensor for the pseudospin model Eq. (6) in the RPA. RPA is equivalent to the linearization approximation introduced above. According to Eqs. (15) and (23), $\chi_{11}(0)^{-1} = 0$ at $T = T_c$. In Eq. (20) we have also introduced phenomenological damping constants $\Gamma_{n\alpha}(q)$ in the phonon equations of motion.

We consider first the set of equations corresponding to $\alpha = 1$. Introducing

$$\begin{aligned}&G_\alpha^{-1}(q\omega) = -i\omega(\tau_\alpha/A_\alpha) + \chi_{\alpha\alpha}^{-1}(q) + J_\alpha(q)\,,\\&D_{n\alpha}^{-1}(q\omega) = \omega_{n\alpha}^2(q) - \omega^2 - i\Gamma_{n\alpha}(q)\,.\end{aligned} \tag{24}$$

The coupled-mode dispersion equation may be written

$$G_1^{-1}(q\omega) - \sum_n |\xi_{n1}(q)|^2 D_{n1}(q\omega) = 0 \tag{25}$$

for an arbitrary number of modes n. If we introduce a force $F_{a1}(q)$ acting on the acoustic-phonon normal-mode coordinate $Q_{a1}(q)$,

$$H_F = -\sum_q Q_{a1}(-q)\,F_{a1}(q)\,,$$

we can calculate the corresponding susceptibility

$$\chi_{a1}^{-1}(q\omega) = D_{a1}^{-1}(q\omega) \times \frac{|\xi_{a1}(q)|^2}{G_1^{-1}(q\omega) - \sum_n' |\xi_{n1}(q)|^2 D_{n1}(q\omega)}\,, \tag{26}$$

where the prime on the summation indicates that the summation is over the optic modes only. If we consider frequencies ω much smaller than the optic-phonon frequencies this expression reduces to

$$\chi_{a1}^{-1}(\omega)=\omega_{a1}^2-\omega^2-i\omega\Gamma_{a1}-\delta^2\gamma/(\gamma-i\omega)\ , \tag{27}$$

where

$$\begin{aligned}\gamma(q)&=(A_1/\tau_1)[\chi_{11}^{-1}(q)+J_{a1}(q)]\,,\\ \delta^2(q)&=|\xi_{a1}(q)|^2/[\chi_{11}^{-1}(q)+J_{a1}(q)]\,,\end{aligned} \tag{28}$$

and where we have introduced the definition

$$J_{a\alpha}(q)=|\xi_{a\alpha}(q)|^2/\omega_{a\alpha}^2(q)\,. \tag{29}$$

From Eq. (13) it follows that

$$J_{a\alpha}(0)=\eta_\alpha^2/c_\alpha v\,. \tag{30}$$

Equation (27) has precisely the form used to describe the central peak and critical neutron scattering intensity at other structural transitions.[4-6] Equation (27) yields different values for the low-frequency response $\omega<\omega_{a1}$, $\omega<\Gamma$, depending on the relative values of ω and γ:

$$\begin{aligned}\chi^{-1}&=\omega_{a1}^2\,, &&\omega\gg\gamma\\ &=\tilde{\omega}_{a1}^2\,, &&\omega\ll\gamma\end{aligned} \tag{31}$$

where

$$\tilde{\omega}_{a1}^2=\omega_{a1}^2-\delta^2\,. \tag{32}$$

From Eq. (27) the half-width of the central component Γ_{c1} is determined by

$$\Gamma_{c1}=\gamma(\tilde{\omega}_{a1}^2/\omega_{a1}^2)\,. \tag{33}$$

From Eqs. (28) and (32) the low-frequency elastic constant is given by

$$\tilde{c}_1/c_1=\chi_{11}^{-1}(0)/[\chi_{11}^{-1}(0)+J_{a1}(0)]\ ,\quad \omega\ll\gamma. \tag{34}$$

For $\omega\gg\gamma$ the coupling to the electronic levels has no effect on the elastic constant,

$$\tilde{c}_1/c_1=1 \tag{35}$$

The half-width of the central peak may be written

$$\Gamma_{c1}(q)=(A_1/\tau_1)\chi_{11}^{-1}(q)\,. \tag{36}$$

Equations (34) and (36) show that both the low-frequency elastic constant and the width of the central peak (at $q=0$) goes to zero as $T\to T_c$.

The preceding expressions have been expressed in terms of the static wave-vector-dependent susceptibility of the pseudospin model Eq. (6). Although derived only in RPA, these expressions may be shown to be exact[14] with $\chi_{\rm RPA}$ replaced by the exact χ.

We consider next the set of coupled equations (20) corresponding to $\alpha=2$. The dispersion relation takes the form

$$(-i\omega+1/\tau_3)\frac{A_2}{\tau_2}\left(G_2^{-1}(q\omega)-\sum_n|\xi_{n2}(q)|^2D_{n2}(q\omega)\right)+4J_1^2(0)\langle\sigma_1\rangle^2\left(1-\sum_n|\xi_{n2}(q)|^2D_{n2}(q\omega)\right)=0\,. \tag{37}$$

The corresponding acoustic-phonon susceptibility for frequencies smaller than the optic-phonon frequencies is given by

$$\begin{aligned}\chi_{a2}^{-1}(q\omega)=\omega_{a2}^2(q)-\omega^2-i\omega\Gamma_{a2}(q)\\ -|\xi_{a2}(q)|^2Z_1(\omega)/Z_2(q\omega)\,,\end{aligned} \tag{38}$$

where

$$\begin{aligned}Z_1(\omega)&=4J_1(0)\langle\sigma_1\rangle^2+(A_2/\tau_2)(-i\omega+1/\tau_3)\,,\\ Z_2(q\omega)&=(-i\omega+1/\tau_3)\{-i\omega+(A_2/\tau_2)\\ &\quad\times[\chi_{22}^{-1}(q)+J_{a2}(q)]\}+4J_1(0)\langle\sigma_1\rangle^2\\ &\quad\times[J_1(0)-J_2(q)-J_{a2}(q)]\,.\end{aligned} \tag{39}$$

For $T>T_c$, χ_{a2}^{-1} has the identical structure as χ_{a1}^{-1}, given by Eqs. (27) and (28) with a central peak of width

$$\Gamma_{c2}(q)=(A_2/\tau_2)\chi_{22}^{-1}(q) \tag{40}$$

and high- and low-frequency elastic constants given by

$$\frac{\tilde{c}_2}{c_2}=1,\quad \frac{\tilde{c}_2}{c_2}=\frac{\chi_{22}^{-1}(0)}{\chi_{22}^{-1}(0)+J_{a2}(0)}\,, \tag{41}$$

respectively.

For $T<T_c$, χ_{a2}^{-1} has a much more complicated structure. The electronic levels are no longer degenerate, and the amount of level separation introduces an additional energy scale. However, in the limits $\tau_2,\tau_3\to0$, and $\tau_2,\tau_3\to\infty$ simple expressions for the elastic constants are obtained. We find

$$\frac{\tilde{c}_2}{c_2}=\frac{\chi_{22}^{-1}(0)}{\chi_{22}^{-1}(0)+J_{a2}(0)}\,, \tag{42}$$

$$\frac{\tilde{c}_2}{c_2}=\frac{J_1(0)-J_2(0)}{J_1(0)-J_2(0)+J_{a2}(0)}\,, \tag{43}$$

respectively, in the two limits. According to Eqs. (22) and (23),

$$\chi_{22}^{-1}(0)=J_1'(0)-J_2'(0),\quad T<T_c\,. \tag{44}$$

Thus in the low-temperature phase, the relative elastic constant $\tilde{c}_2/c_2$ is independent of temperature in both limits.

The low-frequency elastic constants calculated above correspond to isothermal elastic constants. The adiabatic elastic constants, measured ultrasonically, are related to the isothermal constants by well-known thermodynamic relations. Dynamically the relationship between the adiabatic and isothermal elastic constants is obtained by considering the coupling of the sound wave with the heat-diffusion mode.[15] The adiabatic and isothermal elastic constants are equal in the high-temperature phase[16] and at $T = 0$.

IV. LIMITING CASES

For coupling to elastic strain and optic phonons of a single irreducible representation, we obtain the results given by Eqs. (27)–(35) where $\chi_{11}(q)$ now denotes the static susceptibility of the Ising model,

$$-\frac{1}{2}\sum_{l\neq l'} J_1(ll')\sigma_1(l)\sigma_1(l'). \tag{45}$$

The results for coupling to doubly degenerate phonon modes (E modes), as in the case of tetragonal distortion of octahedrally coordinated Jahn-Teller ions,[1] are obtained by setting $\xi_{n1}=\xi_{n2}=\xi_n$, $\tau_1=\tau_2=\tau$, and $J_1=J_2=J$. The elastic constants are then given by

$\tau\to\infty$:

$$\begin{aligned} \frac{\tilde{c}_1}{c} &= 1, \quad \text{all } T \\ \frac{\tilde{c}_2}{c} &= 1, \quad T > T_c \\ \frac{\tilde{c}_2}{c} &= 0, \quad T < T_c; \end{aligned} \tag{46}$$

$\tau\to 0$:

$$\begin{aligned} \frac{\tilde{c}_1}{c} &= \frac{\chi_{11}^{-1}(0)}{\chi_{11}^{-1}(0)+J_a(0)}, \\ \frac{\tilde{c}_2}{c} &= \frac{\chi_{22}^{-1}(0)}{\chi_{22}^{-1}(0)+J_{a1}(0)}; \end{aligned} \tag{47}$$

where χ_{11} and χ_{22} are the susceptibilities for the pseudospin model,

$$-\frac{1}{2}\sum_{l\neq l'} J(ll')[\sigma_1(l)\sigma_1(l')+\sigma_2(l)\sigma_2(l')]. \tag{48}$$

In RPA,

$$\begin{aligned} \chi_{11}(q) &= \frac{A_1}{1-J'(q)A_1}, \\ \chi_{22}(q) &= \frac{A_2}{1-J'(q)A_2}, \end{aligned} \tag{49}$$

with

$$A_1 = \beta(1-\langle\sigma_1\rangle^2),$$

$$A_2 = \frac{\tanh\beta J'(0)\langle\sigma_1\rangle}{J'(0)\langle\sigma_1\rangle}.$$

The width of the central peak for $T > T_c$ is given by

$$\Gamma_c(q) = (\beta/\tau)\chi^{-1}(q), \tag{50}$$

where $\chi_{11}=\chi_{22}=\chi$. The elastic constants in (46) and (47) agree with results previously derived in these limits.[2,17] The general expressions provide an interpolation between these limits.

$TmVO_4$ is an example for which the doubly degenerate electronic levels couple strongly to two different symmetry modes.[8] These are optic phonons and elastic strains of B_{2g} and B_{1g} irreducible representations of the D_{2d} point group, respectively. The corresponding elastic constants are $c_1 = c_{66}$ and $c_2 = \frac{1}{2}(c_{11}-c_{12})$ with c_1 going to zero at the phase transition. The elastic constants in $TmVO_4$ have been measured ultrasonically.[8] Neutron studies of the soft acoustic modes and of the central peak have not yet been carried out. In soft mode systems which have been studied so far the central peak has been too narrow to be resolved by neutron scattering. Brillouin scattering data on $TbVO_4$,[18,19] which measure the elastic constants at intermediate frequencies, suggest that the width of the central peak may be $\sim 10^{11}$ rad/sec. This is estimated from the frequency for which deviation from the low-frequency elastic constant appears. In $TmVO_4$ it may therefore be possible to resolve the central peak structure and to study the predicted narrowing of the central peak directly.

V. CONCLUDING REMARKS

In the models discussed above it was assumed that the electronic levels couple to both acoustic and optic modes. In that case the acoustic mode is the soft mode. If there is no coupling to acoustic modes then the optic phonon susceptibility will diverge at the transition point. For example, if we consider coupling to a single nondegenerate optic-phonon mode, its susceptibility will have the form of Eq. (27),

$$\chi_0(\omega) = \omega_0^2 - \omega^2 - i\omega\Gamma_0 - \delta^2\gamma/(\gamma - i\omega). \tag{51}$$

As before, the low-frequency susceptibility $\omega \ll \omega_0$, $\omega \ll \Gamma_0$ has different values depending on the relative values of ω and γ,

$$\begin{aligned} \chi_0 &= 1/\tilde{\omega}_0^2 \quad (\omega \ll \gamma) \\ &= 1/\omega_0^2 \quad (\omega \gg \gamma), \end{aligned} \tag{52}$$

where

$$\frac{\tilde{\omega}_0^2(q)}{\omega_0^2(q)} = \frac{\chi^{-1}(q)}{\chi^{-1}(q)+J(q)} \tag{53}$$

and the width of the central component is given by

$$\Gamma_c = (A/\tau)\chi^{-1}(q), \tag{54}$$

where A is defined by the first of Eqs. (21). However, in order to have a divergence in the optic-phonon susceptibility, it is in addition required that there is no direct coupling between the optic mode and the elastic strain of the form

$$g\sum_q e(q)Q(-q). \tag{55}$$

This interaction induces a strain distortion even if only optic phonons couple to the electrons. It gives rise to an acoustic-phonon susceptibility given by

$$\chi_a^{-1}(\omega)=(\omega_a^2-\omega^2-i\omega\Gamma_a)-g^2q^2\chi_0(\omega)\ , \tag{56}$$

where $\chi_0(\omega)$ is defined by Eq. (51). From χ_a we obtain the low-frequency elastic constant,

$$\tilde{c}=c-g^2/\tilde{\omega}_0^2(0)\ . \tag{57}$$

Thus as $\tilde{\omega}_0^2(0)$ approaches zero according to Eq. (53) $\tilde{c}$ will go to zero before $\tilde{\omega}_0^2$, and the acoustic mode will again be the soft mode.

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