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Critical dynamics of displacive structural phase transitions and light and neutron scattering below T_c

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Abstract. The critical dynamics of the displacive structural phase transitions with the short-range interaction and one-component order parameter in the scaling region below T_c has been studied.

The two-point retarded correlators of the order parameter and its square describing the spectral shape of light and neutron scattering have been calculated. It is shown that the observed anomalies in the low-frequency scattering near T_c are caused by the critical slowing down of two-particle collective excitations—the fluctuations of the phonon density, and their coupling with soft phonons.

1. Introduction

Structural transitions in weakly anharmonic crystals are referred to as displacive transitions when lattice distortions result from the 'freezing' of atomic displacements corresponding to some phonon coordinate. These transitions are singled out in a special class, mainly owing to the dynamics of the order parameter fluctuations. In the case of displacive transitions these fluctuations, unlike other known types of phase transitions, are propagating excitations (at least away from T_c), i.e. soft optical phonons. The lines with decreasing (at $T \to T_c$) frequency (the soft modes) corresponding to these excitations are found in the light and neutron scattering spectra in many crystals undergoing second-order phase transitions (Scott 1974, Fleury and Lyons 1981). However, the concept of quasi-harmonicity of soft phonons appeared to be invalid near T_c, since in approaching the transition point the soft-mode frequency does not tend to zero and it is smeared out accompanied by the formation and growth of the dynamic central peak; this is especially evident in the light scattering spectra (Fleury and Lyons 1981). These changes in the low-frequency spectrum may be qualitatively described as a result of the coupling of soft phonons with some relaxing degree of freedom (Ginzburg *et al* 1980), but it seems impossible to interpret this sensibly since the relaxation processes really occurring in solids such as the diffusion of impurities and thermal conductivity are extremely slow and the anomalies in the scattering spectrum up to Raman frequencies cannot account for this influence.

At the same time, together with the fluctuations of the order parameter φ for soft phonons, there are some other slow critical excitations with characteristic phonon frequencies—the φ^2 fluctuations. (We shall consider only the transitions with one-component φ.) Moreover, the critical light scattering results from dielectric tensor fluctuations of the following form:

$$\delta\varepsilon(x) = a[\varphi^2(x) - \langle\varphi^2(x)\rangle] \equiv a\,\delta\varphi^2(x)$$

where a is an invariant tensor and the term linear in φ is, as a rule, prohibited by the symmetry of a disordered phase. The validity of this relation at all temperatures should be emphasised (note that the disordered phase symmetry group also defines a φ-dependence of the Landau–Ginzburg Hamiltonian at all T). At $T > T_c$, such $\delta\varepsilon$ implies two-particle scattering which can be described in the mean-field region as difference and summation two-phonon processes. Then the appearance of the soft mode below T_c results from the linearisation of $\delta\varphi^2$, as $\varphi_s \equiv \langle\varphi\rangle$ is non-zero:

$$\delta\varphi^2 = \delta\varphi'^2 + 2\varphi_s\varphi' \qquad \varphi' = \varphi - \varphi_s \qquad \delta\varphi'^2 = \varphi'^2 - \langle\varphi'^2\rangle.$$

Thus, at $T < T_c$ the tensor of full intensity scattering turns into the sum of the one- and two-particle contributions along with the inference term:

$$I(k) \sim \langle\delta\varepsilon(k)\,\delta\varepsilon^*(k)\rangle = a^2 \int d^3x\,\exp(\mathrm{i}\,kx)$$
$$\times\,[\langle\delta\varphi'^2(x)\,\delta\varphi'^2(0)\rangle + 4\varphi_s\langle\varphi'(x)\,\delta\varphi'^2(0)\rangle + 4\varphi_s^2\langle\varphi'(x)\varphi'(0)\rangle].$$

In the mean-field region $T_c - T \gg T_c\mathrm{Gi}$ (Gi is the Ginzburg parameter (Patashinsky and Pokrovsky 1982)) the last one-phonon term (soft-mode intensity) becomes much larger than the intensity of the two-phonon summation and difference scattering processes $\langle\delta\varphi'^2(x)\,\delta\varphi'^2(0)\rangle$ and the interference term $\varphi_s\langle\varphi'(x)\,\delta\varphi'^2(0)\rangle$. In this region, simple perturbation theory estimates give

$$\varphi_s \int d^3x\,\langle\varphi'(x)\,\delta\varphi'^2(0)\rangle \sim \int d^3x\,\langle\delta\varphi'^2(x)\,\delta\varphi'^2(0)\rangle$$
$$\sim \left(\frac{T_c\mathrm{Gi}}{T_c - T}\right)^{1/2} \varphi_s^2 \int d^3x\,\langle\varphi'(x)\varphi'(0)\rangle.$$

It follows from this estimate that, on the boundary of the scaling region $T_c - T \sim T_c\mathrm{Gi}$, the contributions from one- and two-particle processes become of the same order. As shown by Sakhnenko and Timonin (1983), the merging of the thresholds of difference and summation two-phonon processes in the scaling region at $T > T_c$ gives rise to new two-particle excitations—phonon package density fluctuations—the critical slowing down of which leads to the appearance of an abnormally growing central dynamic peak in the Raman spectrum of the disordered phase. In the critical region below T_c the spectrum will represent the superposition of the contributions, which are comparable in magnitude, produced by the scattering by soft phonons and by the fluctuations of the phonon density, and it seems reasonable to suppose that this superposition results in the soft-mode smearing observed in the Raman spectra near T_c.

Similar but less pronounced anomalies in neutron scattering spectra may also be accounted for by the influence of the phonon density fluctuations. The immediate scattering of neutrons by soft phonons, with larger transferred wavevectors occurring both above and below T_c, indicates the deviations from quasi-harmonicity in the soft phonon's dynamics directly observed in this case. These deviations, which manifest themselves in the appearance of the anomalous low-frequency contributions, and the broadening and saturation of soft mode may be interpreted as the results of the interaction of soft phonons with the phonon density fluctuations, this interaction increasing near T_c owing to the convergence of their characteristic frequencies.

This concept of the critical dynamics of displacive phase transitions and of the spectra anomalies in the scaling region was considered by Bruce and Cowley (1980) in the framework of the dynamic scaling hypothesis. However, such a qualitative approach is obviously insufficient to interpret the experimental data which are indicative of the existence of various characteristic times in the critical fluctuation dynamics. In such cases the quantitative evaluation of the spectra shape requires the calculation of the two-point retarded correlators of the order parameter and its square in the scaling region. In the present work such calculations at $T < T_c$ have been carried out for the displacive structural phase transitions with one-component order parameters by the renormalisation group technique in three dimensions using the results of Sakhnenko and Timonin (1983) for $T > T_c$.

2. Temperature vertices and correlators below T_c

The structural phase transition with the one-component order parameter φ proportional to some phonon coordinate may be described by the following effective Hamiltonian:

$$\mathcal{H} = \int d^3x \left(\frac{\pi^2(x)}{2M} + \tfrac{1}{2}\tau\varphi^2(x) + \tfrac{1}{2}[\nabla\varphi(x)]^2 + \tfrac{1}{4}u\varphi^4(x)\right) \tag{2.1}$$

where $\pi(x)$ is the momentum density field canonically conjugated with the fluctuation field of order parameter $\varphi(x)$ and $\tau \sim T - T_c$.

Following the approach of Sakhnenko and Timonin (1983), we shall also utilise quantum formalism with its rather simple diagram technique for calculating Matsubara's temperature correlators. In the retarded correlators obtained by means of analytical continuation from the discrete imaginary frequencies, we shall proceed to the classical limit in the region of small frequencies $\omega \ll T_c\hbar^{-1}$ which we intend to study. At the same time, it should be noted that the absence of the coupling of $\varphi(x)$ with the acoustical phonons and energy density fluctuations in $\mathcal{H}$ (equation (2.1)) limits the range of applicability of the present theory to frequencies much greater than the characteristic acoustic and thermal frequencies.

The phase transition described by the quantum Hamiltonian $\mathcal{H}$ (equation (2.1)) with the conjugate field $\pi(x)$ and $\varphi(x)$ is the Bose condensation of $\varphi(x)$. Subtracting the classical condensate $\varphi = (1/V)\int d^3x\, \varphi(x)$ from $\varphi(x)$, $\varphi(x) = \varphi + \varphi'(x)$, we obtain

$$\mathcal{H} = VF_0(\tau, \varphi) + \mathcal{H}'$$

$$F_0(\tau, \varphi) = \tfrac{1}{2}\tau\varphi^2 + \tfrac{1}{4}u\varphi^4 \tag{2.2}$$

$$\mathcal{H}' = \int d^3x \left(\frac{\pi^2}{2M} + \tfrac{1}{2}(\tau + 3u\varphi^2)\varphi'^2 + \tfrac{1}{2}(\nabla\varphi')^2 + u\varphi\varphi'^3 + \tfrac{1}{4}u\varphi'^4\right) \tag{2.3}$$

$$F(\tau, \varphi) = F_0(\tau, \varphi) - (1/\beta V)\ln\{\mathrm{Tr}[\exp(-\beta\mathcal{H}')] \qquad \beta = T^{-1}. \tag{2.4}$$

The thermodynamic potential density $F(\tau, \varphi)$ determines the static properties of our model, and in particular the values of the spontaneous order parameter $\varphi_s = \varphi_s(\tau)$ and the susceptibility $\chi(\tau)$:

$$\partial F(\tau, \varphi_s)/\partial\varphi_s = 0 \qquad \chi^{-1}(\tau) = \partial^2 F(\tau, \varphi_s)/\partial\varphi_s^2. \tag{2.5}$$

To describe the dynamic properties of the model in the ordered phase ($T < T_c$, $\varphi_s \neq 0$), it is necessary to find Matsubara's correlators of the field $\varphi'(x)$ with the density matrix $\exp(-\beta\mathcal{H}')$ where, in $\mathcal{H}'$ (equation (2.3)), $\varphi = \varphi_s$. However, we shall consider the correlators at arbitrary φ that enable us to use their values at $\varphi = 0$ (Sakhnenko and Timonin 1983) and to find the equation of state (2.5) and $\varphi_s(\tau)$.

Then we consider the following one-particle irreducible vertex functions:

$$\left[\Gamma_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n) - \frac{\partial^{m+n} F_0(\tau, \varphi)}{\partial\varphi^m\,\partial\tau^n}\right] \delta\left(\sum_{i=1}^{m} P_i + \sum_{j=1}^{n} K_j\right)$$
$$= \frac{(-1)^{n+1}}{2^n \beta V}\left\langle T_\sigma\left(\prod_{i=1}^{m} G^{-1}(P_i)\int \mathrm{d}^4X_i \exp[\mathrm{i}(P_iX_i)]\,\varphi'(X_i)\prod_{j=1}^{n}\int \mathrm{d}^4Y_j \right.\right.$$
$$\left.\left.\times \exp[\mathrm{i}(K_jY_j)]\,\varphi'^2(Y_j)\right)\right\rangle'_{1\mathrm{PI}} \tag{2.6}$$

$$G(P) = \int \mathrm{d}^4X \exp[\mathrm{i}(PX)]\langle\varphi'(X)\varphi'(0)\rangle' \tag{2.7}$$

$$X = \{x, \sigma\} \qquad 0 < \sigma < \beta \qquad P = \{p, \omega_l\} \qquad (PX) = px - \sigma\omega_l$$

$$\varphi'(X) = \exp(\sigma\mathcal{H}')\,\varphi'(x)\exp(-\sigma\mathcal{H}').$$

The angular brackets with the prime on the right-hand side of (2.6) denote averaging with the density matrix $\exp(-\beta\mathcal{H}')$ and the subscript 1PI to these angular brackets denotes that the diagrams that can be cut into two parts across internal line and those without internal lines (their contribution corresponds to the term $\partial^{m+n}F_0(\tau, \varphi)/(\partial\varphi^m\,\partial\tau^n)$ in the left-hand side of (2.6)) are absent in the corresponding sum of the diagrams with m tails and n angles.

In the definition of vertices (equation (2.6)), m and n are arbitrary non-negative integers, except that $(m, n) = (0, 0), (1, 0), (2, 0)$. The vertex $\Gamma_{1,0}$ cannot be found via the average of φ' since $\langle\varphi'(x)\rangle' = 0$ and the 1PI vertex $\Gamma_{2,0}(P)$ is the inverse correlator $G(P)$ (equation (2.7)):

$$\Gamma_{2,0}(P) \equiv G^{-1}(P) = \tau + 3u\varphi^2 + p^2 + M\omega_l^2 - \Sigma(P) \tag{2.8}$$

as the self-energy part $\Sigma(P)$ is the sum of the 1PI diagrams with two tails and $\tau + 3u\varphi^2 = \partial^2F_0(\tau, \varphi)/\partial\varphi^2$.

The following equalities serve as the basis for our further considerations:

$$F_{m,n}(\tau, \varphi) \equiv \partial^{m+n}F(\tau, \varphi)/\partial\varphi^m\,\partial\tau^n$$
$$= \lim_{\substack{p_i\to 0\\ k_j\to 0}} [\Gamma_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n)|_{\omega_l^i=\omega_l^j=0}] \tag{2.9}$$

$$\partial\Gamma_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n)/\partial\varphi$$
$$= \lim_{p_{m+1}\to 0} [\Gamma_{m+1,n}(P_1, \ldots, P_{m+1}; K_1, \ldots, K_n)|_{\omega_l^{m+1}=0}] \tag{2.10}$$

$$\partial\Gamma_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n)/\partial\tau$$

$$= \lim_{k_{n+1}\to 0} [\Gamma_{m,n+1}(P_1, \ldots, P_m; K_1, \ldots, K_{n+1})|_{\omega_l^{n+1}=0}]. \tag{2.11}$$

They are valid at all values of τ and φ, complying with the condition $\partial^2 F(\tau, \varphi)/\partial\varphi^2 > 0$. The proof of the relations in (2.9) as for $\varphi = 0$ (Itzykson and Zuber 1980) and $\varphi = \varphi_s$ (Patashinsky and Pokrovsky 1982) is based on the possibility of representing $F(\tau, \varphi)$ at the region of τ, φ values defined above as the Legendre transform of the potential $\phi(\tau, h)$ (at $V \to \infty$):

$$\phi(\tau, h) = -(1/\beta V) \ln \left(\mathrm{Tr} \left[\!\left[\exp\left\{ \beta \left[h \int d^3x\, \varphi(x) - \mathcal{H} \right] \right\} \right]\!\right] \right) \tag{2.12}$$

$$F(\tau, \varphi) = \phi(\tau, h) + h\varphi \qquad h = \partial F/\partial\varphi = \partial F_0/\partial\varphi + \langle \partial\mathcal{H}'/\partial\varphi \rangle'. \tag{2.13}$$

The zero Fourier component of $\varphi(x)$ in (2.12) is a C-variable and the trace also contains the integration over this variable. By means of (2.13) the derivatives $F_{m,n}$ may be expressed in terms of $\phi_{m,n} \equiv \partial^{m+n}\phi/(\partial h^m\, \partial\tau^n)$. For example

$$\begin{aligned}
&F_{0,1} = \phi_{0,1} \qquad F_{2,0} = -\phi_{2,0}^{-1} \qquad F_{1,1} = -\phi_{1,1}\phi_{2,0}^{-1} \\
&F_{3,0} = -\phi_{3,0}\phi_{2,0}^{-3} \qquad F_{0,2} = \phi_{0,2} - \phi_{1,1}^2\phi_{2,0}^{-1} \\
&F_{2,1} = \phi_{2,1}\phi_{2,0}^{-2} - \phi_{3,0}\phi_{1,1}\phi_{2,0}^{-3} \\
&F_{1,2} = -\phi_{1,2}\phi_{2,0}^{-1} + 2\phi_{1,1}\phi_{2,1}\phi_{2,0}^{-2} - \phi_{1,1}^2\phi_{3,0}\phi_{2,0}^{-3} \\
&F_{4,0} = \phi_{4,0}\phi_{2,0}^{-4} - 3\phi_{3,0}^2\phi_{2,0}^{-3}.
\end{aligned} \tag{2.14}$$

In turn, the derivatives $\phi_{m,n}$ can be expressed via the connected field correlators $\varphi(x)$ and $\frac{1}{2}\varphi^2(x)$ thermodynamically conjugated with h and τ, respectively:

$$\phi_{m,n} = \frac{(-1)^{n+1}}{2^n \beta V} \left\langle T_\sigma \left(\prod_{i=1}^{m} \int d^4X_i\, \varphi(X_i) \prod_{j=1}^{n} \int d^4Y_j\, \varphi^2(Y_j) \right) \right\rangle_c. \tag{2.15}$$

Here the angular brackets denote averaging with the density matrix from (2.12) where $h = \partial F/\partial\varphi$.

When considering the diagram expansion of $F_{m,n}$ determined by (2.14) and (2.15) it should be taken into account that the Wick theorem is not valid for the field $\varphi(x)$, as its average is non-zero. In fact, from (2.13)–(2.15) it follows that

$$\varphi = -(\partial\phi/\partial h)|_{h=\partial F/\partial\varphi} = \langle \varphi(x) \rangle.$$

Therefore, we may shift the $\varphi(x)$ field in (2.15): $\varphi(x) = \varphi + \tilde{\varphi}(x)$ which gives

$$\phi_{m,n} = \sum_{k=0}^{n} \binom{n}{k} (-\varphi)^k \tilde{\phi}_{m+k,n-k} \tag{2.16}$$

where $\tilde{\phi}_{m,n}$ are the correlators obtained from (2.15) by substitution of $\varphi(x)$ by $\tilde{\varphi}(x)$. The Hamiltonian of the field $\tilde{\varphi}(x)$ becomes

$$\tilde{\mathcal{H}} = \int d^3x \left[\left(\frac{\partial F_0}{\partial\varphi} - \frac{\partial F}{\varphi} \right) \tilde{\varphi} + \tfrac{1}{2}(\tau + 3u\varphi^2)\tilde{\varphi}^2 + \tfrac{1}{2}(\nabla\tilde{\varphi})^2 + u\varphi\tilde{\varphi}^3 + \tfrac{1}{4}u\tilde{\varphi}^4 \right]. \tag{2.17}$$

Substituting (2.16) into (2.14), one can make certain that the results of such a substitution are only the replacement of $\phi_{m,n}$ by $\tilde{\phi}_{m,n}$ in (2.14), except for the expressions for $F_{0,1}$ and $F_{1,1}$ where the extra terms $\frac{1}{2}\varphi^2 = \partial F_0/\partial\tau$ and $\varphi = \partial^2 F_0/(\partial\varphi\partial\tau)$ appear. Thus the

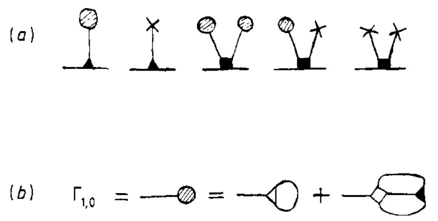

Figure 1.

derivatives $F_{m,n}$ are expressed by the connected correlators of the (2.15) type, and owing to the structure of the right-hand sides of (2.14) their diagram expansion does not contain the diagrams that can be cut across one line into two parts, both containing tails and angles. Yet, this expansion contains one-particle reducible diagrams where a part without external tails and angles or the external field $\partial F/\partial\varphi - \partial F_0/\partial\varphi = \langle\partial\mathcal{H}'/\partial\varphi\rangle'$ may be cut off across one line. Such diagrams in expansion of $F_{2,0}$ are shown in figure 1(*a*), where $\partial F/\partial\varphi - \partial F_0/\partial\varphi$ is indicated by a cross and the sum of a one-tail 1PI diagram—$\Gamma_{1,0}$—by a shaded circle. The block diagram representation of $\Gamma_{1,0}$ in figure 1(*b*) gives

$$\Gamma_{1,0} = u\int \mathrm{d}^3x\,\langle 3\varphi\tilde{\varphi}^2(x) + \tilde{\varphi}^3(x)\rangle_{1\mathrm{PI}} = \left\langle\frac{\partial\mathcal{H}''}{\partial\varphi}\right\rangle''_{1\mathrm{PI}}$$

where $\mathcal{H}''$ is obtained from $\tilde{\mathcal{H}}$ (equation (2.17)) by dropping the term linear in $\tilde{\varphi}$ and the angular brackets with two primes denote the averaging with the density matrix $\exp(-\beta\mathcal{H}'')$. As $\mathcal{H}''$ is of the same form as $\mathcal{H}'$ in (2.3) and only 1PI diagrams are present in the expansion of the external field $\langle\partial\mathcal{H}'/\partial\varphi\rangle'$ (owing to the absence of the zero Fourier component in the field $\varphi'(x)$) then, in the thermodynamic limit $V\to\infty$ when the constraint $\int \mathrm{d}^3x\,\varphi'(x) = 0$ becomes insignificant in calculating diagram contribution, we obtain $\Gamma_{1,0} = \langle\partial\mathcal{H}'/\partial\varphi\rangle'$. Meanwhile, it is quite evident that $\Gamma_{1,0}$ enters into the diagram with a sign opposite to that of the external field that leads to the mutual cancellation of such one-particle reducible diagrams as those in figure 1(*a*).

Thus the diagram expansion of $F_{m,n}$ is obtained by dropping all one-particle reducible diagrams in the expansion of $\tilde{\phi}_{m,n}(-\tilde{\phi}_{2,0})^{-m}$, including the diagrams with the external field. The remaining diagrams differ from the 1PI diagrams appearing in the definition of Γ_{mn} (equations (2.6)–(2.8)) only in the zero external momenta and frequencies, because $\mathcal{H}''$ ($\tilde{\mathcal{H}}''$ in (2.17) without the external field) is of the same form as $\mathcal{H}'$ in (2.3). As in (2.6)–(2.8) the 'non-diagram' contribution is equal to $\partial^{m+n}F_0/(\partial\varphi^m\,\partial\tau^n)$, which results in (2.9). It should be noted that the limit in (2.9) is taken after the limit $V\to\infty$.

The important result of the consideration given above is the possibility of substituting $\varphi'(x)$ correlators in (2.6)–(2.8) by $\varphi(x)$ correlators with the Hamiltonian $\mathcal{H} - (\partial F/\partial\varphi)\int \mathrm{d}^3x\,\varphi(x)$. After such a substitution, $\Gamma_{m,n}$ is easily differentiated with respect to φ. As a result the factor $F_{2,0}\int \mathrm{d}^4x\,\varphi(x)$ appears in the correlator that corresponds to the addition of the tail with zero momentum and frequency. Thereby (2.10) is justified and (2.11) is obtained by the direct differentiation of (2.6) and (2.8).

The right-hand sides of the relation in (2.10), (2.11) may be represented as the expansion in full vertices $\Gamma_{4,0}$, $\Gamma_{3,0}$, $\Gamma_{2,1}$, $\Gamma_{2,0}^{-1}$, by excluding the bare vertices $\Gamma_{4,0}^{(0)} = 6u$, $\Gamma_{3,0}^{(0)} = 6\varphi u$ and the bare angle vertex $\Gamma_{2,1}^{(0)} = 1$ from the perturbation expansions and summing the self-energy parts. The resulting equations for $\Gamma_{m,n}$ determine their dependence upon τ and φ in the region $F_{2,0} > 0$. It is convenient to use the expressions

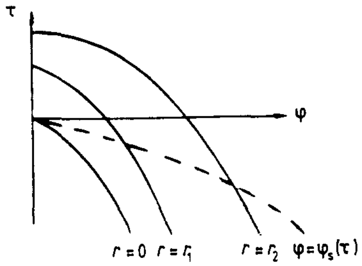

Figure 2.

previously obtained for $\Gamma_{m,n}$ at $\varphi = 0$, $\tau > 0$ (Sakhnenko and Timonin 1983) as the boundary conditions (at $\varphi = 0$, 1PI vertices coincide with the ordinary connected vertices). To find $\Gamma_{m,n}$ at $\tau < 0$ we proceed from the variables τ, φ to the variables r, φ:

$$r = F_{2,0}(\tau, \varphi) = \lim_{p \to 0} [G^{-1}(p, \omega_l = 0)]. \tag{2.18}$$

As $r(\tau, \varphi_s) = \chi^{-1}(\tau)$, the lines $r(\tau, \varphi) =$ constant have the form shown in figure 2. From the equality

$$(\partial \tau / \partial \varphi)_r = -(\partial r / \partial \varphi)_\tau (\partial r / \partial \tau)_\varphi^{-1} = -F_{3,0} F_{2,1}^{-1} \tag{2.19}$$

the derivatives of $\Gamma_{m,n}$ with respect to φ along these lines are obtained from (2.10), (2.11):

$$(\partial / \partial \varphi)_r \Gamma_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n) = \Gamma_{m+1,n}(P_1, \ldots, P_m, 0; K_1, \ldots, K_n)$$
$$- F_{3,0} F_{2,1}^{-1} \Gamma_{m,n+1}(P_1, \ldots, P_m; K_1, \ldots, K_n, 0). \tag{2.20}$$

The dependence of $\Gamma_{m,n}$ upon r, φ at $r > 0$ may be obtained by integration of (2.20) regarding the relations in (2.9) for $F_{3,0}$ and $F_{2,1}$ with the boundary conditions

$$\Gamma_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n; r, \varphi = 0) = \tilde{\Gamma}_{m,n}(P_1, \ldots, P_m; K_1, \ldots, K_n; r) \tag{2.21}$$

where $\tilde{\Gamma}_{m,n}$ are the vertices at $\varphi = 0$, $\tau > 0$, represented as the functions of $r = r(\tau, 0)$. Then $r = r(\tau, \varphi)$ may be determined implicitly by integration of (2.19) using the obtained $F_{3,0}(r, \varphi)$ and $F_{2,1}(r, \varphi)$:

$$\tau = \int_0^r \frac{\mathrm{d}r'}{F_{2,1}(r', 0)} - \int_0^\varphi \mathrm{d}\varphi' \frac{F_{3,0}(r, \varphi')}{F_{2,1}(r, \varphi')}. \tag{2.22}$$

In addition, we assume that on the lines $r(\tau, \varphi) =$ constant up to $\varphi = \varphi_s$ the following estimates are valid:

$$\Gamma_{4,0} \sim \tilde{\Gamma}_{4,0} \qquad G\Gamma_{3,0}^2 \sim \tilde{\Gamma}_{4,0} \qquad \varphi G \Gamma_{3,0} \sim 1 \qquad \Gamma_{2,1} \sim 1. \tag{2.23}$$

Therefore, when expanding the right-hand sides of (2.20), we may consider the vertices $\Gamma_{4,0}$ and $\Gamma_{3,0}$ to be small owing to a small value of $\tilde{\Gamma}_{4,0}$ proportional to invariant charge

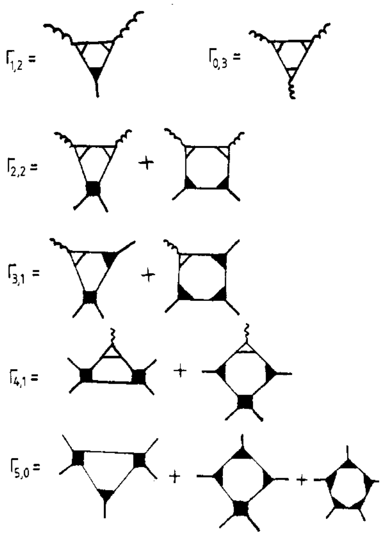

Figure 3.

(Patashinsky and Pokrovsky 1982). Then, having substituted in (2.20) the contributions of the lowest order in $\Gamma_{4,0}$ and $\Gamma_{3,0}$ depicted by the diagrams in figure 3 we obtain

$$(\partial\Gamma_{4,0}/\partial\varphi)_r = \mathrm{O}(\Gamma_{3,0}\tilde\Gamma^2_{4,0}) \qquad (\partial\Gamma_{2,1}/\partial\varphi)_r = \mathrm{O}(\Gamma_{3,0}\Gamma_{4,0})$$

$$(\partial/\partial\varphi)_r\Gamma_{3,0}(P_1, P_2, P_3) = \Gamma_{4,0}(P_1, P_2, P_3, 0) + \mathrm{O}(\tilde\Gamma^2_{4,0}).$$

Using (2.23), this results in

$$\begin{aligned}
&\Gamma_{4,0}(P_1, \ldots, P_4; r, \varphi) = \tilde\Gamma_{4,0}(P_1, \ldots, P_4; r) + \mathrm{O}(\tilde\Gamma^2_{4,0})\\
&\Gamma_{3,0}(P_1, P_2, P_3; r, \varphi) = \varphi\tilde\Gamma_{4,0}(P_1, P_2, P_3, 0; r) + \mathrm{O}(\varphi\tilde\Gamma^2_{4,0})\\
&\Gamma_{2,1}(P_1, P_2; K; r, \varphi) = \tilde\Gamma_{2,1}(P_1, P_2; K; r) + \mathrm{O}(\tilde\Gamma_{4,0}).
\end{aligned} \tag{2.24}$$

The equations for other vertices obtained by substitution in (2.20) of the contribution shown in figure 3 and the expressions from (2.24) are easily integrated and in the lowest order in $\tilde\Gamma_{4,0}$ give

$$\begin{aligned}
&\Gamma_{0,2}(K; r, \varphi) = \tilde\Gamma_{0,2}(K; r) \qquad \Gamma_{1,1}(K; r, \varphi) = \varphi\tilde\Gamma_{2,1}(0, K; -K; r)\\
&G^{-1}(P; r, \varphi) = G^{-1}(P; r, 0) + \tfrac{1}{2}\varphi^2\tilde\Gamma^{-1}_{2,1}(r)[\tilde\Gamma_{4,0}(P, r)\tilde\Gamma_{2,1}(r) - \tilde\Gamma_{4,0}(r)\tilde\Gamma_{2,1}(P, r)]\\
&\tilde\Gamma_{4,0}(P, r) \equiv \tilde\Gamma_{4,0}(P, -P, 0, 0; r) \qquad \tilde\Gamma_{2,1}(P, r) \equiv \tilde\Gamma_{2,1}(P, -P; 0; r)\\
&\tilde\Gamma_{4,0}(r) \equiv \tilde\Gamma_{4,0}(0, r) \qquad \tilde\Gamma_{2,1}(r) \equiv \tilde\Gamma_{2,1}(0, r).
\end{aligned} \tag{2.25}$$

The equations for $G(P; r, 0)$ and the expressions in terms of $G(P; r, 0)$ for vertices

$\tilde{\Gamma}_{m,n}$ present in (2.25) were obtained by Sakhnenko and Timonin (1983) where they had the following designations:

$$\begin{aligned}&\tilde{\Gamma}_{4,0}(P,r)=6\Gamma_1(P,r) \qquad \tilde{\Gamma}_{2,1}(P,r)=T(P,r) \qquad \tilde{\Gamma}_{0,2}(K,r)=-C(K,r)\\&\tilde{\Gamma}_{2,1}(0,K;-K;r)=R(k,r) \qquad \tilde{\Gamma}_{2,1}(r)=T_0(r) \qquad \tilde{\Gamma}_{4,0}(r)=6\Gamma_0(r).\end{aligned} \tag{2.26}$$

The equality determining $r = r(\tau, \varphi)$ follows from (2.9), (2.20) and (2.24):

$$\tau=\tilde{\Gamma}_{2,1}^{-1}(r)[r--\tfrac{1}{2}\varphi^2\tilde{\Gamma}_{4,0}(r)] \tag{2.27}$$

and from the identity $(\partial F_{1,0}/\partial\varphi)_r = r - F_{2,1}^{-1}F_{3,0}F_{1,1}$ with $F_{m,n}(r,\varphi)$ from (2.9), (2.24) and (2.25) the equation of state is obtained:

$$F_{1,0}=r\varphi-\tfrac{1}{3}\varphi^3\tilde{\Gamma}_{4,0}(r)=0. \tag{2.28}$$

When $r > 0$ the solution of the system of (2.27), (2.28) gives the values of the spontaneous order parameter $\varphi_s(\tau)$ and $r(\tau,\varphi_s)=\chi^{-1}(\tau)$. At $\tau < 0$, we have

$$\varphi_s^2=3r\tilde{\Gamma}_{4,0}^{-1}(r) \qquad -2\tau=r\tilde{\Gamma}_{2,1}(r). \tag{2.29}$$

(2.24) and (2.29) justify the assumed estimates in (2.23). According to Sakhnenko and Timonin (1983) at $r_0\exp(-2/\eta) \ll r < r_0 = (9u/8\pi)$, $\eta \simeq 0.02$,

$$\tilde{\Gamma}_{4,0}(r)=(16\pi/3)\sqrt{r} \qquad \tilde{\Gamma}_{2,1}(r)=(r/r_0)^{1-1/2\nu} \qquad \nu\simeq 0.60 \tag{2.30}$$

so that

$$\begin{aligned}&r(\tau,\varphi_s)=\chi^{-1}(\tau)=r_0^{1-\gamma}(-2\tau)^{\gamma} \qquad \gamma=2\nu\\&\varphi_s(\tau)=(3/4\sqrt{\pi})r_0^{(1-\gamma)/4}(-2\tau)^{-\nu/2}.\end{aligned} \tag{2.31}$$

The substitution of (2.31) into (2.25) completes the calculation of the vertices at $\tau < 0$. From now on we shall use the expressions for these vertices as a function of r, having eliminated φ_s by means of (2.29) and keeping in mind that in the final expressions r should be substituted by $r(\tau,\varphi_s)$ from (2.31).

3. Retarded correlators and scattering spectra below T_c

As has been explained in § 1, the dielectric tensor fluctuations complying with the disordered phase symmetry at all temperatures have the form

$$\delta\varepsilon(x,t)=a[\varphi^2(x,t)-\langle\varphi^2(x,t)\rangle].$$

Hence, in both phases the tensor of light scattering spectral intensity is

$$\begin{aligned}I_2(k,\omega)=a^2\int \mathrm{d}^3x\,\mathrm{d}t\exp[\mathrm{i}(kx-\omega t)]\\ \times[\langle\varphi^2(x,t)\varphi^2(0,0)\rangle-\langle\varphi^2(0,0)\rangle^2]\equiv a^2\mathcal{D}_2(k,\omega).\end{aligned}$$

To obtain $I_2(k,\omega)$ in the framework of the quantum formalism adopted in our paper, we should consider Matsubara's Green function

$$G_2(K)=\tfrac{1}{4}\int \mathrm{d}^4X\exp[\mathrm{i}(KX)][\langle\varphi^2(X)\varphi^2(0)\rangle-\langle\varphi^2(0)\rangle^2]. \tag{3.1}$$

The analytical continuation of $G_2(K)$ from discrete imaginary frequencies $\mathrm{i}\omega_l$, $\omega_l > 0$,

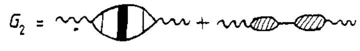

Figure 4.

gives the retarded Green function $G_2^{\mathrm{R}}(k, \omega)$, at $\omega \ll T_{\mathrm{c}} \hbar^{-1}$ related to $\mathscr{D}_2(k, \omega)$ by the classical fluctuation-dissipative theorem

$$\mathscr{D}_2(k, \omega) = \omega^{-1} \operatorname{Im}[G_2^{\mathrm{R}}(k, \omega)].$$

To obtain the diagram expansion of $G_2(K)$ below T_{c} we have to express it through φ'-correlators by extracting the average over the condensate:

$$G_2(K) = \langle G_2(K, \varphi) \rangle_{\varphi} = G_2(K, \varphi_{\mathrm{s}})$$

$$\begin{aligned} G_2(K, \varphi) = \tfrac{1}{4} \int \mathrm{d}^4 X \exp[\mathrm{i}(KX)]\{&\langle \varphi'^2(X) \varphi'^2(0) \rangle' - \langle \varphi'^2(0) \rangle'^2 \\ &+ 2\varphi[\langle \varphi'^2(X) \varphi'(0) \rangle' + \langle \varphi'(X) \varphi'^2(0) \rangle'] + 4\varphi^2 \langle \varphi'(X) \varphi'(0) \rangle' \}. \end{aligned}$$

Examining the diagram expansion of $G_2(K, \varphi)$, we can represent it via the 1PI vertices considered in § 2. In so doing we notice that reducible diagrams with angles have 1PI parts attached to the angles and are connected with the rest of the diagram by a tail, thus concurring with the contribution to the $\Gamma_{1,1}$ vertex. Taking into account that $\varphi = \partial F_0 / \partial \varphi \, \partial \tau$ is the non-diagram contribution to $\Gamma_{1,1}$ (cf (2.6)) and that the sum of the diagrams with two tails represents the one-particle Green function $G(K, \varphi)$, we finally obtain the $G_2(K, \varphi)$ representation depicted in figure 4.
The corresponding analytical expression reads

$$G_2(K, \varphi) = -\Gamma_{0,2}(K; r, \varphi) + \Gamma_{1,1}^2(K; r, \varphi) G(K; r, \varphi).$$

Hence, using the designation of (2.26),

$$G_2(K) = G_2(K, \varphi_{\mathrm{s}}) = C(K, r) + \tfrac{1}{2} r \Gamma_0^{-1}(r) R^2(K, r) G(K; r, \varphi_{\mathrm{s}}). \tag{3.2}$$

The analytical continuation of (2.35), (3.2) gives $G^{\mathrm{R}}(k, \omega)$ and $G_2^{\mathrm{R}}(k, \omega)$ at $T < T_{\mathrm{c}}$. Having applied the expressions obtained by Sakhnenko and Timonin (1983) for the vertices (equation (2.26)) and $G^{\mathrm{R}}(k, \omega, r, \varphi = 0)$ at

$$r_0 \exp(-2/\eta) \ll \max(r, k^2, M\omega^2, 2L\omega) < r_0 \tag{3.3}$$

we get

$$\begin{aligned} G_{\mathrm{R}}^{-1}(k, \omega) = {} & r + k^2 - 2\mathrm{i} L\omega - M\omega^2 + \tfrac{3}{2} r \{[\Gamma_0^{-1}(r) \Gamma_{\mathrm{R}}(k, \omega, r)]^{1/\nu - 1} - 1\} \\ & + \left(1 - \frac{1}{2\nu}\right) \int_r^{r_0} \mathrm{d}r' \, [1 + \tfrac{1}{2}(r/r')^{1/2\nu}] \{[\Gamma_0^{-1}(r') \Gamma_{\mathrm{R}}(k, \omega, r')]^{1/\nu - 1} - 1\} \end{aligned} \tag{3.4}$$

$$\Gamma_{\mathrm{R}}^{-1}(k, \omega, r) = g[\Pi(k, \omega, r) - \Pi(k, \omega, r_0)] + u^{-1}$$

$$\begin{aligned} \Pi(k, \omega, r) = (1/4\pi \mathrm{i} k) \ln\{&[(S + 2\sqrt{r})(P + 2L - \mathrm{i} M\omega) \\ &+ \mathrm{i} k (P + 2L)] / [(S + 2\sqrt{r})(P + 2L - \mathrm{i} M\omega) - \mathrm{i} k (P + 2L)]\} \end{aligned}$$

$$S = (4r + k^2 - 4\mathrm{i} L\omega - M\omega^2)^{1/2} \qquad P = [Mk^2 + (2L - \mathrm{i} M\omega)^2]^{1/2}$$

$$\begin{aligned} G_2^{\mathrm{R}}(k, \omega) = {} & (\nu u^{\alpha/\nu - 1} / 18\alpha) \{\Gamma_{\mathrm{R}}^{-\alpha/\nu}(k, \omega, r) \\ & + [g\alpha r / \nu \Gamma_0(r)] \Gamma_{\mathrm{R}}^{1 - \alpha/\nu}(k, \omega, r) G^{\mathrm{R}}(k, \omega)\} \end{aligned} \tag{3.5}$$

where $\alpha = 2 - 3\nu$ is the heat capacity exponent, and the power functions and logarithm denote the principal branches of these functions that are analytical on the plane with the cut along the negative semi-axis. We recall that, in (3.3)–(3.5), $r = r(\tau, \varphi_s)$ is given by (2.31).

According to (3.5) the spectral density of Raman scattering,

$$I_2(k, \omega) = a^2\omega^{-1}\,\mathrm{Im}[G_2^R(k, \omega)] \tag{3.6}$$

together with the contribution from two-particle fluctuations (the same as at $T > T_c$) contains a contribution from the one-particle Green function. However, unlike the mean-field region ($r > r_0$) this contribution from the one-particle Green function is distorted in the scaling region owing to the presence of the factor $\Gamma_R^{1-\alpha/\nu}(k, \omega, r)$ at $G^R(k, \omega)$. The intensity of such modified one-particle scattering is comparable in magnitude with the two-particle scattering. Indeed, the integral scattering intensity

$$I_{2\,\mathrm{int}}(k) \sim G_2^R(k, 0) \sim [(1/k)\tan^{-1}(k/2\sqrt{r})]^{\alpha/\nu} \times \{1 + (9\alpha/2\nu)[k\sqrt{r}G^R(k, 0)/\tan^{-1}(k/2\sqrt{r})]\}. \tag{3.7}$$

At $r \gg k^2$, both terms in (3.7) are of the same order, $I_{2\mathrm{int}} \sim |\tau|^{-\alpha}(1 + 9\alpha/\nu)$ while, at $r \ll k^2$, the contribution from the term with $G^R(k, 0)$ becomes small. Thus the spectrum in the scaling region is the result of the interference of scattering by the one- and two-particle excitations. Also, one-particle excitations themselves are no longer quasi-harmonic phonons owing to the presence of the self-energy contributions in $G^R(k, \omega)$ (equation (3.4)). In this case the term with the integral over r' in (3.4) analogous to that present in $G^R(k, \omega)$ at $T > T_c$ (Sakhnenko and Timonin 1983) may be considered to be the contribution from the processes of multiple scattering of soft phonons by the phonon density fluctuations $\delta\varphi'^2 = \varphi'^2 - \langle\varphi'^2\rangle$, whereas the term $\frac{3}{2}r[(\Gamma_R/\Gamma_0)^{1/\nu-1} - 1]$ appearing below T_c is associated with the three-phonon processes allowed in the ordered phase.

To elucidate the character of the one-particle excitations near T_c, we would consider $G^R(k, \omega)$ at small r:

$$r \ll \max(k^2, M\omega^2, 2L\omega) \ll r_0. \tag{3.8}$$

In this region, $G^R(k, \omega)$ does not depend upon r any longer:

$$v^2 G_R^{-1}(k, \omega) \simeq v^2k^2 - 2\,\mathrm{i}\,\Gamma\omega - \omega^2 + (1 - 1/2\nu)\Omega_0^2 \times [\![\{1 - \mathrm{i}\omega/[\sqrt{v^2k^2 + (2\Gamma - \mathrm{i}\omega)^2} + 2\Gamma]\}^{1/\nu-1} - 1]\!] \tag{3.9}$$

$$v^2 = M^{-1} \qquad \Gamma = LM^{-1} \qquad \Omega_0^2 = r_0M^{-1}.$$

The last term in (3.9) is the contribution from the integral over r' in (3.4). Its presence changes considerably the structure of the singularities of $G^R(k, \omega)$ in the lower semi-plane of the complex ω, $\mathrm{Im}\,\omega < 0$, where a cut between the branching points $\pm vk - \mathrm{i}2\Gamma$ corresponding to the thresholds of two-phonon difference scattering appears and the position of the poles changes. The latter effect is especially noticeable at small k and Γ:

$$4\Gamma^2 + v^2k^2 \ll (1 - 1/2\nu)\Omega_0^2. \tag{3.10}$$

In this case $G^R(k, \omega)$ has three poles:

$$\omega_0 \simeq -\mathrm{i}(v^2k^2/6\eta\Omega_0^2)[(4\Gamma^2 + v^2k^2)^{1/2} + 2\Gamma] \qquad \eta = \tfrac{1}{6}(1 - 1/2\nu)(1/\nu - 1)$$

$$\omega_\pm \simeq \pm\,\Omega_0(1 - 1/2\nu)^{1/2}(2^{1/\nu-1} - 1)^{1/2} - \mathrm{i}\,\Gamma(\nu^{-1} - 2^{1-1/\nu})/(1 - 2^{1-1/\nu}) \simeq \pm 0.3\Omega_0 - 2.8\,\mathrm{i}\Gamma$$

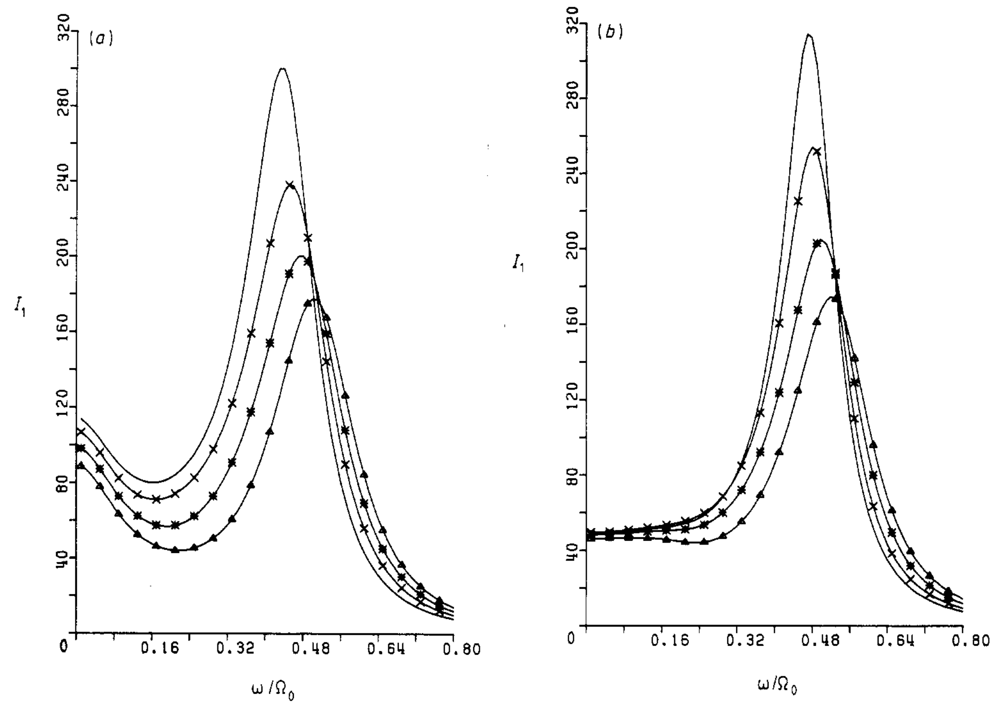

Figure 5. Neutron scattering cross sections at $T < T_c$ for (a) $\Gamma = 5 \times 10^{-2}\,\Omega_0$, $vk = 10^{-2}\,\Omega_0$ ($\triangle$, $r = 4 \times 10^{-2}\,r_0$; $*$, $r = 2.25 \times 10^{-2}\,r_0$; $\times$, $r = 10^{-2}\,r_0$; —, $r = 2.5 \times 10^{-3}\,r_0$) and ($b$) $\Gamma = 5 \times 10^{-2}\,\Omega_0$, $vk = 0.2\,\Omega_0$ ($\triangle$, $r = 2.25 \times 10^{-2}\,r_0$; $*$, $r = 10^{-2}\,r_0$; $\times$, $r = 2.5 \times 10^{-3}\,r_0$; —, $r = 10^{-4}\,r_0$).

so that at low frequencies $|\omega| \lesssim |\omega_0|$ the one-particle fluctuations are relaxing excitations, while at $|\omega| \sim |\omega_\pm|$ they are propagating excitations. The poles $\omega_\pm$ correspond to the 'saturated' soft phonons with approximately three times the frequency and three times the damping of the corresponding values on the boundary of the mean-field region where $\omega_\pm \simeq \pm\,\Omega_0 - \mathrm{i}\Gamma$. The frequency dependences of the neutron scattering cross sections $I_1(k,\omega) \sim \omega^{-1}\,\mathrm{Im}[G^{\mathrm{R}}(k,\omega)]$ calculated by means of (3.4) are depicted in figure 5(a), demonstrating the process of the soft-mode 'saturation' accompanied by the formation of the relaxing excitations as $T \to T_c$.

If the inequality in (3.10) is not valid, the phonon density fluctuations influences the displacement of the two quasi-harmonic poles of $G^{\mathrm{R}}(k,\omega)$. The scattering cross sections for this case are shown in figure 5(b).

At $T < T_c$ the behaviour of the spectral density of light scattering determined by the imaginary part of the correlator $G_2^{\mathrm{R}}(k,\omega)$ (equations (3.5) and (3.6)) is shown in figure 6. The changes in the spectrum are associated with the crossover from one-particle scattering by soft phonons to the two-particle scattering by the phonon density fluctuations at $T = T_c$. In this case the appearance of the central peak at $v^2k^2 \ll 4\Gamma^2$ (figure 6(a)) is associated with the relaxational character of the two-particle excitations while the peak at the frequency $\omega \simeq vk$ when $v^2k^2 \gg 4\Gamma^2$ (figure 6(b)) corresponds to the

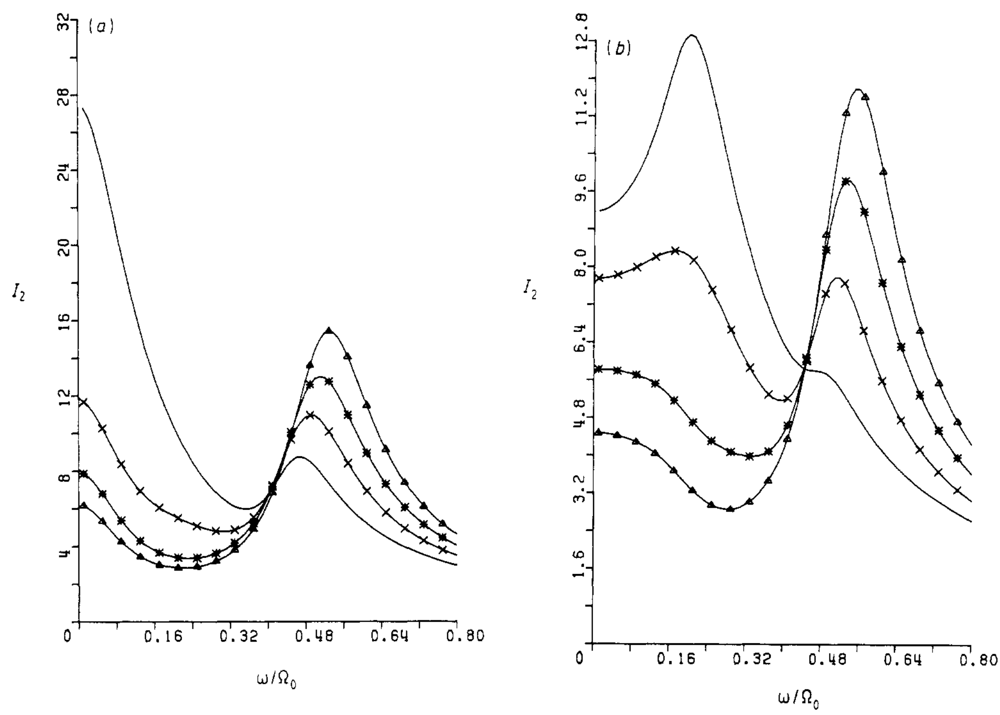

Figure 6. Light scattering spectrum at $T < T_c$ for (*a*) $\Gamma = 5 \times 10^{-2}\,\Omega_0$, $vk = 10^{-2}\,\Omega_0$ (Δ, $r = 4 \times 10^{-2} r_0$; $*$, $r = 2.25 \times 10^{-2} r_0$; $\times$, $r = 10^{-2} r_0$; —, $r = 2.5 \times 10^{-3} r_0$) and (*b*) $\Gamma = 5 \times 10^{-2}\,\Omega_0$, $vk = 0.2\,\Omega_0$ (Δ, $r = 2.25 \times 10^{-2} r_0$; $*$, $r = 10^{-2} r_0$; $\times$, $r = 2.5 \times 10^{-3} r_0$; —, $r = 10^{-4} r_0$).

scattering by the propagating phonon density fluctuations (Sakhnenko and Timonin 1983).

Thus, in contrast to the mean-field region $r > r_0$ where $G_2^{\rm R}(k, \omega) \simeq \varphi_s^2 G^{\rm R}(k, \omega)$ and the spectra of the light and neutron scattering coincide, they become different in the scaling region. Such differences caused by rapid smearing of the soft mode in the optical spectra and indicating the presence of the scaling region in the vicinity of T_c are found in crystals of lead germanate (Satija and Cowley 1982) and strontium titanate (Bruce and Stirling 1983). Although transitions in these crystals do not enter into the class under consideration (lead germanate is a ferroelectric and strontium titanate has a three-component order parameter), the qualitative similarity of the observed singularities is quite obvious.

The quantitative description of the spectra shapes by means of (3.4) and (3.5) is possible for spectral phase transitions with a one-component order parameter in the crystals of quartz (Bruce and Cowley 1980), $AlPO_4$ (Scott 1978) as well as in uniaxial ferroelectrics with the abnormally small dipole interaction of soft phonons, e.g. TSCC (Sugo *et al* 1984) and $Li_2Ge_7O_{15}$ (Wada and Ishibashi 1983). Nevertheless, the experimental light scattering data (Bruce and Cowley 1980, Scott 1978; Sugo *et al* 1984, Wada and Ishibashi 1983) do not allow us to make definite conclusions on the validity of the expression obtained for $G_2^{\rm R}(k, \omega)$ (equation (3.5)). It is possible only to notice the qualitative agreement with the behaviour shown in figure 6(*a*). A more detailed study of the light and neutron scattering spectra in these crystals near T_c and comparison with the results obtained in the present work will make it possible to determine the boundaries of the scaling region and the features of critical dynamics in this region.

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