# Surface-Temperature Determination in the Kapitza Problem*

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It is shown that surface superconductivity can be used as a localized thermometer. Its application to the study of the thermal boundary resistance at a Pb-$^4$He interface provides the first experimental evidence for the existence of a rapid temperature variation within microscopic distances from the surface.

It is well known[1] that the experimental results for the Kapitza resistance between liquid helium and solids, at temperature above 0.1 K, are in strong disagreement with Khalatnikov's acoustic mismatch theory.[2] Peterson and Anderson[3,4] modified the Khalatnikov theory by introducing phonon attenuation in an interface region near the solid surface. This process improves the heat transfer between the media and is able to bring the theory into agreement with experimental results at low temperatures. To explain the results for temperatures of the order of 1 K, however, it is necessary to assume that the attenuation occurs at atomic distances from the surface, and, as the authors point out,[3] it is difficult to think of any mechanism which could produce such a strong attenuation.

It is generally accepted that other mechanisms may contribute to the heat-transfer process. It is then important to ask where these processes take place. The usual extrapolation method to determine the surface temperature does not provide an answer to this question, since the thermometers, because of their macroscopic size, are located far from the interface region. Consequently, it should be interesting to measure the actual temperature of the solid surface. Under the assumption that this temperature can be defined, a zero-thickness thermometer would be needed. No such ideal thermometer exists, but we show in this Letter that the nucleation of surface superconductivity can be used as a thermometer which is localized at the surface and which, having a thickness of the order of the coherence length, is orders of magnitude smaller than standard thermometers.

Surface superconductivity appears at temperatures and magnetic fields related by a well-defined function of the solid, $H_{c3}(T)$. Once this function is known, the measurement of $H_{c3}$ determines the surface-sheath temperature. Our main effort has been directed towards showing that this thermometer can be used to study the temperature near the surface of the solid.

Figure 1 shows schematically the experimental arrangement used for our measurements. The sample rod is glued to the bottom of the $^4$He pot by use of Epibond 100A, and the surface of the sample is leveled to the stainless-steel surface by spark erosion. The superconducting transition was measured by a mutual-inductance technique. The secondary coil was located with its lower end at approximately 0.2 mm from the stainless-steel surface that was in contact with the liquid helium. The primary ac field was always less than 0.1 Oe. The dc magnetic field was

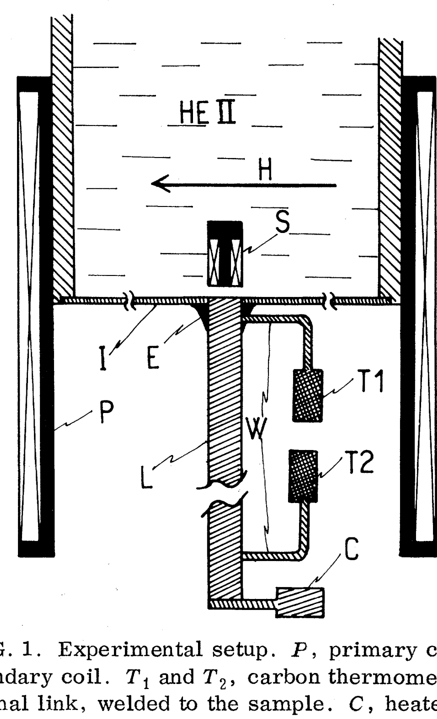


FIG. 1. Experimental setup. $P$, primary coil. $S$, secondary coil. $T_1$ and $T_2$, carbon thermometers. $W$, thermal link, welded to the sample. $C$, heater. $L$, lead rod, 3 mm diameter, 30 mm long. $I$, stainless-steel disk, 0.1 mm thick. $E$, epibond seal. $H$, external dc field.

applied parallel to the sample surface in contact with the liquid helium. The two carbon resistors could be used to obtain the surface temperature by extrapolation. The sample used was polycrystalline lead,[5] a type-I superconductor which shows surface superconductivity.[6]

To obtain the calibration curve $H_{c3}(T)$, the mutual inductance $M$ was measured as a function of decreasing dc magnetic field for different temperatures as determined by the $^4$He vapor pressure. At the same time calibrations of the carbon resistors were made. Typical results are shown in Fig. 2 (curves $a$–$c$). Their main characteristics are as follows:

(1) The smooth variation of $M(H)$ near $H_{c3}$ does not allow a precise determination of the absolute value of $H_{c3}$.

(2) Variation of the skin depth due to magnetoresistance leads to a field-dependent mutual inductance in the normal state. This effect disappears for ac frequencies less than 30 Hz, making the high-field part of the curves horizontal. However, in order to maintain a convenient signal-to-noise ratio, we have chosen a frequency of about 300 Hz.

(3) The field at which the sharp peak in each curve occurs, $H_p$, coincides with the thermodynamic critical field $H_c$. This effect deserves further study, but indicates a sudden change in magnetic flux due to perturbations of the surface superconductivity by the bulk transition. Immediately after the peak a finite change in mutual inductance is observed, and is probably related to the appearance of macroscopic domains in the intermediate state. The inset in Fig. 2 shows this phenomenon for a reduced sweep velocity of 0.3 Oe/min. The peak occurs in such a short time that the amplitude and width shown in Fig. 2 are fixed by the time constants of the electronic circuit.

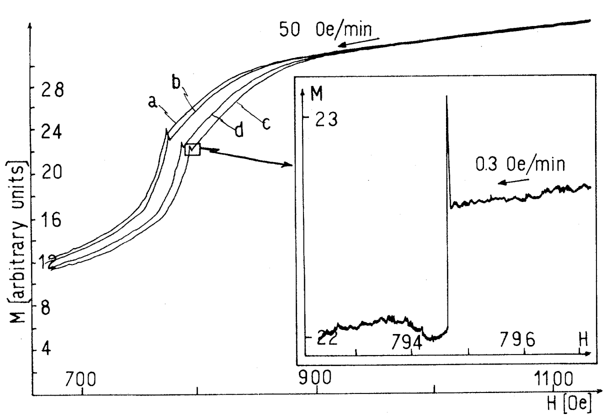


FIG. 2. Mutual inductance as a function of magnetic field. Curves $a$–$c$ correspond to $T=2.15$, $T=2.00$, and $T=1.70$ K, respectively, with no heat flux. An applied heat flux displaces curve $c$ to $d$. Inset shows the peak in an expanded scale.

(4) The shape of $M(H)$ for $H > H_p$ does not change for different temperatures in our range of interest (1–2 K). Using this experimental fact we are able to determine the temperature variation of $H_{c3}$ with greater precision by measuring the shift of the curves with respect to one taken as reference. The temperature variation of $H_{c3}$ obtained in this way is in good agreement with previous experimental results.[7] The magnetic field $H_p$ associated with the peak was found to correspond to $H_c$,[8] the bulk critical field. $H_{c3}$ as obtained by shifting the $M(H)$ curves allows a temperature determination with a precision of 10 mK; the use of the peak improves the precision to 3 mK.

Once the calibration is obtained, heat is applied to the sample. The temperature gradient measured by the two carbon resistors determines an extrapolated surface temperature $T_K$. The heat fluxes were chosen so that in no case did $T_K$ surpass $T_{\rm He}$, the helium temperature, by more than 5%. Under this condition, $T_K - T_{\rm He}$ was found to be proportional to the heat flux. The magnetic field is then varied and a typical curve of inductance versus $H$ is shown in Fig. 2, curve $d$. It is important to remark that the shape of the $M(H)$ curve is then used to determine $H_{c3}$ and $H_p$ as described in (4) above.

From $H_{c3}$ and $H_p$ two temperatures are obtained, which coincide within the experimental error, showing that the phenomenon which produces the peak takes place close to the surface. This is consistent with point (3) above. The temperature so obtained, $T_M$, is that of the interface region in which superconducting nucleation occurs. With use of the temperature of the helium bath and $T_K$ and $T_M$, two boundary thermal resistances, $R_K$ and $R_M$, respectively, are defined.

The experiments were carried out on a sample submitted to four different surface treatments. In experiment I the surface of the sample, after being spark-eroded,[9] was exposed to air for about 1 h before cooling to liquid-air temperatures. After one month of further exposure to air experiment II was performed. In experiment III the sample surface was electropolished and exposed to air for about 10 min, whereas in experiment IV the sample, after similar electropolishing, was left in contact with air for 24 h.

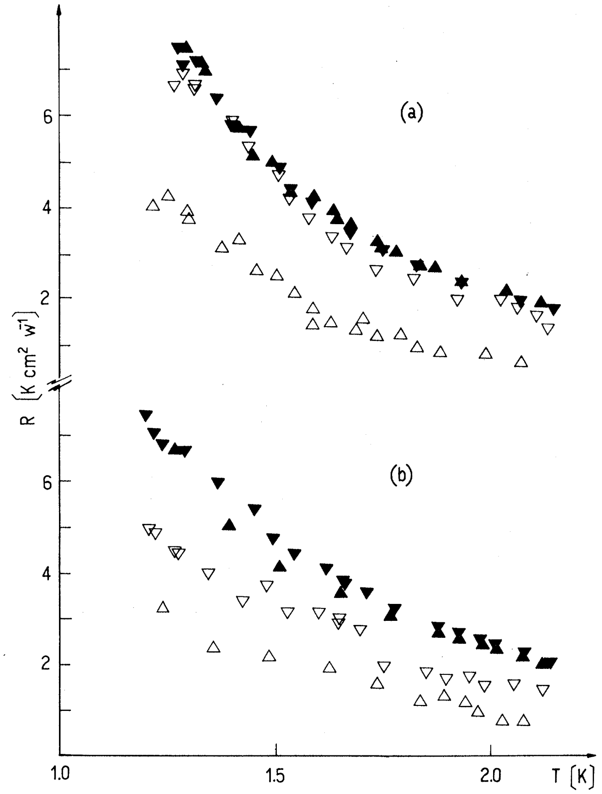


FIG. 3. (a) Kapitza resistance, $R_K$ (▲ and ▼) and $R_M$ (△ and ▽) defined in the text, as a function of temperature. ▲ and △, surface spark eroded and exposed 1 h to air; ▼ and ▽, the same surface treatment, one-month exposure to air. (b) Kapitza resistance, $R_K$ (▲ and ▼) and $R_M$ (△ and ▽), as a function of temperature. ▲ and △, surface electropolished, exposed to air 10 min; ▼ and ▽, the same surface treatment, 24-h exposure to air.

The experimental results, Fig. 3, show the following main features: (i) The extrapolated Kapitza resistance $R_K$ is in general agreement with previous results[1] and is practically the same for all four experiments. (ii) Unlike $R_K$, $R_M$ shows strong dependence on surface conditions. (iii) In all cases the temperature of the surface as determined by the superconducting sheath is closer to the temperature of the liquid than that obtained by extrapolation from the bulk gradient. Consequently for all experiments $R_K/R_M > 1$. (iv) The ratio $R_K/R_M$ increases with temperature for each experiment and it decreases with increasing exposure to air.

With the present theoretical knowledge of the processes that contribute to the heat transfer at microscopic distances from the surface we cannot explain why $T_M$ is more sensitive than $T_K$ to surface treatment. Nevertheless our experiments show that $T_M$ is much more a fundamental parameter than merely a local temperature determined by a different thermal resistivity (caused by different surface treatments) between the thermometer $T_1$ and the surface. This different resistivity would be in series with the bulk one, and consequently $T_K$ should follow the variation of $T_M$, in contradiction with the experimental results [see (i) above]. This conclusion is further supported by the fact that $H_{c3}(T)$ is the same for all experiments. Since $H_{c3}(T)$ is sensitive to electron mean-free-path variations,[10] it is hard to think of a strong change of mean free path in our samples.

Although for practical heat-transport problems $R_K$ can be taken as the relevant quantity, i.e., can tell us how much heat goes through the interface, $R_M$ is related to how the heat gets through. Any microscopic theory ought to be able to explain the difference between $R_K$ and $R_M$, a nontrivial requirement.

Theoretical papers[11,12] have pointed out the possibility of observing a rapid temperature variation within microscopic distances from the surface. When the heat flux is from the solid into the helium it is predicted that the temperature at the surface of the solid should be smaller than estimated by an extrapolation of temperatures in the bulk. Our observations are in agreement with this prediction. It is clear that this temperature profile cannot be observed by standard thermometry.

Further measurements over an extended temperature range and with different superconducting thermometer thicknesses are in progress.

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# Valence Charge Density in Indium Antimonide*

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Nine x-ray structure factors have been accurately measured in good-quality InSb single crystals and compared with various model calculations of the valence-electron charge distribution. Excellent agreement is obtained with a model based on tetrahedral distortion and ionic charge transfer, whereas Phillips's bond-charge model gave the poorest fit of all.

The nature of chemical bonding in tetrahedrally coordinated semiconductors is related to the spatial distribution of the valence-electron charge density. In $A_{\mathrm{III}}B_{\mathrm{V}}$ semiconductors the bonding is expected to be partly ionic and partly covalent, the latter component being responsible for departure from spherical symmetry.

Among the various models proposed for describing valence-electron charge densities, the most attractive for its simplicity is the so-called "bond-charge model," in which an interstitial accumulation of charge with spherical symmetry is supposed to exist along the bonds between nearest neighbors. Such a model, originally proposed by Ewald and Hönl[1] in 1936 in order to explain the nonzero intensity of the (222) x-ray reflection in diamond, and subsequently refined by Brill,[2] was recently redeveloped and extensively elaborated by Phillips who applied it to the III-V semiconductors and other crystals.[3,4] A number of properties such as ionicity, piezoelectricity, and lattice dynamics have been investigated using this model. Theoretical calculations of valence charge densities of various III-V crystals have also been recently performed using pseudopotential methods, and contour plots have been presented in the literature.[5] So far no direct experimental tests of these theoretical models have been available for III-V crystals. In the case of silicon, a comparison between experimental and calculated electron densities shows an overall fairly good agreement except for the shape of the bond maximum.[6]

It was found by one of us, a few years ago,[7] that x-ray reflections for which the two fcc lattices of the GaAs structure scatter out of phase (called in this paper "quasiforbidden") contain a good deal of information of about valence-electron asphericities. The reason for this is that the intensities of such weak reflections involve the difference between the Ga and As scattering form factors. Since the core charge densities are very similar for Ga and As, their scattering factors almost cancel out and a large fraction of the measured intensity is contributed by valence electrons. The core contribution can be accurately evaluated using Hartree-Fock (free-atom) wave functions. Any departure from spherical symmetry of the valence electrons is expected to produce appreciable effects on the measured intensities. These considerations are valid for all $A_{\mathrm{III}}$-$B_{\mathrm{V}}$ crystals when $A_{\mathrm{III}}$ and $B_{\mathrm{V}}$ are in the same row of the periodic table.

This work reports a series of accurate x-ray measurements of quasiforbidden structure factors in InSb, with the specific purpose of comparing experimental results with different model calculations.

The integrated intensities of nine reflections with $h+k+l=4n+2$ ($n$ 00, 1, 2, ...) have been measured in two large InSb single crystals of high quality,[8] cut parallel to the (100) and (111) planes.