Fermi Momentum of Aluminum from 0 to 100 kbar

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The Fermi momentum of aluminum was studied from 0 to 100 kbar at room temperature through the utilization of positron annihilation. Over this pressure range, the Fermi momentum of aluminum is very well described by the free-electron model which predicts that the Fermi momentum of a metal is given by $p_F=\hbar(3\pi^2Z/V)^{1/3}$, where Z is the number of conduction electrons per unit cell in the lattice and V is the volume of the unit cell.

INTRODUCTION

INFORMATION about the properties of solids at very high pressures is essential for testing theories of solids, since this is the only manner in which the interatomic distance can be varied at constant temperature. Unfortunately, low temperatures, ~4°K, are required for most studies of the electronic structure of metals. This requirement has restricted the investigations to relatively low pressures.[1] Positron annihilation is a tool that has been used for the study of the distribution of the momentum of the conduction electrons.[2] Positron annihilation can be employed at room temperature, which is low compared to the characteristic Fermi temperature of the conduction electrons (10 000–50 000°K). Positron annihilation is not as precise or as well understood as those techniques available at very low temperatures; however, it is useful for obtaining some information about the electronic structure of metals and can be used at room temperature where very high pressures are attainable.

We have studied positron annihilation in aluminum in the range of 0 to 100 kbar at room temperature. Aluminum was chosen for this study since it is known not to exhibit any phase transition in the region of interest, and the results are simply interpretable.

From the positron-annihilation data we have obtained the variation of the Fermi momentum with pressure. It was found that the Fermi momentum p_F of aluminum increases with pressure in accordance with the prediction of the free-electron model:

$$p_F=\hbar(3\pi^2Z/V)^{1/3},$$

where Z is the number of conduction electrons per unit cell and V is the volume of the unit cell.

Melz[1] has studied the detailed topology of the Fermi surface of aluminum to 7 kbar using the de Haas–van Alphen effect. He found that the Fermi surface does not expand isotropically with compression and attributed this to a change in the band gaps. We will show that Melz's results are consistent with the present work, as the accuracy of this experiment is not nearly great enough to see the effect observed by Melz.

It is well known that the angular distribution of two-quantum annihilation of positrons in alkali metals, alkaline earths, and aluminum is characterized by a central parabola with a tail at large angles.[2] The interpretation of the tail is not yet certain; it has been explained by assuming that the positrons have a large effective mass or temperature.[3] It could also be due to the population of states above the Fermi level at absolute zero which was found by Luttinger for a system of interacting electrons.[4] However, it is well established that the angle at which the central parabola extrapolates to zero, θ_F, is related to the Fermi momentum by

$$\sin\theta_F=p_F/mc,$$

where m is the mass of an electron and c is the velocity of light.[5] A study of the variation of θ_F with pressure yields the pressure dependence of the Fermi momentum.

EXPERIMENTAL

We used 500 μCi of Na^{22} in this form of NaCl as the source of positrons. The NaCl was placed between two $\frac{1}{8}$-in.-diam disk of $\frac{1}{2}$-mil Mylar. This source was put between two disks of 99.9999% Al which were $\frac{5}{16}$ in. in diam and 7 mil thick to form a sandwich. The entire sandwich was contained by a $\frac{1}{2}$-in.$\times\frac{3}{32}$-in.$\times$20-mil pyrophyllite ring and placed between $\frac{1}{2}$-in. Bridgman anvils (Fig. 1). 2-in. NaI detectors located 1 m from the source with lead slits $1\frac{1}{2}$ in. high and 20 mil wide were used. The slits were perpendicular to the plane of the sample.

The zero-pressure annihilation spectrum of the positron source was obtained initially. It was assumed that pressure changes in the annihilation spectrum of the source were small in comparison to the total pressure change in the spectrum and possible changes in the source spectrum were neglected in analyzing the data. The source contributed roughly one-third of the total annihilation spectrum.

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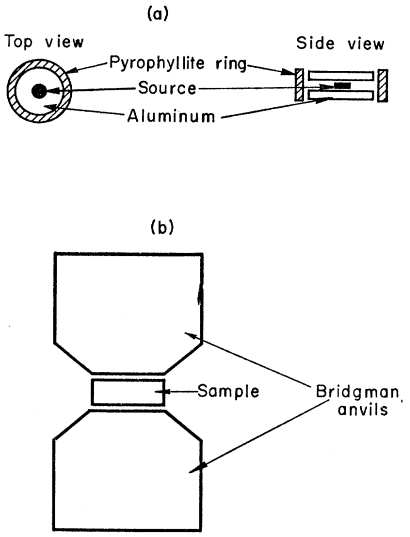

FIG. 1. (a) The positron source, aluminum, and pyrophyllite containing ring. (b) The assembled sample sandwich between Bridgman anvils.

Annihilation spectra were taken at 0, 27, 54, 81, and 108 kbar. The pressure calibration was based on the phase transitions of bismuth in the same geometry using silver chloride as the pressure transmitter. In going from 0 to 100 kbar, the γ emission rate of the source was found to decrease by 20%; this was not surprising as in Mössbauer studies to 100 kbar the intensities of the Fe^{57} lines have been found to decrease as much as 80% because of deformation of the anvils. The raw data at each pressure were corrected for the pressure and time decrease in count rate. The aluminum spectrum at each pressure was obtained by subtracting the source spectrum from the corrected data assuming that the ratio of total source to total aluminum counts was constant over the pressure range of interest and that the shape of the source spectrum did not vary with pressure. The results for the Fermi momentum at each pressure did not depend greatly on these two assumptions.

At each pressure studied, the central portion of the observed annihilation spectrum of the aluminum was fitted with a parabola. In fitting the data with a parabola we neglect the effects due to the finite resolution of the experimental apparatus and the finite width of the source; errors introduced by this assumption are small compared to the statistical uncertainty in the results.

RESULTS

The Fermi momenta at the five pressures examined are plotted in Fig. 2 as a function of pressure. The line in this figure is the Fermi-momentum curve predicted by the free-electron model assuming three free electrons per atom and using the volume data for aluminum of

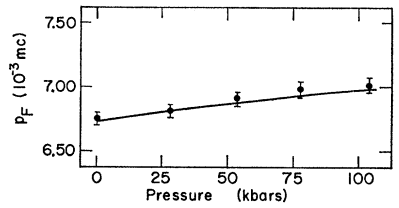

FIG. 2. The Fermi momentum of aluminum as a function of pressure. The line is that predicted by the free-electron model.

Bridgman[6] and Jamieson.[7] Figure 2 shows that the Fermi momentum of aluminum is very accurately described by the free electron model within the errors of this experiment.

We have found that the Fermi momentum of aluminum to 100 kbar is very accurately described by the free-electron model. This is intuitively reasonable as the Fermi surface of aluminum at zero pressure is fairly close to that predicted by the free-electron model assuming three electrons per atom. We would not expect the number of free electrons to increase with pressure as this would require promoting electrons from the atomic core to the conduction band which is energetically unfavorable.

Melz[1] has found in experiments to 7 kbar that the Fermi surface of aluminum does not grow as predicted by the free-electron model. Ashcroft[8] showed that the zero-pressure Fermi surface of aluminum can be fitted with two Fourier coefficients of a weak pseudopotential:

$$V_{111}=0.0179 \text{ Ry},$$
$$V_{200}=0.0562 \text{ Ry},$$

in an orthogonalized-plane-wave (OPW) calculation. Melz obtained the pressure derivatives of the pseudopotential coefficients based on form factors proposed by Harrison[9]

$$dV_{111}/dP=1.6\times10^{-4} \text{ Ry/kbar},$$
$$dV_{200}/dP=2.1\times10^{-4} \text{ Ry/kbar}.$$

He found that these pressure derivatives adequately explained his data.

Using Melz's[1] values of the derivatives of the pseudopotential coefficients we have calculated the average value of the momentum at the Fermi surface for a number of pressures between 0 and 100 kbar assuming that the coefficients are linear in pressure. In this calculation we have followed the procedure developed by Ashcroft. At a given pressure, a value of the Fermi energy was assumed and the volume enclosed by the surface of the assumed energy was computed. The Fermi energy was adjusted until the volume enclosed by the constant energy surface could hold exactly three elec-

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trons. The Fermi momentum at the given pressure was assumed to be the average electron momentum at the Fermi surface. The caluclated average momenta at the Fermi surface agreed with the values obtained from the free electron model to within 0.1% over the entire range. This difference is much smaller than the errors of our experiment, ±1%. Though the band gaps of aluminum are pressure-dependent, very little of the Fermi surface lies close to the zone boundaries. Thus the average momentum at the Fermi surface does not depend strongly on the band gaps so long as the band gaps are small.

This work suggests that positron annihilation studies of Fermi momenta may be an excellent tool for investigating either the pressure dependence of the volume of free-electron metals or the high-pressure electronic properties of metals for which the pressure dependence of the volume is well established. Positron annihilation would be most useful for studying metals with a large compressibility because of the difficulties involved in getting sufficient accuracy in the data and the weak expected volume dependence of the results.

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Ionic, Covalent, and Metallic Surface States of a One-Dimensional Mathieu Potential with Arbitrary Termination

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A model of surface states on a one-dimensonal crystal with sinusoidal potential has been simply solved for *arbitrary* values of the sinusoidal amplitude q, lattice termination z_0, surface step height ξ, and band-gap index m. The Schrödinger equation inside the crystal is equivalent to the Mathieu equation, whose eigenfunctions and eigenvalues are well known. Surface states are constructed as hybrids of bulk states [incorporating the proper nearly free-electron (NFE) and linear-combinations-of-atomic-orbitals (LCAO) limits], and they move towards the centers of the band gaps the stronger the surface "perturbation." It is shown that ionic surface states occur for $m=2$, $z_0=\frac{1}{4}\pi$, and $m=2$, $z_0=\frac{3}{4}\pi$; covalent surface states occur for $m=1$, $z_0=\frac{1}{2}\pi$; and metallic (or virtual) surface states occur in the allowed band between $m=0$ and $m=1$. Shockley states (looked for at $z_0=0$, all m) do not appear. This work has been favorably related to the author's semiclassical ionic model, to previous NFE and LCAO approximation methods, to the theories of Shockley and Statz, and to real surfaces in accordance with low-energy-electron-diffraction studies. In addition, it has uncovered a new linkage between the NFE and LCAO terminologies.

I. INTRODUCTION

THE properties of surface states in one dimension have been carefully studied using exact solutions, symmetry arguments, and approximation methods. Certain exact solutions have been carried out for crystals with Dirac-δ-function repulsive[1] and attractive[2] potentials, but these potentials seem to be highly unrealistic. General conclusions have been drawn from symmetry arguments for crystals terminated in either a symmetric or antisymmetric manner,[3–5] but the most interesting case of arbitrary termination has not been considered. Approximation methods which have been used most extensively include the nearly-free-electron (NFE) method[6,7] and the tight-binding [linear-combination-of-atomic-orbital (LCAO) or molecular-orbital (MO)] method.[8–10] At present, however, there seems to be no one-dimensional (1-D) surface-state model which is exactly solvable, sufficiently realistic and general, and independent of NFE or LCAO approximation methods.

The purpose of this paper is to present such a surface-state model. To be specific, the crystal potential will be sinusoidal with variable amplitude. The surface will be terminated by a potential step with variable height and variable location within the last unit cell. This will encompass symmetric and antisymmetric terminations as special cases. For this problem, the Schrödinger

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