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Positron deformation potential and the temperature dependence of the electron and positron work functions

E. M. Gullikson and A. P. Mills, Jr.
AT&T Bell Laboratories, Murray Hill, New Jersey 07974-2070
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The sum of the electron and positron work functions, ϕ_0, has been measured for a clean Al(100) surface. As a function of temperature, ϕ_0 is found to be proportional to the thermal expansion of the lattice. Using the known thermal expansion coefficient of Al we obtain a value for the positron deformation potential, $\varepsilon_d = -11.7 \pm 0.5$ eV, in poor agreement with theoretical calculations but consistent with the known positron diffusion coefficient.

The electron work function[1] of a metal, ϕ_-, is the difference between the electrostatic dipole potential at the surface, Δ, and the electron chemical potential, μ_-, i.e., $\phi_- = \Delta - \mu_-$. The chemical potential is a bulk ground-state property of a solid and can be computed with high precision. Electron work-function measurements cannot be directly related to such computations without including the dipole term with its much larger computational uncertainty associated with the loss of symmetry at the surface.[2] Since the contribution to the positron work function of a metal, ϕ_+, from the dipole potential is of the opposite sign, $\phi_+ = -\Delta - \mu_+$, the sum of the electron and positron work functions, $\phi_0 = \phi_+ + \phi_-$, is not dependent on Δ and is therefore a bulk property. Measurements of ϕ_0 have the practical advantage of being independent of surface contamination, a problem which can make systematic measurement of ϕ_- or ϕ_+ difficult.

The sum of the electron and positron work functions may be obtained from independent measurements of the two quantities[3,4] or from a single measurement of the positronium work function[5] given by $\phi_{\mathrm{Ps}} = \phi_0 - \frac{1}{2}R_\infty$, where $\frac{1}{2}R_\infty$ is the electron-positron binding energy. Recently, Rosenberg, Howell, and Fluss[6] reported measurements of ϕ_{Ps} as a function of temperature for several materials and noticed that, unlike the nearly constant electron work function, ϕ_{Ps} has a significant temperature dependence. In agreement with this, we find for Al that the temperature dependence of ϕ_0 is mostly due to a change in ϕ_+ and that the change in the electron work function is relatively small. It is likely that this results from a cancellation of the changes in μ_- and Δ.[2]

Following Herring and Nichols[1] we separate the temperature variation into a part due to thermal expansion of the solid and a part due to intrinsic thermal effects,

$$\frac{d\phi_0}{dT} = 3\alpha\left[V\frac{\partial\phi_0}{\partial V}\right] + \left[\frac{\partial\phi_0}{\partial T}\right], \tag{1}$$

where α is the coefficient of thermal linear expansion. Our experiments show that the temperature dependence ϕ_0 is proportional to the thermal expansion of the lattice. This implies that the second term in Eq. (1) is small or else happens to have the same dependence on temperature as does α. According to Herring and Nichols[1] the main contribution to $\partial\phi_-/\partial T$ is due to the effect of lattice vibrations on the internal electrostatic potential. This results in a linear temperature dependence of the potential change at the surface Δ. While this effect can be quite large (-0.48 meV/K for Al) (Ref. 7) it should cancel in the summation over ϕ_+ and ϕ_-. This leaves the smaller effect of lattice vibrations on the chemical potential (< 0.1 meV/K).[1] From a measurement of $\phi_0(T)$ and the known thermal expansion of the solid we can therefore determine the positron deformation potential,[8] $\varepsilon_d = -V\partial\phi_0/\partial V$. The deformation potential is used to calculate the long-wavelength longitudinal phonon contribution to the positron scattering rate in a solid. We present here a measurement of $d\phi_0/dT$ and an implied value of ε_d for Al accurate to a few percent. We would like to emphasize that ε_d is a bulk ground-state property of a solid containing one positron and that it may thus be calculated with high precision and compared with our new measurement as a test of the theoretical assumptions such as the local-density approximation. Unfortunately, theoretical estimates for ε_d are in poor agreement with our measurement.

The experiments were performed using a magnetically guided beam of slow positrons in an ultrahigh-vacuum chamber with a base pressure of 2×10^{-10} Torr. The Al crystals were cleaned by repeated Ne-ion bombardment and annealing at 600 °C and were characterized using low-energy electron diffraction and Auger spectroscopy prior to measuring the positron reemission spectra. During ion bombardment impurity gases were pumped by a titanium sublimation pump and liquid-nitrogen-cooled cryopanel. After the cleaning cycle there was no surface contamination measurable with a double-pass cylindrical-mirror-analyzer Auger spectrometer. The samples were cooled to 20 K using a closed-cycle refrigerator with a liquid-nitrogen-cooled cold shield. A temperature run required approximately 80 min to cool from room temperature to 20 K. The slow positrons produced by a W(110) moderator and 5-mCi ^{58}Co source were accelerated to a few keV and implanted into the Al sample. Positrons reemitted from the Al were detected with a spiraltron channel electron multiplier as in Ref. 9. The energy spectra were obtained by applying a retarding potential to a set of cylindrical-analyzer electrodes located between the sample and the detector. The analyzer was maintained at room temperature to avoid having its electron work function, ϕ_a, change during the experiment.

In Fig. 1, the number of reemitted positrons is plotted versus the voltage applied to the analyzer relative to the sample, $V_a - V_s$. Spectra are shown for Al(111) and Al(100) at temperatures $T=300$ K and $T=20$ K. Vertical lines are drawn at the points where the electric field between the sample and the analyzer is zero, $V_a - V_s = \phi_a - \phi_-$. When $E=0$, there is a discontinuity in the slope of the integral spectra because some positrons are prevented from reaching the detector if $V_a - V_s > \phi_a - \phi_s$, whereas all the positrons are collected otherwise. It should be noted that the $E=0$ points, and thus the electron work functions, are independent of temperature within our experimental uncertainty of ±30 meV. We determined in a separate positron reflection experiment that the $E=0$ point for Al(100) does not vary by more than 20 meV between 20 and 300 K. The difference in the $E=0$ points for the two faces of Al gives us the difference in electron work functions

$$\phi_-(100) - \phi_-(111) = 220 \pm 50 \text{ meV}$$

in agreement with the previous measurements of Grepstad, Gartland, and Slagsvold[10] and the calculations of Lang and Kohn[2] and in disagreement with the empirical rule[11]

$$\phi_-(111) > \phi_-(100) > \phi_-(110) \; .$$

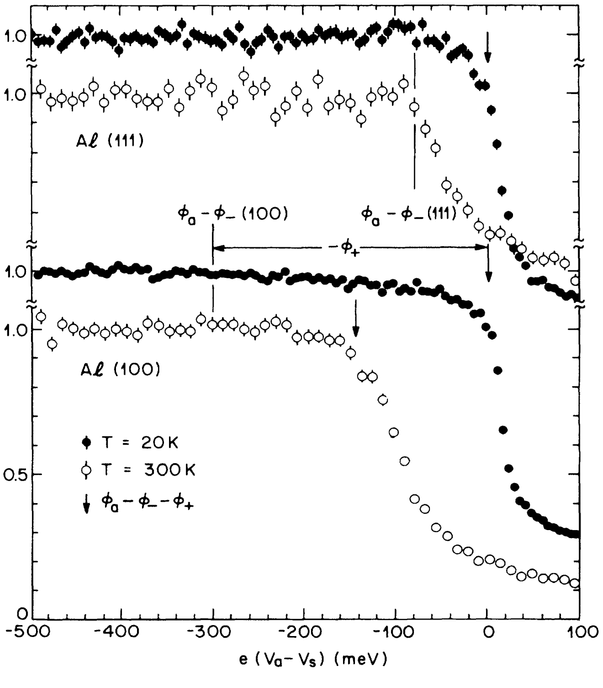

FIG. 1. Integral spectra of positrons reemitted from Al(111) and Al(100) surfaces at two temperatures, 20 K (●) and 300 K (○). The incident positron energy was 1500 eV at 300 K and 4800 eV at 20 K. At 300 K $\phi_+ = +65 \pm 30$ meV for Al(111) and $\phi_+ = -155 \pm 30$ meV for Al(100). At 20 K, $\phi_+ = -80 \pm 30$ meV for Al(111) and $\phi_+ = -300 \pm 30$ meV for Al(100).

The positron work function is given by the difference in retarding potential from the $E=0$ point to the point

$$V_a - V_s = \phi_a - \phi_- - \phi_+ = \phi_a - \phi_0 \; ,$$

where another change in slope occurs when the elastically emitted positrons are prevented from reaching the detector. Because of the quasithermal distribution of the positrons in the sample the latter point is kT^* less than the point of maximum slope.[12] The $V_a - V_s = \phi_a - \phi_0$ point is indicated in Fig. 1 by the vertical arrows. Since $\phi_a - \phi_0$ is independent of the Al dipole layer, it is the same for both Al(100) and Al(111). At 20 K, $\phi_+ < 0$ for both samples, $\phi_+ = -300 \pm 30$ meV for Al(100) and $\phi_+ = -80 \pm 30$ meV for Al(111). At 300 K the work function for the (100) face is still negative, $\phi_+ = -155 \pm 30$ meV, but it is positive for the (111) face. The spectrum for Al(111) at 300 K, which is due to nonthermalized positrons, is clearly different from the others. In particular, there is only one change in slope which occurs at the $E=0$ point, i.e., no vertical arrow for this curve. The work function for Al(111) at 300 K, $\phi_+ = +65 \pm 30$ meV, was obtained using the $\phi_a - \phi_0$ point determined from the Al(100) sample at 300 K. The value of ϕ_+ for Al(100) is in agreement with the measurements of Murray and Mills[13] if the previous value is corrected for the thermal shift of the reemission spectra.[12] The positron work function for Al(111) was previously thought to be negative because the emission of nonthermalized positrons was incorrectly interpreted.[14,15]

The 145 meV change in ϕ_0 between 20 and 300 K is much larger than the electron work-function change (< 20 meV). As explained by Heine and Hodges,[2] the dipole layer is driven by changes in the chemical potential so that their sum, ϕ_-, is nearly constant. On the other hand, ϕ_+ is directly affected by changes in the dipole layer, while ϕ_0 reflects changes in the chemical potentials. In Fig. 2 we present our measurements of the change of ϕ_0, $\Delta(-\phi_+ - \phi_-) = -\Delta\phi_0$, versus temperature. We have arbitrarily set $\Delta\phi_0 = 0$ at $T=0$. The nonlinear temperature

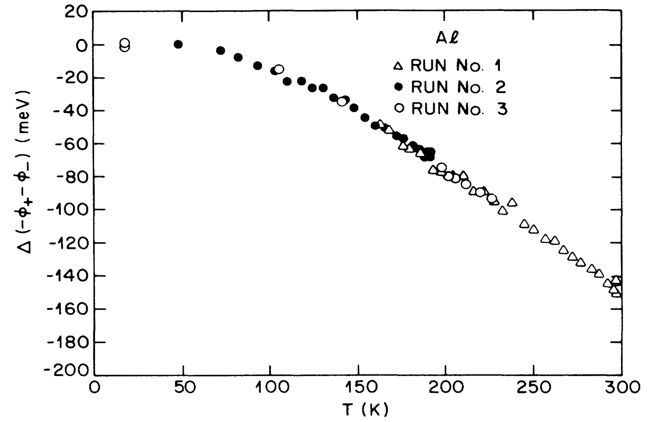

FIG. 2. Change in the sum of the electron and positron work functions for Al vs temperature.

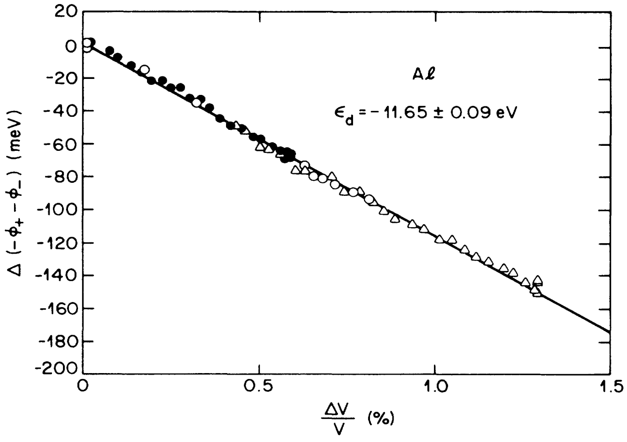

FIG. 3. The data from Fig. 2 plotted vs thermal expansion. The solid line is a least-squares fit and its slope is the positron deformation potential.

dependence appears to be attributable to thermal expansion. For temperatures less than 50 K the thermal expansion is small and a linear temperature dependence due to the second term in Eq. (1) should be evident. The data indicate that such a linear term must be less than 0.1 meV/K.

Figure 3 is a plot of $-\Delta\phi_0$ versus volume computed from the known thermal expansion of Al.[16] Assuming that the entire change of ϕ_0 is due to thermal expansion, we can calculate the positron deformation potential from the slope in Fig. 3,

$$\varepsilon_d = -V\partial\phi_0/\partial V = -11.7 \pm 0.5 \text{ eV} , \qquad (2)$$

where the error bar has been increased to account for systematic effects. Bergersen *et al.*[8] have calculated values of ε_d for several metals and for Al they obtain $\varepsilon_d = -8.6$ eV, in disagreement with our measured value. More recently Farjam and Shore[17] have calculated a value of ε_d in better agreement with our measurement than that in Ref. 8.

We now compare our value for ε_d with the measured positron diffusion coefficient in Al at 300 K,[18] $D = (0.76 \pm 0.14)$ cm^2 sec^{-1}. The Bardeen-Schockley[19] expression for the mobility of a charge carrier interacting with long-wavelength acoustic phonons gives us

$$D = (8\pi/9)^{1/2}\hbar^4 \langle c_{ii}\rangle m_+^{-5/2} \varepsilon_d^{-2} (kT)^{-1/2} . \qquad (3)$$

Here

$$\langle c_{ii}\rangle = 1.120\times 10^{12} \text{ dyne cm}^{-2}$$

is the elastic constant[20] associated with longitudinal waves averaged over the direction of propagation, and m_+ is the positron effective mass. For the positron effective mass we use the band effective mass,[21] $m_B = 1.15 m_e$, and the effective mass arising from correlation,[22] $m_{\text{corr}} = 1.07 m_e$. The combined effective mass, $1.22 m_e$, is in agreement with the experimental determination of Kubica and Stewart.[23] Substituting into Eq. (3) using $T = 300$ K gives us $|\varepsilon_d| = 12.0 \pm 1.1$ eV in good agreement with our experiment.

Recently, Schultz, Lynn, and Nielsen[24] reported an order of magnitude drop in D_+ for Al at 40 K relative to what one would expect based on an extrapolation of the room-temperature measurement using Eq. (3). Since the slope in Fig. 3, and hence ε_d, is constant over the 20–300 K temperature range, the temperature dependence of D_+ cannot be explained by a temperature-dependent ε_d. Instead, it has been suggested that the nonthermalization of the positrons could be responsible for the apparent decrease in D_+.[25]

The relation between $d\phi_0/dT$ and the positron diffusion coefficient can be used to understand the interactions of positrons in other solids besides Al. For example, based on the measurements of Schultz and Lynn[4] we calculate $\varepsilon_d = -13 \pm 1$ eV for Cu. The diffusion coefficient measurements of Ref. 18 then implies that the positron effective mass in Cu is $(1.3 \pm 0.1) m_e$. As a further example, the positron mobility in Ge (Ref. 26) implies $|\varepsilon_d| = 19$ eV if we use $m_+ \approx m_e$;[27] from the known thermal expansion coefficient at 300 K we then find $d\phi_0/dT \approx 0.34$ meV/K.

In conclusion, we have explained the temperature dependence of the positronium work function and used it to derive the positron deformation potential for Al. Our measurement is in good agreement with the known value of the positron diffusion coefficient but in poor agreement with current calculations of the electron and positron chemical potentials.

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