# Explanation of the Glasslike Anomaly in the Low-Temperature Specific Heat of Incommensurate Phases

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An explanation of the glasslike anomaly observed in the low-temperature specific heat of incommensurate phases is proposed. The key point of this explanation is to properly account for the phason damping when computing the thermodynamic magnitudes. The low-temperature specific heat of the incommensurate phases is discussed within three possible scenarios for the phason dynamics: no phason gap, static phason gap, and a phason gap of dynamical origin. Existing NMR and inelastic scattering data indicate that these scenarios are possible in biphenyl, blue bronze $K_{0.30}MoO_3$, and bis (4-chlorophenyl) sulfone respectively. Estimates of the corresponding low-temperature specific heat are in reasonable agreement with the experiments.

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Deviations from the Debye law observed in the low-temperature specific heat of incommensurate (IC) phases have attracted some attention [1–4]. It is observed, in particular, a quasilinear-in-$T$ contribution to this specific heat ($C$) at the lowest temperatures ($\lesssim 1$ K) and, in some compounds [1,2,4], a maximum in $C(T)/T^3$ at temperatures $\sim$10 K. It is expected that these peculiarities are connected to the low-energy excitations specific to IC phases, i.e., the phasons. Theoretical discussions try to relate these phasons and the mentioned maximum in $C(T)/T^3$ [2,4]. As a result, this maximum is usually ascribed to the existence of a gap in the corresponding phason spectrum. The origin of the quasilinear-in-$T$ contribution, in contrast, has not been elucidated. Nevertheless, due to its similitude to that observed in glasses, this contribution inspires some speculation [4].

In this Letter we show that these peculiarities in the low-temperature specific heat of IC phases have a natural explanation. This explanation is based on a specific feature of phasons that is overlooked in the above cited works: the phason damping. In fact, it follows from general considerations that, even in perfect crystals, phasons will be overdamped for small enough wave vectors [5]. The phason damping in perfect crystals has a strong dependence on temperature and vanishes for $T = 0$ [6]. Such a temperature-dependent damping, however, is not consistent with the NMR [7,8] and the inelastic neutron scattering data [9–11] reported for the IC phases in which we are interested: the damping inferred from these experiments is 12 orders of magnitude stronger than that calculated for perfect crystals (see below). The phason damping in real crystals may then be due to defects. To the best of our knowledge, there is no theoretical calculations of such a defect-induced phason damping. Anyway, for our purposes, this damping can be estimated from the above mentioned experiments.

It is worth mentioning that, in some sense, phasons in IC phases are analogous to vortons in superconducting vortex lattices. They both represent acoustical vibrations of the corresponding IC structure that, unlike the ordinary acoustical vibrations, are overdamped for small enough wave vectors. In fact, it has already been found that vortons yield a linear-in-$T$ contribution to the low-temperature specific heat of the corresponding vortex lattice [12,13]. The key point of the theory we present below coincides with that of the mentioned works on vortex lattices: the proper accounting of the specific thermodynamic features of the damped oscillators (phasons in our case). We mention also that these specific features have been found relevant even for "ordinary" crystals: optical vibrations in real crystals remain damped due to defects down to zero temperature, so they may give an important contribution to the corresponding low-temperature specific heat [14]. In this Letter we shall concentrate on IC phases mainly because (i) the smallness of the phason frequencies increases the importance of the phason damping and, consequently, of the corresponding contribution to the specific heat and (ii) the existence of experimental data characterizing the phason dynamics makes it possible to estimate in orders of magnitude.

Let us first consider, for the sake of illustration, the simple case in which the phason spectrum shows a *static phason gap* (due to "phase pinning," see, e.g., Ref. [15]). If the corresponding wave vector dispersion is negligible, the phason branch becomes analogous to an optical one within the Einstein approximation. The corresponding contribution to the specific heat can then be computed from the specific heat of an harmonic oscillator $C_{\rm osc}$. But to describe correctly the low-temperature regime in the IC phases, it is crucial to take into account the phason damping. This can be done by using the formalism of Refs. [12,13,16]. As a result one obtains

$$C_{\rm osc} = k_B\left[\sum_{i=1}^{3}\left(\frac{\lambda_i}{\nu}\right)^2\psi'\left(\frac{\lambda_i}{\nu}\right) - \left(\frac{\omega_D}{\nu}\right)^2\psi'\left(\frac{\omega_D}{\nu}\right) - 1\right], \quad (1)$$

 

where $\nu = 2\pi k_B T/\hbar$, $\psi'(x) = d^2[\ln\Gamma(x)]/dx^2$ is the trigamma function, and the $\lambda$'s are the roots of the equation $\lambda^3 + \omega_D\lambda^2 + (\omega_0^2 + \gamma\omega_D)\lambda + \omega_D\omega_0^2 = 0$, with $\omega_0$ the natural frequency of the oscillator and $\gamma$ the damping coefficient, i.e., the viscosity coefficient divided by the mass of the oscillator. Here $\omega_D^{-1}$ is a memory time associated with a Drude regularization of this viscosity damping (see, e.g., Ref. [16] and accompanying paper [17]). In further considerations the limit $\omega_D \to \infty$ is taken. In this limit, the regularized viscosity damping reduces to the ordinary one.

The dependence on temperature of the specific heat in Eq. (1) of a damped harmonic oscillator can be seen in Fig. 1 for different values of the damping coefficient $\gamma$. In the undamped case ($\gamma = 0$), it is well known that this specific heat is exponentially small at low enough temperatures ($k_B T \ll \hbar\omega_0$). For any finite damping, however, the low-temperature asymptotic for the specific heat Eq. (1) is linear in $T$:

$$C_{\rm osc} \underset{T\to 0}{\approx} \frac{\pi k_B}{3}\frac{\gamma}{\omega_0}\frac{k_B T}{\hbar\omega_0}. \tag{2}$$

In an overdamped case ($\gamma \gg \omega_0$) this expression is valid for $k_B T \ll \hbar\omega_0^2/\gamma$ while in an underdamped one ($\gamma \ll \omega_0$) it is for $k_B T \ll \hbar\omega_0$.

It is worth noticing the qualitative similitude, already at this level of consideration, between this specific heat and that reported in IC phases [1–4]. Of particular interest is what concerns the linear-in-$T$ dependence at the lowest temperatures, which is connected to the damping as we have seen. In the following we shall make more concrete considerations.

Let us then consider the Landau thermodynamic potential (see, e.g., Ref. [6,10])

$$\phi = \phi_0 + \frac{a}{2}(\eta_1^2 + \eta_2^2) + \frac{b}{4}(\eta_1^2 + \eta_2^2)^2 + \frac{c}{2}[(\nabla\eta_1)^2 + (\nabla\eta_2)^2], \tag{3}$$

where $\eta_1$ and $\eta_2$ are the real and imaginary part of the complex order parameter $\eta = (\eta_1, \eta_2)$. In the IC phase ($a < 0$), the equilibrium values can be taken such that $\eta_1^{(\rm eq)} = (-a/b)^{1/2}$ and $\eta_2^{(\rm eq)} = 0$. Within the *scheme of no phason gap*, which seems to be valid for biphenyl (see below), for small deviations of the order-parameter components from their corresponding equilibrium values: $\eta_i = \eta_i^{(\rm eq)} + \eta_i'$, one has the following equations of motion (see, e.g., Ref. [6]):

$$m\ddot{\eta}_1' + \gamma_\eta\dot{\eta}_1 - 2a\eta_1' - c\nabla^2\eta_1' = 0, \tag{4a}$$

$$m\ddot{\eta}_2' + \gamma_\eta\dot{\eta}_2 - c\nabla^2\eta_2' = 0. \tag{4b}$$

Hence $\eta_1$ is associated with longitudinal (amplitude) fluctuations while $\eta_2$ does with transverse (phase) ones.

In Eq. (4b) it is implicit that, as stated above, for small enough wave vectors phasons are overdamped due to the viscosity term $\gamma_\eta\dot{\eta}_2$. At low enough temperatures the phason contribution to the specific heat will be linear in $T$. From Eq. (2), this contribution can be estimated as

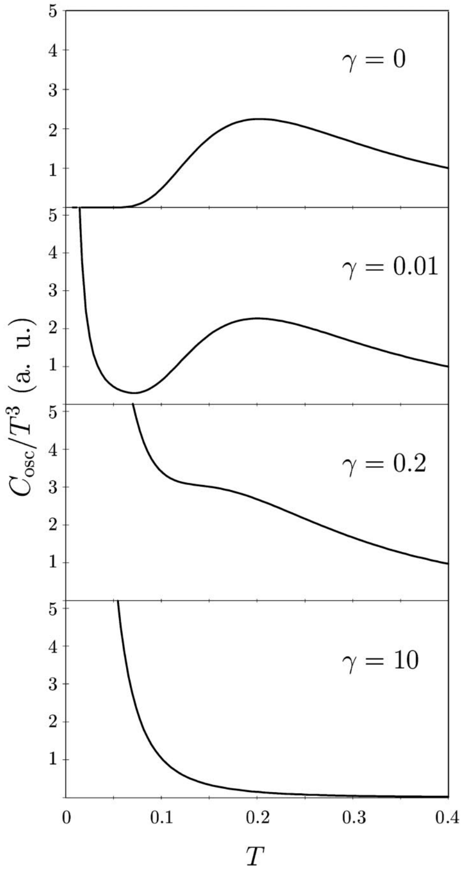


FIG. 1. Specific heat from Eq. (1) divided by $T^3$ of a harmonic oscillator for different dampings. Here units have been chosen such that $k_B = \hbar = 1$, the natural frequency of the oscillator as $\omega_0 = 1$ and the Drude frequency [16,17] $\omega_D = 10^4$. A different scale in $C_{\rm osc}/T^3$ is used in the plot for $\gamma = 10$.

$$C \approx \frac{k_m^3}{6\pi}\frac{k_B^2 T}{\hbar(v^2k_m^2/\gamma)} \tag{5}$$

for $k_B T/\hbar \ll \min(vk_m, v^2k_m^2/\gamma)$, where $v^2 = c/m$, $\gamma = \gamma_\eta/m$ and $k_m$ represents the radius of the Brillouin zone. (Here it has been assumed that the damping is the same for the whole phason branch.) This linear-in-$T$ contribution to the low-temperature specific heat of IC phases due to phasons will prevails over the Debye one (due to acoustic phonons) at temperatures $T \lesssim [\gamma/(vk_m)]\theta$, where $\theta = (\hbar/k_B)(V^3/v)^{1/2}k_m$ with $V$ the velocity of sound.

Let us mention that, in accordance with Eq. (4a), amplitude fluctuations of the order parameter (amplitudons) are also damped. In consequence, they also give a linear-in-$T$ contribution to the specific heat at the lowest temperatures. However, this contribution is always smaller than the phason one because, for a given wave vector, the amplitudon frequency is always greater than the phason one, the corresponding damping coefficient being similar in both cases [see Eqs. (4) and (2)]. Indeed the amplitudon contribution to the low-temperature specific heat of the IC phases of interest is not essential to describe the corresponding experimental data, so we shall omit this contribution if further considerations.

Let us now consider the *scheme of a dynamic phason gap* [10] (see also accompanying paper [17] for further details). In some compounds such as bis (4-chlorophenyl) sulfone (BCPS) the coefficients of the equations of motion for the order parameter may have some frequency dispersion. A relatively simple dispersion is described by the phenomenological model developed in Ref. [18] to reproduce the central peak in the high-temperature phase of strontium titanate. Such a dispersion can be understood as a result of the linear coupling between the order parameter and a relaxational variable $\xi = (\xi_1, \xi_2)$ [10,19]. The phason dynamics is then governed by the following equations of motion:

$$m\ddot{\eta}_2' + d^2\eta_2' - c\nabla^2\eta_2' + d\xi_2' = 0, \tag{6a}$$

$$\gamma_\xi \dot{\xi}_2' + \xi_2' + d\eta_2' = 0. \tag{6b}$$

Let us emphasize that the most important part of these equations is not the extra variable $\xi_2$ by itself, but the frequency dispersion that this variable yields in the phason dynamics. Strictly speaking, to reproduce the specific features of, e.g., BCPS, i.e., gapped phasons and a central peak, it is only this dispersion that would be necessary (see below). However, to calculate the corresponding phason contribution to the specific heat, it is convenient to bear in mind that such a dispersion is obtainable from Eqs. (6) (see accompanying paper [17] for further details). Anyway, the resulting phason dynamic is different at different time scales: while the slow oscillations are overdamped (central peak), the rapid ones are not (gapped phasons) [19]. The phason contribution to the low-temperature specific heat then naturally splits into two terms: $C = C_{\rm gph} + C_{\rm cp}$ (see accompanying paper [17] for further details). The former,

$$C_{\rm gph} = k_B \int \frac{d^3k}{(2\pi)^3}\left(\frac{\hbar\omega_2(k)}{2k_BT}\right)^2\left[\coth^2\left(\frac{\hbar\omega_2(k)}{2k_BT}\right) - 1\right]$$
$$= k_B \frac{k_m^3}{2\pi^2}\left(\frac{T}{\Theta}\right)^3 \int_{x_0(T)}^{\Theta/T} dx \frac{[x^2 - x_0^2(T)]^{1/2}x^3e^x}{(e^x - 1)^2}, \tag{7}$$

is the contribution that can be ascribed to the (undamped) gapped phasons [20]. Here $\omega_2^2(k) = \delta^2 + v^2k^2$, $\delta^2 = d^2/m$, $x_0(T) = \hbar\delta/(k_BT)$ and, assuming that $\delta \ll vk_m$, $\Theta \simeq \hbar v k_m/k_B$. The latter contribution,

$$C_{\rm cp} = k_B \int \frac{d^3k}{(2\pi)^3}\left[\left(\frac{\hbar\omega_{\rm cp}(k)}{2\pi k_BT}\right)^2 \psi'\left(\frac{\hbar\omega_{\rm cp}(k)}{2\pi k_BT}\right) - \left(\frac{\hbar\omega_{\rm cp}(k)}{2\pi k_BT}\right) - \frac{1}{2}\right], \tag{8}$$

is due to damped phasons (central peak). Here $\omega_{\rm cp}^2(k) = v^2k^2/(\delta^2\gamma_\xi/m)$. The low-$T$ asymptotic of this latter contribution is given by Eq. (5) by replacing $\gamma_\eta \to \gamma_\xi\delta^2$.

We are now in a position to estimate the phason contribution to the low-temperature specific heat in the IC phases of biphenyl and BCPS, and in the charge-density-wave system blue bronze $K_{0.30}MoO_3$:

(i) *Biphenyl.* In accordance with NMR [7] and inelastic neutron scattering data [9], the scheme of no phason gap seems to be valid here (the corresponding gap should be $\lesssim 10$ MHz at least). From neutron scattering, the phason damping can be inferred such that $\gamma \sim 50$ GHz. Notice that such a damping, obtained at $T = 3$ K, is 12 orders of magnitude stronger than the one expected for a perfect crystal [6]. So defects must be the cause of the real damping, although inducing no (observable) phason gap. Both phason and sound velocities can be estimated as $\sim 10^5$ cm s$^{-1}$ [3,9,21]. Therefore, the linear-in-$T$ contribution due to phasons will prevail over the Debye one at temperatures $\lesssim 1$K. In addition, according to Eq. (5), it is obtained $C/T \sim 10$–$10^2$ erg cm$^{-3}$ K$^{-2}$ for these low temperatures. The experimental value $\sim 0.5$ erg cm$^{-3}$ K$^{-2}$ [3] is smaller in this case, what shall be commented below.

(ii) *BCPS (dynamic phason gap).* From NMR and inelastic neutron scattering experiments [8,10,11], the corresponding damping can be inferred such that $\gamma_\xi\delta^2 \sim 10$ GHz. Notice that no temperature dependence is observed in this damping down to $T = 19$ K, so it seems very probable that it remains unaltered down to very low temperatures (as in biphenyl). According to Refs. [2,21], both phason and sound velocities are also $\sim 10^5$ cm s$^{-1}$. Then, the linear-in-$T$ contribution due to damped phasons is expected to be the leading one at temperatures $\lesssim$ 1K. It would be such that $C/T \sim 10$–$10^2$ erg cm$^{-3}$ K$^{-2}$, which agrees in order of magnitude with what was observed experimentally [2]. In this case (dynamic phason gap), in addition to this linear-in-$T$ contribution there is a contribution due to undamped gapped phasons [Eq. (7)]. The phason gap can be estimated as $\delta \sim 100$ GHz [10], so the maximum in $C/T^3$ observed at $\sim$1–2K can then be explained as a result of this latter contribution [20]. This has already noticed in Ref. [2].

(iii) *Blue bronze* $K_{0.30}MoO_3$. Here, as well as in other charge-density-wave systems (see, e.g., Ref. [22] and the references therein), the scheme of a static phason gap seems to be the most appropriate one. In accordance with inelastic scattering data [23] this gap is $\sim$200 GHz, and the phason damping can be estimated as $\gamma \sim 800$ GHz. Both phason [24] and sound [1,4] velocities can be inferred as $\sim 3 \times 10^5$ cm s$^{-1}$. Consequently, a linear-in-$T$ contribution with $C/T \sim 10$–$10^2$ erg cm$^{-3}$ K$^{-2}$ is expected due to the damping of the phasons (prevailing over the Debye one at temperatures $\lesssim 1$ K) and, in addition, a maximum in $C/T^3$ at $T \simeq 10$ K due to the phason gap. This qualitatively describes the experimental observations [1,4]. We mention that in Ref. [25] the explanation that the maximum in $C/T^3$ can be ascribed to gapped phasons is questioned: it is shown that a similar maximum arises due to the acoustic anisotropy of this system. In Ref. [26] it is pointed out that, however, acoustic anisotropy cannot explain why this maximum is so sensitive to the crystal quality. It is also worth mentioning that, given the relatively large anisotropy in the phason velocity [24], phasons can behave as overdamped oscillations along certain directions. Bearing in mind that an overdamped phason branch in the one-dimensional case gives rise to a contribution $\sim T^{1/2}$ in the low-temperature specific heat [13]; this could explain the deviation of the linear-in-$T$ behavior observed experimentally in Ref. [4] at the lowest temperatures.

It is worth mentioning that, at present, the above contributions to the low-temperature specific heat due to the damping of the phason can only be estimated in order of magnitude. It is true that the corresponding phason dynamics has been extensively studied in a number of papers. However, the quantitative comparison of our results with the experimental data would require, in particular, the knowledge of the phason damping for the whole phason branch. Unfortunately, these latter data are not reported in the literature and consequently, when carrying out the comparison of our results with experimental data, we are forced to make some assumptions—for instance that the phason damping is the same for the whole phason branch. These types of assumptions, at the present unavoidable, could be the reason for the overestimation of the linear-in-$T$ contribution due the phason damping when compared with experimental observations. More precise estimates require more complete experimental data (see Ref. [14] for a detailed discussion). Nevertheless, the agreement in order of magnitude we have obtained indicates that the deviations from the Debye law observed in the very low-temperature specific heat of IC phases are, most probably, in regard to the phason damping. We mention also that the resulting linear-in-$T$ behavior is expected as a low-temperature asymptotic limit. It is well possible in the above mentioned experiments that this limit has not been fully achieved but rather are in an intermediate (crossover) region, which could be the reason for the different power laws observed there.

In summary, we have shown that the glasslike anomaly observed in the low-temperature specific heat of incommensurate phases can be explained as a result of the phason damping. Three possible scenarios for the phason dynamic, reproducing the corresponding NMR and inelastic scattering data for biphenyl, BCPS, and blue bronze $K_{0.30}MoO_3$, are discussed. Estimates of the corresponding low-temperature specific heat are in reasonable agreement with those observed experimentally.

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