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Angle-resolved photoemission study of the Ag band structure along the ΓΛL line

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Abstract. Angle-resolved photoemission experiments have been carried out on Ag{111} in the photon energy range from 13 to 65 eV with both s- and sp-polarised synchrotron radiation. The valence band structure of Ag has been determined along the high-symmetry Λ line by normal emission measurements. For photon energies smaller than 24 eV, the photoemission peaks could be interpreted as results of direct transitions between initial and final energy bands in the relativistic band structure calculated by Christensen and by Eckardt *et al*. For photon energies larger than 24 eV, the final band was obtained by fitting a free-electron-like final band ($m_e^* = 1.10$, $V_0 = -5.0$ eV) to experimentally verified calculated symmetry points. The experimental valence bands deviate by about −0.3 eV from theory. Measured critical-point energies (in eV, band indices in parentheses) are: $E[\Gamma_8^+(2,3)] = -6.15 \pm 0.10$, $E[\Gamma_7^+(4)] = -5.70 \pm 0.10$, $E[\Gamma_8^+(5,6)] = -4.95 \pm 0.10$, $E[L_6^+(1)] = -7.10 \pm 0.10$, $E[L_{4,5}^+(2)] = -6.25 \pm 0.10$, $E[L_6^+(3)] = -5.80 \pm 0.10$, $E[L_6^+(4)] = -4.55 \pm 0.10$ and $E[L_{4,5}^+(5)] = -4.30 \pm 0.10$. A surface resonance was detected 4.2 eV below the Fermi level at the centre of the surface Brillouin zone $\bar{\Gamma}$, which has an origin similar to that of the surface resonances that have been observed on Cu{001} and Ag{001}. A possible surface state is also reported at about 7.4 eV below the Fermi level at the $\bar{\Gamma}$ point. The nature of this impurity-sensitive structure is not understood.

1. Introduction

We report here the results of an extensive study of the photoemission from a clean Ag{111} surface at normal emission. These results are: (i) a new experimental determination of the band structure of Ag along the ΓΛL line, confirming the work of other authors and extending the range up to the L point; (ii) the identification of a surface resonance located at the $\bar{\Gamma}$ point 4.2 eV below the Fermi level; and (iii) the discovery of a surface state also at the $\bar{\Gamma}$ point, but 7.4 eV below the Fermi level.

An important factor in experimental studies of the electron band structure that are based on the interpretation of angle-resolved photoemission spectra is the choice of the final electron states. In photoemission experiments on Ag{111} with low photon energy ($h\nu < 23$ eV) a single band, band 7 of the calculations of Christensen [1] and of Eckardt *et al* [2], is the only energetically accessible final-state band. Within this low photon energy range, experiment and theory are in satisfactory agreement with one another.

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Courths *et al* [3] have found experimental points for the occupied band 7 in good agreement with theory [1, 2] by using the triangulation method of angle-resolved photoemission to determine the absolute location of direct transitions in k-space. Nelson *et al* [4] have fitted the band 7 calculated by Christensen to a plane wave and used it as a final state for the direct photoemission transitions. The experimental bands are generally in good agreement with the calculations of Eckardt *et al* [2] within 0.3 eV. Wern *et al* [5] have gone a step further by studying the intensity behaviour of direct transitions with photon energy and comparing it with calculated bulk momentum matrix elements for Ag in the vicinity of the Γ point. The energy positions and the dispersion of the final bands 7 and 8 near the Γ point were found to agree very well with the calculation of Eckardt *et al* [2]

All experimental studies of photoemission from Ag{111} have been limited, so far, to the low photon energy range below 25 eV. Under normal-emission conditions this photon energy limit does not allow band mapping to be extended to the L point. The critical-point energies at L can only be obtained by extrapolation [4] or by combination with off-normal emission data from UV resonance light sources [5]. One goal of the present study was to extend normal-emission measurements on Ag{111} to $h\nu = 65$ eV, thereby probing directly the electron band structure near the L point. For $h\nu > 24$ eV, the final band was obtained by fitting a free-electron-like band to experimentally verified calculated critical points. The final-state energy E_f at the Γ point ($k_\perp = 2\Gamma L$) along the $\Gamma\Lambda L$ line was derived from the behaviour with photon energy of the intensities of direct transitions from the initial states near the Γ point. The final-state energy at the L point ($k_\perp = 3\Gamma L$) was obtained by searching for an extremum in binding energy of initial states with different photon energy. The experimental valence bands thus obtained show quite good agreement with the theoretical results of Eckardt *et al* [2] except that a rigid shift of the theoretical d bands downwards in energy by 0.3 eV is required.

An occupied surface resonance is identified at the $\bar{\Gamma}$ point 4.2 eV below the Fermi level. The intensity dependence of the emission from this state upon the polarisation of the light has the characteristics of $\Lambda_{4,5}$ symmetry, allowing the conclusion that this surface resonance has a similar origin to the surface resonances which have been observed on Cu{001} [6] and Ag{001} [7]. In addition, a surface state is found 7.4 eV below the Fermi level at the $\bar{\Gamma}$ point. The intensity dependence of the corresponding emission upon the polarisation of the light has the characteristics of Λ_6 symmetry. The nature of this impurity-sensitive state is not yet clear.

This paper is organised as follows. We give experimental details in §2, and we present the experimental band structure of Ag along $\Gamma\Lambda L$ in §3. We discuss experimental evidence for a surface resonance and surface state at $\bar{\Gamma}$ in §4 and we summarise the conclusion of this work in §5.

2. Experimental procedure

The photoemission experiments were carried out at beam line U7B of the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory. A plane-grating monochromator was used to disperse the synchrotron light, and the experiments were carried out in the range of photon energies from 13 to 65 eV. The electron energies were analysed with an angle-resolved double-pass cylindrical mirror analyser with an angular resolution of 2°. The combined energy resolution (monochromator plus analyser) was

estimated to be 0.35 eV at 100 eV. The sample was mounted on a manipulator that allowed rotations around three mutually perpendicular axes, so that measurements could be done with either pure s- or 25% p-polarised radiation.

A clean Ag{111} surface was prepared by sequences of argon ion bombardment and annealing cycles. The cleaning process was continued until Auger electron spectra of the surface showed no C, O and S signals, although the C line was somewhat obscured by an Ag peak near 260 eV. The crystallinity of the surface was checked and found to be good by low-energy electron diffraction.

3. Experimental determination of band structure: results and discussion

3.1. Low photon energy range: 13 < hν < 30 eV

Normal-emission spectra from Ag{111} taken in the photon energy range between 13 and 30 eV with pure s- and 25% p-polarised radiation are shown in figure 1 (*a*) and (*b*). The broken lines show direct-transition structures and their dispersion with photon energy. The numbers in parentheses identify the band indices of the initial (occupied) bands 1 to 6.

Figure 2 depicts the results of theoretical determinations of the electron band structure of Ag along the ΓΛL line of the Brillouin zone, the full curves are from Eckardt *et al* [2] and the broken curves from Christensen [1]. The experimental data points were determined as follows. The energy of the initial states was determined by subtraction of the value of the measured final-state energy E_k (relative to the Fermi level) from the value of the photon energy. The perpendicular momentum $k_\perp$ along the ΓΛL line was determined with different final states in different ranges of the ΓL axis. In the central region (between approximately 0.2 and 0.8 ΓL), the final state was chosen as band 7 of the calculation of [2]. Since in the central region the dispersion of band 7 is very large, the choice of band 7 from either Eckardt *et al* [2] or Christensen [1] produces essentially the same results.

Near the symmetry point Γ, the experimental final-state bands were obtained from considerations of the photon energy dependence of the emission in the following way. In figure 1 we recognise strong intensity variations with photon energy of states with little or no dispersion between 20 and 28 eV. These intensity variations are more directly exhibited by constant initial-state (CIS) spectra, in which the emission from a given state is monitored as the photon energy is varied. Figure 3 shows such CIS spectra (normalised to constant incident photon flux) for the states at 6.2, 5.7 and 4.95 eV below the Fermi level. These spectra are wholly consistent with the theoretically expected intensity resonances (i.e. the direct product of transition matrix elements and surface transmission factor) calculated by Wern *et al* [5] (see figure 13 in [5]). The positions of the maxima in those theoretical curves give the momentum values of the final states, while the peaks in the CIS spectra give the energy values of the final states. With this information we obtain the data marked with an asterisk and a rhombus (in figure 2) for the final bands 7 and 8, respectively. Extrapolation of the Eckardt *et al* [1] band through these data points gives us then the experimental final-state band, between 17 and 20 eV above the Fermi level, that we need in order to map the initial-band structure near Γ. In figure 2 the pertinent experimental results for the initial bands are shown with full and open circles to indicate strong and weak peaks, respectively, of the electron distribution curves (EDC) in figure 1.

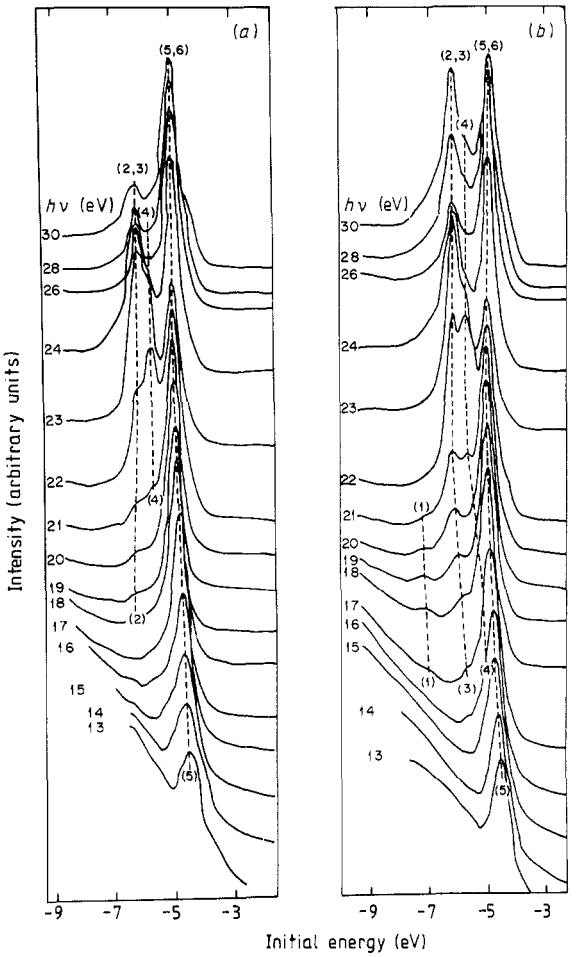

Figure 1. Normal-emission spectra from Ag{111} for photon energies $h\nu$ between 13 and 30 eV. (*a*) s-polarised light; (*b*) 25% p-polarised light.

3.2. High photon energy range: 35 < hν < 65 eV

In order to investigate the band structure along the ΓΛL line up to the L point we have extended our normal-emission measurements to photon energies of 65 eV. Figure 4 shows normal-emission EDC from Ag{111} taken in the photon energy range between 35 to 65 eV with s-polarised (figure 4(*a*)) and 25% p-polarised (figure 4(*b*)) radiation. As in figure 1, the broken lines mark direct transitions and their dispersion with photon energy, while the numbers in parentheses label the band indices of the initial occupied bands 1 to 5.

In figure 4(*a*), the line marking the positions of band 5 is slightly shifted away from the maxima. The reason for this is that bands 4 and 5 are energetically close to each other and it is very difficult to decompose the main peak of the EDC in figure 4(*a*) into two peaks. According to the relativistic dipole selection rule [8], transitions from the initial state with Λ_6 symmetry (band 4 in our case) at normal emission are not forbidden with s polarisation. However, the transitions from the initial state with $\Lambda_{4,5}$ symmetry (band 5 in our case) are only sensitive to the s component of the light [8].

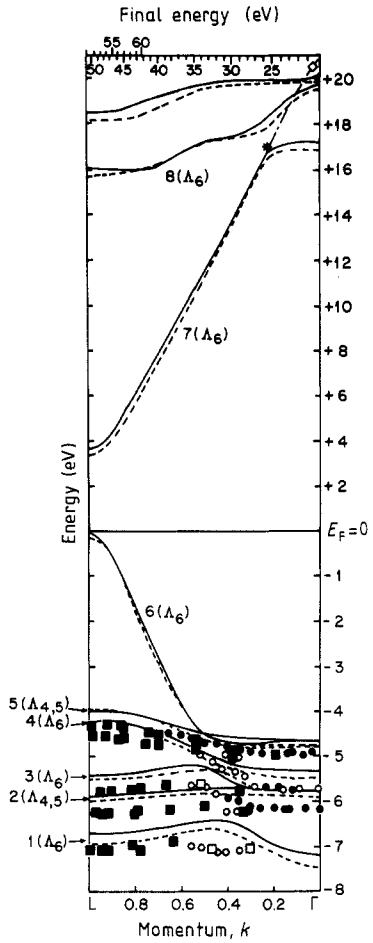

Figure 2. Electron band structure of Ag along the ΓΛL line. Full curves: calculations of Eckardt *et al* [2]; broken curves: calculations of Christensen [1]. Full (open) circles and squares refer to strong (weak) peaks in the EDC measured with photon energies lower than 30 eV and higher then 35 eV, respectively. The energy scale on the abscissa at the top of the figure gives the experimental final-state energies for k_f-values larger than 2ΓL.

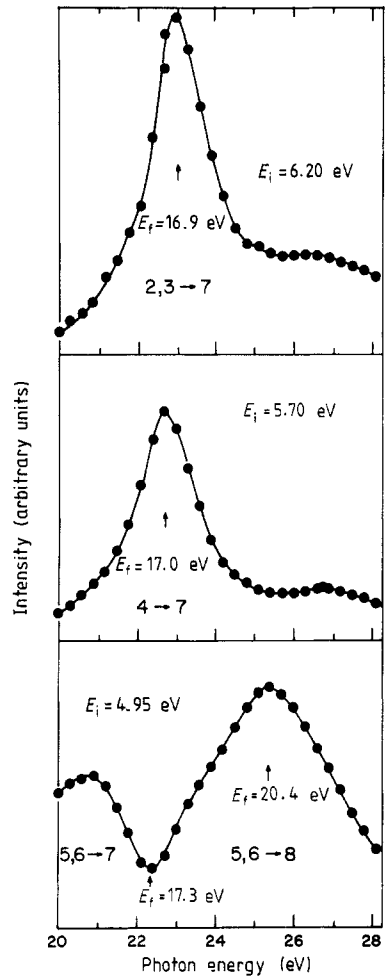

Figure 3. Constant initial-state spectra (CIS) for the states at 6.2 eV (top), 5.7 eV (middle) and 4.95 eV (bottom) below the Fermi level.

Hence, the contribution from the initial state of band 5 is expected to decrease when the s component of the light decreases.

The maxima of difference curves between the EDC with the same photon energy in figure 4 (*a*) and (*b*) should give the position of the $\Lambda_{4,5}$ symmetry band 5. As an example, figure 5 superimposes the EDC for 65 eV photon energy and exhibits the difference curve between them, which has a minimum at a little lower binding energy than the main peak in the EDC labelled A.

Figure 5 also provides the opportunity of judging, from the effects of polarisation of the light, that band 1 and band 4 have Λ_6 symmetry while band 5 has $\Lambda_{4,5}$ symmetry. As

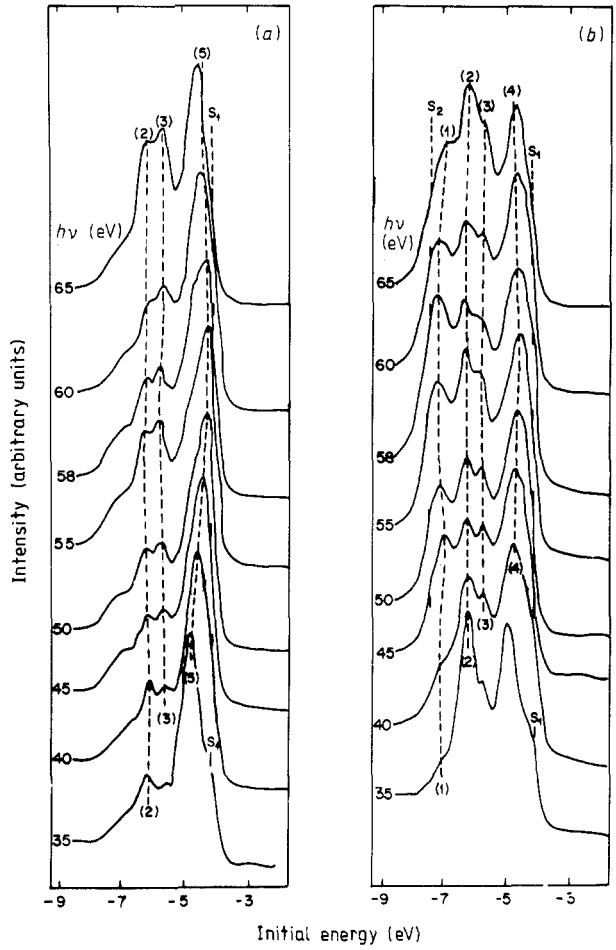

Figure 4. Normal-emission spectra from Ag{111} for photon energies $h\nu$ between 35 and 65 eV. (*a*) s-polarised light; (*b*) 25% p-polarised light.

far as bands 2 and 3 are concerned, visual estimation of the polarisation dependence in figure 5 seems to indicate that band 2 has Λ_6 symmetry and band 3 has $\Lambda_{4,5}$ symmetry. This indication is not unique to figure 5, but is apparent in all EDC depicted in figure 4 and is therefore not attributable to experimental error. It is, however, in contrast with the results of theoretical determinations (figure 2). In fact, theory [1, 2] shows that in the binding-energy range between 5.5 and 6.5 eV below the Fermi level, the photoemission peaks on the lower binding-energy side can originate only from bands 3 or 4 (and both these bands have Λ_6 symmetry) while the peaks on the higher binding-energy side should stem from the $\Lambda_{4,5}$ symmetry band 2, which overlaps band 3 near the Γ point. These discrepancies with theory are not understood at this time†. A possible explanation may be that, because bands 1, 2 and 3 are all close to each other, when the p component of the light increases, the intensity enhancement in the peak from band 1 affects the peak from band 2. However, deconvolution of the EDC

† At photon energies between 21 and 23 eV, in figure 1, the intensity of the peak from band 4 increases when the s component of the light is increased. This observation is also in contrast with the Λ_6 symmetry of band 4. A similar phenomenon can be seen in figure 16 of [5].

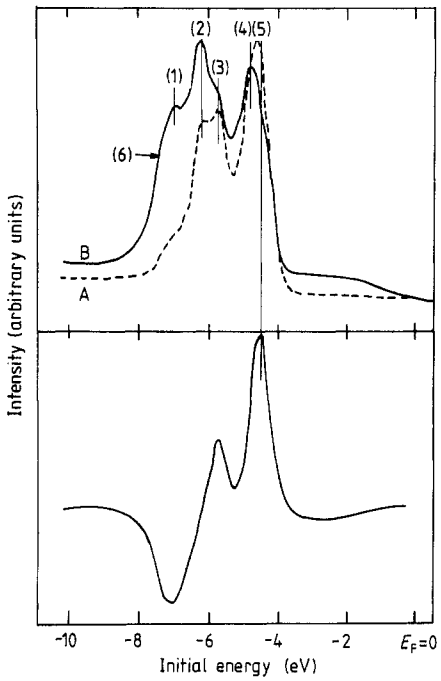

Figure 5. Effect of polarisation of the light on the electron distribution curves for $h\nu = 65$ eV. Curve A: pure s-polarised; curve B: 25% p-polarised light. The bottom panel shows the difference (A − B) between the curves drawn in the top panel.

in figure 5 (not shown here) shows that this explanation is not valid. Thus, we can only conclude that a quantitative interpretation of the dependence of photoemission intensity upon energy and polarisation of the light appears to be a challenge for theory. Experimentally, high energy resolution and high momentum resolution spectrometers should be used to shed light on this subject.

For photon energies greater than 24 eV the final band for direct transitions was obtained from a free-electron-like final band fitted to experimentally verified calculated critical points. At the Γ point ($k_\perp$=2ΓL) the final-state energy was estimated by means of the photon energy dependence of direct transitions from the initial states in the vicinity of Γ. Figure 2 shows that the final-state energy is about 20 eV above the Fermi level. At the L point ($k_\perp$ = 3ΓL), the final-state energy is obtained by searching for an extremum in binding energy of any initial state when the photon energy was varied. For example, in figure 4(*a*) the peaks from band 5 reach a minimum in binding energy when $h\nu$ = 55 eV. Hence, at $h\nu$ = 55 eV direct transitions occurred near the zone boundary (L point) and therefore the final-state energy is about 51 eV above the Fermi level. We have used the above two values to fit a free-electron-like final band at $k_\perp > 2\Gamma L$ with the equation

$$E_f - V_0 = \frac{\hbar^2}{2m_e^*} k_f^2 \tag{1}$$

where E_f is the final-state energy, V_0 is the inner potential and k_f is the final-state momentum. From equation (1), we find that the effective mass m_e^* of the electron is $1.10m_e$ (m_e is the rest mass of the electron) and the inner potential V_0 is −5.0 eV in

the free-electron-like final band. With this final-state band we have mapped out the band structure along the ΓΛL line for photon energies between 35 and 65 eV. On the top abscissa in figure 2 we have drawn an energy scale that gives the experimental final-state energy for k_f values larger than 2ΓL. The experimental data points for the initial states have been drawn with full (open) squares to refer to strong (weak) peaks in the EDC of figure 4. The overall agreement between our experimental results and the theoretical results of Christensen [1] (broken curves in figure 2) and of Eckardt *et al* [2] (full curves) is satisfactory to within about 0.3 eV, demonstrating that a free-electron-like final state as used here works well for band mapping up to photon energy of 65 eV. The experimental values of the critical points are listed in table 1, where they can be compared with both experimental and theoretical values determined by others. A better comparison of the theoretical results of Christensen [1] and of Eckardt *et al* [2] with our experimental results is made in figure 6, where the theoretical bands have been shifted downwards in energy by 0.2 eV and 0.3 eV, respectively. Figure 6 shows that the agreement is good, except perhaps in the central region between Γ and L.

Table 1. Experimental and theoretical values of critical points (in eV) of Ag at Γ and L (band indices in parentheses).

Symmetry label	Experiment			Theory	
	This work	[5]	[4]	[2]	[1]
$\Gamma_8^+(2,3)$	−6.15	−6.19	−6.23	−5.82	−5.90
$\Gamma_7^+(4)$	−5.70	−5.76	−5.80	−5.37	−5.46
$\Gamma_8^+(5,6)$	−4.95	−4.95	−4.95	−4.69	−4.75
$L_6^+(1)$	−7.10	−7.30	−7.13	−6.74	−6.94
$L_{4,5}^+(2)$	−6.25	−6.27	−6.28	−5.91	−5.99
$L_6^+(3)$	−5.80	−5.74	−5.74	−5.44	−5.53
$L_6^+(4)$	−4.55	−4.40	−4.31	−4.23	−4.20
$L_{4,5}^+(5)$	−4.30	−4.11	−4.06	−4.01	−3.97

4. Surface resonance and surface state at $\bar{\Gamma}$

4.1. Surface resonance

The shoulder (labelled s_1) on the lower-binding-energy side of the main peak in figure 4(*a*) has attributes which are characteristic of a surface state or a surface resonance. (i) The energy at which the shoulder appears is independent of the photon energy. Since the EDC were taken at normal emission this fact indicates that the s_1 feature has no dispersion with respect to $k_\perp$. (ii) The intensities of all peaks in the EDC decrease when the surface is covered with foreign atoms, but the intensity of the s_1 feature decreases more markedly. Figure 7 shows the effect of deposition of about 0.2 layers of Ti on the Ag{111} surface: the full curve is the 55 eV EDC already displayed in figure 4, while the broken curve is the EDC measured after deposition of Ti (the two curves have been scaled to equal background at −10 eV). Figure 7(*b*) depicts the difference between the two EDC and emphasises the strong decrease of the s_1 feature at −4.25 eV. This strong decrease proves that s_1 is very sensitive to the presence of impurities on the surface. (iii) There seems to be a dependence of the intensity of the s_1 feature upon the photon energy. It is apparent from figure 4(*a*) that the intensity of s_1 relative to

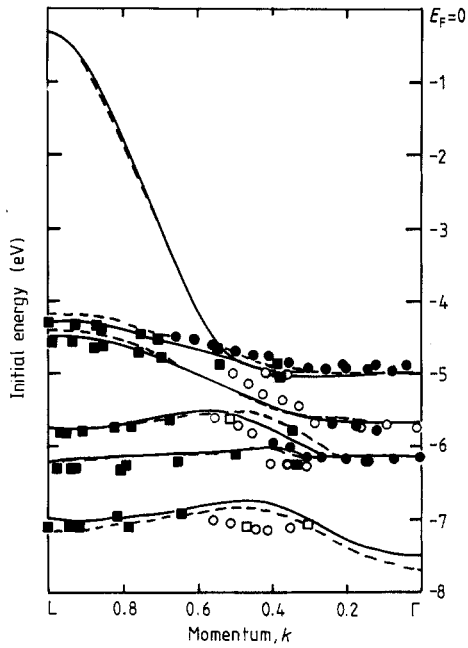

Figure 6. Initial bands of Ag along ΓΛL, as in figure 2, but the theoretical bands calculated by Christensen [1] (broken) and by Eckardt *et al* [2] (full) have been shifted downwards in energy by 0.2 and 0.3 eV, respectively.

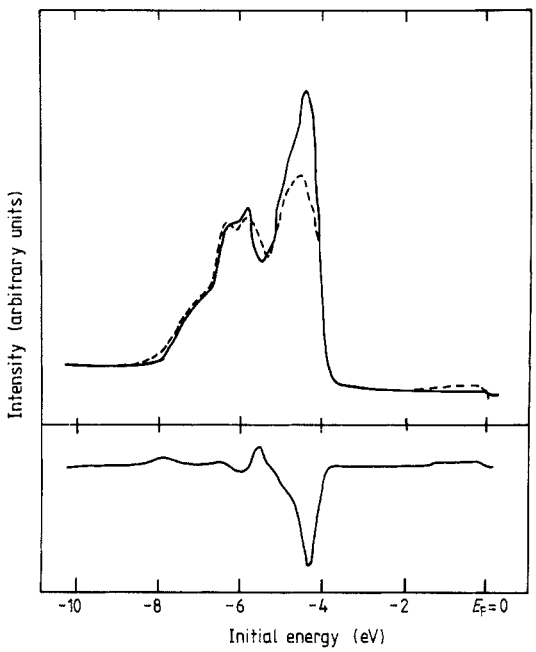

Figure 7. Effect of foreign atoms (Ti) on the 55 eV EDC from Ag{111} measured with s-polarised light. Full curve : Ag{111} clean; broken curve: Ag{111} + 0.2 layers Ti. The curve in the bottom panel is the difference between the two curves shown in the top panel.

that of the neighbouring peak (bulk peak from band 5) increases with photon energy from 35 eV, and reaches a maximum of about 55 eV. We note that the bulk peak is expected to be strongest at this photon energy (Γ point), and hence it is possible that the increase of s_1 is only apparent owing, at least in part, to the proximity of the bulk peak. However, a real maximum of s_1 at 55 eV would be consistent with the expected behaviour of surface states as originally demonstrated by Louie *et al* [9] and later discussed by others [10, 11]. The theoretical argument states that the emission from a surface state that is derived from a single band is periodic in $k_{f\perp}$ (the perpendicular component of the final-state momentum) and, in particular, should be a maximum at photon energies that correspond to the $k_{f\perp}$-value at which the energy of the bulk band is closest to the energy of the surface state. In the present case, where the latter energy is -4.25 eV and the maximum emission occurs at $h\nu = 55$ eV, the final-state energy is about 51 eV. Thus, the final-state momentum is near the L point of the bulk Brillouin zone (see §3), and it is just at the L point that the energy of band 5 is closest to s_1. We conclude that the photon energy dependence of s_1 is consistent with the description of Louie *et al* of surface state behaviour.

However, since there is no energy gap in this energy range, s_1 is more properly called a surface resonance rather than a surface state. Information about the symmetry and the origin of this surface resonance can be obtained from the dependence of its intensity upon the polarisation of the light. Comparing the EDC in figure 4 (*a*) and (*b*), we see that the intensity of s_1 is largest with s-polarised radiation and decreases when the p component of the light is increased. Thus, the corresponding wavefunction is $\Lambda_{4,5}$-like and the state is just a little above the critical point $L^+_{4,5}(5)$, (-4.30 eV in our band mapping) originating from the $\Lambda_{4,5}$ symmetry band 5. This assignment is consistent with the expectation that, in noble metals, the surface valence band is shifted toward lower binding energies with respect to the bulk [12]. We suggest that this surface resonance on Ag{111} has the same origin as the surface resonances observed on Cu{001} [6] and Ag{001} [7].

4.2. Surface state

Our data provide indirect evidence for a new surface state at $\bar{\Gamma}$ on the Ag{111} surface, which is located at 7.4 eV below the Fermi level. Figure 4(*b*) shows normal-emission EDC from Ag{111} for photon energies between 35 and 65 eV and 25% p-polarised radiation. The highest binding-energy peaks originate from band 1 (Λ_6 symmetry). After deposition of 0.2 layers of Ti, the intensity of the peak from band 1 decreases dramatically, especially on the higher-binding-energy side of the peak from band 1. Figure 8 shows an example of this effect for a photon energy of 57 eV (similar effects were found for six other photon energies, not shown here). The minimum in the difference between the EDC from Ti-contaminated and clean surfaces (bottom panel in figure 8) is at -7.4 eV and was found to be independent of photon energy. This surface-sensitive feature (labelled s_2 in figure 4(*b*)) is sensitive to the p component of the light, as a comparison between figure 4(*a*) and (*b*) shows, and its position is quite close to the bulk band 1. The energy resolution of the light and the angular resolution of our analyser are not good enough to separate the s_2 peak from the bulk peak. Experiments with highly p-polarised radiation and high energy resolution and high momentum resolution are desirable in order to determine the photon energy dependence of the intensity and the $k_{\parallel}$ dispersion of this s_2 state. We note that the energy position of this surface state is different from the surface state at $\bar{\Gamma}$ in the s–d

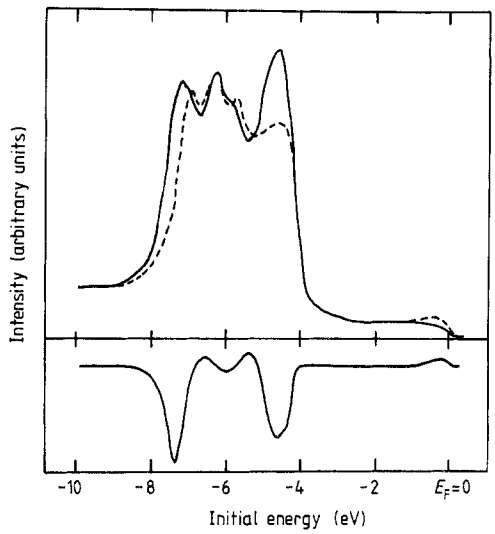

Figure 8. Effect of foreign atoms (Ti) on the 55 eV EDC from Ag{111} measured with 25% p-polarised light. Full curve: Ag{111} clean; broken curve: Ag{111} + 0.2 layers Ti. The curve in the bottom panel is the difference between the two curves shown in the top panel.

gap (above the lowest Λ_1 band) which was discovered by Louie *et al* [9]. This s_2 state is not in the s–d gap, but below the lowest Λ_6 symmetry band: its origin is not clear at this time.

5. Conclusion

Angle-resolved photoemission experiments on clean Ag{111} with s-polarised and 25% p-polarised synchrotron radiation have made possible an experimental determination of the band structure of Ag along the $\Gamma\Lambda L$ line, especially near the Γ point. For photon energies smaller than 24 eV the photoemission peaks can be interpreted as the result of direct transitions between initial and final energy bands in the relativistic band structure calculated by Christensen [1] or Eckardt *et al* [2]. In the photon energy range between 24 and 65 eV the final band was obtained by fitting a free-electron-like final band ($m_e^* = 1.10$ and $V_0 = -5.0$ eV) to experimentally verified calculated symmetry points. The dispersions and the relative positions of the initial bands are in good agreement with the calculations [1, 2] if the latter are shifted downwards in energy by 0.3 eV.

A surface resonance was detected at 4.25 eV below the Fermi level at the centre of the surface Brillouin zone. This surface resonance is believed to have the same origin as the surface resonances that have been observed on Cu{001} [6] and Ag{001} [7]. A probable surface state is also reported at about 7.4 eV below the Fermi level at $\bar{\Gamma}$, but the origin of this state is not understood at the present time.

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