Effect of the finite size of the scattering region on the light-scattering and neutron-scattering line shape near a second-order phase transition

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The scattering of light and neutrons by fluctuations of the order parameter is considered. As usual, the fluctuations relax as a result of a certain diffusion process. In those conditions in which the diffusion-relaxation length becomes greater than the size of the scattering region the scattering lineshape changes—instead of a broad, smeared-out line, a narrow central peak appears.

The correlator of the fluctuations of the order parameter near the point of a second-order phase transition has, in the Landau theory, the following dependence on the frequency ω and wave vector $\mathbf{q}$ (Ref. 1):

$$G_{\omega,\mathbf{q}}=[\omega^2+(\alpha\tau+\beta q^2)^2]^{-1}, \quad \tau=|(T-T_c)/T|, \tag{1}$$

where T is the temperature, T_c is the transition temperature, and α and β are phenomenological constants. This dependence on the wave vector is analogous to a diffusion process of relaxation of the order parameter.

If a change of the order parameter induces a change of the permittivity or density, then light or neutrons, respectively, are scattered by fluctuations of the order parameter. Here it is assumed that the scattering cross section and extinction coefficient obey the dependence (1), where ω is the frequency transfer and $\mathbf{q}$ is the wave-vector transfer:

$$\mathbf{q}=\mathbf{K}-\mathbf{K}' \tag{2}$$

($\mathbf{K}$ and $\mathbf{K}'$ are the wave vectors of the incident and scattered waves). In addition, the scattering cross section is proportional to the volume of the scattering system. Since the characteristic size L of the system is much greater than the scattering wavelength $1/q$, it would seem that the corrections to the dependence (1) should be extremely small. We shall show, however, that this is not so, and that in certain cases the scattering lineshape differs substantially from the form (1).

The frequency dependence of the scattering line is determined by the expression[2]

$$J=\int d^3k G_{\omega,\mathbf{k}} \left| \int e^{i(\mathbf{q}-\mathbf{k})\mathbf{r}} \frac{d^3r}{(2\pi)^3} \right|^2. \tag{3}$$

The usual result is obtained when the square of the modulus of the integral over the scattering volume in this expression is taken to be equal to $(L/2\pi)^3\delta(\mathbf{q}-\mathbf{k})$. We shall perform the analysis of the full dependence on the size of the region for a spherical region of radius L:

$$J=\frac{L^2}{4\pi^4}\int \frac{G_{\omega,\mathbf{k}}}{|\mathbf{q}-\mathbf{k}|^4}\left\{\frac{\sin|\mathbf{q}-\mathbf{k}|L}{|\mathbf{q}-\mathbf{k}|L}-\cos|\mathbf{q}-\mathbf{k}|L\right\}^2 d^3k. \tag{4}$$

Substituting the expression (1) we perform the integration over the angles of $\mathbf{k}$, after which the expression will depend only on the modulus q of the wave vector (and, of course, on the frequency):

$$J=\frac{L^2}{4\pi^3\beta\omega q}\int_{-\infty}^{+\infty}(k-q)^{-3}\operatorname{arctg}\left(\frac{\alpha\tau+\beta k^2}{\omega}\right) \times\left[\frac{\sin(k-q)L}{(k-q)L}-\cos(k-q)L\right]^2 dk. \tag{5}$$

One can verify that this integral can be calculated exactly, but the full expression turns out to be cumbersome and difficult to picture. We shall describe it only in the rather interesting case when

$$\beta q^2 \gg \omega,\ \alpha\tau. \tag{6}$$

The condition (6) is necessary but not sufficient for the existence of substantial corrections to the dependence (1); when the opposite inequalities are fulfilled all the corrections to (1) are small.

If the inequality (6) is fulfilled, the expression for J consists of three terms. The first of them is the usual term

$$J_1=G_{\omega,q}L^3/6\pi^2=L^3\{6\pi^2[\omega^2+(\alpha\tau+\beta q^2)^2]\}^{-1}. \tag{7}$$

We note that by virtue of (6) we should neglect its frequency dependence, so that J_1 reduces to the constant $L^3/6\pi^2 \times(\beta q^2)^2$. The second term

$$J_2=\frac{2^{1/2}L^2}{8\pi^2(\beta q^2)^2}\frac{\beta^{1/2}}{[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]^{1/2}}. \tag{8}$$

The corrections to this expression are either small because of the inequality (6) or small in the small parameter $1/qL$.

There is also a third term, containing rapidly oscillating terms:

$$J_3=(4\pi^2\beta q^4)^{-1}L\exp\left\{-\left(\frac{2L^2}{\beta}\right)^{1/2}[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]^{1/2}\right\}\left\{\cos 2qL\sin\left(\frac{2L^2}{\beta}\right)^{1/2}[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]^{-1/2} -3\sin 2qL\cos\left(\frac{2L^2}{\beta}\right)^{1/2}\omega[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]^{-1/2}\{2(\beta q^2)^{1/2}[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]^{1/2}\}^{-1}\right\}. \tag{9}$$

We can convince ourselves that the maximum value of J_3, according to (6), is always smaller than the largest of the terms J_1 and J_2.

The terms J_1 and J_2 can be obtained easily in the following way: The integrand in formula (5) as a function of the

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integration variable k has maxima, and it is easy to take the integral approximately by taking into account only the contribution of each of the maxima. The maximum at $k_1 = q$ corresponds to the usual Bragg diffraction conditions; it is this maximum which gives the usual term J_1 (see (7)). However, when the inequality (6) is fulfilled there is also another maximum at the point

$$k_2=\{[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]/\beta\}^{1/2}.$$

The contribution of this additional maximum corresponds to non-Bragg diffraction, and therefore J_2 has a different dependence on the size of the scattering region.

We now write out these terms near the transition point for frequencies $\omega \gg \alpha\tau$:

$$J_1+J_2=\frac{L^3}{6\pi^2(\beta q^2)^2}+\frac{2^{1/2}L^2\beta^{1/2}}{8\pi^2(\beta q^2)^2\omega^{1/2}}. \qquad (10)$$

We arrive at the criterion that forms the central point of our analysis:

$$1)\ L>(\beta/\omega)^{1/2};\quad 2)\ L<(\beta/\omega)^{1/2}. \qquad (11)$$

In the case 1) the term J_1 is decisive and all the corrections to it are small. In the case 2) the lineshape is determined just by the term J_2, while the constant J_1 is small in comparison. The term J_3 in this case is found to be exponentially small. The criterion (11) is entirely analogous to the mesoscopic criteria for an electron system[3]: The effect is manifested when the size of the scattering region is comparable with the diffusion-relaxation length of the order parameter. In the case 2) the shape of the central peak corresponds to a square-root increase of the intensity with decrease of the frequency. This increase stops at frequencies ω of the order of $\alpha\tau$, while, according to (8), in formula (10) it is necessary to replace ω by $\alpha\tau$.

It should be said that the restriction to the Landau theory is not essential for our analysis. It can be seen from the simplified derivation of the term J_2 that for this term to be present it is necessary that $G_{\omega,k}$ be inversely proportional to k^4. In the scale-invariant theory of phase transitions, instead of k^4 we have $k^{4-2\eta}$, where η is the Fisher index. It is well known that the Fisher index is always numerically small. We can neglect it, after which it is always possible to guarantee the presence of a term of the type J_2. We cannot determine the power of the frequency appearing in this term, but it is clear that when the criterion (11) is fulfilled this term is large.

The above analysis can be applied to one- and two-dimensional systems, in which, strictly speaking, a second-order phase transition does not occur but there are order-parameter correlations extending over large distances. These correlations are described by formula (1) with a wave vector of the appropriate dimensionality, and this is entirely adequate for our analysis. When the inequality (6) is fulfilled we have for $d = 1$

$$J=\frac{L}{\pi(\beta q^2)^2}+\{2\pi 2^{1/2}q^2\beta^{1/2}(\omega^2+\alpha^2\tau^2)^{1/2}[(\omega^2+\alpha^2\tau^2)^{1/2}+\alpha\tau]^{1/2}\}^{-1}, \qquad (12)$$

and for $d = 2$

$$J=\frac{L^2}{4\pi(\beta q^2)^2}+\frac{L}{4\pi^2\beta q^3}\frac{\operatorname{arctg}\omega/\alpha\tau}{\omega}. \qquad (13)$$

Comparison of the terms in these expressions makes it possible to extend the criterion (11) to low dimensionalities.

The frequency dependences (8), (12), and (13) can be observed experimentally if the frequency $\omega_c = \beta L^2$ determined by the criterion (11) is not smaller than the instrumental frequency width of the line. Unfortunately, the parameter β for structural and other second-order phase transitions is not very well known. Nevertheless, it is clear that it has the structure

$$\beta=l^2/t_r, \qquad (14)$$

where t_r is the relaxation time of the order parameter and l is the size of the region over which spatial dispersion is manifested. Taking for these quantities the values $t_r = 10^{-13}$ sec, $l = 4\cdot 10^{-7}$ cm, and $L = 0.1$ cm, we obtain $\omega_c = 160$ Hz. It is necessary to note one further circumstance: Near the point of a second-order phase transition the relaxation of the parameter is slowed down, and this improves the opportunities for experiment. This fact, however, is not described by the simple phenomenological theory. Observation of the dependence of the light-scattering linewidth on the size of the scattering region can, in certain cases, assist in the analysis of the appearance of such phenomena as the light-scattering central peak.

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