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# Ultra-low-temperature anomalies in heat capacities of metals caused by charge-density waves

M. L. Boriack and A. W. Overhauser
*Department of Physics, Purdue University, West Lafayette, Indiana 47907*
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A new fruitful area for low-temperature research is discussed. The contribution of phase excitations of incommensurate charge-density waves to the low-temperature heat capacities of metals is calculated, and the characteristic signature of phasons in calorimetric measurements is illustrated. The importance of anisotropy in the phason dispersion relation to the magnitude of the phason heat capacity and to the temperature at which the signature of the phasons can be seen is emphasized. Unless measurements are done at sufficiently low temperatures to *freeze-out* the phasons, it is possible to make significant errors in determining the electronic heat capacity. In particular the possibility of detecting phasons in potassium is discussed.

## I. INTRODUCTION

The electronic ground state of a metal need not be the mere occupation of the $N$ lowest-energy Bloch states. Exchange interactions and electron-electron correlations can cause a modulated, collective deformation of the electronic charge density to have a lower total energy.[1] For such a charge-density-wave (CDW) ground state the conduction-electron density is

$$\rho = \rho_0[1 + p\cos(\vec{Q}\cdot\vec{r} + \varphi)], \tag{1}$$

where $\rho_0$ is the average density and $p$ is the fractional modulation. The CDW wave vector $\vec{Q}$ is very nearly equal to the diameter $2k_F$ of the Fermi surface. The phase $\varphi$ will be discussed at length below.

A CDW instability can occur only if the electronic charge density is locally neutralized by an accompanying lattice distortion.[1] Each positive ion will be displaced from its equilibrium lattice site $\vec{L}$ by

$$\vec{u}(\vec{L}) = \vec{A}\sin(\vec{Q}\cdot\vec{L} + \varphi). \tag{2}$$

Since $\vec{Q}$ is controlled by Fermi surface dimensions, the wavelength of a CDW will generally be unrelated to lattice periodicities, i.e., the CDW is incommensurate with the lattice. In this case, the resulting structure is multiply periodic, no two ions are equivalent, and the crystal no longer has a translation group. Such a loss in symmetry means that the energy of the system is independent of phase $\varphi$. It then follows that there will be low-frequency collective excitations corresponding to $\varphi$ varying slowly in space and time. These elementary excitations are called phasons[2,3] and have important consequences for experiments that try to detect a CDW with diffraction[2] or through Knight-shift or hyperfine-field effects.[3]

The purpose of this paper is to discuss the contribution of phasons to the low-temperature heat capacity of metals containing incommensurate CDW's. We illustrate the type of anomalies which occur in plots of $C/T$ vs $T^2$, where $C$ is the total heat capacity. Such anomalies have recently been seen by Sawada and Satoh[4] and led to the identification of CDW's in La-Ge systems. It is expected that the more isotropic the metal, the more anisotropic the phason dispersion relation.[3] We demonstrate that this anisotropy can have profound effects on the size of the phason anomaly and on the temperature at which the anomaly is observed. As an example we discuss the possibility of a phason contribution to the heat capacity of potassium in particular. The search for this type of anomaly in metals provides a new area for ultra-low-temperature research.

## II. PHASON HEAT CAPACITY

Consider an incommensurate CDW described by Eqs. (1) and (2). Since the energy of the system is independent of phase $\varphi$, it follows that there will be low-frequency collective excitations corresponding to $\varphi$ varying slowly in space and time.[2,3] We express $\varphi(\vec{L}, t)$ by an expansion in running waves

$$\varphi(\mathrm{L}, t) = \sum_{\vec{q}} \varphi_{\vec{q}} \sin(\mathrm{q}\cdot\mathrm{L} - \omega_{\vec{q}} t). \tag{3}$$

This approach is analogous to treating lattice dynamics in the continuum approximation. The wave vectors $\{\vec{q}\}$ are assumed small compared to the Brillouin zone, and $\{\varphi_{\vec{q}}\}$ are the amplitudes of the phasons. If $\vec{q}\perp\vec{Q}$ the local direction of the CDW vector is slightly rotated. If $\vec{q}\parallel\vec{Q}$ the local magnitude of $\vec{Q}$ is periodically modulated.

The dynamics of these modes have been studied in the small-$q$ limit, assuming negligible damping.[2,3] The phason frequencies vary linearly with $q$. The frequency spectrum is plotted schematically in Fig. 1. Phasons are a type of lattice vibration and are linear combinations of old phonons (in a crystal with no CDW) having

 

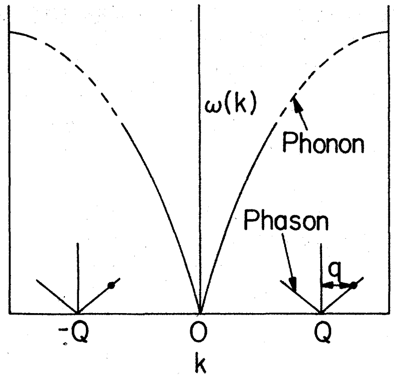


FIG. 1. Schematic illustration of the vibrational modes of a metal having an incommensurate CDW structure. The frequency of the phason branch goes to zero at $\pm\vec{Q}$, the location of CDW satellite reflections in $k$ space. Such a diagram has only approximate meaning since an incommensurate CDW structure does not have a Brillouin Zone.

wave vectors near $\vec{Q}$ and $-\vec{Q}$. That the phason frequency goes to zero at $\pm\vec{Q}$ is a consequence of the incommensurate nature of the CDW. Since there can be only a given number of vibrational normal modes, phasons and phonons must share the spectral density in the regions near $\pm\vec{Q}$ as is indicated by the dashed region of the phonon dispersion curve in Fig. 1.

It is expected that $\omega$ vs $\vec{q}$ will be highly anisotropic because a local rotation of $\vec{Q}$ requires less energy than a change in its magnitude. Consequently, the surfaces of constant frequency for the phasons will be flat (pancake-shaped) ellipsoids. Taking $\vec{Q}$ in the $z$ direction and letting $q_z$ and $q_\perp = (q_x^2 + q_y^2)^{1/2}$ be components of $\vec{q}$ parallel and perpendicular to $\vec{Q}$, respectively, we can write

$$\omega_{\vec{q}}^2 = c_z^2 q_z^2 + c_\perp^2 q_\perp^2 . \tag{4}$$

The ratio of $c_z$ to $c_\perp$ may be as high as 100 to 1, especially if the Fermi surface is nearly spherical. (For the ideal metal "jellium," which has no preferred direction, the ratio would be $\infty$.) For more anisotropic materials the ratio of $c_z$ to $c_\perp$ may be much smaller.

Since phasons are (a special type of) lattice vibrations and their frequency varies linearly with $q$, it is natural to evaluate the phason heat capacity using a Debye model. For phasons the parameters of this model are the radius $q_\Phi$ of the phason ("Debye") sphere in $k$ space, the speed $c_z$ of phasons with $\vec{q} \parallel \vec{Q}$, the anisotropy factor $\eta \equiv c_\perp / c_z$. Because of the anisotropy, definition of the phason Debye temperature is somewhat arbitrary. We choose to define the phason Debye temperature in terms of the velocity parallel to $\vec{Q}$, i.e.,

$$\Theta_\Phi \equiv \hbar c_z q_\Phi / k_B , \tag{5}$$

where $k_B$ is Boltzmann's constant. Thus the free parameters are $q_z$, $c_z$, and $\eta$.

The total energy $E$ of the phasons is evaluated by summing the contributions $\hbar\omega_{\vec{q}}$ of each normal mode weighted by the Bose-Einstein occupation factor. For a sample with volume $\Omega$,

$$E = \frac{\Omega}{(2\pi)^3} \int_0^{2\pi} d\varphi \int_{-q_\Phi}^{q_\Phi} dq_z \int_0^{(q_\Phi^2 - q_z^2)^{1/2}} dq_\perp \times q_\perp \hbar \omega_{\vec{q}} (e^{\hbar \omega_{\vec{q}} / k_B T} - 1)^{-1} . \tag{6}$$

In Eq. (6) a choice of cylindrical coordinates was made.

Note that for phasons there is only one "branch" with the angle between $\vec{q}$ and $\vec{Q}$ varying continuously from 0° to 180°. Thus the factor of 3 present for phonons is absent for phasons.

Changing the variables of integration to dimensionless units, the phason heat capacity at constant volume $C_\Phi = (\partial E / \partial T)_v$ is given by

$$C_\Phi = \frac{\Omega q_\Phi^3 k_B T^3}{2\pi^2 \Theta_\Phi^3 \eta^2} \int_0^{\Theta_\Phi / T} dz \int_0^{\rho_{\max}} d\rho\, \rho r^2 \times [(e^r - 1)(e^{-r} - 1)]^{-1} , \tag{7}$$

where $r^2 \equiv z^2 + \rho^2$ and

$$\rho_{\max} = \eta (\Theta_\Phi / T) [1 - (Tz/\Theta_\Phi)^2]^{1/2} . \tag{8}$$

It is interesting to evaluate (7) in the high- and low-temperature limits. For high temperatures $\Theta_\Phi / T \ll 1$, and (since $\rho, z, r \ll 1$) we can expand the exponentials to lowest order in $r$. Elementary integrations yield the high temperature Dulong-Petit result $C_\Phi = N_\Phi k_B$, where $N_\Phi$ is the number of phason modes contained by the phason "Debye" sphere, i.e., $N_\Phi = \Omega q_\Phi^3 / 6\pi^2$.

To evaluate the phason heat capacity in the $T \to 0$ limit it is convenient to express Eq. (6) in ellipsoidal coordinates: $q_z = (r/c_z)\cos\theta$; $q_x = (r/c_\perp)\sin\theta\cos\varphi$; $q_y = (r/c_\perp)\sin\theta\sin\varphi$. A change to dimensionless variables then obtains (in the limit $T \to 0$) the usual integral which arises in the Debye theory. It follows that in the low-temperature limit

$$C_\Phi = (4\pi^4 N_\Phi k_B / 5\eta^2)(T/\Theta_\Phi)^3 . \tag{9}$$

It is important to note the presence of $\eta^2$ in the denominator. This factor is not surprising since through $\Theta_\Phi$ three powers of the phason velocity appear in the denominator of $C_\Phi$. Thus, $C_\Phi \sim c_z^{-1} c_\perp^{-2}$. The anisotropy of the phason velocity and the magnitude of the velocity have a strong effect on the size of $C_\Phi$ at low temperatures. In Sec. III we discuss the additional and profound effect of $\eta$ on the temperature at which the presence of phasons can be identified in measurements of the total heat capacity. Both these effects of the phason anisotropy are extremely important to the question of whether phasons can be identified in low-temperature calorimetric measurements on a metal with an incommensurate CDW ground state.

### III. TOTAL HEAT CAPACITY

In this section we discuss the "signature" of the phasons in a measurement of the total heat capacity. The effects we describe have been seen recently by Sawada and Satoh[4] in a study of heat capacity anomalies in La-Ge systems and analyzed by them in terms of phasons. In their analysis, Sawada and Satoh took the phason velocity to be isotropic. Although in an anisotropic metal the phason dispersion relation is expected to be more isotropic than in an isotropic metal, it is likely that their fit to the experimental data could be made even better with this additional parameter.

For our discussion, however, we shall deal with the possibility of detecting the presence of the proposed CDW in potassium[2] by measurements of the low-temperature heat capacity. In this case, phason anisotropy has crucial significance, as will be shown below. The CDW hypothesis has been successful in quantitatively explaining the anomalous optical absorption,[5] the anisotropic conduction-electron $g$ factor,[6] and the anisotropic residual resistivity[7] of potassium. Although these results and many others[2,8] present convincing evidence that potassium has a CDW structure, a persistent question is: Where are the CDW diffraction satellites? Actually, a careful search for them has never been reported. It is likely, however, that the phason Debye-Waller factor reduces the satellite intensity to a point where the satellites would be extremely difficult to find.[2,3] Total intensity is not lost, but is transferred into a pancake-shaped phason cloud. Special techniques that integrate over the diffuse pancake may be required to detect CDW's in potassium in diffraction experiments. The existence of phasons also offers an explanation of the "null" result of attempts to detect a CDW in potassium with Knight shift and hyperfine-field effects.[3] Consequently, it is of great interest to investigate whether or not phasons might make their presence known in calorimetric measurements.

To estimate the phason specific heat it is necessary to make reasonable guesses for the three parameters mentioned previously: the anisotropy factor $\eta$; the phason-sphere ("Debye") radius $q_\Phi$; and the velocity $c_z$ of phasons with $\vec{q}$ parallel to $\vec{Q}$. To illustrate the importance of $\eta$, we shall use the two values $\eta=0.1$ and $\eta=0.01$ in our calculations. That is, we discuss phasons with $\vec{q}$ perpendicular to $\vec{Q}$ 10 and 100 times softer than those with $\vec{q}$ parallel to $\vec{Q}$.

Because the number of phason modes $N_\Phi$ and, therefore, the magnitude of the phason contribution varies with $q_\Phi^3$, the size of this parameter is of crucial importance. For our discussion we *guess* the radius $q_\Phi$ of the phason sphere to be approximately 5% of the phonon Debye radius. The value of $\vec{Q}$ suggested for potassium is very close in $k$ space to the [110] reciprocal-lattice vector. Taking half the difference between $Q$ and the [110] reciprocal lattice vector gives the above estimate for $q_\Phi \sim 4.6\times10^6\ \mathrm{cm}^{-1}$. The ratio of the volume of phason and phonon Debye spheres is $1.3\times10^{-4}$ with this estimate.[9] This compares with the Sawada and Satoh experimental result of $8\times10^{-5}$ in La-Ge.[4]

The phason velocity $c_z$ (for $\vec{q}$ parallel to $\vec{Q}$) also appears with the third power in the expression for the low-temperature phason heat capacity. Since phasons are a special type of lattice vibration, it might be guessed that phason-vibration velocities are of the same order as phonon velocities.

It is possible to arrive at a rough lower limit for $c_z$ given the choices of $q_\Phi$ and $\eta$ above. As will be discussed in detail below, heat-capacity measurements at temperatures above the low-temperature limit for the phasons can lead to errors in the experimentally determined thermal effective mass of the electrons. By requiring that this error be less than, say, 30% of the true thermal effective mass for $\eta=0.01$, a lower limit on the phason velocity of $c_z \cong 2.7\times10^5$ cm/sec is obtained. To illustrate the effect of phason velocity we shall compute the phason heat capacity using $c_z=2.7\times10^5$ cm/sec and twice this value.

Neglecting phasons for the moment, at low temperatures the heat capacity of a metal is the sum of electronic and lattice contributions. The heat capacity is given by

$$C=\gamma T+AT^3\,, \tag{10}$$

where $\gamma T$ is the electronic contribution and $AT^3$ is the lattice heat capacity. A plot of $C/T$ vs $T^2$ is a straight line. The intercept at $T=0$ gives $\gamma$ and the thermal effective mass $m_t^*$ defined by $m_t^*/m_0=\gamma/\gamma_0$, where $m_0$ is the free-electron mass and $\gamma_0$ is the free-electron value. The slope of the straight line determines the Debye temperature. A CDW leads[1] to a 5%–10% enhancement in $\gamma$, which cannot be distinguished experimentally

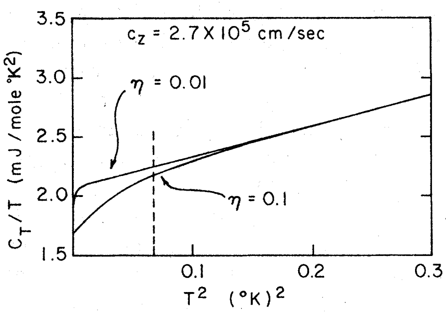


FIG. 2. Total heat capacity $C_T$ including phasons for potassium plotted as $C_T/T$ vs $T^2$. $c_z$ is the velocity of phasons with wave vector $\vec{q}$ parallel to the CDW wave vector $\vec{Q}$. $\eta$ gives the anisotropy of the phason velocity. See text for details.

from band-structure, phonon-interaction, or many-body effects.

The heat capacity of potassium has been measured between 0.26 and 4.2°K by Lien and Phillips.[10] They found the heat capacity was well fit by the expression

$$C = 2.08\ T + 2.57\ T^3 \tag{11}$$

with units of mJ/(mol K).

If phasons are present, the total heat capacity $C_T$ is given by the sum of (7) and (10) less $C_E$,

$$C_T = \gamma T + AT^3 + C_\Phi - C_E\ . \tag{12}$$

Here $C_E$ is an "Einstein" specific heat subtracted from the phonon specific heat to keep the total number of normal modes equal to $3N$. These phonons correspond to the dashed region of Fig. 1, where the phonons have lost spectral density at the expense of the phasons. For potassium the Einstein temperature of these phonons is ~20°K and the contribution of $C_E$ is negligible in the temperature range of interest in this work.

The most convenient way to illustrate the "signature" of phasons in the total heat capacity

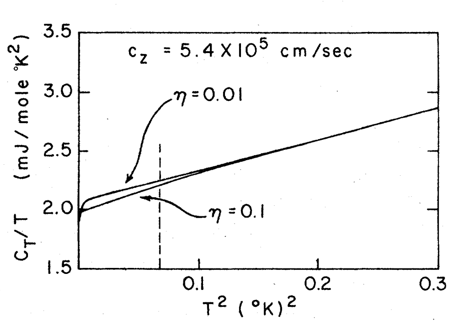


FIG. 3. $C_T/T$ vs $T^2$ with the phason velocity twice that in Fig. 2.

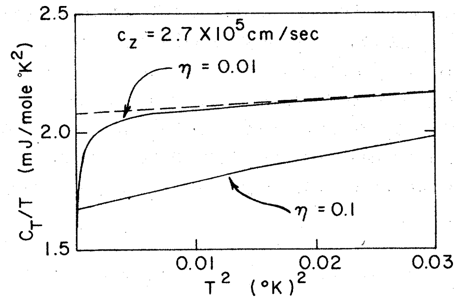


FIG. 4. Low-temperature range of Fig. 2.

is to adjust $\gamma$ and $A$ in (12) so that $C_T$ agrees with the measured heat capacity, say, from $T^2 = 0.2$ to 0.4°K². The curves of $C_T/T$ vs $T^2$ in Figs. 2–5 have been plotted in this way. It should be noted that for $\eta = 0.01$ the value of $A$ (or, equivalently, the Debye temperature) needed to fit the measured data differs insignificantly from the value found by Lien and Phillips. For $\eta = 0.1$ the change in the Debye temperature needed to fit the experimental data is $\lesssim 2\%$. Thus, phasons have little effect on the slope of the line in this region. As will be discussed in detail below, it was necessary to change $\gamma$ significantly from the value found by Lien and Phillips.

Plots of $C_T/T$ vs $T^2$ are shown in Fig. 2 for $c_z = 2.7 \times 10^5$ cm/sec and $\eta = 0.1, 0.01$. The dashed vertical line indicates the lowest temperatures, $T \sim 0.26$°K, measured by Lien and Phillips. The signature of the phasons is the departure of the curve from a straight line as $T$ decreases.[11] For the lowest temperatures, the curve is once again a straight line with a slope determined by the sum of the phonon and phason contributions. The curve with $\eta = 0.1$ can be seen to depart significantly from a straight line by $T^2 = 0.07$°K². (For smaller $q_\Phi$ this departure would be less). The curve for $\eta = 0.01$ does not depart significantly from a straight line until $T^2 = .005$°K² or $T \cong 70$ mK.

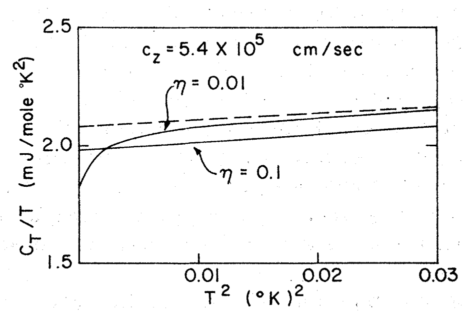


FIG. 5. Low-temperature range of Fig. 3.

Similar results for $c_z = 5.4\times10^5$ cm/sec are shown in Fig. 3. Note that although the curve for $\eta = 0.1$ does fall below the straight line, the curvature is much less than for the curves in Fig. 2 since the phason Debye temperature is greater. Such a curve could be difficult to distinguish experimentally from a straight line.

Figures 4 and 5 are expanded plots of the low temperature parts of Figs. 2 and 3, respectively. In Fig. 4 the curve for $\eta = 0.01$ indicates that the heat capacity of the phasons does not reach $T^3$ behavior until $T \leq 20$ mK in this case. Comparison of the straight-line portions for $\eta = 0.01$ and $\eta = 0.1$ clearly indicates the effect of phason anisotropy on the slope of the line, i.e., the magnitude of the phason contribution to the low-temperature heat capacity. It is also evident that phason anisotropy is extremely important in determining where the $C/T$ vs $T^2$ curve departs from the higher temperature straight-line behavior. Where this "turn down" occurs is extremely important. Although the Dulong-Petit contribution of the phasons is small and would be easily missed in comparison with the electron and phonon contributions at higher temperatures, the anisotropy of the phasons allows the soft phasons to fill up at much lower temperatures where $C_\Phi$ is sizeable in comparison to the other contributions. Thus, for $c_z = 2.7\times10^5$ cm/sec and $\eta = 0.01$, at $T = 0.07$°K the phason heat capacity is ~25% of the total, whereas for $c_z = 5.4\times10^5$ cm/sec and $\eta = 0.1$, the phason contribution might be missed entirely. This point is crucial for experimental measurements of the phason heat capacity.

The dashed line in Figs. 4 and 5 is the extrapolation of the measurements of Lien and Phillips. The intercept at zero temperatures gives $\gamma = 2.08$ mJ/(mol K$^2$). It is evident from the curves in Figs. 4 and 5 that the presence of phasons can lead to a significant error in the determination of $\gamma$. Unless calorimetric measurements are performed at temperatures sufficiently low so that the phason heat capacity is in the $T^3$ regime, extrapolation of the $C/T$ vs $T^2$ curve to zero temperature can lead to a fictitious and erroneous apparent contribution to $\gamma$. In other words, it is necessary to freeze-out the phasons before the electronic heat capacity can be identified with certainty. As discussed above in arriving at a rough lower limit to the phason-velocity, for $\eta = 0.01$ and $c_z = 2.7\times10^5$ cm/sec the error in $\gamma$ is ~30%. Equivalently, for this case the thermal effective electron mass $m_t^*$ would be ~30% too large. From the rest of the curves it can be seen how the "fictitious" $\gamma$ varies with phason anisotropy and speed.

The possibility that the existence of phasons can lead to an erroneously large thermal effective mass is very interesting in light of the controversy surrounding the theoretical value of $m_t^*$. It is convenient theoretically to write $m_t^*$ in terms of the free electron mass $m_0$ and various correction terms so that

$$m_t^* = m_0(1 + \delta_b + \delta_{ep} + \delta_{ee} + \delta_{e\Phi} + \delta_f)\,, \tag{13}$$

where $\delta_b$ arises from energy-band effects, (including that of the CDW), $\delta_{ep}$ is the enhancement due to electron-phonon interaction, $\delta_{e\Phi}$ is the analogous (and as yet undetermined) enhancement arising from electron-phason interactions, $\delta_{ee}$ arises from electron-electron interactions, and $\delta_f$ is the "fictitious" contribution from the phasons discussed above. Of course, if the phasons are observed in the $T^3$ regime it is possible to extrapolate correctly to zero temperature and $\delta_f = 0$. Unless the phasons are frozen out, $\delta_f \neq 0$.

The contribution to $m_t^*$ subject to the most disagreement is $\delta_{ee}$. For an ideal electron gas the deviation of the density-of-states effective mass $m^*$ at the Fermi surface from $m$ is not large, but this is caused by a cancellation of inherently much larger effects.[12] Unfortunately, there is disagreement even as to the sign of $\delta_{ee}$. Rice[13] has shown that Hubbard's theory[14] leads to $m^* > m$ by about 10% for typical metallic densities. In contrast, several other calculations[15] find $m^* < m$ by a few percent. Clearly, for meaningful comparison between experimental and theoretical values of $m_t^*$, it is essential that the fictitious phason contribution be known to be zero.

Heat capacity measurements of incommensurate CDW systems at ultralow temperature are, therefore, important for two reasons. First, such measurements can yield important evidence of the existence and properties of phasons. Second, by freezing-out the phasons the true electronic contribution to the heat capacity can be measured.

In this study we have assumed that the phason spectrum is that of a pure metal. It is possible for phason distortions of a CDW to be pinned at impurities, and this could alter slightly the phason frequency spectrum. This effect is unlikely for the case of potassium, since the root-mean-square phase fluctuation is estimated to be several radians, even at very low temperatures.[3] A pinned CDW would lead to broadening of the nuclear magnetic resonance, and this has been found to be absent in potassium.[16]

## IV. CONCLUSION

We have discussed the phason contribution to the low-temperature heat capacity of metals containing incommensurate CDW's and illustrated the characteristic signature of phasons in measurements of the total heat capacity. By way of example, we have discussed the possible contribution of phasons to the heat capacity of potassium. It is important to note that the parameters used for the phasons were guessed. Anisotropy of the phasons is extremely important in determining both the magnitude of the low-temperature phason heat capacity and the temperature at which a plot of $C/T$ vs $T^2$ departs significantly from a straight line. If heat-capacity measurements are not done at sufficiently low temperatures so that the phasons are frozen out, the electronic heat capacity can be significantly overestimated.

It is likely that a search for phasons in potassium must be performed at temperatures significantly below 0.1°K. These measurements would be difficult, due to the small value of the heat capacity at these temperatures.

Throughout the calculation presented above damping of the phasons has been neglected. This damping has recently been calculated.[17] For potassium phasons with $\vec{q}$ parallel to $\vec{Q}$ are strongly attenuated and nearly critically damped. However, phasons with $\vec{q}$ perpendicular to $\vec{Q}$ are only very weakly attenuated. The effects of damping on phason heat capacity have been investigated. The differences between the finite-damping case and the zero-damping case discussed above are not substantial enough to warrant a separate discussion at this time. Since the characteristic signature of phasons is primarily determined by those phasons with $\vec{q}$ nearly perpendicular to $\vec{Q}$ and these phasons are only very slightly damped, it is reasonable that the effects of phason damping on the heat capacity should be relatively small.


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