Evidence for the Negative Work Function Associated with Positrons in Gold†

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Positrons which have been thermalized in various moderators and coated with ~200-Å gold leave the gold surface with an energy which peaks between 0.75 and 2.90 eV. This energy is thought to be associated with a positron or "negative" work function of gold.

INTRODUCTION

Until we reported[1] our observation of low-energy positrons, the negative results of Madanski and Rasetti[2] had been widely quoted as evidence for the lack of emission of low-energy positrons from surfaces of various materials. In their experiments, they used a ^{64}Cu positron source and looked for the emission of positrons with energies less than 150 eV from various metallic and dielectric materials. Later, in an unpublished thesis, Cherry[3] did observe positrons from a ^{22}Na source with energies less than 10 eV transmitted through mica which had been coated with a thin conducting layer of chromium. Following the method of Cherry, Madey[4] found emission of slow positrons from polyethylene with energies peaked near 20 eV. He speculated that the emission process was enhanced by an electric field built up within the dielectric by stopped positrons and their annihilation γ rays.

Our interest in the problem of emission of slow positrons first came from a desire to use them as projectiles in a study of positron-atom–scattering cross sections. On the basis of the experiments on the slowing-down spectrum of positrons with energy above 10 keV of Birkhoff and Wilkie[5] and the "continuous-slowing-down" theory of Spencer and Fano,[6] it was felt that, with a moderated high-energy source, one might be able to produce sufficient flux with energies below 100 eV to measure total scattering cross sections for various target gases.[1]

Repeated attempts to observe positrons in the interval 10–100 eV were unsuccessful although Paul *et al.*[7] using a very similar approach, found a signal associated with positrons between 0.3 and 1.5 keV. Following the independent suggestions of Kohn[8] and Callaway[9] that the positrons may be thermalized and then "thrown" from the material with several eV, attention was focussed on the energy interval ~1.0–10.0 eV. It was in this interval that we did find positrons, all in a peak near 1 eV. We associate this energy with what we call the positron work function or "negative" work function. This is the energy of the positrons which has been thermalized in the moderator and thrown out of a material. Detailed calculations of the mechanism have recently been completed by Tong.[10] A comparison of our results with his theory follows. More recently Jaduszliwer *et al.*[11] have verified the existence of this low-energy peak.

EXPERIMENT

Rather than using a radioactive source as our source of positrons, we used a 1-A peak current 55-MeV electron linear accelerator (LINAC). The positrons were created through pair production by the bremsstrahlung from a 20-nsec burst of electrons, at a repetition rate of 500 Hz. Energy analysis of the positrons was accomplished by fast time-of-flight techniques using a 3-long flight path. The coincident γ radiation from annihilation in a metal target at the end of the flight path determined the time of arrival of a positron. A schematic diagram of the source and flight path are shown in Fig. 1.

The bremsstrahlung target consisted of a water-cooled 0.3-radiation-length-thick tantalum target within a lead collimator. The forward-directed bremsstrahlung radiation impinged on the converter where the positron-electron pair was formed. This normally consisted of a "sandwich" of tantalum, in

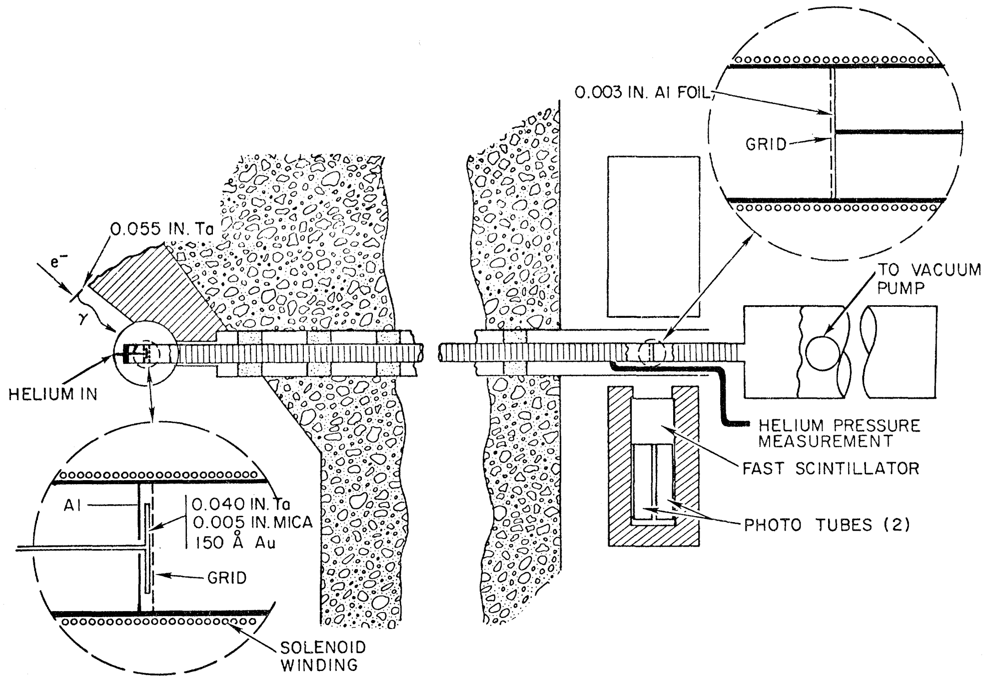

FIG. 1. Scattering chamber and time-of-flight energy analyzer, shown with details of positron-source region and positron annihilator.

which positrons were born, and a low-Z material, in which the positron energy was moderated and from which they were emitted. Several materials have been used for the moderator: aluminum (with an oxide coating of undetermined thickness) with a 200-Å gold coating; mica with a 150-Å gold coating; a CsBr crystal with a 150-Å gold coating. It was our thought at the time of the experiment that we were dealing with a bulk process and that the gold coating simply gave a gold equipotential surface. To make sure this was so, the coating was made as thin as possible so that it would still have a good uniform-conducting surface but not appreciably thermalize the positrons coming through it. However, Tong[10] has shown that the negative work function is largely a surface process. Thus we would expect all of our results to be somewhat similar and to reflect primarily the gold coating.

Because the positrons leave the moderator with essentially one energy, the converter-moderator sandwich could be biased at various potentials with an external power supply (refer to Fig. 2 for an example). A grounded aluminum plate immediately behind the converter target served as part of a 20-pF vacuum capacitor to prevent fluctuations in the potential on the converter target due to Compton and/or photoelectron currents. A grounded gold-plated copper grid was placed on the downstream side of the converter target. With this arrangement it was possible to accelerate or decelerate any positrons coming from the surface of the converter target. However, many of the positrons (between 30 and 60%) which appeared in the low-energy peak (near 1 eV) came from the gold-coated copper grid, perhaps the end of the flight tube, and other sources. For this reason, we separated the positrons coming through the gold-plated moderator from the others by appropriately biasing the target.

In Fig. 2, we show the positron spectrum when +15.00 V were placed on a tantalum converter with a gold-coated mica moderator. When one subtracts 15 eV from the energy of the distribution, a few positrons appear to come from energies less than 0 eV. This reflects only the resolution of the time-of-flight apparatus. It was unfortunate that we had to bias the converter moderator with respect to the grid, since in the time-of-flight experiment the resolution of the emitted positrons was approximately 25 times worse at 15 eV than it was just below 1 eV.

Although the flight path was magnetically shielded, a weak solenoidal field was required to prevent any residual ac or dc magnetic fields from deflecting the low-energy positrons into the walls of the flight tube. The annihilation target at the end of the flight path was also biased with respect to a grounded grid at its upstream side. This bias was required to drive the positrons into the surface of the annihilation foil. The target was an aluminum foil with a thin coating of gold black used to minimize back scattering. High-energy positrons, γ rays, neutrons, etc., could pass through the annihilation foil and proceed into a beam dump downstream.

The vacuum along the flight path was maintained

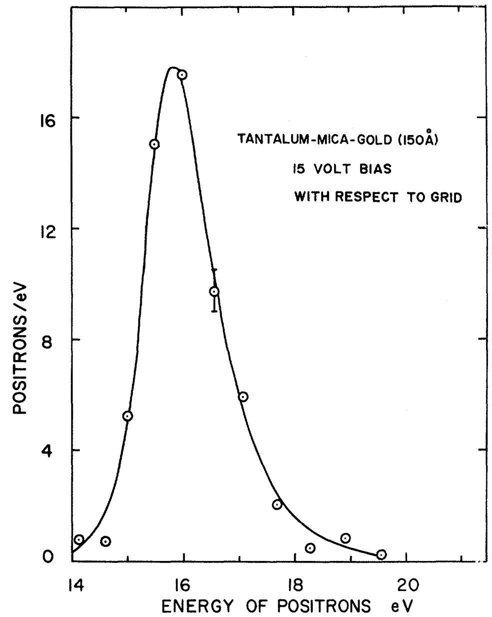

FIG. 2. Average positron energy distribution coming from a tantalum-mica-gold moderator sandwich biased 15 V with respect to the collision chamber. The error is the standard deviation associated with the average of eight runs.

at 2×10^{-7} Torr or better. Large amounts of lead and heavy concrete shielding separated the converted end of the flight path and the detectors. Each detector was a 6-in.-diam ×6-in.-long Ne 211 fast-liquid scintillator that was viewed by both a 56 AVP and a 6810-Å phototube. The multiple phototubes were required to suppress the effects of spurious "after pulses" from the prompt bremsstrahlung flash. The detectors had an efficiency of 1.3% for annihilation radiation for fourway phototube coincidence.

RESULTS AND DISCUSSION

As has been seen in Fig. 2, the energy at which the plot of the number of positrons/eV peaks with a 15-V bias on the moderator corresponds to a negative work function peaking near 1 eV. When displayed as a function of the time of flight, the number of positrons/time interval peaks closer to 2 eV. This apparent discrepancy originally led to some confusion.[1] The values from many runs for various (gold-coated) moderators are summarized in Table I in comparison with other calculated and measured values. The agreement for gold is good when one allows for the incomplete thermalization of the positrons in the thin gold layer.

In essence, the simple theory of the "negative" work function, which is discussed in the following paper,[10] describes the surface of the metal as a dipole layer with the electron density extending beyond the surface, resulting in residual positive charge within the surface. As the positron is brought from infinity into the dipole it eventually feels only the residual repulsive potential, which primarily sets the value of the "negative" work function. In the treatment by Tong,[10] allowance is also made for electron-positron correlation. It is important to realize that the value for gold can be related to part of the electron work function for the same material. This fact allows us to separate the electrostatic part and perhaps some of the electron-electron correlation parts of the work function from other quantum effects such as electron exchange which does not exist for positrons.

It has also been observed experimentally by us that the angular distribution of the positrons leaving the surface appears to be less than $\pm 2°$, much less than the $(1/40)^{1/2}[=(kT/1\text{ eV})^{1/2}]$ rad one might expect for electrons in thermal equilibrium with the solid and "thrown" from it with an energy of ~1 eV. This limit follows from experiments where the axial magnetic field was changed drastically without changing the time-of-flight distribution of the transmitted positrons. If the positrons had *a larger angular distribution*, then the transmitted peak would shift towards lower energies with increased magnetic field strength. This results from the fact that positrons which would normally be lost to the walls of the time-of-flight spectrometer would now spiral down the tube and be collected. This was not found. The reason for the small angle of emission is not yet understood.

The energy distribution of transmitted positrons near 1 eV is ~1 eV full width at half-height with a tail on the high-energy side. This width, unlike the case where the positrons are accelerated to higher

TABLE I. Values for gold-coated moderators in comparison to calculated and theoretical values.

Material	Values of negative work functions Φ^P Experimental (eV)	Theoretical (eV)
Gold	···	-0.77[a]
Mica (150 Å) gold	-0.75 ± 0.5[b]	···
	<5[c]	
CsBr (150 Å) gold	-2.90 ± 1.0[b,d]	···
Al (200 Å) gold	-1.25 ± 0.5[b]	···
Mica-chromium	<5[e]	
Polyethylene	-20.7[f]	···
Cu	···	-1.8[a]

[a]B. Y. Tong in Ref. 10.
[b]This work.
[c]D. A. L. Paul (private communication).
[d]The accuracy of this measurement is small due to experimental factors. However, the higher value may reflect the CsBr substrate since the gold foil is thin.
[e]W. H. Cherry in Ref. 3.
[f]J. M. J. Madey in Ref. 4.

energies, is not likely to be instrumental since at 1 eV the resolution of the spectrometer is very much smaller than the measured width. One can deduce a width of this magnitude if one assumes that the positrons leave an oscillating platform with a mass near that of the electron and with an energy near 1/40 eV. Then in the laboratory frame, the energy of the positrons would be $\sim 1 \pm 2\,(1\times 0.025)^{1/2}$ eV. Accurate measurements of this width, the angular distribution, and the energy as functions of the source temperature and film thickness are needed. Since the negative work function is largely a surface effect, extreme care with surface conditions will have to be taken.

The yield of positrons was between 1 and 10 positrons/sec, or between 10^{-7} and 10^{-6} of the total positron yield from the source. This is in essential agreement with the unpublished results of Cherry.[3]

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Negative Work Function of Thermal Positrons in Metals

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Theoretical study shows that thermalized positrons are thrown out of a metal with an energy of the order of several eV. This phenomenon is shown to be closely related to the electron work function of metals. Since energy is emitted when positrons leave the metal surface, it is named "negative work function." The negative work function of thermal positrons is $\Phi^p \simeq \Delta\phi^e - \mu_c^p + O(N_p/N)$, where N_p and N are the total number of positrons and electrons, respectively, in the metal. $\Delta\phi^e$ is the electrostatic potential across the metal surface due to the double layer taken from the electron work-function calculation, and μ_c^p is the correlation contribution to the positron chemical potential at the mean electrostatic potential.

Recently observations of low-energy positrons of several eV emitted from metallic surfaces when a high-energy positron source is directed onto the other side of the slab were reported.[1,2] Up to now the accuracy of such experiments[2] only allows us to take these results as qualitative indications of the existence of such low-energy positron sources. Metals and dielectrics like mica or polyethylene were used. Without realizing that such emission is mainly a surface phenomenon, in nearly all cases the material has been coated with a layer of metal, usually gold or chromium. The only quantitative measurement is reported in Ref. 1.

It is well known that high-energy positrons are easily thermalized in metals after a few collisions.[3] The low-energy emission of several eV, which is much larger than the kinetic energy of thermal positron (~0.025 eV), must therefore be related to the energy that a positron receives when it leaves the metallic surface. We call this the negative work function of the thermal positrons in metals. It is qualified by the word "negative" because unlike the electrons, energy is emitted on leaving the metal.

Let us first return to our understanding of the ordinary work function of a metal. The jellium model used in such theory says that the positive ions are replaced by a rigid uniform positive jelly. The electron cloud fills up the whole interior of the metal but it spills over a little near the edge [Fig. 1(a)]. This leakage of electrons and the excess positive background form a double layer [Fig. 1(b)] first suggested by Frenkel[4] and used by Wigner and