Neutron scattering by polaritons: Mechanical excitation of the polariton*

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The displacement-displacement correlation function in the case of an isotropic ionic crystal with several optic branches has been calculated to determine the inelastic-scattering neutron cross section. We have examined the feasibility of an experiment to measure the dispersion curves and the density of states of the polar modes. The infrared electromagnetic field outside the crystal associated with the polariton excitation has been considered and found directly related to the polariton density of states. Though relevant information can be gained by this kind of experiment, its feasibility seems to be subordinate to the planned fulfilment of the high-energy pulsed neutron sources.

I. INTRODUCTION

As is well known, polaritons[1] are composite quasiparticles formed, in a crystal, by coupling the electromagnetic field with elementary excitations. We shall be concerned with the coupling in ionic crystals of long-wavelength transverse-optic (TO) phonons and the electromagnetic (EM) field.

This kind of polariton connects the optical and vibrational properties that are very important in the understanding of the macroscopic behavior in terms of a microscopic picture.

A large amount of experimental work has been done on the dispersion relation of polaritons,[1] mainly using Raman scattering, by which a very good resolution in energy-momentum space can be obtained though the upper branch cannot be easily measured.

However, the double nature of the polariton allows it to be excited by interacting with its mechanical part. In principle at least this can be done using both thermal-neutron and x-ray scattering, but the x-ray scattering has a very bad resolution in the phonon energy range.

We consider the neutron scattering by polaritons by exploring the feasibility of the experiment, which is seriously limited by the very low energy-momentum ratio of the neutron. However, these limitations concern the coherent scattering only, while the incoherent scattering has no limitations connected to the momentum resolution as it will be shown later.

Another stimulating aspect of the polariton excitation by neutrons is the possibility to obtain information by the measurement of the infrared EM field which leaks out of the crystal. This EM field might be experimentally detected, since no other source is present in the infrared region at low temperature, so that the background is expected to be small.

II. NEUTRON CROSS SECTION

As is well known, for thermal neutrons the cross section can be calculated in the Born approximation. Let us consider the nuclear inelastic scattering neglecting the magnetic contribution, which does not change the general characteristics of the cross section; for processes involving only one excitation we have[2]

$$\frac{d^2\sigma}{d\Omega\, d\epsilon} = \frac{1}{2\pi\hbar}\frac{K'}{K}\sum_{\substack{\vec{l}\,\vec{s}\\ \vec{l}'\vec{s}'\\ \alpha\beta}} b_{\vec{l}\,\vec{s}} b_{\vec{l}'\vec{s}'}\, e^{-W_{\vec{l}\,\vec{s}}} e^{-W_{\vec{l}'\vec{s}'}} \times \exp[-i\vec{\chi}\cdot(\vec{l}+\vec{s}-\vec{l}'-\vec{s}')]\chi_\alpha\chi_\beta \times \int_{-\infty}^{+\infty} dt\, e^{i\omega t}\langle u^\alpha_{\vec{l}\,\vec{s}}(0) u^\beta_{\vec{l}'\vec{s}'}(t)\rangle\,, \tag{1}$$

where $\vec{l}$ is the unit-cell index, $\vec{s}$ is the position of the atoms in the unit cell, $b_{\vec{l}\,\vec{s}}$ is the nuclear scattering amplitude, and $e^{-W_{\vec{l}\,\vec{s}}}$ is the corresponding Debye-Waller factor. $\vec{K}$ is the wave vector of the incoming neutron and $\vec{K}' = \vec{K} - \vec{\chi}$ is the wave vector of the scattered neutron. $u^\alpha_{\vec{l}\,\vec{s}}(t)$ is the Heisenberg representation of the α component of the displacement from the equilibrium position of the atom at the site $\vec{l}+\vec{s}$. $\epsilon = \hbar\omega$ is the neutron energy change. The angular brackets indicate the thermal average.

The cross section [Eq. (1)] gives rise to two different processes: the coherent and incoherent scattering. The former is related with interference phenomena produced by the crystal as a whole because of its periodicity; the latter is due to local fluctuations.[2]

The two contributions can be easily separated introducing the average scattering amplitude $\bar{B}_{\vec{s}} = \langle b_{\vec{l}\,\vec{s}} e^{-W_{\vec{l}\,\vec{s}}}\rangle_c$, independent of $\vec{l}$ in a perfect crystal, and the fluctuations

$$\Delta_{\vec{l}\,\vec{s}} = b_{\vec{l}\,\vec{s}} e^{-W_{\vec{l}\,\vec{s}}} - \bar{B}_{\vec{s}}\,.$$

$\Delta_{\vec{l}\,\vec{s}}$ is a random quantity such that

$$\langle \Delta_{\vec{l}\,\vec{s}}\rangle_c = 0, \quad \langle \Delta_{\vec{l}\,\vec{s}}\Delta_{\vec{l}'\vec{s}'}\rangle_c = \delta_{\vec{l}\,\vec{l}'}\delta_{\vec{s}\,\vec{s}'}\Delta^2_{\vec{s}}\,,$$

where $\langle\cdots\rangle_c$ indicates the configurational average over the Gibbs ensemble. We have

$$\left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\rm coh} = \frac{1}{2\pi\hbar}\frac{K'}{K}\sum_{\substack{\vec{l}\vec{l}'\\ \vec{s}\vec{s}'\\ \alpha\beta}} \bar{B}_{\vec{s}}\bar{B}_{\vec{s}'}\exp[-i\vec{\chi}\cdot(\vec{l}+\vec{s}-\vec{l}'-\vec{s}')] \times \chi_\alpha\chi_\beta\int_{-\infty}^{+\infty} dt\, e^{i\omega t}\langle u^\alpha_{\vec{l}\vec{s}}(0)u^\beta_{\vec{l}'\vec{s}'}(t)\rangle \quad , \tag{2}$$

$$\left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\rm inc} = \frac{1}{2\pi\hbar}\frac{K'}{K}\sum_{\substack{\vec{l}\vec{s}\\ \alpha\beta}} \Delta^2_{\vec{s}}\,\chi_\alpha\chi_\beta \times\int dt\, e^{i\omega t}\langle u_{\vec{l}\vec{s}}(0)u^\beta_{\vec{l}\vec{s}}(t)\rangle \ . \tag{3}$$

Then we develop the atomic displacement in normal coordinates[3]

$$u^\alpha_{\vec{l}\vec{s}}(t) = (NM_{\vec{s}})^{-1/2}\sum_{\vec{q}j} e^\alpha_{\vec{q}j}(\vec{s})Q_{\vec{q}j}(t)\, e^{i\vec{q}\cdot\vec{l}} \ , \tag{4}$$

where $M_{\vec{s}}$ is the mass of the atom in the $\vec{s}$ site, $e_{\vec{q}j}(\vec{s})$ and $Q_{\vec{q}j}(t)$ are, respectively, the polarization vector and the normal coordinate of the $(\vec{q},j)$ mode. In Eq. (4) $\vec{q}$ is a vector of the first Brillouin zone.

Substituting Eq. (4) into Eqs. (2) and (3), we can see that both cross sections contain the correlation functions $\langle Q_{\vec{q}j}(0)Q_{\vec{q}j'}(t)\rangle$.[4] To obtain their explicit expression we use the Green's-function method in a way similar to Benson and Mills.[5] We assume the Hamiltonian to be

$$H = H_{\rm ph} + H_{\rm EM} + H_{\rm EM\text{-}ph} + H_{\rm anh} \ , \tag{5}$$

while

$$H_{\rm ph} = \frac{1}{2}\sum_{\vec{q}\lambda n}(P^\dagger_{\vec{q}\lambda n}P_{\vec{q}\lambda n} + \omega^2_{\vec{q}\lambda n}Q^\dagger_{\vec{q}\lambda n}Q_{\vec{q}\lambda n}) \tag{6}$$

describes[6] the phonons in the harmonic approximation, $P_{\vec{q}\lambda n}$ and $\omega_{\vec{q}\lambda n}$ being, respectively, the momentum conjugate to $Q_{\vec{q}\lambda n}$ and the normal-mode frequency; we have substituted the branch index j with λn, where λ specifies the polarization and n indicates the nth equivalent oscillator of the medium.[7]

$$H_{\rm EM} = \frac{1}{2}\sum_{\vec{q}\lambda_\perp}\left(\Pi^\dagger_{\vec{q}\lambda}\Pi_{\vec{q}\lambda} + \frac{c^2q^2}{\epsilon_0}A^\dagger_{\vec{q}\lambda}A_{\vec{q}\lambda}\right) \tag{7}$$

is the Hamiltonian of the EM transverse field in an isotropic medium in the absence of coupling,[8] ϵ_0 being the high-frequency dielectric constant, $A_{\vec{q}\lambda}$ and $\Pi_{\vec{q}\lambda}$ are the vector potential and electric field operators, respectively.

$$H_{\rm EM\text{-}ph} = -\omega_p\sum_{\vec{q}\lambda_\perp n} f_n^{1/2}P^\dagger_{\vec{q}\lambda n}A_{\vec{q}\lambda} + \tfrac{1}{2}\omega_p^2\sum_{\vec{q}\lambda_\perp}A^\dagger_{\vec{q}\lambda}A_{\vec{q}\lambda} \tag{8}$$

is the interaction Hamiltonian[7,8] in the long-wavelength limit for an isotropic medium. ω_p is the usual plasma frequency,

$$\omega_p^2 = \sum_{\vec{s}}\frac{4\pi\epsilon^2_{\vec{s}}}{\epsilon_0 V_c M_{\vec{s}}} \ ,$$

$\epsilon_{\vec{s}}$ being the charge of the sth ion and V_c is the unit-cell volume; f_n is the "oscillator strength" of the nth equivalent oscillator, which in the long-wavelength limit and for an isotropic medium we take independent of λ,[9] and $\vec{q}$. f_n obeys the sum rule $\sum_n f_n = 1$, for each transverse polarization. $H_{\rm anh}$, which will cause the normal modes to acquire a finite lifetime, is assumed to depend only on the normal coordinates $Q_{\vec{q}\lambda n}$.

Let us consider two operators, A and B; the Fourier transform of their correlation function is

$$(2\pi)^{-1}\int_{-\infty}^{+\infty} dt\, e^{i\omega t}\langle A(0)B(t)\rangle = i[1+n(\omega)][G_r(\omega+i\epsilon) - G_a(\omega-i\epsilon)] \ , \tag{9}$$

where $n(\omega) = (e^{\beta\hbar\omega}-\eta)^{-1}$ ($\eta = +1$ and $\eta = -1$ for bosons and fermions, respectively), and G_r and G_a are the usual retarded and advanced Green's functions.[5,10]

We can obtain the Green's function from the equation of motion

$$i\frac{d}{dt}G(t) = \delta(t)\langle[A(t),B(0)]\rangle + \langle\langle[A(t),H],B(0)\rangle\rangle \ , \tag{10}$$

where H is the Hamiltonian of the system.

Thus the correlation function $\langle Q_{\vec{q}\lambda n}(0)Q_{\vec{q}\lambda' n'}(t)\rangle$ is related to the Green's function

$$D^{\lambda n,\lambda' n'}_{QQ}(\vec{q},\omega) = \int_{-\infty}^{+\infty}\frac{dt}{2\pi}\, e^{i\omega t}\langle\langle Q^\dagger_{\vec{q}\lambda n}(t), Q_{\vec{q}\lambda' n'}(0)\rangle\rangle \ , \tag{11}$$

which is given by

$$(\omega^2_{\vec{q}\lambda} - \omega^2)D^{\lambda n,\lambda' n'}_{QQ}(\vec{q},\omega) - \sum_m\frac{\Omega_{p,\lambda n}\Omega_{p,\lambda m}\omega^2_{\vec{q}\lambda m}}{(c^2q^2/\epsilon_0)+\omega_p^2-\omega^2}D^{\lambda m,\lambda' n'}_{QQ}(\vec{q},\omega) + \sum_m\frac{\Omega_{p,\lambda n}\Omega_{p,\lambda m}}{(c^2q^2/\epsilon_0)+\omega_p^2-\omega^2}\mathcal{G}^{\lambda m,\lambda' n'}_{PQ}(\vec{q},\omega) - \mathcal{G}^{\lambda n,\lambda' n'}_{PQ}(\vec{q},\omega) = -\frac{1}{2\pi}\delta_{\lambda\lambda'}\delta_{nn'} + \frac{1}{2\pi}\frac{\Omega_{p,\lambda n}\Omega_{p,\lambda' n'}\delta_{\lambda\lambda'}}{(c^2q^2/\epsilon_0)+\omega_p^2-\omega^2} \ , \tag{12}$$

where

$$\Omega_{p,\lambda n} = \begin{cases}\omega_p f_n^{1/2} & \text{for the transverse-optic modes,}\\ 0 & \text{otherwise,}\end{cases} \tag{13}$$

and

$$\mathcal{G}^{\lambda n,\lambda' n'}_{PQ}(\vec{q},\omega) = -i\int\frac{dt}{2\pi}\, e^{i\omega t} \times\langle\langle[P_{\vec{q}\lambda n}(t), H_{\rm anh}], Q_{\vec{q}\lambda' n'}(0)\rangle\rangle \ . \tag{14}$$

It is important to note that Eq. (12) implies a mixing of the different transverse-optic branches of the same polarization, even in the absence of anharmonic coupling. The mechanism of this coupling has a clear physical interpretation: in fact the TO normal modes couple through their own EM field, behaving like classical forced oscillators. However, the coupling is small when the different branches have large energy separation, as we expect. If, for instance, we consider two optic branches with $\lambda = \lambda'$, $\omega_{\mathrm{TO},2}^2 \gg \omega_{\mathrm{TO},1}^2$, in the absence of anharmonic terms, we find, for $\omega = \omega_{\mathrm{TO},1}$:

$$D_{QQ}^{11} \sim \frac{1}{\pi} \frac{\Omega_{p2}^2}{\Omega_{p1}^2} \frac{\omega_{\mathrm{TO},2}^2}{\omega_{\mathrm{TO},1}^2} \frac{1}{\omega_{\mathrm{TO},1}^2}, \quad D_{QQ}^{12} \sim \frac{1}{\pi} \frac{\Omega_{p2}}{\Omega_{p1}} \frac{1}{\omega_{\mathrm{TO},1}^2},$$

$$D_{QQ}^{22} \sim -\frac{1}{2\pi} \frac{1}{\omega_{\mathrm{TO},2}^2},$$

and thus $|D_{QQ}^{11}| \gg |D_{QQ}^{12}| \gg |D_{QQ}^{22}|$ if we assume $\Omega_{p,1} \sim \Omega_{p,2}$.

We note that if the anharmonic term (14) is diagonal with respect to the polarization index λ, then the Green's function is diagonal too; in other words, the coupling between different polarizations takes place only through $\mathcal{G}_{PQ}$. Equation (12) is very useful in neutron scattering studies, as it contains also the contributions of longitudinal and acoustic waves, while Benson and Mills[5] consider a single TO branch only. Moreover, for the modes that do not couple to the EM field,[9] Eq. (12) reduces to the usual equation of motion for the Green's function.

It is well known that the main effect of the anharmonic term is a shift of the polariton frequencies and the appearance of a finite lifetime, as it has been extensively discussed by Wallis and Maradudin.[6] However, because the anharmonic width is very small compared to the energy resolution generally achieved in neutron scattering experiments, we shall neglect these effects and thus consider D_{QQ} diagonal with respect to the polarization index.

Let us now consider a crystal with two atoms per unit cell. In this case $f_n = 1$ for the two TO branches, degenerate in the isotropic medium we consider, and $f_n = 0$ for the longitudinal and acoustic branches. Using Eqs. (12) and (9) for the correlation function, we obtain

$$\int \frac{dt}{2\pi} e^{i\omega t} \langle Q_{\vec{q}\lambda n}(0) Q_{\vec{q}'\lambda' n'}^{\dagger}(t) \rangle = \delta_{\lambda\lambda'} \delta_{nn'} \frac{n(\omega)+1}{2\omega_{\vec{q}\lambda n}} S_{\mathrm{ph}}^{\lambda n}(\omega) \times \{[\delta(\omega-\omega_-) - \delta(\omega+\omega_-)] + [\delta(\omega-\omega_+) - \delta(\omega+\omega_+)] f_n\}, \tag{15}$$

where $S_{\mathrm{ph}}^{\lambda n}(\omega)$ is the phonon strength function, which is

$$S_{\mathrm{ph}}^{\lambda n}(\omega) = \begin{cases} \dfrac{\omega \omega_p^2 \omega_{\mathrm{TO}}}{(\omega_{\mathrm{TO}}^2 - \omega^2)^2 + \omega_p^2 \omega_{\mathrm{TO}}^2} & \text{for the TO branches (Ref. 1),} \\ 1 & \text{otherwise,} \end{cases} \tag{16}$$

and

$$\omega_{\pm}^2(\vec{q}) = \tfrac{1}{2}\{(c^2q^2/\epsilon_0 + \omega_{\vec{q}\lambda n}^2 + \omega_p^2) \pm [(c^2q^2/\epsilon_0 + \omega_p^2 - \omega_{\vec{q}\lambda n}^2)^2 + 4 f_n \omega_p^2 \omega_{\vec{q}\lambda n}^2]^{1/2}\}. \tag{17}$$

We note that for $f_n = 1$, $\omega_{\pm}^2(\vec{q})$ gives the usual polariton dispersion relation, while for $f_n = 0$, $\omega_-^2(\vec{q}) = \omega_{\vec{q}\lambda n}^2$, i.e., the unperturbed phonon frequency, and $\omega_+^2(\vec{q}) = \omega_p^2 + c^2q^2/\epsilon_0$, which is the well-known dispersion relation for the transverse EM field in a plasma. However, the Green's function D_{QQ} has no pole at this latter frequency, as D_{QQ} is related to the mechanical excitation only.

Thus, using Eqs. (2) and (15), we have

$$\left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\mathrm{coh}} = \frac{N}{2\pi\hbar} \frac{K'}{K} \sum_{\vec{q}\lambda n} |\vec{F}_{\lambda n}(\vec{\chi}) \cdot \vec{\chi}|^2 \frac{n(\omega)+1}{2\omega_{\vec{q}\lambda n}} \times S_{\mathrm{ph}}^{\lambda n}(\omega) \delta(\vec{\chi} - \vec{q} - \vec{G}_0) \{[\delta(\omega-\omega_-) - \delta(\omega+\omega_-)] + [\delta(\omega-\omega_+) - \delta(\omega+\omega_+)] f_n\}. \tag{18}$$

$\vec{G}_0$ being the reciprocal-lattice vector which takes $\vec{\chi}$ in the first Brillouin zone and

$$\vec{F}_{\lambda n}(\vec{\chi}) = \sum_{\vec{s}} M_{\vec{s}}^{-1/2} \bar{B}_{\vec{s}} e^{-i\vec{\chi}\cdot\vec{s}} \vec{e}_{\vec{\chi}\lambda n}(\vec{s}). \tag{19}$$

From Eq. (3), we get

$$\left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\mathrm{inc}} = \frac{1}{2\pi\hbar} \frac{K'}{K} \sum_{\substack{\vec{s} \\ \vec{q}\lambda n}} |\vec{\chi} \cdot \vec{e}_{\vec{q}\lambda n}(\vec{s})|^2 \frac{\Delta_{\vec{s}}^2}{M_{\vec{s}}} \frac{n(\omega)+1}{2\omega_{\vec{q}\lambda n}} S_{\mathrm{ph}}^{\lambda n}(\omega) [2\omega_-(\vec{q}) \delta(\omega^2 - \omega_-^2(\vec{q})) + 2\omega_+(\vec{q}) \delta(\omega^2 - \omega_+^2(\vec{q})) f_n] \text{ when } \omega \geq 0, \tag{20}$$

which in the case of cubic crystals reduces to

$$\left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\mathrm{inc}} = \frac{N}{2\pi\hbar} \frac{K'}{K} \chi^2 [n(\omega)+1] \sum_{\vec{s}} \frac{\Delta_{\vec{s}}^2}{M_{\vec{s}}} \left(\frac{g_{\mathrm{TO}}(\omega)}{\omega_{\mathrm{TO}}} + \frac{g_{\mathrm{ph}}(\omega)}{\omega}\right) \quad (\omega \geq 0), \tag{21}$$

where $g_{\mathrm{ph}}(\omega)$ is the density of states for the pure-phonon branches and

$$g_{\mathrm{TO}}(\omega) = \frac{2\omega}{3N} \sum_{\vec{q}} \frac{\omega_{\mathrm{TO}}}{\omega_{\vec{q}\lambda_\perp 2}} S_{\mathrm{ph}}^{\mathrm{TO}}(\omega)$$

$$\times [\delta(\omega^2 - \omega_-^2(\vec{q})) + \delta(\omega^2 - \omega_+^2(\vec{q}))], \tag{22}$$

$\omega_{\vec{q}\lambda_\perp 2}$ being the dispersion relation of the TO phonon in the absence of interaction with the EM field.

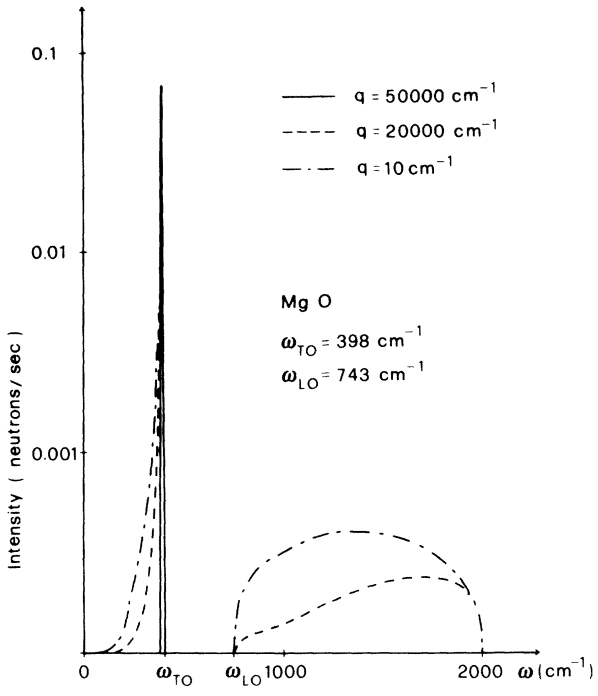

FIG. 1. Coherently scattered neutron flux in typical q-constant scans, $q = 10$, 20 000, and 50 000 cm^{-1}. The corresponding frequencies are $\omega_-(10) = 0.5$ cm^{-1}, $\omega_+(10) = 744$ cm^{-1}, $\omega_-(20\,000) = 375$ cm^{-1}, $\omega_+(20\,000) = 1960$ cm^{-1}, $\omega_-(50\,000) = 395$ cm^{-1}, $\omega_+(50\,000) = 4650$ cm^{-1}. The energy of the incident neutron is 2000 cm^{-1}. In the calculation an isotropic source of 10^{17} neutrons/(cm^2 sec eV) has been used.

From Eq. (18) we can see that in an ideal scattering experiment the polariton dispersion relation can be easily measured by properly choosing the scattering vector $\vec{\chi}$. However, good resolution in $\vec{q}$ space is not yet available and this is obviously the main limit to the use of neutron coherent scattering in polariton experiments. Anyhow the feasibility of such an experiment is shown in Fig. 1, where the scattered neutron flux is plotted vs ω in a $\vec{q}$ constant scan in the case of MgO,[11] which is suitable material for this kind of experiment, because of its large $\omega_{LO}^2/\omega_{TO}^2 \equiv (\omega_{TO}^2 + \omega_p^2)/\omega_{TO}^2$ ratio. The intensity has been calculated using a resolution function

$$R(\vec{q}, \omega) = (2\pi\sigma_q^2)^{-1/2}(2\pi\sigma_\omega^2)^{-1/2} \times \exp(-q^2/2\sigma_q^2)\exp(-\omega^2/2\sigma_\omega^2) ,$$

where $\sigma_q = 10\,000$ cm^{-1} and $\sigma_\omega = 5$ cm^{-1}. Figure 1 shows first that a neutron scattering experiment allows a quite accurate measurement of ω_{TO} and ω_{LO}, which together with ϵ_0 determine completely the polariton dispersion curves. Moreover, one can see a strong variation of the peak shapes varying $\vec{q}$. That variation can be qualitatively correlated to the shape of the dispersion curves. The incident neutron energy was 2000 cm^{-1}. The absolute intensity scale in Fig. 1 has been approximately evaluated using an isotropic source of 10^{17} neutrons/(cm^2 sec eV).[12] The neutron inelastic scattering kinematic is extensively discussed in the literature.[13–15] However, we preferred to use a simplified resolution function, depending only on the square modulus of the $\vec{q}$ vector, since the qualitative features of the neutron scattering do not depend on its detailed shape. The assumed $\vec{q}$-space resolution is quite high as compared to the presently available neutron spectrometers. However, such a resolution corresponds to an angular spread of about 0.5×10^{-4} rad. Thus a suitable spectrometer is limited by the present state of art. Because of the high value of ω_{LO} in MgO and in similar materials, the neutron energy must be high, as compared to the average energies available in the high-flux reactors. However, high-energy neutron sources are already available, which give intense pulsed neutron beams obtained by conversion of charged particles (electrons or protons). Obviously these pulsed sources are particularly suitable to be used in chopper spectrometers and they have been already employed in condensed matter studies.[12,16] However, the beam intensity we used can be obtained only with the planned sources.[12]

Complementary information could be gained from the incoherent scattering, but it is obvious that the structure of g_{TO} is generally obscured by the contributions of g_{ph}.

III. INFRARED EM FIELD

Let us consider the equation which determines the rate of variation, inside the crystal, of the energy density of the polaritons excited by some external source (neutrons in our case)[1,17]

$$\operatorname{div}\vec{S} = -\frac{dU}{dt} , \tag{23}$$

$\vec{S}$ being the Poynting vector, $\vec{S} = (c/4\pi)\vec{E}\times\vec{H}$, and U is the energy density, which will contain both a mechanical and an electromagnetic contribution. We note that Eq. (23) holds in the absence of anharmonic dissipation processes.

Let us consider now the following stationary process: a constant flux of neutrons is impinging on the crystal which is in thermal equilibrium with a bath of external radiation, at constant temperature. In this case $dU/dt = 0$ so that a constant flux of electromagnetic energy leaks out of the crystal. In this ideal situation the photon flux which leaves the crystal is equal to the polariton flux created by the neutrons times the "electric field strength function." Thus, for the photon flux in the energy interval $d\epsilon$, we can write

$$\frac{d\mathfrak{N}}{d\epsilon} = \left(\frac{d\mathfrak{N}}{d\epsilon}\right)_{\text{coh}} + \left(\frac{d\mathfrak{N}}{d\epsilon}\right)_{\text{inc}}$$

$$= \int \left[\left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\text{coh}} + \left(\frac{d^2\sigma}{d\Omega\, d\epsilon}\right)_{\text{inc}}\right] S_{\text{EM}}(\omega)\, d\Omega \ , \quad (24)$$

where $S_{\text{EM}}(\omega)$ is the electron field strength function, given by[1]

$$S_{\text{EM}}(\omega) = \left(1 + \frac{\omega_p^2\, \omega_{\text{TO}}^2}{(\omega_{\text{TO}}^2 - \omega^2)^2}\right)^{-1} . \quad (25)$$

By substituting Eqs. (18) and (21) in (24), we find

$$\frac{d\mathfrak{N}}{d\epsilon} = \frac{N}{\hbar}\frac{\epsilon_0}{c^2}\frac{1}{K^2}\left(\sum_{\vec{G}_0}\frac{1}{X}\left|\vec{F}_{\text{TO}}(\vec{G}_0)\cdot\vec{G}_0\right|^2\right)\frac{n(\omega)+1}{\omega_{\text{TO}}}\,\omega S_{\text{ph}}^{\text{TO}}(\omega)$$

$$+\frac{2N}{\hbar}\left(\sum_{\vec{s}}\frac{\Delta_{\vec{s}}^2}{M_{\vec{s}}}\right)\frac{K'}{K}(K^2+K'^2)\frac{n(\omega)+1}{\omega_{\text{TO}}}g_{\text{TO}}(\omega)S_{\text{EM}}(\omega), \quad (26)$$

when $0 \le \omega < \omega_{\text{TO}}$, $\omega > \omega_{\text{LO}}$; $d\mathfrak{N}/d\epsilon = 0$ elsewhere.

The sum in the first term is extended over all of the reciprocal-lattice vectors $\vec{G}_0$, which take the scattering vector $\vec{\chi} = \vec{K} - \vec{K}'$ in the first Brillouin zone and $X = \cos\beta + \sin\beta\,\tan\alpha$ with $\beta = \arccos(\hat{K}\cdot\hat{K}_B)$, $\alpha = -\arcsin(\hat{G}_0\cdot\hat{K}_B)$, and $\vec{K}_B = \vec{K} - \vec{G}_0$. Calculating the coherent contribution to the electromagnetic emission we used the approximation $q \ll \chi \simeq G_0$, which verifies only when ω is not too close to ω_{TO}.

We note that the coherent contribution to the EM field emission is independent of the shape of the dispersion relation, while the incoherent part is proportional to the polariton density of states. It is evident that by a proper choice ($\hat{G}_0\cdot\hat{K}_B = 1$) of the scattering geometry the former contribution will vanish and thus we can obtain direct information on g_{TO}.

In Fig. 2 we have plotted $d\mathfrak{N}/d\epsilon$ for MgO, assuming a vanishing coherent contribution in Eq. (26). As is well known, this can always be achieved by a proper choice of the crystal orientation. For comparison, in Fig. 2 we have plotted also the polariton density of states $g_{\text{TO}}(\omega)$. As we expect, g_{TO} shows a large peak at $\omega = \omega_{\text{TO}}$ and a slowly varying contribution above ω_{LO}. Though the EM field emission is proportional to g_{TO}, it behaves quite differently. In fact, the largest contribution comes from the upper branch, while the lower branch gives a nearly negligible emission. The particular behavior of the lower branch is due to the fact that the corresponding modes are mainly mechanical in character.

The expected intensity for the EM fields is probably outside the sensibility limit of the presently available infrared detectors. However, a chopper system can be used to reduce the detector noise and make appreciable the emission.

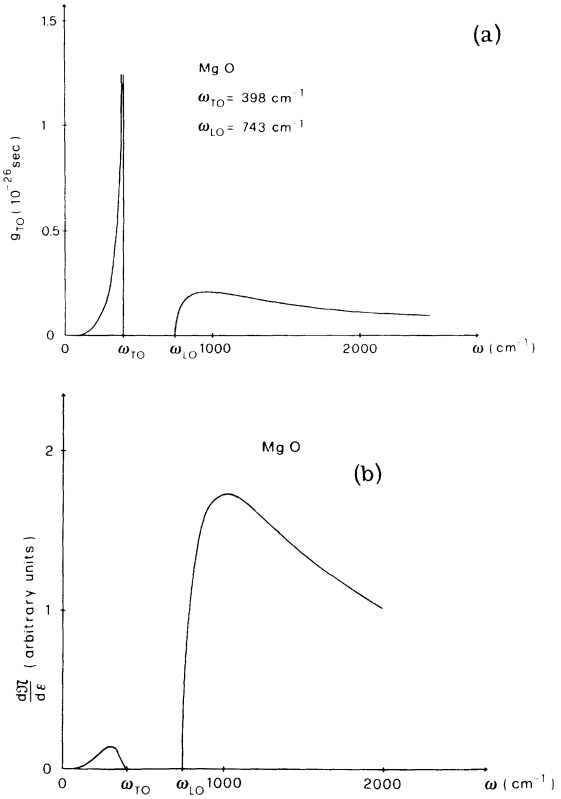

FIG. 2. (a) Polariton density of states $g_{\text{TO}}(\omega)$. (b) Infrared EM field emission.

IV. CONCLUSIONS

In conclusion, our results indicate the possibility of obtaining information on the polariton dispersion curves by neutron coherent scattering experiments. Though this kind of experiment does not have a good accuracy, one can suggest it for those materials which are not suitable for Raman scattering studies. The incoherent scattering processes give information not affected by the bad momentum resolution, however, the polariton contribution is generally masked by that from the nonpolar modes. It is important to note that the neutron scattering experiments also allow one to gain information about the upper polariton branch, which is generally badly covered by the Raman scattering.[1,5] Finally the neutron scattering can give further useful data by means of the electromagnetic field emission, associated with the polariton excitation in both branches.

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