Effect of fluctuations on the NQR spectrum in the incommensurate phase of a Cs_2ZnI_4 crystal

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The direct spin–lattice relaxation due to local thermal fluctuations of the amplitude of the atomic-displacement wave in the incommensurate phase of a crystal can strongly influence the lineshape of the resonant spectrum, reducing the intensity of the outermost temperature-dependent peaks in the frequency distribution, while having essentially no effect on the temperature-independent features of this distribution. This effect is apparently the reason for the anomalous behavior of the NQR spectrum of ^{127}I in Cs_2ZnI_4 in a certain temperature interval near the transition from the "normal" phase to the incommensurate phase.

A study[1] of the incommensurate phase in Cs_2ZnI_4 ($T_i = 118$ K, $T_c = 108$ K) by the NQR method has revealed an unusual temperature dependence of the spectrum of ^{127}I. Below T_i, instead of the splitting of the line of the "normal" phase into two peaks, which bound the continuous spectrum of resonant frequencies (see Ref. 2 and the papers cited there), one observes a single asymmetric peak, just below the transition. The other outermost structural feature, on the other hand, gradually emerges from the noise level as the temperature is lowered. Roughly at the center of the region in which the narrow incommensurate phase exists, the NQR spectrum assumes a shape typical of the model of a static plane-wave modulation.[2] In the spectrum of one of the structurally nonequivalent iodine nuclei, at a lower temperature, a third structural feature gradually emerges from the noise level in the frequency distribution, at a higher frequency.

As was shown in Ref. 3, the deviation of the shape of the NMR line from the static model[2] just below T_i can be attributed to thermal fluctuations. However, the case of a linear relationship between the resonant frequency and the order parameter, which was the case studied in Ref. 3, does not describe the particular features of the NQR spectrum in Cs_2ZnI_4.

We use the representation in the adiabatic approximation,[4]

$$I(\omega) = (2\pi L)^{-1} \int_{-\infty}^{\infty} \int_0^L \left\langle \exp\left\{ i \left[\omega t - \int_0^t \Omega(\eta(x,\tau))d\tau \right] \right\} \right\rangle dx dt, \qquad (1)$$

where Ω is the resonant frequency, which depends on the local order parameter η, the angle brackets mean an average over an ensemble, and the integration over x is carried out in order to average the spectrum over the 1D modulation assumed in this paper.

Since the fluctuation region of structural phase transitions is narrow ($|T-T_c|/T_c \ll 10^{-3}$–10^{-4}; Ref. 5), we can use the Landau theory, and we can assume that the distribution of fluctuations in the order parameter is Gaussian. We restrict the calculations below to (a) the plane-wave region,

$$\eta(x,t)=\eta_1(x,t)\cos(qx)+\eta_2(x,t)\sin(qx)$$

$$=[\eta_0+\eta_1'(x,t)]\cos(qx)+\eta_2'(x,t)\sin(qx),$$

where $q=2\pi/L$, $\eta_0=\langle n_1(x,t)\rangle$, the wave phase is given by $\langle\eta_2(x,t)\rangle=0$, and the fluctuations $\eta_1'(x,t)$ and $\eta_2'(x,t)$ result from thermal excitation of respectively amplitudon modes and phason modes; and (b) direct spin–lattice relaxation, for which it is sufficient to retain the following expression in the expansion of $\Omega(\eta)$ in the fluctuations:

$$\Omega(\eta)\approx\Omega[\eta_0\cos(qx)]+\Omega'[\eta_0\cos(qx)][\eta_1'(x,t)\cos(qx)+\eta_2'(x,t)\sin(qx)],$$

where $\Omega'(\eta)=d\Omega(\eta)/d\eta$. Since the NQR frequencies are small in comparison with the frequencies of crystal modes, the correlation functions of the Fourier components of these fluctuations which dominate (1) can be written in Debye form:

$$\langle|\eta_1'(\mathbf{k},\omega)|^2\rangle=2\gamma\, k_BT/[\gamma^2\omega^2+(\alpha+\delta\mathbf{k}^2)^2],$$

$$\langle|\eta_2'(\mathbf{k},\omega)|^2\rangle=2\gamma k_BT/[\gamma^2\omega^2+\delta\mathbf{k}^4],$$

where $\alpha\sim|T-T_i|$. Under these assumptions, expression (1) becomes

$$I(\omega)\approx(2\pi L)^{-1}\int_{-\infty}^{\infty}\int_0^L \exp\{i[\omega-\Omega(\eta_0\cos(qx))]t-|\Omega_1(x)t|$$

$$-|\Omega_2(x)t|^{3/2}\}dxdt, \qquad (2)$$

where

$$\Omega_1(x)=\gamma k_BTr_c\Omega'[\eta_0\cos(qx)]^2\cos(qx)^2/4\pi\delta^2, \quad r_c=(\delta/\alpha)^{1/2},$$

$$\Omega_2(x)=\gamma^{1/3}[k_BT\Omega'(\eta_0\cos(qx))^2\sin(qx)^2/6]^{2/3}/\pi\delta.$$

To pursue the calculations would require numerical methods. We will accordingly analyze the expression derived here. If we ignore the spin–lattice relaxation through amplitudons and phasons, then we have $\Omega_1(x)\approx\Omega_2(x)\approx0$, and expression (2) becomes the known expression for the static model: $I(\omega)=L^{-1}\Sigma|\partial\Omega[\eta_0\cos(qx)/\partial x|^{-1}]$, where the summation is over all the solutions of the equation $\omega=\Omega[\eta_0\cos(qx)]$. The continuous frequency distribution contains peaks corresponding to extrema of the frequency distribution $\partial\Omega[\eta_0\cos(qx)/\partial x=\Omega'(\eta_0\cos(qx))(-q\sin(qx))]=0$. The frequencies of the peaks corresponding to $\sin(qx)=0$ vary with the temperature in accordance with $\Omega(\pm\eta_0)$, while those corresponding to $\Omega'[\eta_0\cos(qx)]=0$ do not vary.

The relaxation through phasons has no practical effect on any of these peaks, since the condition $\Omega_2(x)=0$ holds at their frequencies. The effect of the relaxation through amplitudons, in contrast, is selective: We have $\Omega_1(x)=0$ for peaks with temperature-independent frequencies but $\Omega_1(x)\neq 0$ for peaks with temperature-dependent frequencies. As a result, the latter may be shrunk or completely suppressed. Because of the broadening of each individual spin packet in this part of the spectrum, an intensity peak may disappear completely in the noise. This circumstance may account for the fact that the usual splitting of an NQR spectral line was not observed below T_i in Ref. 1. The initial increase in the second and third peaks from the noise level and the weak temperature dependence of their frequencies may be due to the decay of $\Omega_1(x)$ as the temperature-dependent peak approaches the frequency of a corresponding temperature-independent peak.

If the resonating atoms occupy common sites in the crystal cell at $T \geqslant T_i$, and if $\Omega'(0)$ dominates the expansion of $\Omega'(\eta_0)$ in η_0, then the relaxation rate of $\Omega_1(x)$ falls off in proportion to r_c as the temperature is lowered below T_i, and the peaks with temperature-dependent frequencies may be restored with distance from the transition. In the case of a particular position, $\Omega'(\eta_0)$ increases with decreasing temperature no more slowly than $\eta_0 \sim r_c^{-1}$. This behavior leads to an increase in $\Omega_1(x)$ and to a pronounced attenuation of these peaks. The peaks begin to be restored only in the η_0 saturation region.

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[1] I. P. Aleksandrova *et al.*, Fiz. Tverd. Tela (Leningrad) **33**, 1344 (1990) [Sov. Phys. Solid State **33**, 758 (1990)].
[2] I. P. Aleksandrova, in: *Incommensurate Phases in Dielectrics*, ed. by R. Blinc and A. P. Levanyuk (North-Holland, Elsevier Sci. Publ., Amsterdam, 1986), p. 277.
[3] A. M. Fajdiga *et al.*, Phys. Rev. Lett. **69**, 2721 (1992).
[4] A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon Press, Oxford, 1961).
[5] V. L. Ginzburg, Fiz. Tverd. Tela (Leningrad) **2**, 2031 (1960) [Sov. Phys. Solid State **2**, 1824 (1960)].

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