sional-multichannel method and to demonstrate that it works well, it is of some relevance to further consider the comparison between the CPA and the space-time Magnus methods. The CPA starts to break down for nonresonant inelastic processes where the energy transfered is a large fraction of the kinetic energy in one of the channels.[1] Consequently, a comparison can be made with a simple model like the following: W_{11}, $W_{00}=\infty$ for $x<0$, $W_{00}=0$ and $W_{11}=1$ for $x>0$, and $W_{01}=V$ (a constant) for $0\leqslant x\leqslant A$ and $W_{01}=0$ for $x>A$.

The full quantum method, the CPA method, and our method (for a single interaction interval) may be solved analytically for this model. We find that at high energies the space-time Magnus method and the CPA both agree with the exact quantum results, and at low energies both methods give good results for the average transition probability. However, at low energies the space-time Magnus method very accurately reproduces the correct oscillations in the transition probability while the CPA is badly out of phase with the correct oscillations. (In the very-low-energy limit, the single-interaction-interval space-time Magnus method begins to develop some discrepancies.)

The reason that the semiclassical method provides an improvement over the CPA can readily be understood. The only approximation inherent in the semiclassical method involves the choice of a finite interval size, as the Magnus decomposition and the semiclassical approximation to the space-time propagator both become exact as the interval size decreases. It is the use of heavy masses and slowly varying potentials that permits the use of rather large intervals, thereby making the method useful.

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Thermalization of Positrons and Positronium

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Positrons in metals and positronium in quartz have been observed to thermalize down to nearly liquid-helium temperatures before annihilation. The achievement of such low temperatures by both positrons and positronium in approximately 10^{-10} sec indicates that phonon scattering plays a major role in thermalization. These results have important implications for the ability to achieve high precision in Fermi-surface studies by the positron-annihilation technique.

For many years the annihilation of positrons in solids has been used to investigate various characteristics of electrons in matter. It has usually been assumed that at the time of annihilation the positron, or sometimes the positronium atom, was in its lowest state in thermal equilibrium with its environment, although the experiments of Kim, Stewart, and Carbotte[1] suggested that this was not the case at very low temperatures. This Letter presents direct experimental evidence that, in certain cases at least, positrons and positronium atoms at the time of annihilation can be close to thermal equilibrium when in the liquid-helium temperature range.

The earliest calculations of positron thermalization by electron scattering[2,3] yielded about 10^{-12} sec for the time to reach room temperature. More recent and detailed calculations by Carbotte and Arora[4] led to a much longer thermalization time at low temperatures. Experiments on alkali metals by Kim, Stewart, and Carbotte[1] appeared to confirm the suggestion that positrons in metals annihilate with a certain minimum temperature below which they cannot thermalize. Although the agreement of theory[4] and experiments was only qualitative, the minimum positron effective temperatures were observed to increase with the density of the valence electrons as suggested by theory, and thus metals like Mg and Al would be expected to have even higher minimum temperatures. However more recently Perkins and Carbotte[5] predict that when positron-phonon interactions are included in the thermalization process, the positron should indeed be thermalized—even to about 20°K—at the time of annihilation.

Positron motion in metals can be directly observed from an experiment measuring the angular correlation of the annihilation photons, which carry off the momentum of the annihilating positron-electron pairs. The small contribution from the thermal motion of the positron can be seen as a smearing of the sharp corner of the electron momentum distribution at a momentum corresponding to the Fermi cutoff. However, the thermal motion of positrons is not the only source of smearing. Localization of some of the positrons at any traps also generates momentum smearing. Vacancies, voids, grain boundaries, dislocations, and surfaces have all been demonstrated to trap positrons. The total observed smearing also includes the contribution from the optical resolution of the instrument, and uncertainty in the actual resolution function can generate major systematic errors if the observed smearing and the resolution function are of comparable magnitude. Therefore, a very high resolution is necessary to observe the positron motion at low temperatures. Angular-correlation apparatus capable of achieving a resolution comparable with the thermal motion of positrons at liquid-helium temperature was recently built by us[6] and hence we attempted to resolve the discrepancy between the most recent theoretical predictions[5] and previous measurements.[1]

In a series of carefully annealed metals of widely different valence-electron densities (K, Mg, and Al) we have observed that positrons are close to thermal equilibrium with the sample at liquid-helium temperature. Assuming that the best fit to the total smearing of the angular-correlation curve reflects just the optical resolution, positron penetration into the sample, and the Maxwell-Boltzmann momentum distribution of free positrons at temperature T, we find $T = 25 \pm 25$°K in K, $T = 10 \pm 10$°K in Mg, and $T = 30 \pm 25$°K in Al. Only in the case of Na, where $T = 50 \pm 30$°K, did we find some evidence of nonthermalization or/and localization. Since Na undergoes a martensitic transformation at approximately 30°K we do not feel justified in ascribing this smearing to a lack of thermalization. In this paper we will present only Mg data, which was taken with the highest instrumental resolution (0.1 mrad slit width) and which has the smallest statistical uncertainty.

A high-purity (99.999%) single crystal of Mg was oriented to within 1° along the c axis, spark-machined flat, and carefully annealed. The experiment was performed at the sample temperature of 4.2°K by using ^{58}Co as positron emitter (400 mCi total activity). More than 10^4 counts per channel were accumulated on the peak of the angular-correlation curve, and data collection was heavily concentrated around the Fermi cutoff. The results, taken at both positive and negative angles, are displayed in Fig. 1, folded about

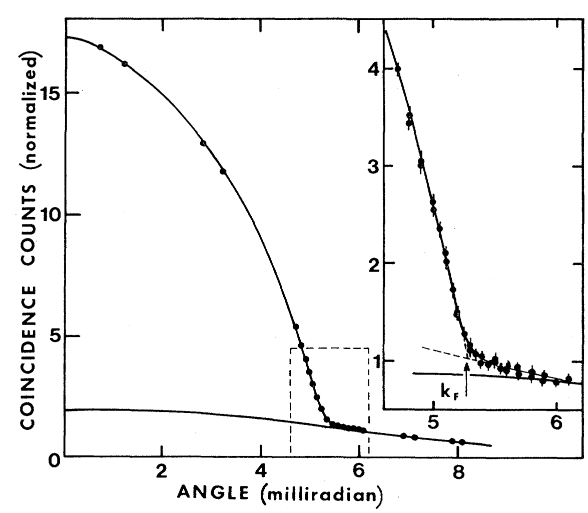

FIG. 1. Angular correlation of photons from positrons annihilating in a Mg single crystal at 4.2°K with the c axis in the direction of the observed component of momentum. The inset shows an enlarged region near k_F. The observed slope of the cutoff, the higher-momentum component shown beyond k_F, and the Gaussian fit to smearing are all discussed in the text. The smearing corresponds to a free-positron temperature of 10 ± 10°K.

the centroid of the curve. They clearly show two major parts of the curve: the parabolic part due to annihilation with the valence electrons and the broad component which is mainly due to annihilation with the core electrons. The broad component was fitted with a Gaussian function in the interval from 6 to 8 mrad. It was discovered that the Gaussian fit to the broad component could not account for all the annihilations beyond the Fermi cutoff. Another component of small amplitude (approximately 1% of the peak counts) just beyond the Fermi cutoff was observed. This component could be accounted for by higher-momenta annihilations of valence electrons scattered by the presence of the [0002] Brillouin zone boundary and would have an amplitude related to the energy gap at the zone boundary. In order to account for this component we have fitted the data with a two-orthogonalized-plane-wave calculation of the annihilating rate in the manner described by Berko and Plaskett.[7] A good fit was obtained using a value for the energy gap of $\Delta E = 0.05$ Ry, which is in excellent agreement with de Haas-van Alphen observations.[8] Below the Fermi cutoff the data indicated a somewhat steeper slope than predicted by the single-particle two-orthogonalized-plane-wave model. We fitted the data with a phenomenological enhancement of the form $1 + 0.25 \times (P/P_F)^2 + 0.38(P/P_F)^4$ which is larger than the enhancement resulting from the free-electron-gas calculation,[9] $1 + 0.19(P/P_F)^2 + 0.13(P/P_F)^4$. The fitted Fermi momentum for Mg at 4.2°K was found to be $(5.29 \pm 0.005) \times 10^{-3} mc$, corresponding to $k_F = (0.725 \pm 0.001)a_0^{-1}$, where a_0 is the Bohr radius. This is a little less than the free-electron value $(0.727 a_0^{-1})$ and in reasonable agreement with the result, $k_F = 0.722 a_0^{-1}$, obtained from magnetoacoustic-attenuation measurements.[10]

The smearing of the data was fitted with a convolution of the zero-temperature angular-correlation curve (calculated from the described model) and a Gaussian function representing the total resolution function of full width at half-maximum of 0.16 ± 0.02 mrad. Accounting for the instrumental resolution (including the slit width and positron penetration profile in the sample) this corresponds to the momentum distribution of a free positron in thermal equilibrium at a temperature of 10 ± 10°K. This result is virtually independent of the model used for fitting the data because the positron motion and instrumental resolution influence the angular-correlation curve only in the region within a fraction of a milliradian from the Fermi cutoff. The use of a more realistic mass[11,12] for the positron would reduce the temperature by a factor m^*/m (about 1.2–1.6).

Our experiments with the alkali metals yield lower minimum positron effective temperatures than previously observed[1] (100 ± 50°K in K and 160 ± 60°K in Na). Our new results have the advantages of superior instrumental resolution (by a factor of 2) and a finer spacing of data points in the region of the Fermi cutoff. As mentioned before, Na undergoes a martensitic phase transformation at low temperatures which may well introduce positron traps and a resultant momentum broadening. Until the importance of the defects caused by the martensitic transformation in Na (and in Li) is assessed, data from low-temperature positron experiments in these two metals must be viewed with caution. It should be emphasized that all the complications which we can envisage—whether they are instrumental artifacts, positron localization at trapping sites, or positron effective-mass ratio greater than unity—tend to increase the smearing of the momentum distribution. Thus an experiment to measure positron motion obtains an upper limit, and our present values for the minimum positron effective temperature, as well as the previous higher values, should be viewed from this perspective.

We have also observed the angular correlation of photons from positrons annihilating in oriented single crystals of quartz at 4.2°K. Previous experiments on this substance[13] clearly revealed two contributions to the angular-correlation curve: a broad Gaussian component due mainly to core-electron annihilations and a series of narrow components due to self-annihilation of positronium atoms. The width of the central most intense positronium peak was observed[14,15] to depend on the temperature of the sample and can be simply related to the momentum distribution of positronium atoms. Figure 2 shows an angular-correlation curve of quartz (k_z momentum along the x axis of the crystal) taken at 4.2°K. After subtraction of the broad component, the central peak was fitted well with a Gaussian function having a full width at half-maximum equal to 0.19 ± 0.02 mrad. Accounting for the width of the slits (0.1 mrad) and the positron penetration profile in the sample, this corresponds to the momentum distribution of free positronium in thermal equilibrium at the temperature $T = 10^{+2}_{-5}$°K. If the effective mass of a positronium atom were larger than its free-particle mass[14,15] this minimum temperature would be even lower. There have

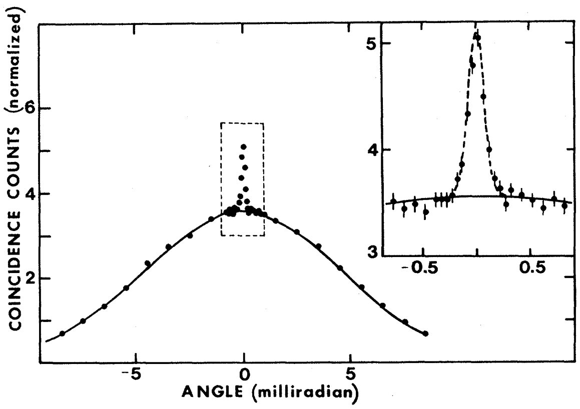

FIG. 2. Angular correlation of photons from positrons annihilating in a single crystal of quartz at 4.2°K with the x axis in the direction of measurement. The narrow peak has a width which corresponds to a free positronium atom with a temperature of 10^{+2}_{-5} °K.

been no calculations of positronium thermalization times in nonmetals at low temperature, but in view of the limited electron scattering it is clear that phonon contributions must be very important.

In conclusion, our experimental results in different materials clearly demonstrate that both the positron and the positronium atom are close to thermal equilibrium, even at liquid-helium temperatures, within times of the order of 10^{-10} sec. This is a very important observation, because it allows the high-resolution study of fine features of the Fermi surface in metals and alloys by the positron-annihilation technique. In particular, high-resolution angular-correlation measurements of annihilation photons can provide a direct caliper measurement of the Fermi momentum in metals and alloys to an accuracy of 0.1%. The precision of the results is mainly determined by the instrumental resolution rather than by positron motion as was suggested by earlier results.[1] Further, the results of our measurements of the thermalization of positrons in metals are in agreement with theoretical calculations which include the positron-phonon interactions. The thermalization of positronium atoms in quartz at low temperatures also emphasizes the importance of interactions with phonons.

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