Distortion of an Unoccupied Band in Be by the Electron-Plasmon Interaction

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By means of angle-resolved photoemission, the unoccupied band structure of Be along the Δ axis has been determined. While the data are in generally good agreement with the results of a self-consistent pseudopotential calculation, additional structure near the threshold for plasmon production is seen. It is argued that this structure is a many-body distortion caused by the electron-plasmon interaction.

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It is common in photoemission studies of solids to identify spectral features with ordinary band energies. At the same time, it is well known that many-body effects can complicate and even invalidate this identification. These complications have been observed, for example, in narrow-band metals, where atomiclike correlation effect can reduce bandwidths and generate extra peaks. It has also been predicted[1] that the band structure of simple metals should be distorted from its free-electron-like behavior at energies near the threshold for plasmon production. The origin of this effect is the electron-plasmon interaction. The one-electron effective potential and the excited-state lifetime are closely related to the real and imaginary parts of the electron self-energy, respectively, and thus coupled by a Kramers-Kronig type relation.[1] Whenever a new decay mode appears, e.g., creation of plasmons, additional structure in the energy-momentum relation for the electrons is also generated.

We have recently performed an extensive photoemission study of the electronic structure of Be.[2] In this Letter, we describe our results for the unoccupied states along Δ up to 22 eV above the Fermi level. While our data are in generally good agreement with the results of a recent self-consistent band calculation,[3] we see additional sharp structure near the threshold for plasmon production. We interpret these results as being the first observation of the distortion of electronic band structure due to the electron-plasmon interaction.

There are several reasons why this effect has not been observed previously. First, the effect occurs in the unoccupied part of the band, and attention has typically focused on the occupied part. Second, the material studied must meet two seemingly contradictory requirements. On the one hand, it must have strong interband transitions to final energies near the plasmon energy, or else photoemission into the band of interest will not be observed. On the other hand, interband transitions dampen the plasmon excitation, and obscure the interesting structure. Thus a metal intermediate between transition metals and free-electron metals is required. Finally, very accurate band calculations must exist, so that deviations can be clearly identified. This identification is easiest if the calculated bands are smooth in the region of interest.

Beryllium meets these requirements. The plasmon energy is[4] approximately 19 eV and strong direct transitions along Δ to final energies in the range 10–22 eV above E_f exist.[2] It is a wide-band *sp* metal, and recent self-consistent pseudopotential calculations have very successfully described its electronic structure. A single smooth band along Δ is predicted for the energies of interest.[3]

The experiments were performed at the Synchrotron Radiation Center of the University of Wisconsin in Madison, Wisconsin. All data in this Letter were taken with *p*-polarized light incident at 45° from the sample normal, collecting electrons along the normal. The experimental equipment as well as procedures specific to Be are described in detail elsewhere.[2,5,6] Total energy resolution is typically 0.2 eV.

Normal-emission spectra are presented in Fig. 1. The stationary peak at 2.8-eV binding energy is a surface state in the Γ_3^+-Γ_4^- gap. The dispersing peak at higher binding energies is the bulk transition, which is the subject of this Letter.

Conservation of momentum parallel to the sample surface restricts transitions to the Δ line in the Brillouin zone. Our procedure for obtaining the energy-momentum relationship along this line is as follows. First, we determine the top and bottom of the occupied band by measuring the extrema in binding energy of the bulk peak as a function of photon energy. This determination, which is *completely independent* of the form of the final state, gives the bottom of the band, Γ_1^+, at 11.1 ± 0.1 eV,

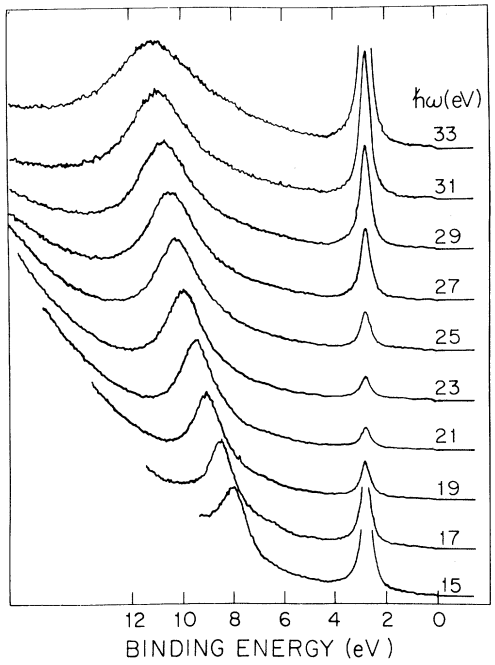

FIG. 1. Normal-emission spectra of Be(0001). All spectra have been scaled so that the dispersing peak maintains a constant height.

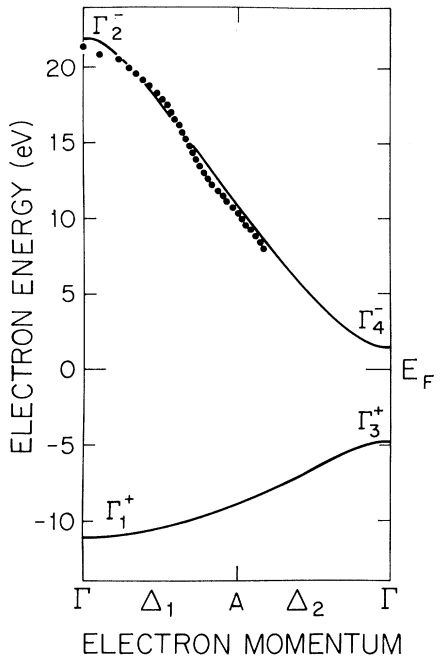

FIG. 2. Band structure of Be along Δ. The lower curve is the empirically determined occupied band. The upper curve is the calculated result from Ref. 3. The dots are the experimental unoccupied band.

and the top, Γ_3^+, at 4.8 ± 0.1 eV. These compare to the calculated values of 11.16 and 4.32 eV, respectively. The discrepancy at Γ_3^+ is discussed extensively elsewhere.[2] We then construct an empirical occupied band with the same shape as the calculated band, but scaled to agree with the experimental symmetry points. This scaling follows the expression

$$E(k) = E_{\text{calc}}(k) - 0.07[E_{\text{calc}}(k) - E_{\text{calc}}(k=0)],$$

where $E_{\text{calc}}(k)$ is the calculated binding energy. The resulting band is shown as a solid line in Fig. 2.[3] Once the occupied band is determined, the unoccupied band follows by subtraction of the binding energy from the photon energy at which a transition is observed. These experimental points, taken at $\frac{1}{2}$-eV photon-energy intervals, are shown in the upper half of Fig. 2.[7,8] The solid line is the calculated band. The general agreement is clearly good. Transitions to energies higher than those shown are mostly broad and weak, due to small dipole matrix elements, until Γ_4^- is approached at 92 eV.

Interesting structure in the experimental points is visible near 15 eV. In Fig. 3, we present the difference between the experimental and theoretical unoccupied bands as a function of reduced momen-

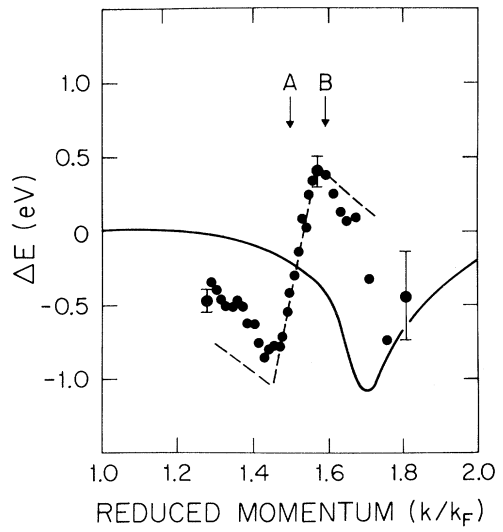

FIG. 3. Difference between theory and experiment in Fig. 2. The solid line is the predicted energy dependence of the effective one-electron potential for a homogeneous electron gas with the Be average density (appropriately scaled from Ref. 9). k_F is the free electron Fermi momentum for the Be density, 1.94 Å^{-1}. The dashed line shows how the experimental points change under a different data reduction (see text). The indicated uncertainties arise from the peak-position determination, and get larger as the initial band gets flatter.

tum. In this figure, point A is the wave vector of a free electron at the Be plasmon energy, while point B is the wave vector of an electron propagating along Δ in Be at an energy equal to the Fermi energy plus the plasmon energy. In an electron gas the two points would be identical.

We believe that the structure shown in Fig. 3 is a many-body distortion caused by the electron-plasmon interaction. It is important, however, to consider some other possible explanations. First, it is not an experimental artifact or caused by our peak-position determinations. These are very simple, one-component peaks, well separated from any other structure. Second, it is not caused by our data reduction. As an illustration, we show in Fig. 3 how the structure changes if the original, unscaled occupied band is used to determine the momentum. This curve still has all of the essential structure, although its shape has changed slightly. Finally, this structure is too sharp to be explained as errors in the calculation of the final state. Modifications to the calculation that are expected to increase its numerical accuracy in this energy range do not introduce additional structure.[3] We conclude that the existence of the effect in no way is a result of the particular theoretical states used.

A calculation of this effect for real metals would be difficult. As an approximation to what is expected, we show in Fig. 3 the calculated results for the homogeneous electron gas.[9,10] This comparison shows that the observed structure is close in magnitude, position, and width to what is expected. However, the experimental curve is clearly more complex, and begins at a smaller momentum, than predicted.

These disagreements could be due to errors in the theory, which are expected in regions such as these where the electron self-energy has a strong energy dependence.[1] They could also be due to the fact that electron-gas theory explicitly ignores interband transitions which are an important decay mode for plasmons. Finally, the electron gas is a poor approximation to Be in other ways. Be has large band gaps, and a very small density of states at the Fermi level. The difference between points A and B in Fig. 3 shows the importance of these band-structure effects and the difficulty of making a detailed comparison with electron-gas results.

There are other indications that the interaction between plasmons and electrons in real metals is inadequately described by electron-gas theory. For example, final-state lifetimes in Be, which should be dominated by plasmon generation, are significantly longer than predicted.[2] Also, plasmon loss satellites in Al[11] and Be[2] are smaller than calculated. Finally, the Be plasmon has a large (3–5 eV) width.[4] These points are particularly relevant because of the Kramers-Kronig relation between band distortion and plasmon generation.

In summary, we have observed anomalous structure in an unoccupied band of Be, and we have argued that this structure is caused by the electron-plasmon interaction. Electron-gas theory makes qualitatively similar predictions, but a quantitative description will require a calculation incorporating the effects of charge inhomogeneity and interband transitions. This phenomenon may also be observable in other electron spectroscopies that probe unoccupied bands, such as low-energy electron diffraction or inverse photoemission.

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[1]L. Hedin and S. Lundqvist, in *Solid State Physics, Vol. 23,* edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Holt, Rinehart, and Winston, New York, 1969), p. 1.

[2]E. Jensen *et al.* (unpublished).

[3]M. Y. Chou, P. K. Lam, and M. L. Cohen, Phys. Rev. B **28**, 4179 (1983); M. L. Cohen and M. Y. Chou, private communication.

[4]H. Raether, *Excitation of Plasmons and Interband Transitions by Electrons,* Springer Tracts in Modern Physics Vol. 88 (Springer-Verlag, Berlin, 1980), p. 1, and references therein.

[5]B. P. Tonner, Nucl. Instrum. Methods **172**, 133 (1980).

[6]C. L. Allyn, T. Gustafsson, and E. W. Plummer, Rev. Sci. Instrum. **49**, 1197 (1978).

[7]Peak positions were determined by first subtracting a smooth background, and then fitting a Lorentzian. Good fits were obtained at lower photon energies. The observed widths were very close to those predicted by the imaginary part of the self-energy for holes in jellium (Ref. 2). At higher photon energies, some asymmetry was observed, and the peak position reported is the one that gave the best fit to the top half of the peak. Essentially identical results, but with more scatter, were obtained by picking peak maxima by eye.

[8]Hcp lattices are complicated by having two atoms per unit cell. The observable bands along Δ have Δ_1 and Δ_2 symmetry and join smoothly at A. Since dipole-allowed transitions do not break this symmetry, it is convenient to unfold the bands as in Fig. 2.

[9]L. Hedin and B. I. Lundqvist, J. Phys. C **4**, 2064 (1971).

[10]This theory is tabulated for integral multiples of $0.2k/k_f$ for $r_s = 2$ in Ref. 9. We scaled to the Be density ($r_s = 1.87$) by the ratio of the Fermi energies. The curve drawn is a smooth interpolation between the tabulated points except for the minimum at $k = 1.7k_F$, which is estimated from a plot in Ref. 9.

[11]S. A. Flodstrom, R. Z. Bachrach, R. S. Bauer, J. M. McMenamim, and S. B. M. Hagstrom, J. Vac. Sci. Technol. **14**, 303 (1977).