Discrete lattice effects and the phason gap of incommensurate systems

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The effect of thermal fluctuations and discreteness of the crystal lattice on the pinning of the modulation wave and the phason gap of structurally incommensurate systems is examined. Very close to the paraelectric-to-incommensurate transition temperature T_I, the phason gap is vanishingly small, but at slightly lower temperatures it takes on a nonzero value. Near the lower incommensurate-to-commensurate transition temperature T_C, where floating effects are negligible, the pinning may lead to a "devil's staircase"-like temperature dependence of the phason gap.

I. INTRODUCTION

A basic conclusion of the continuum-model theory for incommensurate (I) systems is the existence of a gapless ($\Delta_\varphi=0$) phase excitation spectrum,[1] i.e., a frictionless sliding of the modulation wave, representing the Goldstone mode which recovers the broken translational symmetry.

Detailed experimental investigations made by us and other groups[2-4] do not seem to confirm the above results even in the case of nominally pure crystals. In all incommensurate systems studied until now, a phason gap always exists. This seems to be due to two reasons: (i) In all nominally pure crystals, a small number of impurities always exist, which "pin" the modulation wave. This impurity-induced pinning destroys long-range order and, after a certain impurity concentration, "melts" the incommensurate lattice.[5] (ii) Even in the absence of impurities, the modulation wave is pinned due to the discreteness of the crystal lattice at temperatures lower than a locking temperature T_L, where $T_L < T_I$.[6,7] In the temperature region between T_I and T_L, the solitons are very thick and overlap strongly, resulting in a zero-pinning energy. The "frictionless sliding" to "locking" transition at T_L is equivalent to the appearance of a finite-pinning potential barrier which prohibits the free sliding of the modulation wave. The existence of the pinning barrier makes the soliton configuration defective in the sense that the solitons may lock at random positions. This indicates the possible existence of metastable soliton configurations.

Recent experimental and theoretical work has shown[8-11] that, close to the critical temperature T_I, pinning effects are reduced due to thermal fluctuations. What really happens is the creation of regions where the modulation wave is floating, trapped among an effective number of impurities, thus causing a time-averaged decrease of the phason gap. Indeed, Kaziba and Fayet reported EPR measurements in Rb_2ZnCl_4:Mn^{2+}, indicating a sharp decrease of Δ_φ very close to T_I.[12]

In this work, we examine the case of a one-dimensional discrete incommensurately modulated pure system, calculate the phason gap Δ_φ, and the influence of thermal fluctuations. The reason is twofold: (i) Although the predominant contribution to Δ_φ in real systems is caused by impurities, the threshold for activating floating effects should be determined by discrete lattice effects. (ii) The strength of the discrete lattice pinning is $\propto \exp(-\mathcal{d}/a_0)$, where $\mathcal{d}$ is the soliton width, and a_0 is the lattice constant.[13] This relation indicates that lattice pinning may dominate in the narrow multisoliton limit (MSL) temperature region where the thermally excited motions are smaller due to lower temperatures.

II. DISCRETE HAMILTONIAN AND THE PHASON GAP

A. Hamiltonian

Following the formalism of Bruce and Cowley[14], we consider a quasi-1D model where the displacement field depends only on the z spatial coordinate $\mathbf{u}(z)$. We assume the following: (i) The low-temperature commensurate phase locks at a wave vector τ_1/p, where τ_1 is a reciprocal-lattice vector and p is an integer. (ii) The incommensurate soft-mode vector $\mathbf{q}_S$ is close to τ_1/p. In this case, the Hamiltonian contains a kinetic-energy term, two- and four-phonon anharmonic terms, and a p-phonon Umklapp term

$$\mathcal{H}=\mathcal{H}_k+\mathcal{H}_2+\mathcal{H}_4+\mathcal{H}_p \ , \tag{1a}$$

$$\mathcal{H}_k=\tfrac{1}{2}\int_{\mathbf{q}}|\dot{Q}(\mathbf{q})|^2 \ , \tag{1b}$$

$$\mathcal{H}_2=\tfrac{1}{2}\int_{\mathbf{q}}\omega^2(\mathbf{q})|Q(\mathbf{q})|^2 \ , \tag{1c}$$

$$\mathcal{H}_4=\int_{\mathbf{q}_1}\int_{\mathbf{q}_2}\int_{\mathbf{q}_3}\int_{\mathbf{q}_4}u_4(\mathbf{q}_1\mathbf{q}_2\mathbf{q}_3\mathbf{q}_4)\times Q(\mathbf{q}_1)Q(\mathbf{q}_2)Q(\mathbf{q}_3)Q(\mathbf{q}_4)\times\Delta(\mathbf{q}_1+\mathbf{q}_2+\mathbf{q}_3+\mathbf{q}_4)\,, \quad (1d)$$

$$\mathcal{H}_p=\int_{\mathbf{q}_1}\cdots\int_{\mathbf{q}_p}v_p(\mathbf{q}_1\cdots\mathbf{q}_p)Q(\mathbf{q}_1)\cdots Q(\mathbf{q}_p)\times\Delta(\mathbf{q}_1+\cdots\mathbf{q}_p)\,. \quad (1e)$$

The following notation is used:

$$\int_{\mathbf{q}}=\sum_{\pm q_S}(a/2\pi)^d\int d^dq\,,\quad 0<|\mathbf{q}-\mathbf{q}_S|<\Lambda\,,$$

where Λ is some small cutoff and $(2\pi/a)^d$ is the volume of the Brillouin zone. In case of a 1D system, the star of the soft mode has only a double wave vector $\pm q_S$ and the soft-mode dispersion is given by the following form:

$$\omega^2(\mathbf{q})=c(T-T_I)+|\mathbf{q}-\mathbf{q}_S|^2\,. \quad (2)$$

The Δ functions express wave-vector conservation modulo a reciprocal-lattice vector $\boldsymbol{\tau}$

$$\Delta(\mathbf{q}_1+\mathbf{q}_2+\cdots)=\sum_{\tau}(2\pi)^d\delta^d(\mathbf{q}_1+\mathbf{q}_2+\cdots+\boldsymbol{\tau})\,.$$

Proceeding in exactly the same way as in Ref. 14, we get, after projecting out the noninteresting terms in real space,

$$\mathcal{H}=\mathcal{T}+\mathcal{V}\,, \quad (3a)$$

where

$$\mathcal{T}=\frac{|\mathbf{u}_0|^2}{2L}\int_0^L|\dot\phi(z)|^2dz \quad (3b)$$

and

$$\mathcal{V}=\frac{|\mathbf{u}_0|^2}{L}\int_0^L\frac{1}{2}\left[\frac{d\phi(z)}{dz}-\delta_0\right]+v(1-\cos p\phi(z))dz\,, \quad (3c)$$

$|\mathbf{u}_0|$ and ϕ are the amplitude and the phase of the incommensurately nuclear displacement

$$\mathbf{u}=|\mathbf{u}_0|\cos[\phi(z)]\,, \quad (4a)$$

$$|\mathbf{u}_0|\propto(T_I-T)^\beta\,, \quad (4b)$$

δ_0 defines the position of the soft-mode minimum at $\mathbf{q}_S=\tau_1/p+\boldsymbol{\delta}_0$, and

$$v=-(|\mathbf{u}_0|^{p-2}v_p)\,. \quad (4c)$$

In our case, we assume that τ_1 and δ_0 both lie on the z axis. This model predicts, in case of a system without impurities, the existence of a gapless phason mode.[1]

Things are totally different if we consider a discrete lattice model. The continuous Hamiltonian of relation (3a) now becomes

$$\mathcal{H}=\frac{1}{N}|\mathbf{u}_0|^2\sum_{i=1}^{N}1/2\dot\phi_i^2+\tfrac{1}{2}(\phi_{i+1}-\phi_i-\delta_0)^2+v(1-\cos p\phi_i)\,. \quad (5)$$

The ground states of the lattice are the solutions of the equations $\partial\mathcal{H}/\partial\phi_i=0$ which minimize the above Hamiltonian,[15]

$$\phi_{i+1}-\phi_i=\phi_i-\phi_{i-1}+vp\sin(p\phi_i)\quad\text{(standard map)}\,. \quad (6)$$

In Fig. 1 we see numerically calculated ground-state configurations. The ground states were found by a simultaneous slow variation of δ_0 and the initial phases ϕ_0 and ϕ_1 at constant v values, choosing the configuration with minimum energy as the ground-state configuration. Previous calculations with the same method, by keeping both v and δ_0 constant,[7,15] do not seem to apply in the case of structurally incommensurate systems due to the fact that, in a real system, the wave-vector misfit δ_0 is temperature dependent.

Stable ground-state trajectories could be produced only close to T_I for small-v values. By getting lower into the incommensurate phase — by increased v values — the trajectories were highly unstable, showing an extreme sensitivity to the initial conditions. Small initial numerical errors increase exponentially and the method proves to be inefficient in calculating ground state configurations.

The mean wave number $\mathbf{q}_S$ of the calculated trajectories, which, in the case of a discrete lattice treatment, corresponds to the winding number $\rho=(\phi_N-\phi_0)/N$, increases by decreasing temperature in contrast to Ref. 7. This is in partial agreement with experimental results in 1D modulated systems. In the case of K_2ZnCl_4 and Rb_2ZnCl_4 (Ref. 16), where $q_S\propto c^*(1-\delta_0)$, δ_0 remains initially almost constant, but on approaching T_C goes fast to zero, $\delta_0\to 0$ as $T\to T_C$. However, neutron-scattering experiments in deuterated $TMATC-Zn$[17], as well as more recent experiments in Rb_2ZnCl_4 (Ref. 18) show a nonmonotonous variation of δ_0 by decreasing temperature.

In Fig. 1 we observe that, very close to T_I, the phase consists of thick overlapping solitons and the displacement of the modulation wave can be described by a plane-wave modulation (PWM).[19] As we get lower into the incommensurate phase, things are changing dramatically and the ground-state configurations are now described by a multisoliton lattice, e.g., walls of rapid phase

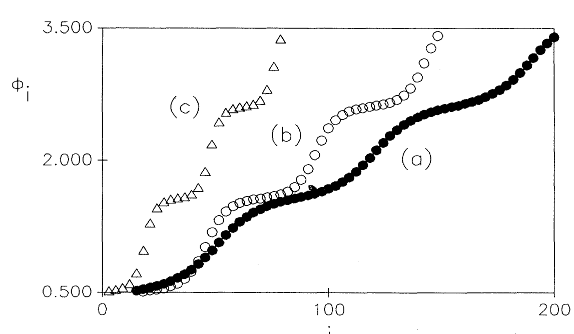

FIG. 1. Ground-state trajectories for $p=6$ and different v values: (a) $v=0.0002$, (b) $v=0.0008$, (c) $v=0.003$.

change, that separate almost commensurate regions. By further decreasing the temperature, more and more, narrower solitons are created. This result was also confirmed by experimental work.[14] We therefore conclude that the ground-state configurations can be satisfactorily described by well-separated solitons of thickness d and intersoliton distance l almost in the whole incommensurate phase. The phase ϕ varies almost linearly with the distance only very close to T_I in the PWM limit.

B. Phason gap

The basic characteristic of an incommensurate system is that the modulating wave vector is irrationally related to the underlying lattice. In the case of a 1D system modulated in the z direction, this means that $\mathbf{q}_S/\mathbf{c}^* \neq m/n$: The reciprocal lattice can be described by vectors of the form

$$\sum_{i=1}^{4} m_i \mathbf{q}_i = m_1 \mathbf{a}^* + m_2 \mathbf{b}^* + m_3 \mathbf{c}^* + m_4 \mathbf{q}_S \ , \qquad m_i \text{ is an integer} .$$

We regard these vectors as projections of vectors of a lattice in a 4D space with basis[20,21]

$$\begin{aligned} \mathscr{q}_i &= (\mathbf{q}_i \ , 0) \quad (i = 1,2,3) \ , \\ \mathscr{q}_4 &= (\mathbf{q}_S \ , \mathbf{q}_\perp) \ , \end{aligned} \tag{7}$$

where $\mathbf{q}_\perp$ is a base vector in a direction perpendicular to the 3D position space. The translational invariance, which is lost in three-dimensional space, is recovered in a four-dimensional superspace reciprocal to the one generated by $\mathscr{q}_i$ with primitive basis (1,0,0,0), (0,1,0,0), $(0,0,1,-q_S)$, (0,0,0,1).

If we take the polarization of the modulation along the b axis, the positions of the atoms in the incommensurate phase are described by

$$\mathbf{r}_i = \mathbf{n}_i + |\mathbf{u}_0| \cos(\mathbf{q}_S \mathbf{n}_{zi}) \mathbf{b} \ . \tag{8}$$

Acting on (8) with the translations of the above superlattice, we obtain a translationally invariant set of points in the four-dimensional space,

$$\mathscr{R}(\mathbf{r}_i \ , f_i) = [\mathbf{n}_i + |\mathbf{u}_0| \cos(\mathbf{q}_S \mathbf{n}_{zi} + 2\pi f_i) \mathbf{b} \ , f_i] \tag{9}$$

It is clear that "f" corresponds to the phase of the modulation wave and the positions of the ions in the real crystal are obtained from an intersection of the superspace with $f = 0$.

In case of a soliton lattice,[21] the modulation function is equal to

$$\mathbf{u} = |\mathbf{u}_0| \cos[(\tau_1/\mathbf{p}) \mathbf{n}_{zi} + \phi(\mathbf{n}_{zi})] \mathbf{b} \ , \tag{10}$$

where $\phi(z)$ describes a soliton lattice with $\phi(z + L/p) = \phi(z) + 2\pi/p$ and $L/p = d + l$. The 4D primitive basis of the position space is now equal to (1,0,0,0), (0,1,0,0), $(0,0,1,-1/p-1/L)$, (0,0,0,1).

The phase mode is connected with the translations in the "phase" direction and the corresponding group representations. The phase fluctuation at the ith lattice site is thus described by the relation

$$\delta\phi_i = \left[\frac{1}{N} \right]^{1/2} \exp[i(\mathscr{Q}_\varphi \cdot \mathscr{R}_{\varphi i} - \omega t)] \ , \tag{11}$$

where

$$\mathscr{Q}_\varphi = (0,0,0,k), \quad \mathscr{R}_{\varphi i} = (0,0,0,f_i) \ .$$

The dispersion relation of the phason branch is a solution of the equation

$$\omega_\varphi^2 = \frac{1}{|u_0|^2} \sum_{i,j} \frac{\partial^2 \mathcal{V}}{\partial\phi_i \partial\phi_j} e^{i\mathbf{k}\cdot(\mathbf{f}_i - \mathbf{f}_j)} \ , \tag{12}$$

which, with the help of Eqs. (3c) and (5), becomes

$$\omega_\varphi^2 = 2(1 - \cos k\xi) - p^2 v \mathscr{F} \ . \tag{13a}$$

Here,

$$\mathscr{F} = \lim_{N\to\infty} \frac{1}{N} \sum_{i=1}^{N} \cos p\phi_i \ , \tag{13b}$$

and ξ is the superlattice constant in the "phase" direction. The phason gap Δ_φ is defined by the lowest excitation frequency $\omega_\beta (k=0)$:

$$\Delta_\varphi^2 = -p^2 v \mathscr{F} \ . \tag{14}$$

III. FLOATING EFFECTS AND THE PHASON GAP

A. Floating effects ($T > T_L$)

As we see from Fig. 1, thick solitons seem to exist only very close to T_I. In this temperature region, the phase varies, as already mentioned above, linearly with the distance ($\phi_i = \mathbf{q}_S \cdot \mathbf{n}_{zi}$), so that $\mathscr{F}$ is taken to be $\mathscr{F} \cong 0$, resulting in a vanishing small phason gap $\Delta_\varphi \cong 0$. However, a little further into the incommensurate phase, the solitons seem to be well separated. This indicates that the "frictionless sliding" to "locking" transition temperature T_L must lie close to T_I. Taking into consideration that the phase ϕ_i between two well-separated solitons remains almost constant and equal to $\phi_i = (2\pi/p)m$ ($m = 1,2,3,\ldots$), we find that, to a good approximation,

$$\mathscr{F} \cong \frac{\ell}{d + \ell} \ . \tag{15}$$

We conclude that, very close to T_I and for temperatures $T > T_L$, $\Delta_\varphi = 0$ and the phase ϕ of the modulation wave slides freely. For $T < T_L$, there is an energy cost to translate the phase ϕ of the modulation wave by $\delta\phi$ ($\delta\phi \in [-\pi/p, \pi/p]$). This energy cost is equal to

$$\overline{\Delta\varepsilon} = \frac{1}{N} |\mathbf{u}_0|^2 v \left[\sum_i [\cos p(\phi_i + \delta\phi) - \cos p\phi_i] \right] . \tag{16}$$

We can prove that

$$\lim_{N\to\infty} \frac{1}{N} \sum_{i=1}^{N} \sin p\phi_i = 0 \ .$$

By transforming Eq. (6) to

$$\phi_{N+1}-\phi_N=\phi_1-\phi_0+vp\sum_{i=1}^{N}\sin p\phi_i$$

and taking into consideration that

$$\lim_{N\to\infty}\frac{\phi_N-\phi_1}{N}=\lim_{N\to\infty}\frac{\phi_{N-1}-\phi_0}{N}=\rho\ ,$$

we get

$$\lim_{N\to\infty}\frac{\phi_N-\phi_1}{N}-\lim_{N\to\infty}\frac{\phi_{N-1}-\phi_0}{N}=\lim_{N\to\infty}\frac{1}{N}\sum_{i=1}^{N}\sin p\phi_i=0\ ,$$

where it is known[22] that the winding number ρ is always finite for every ground-state configuration.

As a result of the above treatment, Eq. (16) can be transformed into

$$\overline{\Delta\varepsilon}=|\mathbf{u}_0|^2 v\mathcal{F}(\cos p\delta\phi-1)\ . \tag{17}$$

The potential barrier which prohibits the free sliding of the modulation wave is thus equal to

$$\bar{E}_b=-2|\mathbf{u}_0|^2 v\mathcal{F}\ . \tag{18}$$

However, close to T_L, where the intersoliton distance is very small and the solitons are still "thick," the pinning energy is, according to relations (15) and (18), small enough to be overcome by thermal fluctuations. This results in a time-averaged decrease of the phason gap.

B. Floating effects ($T<T_L$)

In the case where the thermal energy is larger than $\bar{E}_b$, the phase is sliding inside the crystal so that the normally frozen modulation wave travels "freely." After reflecting at the crystal edges, standing waves are created.[8,9]

The phase ϕ_i of the modulation wave at the ith lattice site is now time dependent and equal to

$$\phi_i(t)=\phi_{i0}+\sum_k\phi_k(\mathbf{n}_i,t)\ . \tag{19}$$

ϕ_k are the phase fluctuations with wave vector k,

$$\phi_k(\mathbf{n}_i,t)=\phi_{0k}\sin(\omega_{\varphi k}t+\alpha_k)\mathcal{I}(\mathbf{k}\cdot\mathbf{n}_i)\ , \tag{20}$$

where

$$\mathcal{I}(\mathbf{k}\cdot\mathbf{n}_i)=\sin(\mathbf{k}_x\cdot\mathbf{n}_{xi})\sin(\mathbf{k}_y\cdot\mathbf{n}_{yi})\sin(\mathbf{k}_z\cdot\mathbf{n}_{zi})\ ,$$

ϕ_{i0} is the static part of the phase of the modulation wave and α_k is an initial phase.

Assuming a random distribution of the initial phases α_k (random-phase approximation), it is possible to substitute the time average by an average over α_k,

$$(\,\bar{}\,)\longrightarrow\frac{1}{2\pi}\int_0^{2\pi}(\ )d\alpha_k\ .$$

Since $\cos p\phi_i(t)=(e^{ip\phi_i(t)}+e^{-ip\phi_i(t)})/2$, we calculate the average of $e^{\pm ip\phi_k(\mathbf{n}_i,t)}$ over α_k. This is easily proven to be equal to

$$\overline{e^{\pm ip\phi_k(\mathbf{n}_i,t)}}=J_0[p\phi_{0k}\mathcal{I}]\ , \tag{21}$$

where J_0 is the zeroth-order Bessel function. With the help of relations (19)–(21), we get

$$\cos p\phi_i(t)=\cos p\phi_{i0}\left[\prod_k J_0[p\phi_{0k}\mathcal{I}]\right]\ . \tag{22}$$

Using the equipartition theorem for the contribution of the kth mode

$$\sum_i\overline{\dot{u}_i/2}=k_BT/2\ ,$$

and relation (20), we get

$$\langle\phi_{0k}^2\rangle\propto\frac{1}{u_0^2}\ . \tag{23}$$

If we assume small phase fluctuations $\phi_{0k}\ll 1$,

$$\cos p\phi_i(t)=[1-\Omega^2]^{N_\phi}\cos p\phi_{i0}$$
$$=\exp(-N_\phi\Omega^2)\cos p\phi_{i0} \tag{24a}$$

and

$$\mathcal{F}(t)=\exp(-N_\phi\Omega^2)\mathcal{F}\ . \tag{24b}$$

Here

$$\Omega^2=\frac{\langle p^2\phi_{0k}^2\rangle}{32}\quad(\langle\mathcal{I}^2\rangle=\tfrac{1}{8})$$

and

$$N_\phi=\left[\frac{L}{2\pi}\right]^3\int_{k_{\min}}^{k_{\max}}dk^3\ .$$

According to experimental results,[16] the wave vector $\mathbf{q}_S$ and, consequently, the $\mathcal{F}$ term vary slowly with temperature except very close to T_C. Taking this into consideration and relations (4b), (14), and (24b), the phason gap can be expressed as (Fig. 2)

$$\Delta_\varphi^2=|T_I-T|^{\beta(p-2)}p^2v_p\mathcal{F}\exp[-C/(T_I-T)^{2\beta}]\ , \tag{25}$$

where, in the neighborhood of T_L, we make the following approximation:

$$\mathcal{F}=\begin{cases}0, & T_L<T\\ \text{const}(T_L-T), & T<T_L\end{cases}\ .$$

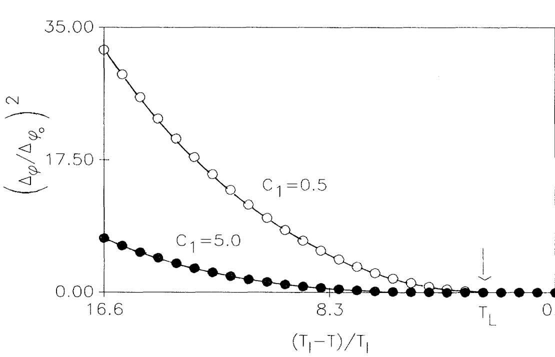

FIG. 2. The phason gap Δ_φ as a function of temperature close to the "pinning" transition temperature T_L where $\Delta_{\varphi0}=(T_I)^{\beta(p-2)}p^2u_p\mathcal{F}$ and $C_1=T_I^{-2\beta}C$.

The predictions of the above theoretical model seem to agree qualitatively with NMR and NQR experimental results on spin-lattice (T_1) and spin-spin (T_2) relaxation time measurements[23,24] very close to T_I, where a sharp decrease of both (T_1) and (T_2) and hence Δ_φ is observed. However, in the high- as well as in the intermediate-temperature range below T_L and above T_C, the phason gap seems to be dominated by impurities and lattice defects rather than by discrete lattice effects. Only close to T_C where, according to experimental results, the multisoliton lattice appears and a sharp increase of (T_1), (T_2), and Δ_φ is observed,[23,25] discrete lattice pinning effects should dominate.

C. Multisoliton limit

According to relations (13) and (14), the phason gap Δ_φ is, close to T_C, equal to

$$\Delta_\varphi^2 \cong \Delta_{\varphi c}^2 \frac{l}{\mathcal{d}+l} , \tag{26}$$

where $\Delta_{\varphi c}$ is the phason gap at T_C. As we approach T_C from above, the intersoliton distance l increases with decreasing temperature in the sense that solitons start to convert into domain walls of the low-temperature commensurate phase. The intersoliton distance is then given by relation[19]

$$l \propto -l_0 \ln(1-T_c/T) . \tag{27}$$

With the help of relations (15) and (27), Δ_φ is equal to

$$\Delta_\varphi^2 = \Delta_{\varphi c}^2 \left[1 + \frac{\mathcal{d}}{l_0 \ln(1-T_c/T)} \right] . \tag{28}$$

The predicted temperature dependence of the phason gap in the multisoliton limit is seen in Fig. 3.

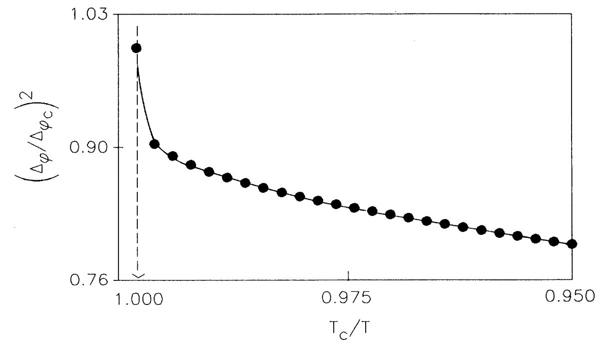

FIG. 3. The phason gap Δ_φ as a function of temperature close to the incommensurate-to-commensurate transition temperature T_c.

IV. CONCLUSIONS

The temperature dependence of the phason gap due to discrete lattice pinning in the presence of thermal fluctuations, as shown in Figs. 2 and 3, represents the main result of this study. The predicted rapid increase of Δ_φ close to the incommensurate-to-commensurate transition at T_C has been experimentally observed.[23,25] In the intermediate- and high-temperature range, discrete lattice pinning seems to be masked by impurity pinning. It might be interesting to see if discrete lattice pinning will become dominant in very pure crystals.

We also want to note that, in systems where the wave vector of the modulation wave exhibits a stepwise change as a function of temperature,[26] Δ_φ should also change in exactly the same way. In the case where solitons are narrow and far away from each other, each of them tends to be centered at a particular point of the unit cell. The variation of temperature will then induce a whole series of first-order phase transitions characterized by neighboring **q** values and the density of these phase transitions will increase as the commensurate phase is approached.[27] Moreover, Aubry[6] pointed out that the variation of l as a function of temperature constitutes, for temperature $T > T_L$, an incomplete devil's staircase and for $T < T_L$ a complete devil's staircase. In this case Δ_φ should also constitute, in the multisoliton limit, a complete devil's staircase. Recent experiments indicate that this might indeed be the case.[25,28]

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