Thermal fluctuations and the lineshape of a magnetic resonance in the incommensurate phase of a crystal

M. A. Popov

Krasnoyarsk State University, 660041 Krasnoyarsk, Russia
(Submitted 20 October 1995; resubmitted 5 January 1996)
Zh. Éksp. Teor. Fiz. **110**, 218–226 (July 1996)

The adiabatic approximation is used to study the effect of the linear and quadratic contributions of the thermal lattice fluctuations to the magnetic resonance frequency on the resonance lineshape in the incommensurate phase of a crystal. An analysis of the temperature behavior of the peaks in the quasicontinuous resonance-frequency distribution characteristic of this phase is carried out. The decay of fluctuations in the lattice-wave amplitude that accompanies a drop in temperature usually makes these peaks taller, although the increase in the height of the peaks with temperature-independent frequencies is limited by the quadratic contribution of the fluctuations in the phase of the lattice wave. But for atoms occupying particular positions in the crystal cell in the high-temperature phase the linear contribution of fluctuations increases as the temperature is lowered, which makes the peaks with temperature-dependent frequencies shorter. The height of these peaks increases as the order parameter becomes saturated or as the region of a peak with a temperature-independent frequency is approached. [S1063-7761(96)01707-6]

1. INTRODUCTION

Crystals with an incommensurate phase are currently an object of widespread study.[1] In such a phase, the displacements of the atoms from the positions in a high-temperature translationally symmetric phase form a spatial wave with a wave vector not coinciding with any of the highly symmetric (singular) points of the Brillouin zone.

In the translationally symmetric phase, the magnetic and electric fields in atoms occupying the same positions in the unit cells of the crystals have the same time averages. Hence the magnetic resonance spectrum in such a phase is discrete (the number of lines is determined by the number of nonequivalent positions in the cell occupied by the resonating atoms and the magnitude of the atomic spin), which makes it possible to determine the local symmetry from the angular dependence of the resonance frequency and therefore to localize the atom in a cell.[2] The shape of an individual peak in the spectrum can be used to determine the type of atomic mobility.[3]

In an incommensurate phase the translational degeneracy of resonance frequencies is lifted, and each individual peak becomes a quasicontinuous distribution of resonance frequencies in an interval that depends on the amplitude of the lattice modulation wave.[4] The distribution has singularities at least at the end of the interval. For a long time the experimentally observed resonance lines were interpreted by a static model: the lattice displacement wave $\eta(\mathbf{r},t)$ was assumed "frozen," i.e., $\eta(\mathbf{r},t)\equiv\eta(\mathbf{r})$. Within this model, the magnetic resonance line below the phase transition temperature T_i is the convolution of this resonance frequency distribution and the line shape in the high-temperature phase; just below T_i the line consists of two peaks that bound the continuously distributed resonance frequencies (i.e., the temperature range below T_i, in which the fluctuations of the magnetic and electric fields exceed the effect of the variations of local static electric fields on the resonance frequencies and are the only source of a resonance line above T_i, is fairly narrow). This line splitting in the transition to the incommensurate phase was observed in many NMR studies involving quadrupole nuclei, in NQR studies, and in EPR studies (see the review by Cummins[1]). In interpreting the experimental data, however, emphasis was placed both on the form of $\eta(\mathbf{r})$ (the wave or soliton approximation) and on the type of dependence of the resonance frequency Ω on η (the number of terms taken into account in the expansion of Ω in powers of η, and the local or nonlocal approximation).[5,6]

For a long time the effect of the lattice mobility of spins on the resonance lineshape in the incommensurate phase of a crystal was ignored. The first papers to consider this effect dealt with slippage of the lattice modulation wave along the crystal[7] and thermal fluctuations of the phase of the wave with a given Gaussian distribution.[8] Only fairly recently have rigorous studies of the effect of thermal fluctuations on the resonance line emerged.[9–11] Fajdiga *et al.*[9] and Dolinšek *et al.*[11] examined the effect of the linear contribution of fluctuations, $\delta\Omega=(\partial\Omega/\partial\eta)\delta\eta$, for, respectively, a linear relationship between Ω and η, or $\Omega=\Omega_0+\Omega_1\eta$, and a quadratic relationship, $\Omega=\Omega_0+\Omega_2\eta^2/2$. The result was an explanation of the lack of splitting of the resonance line over a certain temperature range below T_i, a phenomenon observed by NMR on ^{87}Rb in Rb_2ZnCl_4 and on ^{39}K in K_2SeO_4.

Studying the NQR spectrum of ^{127}I in Cs_2ZnI_4, Aleksandrova *et al.*[12] found that as the temperature is lowered below T_i the resonance line becomes asymmetric instead of splitting in the ordinary way, and only after that a second peak emerges above the noise level. In Ref. 10 a generalization to the case of an arbitrary relationship between Ω and η suggested a way of explaining the unusual behavior of the resonance line by the effect of the linear contribution of thermal lattice fluctuations to the resonance frequency.

The present paper develops the approach suggested in Ref. 10 and generalizes it to the case of a quadratic contribution of thermal lattice fluctuations to the resonance frequency, $\delta\Omega=(\partial\Omega/\partial\eta)\delta\eta+(\partial^2\Omega/\partial\eta^2)(\delta\eta)^2/2$.

2. THE LINEAR CONTRIBUTION OF FLUCTUATIONS

In the adiabatic approximation[2,3] and with the inhomogeneity of the crystal in the incommensurate phase taken into account, the resonance line can be represented by the function

$$g(\omega)=(2\pi V)^{-1}\int d\mathbf{r}\int_{-\infty}^{\infty}dt \times\left\langle \exp\left\{ i\left[\omega t-\int_0^t dt'\Omega(\mathbf{r},t')\right]\right\}\right\rangle, \tag{1}$$

V is the volume occupied by the sample, and the angle brackets stand for averaging over the random process tested by the resonance frequency.

Except for a narrow neighborhood of the transition temperature,[13] the behavior of the lattice subsystem is successfully described by Landau's thermodynamic theory, which makes it possible to consider atomic motion as being a Gaussian process. Without loss of generality, we restrict our problem to the case of a crystal with an incomplete thermodynamic potential depending on a two-component order parameter $\eta_\mathbf{r}=(\eta_{1\mathbf{r}},\eta_{2\mathbf{r}})$ (see Ref. 14):

$$\Phi[\eta_\mathbf{r}]=\Phi_0+\int d\mathbf{r}\left\{\frac{A}{2}(\eta_{1\mathbf{r}}^2+\eta_{2\mathbf{r}}^2)+\frac{B}{4}(\eta_{1\mathbf{r}}^2+\eta_{2\mathbf{r}}^2)^2 +\frac{D}{2}((\nabla\eta_{1\mathbf{r}})^2+(\nabla\eta_{2\mathbf{r}})^2) +D'\left(\eta_{1\mathbf{r}}\frac{\partial\eta_{2\mathbf{r}}}{\partial z}-\eta_{2\mathbf{r}}\frac{\partial\eta_{1\mathbf{r}}}{\partial z}\right)\right\}, \tag{2}$$

where B and D are strictly positive, and ∇ is the nabla operator. By applying the transformation

$$\xi_{1\mathbf{r}}=\eta_{1\mathbf{r}}\cos(qz)-\eta_{2\mathbf{r}}\sin(qz),$$

$$\xi_{2\mathbf{r}}=\eta_{1\mathbf{r}}\sin(qz)+\eta_{2\mathbf{r}}\cos(qz)$$

with $q=D'/D$ to (2), we can cancel out the Lifshitz invariant. Equation (2) becomes

$$\Phi[\xi_\mathbf{r}]=\Phi_0+\int d\mathbf{r}\left\{\frac{A'}{2}(\xi_{1\mathbf{r}}^2+\xi_{2\mathbf{r}}^2)+\frac{B}{4}(\xi_{1\mathbf{r}}^2+\xi_{2\mathbf{r}}^2)^2 +\frac{D}{2}((\nabla\xi_{1\mathbf{r}})^2+(\nabla\xi_{2\mathbf{r}})^2)\right\},$$

with $A'=A-Dq^2$, and the following correlation functions correspond to the solution $\langle\xi_{1\mathbf{r}}\rangle=\mathrm{Re}\sqrt{-A'/B}\equiv\xi_0$ (here $\langle\xi_{2\mathbf{r}}\rangle=0$ follows from the conditions of equilibrium $\delta\Phi[\xi_\mathbf{r}]/\delta\xi_\mathbf{r}=0$ below the temperature $T_i=T_0+Dq^2/\alpha$, with $A=\alpha(T-T_0)$ and $\alpha>0$, of the transition from the disordered phase to the incommensurate phase with relaxation-type crystal dynamics):

$$\langle\xi_1'\xi_1'\rangle_{\mathbf{k}\omega}=\frac{T\Gamma}{(A_0+Dk^2)^2+\Gamma^2\omega^2},$$

$$\langle\xi_2'\xi_2'\rangle_{\mathbf{k}\omega}=\frac{T\Gamma}{D^2k^4+\Gamma^2\omega^2},\quad \langle\xi_1'\xi_2'\rangle_{\mathbf{k}\omega}=0,$$

where

$$\xi'=\xi-\langle\xi\rangle,$$

$$\langle\xi_1'\xi_2'\rangle_{\mathbf{k}\omega}=\int d\mathbf{r}\int_{-\infty}^{\infty}dt\langle\xi_{1\mathbf{r}}'(t)\xi_{20}'(0)\rangle\exp[i(\omega t-\mathbf{kr})],$$

Γ is the coefficient in the dissipation function

$$\Psi[\eta_\mathbf{r}]=\int d\mathbf{r}\Gamma(\dot\eta_{1\mathbf{r}}^2+\dot\eta_{2\mathbf{r}}^2)/2,$$

$\dot\eta=d\eta/dt$, $A_0=A'+3B\xi_0^2$, and the Boltzmann constant is set to unity.

Suppose that an external magnetic field (or the gradient of an electric field in NQR) identifies $\eta_{1\mathbf{r}}(t)$. The effect of the linear contribution of the lattice fluctuations is reduced to

$$\Omega(\mathbf{r},t)=\Omega_{0\mathbf{r}}+\Omega_{1\mathbf{r}}\eta_{1\mathbf{r}}'(t),$$

where $\Omega_{0\mathbf{r}}=\Omega(\xi_0\cos(qz))$, and $\Omega_{1\mathbf{r}}=\Omega'(\xi_0\cos(qz))$, with $\Omega'(\eta)=d\Omega(\eta)/d\eta$. Since, as noted earlier, the motion of the lattice atoms is described by a Gaussian process, we can write the function (1) representing the resonance line as[2,3]

$$g(\omega)=\frac{1}{2\pi L}\int_0^L dz\int_{-\infty}^{\infty}dt \times\exp\left\{i(\omega-\Omega_{0\mathbf{r}})t-\Omega_{1\mathbf{r}}^2\int_0^t dt'(t-t')\langle\eta_{1\mathbf{r}}'(t')\eta_{1\mathbf{r}}'\rangle\right\}$$
$$=\frac{1}{2\pi L}\int_0^L dz\int_{-\infty}^{\infty}dt \times\exp\left\{i(\omega-\Omega_{0\mathbf{r}})t-\Omega_{1\mathbf{r}}^2\int_{-\infty}^{\infty}d\varepsilon\langle\eta_\mathbf{r}^2\rangle_\varepsilon\frac{1-\cos(\varepsilon t)}{2\pi\varepsilon^2}\right\}, \tag{3}$$

where

$$\langle\eta_\mathbf{r}^2\rangle_\varepsilon=\int_{-\infty}^{\infty}dt\langle\eta_{1\mathbf{r}}'(t)\eta_{1\mathbf{r}}'\rangle\cos(\varepsilon t)$$
$$=\frac{v}{8\pi^3}\int d\mathbf{k}[\langle\xi_1'\xi_1'\rangle_{\mathbf{k}\varepsilon}\cos^2(qz) +\langle\xi_2'\xi_2'\rangle_{\mathbf{k}\varepsilon}\sin^2(qz)], \tag{4}$$

$L=2\pi/q$, integration over the wave vectors $\mathbf{k}$ is done inside the first Brillouin zone, and v is the unit cell volume in the high-temperature phase.

In view of the unwieldy nature of the above expression we restrict our discussion to an estimate of the integral. The principal contribution to the integral is provided by low-

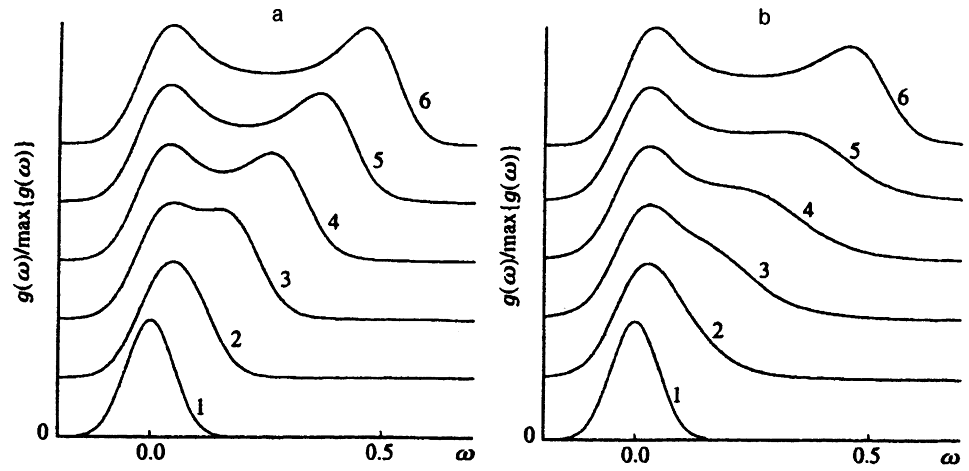

FIG. 1. Intensity-normalized lineshape of a magnetic resonance in the incommensurate phase of a crystal on atoms occupying particular positions, caused by the linear contribution of fluctuations with a saturated order parameter. Notation: (a) $\Gamma=1$, and (b) $\Gamma=10$; curve *1*, $\xi_0^2=0$; curve *2*, $\xi_0^2=0.2$; curve *3*, $\xi_0^2=0.4$; curve *4*, $\xi_0^2=0.6$; curve *5*, $\xi_0^2=0.8$; and curve *6*, $\xi_0^2=1$.

frequency phonons, and the edge of the Brillouin zone in (4) can be ignored, with the result that integration is extended to infinity. This yields

$$\langle \eta_{\mathrm{r}}^2\rangle_\varepsilon \approx \frac{vT\Gamma}{2\pi(2D)^{3/2}} \times \left\{ \frac{\cos^2(qz)}{[(A_0^2+\Gamma^2\varepsilon^2)^{1/2}+A_0]^{1/2}} + \frac{\sin^2(qz)}{|\Gamma\varepsilon|^{1/2}} \right\}. \quad (5)$$

For $T<T_i$ the gap in the spectrum of $\xi'_{1\mathrm{r}}$ is finite. If the relaxation contribution is ignored in comparison to the gap, the estimate of the integral can be simplified:

$$g(\omega)\approx \frac{1}{\pi L}\int_0^L dz \int_0^\infty dt\ \cos[(\omega-\Omega_{0\mathrm{r}})t] \times \exp[-\gamma_{1\mathrm{r}}t-(\gamma_{2\mathrm{r}}t)^{3/2}], \quad (6)$$

where the relaxation rate

$$\gamma_{1\mathrm{r}}=\frac{v\Gamma T r_c \Omega_{1\mathrm{r}}^2}{16\pi D^2}\cos^2(qz)$$

is determined by the thermal fluctuations $\xi'_{1\mathrm{r}}(t)$ of the lattice wave amplitude ("amplitudons"),

$$\gamma_{2\mathrm{r}}=\frac{(2\Gamma)^{1/3}}{\pi D}\left[\frac{vT\Omega_{1\mathrm{r}}^2}{6}\sin^2(qz)\right]^{2/3}$$

is determined by the phase fluctuations $\xi'_{2\mathrm{r}}(t)$ ("phasons"), and $r_c=(D/A_0)^{1/2}$ is the correlation radius.

If we ignore the lattice fluctuations, $\gamma_{1\mathrm{r}}\approx\gamma_{2\mathrm{r}}\approx 0$, Eq. (6) becomes the relationship well known from the static model[4–6]:

$$g(\omega)=L^{-1}\sum |\partial\Omega(\xi_0\cos(qz))/\partial z|^{-1},$$

where the sum is over all solutions of the equation $\omega=\Omega(\xi_0\cos(qz))$. The quasicontinuous distribution of resonance frequencies exhibits peaks corresponding to the extrema of this function:

$$\frac{\partial\Omega(\xi_0\cos(qz))}{\partial z}=-q\Omega'(\xi_0\cos(qz))\sin(qz)=0.$$

The frequencies of these peaks corresponding to $\sin(qz)=0$ vary with temperature according to $\Omega(\pm\xi_0)$, while those corresponding to $\Omega'(\xi_0\cos(qz))=0$ do not vary.

Phasons have essentially no effect on any of the above peaks, since at these frequencies $\gamma_{2\mathrm{r}}=0$. Amplitudons, however, act selectively: $\gamma_{1\mathrm{r}}=0$ for peaks with temperature-independent frequencies, while $\gamma_{1\mathrm{r}}\neq 0$ for peaks with temperature-dependent frequencies. As a result the latter may be weakened or may even be completely suppressed.

While for $T>T_i$ the resonant atoms occupy general positions in the crystal cell and $\Omega'(0)$ dominates in the expansion of $\Omega'(\xi_0)$ in powers of ξ_0, as the temperature is lowered below T_i the relaxation rate $\gamma_{1\mathrm{r}}$ decreases in proportion to the correlation radius r_c and peaks with temperature-dependent frequencies may be restored as we move away from the neighborhood of the transition. In the event of a particular position, $|\Omega'(\xi_0)|$ grows with decreasing temperature no slower than $\xi_0\sim r_c^{-1}$ which leads to a decrease in $\gamma_{1\mathrm{r}}$ and marked suppression of the peak with the frequency $\Omega(\xi_0)$.

There are two reasons why this peak grows as the temperature falls. First, in the region where ξ_0 is saturated the coefficient $\Omega'(\xi_0)$ becomes a constant and the temperature behavior of $\gamma_{1\mathrm{r}}$ is again dominated by the decrease in r_c. This situation is numerically modeled in Fig. 1 by the convolution of (6) and the Gaussian $\exp\{-\omega^2/2\gamma_g^2\}/(2\pi\gamma_g^2)^{1/2}$. At $\Omega(\eta)=\eta^2/2$ the temperature T was determined from the relationship $\xi_0=\tanh(\xi_0/T)$ known from the molecular approximation, and the coefficients were set at $B=D=v=1$ and $\gamma_g=1/20$. Second, the increase in $|\Omega'(\xi_0)|$ was replaced by a decrease as we move closer to the region of a peak with a temperature-independent frequency. This situation is modeled in Fig. 2 at $\Omega(\eta)=\eta^2/2-\eta^4/4$. The relaxation rates were calculated by the formulas

$$\gamma_{1\mathrm{r}}=v\Gamma r_c\Omega_{1\mathrm{r}}^2\cos^2(qz)/16\pi D^2,$$

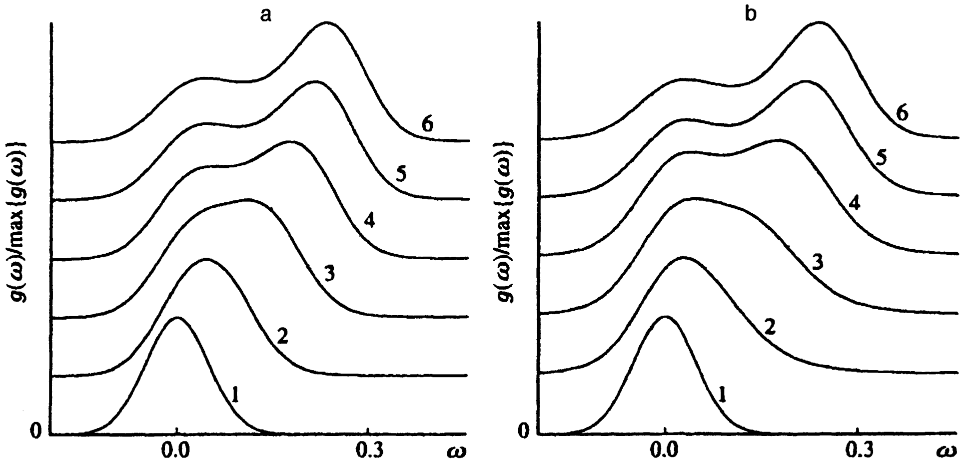

FIG. 2. Intensity-normalized lineshape of a magnetic resonance in the incommensurate phase of a crystal on atoms occupying particular positions, caused by the linear contribution of fluctuations with a temperature intersection of the peaks in the quasicontinuous distribution of the resonance frequencies. Notation is the same as in Fig. 1.

$$\gamma_{2\mathrm{r}}=(2\Gamma)^{1/3}[v\Omega_{1\mathrm{r}}^2\sin^2(qz)/6]^{2/3}/\pi D,$$

and the coefficients were set at the same values.

As ξ_0 increases, the initial decrease in the height of the peak at the frequency $\Omega(\xi_0)$ changes in both cases to an increase in the peak height as we move closer to the region where the above conditions are met. But while for small values of Γ this behavior is observed over the entire temperature range, for large values of Γ the decrease in peak height is accompanied by a change in peak shape (the peak becomes asymmetric and moves to the frequency $\Omega(0)$), while the increase leads to the emergence of a new peak with a slowly varying frequency, growing from under the wing of the asymmetric peak.

Such behavior of the resonance line was observed, as noted in the Introduction, in Cs_2ZnI_4 below T_i. The following estimates can be made on the basis of the data of Ref. 12 for this crystal: $v\approx 1.3\times 10^{-27}$ m^3, the average frequency of "soft" optical phonons $\omega_{\mathrm{ph}}=k_BT_i/\hbar\approx 1.6\times 10^{13}$ s^{-1}, $D\approx m\omega_{\mathrm{ph}}^2/Q^2\approx 4.1\times 10^{-18}$ J (here m is the mass of an iodine atom, and Q is the Debye wave vector), and $\Gamma=m\gamma_{\mathrm{ph}}=m\omega_{\mathrm{ph}}\kappa\approx 3.3\times 10^{-12}$ κ kg s^{-1} (here γ_{ph} is the phonon relaxation rate, and $\kappa\approx 10^{-2}$–10^0; see Ref. 11). If with the observed splitting of resonance frequencies $\Delta\Omega_0\approx 5.6\times 10^6$ s^{-1} we associate $b\xi_0^2$ with $\xi_0\approx 10^{-12}$ m and $r_c\approx 10^{-8}$ m, the estimate of the relaxation rate $\gamma_{1\mathrm{r}}$ amounts to 10^6 κ s^{-1}, which is comparable to $\Delta\Omega_0$. Hence the effect of thermal lattice fluctuations on the resonance frequency may be responsible for the anomalous behavior of the resonance lineshape in the incommensurate phase of Cs_2ZnI_4.

3. THE QUADRATIC CONTRIBUTION OF FLUCTUATIONS

The effect of lattice fluctuations in this case reduces to

$$\Omega(\mathbf{r},t)=\Omega_{0\mathrm{r}}+\Omega_{1\mathrm{r}}\eta_{1\mathrm{r}}'(t)+\Omega_{2\mathrm{r}}\eta_{1\mathrm{r}}'^2(t)/2,$$

where $\Omega_{2\mathrm{r}}=\Omega''(\xi_0\cos(qz))$, with $\Omega''(\eta)=d^2\Omega(\eta)/d\eta^2$. The Gaussian nature of the random process that the lattice atoms undergo allows for the following approach. We transform $G_{\mathrm{r}}(t)$, the Fourier transform of a sample-nonaveraged resonance line, to the following form:

$$G_{\mathrm{r}}(t)=\left\langle \exp\left\{-i\int_0^t dt'\Omega(\mathbf{r},t')\right\}\right\rangle$$
$$=\exp(-i\Omega_{0\mathrm{r}}t)\sum_{n=0}^{\infty}\frac{1}{n!}\left\langle\left\{-i\int_0^t dt'[\Omega(\mathbf{r},t')-\Omega_{0\mathrm{r}}]\right\}^n\right\rangle. \quad (7)$$

As is known,[15] the average of an arbitrary product of lattice variables in a Gaussian process can be represented in the form of the sum of products of pairwise correlation functions obtained by all possible pairings. In the graphic representation of the result of averaging for the nth term in the series (7), the connected diagrams are chains and loops,[16]

$$\left\langle\left\{\int_0^t dt'[\Omega(\mathbf{r},t')-\Omega_{0\mathrm{r}}]\right\}^n\right\rangle_{\mathrm{con}}=\frac{n!C_{n\mathrm{r}}+(n-1)!O_{n\mathrm{r}}}{2},$$

where

$$C_{n\mathrm{r}}=\Omega_{1\mathrm{r}}^2\Omega_{2\mathrm{r}}^{n-2}\times\prod_{j=1}^{n}\left\{\int_0^t dt_j\right\}\langle\eta_{\mathrm{r}1}'\eta_{\mathrm{r}2}'\rangle\langle\eta_{\mathrm{r}2}'\eta_{\mathrm{r}3}'\rangle\cdots\langle\eta_{\mathrm{r}n-1}'\eta_{\mathrm{r}n}'\rangle$$

is the nth-order chain, and

$$O_{n\mathrm{r}}=\Omega_{2\mathrm{r}}^n\prod_{j=1}^{n}\left\{\int_0^t dt_j\right\}\langle\eta_{\mathrm{r}1}'\eta_{\mathrm{r}2}'\rangle\langle\eta_{\mathrm{r}2}'\eta_{\mathrm{r}3}'\rangle\cdots\langle\eta_{\mathrm{r}n}'\eta_{\mathrm{r}1}'\rangle$$

is the nth-order loop, with $\eta_{\mathrm{r}j}'\approx\eta_{1\mathrm{r}}'(t_j)$. The name of each diagram depends on whether or not the initial and final moments of time in the sequence of the multiplied correlation functions coincide. While loops are generated exclusively by the quadratic contribution of fluctuations, chains (starting at $n=3$) are generated by both linear and quadratic contributions of fluctuations. The chain with $n=2$ is generated exclusively by the linear contribution of fluctuations and is represented in (3).

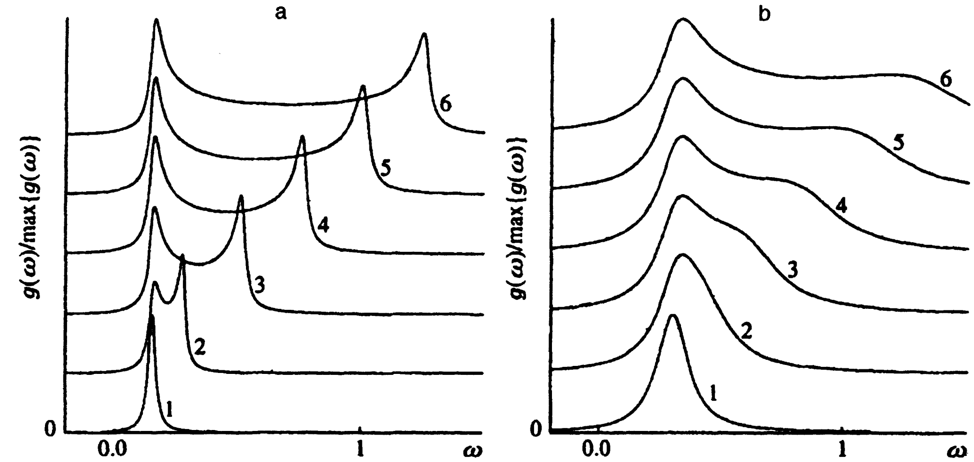

FIG. 3. Intensity-normalized lineshape of a magnetic resonance in the incommensurate phase of a crystal on atoms occupying particular positions, caused by the quadratic contribution of fluctuations. Notation: (a) $\Gamma=1$, and (b) $\Gamma=10$; curve *1*, $\xi_0^2=0$; curve *2*, $\xi_0^2=0.5$; curve *3*, $\xi_0^2=1$; curve *4*, $\xi_0^2=1.5$; curve *5*, $\xi_0^2=2$; and curve *6*, $\xi_0^2=2.5$.

According to Mayer's theorem,[17] the sum of all connected and disconnected diagrams (7) can be written as

$$G_{\rm r}(t)=\exp\{-i\Omega_{0{\rm r}}t+F_{C{\rm r}}(t)+F_{O{\rm r}}(t)\},$$

where

$$F_{C{\rm r}}(t)=\sum_{n=2}^{\infty}\frac{(-i)^nC_{n{\rm r}}}{2},\quad F_{O{\rm r}}(t)=\sum_{n=1}^{\infty}\frac{(-i)^nO_{n{\rm r}}}{2}.$$

A general approach to finding $F_{C{\rm r}}(t)$ and $F_{O{\rm r}}(t)$ for an arbitrary $\langle\eta'_{1{\rm r}}(t)\eta'_{1{\rm r}}\rangle$ is developed in Ref. 16 and involves solving an integral equation for an auxiliary series. A direct estimate for large times yields

$$F_{C{\rm r}}(t)\approx-\Omega_{1{\rm r}}^2\int_{-\infty}^{\infty}d\varepsilon\langle\eta_{\rm r}^2\rangle_\varepsilon\frac{1-\cos(\varepsilon t)}{2\pi\varepsilon^2}[1+i\Omega_{2{\rm r}}\langle\eta_{\rm r}^2\rangle_\varepsilon],$$
$$F_{O{\rm r}}(t)\approx-t\int_{-\infty}^{\infty}d\varepsilon\ln[1+i\Omega_{2{\rm r}}\langle\eta_{\rm r}^2\rangle_\varepsilon]/4\pi. \quad (8)$$

In contrast to (6), for a quadratic contribution of fluctuations to the resonance frequency it is impossible to explicitly separate the contributions of amplitudons and phasons to $F_{C{\rm r}}(t)$ and $F_{O{\rm r}}(t)$. The contribution of the chains $F_{C{\rm r}}(t)$ shifts the resonance line of the rth spin in the direction opposite the sign of $\Omega_{2{\rm r}}$ and narrows it in comparison to (3). At the same time the contribution of the loops $F_{O{\rm r}}(t)$ shifts the resonance line in the direction of the sign of $\Omega_{2{\rm r}}$ and broadens it.

Figure 3 illustrates the case of numerically modeling the variation of the magnetic resonance lineshape with the temperature reduced below T_i with $\Omega(\eta)=\eta^2/2$. The coefficients were set to $B=D=v=1$ and $\gamma_g=0$. The differences from the case of the linear contribution of fluctuations are listed below.

The contribution of loops causes a shift and broadening of the resonance line already at temperatures above T_i. Because of this broadening and the broadening caused by the linear contribution of fluctuations on atoms in general positions of the crystal cell,[9] the quasicontinuous distribution of the resonance frequencies in the incommensurate phase of the crystal remains unobservable over a certain temperature range below T_i.

As Eqs. (4) and (8) imply, essentially only amplitudons affect peaks with temperature-dependent frequencies. The decrease in the amplitudon contribution to $\langle\eta_{\rm r}^2\rangle_\varepsilon$ as the temperature is reduced leads, correspondingly, to a growth of these peaks [the peak at the frequency $\Omega(\xi_0)$ in Fig. 3]. But on atoms in particular positions of the crystal cell the increase in the contribution of chains balances the lowering of these peaks obtained for the linear contribution of fluctuations.

On the other hand, only the loop contribution affects peaks with temperature-independent frequencies. Hence these peaks also grow as the temperature is lowered, but the growth is limited by the phason contribution to $\langle\eta_{\rm r}^2\rangle_\varepsilon$ [Eq. (4)], which is essentially temperature-independent.

I am grateful to I. P. Aleksandrova, V. B. Zobov, and S. V. Primak for useful discussions. This work was supported financially by the Russian Committee of Institutions of Higher Learning (Project No. 94-31.3-14).

[1] H. Z. Cummins, Phys. Rep. **185**, 211 (1990).
[2] A. Abragam, *The Principles of Nuclear Magnetism*, Clarendon Press, Oxford (1961).
[3] P. W. Anderson, J. Phys. Soc. Jpn. **9**, 316 (1954).
[4] A. K. Moskalev, I. A. Belobrova, and I. P. Aleksandrova, Fiz. Tverd. Tela (Leningrad) **20**, 3288 (1978).
[5] R. Blinc, P. Prelovšek, V. Rutar *et al.*, in *Incommensurate Phases in Dielectrics*, Vol. 1, R. Blinc and A. P. Levanyuk (eds.), North-Holland, Amsterdam (1986), p. 143.
[6] I. P. Aleksandrova, in *Incommensurate Phases in Dielectrics*, Vol. 1, Blinc and A. P. Levanyuk (eds.), North-Holland, Amsterdam (1986), p. 277.
[7] R. Blinc, D. C. Ailion, P. Prelovšek, and V. Rutar, Phys. Rev. Lett. **50**, 67 (1983).
[8] R. Blinc, F. Milia, B. Topič, and S. Žumer, Phys. Rev. B **29**, 4173 (1984).
[9] A. M. Fajdiga, T. Apih, J. Dolinšek *et al.*, Phys. Rev. Lett. **69**, 2721 (1992).
[10] M. A. Popov, I. P. Aleksandrova, and S. V. Primak, JETP Lett. **59**, 485 (1994).
[11] J. Dolinšek, A. M. Fajdiga-Bulat, T. Apih *et al.*, Phys. Rev. B **50**, 9729 (1994).
[12] I. P. Aleksandrova, S. V. Primak, E. V. Shemetov, and A. I. Kruglik, Fiz.

Tverd. Tela (Leningrad) **33**, 1344 (1991) [Sov. Phys. Solid State **33**, 758 (1991)].
[13] V. L. Ginzburg, Fiz. Tverd. Tela (Leningrad) **2**, 2031 (1960) [Sov. Phys. Solid State **2**, 1824 (1961)].
[14] A. P. Levanyuk and D. G. Sannikov, Fiz. Tverd. Tela (Leningrad) **18**, 423 (1976).
[15] M. C. Wang and C. E. Uhlenbeck, Rev. Mod. Phys. **17**, 323 (1945).
[16] V. E. Zobov and M. A. Popov, Teoret. Mat. Fiz. **102**, 305 (1995).
[17] G. Uhlenbeck and G. Ford, *Lectures in Statistical Mechanics*, Mir Publishers, Moscow (1965), p. 60.

Translated by Eugene Yankovsky