Width of the critical region at incommensurate phase transitions

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Using the self-consistent phonon theory and the renormalization-group theory we have studied the critical behavior of systems with incommensurately modulated structure and especially the width of the critical region near structural phase transitions, where nonclassical critical exponents are valid. The critical region can be large if the dispersion of the soft mode is small (very ''soft'' mode) and/or if the anharmonic interaction constant is large. Taking parameters appropriate to rubidium tetrachloro zincate, rubidium tetrabromo zincate, and bis(4-chlorophenyl)sulfone we obtain results which are in good agreement with the experimental data.

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I. INTRODUCTION

In recent years the properties of structurally incommensurately (IC) modulated crystals have been frequently investigated by magnetic resonance spectroscopy. Particularly, nuclear quadrupole resonance (NQR) and quadrupolar perturbed nuclear magnetic resonance (NMR) and spin-lattice relaxation studies have been shown to be powerful tools for investigating the statics and dynamics of several compounds of the A_2BX_4 type such as rubidium tetrachloro zincate [Rb_2ZnCl_4 (RZO)], rubidium tetrabromo zincate [Rb_2ZnBr_4 (RZB)], betaine calciumchloride dihydrate [$(CH_3)_3NCH_2COO{\cdot}CaCl_2{\cdot}2H_2O$ (BCCD)], and bis(4-chlorophenyl)sulfone [$(C_6H_4Cl)_2SO_2$) (BCPS)], where transitions from a normal (N) phase to an IC phase occur. The critical behavior close to the second-order phase transition (PT) between the N and IC phases was studied experimentally by several authors.[1–9] The results of NMR studies showed that for a broad variety of crystals the critical behavior can be described by genuine nonclassical critical exponents of the appropriate three-dimensional (3D) XY model. Especially, the values obtained in various papers for the critical exponents reveal a basic argument against the applicability of the Landau theory. These values include, for instance, the critical exponents $\beta=0.35\pm0.01$ and $\overline{\beta}=0.83\pm0.03$ of the order parameter (OP) and its square, respectively, for RZC[1,6] and for RZB,[6,8] the exponent $\zeta^{\mathrm{fml}}=2\gamma-3\nu=0.625\pm0.025$ of the critical contribution T_1 to the spin-lattice relaxation time [in the fast motion limit, fml; for RCZ (Ref. 8), RZB (Refs. 6 and 7), and for BCCD (Ref. 8)], and the critical exponent $z\nu=\gamma=1.317\pm0.03$ of the OP relaxation frequency in the crystals RZC (Ref. 9) and RZB (Refs. 6 and 7). X-ray diffraction measurements resulted in the same exponents $\overline{\beta}$ for BCCD (Ref. 10) and K_2SeO_4 (Ref. 11) as mentioned above. In particular, large critical regions were observed in NMR line shape studies in the IC phase and the spin-lattice relaxation measurements in the N phase close to the N-IC PT. Note that a typical value of the width of the critical region of RZC and RZB from NMR line shape measurements is as large as $\Delta T\approx50$ K (see, e.g., Refs. 6 and 7 and further references cited herein). Diffraction synchrotron measurements[12] gave a critical region of the order of $t=|T-T_i|/T_i\approx0.1$. Specific heat measurements were reported[13] which agree with the expected critical behavior in a range up to $t\approx0.2$. Recently, Martin-Olalla *et al.*[14] have determined the asymptotic critical behavior of the function $\Delta s/Q^2$ near PT's, where Δs is the excess entropy and Q is the order parameter. The results also support the claim that IC PT's in general and the A_2BX_4 compounds in particular, in contrast with most structural PT's, have critical regions large enough to be observable.

However, other authors doubted the existence of the apparently broad critical regions[15–17] and have interpreted the NMR experiments in these compounds within a Landau approach.[18] The Ginzburg-Landau-Levanyuk criterion[19,20] states that only near to the critical temperature should the nonclassical critical exponents be valid. Fluctuations should contribute only near the critical point and change the mean-field picture.

The aim of the present paper is to study the critical behavior of IC systems such as A_2BX_4 and, especially, to evaluate the width of the critical region near structural PT's, where nonclassical critical exponents are valid. The theoretical analysis will be based both on an approach using the self-consistent phonon theory (SCPT) (in Sec. II) and on a renormalization-group (RG) analysis (in Sec. III).

With respect to the SCPT the calculations mainly refer to previous publications by Plakida and co-workers (see, e.g. Refs. 21 and 22) and Bruce and Cowley (independent mode coupling theory[23]). In these calculations the role of correlation effects will be of particular interest and it will be proved how they are determined by the interaction radius and by the strength of anharmonic interaction. The second approach is essentially based on the more recent work by Schwabl and co-workers using the renormalization-group analysis. Schorgg and Schwabl[24,25] have presented a theory for the attenuation of ultrasound close to the N-IC PT based on an order parameter model appropriate for a universality class containing the A_2BX_4 family. It was shown that the phason modes lead to a singularity of the scaling function, which describes the crossover from the critical region to the coex-

istence regime. Moreover, using an extended renormalization scheme Kaufmann *et al.*[26] have studied the critical dynamics at N-IC PT's and the crossover from the nonclassical critical behavior to the mean-field regime. Specifically, the resulting width of the critical region was investigated and compared with NMR relaxation experiments on RZC by Mischo *et al.*[4] The crossover from the nonclassical critical exponents for RZC to the mean-field ones was found[26] to take place between $\Delta T \approx 5$ and 20 K in very good agreement with the experimental NMR data. An important role in this approach plays the strength of the isotropic anharmonic interaction in the Landau-Ginzburg-Wilson free-energy functional. This finding suggests to check the influence of anharmonicity in the calculation of the nuclear spin lattice relaxation rate in more detail (Sec. III).

II. SELF-CONSISTENT PHONON THEORY

Unlike to the mean-field approximation the SCPT takes into account the critical fluctuations of the order parameter, which play an important role by the second-order PT's. That results in the renormalization of the phenomenological Landau expansion,

$$F(\eta) = F_0 + A(T)\,\eta^2 + \frac{1}{2} B\,\eta^4 + D\,\eta^6, \tag{1}$$

for the free energy, in the renormalized harmonic approximation (or SC1):

$$F = \frac{1}{\beta} \sum_{\vec{q}} \ln\left(2 \sinh \frac{\beta \Omega_{\vec{q}}}{2}\right) - \sum_{\vec{q}} \frac{\Omega_{\vec{q}}}{4} \coth \frac{\beta \Omega_{\vec{q}}}{2} + \tilde{U}(\vec{x}_i), \tag{2}$$

where $\eta_i(T)$ are some finite displacements of atoms below the critical temperature of the PT, $\Omega_{\vec{q}}$ denotes the soft mode frequency, and $\tilde{U}(\vec{x}_i)$ is the renormalized potential energy in the SC1.

For the renormalized coefficients one gets

$$A(T) = \left(\frac{dF_1}{d\eta^2}\right)_{\eta=0} = \frac{1}{2} \Omega^2_{\vec{q}_c}(\eta = 0), \tag{3}$$

$$B(T) = \left(\frac{d^2 F_1}{d(\eta^2)^2}\right)_{\eta=0} = B_0(T) + B_1(T), \tag{4}$$

where $B_0(T)$ has no features at $T \to T_C$, whereas $B(T)$ near the PT has the form

$$B_1(T) = -3 B_0 \xi(T)/[1 + \xi(T)], \tag{5}$$

with the correlation parameter

$$\xi(T) = -\frac{1}{2} V_4(\vec{q}_c) \sum_{\vec{q}} \frac{\partial}{\partial \Omega^2_{\vec{q}}} \left[\frac{1}{2\Omega_{\vec{q}}} \coth \frac{\beta \Omega_{\vec{q}}}{2}\right], \tag{6}$$

where V_4 is the renormalized quartic anharmonic interaction. $\vec{q}_c$ indicates the wave vector at the critical temperature T_C (incommensurate wave vector). The correlation length $\xi(T)$ diverges for $\tau = |T - T_C|/T_C \to 0$ due to the critical fluctuations, as the usual estimation shows:[19,20]

$$\xi(T) \approx \frac{b}{N} \sum_{\vec{q}} \frac{T}{\Omega^4_{\vec{q}}} \approx \frac{b T_C}{8\pi} \frac{a^3}{\omega_0 s^3 \sqrt{\tau}} \approx \frac{\omega_0}{\overline{\omega}} \frac{1}{8\pi \overline{R}_0^3 \sqrt{\tau}}, \tag{7}$$

with

$$b = \frac{N}{2} V_4(\vec{q}_c), \quad T_C \approx \frac{1}{b} \omega_0^2 \overline{\omega}^2.$$

Here the soft mode frequency was approximated by the equation

$$\Omega^2_{\vec{q}} = \omega_0^2 \tau + s^2 \vec{q}^2, \tag{8}$$

where here and in the following the wave vector $\vec{q}$ has to be understood as the deviation from the incommensurate wave vector $\vec{q}_c$. The dimensionless radius $\overline{R}_0 = R_0/a$ (or the effective radius $\overline{R}_0$) of the interatomic interaction was defined by the dispersion of the soft mode at $|\vec{q}| \to 0$: $s^2 = R_0^2 \overline{\omega}^2 = \overline{R}_0^2 a^2 \overline{\omega}^2$, where a is the lattice constant and $\overline{\omega}$ is the average frequency,

$$\frac{1}{\overline{\omega}^2} = \frac{1}{N} \sum_{\vec{q}} \frac{1}{s^2 \vec{q}^2} \approx \frac{1}{s^2 q_0^2}, \quad q_0 \approx \frac{2\pi}{a}, \tag{9}$$

and $\omega_0^2 = -\omega_0^2(q_c)$ is the so-called "unstable harmonic frequency:" i.e., the soft mode frequency $\Omega_{\vec{q}_c}(\eta = 0) = \Omega(q_c, T_C)$ can tend to zero at a critical temperature T_C only in that case, if the square of the phonon frequency ω_0^2 in the harmonic approximation for the soft mode frequency is negative. Hence, the lattice in the symmetrical phase in the harmonic approximation is unstable.

In the following discussion we restrict ourselves to consideration of coupled anharmonic oscillators (ϕ^4 model) with the model interaction (see Ref. 22)

$$\varphi_{\vec{q}} = \varphi_0 \exp(-R_0^2 \vec{q}^2), \tag{10}$$

where is $\varphi_{\vec{q}}$ the interaction parameter. In this model the phonon frequency is determined by the relation

$$\Omega^2_{\vec{q}} = \Delta^2 + (\varphi_0 - \varphi_{\vec{q}}), \tag{11}$$

where Δ is the width of the spectrum gap. Then one obtains for $\xi(T)$ in the case of $\Delta \to 0$

$$\xi(T) \approx \frac{3B}{N} \sum_{\vec{q}} \frac{T}{\Omega^4_{\vec{q}}} \approx \frac{3BT}{3\pi \varphi_0^{3/2}} \frac{1}{\Delta} \approx \frac{1}{8\pi \sqrt{\varphi_0/A}} \frac{1}{\overline{R}_0^3 \sqrt{\tau}}. \tag{12}$$

For the dispersion of the soft mode frequency [see Eq. (8)] we have $s^2 \vec{q}^2 = \varphi_0 - \varphi(\vec{q}) \approx \varphi_0 R_0^2 \vec{q}^2$, i.e., $s^2 \approx \varphi_0 R_0^2$ (Ref. 20), and Δ corresponds to $\omega_0^2 \tau$.

Expression (7) reveals that the correlation effects become significant in the temperature region $\tau \leqslant (\omega_0/\overline{\omega})(8\pi \overline{R}_0^3)^{-2}$, when $\xi(T) \geqslant 1$, and are mainly determined by the dimensionless interaction radius r_0 and by the relation $\omega_0/\overline{\omega}$.

Some experiments report large regions in which nonclassical exponents for the temperature dependence of the relaxation rate are observed.[5,17] Diffraction synchrotron

measurements[21] give a critical region of the order of $\varepsilon \approx 0.1$. Specific heat measurements were also reported[22] to agree with the expected critical behavior in a range up to $\varepsilon \approx 0.2$. Equation (7) shows, if $\omega_0 \approx \overline{\omega}$, then for an effective radius $\overline{R_0} \approx 1$

$$\tau \leqslant \left(\frac{1}{8\pi \overline{R}_0^3} \right)^2 \approx \left. \frac{1}{64\pi^2 \overline{R}_0^6} \right|_{\overline{R}_0 = 1} \approx 10^{-3}. \tag{13}$$

But if the dispersion is small (very ''soft'' mode), $s^2 q^2 \ll \omega_0^2$, then the critical region can be large. For $\overline{\omega} \approx 0.1\omega_0$ one gets

$$\tau \leqslant \left(10 \frac{1}{8\pi \overline{R}_0^3} \right)^2 \approx 10^2 \left. \frac{1}{64\pi^2 \overline{R}_0^6} \right|_{\overline{R}_0 = 1} \approx 10^2 \times 1.6 \times 10^{-3}$$

$$= 0.16. \tag{14}$$

In conclusion, the critical region can be large if $\omega_0^2 \gg \overline{\omega}^2$, where $\overline{\omega}^2 \approx s^2 q_0^2 \approx \varphi_0^2 R_0^2 q_0^2$, i.e., if the dispersion of the soft mode frequency is small, $s^2 q^2 \ll \omega_0^2$, or if the restoring forces in the lattice are weak, $\varphi_0 \ll \omega_0^2$.

It is worthwhile to point out that in the critical region where $\xi(T) > 1$ both the Ginzburg-Landau theory and SCPT are not applicable. However, in the former theory the critical fluctuations are ignored and the structural phase transition appears to be of second order. Contrary to that, in the SCPT the critical fluctuations are overestimated, which results in the first-order phase transition since the quartic coefficient becomes negative in the limit $T \to T_c$, $\xi \to \infty$, when $B = B_0 - 3B_0 = -2B_0 < 0$. Therefore, by applying the SCPT we can easily obtain the region of strong fluctuations given by the anomalous behavior of the order parameter, while in the Ginsburg-Landau theory the Levanyuk criterion should be used to specify the critical region. There is an exactly solvable model for structural PT's where an anharmonic interaction has an infinite range (see, e.g., Ref. 21). The SCPT gives an exact description of this model and the structural PT within this theory is of second order in spite of strong critical fluctuations at $T \to T_c$ being rigorously taken into account.[27] For example, taking into account electron-phonon interactions one obtains for the correlation length[22]

$$\xi(T) \approx \sqrt{\lambda - 1} (8\pi r \overline{R}_0^3 \sqrt{t})^{-1}, \tag{15}$$

where λ is the electron-phonon interaction constant; i.e., the critical region could be large for $\lambda \gg 1$:

$$t \leqslant (\lambda - 1) \frac{1}{64\pi^2 \overline{R}_0^6}. \tag{16}$$

Let us consider the width of the critical region ΔT at a PT from a normal to a structurally incommensurately modulated phase (at temperature T_i). IC systems such as A_2BX_4 are characterized by extremely soft modes. For example, for RZB $\overline{\omega} \approx 0.1\omega_0$ holds,[7] i.e., $t = |T - T_i|/T_i = \Delta T/T_i \approx 0.16$. With $T_i = 350$ K one obtains $\Delta T = 56$ K. In the case of RZC, where we also have $\overline{\omega} \approx 0.1\omega_0$ (Ref. 4) and where $T_i = 303$ K, we find $\Delta T = 48$ K. For BCPS it holds $\overline{\omega} \approx 0.17\omega_0$ (Ref. 28), i.e., $\Delta T = 0.06 \times 150 = 9$ K. These results are in good agreement with the experimental data, that the width of the critical region for RZB is $\Delta T \approx 50$–60 K,[7] while for BCPS the value ΔT is smaller: $\Delta T \approx 10$ K (Ref. 29).

III. RENORMALIZATION-GROUP ANALYSIS

Kaufmann *et al.*[26] have studied second-order PT's from a high-temperature N phase to a structurally IC modulated phase at the critical temperature T_i. The real-space displacement field corresponding to the one-dimensional IC modulation can be represented by its normal-mode coordinates $Q(\vec{q})$. Using $Q(\vec{q})$ as a primary order parameter of the N-IC PT in the Landau-Ginzburg-Wilson free-energy functional, the diagonalization leads to[26]

$$H[\{\psi_0^\alpha\}] = \frac{1}{2} \sum_{\alpha = \phi, A} \int_{\vec{k}} (r_0 + k^2) \psi_0^\alpha(\vec{k}) \psi_0^\alpha(-\vec{k})$$

$$+ \frac{u_0}{4!} \sum_{\alpha, \beta = \phi, A} \int_{\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4} \psi_0^\alpha(\vec{k}_1) \psi_0^\alpha(\vec{k}_2)$$

$$\times \psi_0^\beta(\vec{k}_3) \psi_0^\beta(\vec{k}_4) \delta\left(\sum_{l=1}^{4} \vec{k}_l \right), \tag{17}$$

with new Fourier coordinates $\psi_0^\phi(\vec{k})$ and $\psi_0^A(\vec{k})$ in the order parameter space. u_0 characterizes the strength of the isotropic anharmonic interaction. The parameter r_0 is proportional to the distance τ from the critical temperature τ_C and corresponds to $\omega_0^2 \tau$ in Eq. (8). Following the notation of Kaufmann *et al.*, the wave vector $\vec{k}$ indicates the deviation from the incommensurate wave vector. In Ref. 26 the dependence of the relaxation time T_1 on ΔT was only calculated for a fixed coupling constant $u_0 \propto u(1) = 0.6$. But as was already shown by Frey and Schwabl,[30] choice of the values $u(1)$ affects the form of the crossover effective critical exponent for the longitudinal static susceptibility $\gamma_{\text{eff}} = \partial \ln \chi^{-1} / \partial \ln r$. Therefore, using the extended renormalization scheme of Kaufmann *et al.*,[26] we calculate $T_1(\Delta T)$ numerically for different values of the anharmonic interaction constant $u(1)$ which is not small for the compounds of the A_2BX_4 family. We start from Eq. (4.27) in Ref. 26,

$$\frac{1}{T_1} \propto \frac{1}{\mu l} \frac{1}{\lambda} \int \tilde{k}^2 d\tilde{k} \frac{1}{\tilde{\omega}_L^2 + [\tilde{k}^2 + \tilde{\tau}(l)]^2}, \tag{18}$$

where μ, λ, and l are scale and flow parameters, respectively, $\tilde{\tau}(l)$ is a renormalized quantity describing the flow-dependent coupling, $\tilde{\omega}_L = \omega_L / \lambda \mu^2 l^2$ is the renormalized Larmor frequency, and $\tilde{k} = k/\mu l$ is the renormalized wave vector. In Fig. 1 the dependence of $T_1(\tau)$ on the physical temperature[26]

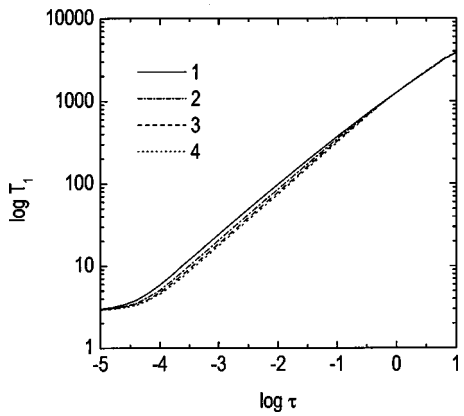

FIG. 1. Plot of the relaxation time T_1 (logarithmic base) vs the quantity $\tau \propto (T-T_i)$ (logarithmic base) for different anharmonic interaction constants $u=0.2$ (1), 0.4 (2), 0.6 (3), 0.75 (4).

$$\tau = \tau(1) \propto T - T_i = \Delta T \tag{19}$$

is shown for different values of $u=u(1)$. Very close to T_i—i.e., for very small values of τ—there is a temperature-independent region, where the OP relaxation frequency $[2\pi\tau_{OP}(0)]^{-1}=\omega_\varphi(0)$ [see Eq. (4.3) in Ref. 26 and Fig. 2 in Ref. 7] is comparable or smaller than the Larmor frequency, the so-called slow-motion case (see also Fig. 3). With increasing τ, where T_1 increases too, a temperature dependence with a nonclassical critical exponent

$$\zeta = -d(\ln T_1)/d(\ln \tau) \tag{20}$$

appears. For still higher temperatures one finds a crossover to the mean-field exponent. With increasing anharmonicity u the relaxation time T_1 decreases whereas the width of the critical region increases. In Fig. 2 the effective exponent—i.e., the slope of the curve in Fig. 1—is plotted versus the logarithm of τ. A crossover takes place from the critical behavior with nonclassical critical exponents of $\zeta=0.625$ to the mean-field regime with a value of 0.5. More clearly than in Fig. 1 it may be inferred from Fig. 2 that with increasing

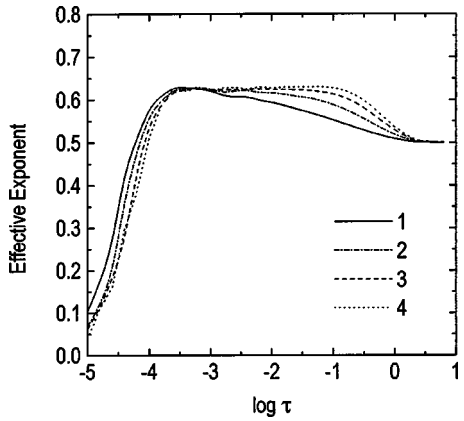

FIG. 2. Dependence of the effective exponent according to Eq. (20) on the renormalized temperature τ (logarithmic base) for different anharmonic interaction constants: $u=0.2$ (1), 0.4 (2), 0.6 (3), 0.75 (4).

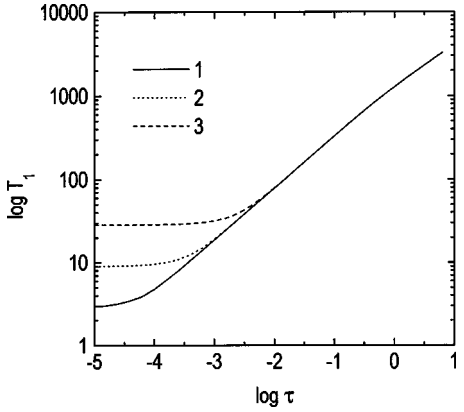

FIG. 3. Dependence of the relaxation time T_1 (logarithmic base) on the renormalized temperature τ (logarithmic base) for $u=0.6$ and for different renormalized Larmor frequencies $\tilde{\omega}_L=1$ (1), 10 (2), 100 (3).

anharmonicity the width of the critical region increases, too. For example, for $u=0.2$ we find a nonclassical critical exponent between $\ln\tau_2=-3$ and $\ln\tau_1=-3.5$. If we choose the constant in Eq. (19) according to $\tau_k=t_k=(T_k-T_i)/T_i$ ($k=1,2$) and if we take $T_i=303$ K (for RZC), we find an interval of $\Delta T\approx 0.2$ K above the crossover to the slow-motion limit, whereas for $u=0.75$ we obtain $\Delta T\approx 30$ K. This increase of ΔT with increasing constant u is in agreement with the expression (7) or (13) obtained from the self-consistent phonon theory (having in mind that u corresponds to b in the SCPT).

The $T_1(\tau)$ dependence for different Larmor frequencies $\tilde{\omega}_L$ is shown in Fig. 3. The transition from the temperature-dependent (fast-motion limit) to the temperature-independent (slow-motion limit) region may be clearly inferred. A similar $T_1(\tilde{\omega}_L)$ behavior was found by Decker *et al.*[7] on the basis of a conventional approach by the numerical simulation of the temperature dependence of the critical relaxation time $T_{1\,\mathrm{critical}}$ for various Larmor frequencies.

IV. CONCLUSION

We have studied the critical behavior of incommensurate systems such as RZC, RZB, and BCPS and especially the width of the critical region near structural phase transitions, where nonclassical critical exponents are valid, using the self-consistent phonon theory and the renormalization-group theory. The calculation reveal that the critical region may be large if the dispersion of the soft mode is small (very ''soft'' mode) or if the anharmonic interaction constant is large. Taking parameters appropriate to these compounds we find that the width of the critical region for RZB/RZC is $\Delta T\approx 30-50$ K. For BCPS a smaller critical region $\Delta T\approx 10$ K, is estimated in good agreement with the experimental data.

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