ly because of their isotropic properties, but because of the time and length scales over which most of the decomposition process takes place.

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## Evidence of Phasons in the Specific Heat of Potassium

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(Received 20 July 1981)

The low-temperature specific heat of potassium has been remeasured and evidence for the existence of phasons is found. The fraction of phonons converted to phasons is $2\times10^{-5}$ and the Debye temperature of these phasons is 6 K.

PACS numbers: 65.40.-f

Traditionally the low-temperature specific heat of a pure, normal metal has been analyzed using the equation

$$C_0=\gamma T+\alpha T^3+\beta T^5. \quad (1)$$

Recently Overhauser and his collaborators studied the effect of a charge-density wave (CDW) on the specific heat of alkali metals.[1,2] The existence of a CDW in simple metals would have important ramifications for many-electron theory, as has been discussed in several publications.[2-5] Therefore it seems important to test the theoretical predictions for the case of a fundamental property such as the specific heat.

A CDW is a broken-symmetry state, accompanied by a lattice distortion. The new collective excitations are low-frequency modes called phasons.[6] Phasons are linear combinations of "old phonons" and produce an extra contribution to the specific heat, $\Delta C_{\mathrm{CDW}}$, to be added to $C_0$. Using this concept Giuliani and Overhauser[2] analyzed the specific heat of rubidium and found a low-temperature anomaly similar to that expected for a CDW structure. However, Taylor *et al.*[7] showed that for Rb Eq. (1) is valid only below 0.4 K. The observed anomaly could be explained by the dispersion of the slow shear acoustic mode. They based their calculation on a lattice-dynamics model that fit the phonon spectrum determined from inelastic neutron scattering. They carried out similar calculations for potassium and found that Eq. (1) should be valid below 3 K. Since a CDW heat-capacity anomaly in potassium is estimated to appear between 0.5 and 2 K, its appearance would be less ambiguous. Prior measurements[8,9] in this temperature range gave signs of experimental difficulty.

We have considered carefully the different sources of error in the specific heat, and these will be discussed elsewhere in more detail.[10] The temperature scale, and also the agreement of the thermometer calibration with the accepted scale, are extremely important. For example, just the change from the $T_{62}$ to the $T_{76}$ scale re-

 

duces the specific heat at 1 K by 1% and at 2 K by 1.2%. We used a nitrogen filled germanium thermometer calibrated by Lake Shore Cryotronics, Inc. between 0.1 and 6 K, but checked this calibration against the National Bureau of Standard's Superconductive Fixed Point Device SMR 767 (corrected to $T_{76}$). Small deviations were found, so the supplied calibration was adjusted smoothly.

With this thermometer we measured the specific heat of a standard copper sample, designated T4.2, kindly given to us by Argonne National Laboratory. A least-squares fit of the specific heat for this sample gave $C_{LS} = 0.68T + 0.048T^3$ mJ/mole K. The two coefficients differ slightly from previous values, but this is a consequence of the change from $T_{62}$ to $T_{76}$. Figure 1 shows the relative deviation from the least-squares fit: $\Delta C/C \equiv (C - C_{LS})/C$. Above 0.6 K the deviations are less than 1%.

The addenda correction for the potassium sample was kept small by using a thin-walled calorimeter made from 2-mil copper foil. Five copper wires, placed inside, were soldered to the bottom for better heat distribution. The heat capacity of a nearly identical, empty, calorimeter was measured together with the thermometer, heater wire, and a small amount of General Electric varnish. The addenda correction was less than 4% of the heat capacity of potassium. The calorimeter was filled under argon with 53 g of potassium by Callery Chemical Company, who also supplied a spectrographic report. All impurities, with the exception of three nonmagnetic ones, were less than 10 ppm by weight and so have only a negligible contribution to the specific heat.

We measured the specific heat of potassium between 0.5 and 5 K, using one calorimeter and compared them with the results of Lien and Phillips.[8] Above 1.2 K the two sets of results are in excellent agreement but below this temperature our results are lower: 1.2% at 1.2 and 10% at 0.5 K. Lien and Phillips[8] employed two calorimeters, one for temperatures above 1.2 K and another for the lower temperature range. They remark that in the overlapping temperature range the results obtained with the low-temperature calorimeter are 1.3% above those of the other. Our results are in excellent agreement with the results of Filby and Martin[9] between 0.9 and 1.5 K, but fall below their results at lower temperatures.

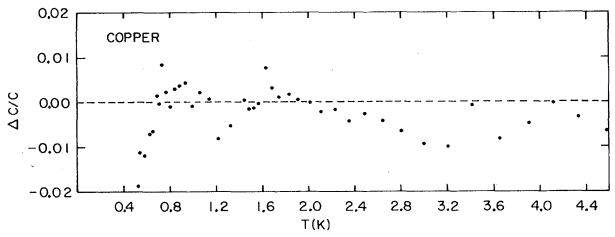


FIG. 1. Relative deviation of the specific heat of pure copper from least-squares fit; $C_{LS} = 0.68\,T + 0.048\,T^3$ mJ/mol K.

In order to allow for possible phason contributions (of a few percent of the total heat capacity) we analyzed the data as discussed below. On treating the copper data in this way we found the same parameters as those obtained from the least-squares fit given above.

We define $y \equiv (C - \gamma T - \beta T^5)/T^3$. If a CDW does not exist, $y$ is equal to $\alpha$, the coefficient of the cubic term in the specific heat, and should be constant up to 3 K (if $\gamma$ and $\beta$ are chosen correctly).[7] However, if a CDW is present, $y$ should be equal to $\alpha$ at higher temperatures, where the extra phason contribution becomes negligible.[2] At lower temperatures, $y$ should increase and at the very lowest temperatures become constant again, but at a higher value than $\alpha$, since the extra phason contribution also becomes proportional to $T^3$.

From the higher temperature we estimated $\beta$ to be 0.051 mJ/mole K$^6$, which agrees with previous experimental results,[8] but is 20% higher than the *a priori* calculation by Taylor *et al.*[7] With this value of $\beta$, we calculated $y$ for many values of $\gamma$, and show, in Fig. 2, $y$ vs $T^2$ for three values of $\gamma$. We choose $\gamma$ so that at the lowest temperatures $y$ becomes constant again; accordingly,

$$C_0 = 1.83T + 2.66T^3 + 0.051T^5 \text{ mJ/mole K}. \quad (2)$$

The coefficients of the linear and cubic terms differ substantially from previous results, which is partly a consequence of the lower specific heat found below 1 K. In addition, if for our results below 3 K only a least-squares analysis is used, ignoring extra phason contributions, $\gamma$ increases by 8%, $\alpha$ decreases by 1%, while $\beta$ stays the same. The experimental coefficients of the cubic term, as well as of the $T^5$ term already discussed above, are both higher than the theoretical values.[7] Although theory reproduces the neutron scattering data very well, it apparently does not reproduce the acoustic range accurately.

In Fig. 3 we show $(C - C_0)/C_0$; a maximum is observed at $T = 0.75$ K, which leads to a maximum

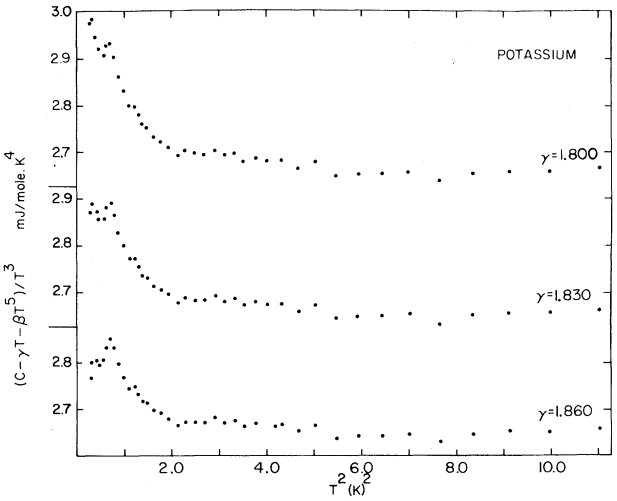


FIG. 2. $(C-\gamma T-\beta T^5)/T^3$ vs $T^2$ of potassium for three values of the constant $\gamma$ in mJ/mol $K^2$; $\beta = 0.051$ mJ/mol $K^6$.

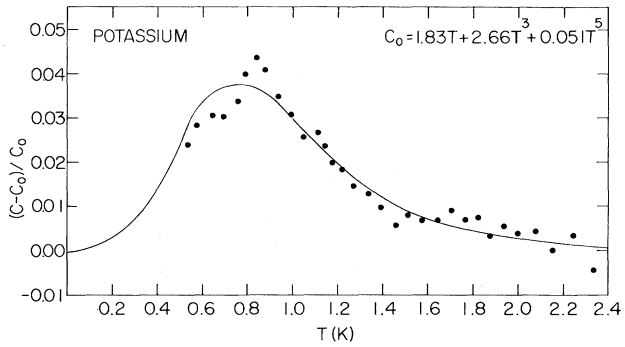


FIG. 3. Relative deviations of the experimental results from $C_0$. The smooth curve is the theoretical heat-capacity anomaly caused by the CDW structure, taken from Ref. 2, with the parameters $\Theta_{CDW} = 6$ K, and the fraction of phonons converted to phasons of $2 \times 10^{-5}$.

in $\Delta C$ vs $T$ at 1.08 K and a phason Debye temperature $\Theta_{CDW} = 6$ K. The maximum phason contribution is $(4 \pm 1)\%$, which corresponds to a fraction of the phonons converted to phasons of $2 \times 10^{-5}$. The solid curve is taken from Ref. 2 and uses the parameters given above. Agreement with the data is surprisingly good. The value found here for $\Theta_{CDW}$ is higher than that (3.25 K) estimated from the low-temperature resistivity.[4] This is not unexpected since the resistivity caused by electron-phason scattering involves the matrix element for electron-phason scattering (which is expected to decrease at higher phason frequencies). The fraction of phonon modes converted to phasons was estimated from the theoretical curve (Fig. 1) of Ref. 2. We obtained a value $2 \times 10^{-5}$, which is approximately the same fraction needed to fit the low-temperature resistivity.[4]

In conclusion we find strong experimental evidence for the existence of phasons in the low-temperature specific heat of potassium.

We are grateful to Dr. S. Nagata for his preliminary work on this research. Also, we thank Professor A. W. Overhauser and Dr. G. F. Giuliani for their insistent encouragement to undertake this research, and Professor N. J. Giordano for his constructive criticism. This work was supported by the National Science Foundation Grant No. DMR-79-08356A1.

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