Light scattering in an incommensurate phase

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The integrated and spectral intensities for light scattering due to fluctuations of the order parameter in an incommensurate phase are evaluated in the framework of the phenomenological theory. It is found that this scattering differs from that in other degenerate systems as well as that near ordinary phase-transition points.

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The spectrum of elementary excitations of an incommensurate phase has a number of features (see, for example, Ref. 1–3), the most important of which is the presence of a Goldstone branch of excitations (a phason), which is characteristic of degenerate systems. Although there have been attempts to investigate these features experimentally by light-scattering techniques (see, for example, Refs. 4–6), there has been practically no theoretical treatment of light-scattering in an incommensurate phase, with the exception of several conclusions of a qualitative nature based on symmetry ideas.[7] We shall show below that the light-scattering properties of the incommensurate phases formed during structural transitions in crystals are different from those of other degenerate systems such as liquid crystals.[8]

For simplicity we examine the region near the transition point from the incommensurate to the disordered (high-temperature) phase. In this case one cannot take the incommensurate superstructure into account in first approximation in the formulas[9] relating the light-scattering intensity to fluctuations of the tensor ϵ_{ik}, since only the higher-order approximations in powers of the order parameter affect the propagation of light.[10]

The way that the above-mentioned elementary excitations are manifested in light-scattering is determined by the nature of the dependence of the tensor ϵ_{ik} on the order parameter; it is convenient to treat the incommensurate phase as a spatial modulation of the order parameter, as was done in Refs. 11 and 12. We treat as an example the incommensurate phase of ammonium fluoroberyllate.[12] The dependence of ϵ_{ik} on the components of the order parameter η and ξ is given by the expression[10]

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$$\epsilon_{ii} = \epsilon_{ii}^{(o)} + a_i \rho^2 , \quad \epsilon_{xy} = b \rho^2 \cos 2\phi ,$$

where ρ and ϕ are determined by the relations $\eta = \rho \cos\phi$ and $\xi = \rho \sin\phi$.

The intensity of scattered light with ($I_{\parallel}$) and without ($I_{\perp}$) a change in the polarization is, by analogy with Ref. 9:

$$I_{\parallel} (\mathbf{q}) \sim < | \Delta \epsilon_{ii} (\mathbf{q}) |^2 > = 4 a_i^2 \rho_o^2 < | \rho (\mathbf{q}) |^2 >,$$

$$I_{\perp} (\mathbf{q}) \sim < | \Delta \epsilon_{xy} (\mathbf{q}) |^2 > = b^2 \rho_o^2 \{ < | \rho (\mathbf{q} - 2\mathbf{k}_o) + \rho (\mathbf{q} + 2\mathbf{k}_o) |^2 >$$

$$+ \rho_o^2 < | \phi (\mathbf{q} - 2\mathbf{k}_o) - \phi (\mathbf{q} + 2\mathbf{k}_o) |^2 > + 2 \rho_o \operatorname{Im} < [\rho (\mathbf{q} - 2\mathbf{k}_o) + \rho (\mathbf{q} + 2\mathbf{k}_o)]$$

$$\times [\phi^* (\mathbf{q} - 2 k_o) - \phi^* (\mathbf{q} + 2\mathbf{k}_o)] > \} . \qquad (1)$$

Where $\rho(\mathbf{q})$ and $\phi(\mathbf{q})$ are the Fourier components of the fluctuations of ρ and ϕ, κ_0 is the wave vector of the superstructure, and ρ_0 is the equilibrium value of ρ.

Using the thermodynamic potential of Ref. 12 and proceeding in the usual way,[9] we find in first approximation in ρ_0^2

$$< | \rho (\mathbf{q}) |^2 > = \frac{k_B T}{V} \frac{1}{2 \beta_1 \rho_o^2 + \delta_1 q_x^2 + \delta_2 q_y^2 + \delta_3 q_z^2} , \quad < | \phi (\mathbf{q}) |^2 >$$

$$= \frac{k_B T}{V} \frac{1}{\rho_o^2 (\delta_1 q_x^2 + \delta_2 q_y^2 + \delta_3 q_z^2)} . \qquad (2)$$

The fact that the fluctuations of the phase ϕ diverge at $\mathbf{q} = 0$ is a manifestation of the degeneracy of the system; there is an additional divergence at the phase transition point where, ρ_0^2.

Substituting Eq. (2) into Eq. (1) and taking into account that $\mathbf{q} \approx 0$ for light, we have

$$I_{\parallel} (\mathbf{q} \approx 0) \sim < | \Delta \epsilon_{ii} (\mathbf{q} \approx 0) |^2 > = \frac{2 a_i^2 k_B T}{\beta_1 V} , \quad I_{\perp} (\mathbf{q} \approx 0) \sim < | \Delta \epsilon_{xy} (\mathbf{q} \approx 0) |^2 >$$

$$= \frac{b^2 \rho_o^2 k_B T}{\delta_1 k_o^2 V} . \qquad (3)$$

The expression for $I_{\parallel}$, to which only the amplitudon (fluctuation of ρ) contributes, coincides with the expression for the light-scattering intensity of fluctuations of the order parameter in the usual case.[9] Of greater interest is $I_{\perp}$, which contains contributions from both the amplitudon and the phason (fluctuation of ϕ). Although the fluctuations of the phase for $\mathbf{q}=0$ are infinite, the intensity $I_\perp$ is finite and depends weakly on $\mathbf{q}$; this distinguishes an incommensurate phase from nematic and smectic liquid crystals. Physically, this is due to the fact that the scattering is governed by a phason with wave vector $\mathbf{q}\pm 2\mathbf{k}_0$ rather than $\mathbf{q}$. The situation is somewhat similar to that in cholesteric liquid crystals, inasmuch as the structure of these crystal can also be treated as a spatial periodicity of the order parameter. However, in contrast to the case at hand, in cholesteric liquid crystals the tensor ϵ_{ik} is coupled linearly to the order parameter, and for them $k_0 \sim q$, leading to a different dependence of the intensity on the scattering geometry and temperature.[8]

Unlike $I_\perp$, the quantity $I_\parallel$ of Eq. (3) is equal to zero at the phase transition point, but because k_0 is small it grows rather rapidly as the temperature is lowered. Thus, light-scattering involving a phason appears only when the system is not too close to the transition point, in contradiction to the conjectures of Refs. 4 and 6.

To evaluate the spectral density of the fluctuations $\Delta\rho$ and $\Delta\phi$ we write the equation of motion (in first approximation) as

$$\mu\frac{\partial^2(\Delta\rho)}{\partial t^2}+\nu\frac{\partial(\Delta\rho)}{\partial t}-\delta_1\frac{\partial^2(\Delta\rho)}{\partial x^2}-\delta_2\frac{\partial^2(\Delta\rho)}{\partial y^2}-\delta_3\frac{\partial^2(\Delta\rho)}{\partial z^2}+2\beta_1\rho_0^2\Delta\rho$$

$$=h_1(\mathbf{r},\ t),$$

$$\mu\frac{\partial^2(\Delta\phi)}{\partial t^2}+\nu\frac{\partial(\Delta\phi)}{\partial t}-\delta_1\frac{\partial^2(\Delta\phi)}{\partial x^2}-\delta_2\frac{\partial^2(\Delta\phi)}{\partial y^2}-\delta_3\frac{\partial^2(\Delta\phi)}{\partial z^2}=h_2(\mathbf{r},t)$$

where μ is the effective mass, ν the coefficient of viscosity, and h_1 and h_2 are generalized forces. Using the standard procedure,[9] we obtain the spectral density of the scattered light

$$J_\parallel(\mathbf{q}\approx 0,\Omega)=2\beta_1\nu\rho_0^2 I_\parallel(\mathbf{q}\approx 0)/\pi\,[(2\beta_1\rho_0^2-\mu\Omega^2)^2+\nu^2\Omega^2],\tag{4}$$

$$J_\perp(\mathbf{q}\approx 0,\Omega)=\frac{2\nu\delta_1 k_0^2 I_\perp(\mathbf{q}\approx 0)}{\pi}\left[\frac{1}{(4\delta_1k_0^2+2\beta_1\rho_0^2-\mu\Omega^2)^2+\nu^2\Omega^2}+\frac{1}{(4\delta_1k_0^2-\mu\Omega^2)^2+\nu^2\Omega^2}\right].\tag{5}$$

The Raman components (4), which correspond to the amplitudon, behave just as in the case of the usual soft modes.[9] Expression (5) has a different dependence on Ω and temperature. Near the phase transistion point the denominators in Eq. (5) coincide, and if $\nu^2/\mu > 8\delta_1 k_0^2$, then $J_\perp(0,\Omega)$ will have only a central peak of width $\sim 8\delta_1 k_0^2/\nu$.. As the temperature is lowered, two lateral components can break away from the central peak as a result of the first term in Eq. (5); these correspond to an amplitudon with wave vector $2\mathbf{k}_0$. If $\nu^2/\mu < 8\,\delta_1 k_0^2$, one should see two lateral components near the transition point (instead of the central peak) which each split into two as the temperature is lowered. The inequalities $\nu^2/\mu \gtrless 8\delta_1 k_0^2$ can be put in the form $\nu/\mu\omega_a \gtrless 2(\sqrt{2})\, k_0/k_b$, by introducing the characteristic phonon frequency $\omega_a \sim 10^{13}$ Hz and the wave vector of the Brillouin zone boundary k_b and taking into account that $\delta_1 k_b^2/\mu \sim \omega_a^2$. The damping factor ν/μ of an optical phonon is usually $\sim(1\text{–}10^{-1})\omega_a$), and the ratio k_0/k_b is $\sim 10^{-1}\text{-}10^{-2}$, so that in different cases either inequality can be satisfied. In cholesteric liquid crystal there is only a central peak.[8]

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