Dipole-active excitations in crystals with one-dimensional disorder

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We consider the spectrum of long-wavelength excitons and optical phonons in crystals with one-dimensional disorder (crystals such as ZnS with alternating layers having the sphalerite and wurzite structures). We find the spectral density and density of states of the dipole-active excitations for crystals with this sort of one-dimensional disorder and with a Coulomb dipole-dipole interaction. We show that because the excitation spectrum is nonanalytic, the exponential factor in the asymptotic density of states $\rho(\varepsilon) \sim \exp(-c_1|\varepsilon|)$ at frequencies in the fluctuation region differs from both the corresponding factor in the asymptotic spectral density $\mathscr{A}(\varepsilon,k) \sim \exp(-c_2|\varepsilon|^{3/2})$ and the asymptotic behavior of $\rho(\varepsilon)$ in the short-range interaction problem ($\varepsilon = \omega - \omega_0$ is the distance from the edge of the continuum).

Zinc sulfide crystals exhibit plane stacking faults due to coexistence of the two structural modifications of ZnS—sphalerite and wurzite. Sphalerite and wurzite layers oriented perpendicular to the hexagonal axis can alternate in a disordered manner. In this case there is one-dimensional disorder along a preferred axis.[1] One-dimensional disorder of this sort arises in many other systems as well.[2–5]

The experimental study of the excitonic reflection spectra in ZnS crystals containing stacking faults and the theoretical interpretation of these spectra[6] have shown that the observed shift and inhomogeneous broadening of the exciton line are described well by a model with one-dimensional disorder.

In the present study we consider the energy spectrum of long-wavelength dipole-active elementary excitations (excitons and optical phonons) in uniaxial crystals with one-dimensional disorder in cases where the Coulomb dipole–dipole interaction is important, leading to nonanalyticity of the spectrum. An additional feature of this problem is that the dipole-active excitations are described by a vector equation for the dipole moment density, unlike the scalar problems usually considered.[7]

We calculate the spectral density and density of states of the dipole-active excitations. In a number of cases the problem of evaluating the spectral density can be reduced to a scalar problem with a short-range interaction.[8] In contrast to Ref. 8, however, for a definite sign of the dispersion of the branches the leading contribution to the asymptotic density of states in the fluctuation region is from the long-wavelength states of the nonanalytic region of the spectrum. In this case one obtains an asymptotic density of states $\rho(\varepsilon)$ which differs from that for the standard one-dimensional problem.[8,9] Specifically,

$$\rho(\varepsilon) = a_3|\varepsilon|^{3/2} \exp(-a_1|\varepsilon|). \tag{1}$$

Here $\varepsilon = \omega - \omega_0$ is the distance from the edge of the continuum. We also find that the exponential factor in the asymptotic density of states (1) has a different energy dependence from that of the asymptotic spectral density $\mathscr{A}(\varepsilon, k_z = 0)$ found in this study:

$$\mathscr{A}(\varepsilon, k_z = 0) = a_4|\varepsilon|^{1/2} \exp(-a_2|\varepsilon|^{3/2}). \tag{2}$$

The phenomenological relations in this paper apply specifically to the optical phonon spectrum in a crystal of the ZnS type with stacking faults.

1. FORMULATION OF THE PROBLEM

In an ideal crystal the long-wavelength dipole-active excitations are described by Maxwell's equations together with the material equations of the medium. After **E** and **D** are eliminated from these equations, the equation for the polarization **P** becomes

$$\left\{ \chi_{ij}^{-1}(\omega, \mathbf{k}) + 4\pi \frac{k_i k_j}{k^2} + 4\pi \frac{\omega^2}{\omega^2 - c^2 k^2} \left[\delta_{ij} - \frac{k_i k_j}{k^2} \right] \right\} P_j(\omega, \mathbf{k}) = 0, \tag{3}$$

where the second and third terms in the braces give the separate contributions due to the longitudinal and transverse electric fields.

To take the dispersion explicitly into account we can expand the function $\chi_{ij}^{-1}(\omega, \mathbf{k})$, which is analytic in **k** (see Ref. 10, p. 47), in a series. To terms of second order we have

$$\chi_{ij}^{-1}(\omega, \mathbf{k}) = \chi_{ij}^{-1}(\omega) + i\gamma_{ijl}(\omega) k_l + \alpha_{ijlm}(\omega) k_l k_m. \tag{4}$$

This approach has certain advantages over the conventional approach of writing an equation for **E** (or **D**) and expanding the tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ [or $\varepsilon_{ij}^{-1}(\omega, \mathbf{k})$] in a series in **k**, since expansion (4) is valid for natural waves of arbitrary polarization.

In a crystal with disorder it is convenient to separate χ_{ij}^{-1} into an average value $\langle \chi_{ij}^{-1} \rangle$ and a fluctuating part $\delta\chi_{ij}^{-1}$. A crystal described by Eq. (3) with $\langle \chi_{ij}^{-1} \rangle$ substituted-in will henceforth be called a virtual crystal. In the case of one-dimensional disorder the fluctuations $\delta\chi^{-1}$ are functions of z and z'. In the Fourier representation we have

$$\begin{aligned} \delta\chi^{-1}(k_z, k_z') &= \int dz \exp(-ik_z z)\, \delta\chi^{-1}(z, z') \exp(ik_z' z')\, dz' \\ &= \int d\left(\frac{z+z'}{2}\right) \left[\exp\left\{ -i(k_z - k_z') \frac{z+z'}{2} \right\} \right. \\ &\quad \left. \times \int d(z-z') \exp\left\{ -i \frac{k_z + k_z'}{2} (z-z') \right\} \delta\chi^{-1}\left(\frac{z+z'}{2}, z-z' \right) \right]. \end{aligned} \tag{5}$$

The fluctuations $\delta\chi^{-1}$, like χ^{-1} itself, fall off rapidly as

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functions of the difference $z - z'$. In the long-wavelength limit ($k_z d \ll 1$, $k'_z d \ll 1$, where d is the distance between crystal planes) the integrand of the inner integral in (5) can therefore be expanded in powers of $(k_z + k'_z)$. In the case of small fluctuations $\delta\chi^{-1}$ it is sufficient to keep only the term of zeroth-order in $(k_z + k'_z)$ in the expansion of the inner integral in (5). In the outer integral, however, the argument of the exponential is large for any k_z or k'_z. This exponential must therefore be taken into account exactly.

Substituting expression (5) for $\delta\chi^{-1}$ in (3), transforming to a coordinate representation in z, and neglecting retardation, we obtain an equation for the natural waves in a crystal with one-dimensional disorder:

$$\Big\{ [\chi_{ij}^{-1}(\omega) + i\gamma_{ijl}(\omega) k_l + \alpha_{ijlm} k_l k_m] P_j(\omega, \mathbf{k}_\perp, z) + 4\pi \int dz' L_{ij}(z-z') P_j(\omega, \mathbf{k}_\perp, z') \Big\} + \delta\chi_{ij}^{-1}(\omega, z) P_j(\omega, \mathbf{k}_\perp, z) = 0, \tag{6}$$

where the operator

$$L_{ij}(z-z') = \int \frac{dk_z}{2\pi} \frac{k_i k_j}{k^2} \exp\{ik_z(z-z')\},$$

like the operator $k_i k_j / k^2$ in $\mathbf{k}$ space, acts on the vector $\mathbf{P}$ to extract its irrotational part; $\mathbf{k}_\perp = (k_x, k_y)$, $k_z = -i\partial/\partial z$.

With an eye toward applications to the ZnS crystal, let us consider the case in which the fluctuations, just as in the virtual crystal, are characterized by a uniaxial symmetry, although a different symmetry from that of the virtual crystal. Let us first consider the spectrum of the virtual crystal and then take fluctuations into account.

The spectrum of the virtual crystal in the small-$\mathbf{k}$ limit (i.e., without allowance for spatial dispersion) has the following properties. The frequency of a wave polarized perpendicular to $\mathbf{k}_\perp$ in the (x, y) plane (the s wave) is determined by the equation

$$\chi_\perp^{-1}(\omega) \equiv \chi_{xx}^{-1}(\omega) = \chi_{yy}^{-1}(\omega) = 0. \tag{7}$$

The solution of system (6) for a wave polarized in the plane passing through $\mathbf{k}_\perp$ and the z axis (a wave of p polarization) is given for different relationships between k_z and $\mathbf{k}_\perp$ by the roots of the following equations:

for $k_z \ll k_\perp$

$$\chi_\parallel^{-1}(\omega) \equiv \chi_{zz}^{-1}(\omega) = 0;$$

$$\mathbf{P} \perp \mathbf{k}, \quad \mathbf{P} = (0, 0, P_z), \tag{8a}$$

$$\chi_\perp^{-1}(\omega) + 4\pi = 0; \quad \mathbf{P} \| \mathbf{k},$$

$$\mathbf{P} = \left(P_\perp \frac{k_x}{k_\perp}, P_\perp \frac{k_y}{k_\perp}, 0 \right); \tag{8b}$$

for $k_z \gg k_\perp$

$$\chi_\perp^{-1}(\omega) = 0; \quad \mathbf{P} \perp \mathbf{k},$$

$$\mathbf{P} = \left(P_\perp \frac{k_x}{k_\perp}, P_\perp \frac{k_y}{k_\perp}, 0 \right), \tag{9a}$$

$$\chi_\parallel^{-1}(\omega) + 4\pi = 0;$$

$$\mathbf{P} \| \mathbf{k}; \quad \mathbf{P} = (0, 0, P_z). \tag{9b}$$

Thus the frequency of a p-polarized wave depends as $\mathbf{k} \to 0$ on the relationship between $k_\perp$ and k_z (on the direction of $\mathbf{k}$), i.e., has a nonanalytic dependence1) on $\mathbf{k}$. In the intermediate region $k_z \approx k_\perp$ the frequencies determined from Eqs. (8) and (9) are joined by smooth curves (with no intersections).

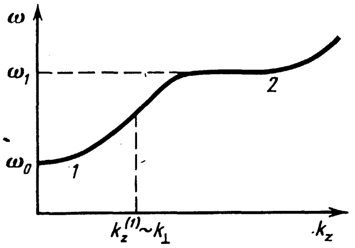

FIG. 1. The function $\omega(k_z)$ in the long-wavelength region of the spectrum: 1) nonanalyticity region, 2) spatial dispersion region.

For the sake of definiteness we shall consider the case when the anisotropy of the short-range interaction is small: $|\chi^{-1}(\omega) - \chi^{-1}(\omega)| \ll 1$. Then the branches of the spectrum join the frequencies ω_0 and ω_1, which are roots of Eqs. (8a) and (9a), and also $\tilde{\omega}_0$ and $\tilde{\omega}_1$, the roots of Eqs. (8b) and (9b). Figure 1 shows the spectrum $\omega(k_z)$ for the case in which the signs of the constants which determine the dispersion of the branch in the nonanalyticity region and in the short-wavelength region coincide; it is seen that the dimension of the nonanalyticity region of the spectrum $\omega(k_z)$ is proportional to $k_\perp$.

Let us turn to a calculation of the spectral density and density of states of the excitations under study.

2. LONG-WAVELENGTH EDGE OF THE NONANALYTICITY REGION ($k_z \ll k_\perp$)

The spectrum near ω_0 for a uniaxial virtual crystal is given to second order in $k_z/k_\perp \ll 1$ and $k_\perp d \ll 1$ by

$$\omega - \omega_0 = \frac{1}{2} A_0 \frac{k_z^2}{k_\perp^2} + \frac{1}{2} B_0 k_\perp^2, \tag{10}$$

where

$$A_0 = -2\left(\frac{\partial \chi_\parallel^{-1}}{\partial \omega}\right) \frac{4\pi}{1 + 4\pi\chi_\perp(\omega_0)},$$

$$B_0 = 2\left(\frac{\partial \chi_\parallel^{-1}}{\partial \omega}\right) \left[\frac{\chi_\perp(\omega_0)}{1 + 4\pi\chi_\perp(\omega_0)} \gamma_{xzx}^2 - \alpha_{zzxx} \right].$$

In the region under study we have $P_\perp \ll P_z$, and so we find from (6) with allowance for (10)

$$\Big\{ \omega - \omega_0 + \frac{1}{2k_\perp^2} A_0 \frac{\partial^2}{\partial z^2} - \frac{1}{2} B_0 k_\perp^2 + \left(\frac{\partial \chi_\parallel^{-1}}{\partial \omega}\right)^{-1} \delta\chi_\parallel^{-1}(z) \Big\} P_z(z) = 0. \tag{11}$$

For considering long-wavelength states we may treat the fluctuations $\delta\chi_\parallel^{-1}(z)$ as δ-correlated[7]:

$$(\partial\chi_\parallel^{-1}/\partial\omega)^{-2} \langle \delta\chi_\parallel^{-1}(z) \delta\chi_\parallel^{-1}(z') \rangle = D_\parallel \delta(z - z'). \tag{12}$$

Equation (11) with correlator (12) has the form of the Schrödinger equation with a "white noise" potential; this problem was studied in Refs. 8 and 9. As in Ref. 8, Eq. (10)

and correlator (11) are rendered simultaneously dimensionless by introducing a dimensionless frequency shift $\omega' = (\omega - \omega_c)/\Omega_0$ and dimensionless coordinate $z' = z/l_0$, where

$$\Omega_0 = D_{\parallel}^{2/3} A_0^{-1/3} k_{\perp}^{2/3} \tag{13}$$

is the scale of the decay of the spectral density and density of states, and

$$l_0 = D_{\parallel}^{-1/3} A_0^{2/3} k_{\perp}^{4/3} \tag{14}$$

is the scale of the localization length near the frequency $\omega_c = \omega_0 + \frac{1}{2} B_0 k_{\perp}^2$ which determines the long-wavelength edge of the spectrum for fixed $k_{\perp}$. The spectral density $\mathscr{A}(\omega', k_z')$ for arbitrary ω' and k_z' ($k_z' = k_z l_0$) was found numerically by Halperin,[8] who also established the asymptotic form of $\mathscr{A}(\omega', k_z')$. In particular, for $k_z' = 0$ and $\omega' > \omega_c$

$$\mathscr{A}(\omega', 0) = 2^{-1/2} \pi^{-1} (\omega')^{-5/2}, \tag{15.1}$$

and in the fluctuation region

$$\mathscr{A}(\omega', 0) = 2^{1/2} \pi |\omega'|^{1/2} \exp\left[-{}^4/_3 |2\omega'|^{3/2}\right]. \tag{15.2}$$

In a crystal with one-dimensional disorder it is convenient to introduce a density of states with fixed $k_{\perp}$. The asymptotic form of this function is the same as that in the standard one-dimensional problem[8,9] with the scale Ω_0 from (13) substituted-in:

$$\rho(\omega, k_{\perp}) = \frac{2|\omega - \omega_0 - {}^1/_2 B_0 k_{\perp}^2|}{\pi D_{\parallel}} \times \exp\left[-\frac{4A_0^{1/2}}{3D_{\parallel}} \left(\frac{2|\omega - \omega_0 - {}^1/_2 B_0 k_{\perp}^2|}{k_{\perp}^{2/3}}\right)^{3/2}\right]. \tag{16}$$

We see from (13) and (16) that because the spectrum is nonanalytic, the characteristic scale of the decay of the density of states with fixed k depends on $k_{\perp}$. The total density of states $\rho(\omega)$ is given as the integral of (16) over all $k_{\perp}$. At a fixed distance $|\omega - \omega_0|$ from the edge of the spectrum, for small values of $k_{\perp}$ the region of nonanalyticity and, consequently, the contribution of these $k_{\perp}$ to $\rho(\omega)$ will also be small. At large $k_{\perp}$ the long-wavelength edge moves away from ω_0, and this decreases the contribution of large $k_{\perp}$ to $\rho(\omega)$ at this frequency. Therefore, the integral of (16) over $k_{\perp}$ can be evaluated by the method of steepest descent:

$$\rho(\omega) = \frac{3^{1/4} |\omega - \omega_0|^{3/2}}{2^{3/2} \pi^{3/2} D_{\parallel}^{1/2} A_0^{1/4} B_0^{5/4}} \exp\left[-\frac{4(3A_0 B_0)^{1/2}}{D_{\parallel}} |\omega - \omega_0|\right]. \tag{17}$$

This expression differs from the asymptotic form of $\rho(\omega)$ in the standard one-dimensional problem because of certain aspects of the behavior of $\rho(\omega)$ in the nonanalytic spectrum of the virtual crystal (for $\omega \gtrsim \omega_0$). In fact, because of the nonanalyticity, the phase volume in $\mathbf{k}$ space which contributes to $\rho(\omega)$ near ω_0 is smaller than it would be in an analytic spectrum: Eq. (10) gives a small distance $(\omega - \omega_0)$ from the edge not only for k of small modulus but also in a narrow interval of angles θ in $\mathbf{k}$ space about $\theta = \pi/2$ (i.e., a small ratio $k_z/k_{\perp}$). Therefore, the singularity in $\rho(\omega)$ turns out to be weaker than the Van Hove singularity near the edge of the analytic spectrum ($\rho(\omega) \propto (\omega - \omega_0)^{1/2}$ in the three-dimensional case) and is of the form $\rho \propto (\omega - \omega_0)$. The slower growth of the density of states of the virtual crystal in the region $\omega > \omega_0$ leads to a slower decay of the density of states in the fluctuation region.

The asymptotic density of states near the branch joining the roots of Eqs. (8b) and (9b) differs from (17) (except in the case of a hexagonal virtual crystal) because the dispersion of the branch is anisotropic in the transverse direction. Specifically, in the expression for the spectrum of this branch

$$\omega = \tilde{\omega}_0 + \frac{1}{2} \tilde{A}_0 \frac{k_z^2}{k_{\perp}^2} + \frac{1}{2} \tilde{B}_0 k_{\perp}^2$$

the quantity $\tilde{B}_0 = \tilde{B}_0(\mathbf{k}_{\perp}/|k_{\perp}|)$ depends on the direction of $\mathbf{k}_{\perp}$. Integration over the directions of $\mathbf{k}_{\perp}$ by the method of steepest descent gives

$$\rho(\omega) \propto |\omega - \omega_0| \exp\left[-\frac{4(3\tilde{A}_0 \tilde{B}_{0\,min})^{1/2}}{D_{\perp}} |\omega - \omega_0|\right], \tag{18}$$

where $D_{\perp}$ is given by a relation analogous to (12).

We note that the presence of other, non-one-dimensional scattering mechanisms in real crystals (e.g., scattering by point defects or scattering with emission of acoustic phonons) should eliminate the one-dimensional localization of excitations. Nevertheless, if the damping Γ due to these mechanisms is sufficiently small the "one-dimensional" asymptotic forms obtained above for the spectral density and density of states are valid. In particular, asymptotic form (16) obtains for $\Gamma \ll \Omega_0$.

3. CONTINUUM REGION

The spectral density near the flat part of spectrum, i.e., in the region where $k_{\perp} \ll k_z$ and $\omega = \omega_1 + \frac{1}{2} B_1 k_{\perp}^2$ (see Fig. 1) can be evaluated in the limit $k_{\perp} \to 0$. Finding the spectrum of the virtual crystal from Eq. (6) to terms of $O(k^2)$ and dropping the terms containing the small quantity $k_{\perp}$, one can reduce the problem of evaluating the spectral density to the problem of short-range forces. The corresponding scales for the decay of the spectral density Ω_1 and localization length l_1 turn out to be different here than in (13) and (14)

$$\Omega_1 = D_{\perp}^{2/3} C_1^{-1/3}, \quad l_1 = D_{\perp}^{-1/3} C_1^{2/3}, \tag{19}$$

where

$$C_1 = 2(\partial \chi_{\perp}^{-1}/\partial \omega)^{-1} \alpha_{xxzz}.$$

In the present limit $k_{\perp} \to 0$ the frequency ω_1 is doubly degenerate. Here the spectral densities of the p and s waves coincide: $\mathscr{A}_{xx}(\omega, k_z) = \mathscr{A}_{yy}(\omega, k_z)$.

At finite $k_{\perp}$ the nonanalytic term in Eq. (6) begins to have an influence on the p wave. In this case the spectrum of the p wave in the virtual crystal is of the form

$$\omega = \omega_1 - \frac{1}{2} A_0 \frac{k_{\perp}^2}{k_{\perp}^2 + k_z^2} + \frac{1}{2} C_1 k_z^2. \tag{20}$$

To determine the boundaries within which one can use the results of the short-range problem, one should estimate the nonanalyticity correction to the frequency of the localized state. To first order this correction is

$$\Delta\omega = -\frac{\pi^2}{8} A_0 \frac{k_{\perp}}{\varkappa}, \quad \varkappa = \left(\frac{2|\omega_1 - \omega|}{C_1}\right)^{1/2}. \tag{21}$$

For

$$k_\perp \ll D_\perp (A_0 C_1)^{-1} \tag{22}$$

correction (21) is small compared to $|\omega_1 - \omega|$, and $\mathscr{A}_{xx}(\omega, k_z = 0)$ is therefore described by Halperin's numerical solution with parameters (19) at all frequencies except, first, in the narrow interval

$$|\omega-\omega_1| \ll (A_0 C_1)^{1/2} k_\perp \tag{23}$$

in which spectrum (20) is substantially nonparabolic and, second, in the far asymptotic reaches of the fluctuation region, where the exponentially decaying quantity $\rho(\omega, k_\perp)$ becomes smaller than the unperturbed density of states in the nonanalyticity region.

At large $k_\perp$, when

$$D_\perp (A_0 C_1)^{-1} \ll k_\perp \ll D_\perp^{2/3} A_0^{-1/2} C_1^{-5/6}, \tag{24}$$

the nonparabolicity of the spectrum becomes important at all frequencies, corresponding to both the diffusion and fluctuation regions. Thus in this case the problem reduces to finding the spectral density for a system with short-range forces and a nonparabolic spectrum. We shall not consider this problem here.

It can be shown, using spectrum (20), that in the region

$$\omega-\omega_1 \gg \Omega_1 \tag{25}$$

$\mathscr{A}(\omega, k_z)$ can be evaluated for all $k_\perp$ by perturbation theory. At still greater values

$$k_\perp \gg D^{2/3} A_0^{-1/2} C_1^{-5/6} \tag{26}$$

the minimum value of the derivative $\partial\omega/\partial k_z$ of spectrum (20) increases; near the frequency ω_1 there is no longer even an approximately flat part of the spectrum, and perturbation theory applies at all frequencies.

It should be noted that all these values of k are nonetheless small compared to the characteristic values of k_z for the localized states, which are of the order of the localization length l_1^{-1}. Therefore, all the states considered above are actually situated in frequency near ω_1.

Excitations whose frequency ω lies in the interval $\omega_0 < \omega < \omega_1$ (when $k_z \approx k_\perp$) at a sufficient distance from the frequencies ω_0 and ω_1 corresponding to the flat parts of the spectrum are weakly scattered by fluctuations $\delta\chi^{-1}$. Therefore, the spectral density tensor $\mathscr{A}_{ij}(\omega, \mathbf{k})$ expressed in terms of the single-particle polarization Green function differs little from the spectral density of the virtual crystal and can be evaluated by perturbation theory.

In an analogous way, the density of states to within parametrically small corrections is the same as the unperturbed density of states of the virtual crystal. This is also true at frequencies near ω_1, since the flat parts of the spectrum for different $k_\perp$ fall at different values of the frequency $\omega = \omega_1 + \frac{1}{2} B_1 k_\perp^2$ and therefore give only a small correction to the total density of states.

4. OPTICAL PHONONS IN ZnS CRYSTALS WITH STACKING FAULTS

In a ZnS crystal with stacking faults there is a random alternation of layers having cubic and hexagonal environments (i.e., having the sphalerite and wurzite structures). Such a crystal can be characterized by the fraction c of hexagonal layers arranged in an uncorrelated manner.[6] The stackings of the three nearest crystalline planes in the sphalerite and wurzite crystals are the same. Differences appear only for stackings of four or more planes. Consequently, the frequencies of optical phonons in the nearest-neighbor approximation are the same in cubic and hexagonal ZnS; a difference arises only when the next nearest neighbors are taken into account, and it is therefore small.

Accordingly, the susceptibilities of pure cubic $(\chi^{(c)})$ and pure hexagonal $(\chi^{(h)})$ crystals are

$$\chi_\perp^{(c)}(\omega) = \chi_\parallel^{(c)} = \frac{(Ze)^2}{\mu(\omega_t^2-\omega^2)}, \tag{27a}$$

$$\chi_{\perp,\parallel}^{(h)}(\omega) = \frac{(Ze)^2}{\mu(\omega_t^2+\Delta_{\perp,\parallel}-\omega^2)}, \tag{27b}$$

where Ze is the ion charge, μ is the reduced mass of the ZnS unit cell, ω_t is the frequency of the *TO* phonon in cubic ZnS; $\Delta_\perp, \Delta_\parallel \ll \omega_t^2$.

The invese susceptibility of the virtual crystal is given by the average of χ^{-1} over the modifications:

$$\chi_{\perp,\parallel}^{-1}(\omega) = \frac{\mu}{(Ze)^2}(\omega_t^2 + c\Delta_{\perp,\parallel} - \omega^2), \tag{28}$$

and the quantities $D_\perp$ and $D_\parallel$ appearing in (12)–(26) are now

$$D_{\perp,\parallel} = c(1-c)\Delta_{\perp,\parallel} d/4\omega^2, \tag{29}$$

where d is the distance between crystalline planes of the same sublattice.

It is a peculiarity of ZnS that the nonanalyticity region, which is absent in one of the pure components (the cubic), is formed only on account of an admixture of the other component. The dispersion of the branch in the nonanalyticity region

$$\omega_1-\omega_0 = c(\Delta_\perp-\Delta_\parallel)/2\omega_t \ll \omega_t$$

is parametrically small by virtue of the small difference in the eigenfrequencies of the initial pure components.

The presence of stacking faults is manifested experimentally in a broadening of the Raman line, for example. Upon excitation of a phonon of s polarization, the fluctuation contribution to the Raman linewidth should, according to (19) and (29), depend on the concentration of defects in a manner characteristic of one-dimensional disorder: $\Gamma_s \approx \Omega_1 \propto [c(1-c)]^{2/3}$. For a phonon of p polarization the nonanalyticity of the spectrum causes the linewidth to depend substantially on the angle θ between the z axis and the direction of the **k** momentum transferred to the phonon. For $\theta \neq 0, \pi/2$ perturbation theory calculations give

$$\Gamma_p = \frac{k\sin\theta(1-c)}{4\omega_t(\Delta_\perp-\Delta_\parallel)}\left[\Delta_\perp^2\frac{\cos^3\theta}{\sin^3\theta} + \Delta_\parallel^2\frac{\sin\theta}{\cos\theta}\right], \quad \Gamma_p \ll \omega_1-\omega_0. \tag{30}$$

The unusual density dependence of Γ_p arises because the density c of the hexagonal layers determines both the correlator of the fluctuations, i.e., (29), and the dispersion of the branch in the nonanalyticity region.

Estimates of the maximum fluctuational linewidth of the optical phonon (for $c = 0.5$) give $\Gamma_s \approx 1 \text{ cm}^{-1}$, a value comparable to the anharmonic linewidth in these crystals.

[1)] Nonanalytic behavior of $\omega(k)$ was first found in Ref. 11 for the phonon spectrum of ideal diatomic ionic crystals. For the virtual crystal under study the nonanalyticity is due to the anisotropy of the initial components.

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