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Electrons in Liquid Metals by Positron Annihilation*

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The angular correlation of photons from positrons annihilating in solid and liquid Na, Ga, In, and Sn has been measured. The results are interpreted in terms of a mean free path of the conduction electrons. These mean free paths are compatible with values derived from electrical conductivity measurements except for Ga which shows anomalous behavior.

In recent years much interest has been shown in the electronic structure of liquid metals, and considerable work, both experimental and theoretical, has been published.[1] One of the experimental techniques which promises some useful information is the observation of the angular correlation of photons from positron annihilation. This technique measures the momentum of the electron-positron pair at annihilation and may thus yield occupation density in electron **k** space if the positron momentum is known. Calculations by Lee-Whiting[2] and Adler[3] have shown that the positron thermalizes in metals before annihilation. This note adds some experimental evidence concerning thermalization of positrons and shows measured results from which the change of electron mean free path upon melting may be estimated for Na, Ga, In, and Sn.

The measurement of the angular correlation of annihilation photons was done in a slit-type apparatus described previously[4] which determines $N(k_z)dk_z$, k_z being selected by the experimental arrangement. The specimen-to-detector distances were 100 in. and the slit width was 0.030 in. for the sodium data and 0.050 in. for the other data. The various specimens were mounted in a small tray under the positron emitter and were heated by an attached automobile cigar lighter. The temperature of the heated specimen was determined to about $\pm 20°$C. The apparatus automatically scanned in steps the angle between annihilation photons coincident (10^{-6}-sec resolution) in the two detectors. The coincidence counting rate plotted against angle is approximately an inverted parabolic curve.[4]

The results have been presented not in the original form of the data but as differences between adjacent points on the angular correlation curve. These differences are called the slopes and are proportional[4] to $k\rho(\mathbf{k})$, where $\rho(\mathbf{k})$ is the density of occupation of states in **k** space. This presentation is thus directly related to momentum-space density. The slope presentation has the further advantage of displaying the statistical errors of the data which would be vanishingly small on the angular correlation presentation.[5] These slopes are presented in Fig. 1.

The geometrical resolution function of the experimental apparatus was folded into the positron mo-

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[1] For a review, see N. E. Cusack, Rept. Progr. Phys. (to be published).

[2] G. E. Lee-Whiting, Phys. Rev. **97**, 1557 (1955).

[3] S. L. Adler (private communication), and Phys. Rev. **130**, 1654 (1963).

[4] A. T. Stewart, Can. J. Phys. **35**, 168 (1957).

[5] See for example, A. T. Stewart, J. B. Shand, J. J. Donaghy, and J. H. Kusmiss, Phys. Rev. **128**, 118 (1962), where because of anisotropy the slope method does not yield $k\rho(k)$ and thus the angular correlation data was presented.

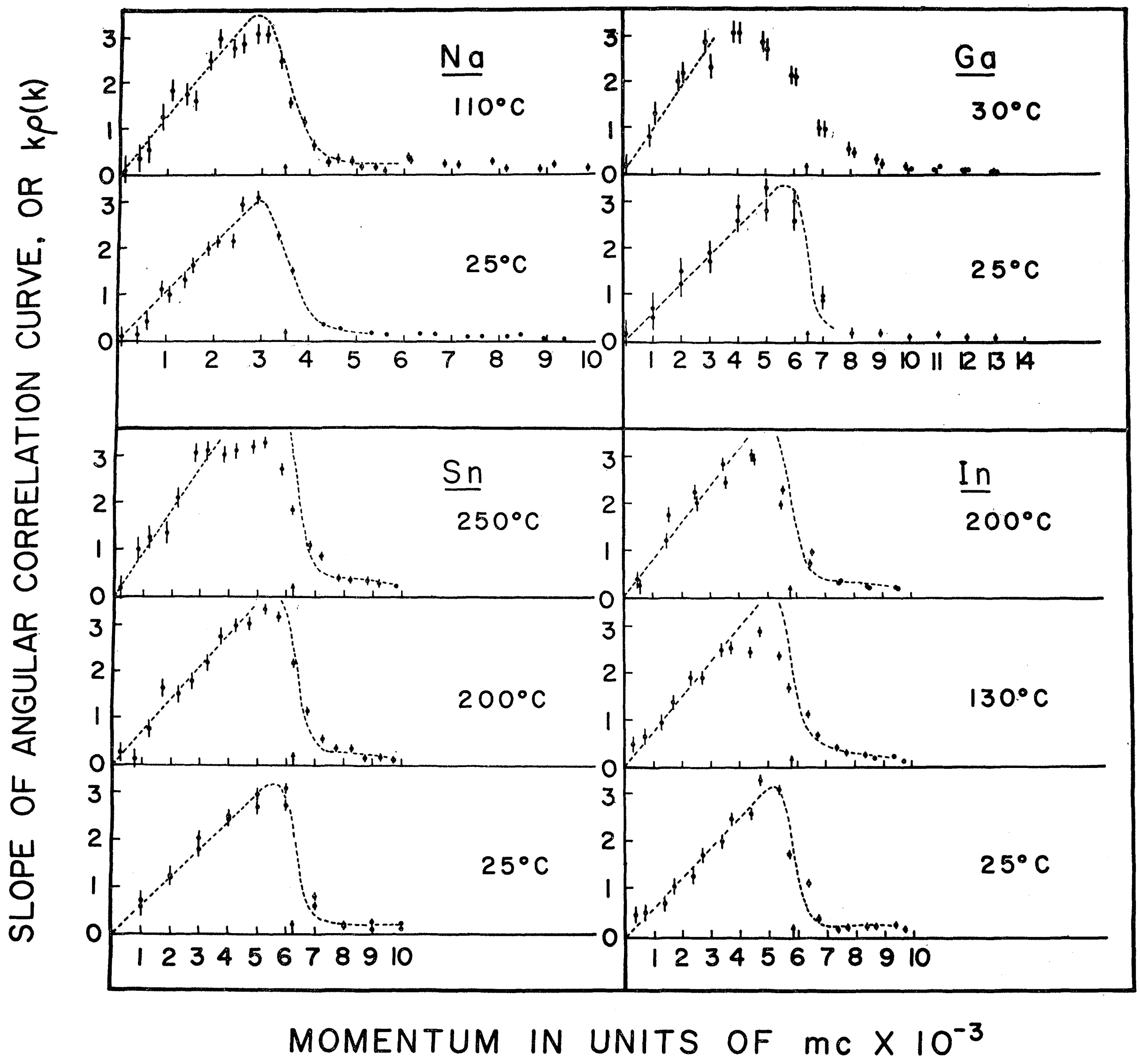

FIG. 1. Slopes of angular correlation data plotted versus electron momentum for solid and liquid Na, Ga, In, and Sn. The dashed curves have been calculated on the basis of the free-electron theory taking positron thermalization temperature and instrumental resolution into account. The small arrows mark the abscissa corresponding to the Fermi momentum. Note that the horizontal scale for Na differs from that of the other metals.

mentum distribution, which was assumed to be the Maxwell-Boltzmann distribution appropriate to the specimen temperature. The dashed curves in the figure are drawn by assuming a free-electron momentum distribution and folding it with the calculated resolution function. The low-momentum points were used to determine the initial straight-line portion of the dashed curves. This latter procedure appears unambiguous for the solid metals but is questionable for the liquids Sn and In, which do not appear to have a well-defined straight-line portion.

In some cases discussed below, smearing of the Fermi cutoff has been interpreted as a decrease in electron mean free path caused by heating or melting the metal. It has been assumed that the mean free path l is related to the distribution of uncertainty Δk in wave vector k_z by $\exp(-\frac{1}{2}\Delta k^2 l^2)$. Thus, $l=3.3/\Delta\theta$ Å-mrad, where $\Delta\theta$ is half the angular difference (near the cutoff) for a change from 20 to 80% of the full height of the electron momentum distribution.

Sodium. The data show a good fit to the calculated dotted line. Therefore, it has been concluded that: (a) The positrons are probably thermal; they certainly have less energy than $3kT$; (b) the calculated resolution function can thus be used for the other metals; and (c) the mean free path of electrons in this liquid specimen is not measurable, i.e., is probably greater than 40 Å. This is, of course, expected for sodium.

Gallium. Solid gallium shows a slight smearing of the maximum k cutoff which is probably due to anisotropy of the Fermi surface and accompanying higher momentum components of the electron wave functions. The results for liquid gallium show a very large "blurring" of the Fermi cutoff, which could imply an electron mean free path of $l \approx 1-4$ Å. It is known that the crystal structure of Ga consists of pairs of ions in the lattice.[6] If these pairs were much more tightly bound in the liquid, the molecular orbital electrons doing the binding would have a momentum distribution somewhat like the observed data. There would certainly be no Fermi cutoff. Knight, Berger, and Heine,[7] and Pashaev[8] have noted many other anomalies in the behavior of liquid and solid Ga.

Indium. The sharpness of the momentum cutoff in indium changes somewhat as the solid is heated from room temperature to about 130°C and does not change much more upon melting. If the increase in smearing above room temperature is interpreted to yield an electron mean free path, $l \approx 4-10$ Å is obtained for the solid and liquid near the melting point.

Tin. The results for tin show a slight change in the sharpness of the cutoff in heating the metal from room temperature to about 200°C. The mean free path obtained from the change in sharpness is $l \approx 25$ Å. Melting further decreases this to a value in the range $l \approx 3$–6 Å.

The mean free paths for In and Sn are approximately in accord with simple estimates from the conductivity,[1] although it is a little surprising that the thermal scattering in In appears in large part before melting. The expected mean free path for liquid gallium (≈ 17 Å) is not observed and it is possible that the measured momentum distribution cannot be usefully characterized by a Fermi cutoff and a mean free path.

We are indebted to J. J. Donaghy and J. B. Shand of this laboratory for invaluable assistance and to Dr. Philip Taylor for helpful discussions regarding positron velocities in metals. Dr. A. MacIntosh has kindly sent us a preprint of his recent measurements in liquid and solid mercury and we are indebted to Dr. N. E. Cusack for an advance copy of his review of the properties of liquid metals.

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[6] H. Hendus, Z. Naturforsch. **2a**, 505 (1947).

[7] W. D. Knight, A. G. Berger, and V. Heine, Ann. Phys. (N.Y.) **8**, 173 (1959).

[8] B. P. Pashaev, Fiz. Tverd. Tela **3**, 416 (1961) [translation: Soviet Phys.—Solid State **3**, 303 (1961)].

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Influence of Collisions on Scattering of Electromagnetic Waves by Plasma Fluctuations*

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The incoherent scattering of electromagnetic waves from electron-plasma oscillations in a thermal plasma is investigated. In particular, the effect of collisions on the shape of the isolated peak in the scattered intensity at a frequency displaced from the incident frequency by the plasma frequency is discussed making use of Nyquist's theorem and recent conductivity calculations.

In recent years considerable attention has been directed to the problem of the incoherent scattering of electromagnetic waves due to electron density fluctuations in a fully ionized plasma.[1–5] It is well established that most of the scattering arises from the low frequency fluctuations whose origin is due to the coupling of the electrons to the thermal motion of the ions. In addition to this dominant effect, there is a very sharp resonance in the vicinity of the electron plasma frequency. (See Fig. 2, Ref. 1.) For the scattering produced by long wavelength fluctuations the frequency width of this contribution is very small. It is the theoretical problem of the determination of the structure of latter isolated peak, for an equilibrium plasma, to which we turn our attention, since recently developed experimental techniques suggest this peak is resolvable.[6]

Now the expression for the differential scattering cross section is

$$\sigma d\Omega d\omega = (e^2/mc^2)^2[1-\sin^2\theta\cos^2(\phi-\phi_0)] \times S(\mathbf{k}-\mathbf{k}_0,\, \omega-\omega_0)\, d\Omega d\omega\,, \quad (1)$$

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[*] Work accomplished under the auspices of the U. S. Atomic Energy Commission.

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[2] E. E. Salpeter, Phys. Rev. **120**, 1528 (1960).

[3] J. A. Fejer, Can. J. Phys. **38**, 1114 (1960).

[4] T. Hagfors, Stanford Electronics Laboratories, Report No. 1, 1960 (unpublished).

[5] M. N. Rosenbluth and N. Rostoker, Phys. Fluids **5**, 776 (1962).

[6] We are indebted to Dr. E. A. Frieman for pointing this fact out to us.