Spin-lattice relaxation due to sliding of the modulation wave in incommensurate systems with impurities

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A theory of spin-lattice relaxation via large-scale fluctuations of the modulation wave in incommensurate systems is developed for the case of impurity pinning. For a simple model used for the description of such fluctuations, the motionally averaged spectrum and the positional dependence of T_1 in the slow-motion regime are calculated and illustrated graphically. In the thermally depinned regime close to the paraelectric-incommensurate transition temperature T_I, an anomalous temperature and Larmor-frequency dependence of the spin-lattice relaxation rate T_1^{-1} is predicted. It is shown that, on crossing T_I from above, T_1 continues to decrease with decreasing temperature until it reaches a shallow minimum in the thermally depinned incommensurate phase.

I. INTRODUCTION

The continuum theory of incommensurate (I) systems predicts the existence of a gapless sliding mode in the I phase.[1] This is the Goldstone mode which recovers the broken continuous phase symmetry of the incommensurate phase.[2] The above result does not correspond to physical reality because of the presence of impurities[3] and discrete lattice effects[4] which are responsible for the pinning of the modulation wave to the underlying lattice. The theory of spin-lattice (T_1) relaxation in incommensurate systems[5] has been therefore developed for the case of small fluctuations of the modulation wave which can be decomposed into phason and amplitudon modes.[1,6] We consider possible occurrence of spin-lattice relaxation via large-scale fluctuations of the modulation wave in the presence of impurity pinning.

Recent NQR, NMR,[3,7] and EPR[8] experiments have shown that thermal depinning of the modulation wave may take place in the high-temperature part of the I phase close to the incommensurate-paraelectric transition temperature T_I. Electric-field-induced depinning has been observed in charge density wave (CDW) systems[9] and the effect of a uniform sliding of the modulation wave on T_1 has been evaluated.[10] Theories for the motional narrowing of the NMR line shape due to sliding and large-scale phase fluctuations of the completely[11] or partially depinned[12] modulation wave have been developed. Here in Sec. II the behavior of the pinned modulation wave system is modeled by fluctuations of the modulation wave in a coherence volume of a square box shape. Theoretical expressions for the NMR spectrum and spin-lattice relaxation are developed in Sec. III. Results for a simplified one-dimensional case are presented graphically.

II. PHASE FLUCTUATIONS IN INCOMMENSURATE SYSTEMS

In the case of a pinned one-dimensional modulation wave we can divide the displacement u_j of the jth nucleus from its position in the paraelectric phase into a large static (or slowly moving) part $\mathbf{u}_{j0}$ and a small rapidly fluctuating part $\delta\mathbf{u}(t)$:

$$\mathbf{u}_j(t)=\mathbf{u}_{j0}+\delta\mathbf{u}_j(t)\ . \tag{1}$$

The rapidly fluctuating part can be in the harmonic approximation decomposed[1,6] into phason and amplitudon modes:

$$\delta\mathbf{u}_j(t)=\sum_{\mathbf{k}}\frac{\mathbf{e}_{\mathbf{k}}(j)}{\sqrt{2Nm_j}}[\cos(\mathbf{q}_s\cdot\mathbf{r}_j+\phi_0)(e^{-i\mathbf{k}\cdot\mathbf{r}_j}P_{A\mathbf{k}}+\text{c.c.})+\sin(\mathbf{q}_s\cdot\mathbf{r}_j+\phi_0)(e^{-i\mathbf{k}\cdot\mathbf{r}_j}P_{\varphi\mathbf{k}}+\text{c.c.})]\ . \tag{2}$$

Here $P_{A\mathbf{k}}$ and $P_{\varphi\mathbf{k}}$ represent the normal coordinates for the amplitudon and phason modes, $\mathbf{q}_s$ is the wave vector of the incommensurate modulation, $\mathbf{k}=\mathbf{q}-\mathbf{q}_s$, $\mathbf{e}_k$ is the polarization vector, m_j the mass of the jth nucleus and N the number of unit cells which are coherently moving.

The mean-square displacement is now obtained as

$$\overline{\delta u_j^2}=\sum_{\mathbf{k}}\frac{1}{Nm_jN_0}[\cos^2(\mathbf{q}_s\cdot\mathbf{r}_j+\phi_0)|P_{A\mathbf{k}}|^2+\sin^2(\mathbf{q}_s\cdot\mathbf{r}_j+\phi_0)|P_{\varphi\mathbf{k}}|^2]\ . \tag{3}$$

Here N_0 is the number of nuclei in the unit cell. Equation (3) leads to the familiar expression[5] for the spin-lattice relaxation rate in I systems:

$$T_1^{-1} \propto J_A \cos^2\phi_{0j} + J_\varphi \sin^2\phi_{0j} , \tag{4}$$

where J_A and J_φ are the local spectral densities of the amplitudon and phason modes and $\phi_{0j} = \mathbf{q}_s \cdot \mathbf{r}_j + \phi_0$. $P_{A\mathbf{k}}$ and $P_{\varphi\mathbf{k}}$ are oscillating with normal mode frequencies

$$\omega_{Ak}^2 = \omega_{A0}^2 + \kappa k^2 \tag{5a}$$

and

$$\omega_{\varphi k}^2 = \omega_{\varphi 0}^2 + \mathcal{K} k^2 . \tag{5b}$$

Here ω_{A0} and $\omega_{\varphi 0}$ are the amplitudon and the impurity-induced phason gap,[6] respectively. We have $\omega_{A0} \gg \omega_{\varphi 0}$ (except for $T \to T_I$) and $\sqrt{\mathcal{K} k_{\max}^2} \gg \omega_{A0}, \omega_{\varphi 0}$, as well as $\omega_{\varphi 0} \gtrsim \sqrt{\mathcal{K} k_{\min}^2}$. Here $k_{\max}$ is determined by the size of the unit cell and $k_{\min}$ by the crystal size in the defect (pinning center) free case and by the size of the defect free regions in real crystals. From the equipartition theorem we find

$$\tfrac{1}{2}\omega_{A\mathbf{k}}^2 |P_{A\mathbf{k}}|^2 = \tfrac{1}{2}\omega_{\varphi\mathbf{k}}^2 |P_{\varphi\mathbf{k}}|^2 = \tfrac{1}{2} kT . \tag{5c}$$

The largest phason contribution to the mean-square displacement is given by the $k_{\min}$ mode:

$$\left[\sqrt{\overline{\delta u_j^2}}\,\right]_{k_{\min}} \lesssim \left[\frac{kT}{N_0 N m_j}\right]^{1/2} \frac{1}{\omega_{\varphi 0}} \sin\phi_{0j} \tag{6}$$

and is inversely proportional to the phason gap. Here

$$N \approx \frac{\xi_\parallel \xi_\perp^2}{V_0} \le N_{\text{ideal crystal}} , \tag{7a}$$

where V_0 is the unit cell volume, $\xi_\parallel$ the coherence length along $\mathbf{q}_s$ and $\xi_\perp$ the coherence length perpendicular to $\mathbf{q}_s$. Both $\xi_\parallel$ and $\xi_\perp$ are k dependent in the case of impurity pinning.

We assume that $\xi_\parallel \approx \xi_\perp \approx \bar{l}$ where $\bar{l}$ is the mean distance between pinning defects

$$\bar{l} = \left[\frac{V_{\text{cryst}}}{N_{\text{def}}}\right]^{1/3} = n_D^{-1/3} \tag{7b}$$

which is inversely proportional to the cube root of the pinning defect density. So the mean-square displacement corresponding to $k_{\min}$ mode in the coherence volume $\bar{l}^3$ can be written as

$$\left[\sqrt{\overline{\delta u_j^2}}\,\right]_{k_{\min}} \lesssim \left[\frac{kTV_0}{N_0 m_j \bar{l}^3}\right]^{1/2} \frac{1}{\omega_{\varphi 0}} \sin\phi_{0j} . \tag{8}$$

The phason gap $\omega_{\varphi 0}$ is close to T_I proportional to a relatively high power of the amplitude of the modulation wave both for impurity[13] and for discrete lattice pinning.[14] In the first case one finds, for instance,[13]

$$\omega_{\varphi 0} \propto |T_I - T|^{\beta(n-2)} , \tag{9a}$$

whereas one gets in the second of the above two cases[14]:

$$\omega_{\varphi 0} \propto |T_I - T|^{\beta n} \exp[-C/(T_I - T)^{2\beta}] . \tag{9b}$$

Here $\beta \sim \frac{1}{3}$ is the critical exponent for the amplitude of the incommensurate modulation wave

$$u_{j0} \propto (T_I - T)^\beta , \tag{9c}$$

whereas the commensurability index n equals 6 for Rb_2ZnCl_4. Since $\omega_{\varphi 0}$ decreases on approaching T_I one may expect a thermal depinning of the modulation wave below the incommensurate-paraelectric transition, i.e., above a certain temperature thermal fluctuations should become large enough to overcome the pinning energy. For the case of a nearly free sliding of the modulation wave, $\omega_{\varphi 0} \to 0$, $\overline{\delta u_j^2}|_{k_{\min}}$ becomes very large, and the amplitudon and phason description of the elementary excitations breaks down. The same is true for the spin-lattice relaxation rate—expression (4)—which diverges[5] for $\omega_{\varphi 0} \to 0$ when the nuclear Larmor frequency $\omega_L \to 0$.

For the above case of large phase fluctuations we have to use the complete expression for the nuclear displacement

$$\mathbf{u}_j(t) = [\mathbf{u}_{j0} + \delta\mathbf{u}_{j0}(t)] \cos[\phi(\mathbf{r}_j, t)] \tag{10a}$$

and abandon the linearized description of the phase fluctuations as used in expressions (1) and (2).

Instead of expanding the nuclear displacement $\mathbf{u}_j(\mathbf{r},t)$ into a large quasistatic and a small rapidly fluctuating part we shall divide the phase $\phi(\mathbf{r},t)$ into a time-independent and a time-dependent part

$$\phi(\mathbf{r}_j, t) = \phi_{0j} + \sum_{\mathbf{k}=\mathbf{k}_{\min}}^{\mathbf{k}_{\max}} \varphi_{\mathbf{k}}(\mathbf{r}_j, t) , \tag{10b}$$

where the time-independent part equals $\phi_{0j} = \mathbf{q}_s \mathbf{r}_j + \phi_0$ and the time-dependent part is—for the case of partial pinning—expressed in terms of standing phase waves $\varphi_{\mathbf{k}}(\mathbf{r}, t)$.

For the case of impurity pinning the $\mathbf{k}_{\min}$ is roughly determined by the inverse average distance between defects. For simplicity we assume that the whole crystal can be viewed as a sum of coherence volumes of equal size $\bar{l}^3$ and further that the $\varphi_{\mathbf{k}}(\mathbf{r},t)$ can be represented by standing waves in a single square box (coherence volume):

$$\varphi_{\mathbf{k}}(\mathbf{r}, t) = \varphi_{0\mathbf{k}} \sin(\omega_{\mathbf{k}} t + \alpha_{\mathbf{k}}) s(\mathbf{k} \cdot \mathbf{r}) , \tag{11a}$$

where

$$s(\mathbf{k} \cdot \mathbf{r}) = \sin(k_x x) \sin(k_y y) \sin(k_z z) . \tag{11b}$$

So we have $(k_x)_{\min} = (k_y)_{\min} = (k_z)_{\min} = \pi/\bar{l}$. Instead of the symbol $\omega_{\varphi k}$ introduced for phason frequency, from now on ω_k will be used for the frequency of the large phase fluctuation modes. In the random phase approximation the phases α_k are randomly distributed in the interval $\alpha_k \in [0, 2\pi]$. Similarly we may assume that the distribution of the phase fluctuation amplitudes φ_{0k} in the interval $0 \le \varphi_{0k} \le \infty$ is Gaussian:

$$f(\varphi_{0k}) = \frac{2}{\pi \overline{\varphi_{0k}^2}} \exp(-\varphi_{0k}^2 / 2\overline{\varphi_{0k}^2}) . \tag{12}$$

The second moment of this distribution $\overline{\varphi_{0k}^2}$ can be obtained from

$$\sum_j \overline{\frac{m_j \dot{u}_j^2}{2}}\bigg|_k = \frac{1}{2}kT \tag{13a}$$

as the φ_{0k} are not normal modes. With

$$\dot{\varphi}_k = \varphi_{0k}\omega_k \cos(\omega_k t + \alpha_k) s(\mathbf{k}\cdot\mathbf{r}) \tag{13b}$$

and Eq. (10a) we find $\overline{\varphi_{0k}^2}$ as

$$\overline{\varphi_{0k}^2} \cong \frac{32kT}{N_0 N m}\frac{1}{u_0^2\omega_k^2}\ , \tag{13c}$$

where m and u_0^2 stand for averages over the nuclei in the unit cell and the average of $s^2(\mathbf{k}\cdot\mathbf{r})$ over the coherence volume has been taken to be equal to $\frac{1}{8}$.

It should be noticed that in the case of an ideal, defect free crystal the sum over $\mathbf{k}$ would contain also the term $\mathbf{k}=0$ describing uniform sliding of the modulation wave. In such a case we would have

$$\varphi_{\mathbf{k}=0} = \Omega t \tag{14a}$$

with $\Omega = \mathbf{q}_s \cdot \mathbf{v}$ being the harmonic-oscillation frequency of the jth nucleus induced by the motion of the modulation wave and $\mathbf{v}$ the uniform sliding velocity. The corresponding contribution to the spin-lattice relaxation rate is just a δ function at Ω:[10]

$$\frac{1}{T_1} \propto \delta(\omega_L - \Omega)\ . \tag{14b}$$

III. SPIN-LATTICE RELAXATION DUE TO LARGE PHASE FLUCTUATIONS IN THE PRESENCE OF IMPURITY PINNING

Let us now assume that we deal with nuclei with a nonzero quadrupole moment as well as that the wavelength of the incommensurate modulation is much larger than the radius of the region from which important contributions to the electric field gradient (EFG) tensor at the nuclear site come from.[5] Expanding the EFG tensor at the lth nuclear site in powers of nuclear displacements we find in the local approximation[5]

$$T_l^{(\mu)}(t) = T_l^{(\mu)} + \mathbf{u}_l(t)\cdot \mathbf{T}_{01}^{(\mu)}(l) + \cdots\ . \tag{15}$$

Here $\mathbf{T}_{01}^{(\mu)}(l)$ represents the change in the μth component of the EFG tensor at the lth nuclear site due to the nuclear displacements $\mathbf{u}_l(t)$ induced by the motion of the modulation wave.

The resulting frequency shift can be to the first order in displacements written as

$$\omega_l = \omega_0 \cos[\phi(\mathbf{r}_l, t)]\ , \tag{16}$$

where ω_0 is the amplitude of the frequency shift. The corresponding spectrum can be calculated using the well-known relation:[11]

$$F(\omega) = \sum_l \int_{-\infty}^{\infty} \left\langle \exp\left[i\int_0^t \omega_l(t')dt'\right]\right\rangle e^{i\omega t} dt\ , \tag{17}$$

where the summation goes over all nuclei which are moving coherently.

In the slow motion regime this formula gives a well-known static spectrum with two edge singularities.[11] With increasing frequencies ω_k (speed of fluctuations) the spectrum becomes more and more motionally averaged. The spectrum gradually transforms from a two peak spectrum to a three peak spectrum and finally to a spectrum with a single central peak. In Fig. 1 this transition is illustrated for a simplified one-dimensional case where 100 nuclei are uniformly distributed in the interval $[0,\bar{l}]$ on ξ axis. Equation (11b) is here reduced to $s=\sin k\xi$ with $k=\pi/\bar{l}, 2\pi/\bar{l}, \ldots, 100\pi/\bar{l}$. The amplitude of the phase fluctuations $(\overline{\varphi_{k_{\min}}^2})^{1/2}$ corresponding to the wave vector $\pi/\bar{l}$ is here chosen to be 1.8 while the ratio of the corresponding frequency $\omega_{k_{\min}}$ to the frequency shift ω_0 is varied. The specific shape of the partially motionally averaged spectrum in the transition region [Figs. 1(b) and 1(c)] is model dependent.

The nuclear spin-lattice relaxation is here determined by the spectral density of the correlation function of $\mathbf{u}_l(t)$, i.e., by

$$G_l(t) = \overline{\mathbf{u}_l(0)\cdot\mathbf{u}_l(t)} \tag{18}$$

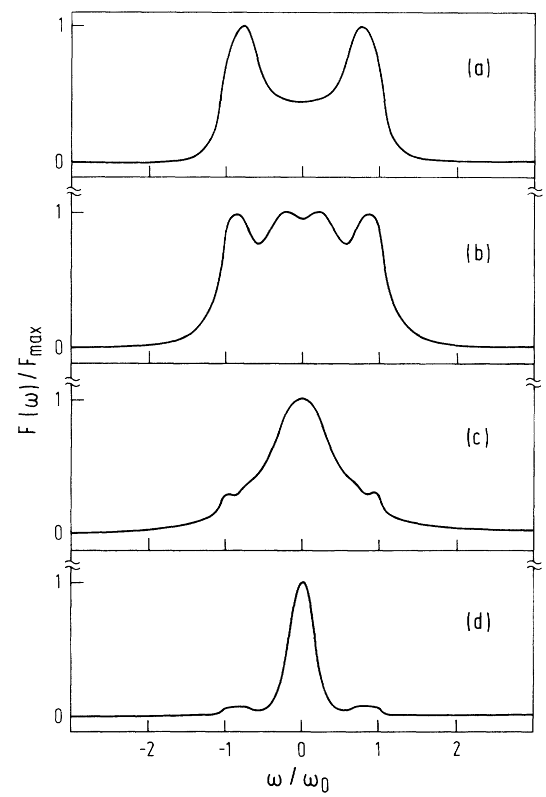

FIG. 1. Motional averaging of incommensurate NMR line shape for large-scale phase fluctuations with $\sqrt{\overline{\varphi}_{k_{\min}}^2}=1.8$ and $\omega_0/\omega_{k_{\min}}=21.6$ (a), 10.8 (b), 5.4 (c), 1.35 (d). Here ω_0 measures the incommensurate frequency shift and $\omega_{k_{\min}}$ the fluctuation rate.

which can be written as a sum of amplitude and phase terms,

$$G_l(t)=G_{Al}(t)+G_{\varphi l}(t) \tag{19a}$$

if amplitude fluctuations are much faster than the large-scale phase fluctuations so that the two modes are essentially uncoupled. Here

$$G_{\varphi l}(t)=u_{0l}^2\overline{\cos[\phi(\mathbf{r}_l,0)]\cos[\phi(\mathbf{r}_l,t)]} \tag{19b}$$

and

$$G_{Al}(t)=\overline{\delta\mathbf{u}_{0l}(0)\cdot\delta\mathbf{u}_{0l}(t)}G_{\varphi l}(0)/u_{0l}^2 \ . \tag{19c}$$

Using Eq. (10b) the autocorrelation function $G_{\varphi l}(t)$ can be in the random phase approximation expressed as

$$G_{\varphi l}(t)=\frac{u_{0l}^2}{2}\{\overline{\cos[\phi(\mathbf{r}_l,0)-\phi(\mathbf{r}_l,t)]}+\overline{\cos[\phi(\mathbf{r}_l,0)+\phi(\mathbf{r}_l,t)]}\}$$
$$=\frac{u_{0l}^2}{4}\left[e^{2i\phi_0}\prod_k \overline{\exp\{i[\varphi_k(0)+\varphi_k(t)]\}}+\prod_k \overline{\exp\{i[\varphi_k(0)-\varphi_k(t)]\}}+\text{c.c.}\right]_l \ . \tag{20a}$$

The amplitude correlation function $G_{Al}(t)$ can be, on the other hand, expressed as

$$G_{Al}=\sum_k \frac{1}{N_0Nm_lu_{0l}^2}\overline{P_{Ak}(0)P_{Ak}^*(t)}G_{\varphi l}(0) \tag{20b}$$

if the normal coordinate expansion, Eq. (2), is used.

In the limit of small phase fluctuations ($\sum_k \varphi_k \ll 1$) one finds

$$G_{\varphi l}(t)=u_{0l}^2\left[\frac{1}{2}\cos(2\phi_{0l})+\sin^2\phi_{0l}\sum_k \overline{\varphi_k(0)\varphi_k(t)}\right] , \tag{21a}$$

$$G_{Al}(t)=\sum_k \frac{1}{N_0Nm_l}\overline{P_{Ak}(0)P_{Ak}^*(t)}\cos^2\phi_{0l} \ . \tag{21b}$$

This leads—after transforming the $\varphi_k(t)$ into the $P_{\psi k}$—to the well-known[5] expression for relaxation via phasons and amplitudons given by Eq. (4).

For large phase fluctuation the situation is different. The standing wave approximation [Eq. (11)] enables us to write

$$\varphi_k(0)-\varphi_k(t)=2\varphi_{0k}\cos\left[\frac{\omega_k t}{2}+\alpha_k\right]\sin\left[\frac{\omega_k t}{2}\right]s \ , \tag{22a}$$

$$\varphi_k(0)+\varphi_k(t)=2\varphi_{0k}\sin\left[\frac{\omega_k t}{2}+\alpha_k\right]\cos\left[\frac{\omega_k t}{2}\right]s \ , \tag{22b}$$

and to find

$$\overline{\exp\{i[\varphi_k(0)-\varphi_k(t)]\}}=\frac{1}{2\pi}\int_0^{2\pi}\int_0^{\infty}f(\varphi_{0k})\exp\{i[\varphi_k(0)-\varphi_k(t)]\}d\varphi_{0k}d\alpha_k \ , \tag{23a}$$

where the Gaussian distribution $f(\varphi_{0k})$ is given by expression (12). The above expression becomes

$$\overline{\exp\{i[\varphi_k(0)-\varphi_k(t)]\}}=\left[\frac{2}{\pi\overline{\varphi_{0k}^2}}\right]^{1/2}\int_0^{\infty}J_0\left[2\varphi_{0k}s\sin\frac{\omega_k t}{2}\right]e^{-\varphi_{0k}^2/2\overline{\varphi_{0k}^2}}d\varphi_{0k}$$
$$=e^{-z_1}I_0(z_1) \tag{23b}$$

with $z_1=4s^2\overline{\varphi_{0k}^2}\sin^2(\omega_k t/2)$. The $\overline{\exp\{i[\varphi_k(0)+\varphi_k(t)]\}}$ term can be treated similarly.

Here J_0 is the zeroth order Bessel function whereas $I_0(z)$ is the zeroth order modified Bessel function defined by

$$I_0(z)=\sum_{m=0}^{\infty}\frac{(z/2)^{2m}}{(m!)^2} \ . \tag{23c}$$

The correlation function (18) now becomes

$$G_{\varphi l}(t)=\frac{u_{0l}^2}{2}\left[\prod_k e^{-z_1}I_0(z_1)+\cos(2\phi_{0l})\prod_k e^{-z_2}I_0(z_2)\right]_l , \tag{24}$$

where $z_2=4s^2\overline{\varphi_{0k}^2}\cos^2(\omega_k t/2)$ and z_1 has been defined above.

Limiting our discussion to the case of large fluctuations $\sum_k s^2\overline{\varphi_{0k}^2}\gg 1$ we note that $G_{\varphi l}(t)$ will decay to zero in a time $t \ll \omega_k^{-1}$ for all k. Therefore we shall investigate only the short time behavior of $G_{\varphi l}(t)$, i.e., the behavior for $\omega_k t \ll 1$. Using $\sin x \approx x$, $\cos x \approx 1$ for $x \ll 1$ and assuming that $z_1 \ll 1$ we get

$$G_{\varphi l}(t)=\frac{u_{0l}^2}{2}[\exp(-p^2t^2/2)+D_2\cos(2\phi_{0l})] , \quad t<\omega_{k_{\max}}^{-1} , \tag{25a}$$

where taking into account Eq. (13c), $(1/NN_0)\sum_k s^2=\frac{1}{8}$ and $u_0=u_{00}(T_I-T/T_I)^\beta$ we have

$$p=\left[\frac{8kT}{m}\right]^{1/2}\frac{l}{u_0}=p_0\left[\frac{T_I-T}{T_I}\right]^{-\beta} \tag{25b}$$

and

$$D_2=\exp\left[-\sum_k \overline{4s^2\varphi_{0k}^2}\right]\prod_k I_0(\overline{4s^2\varphi_{0k}^2})\ll 1 . \tag{25c}$$

In the long time limit, $t>\omega_{k_{\min}}^{-1}$, on the other hand, $G_{\varphi l}(t)$ becomes a constant:

$$G_{\varphi l}(t)\approx u_{0l}^2 D_1^2\cos^2\phi_{0l}\ll 1 , \tag{26a}$$

where

$$D_1=\exp\left[-\sum_k \overline{s^2\varphi_{0k}^2}\right]\prod_k I_0(\overline{s^2\varphi_{0k}^2}) . \tag{26b}$$

Both D_1 and D_2 are zero at T_I and increase on going into the I phase.

The spectral density of the EFG tensor fluctuations induced by large-scale phase fluctuations and small amplitude fluctuations of the modulation wave is now obtained as

$$T_1^{-1}\propto J_l^{(\mu)}(\omega)=\int_{-\infty}^{+\infty}\overline{\Delta T^{(\mu)}(0)\Delta T^{(-\mu)}(t)}e^{i\omega t}dt =[(J_l)_\varphi+(J_l)_A](T_{0l}^{(\mu)})^2 , \tag{27a}$$

where

$$(J_l)_\varphi=\int_{-\infty}^{+\infty}G_{\varphi l}(t)e^{i\omega t}dt=\frac{\sqrt{\pi}}{p}e^{-\omega^2/4p^2}u_0^2 \tag{27b}$$

and

$$(J_l)_A=\int_{-\infty}^{+\infty}G_{Al}(t)e^{i\omega t}dt\propto\omega_{A0}^{-1} . \tag{27c}$$

Here p decreases and u_0^2 increases on cooling into the I phase:

$$u_0^2\propto(T_I-T)^{2\beta},\quad p=p_0[(T_I-T)/T_I]^{-\beta} \tag{28a}$$

so that the relaxation rates become

$$(T_{1\varphi}^{-1})\propto(T_I-T)^{3\beta}\exp\left[-\frac{\omega^2}{4p_0^2}\left[\frac{(T_I-T)}{T_I}\right]^{2\beta}\right] \tag{28b}$$

and

$$(T_{1A}^{-1})\propto(T_I-T)^\beta . \tag{28c}$$

As in this case the large-scale sliding motions are fast enough, the incommensurate NMR frequency distribution will be motionally averaged out and only a single homogeneous central line with a single T_1^{-1} will be observed.[3,11] In such a case it will be experimentally impossible to separate $(T_{1\varphi}^{-1})$ and (T_{1A}^{-1}) and the effective relaxation rate will be determined by the faster process, i.e., by large-scale phase fluctuations. The resulting spin-lattice relaxation time T_1 induced by large-scale phase fluctuations shows—for not too large p_0 values—an exponential dependence on the square of the nuclear Larmor frequency. The predicted temperature dependence of T_1 is also anomalous: On cooling into the I phase T_1 first decreases with increasing T_I-T, goes through an asymmetric minimum and then increases at still larger T_I-T values. The minimum disappears if $\omega/p_0\ll 1$ (Fig. 2). These predictions are in agreement with the preliminary study performed by Papavassiliou *et al.*[15]

The above described behavior is completely different from the behavior expected[5] in the case of small phase fluctuations. Here the phason induced T_1 is essentially T independent and proportional to the phason gap Δ_φ whereas the amplitudon induced T_1 increases with decreasing T as the amplitudon gap: $\omega_{A0}\propto(T_I-T)^\beta$. The minimum value of T_1 is found here at the paraelectric-I transition temperature T_I and not below T_I as in the case of a floating modulation wave, [Eq. (28b)].

A situation may however arise where some of the sliding motions are too slow to affect the NMR spectrum (though they may still influence T_1) whereas the high frequency part of sliding motions and the amplitudon modes have a too small amplitude to be effective in averaging out the NMR spectrum [see Fig. 1(a)]. In such a case $(J)_\varphi$ must be directly numerically calculated from Eq. (24). One observes a variation of T_1 over the incommensurate frequency spectrum which is due to dependence of the nuclear displacements in the pinned modulation wave on the distance from the pinning centers. According to expression (11) these displacements are zero at the pinning centers and reach a maximum in the middle. This situation is illustrated in Fig. 3 where the spatial dependence of the T_1^{-1} is presented for the same one-

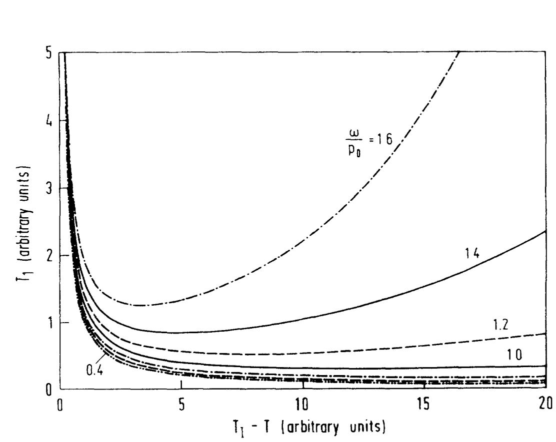

FIG. 2. Temperature and Larmor-frequency dependence of the spin-lattice relaxation time T_I induced by large-scale phase fluctuations of the pinned incommensurate modulation wave. The curves are calculated for different values of ω/p_0.

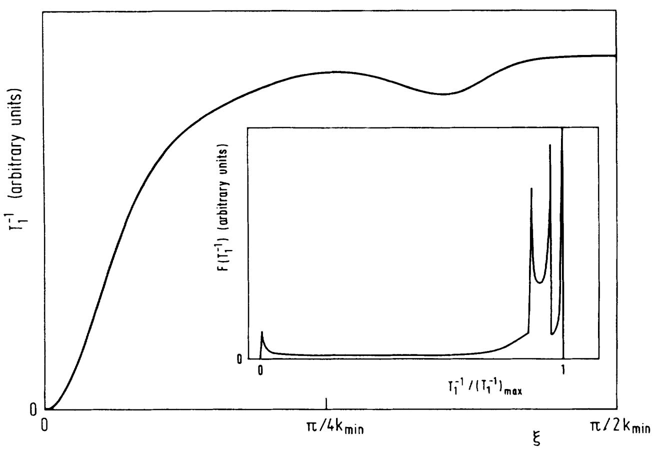

FIG. 3. Variation of T_1^{-1} over the incommensurate frequency distribution for the case of large "slow" phase fluctuations of the pinned modulation wave. The inset is showing the corresponding distribution of relaxation rates.

dimensional model used above in the calculation of the shape of the spectrum. The coordinate ξ measures the position and runs from zero to one half of the distance between two pinning center $\bar{l}/2(=\pi/2k_{min})$. According to our assumption that the resonant nuclei are uniformly distributed along this distance the distribution of the relaxation rates $\mathcal{F}(T_1^{-1})$ is simply obtained from

$$\mathcal{F}(T_1^{-1})dT_1^{-1}=\rho(\xi)d\xi, \quad \rho(\xi)=\text{const} \tag{29}$$

by numerically calculating $d(T^{-1})/d\xi$. This distribution is shown in the inset to Fig. 3. It is significantly different from the one found in conventional incommensurate systems when small phason and amplitudon fluctuations determine the relaxation rate.[6] The group of strong peaks around $T_1^{-1}/(T_1^{-1})_{max}\approx 1$ is due to the fact that a large fraction of nuclei situated near the middle of the line between the two pinning centers move with nearly the same displacement amplitude, whereas the small peak near $T_1^{-1}\approx 0$ corresponds to nuclei close to the pinning centers where the displacement amplitude is very small. $\mathcal{F}(T_1^{-1})$ can be directly obtained from the Laplace transform of the magnetization recovery function:

$$\Delta M(t)/M_0=\int_0^{\infty}\mathcal{F}(T_1^{-1})e^{-t/T_1}dT_1^{-1} . \tag{30}$$

In the intermediate case [Figs. 1(b) and 1(c)] the spectrum is partially motional averaged and there is no one-to-one correspondence between the resonant frequency and the spin-lattice relaxation rate.

[1] A. D. Bruce and R. A. Cowley, J. Phys. C **11**, 3609 (1978).

[2] P. Bak, Rep. Prog. Phys. **45**, 587 (1982).

[3] R. Blinc, D. C. Ailion, P. Prelovšek, and V. Rutar, Phys. Rev. Lett. **50**, 67 (1983); F. Milia, G. Papavassiliou, and A. Anagnostopoulos, Phys. Rev. B **43**, 11464 (1991).

[4] D. A. Bruce, J. Phys. C **13**, 4615 (1980).

[5] S. Žumer and R. Blinc, J. Phys. C **14**, 465 (1981).

[6] See, for instance, J. D. Axe, M. Iizumi, and G. Shirane, in *Incommensurate Phases in Dielectrics,* edited by R. Blinc and A. P. Levanyuk (North-Holland, Amsterdam, 1986), Vol. II, p. 1.

[7] B. Topič, U. Haeberlen, R. Blinc, unpublished.

[8] J. Emery, S. Hubert, and J. C. Fayet, J. Phys. (Paris) **46**, 2099 (1985); see also J. Emery, S. Hubert, and J. C. Fayet, J. Phys. (Paris) Lett. **45**, 693 (1984) and N. Aityakoub, J. Emery, S. Huberts, and J. C. Fayet, J. Phys. C **21**, 3001 (1988).

[9] P. Monceau, J. Richard, and M. Renard, Phys. Rev. Lett. **45**, 43 (1980).

[10] D. C. Ailion and R. Blinc, unpublished.

[11] M. Kogoj, S. Žumer, and R. Blinc, J. Phys. C **17**, 2415 (1984).

[12] R. Blinc, F. Milia, B. Topič, and S. Žumer, Phys. Rev. B **29**, 4173 (1984); F. Milia, R. Blinc, and S. Žumer, Solid State Commun. **50**, 1019 (1984).

[13] R. Blinc, J. Dolinšek, P. Prelovšek, and K. Hamano, Phys. Rev. Lett. **56**, 2387 (1986).

[14] S. Papavassiliou, A. Anagnostopoulos, F. Milia, R. Blinc, and S. Kotsios, Phys. Rev. B **44**, 7283 (1991).

[15] G. Papavassiliou, F. Milia, R. Blinc, and S. Žumer, Solid State Commun. **77**, 891 (1991).