EXPERIMENTAL DETECTION OF AN ANOMALY IN THE SPECIFIC HEAT OF A METAL WITH HEAVY IMPURITY ATOMS

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Submitted to JETP editor March 25, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) **49**, 456-458 (August, 1965)

An anomaly in the behavior of the specific heat, predicted in general form by Kagan and Iosilevskii[2] for crystals containing heavy impurity atoms, was detected experimentally in the alloy MgPb (2.8 at.% Pb). The specific heat was measured in the temperature range 1.2—20°K using an adiabatic calorimeter. The results obtained were in good agreement with the theory.

KAGAN and Iosilevskiĭ[1] established that the vibration spectrum of an isolated heavy impurity atom of mass m′ in an ideal lattice consisting of atoms of mass m, where $m'/m \gg 1$, was strongly resonant, being localized near the frequency

$$\omega_0 = \omega_{max}(k|1 - m'/m|)^{-1/2}.$$

In this expression, ω_{max} is the limiting frequency of the phonon spectrum of an ideal lattice, and k is a coefficient which depends on the actual form of the phonon spectrum and is equal to 3 in the Debye model. Such a deformation of the phonon spectrum affects considerably the thermodynamic and, probably, the transport properties of an alloy.

As shown in[2], the localization of the spectral vibrations of impurity atoms in the region of low frequencies leads to a strong rise in the low-temperature specific heat of an alloy even when the impurity atom concentration is relatively low. Kagan and Iosilevskiĭ associated this with the physically self-evident consideration that the transition to the classical limit occurs much earlier in the frequency region from $\omega = 0$ to $\omega \approx \omega_0$ (which is characteristic of the impurity atom vibrations), than in the main part of the phonon spectrum of the matrix.

The present communication reports the results of an experimental discovery of the anomalous behavior of the specific heat of a MgPb alloy (2.8 at.% Pb).[1)] The measurements were carried out on polycrystalline samples of pure Mg and of the alloy, which was a substitutional solid solution with a hexagonal lattice similar in its parameters to the Mg lattice. The chemical composition and uniformity of the alloy were checked by x-ray diffraction and chemical analyses. It should be mentioned that this alloy had a sufficiently high mass ratio $m'/m = 8.6$ and a relatively low value of the Debye temperature of the matrix ($\Theta_{Mg} = 311$°K), which made it possible to observe the specific heat anomaly in the temperature range 1.2—20°K—convenient from the measurement point of view.

The measurements were carried out using an adiabatic calorimeter. Cylindrical samples, 15 mm in diameter and 50 mm long, were suspended in a vacuum jacket by thin capron filaments. Each sample was heated by a manganin heater, wound bifilarly on the lateral surface of the sample. A carbon resistor, made by the Speer Resistor Company, was used as a thermometer. Good thermal contact between the thermometer and a sample was ensured by soldering the thermometer to a copper foil glued to the ends of the sample. The thermometer was calibrated using the magnetic susceptibility of chromium—potassium alum for the region from 1 to 1.5°K, He^4 vapor pressure from 1.5 to 4.2°K, and a standard germanium resistance thermometer (prepared and calibrated at the All-Union Scientific Research Institute for Physico-technical and Radiotechnical Measurements) for the range from 4.2 to 20°K. Repeated calibrations of our thermometer in the range from 1.2 to 4.2°K over two years showed that it was highly stable: the calibration of the thermometer was fully reproduced, within the experimental error, in the region 1.2—2.8°K and the maximum discrepancy at 4.2°K was 0.02 deg.

To determine temperature, we used an interpolation formula with two constants:

[1)] Using the same alloy, Zemlyanov and Chernoplekov were the first to detect experimentally a strong deformation in the vibrational spectrum and the appearance of a quasilocal level, by investigating the inelastic scattering of neutrons. Preliminary data on the behavior of the specific heat of a similar system have been reported also by Leman et al.

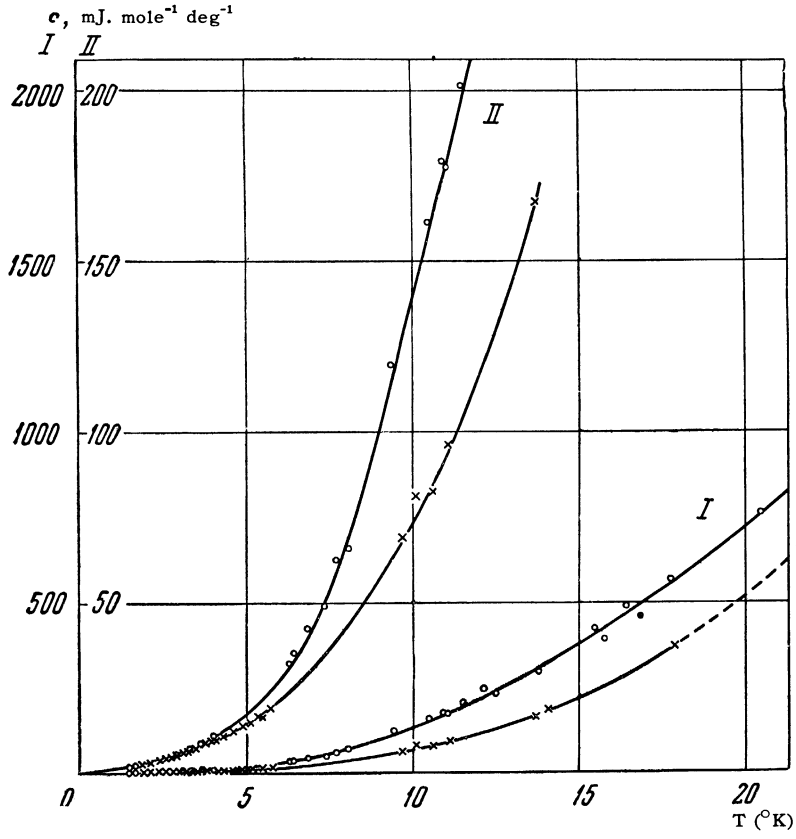

FIG. 1. Temperature dependence of the specific heat of Mg and the alloy MgPb (2.8 at.% Pb): × – Mg; o – MgPb.

$$(\log R/T)^{1/2} = a + b\log R, \quad a = -7.999, \quad b = 2.904.$$

Figure 1 shows the temperature dependence of the specific heat of Mg and MgPb obtained in our experiments. The electronic specific heat coefficients and the Debye temperatures calculated from these data were, respectively:
$\gamma = 1.31$ mJ-mole^{-1} deg^{-2}, $\Theta = 311 \pm 10$°K for Mg and $\gamma = 1.11$ mJ-mole^{-1} deg^{-2} for MgPb (γ and Θ were obtained by analyzing the experimental results by the least-squares method).

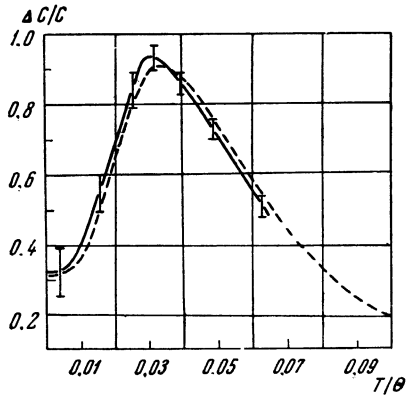

FIG. 2. Experimental and theoretical curves for the quantity $\Delta C/C$ as a function of T/Θ for the alloy MgPb (2.8 at.% Pb): the continuous curve is experimental; the dashed curve is theoretical.

Figure 2 shows the smoothed-out experimental curve which gives $\Delta C/C$ as a function of the ratio T/Θ (ΔC is the difference between the phonon specific heats of the alloy and the matrix, C is the phonon specific heat of the matrix), and includes a dashed theoretical curve, calculated using Eq. (2) given in [2]. Extrapolation of the experimental data to T = 0 gives $\Delta C/C \approx 0.32$. As the temperature is raised, the value of $\Delta C/C$ increases, passes through a maximum $\Delta C/C \approx 0.93$ at $T_{max} \approx 10.6$°K, and then rapidly decreases to 0.5 at 20°K. The theory of Kagan and Iosilevskiĭ[2] gives, at T = 0, the expression $\Delta C/C = (3/2)|\epsilon|\eta$ (where η is the concentration of impurity atoms, $\epsilon = |1 - m'/m|$) which gives $\Delta C/C \approx 0.31$ at T =0. Direct calculations give $T_{max} = 3.5 \times 10^{-2}\Theta = 10.9$°K and the corresponding value $\Delta C/C = 0.91$, i.e., the measured and calculated values are very close. It is evident from Fig. 2 that, in general, the theoretical curve agrees well with the experimental curve over the whole range of temperatures.

Thus, our investigations of the specific heat of the alloy MgPb confirmed experimentally the Kagan and Iosilevskiĭ effect. Small amounts of a heavy impurity in a light-atom lattice alter very greatly the specific heat of the matrix lattice. In our experiment, 2.8 at.% Pb doubled the specific heat in the region of 10.6°K.

In conclusion, the authors thank Yu. M. Kagan and N. A. Chernoplekov for valuable discussions and constant interest in this work; M. N. Andriyanov, V. A. Zinov'ev, V. I. Kutaĭtsev, and N. T. Chebotarev for the preparation and analysis of the samples; M. P. Orlova and D. N. Astrov for supplying the calibrated semiconducting thermometer; V. E. Keĭlin for his help in constructing the apparatus; and V. G. Ovchinnikov for his help in the experiments.

[1] Yu. Kagan and Ya. Iosilevskiĭ, JETP **45**, 819 (1963), Soviet Phys. JETP **18**, 562 (1964).

[2] Yu. Kagan and Ya. Iosilevskiĭ, JETP **42**, 259 (1962), Soviet Phys. JETP **15**, 182 (1962).

Translated by A. Tybulewicz
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