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Size Dependence of the Transport Properties of Bismuth in the Phonon-Drag Region

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The low-temperature size effects in the phonon-drag thermopower and electrical and thermal conductivities of bismuth, measured under identical experimental conditions on a tuning-fork sample of arm thicknesses 2.8 and 1.2 mm, are reported. The thin arm has the highest positive thermopower and a plateau is observed for both thicknesses in a different temperature range. It is shown that the size effect in the thermopower and electrical conductivity occurs in the same temperature range, suggesting that the nonequilibrium phonon distribution should be taken into account in discussing the electrical conductivity at low temperatures. The thermal conductivity exhibits a size effect at temperatures higher than expected.

Throughout the period since the discovery of the peculiar size dependence of the electrical conductivity of bismuth at 4.2 K,[1] its explanation has remained puzzling. The general feature observed[2,3] is first a decrease of conductivity with thickness, then a leveling off, and then another decrease with thickness. Following the observation of phonon-drag humps in the thermopower of bismuth at low temperatures,[4] one of the present authors (J.-P. I.) suggested that both phenomena might be related.[5] The electrons dragging the phonons would cause a departure of the phonon distribution from equilibrium, leading to an enhancement of the electrical conductivity. In the limit, the effective electron mean free path (mfp) would become equal to that of the much larger low-energy-phonon mfp, and the reported effect could then be an indirect observation of a phonon size effect.[2,3] The aim of the present paper is to report observations of the size effect in the electrical conductivity and in the phonon-drag thermopower measured both on the same single crystal and under identical experimental conditions.

The sample of 99.9999% purity and dimensions $42.5\times 6\times 3$ mm^3 was kindly supplied by Koenig and Brown.[6] A single spark cut 36 mm deep along the length transformed it into a tuning fork of arm thicknesses 2.8 and 1.2 mm, respectively, the width being 3 mm. The sample was then etched to remove surface damage and mounted in a specially designed liquid-helium cryostat. Two symmetrical thermal and electrical circuits allowed us to compare the desired transport properties for both arms simultaneously. The heat flux or electrical current was along the sample axis oriented in the binary direction. The temperature sensors were Au(Fe) p-chromel thermocouples, and emf's from these and from voltage probes were measured by means of a Leeds and Northrup K-5 potentiometer and a sensitive null detector. A resolution of better than 5×10^{-9} V was achieved.

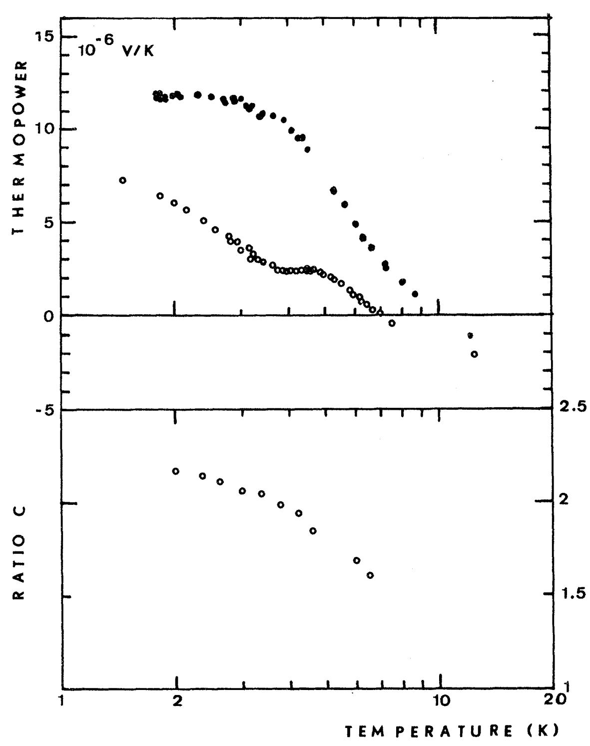

FIG. 1. Temperature variation of the size effect in the absolute thermopower compared to that in the electrical conductivity. The two upper curves (left-hand scale) are relative to the thermopowers of the thick (open circles) and thin (solid circles) arms of the tuning fork. The lower curve (open circles, right-hand scale) represents the conductivity ratios $c=\sigma(2.8)/\sigma(1.2)$ in the same temperature range.

In Fig. 1 the absolute thermopowers of the thick and thin samples are represented versus temperature. As previously observed by Korenblit *et al.*,[4] the thermopowers are positive in the lowest-temperature range. However, these authors found for a sample of circular cross section of diameter nearly 2.5 mm a maximum around 4 K. In our thin sample which has the highest positive thermopower, the latter increases with decreasing temperature down to about 3 K and then reaches an almost constant value down to 1.6 K. For the thickest sample the thermopower has smaller positive values in the range investigated. It first increases with decreasing temperature, levels off around 4 K, and then increases again with decreasing temperature.

Korenblit's theory[4] expresses the partial dragging thermal emf due to carriers of group s in the form

$$s_g=\frac{1}{e^s N^s (kT)^2}\sum_{n=1}^{3}\int_{\Omega_s}\frac{d^3q\, e^{\hbar\omega_q^n/kT}\,\hbar\omega_q^n}{(2\pi\hbar)^3\,(e^{\hbar\omega_q^n/kT}-1)^2}\times\frac{\tau^n(q)}{\tau^{n,s}(q)}\,v_l^n q_k\,,$$

where e^s and N^s are the charge and concentration of the carrier group s, n is the phonon-acoustic-branch polarization number, v_l^n is the lth component of the group velocity of these phonons, $\tau^{n,s}$ is the relaxation time of these phonons with carriers of group s, and τ^n is the total relaxation time of these phonons including that for boundary scattering. The integration is carried out over the region Ω_s occupied by the phonons interacting with carriers of group s. These partial thermopowers, which are highly anisotropic, are combined together to give the total phonon-drag thermopower and, depending upon the relative contribution of each group s and the corresponding mfp of interacting phonons, an increase in the positive value of the thermopower with decreasing thickness can be physically understood. Besides, with the adjustment of some critical parameters, such plateaus as we observed can be qualitatively explained,[4] and further experimental work is now in progress to interpret them quantitatively.

The lower part of Fig. 1 allows a direct comparison between the size effect in the thermopower and in the electrical conductivity σ to be made. It represents the conductivity ratios $c=\sigma(2.8)/\sigma(1.2)$ for both arms versus temperature in the same temperature range, showing an increase in c with de-

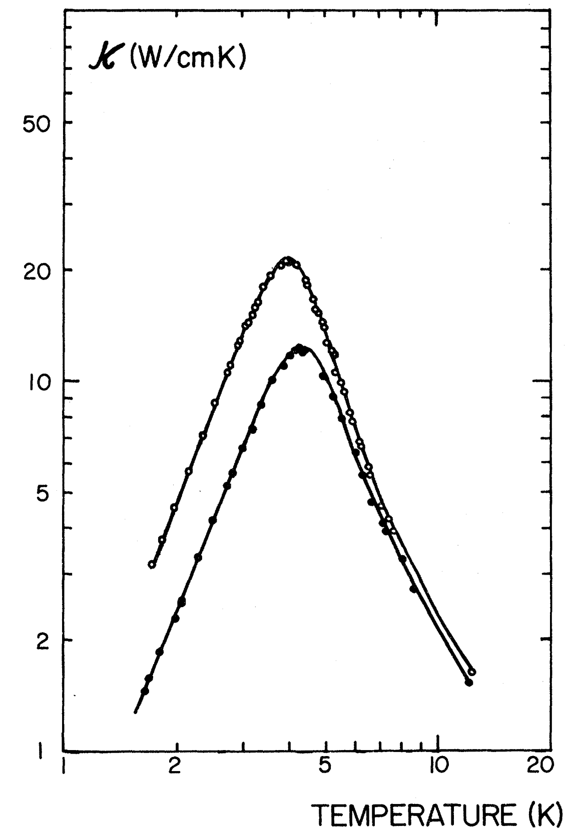

FIG. 2. Thermal conductivities κ of the thick (open circles) and thin (solid circles) arms vs temperature. In the lowest temperature range the slope of the curve for the thickest arm is 2.6 and the maximum lies at 4 K, while it is shifted to 4.3 K for the thinnest arm.

creasing temperature, as expected. The residual resistivity ratio (RRR) before cutting the original sample was $\rho_{300\,K}/\rho_{4.2\,K} = 230$, while those of the thick and thin arms are now 190 and 100, respectively.[7] Besides, it can be seen from Fig. 1 that a reasonable extrapolation of our results indicates that the size effect in the electrical conductivity would probably still be significant around 15 K, as is the case for the phonon-drag thermopower. It should be noted too that a little above this temperature one of the hole mobilities starts to depart from its T^{-2} variation.[8]

In order to have a parallel indication of the size dependence of the mfp of the thermal phonons, the thermal conductivity κ of both arms was measured in the same temperature range. It can be seen from Fig. 2 that at the maximum and in the lowest temperature range we observed the general features common to samples of different sizes. However, a size effect exceeding 10% persists at much higher temperatures than expected.

At the present stage, these observations show that the size effect in the thermopower and electrical conductivity occurs in the same temperature range, thus suggesting that the mfp of the concerned low-energy phonons should be considered in explaining, at least partly, the size effect in the electrical conductivity. As suggested by Parrott[3] a more refined theory might then reconcile theory with experiment. Perhaps it might also give a clue to the peculiar mobility variation, say below 20 K, where the T^{-2} variation might in fact be that of the subthermal-phonon mfp's, which could vary in this way[9] for a given wave number q, fixed by momentum and energy-conservation requirements by the corresponding electron or hole wave numbers.

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