Photoemission of spin-polarized electrons from tungsten: Theoretical predictions*

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We have calculated the spin-polarized spectrum of electrons photoemitted by circularly polarized light in tungsten. Results are given for free and cesiated surfaces along the [100] and [111] directions. In the [110] direction there is no spin polarization. The calculation is based on the three-step model and involves a group-theoretical analysis of the direct optical excitations and their joint density of states. 100% polarization is predicted for certain photon energy regions.

INTRODUCTION

In the photoemission process induced by circularly polarized light in nonmagnetic targets the spin-orbit interaction couples the light angular momentum to the electron spin. Under suitable conditions this coupling leads to the photoemission of spin-polarized electrons. In isolated atoms (in a gas) this process is well understood, both theoretically and experimentally.[1-5]

The photoemission of spin-polarized electrons in solids has been observed in the semiconductor GaAs,[6,7] and in the heavy alkali metals.[8] In solids a calculation of comparable accuracy to that of isolated atoms is not possible at present because of the complexity of the photoemission process itself.[9] Only qualitative predictions within the three-step model have been put forward.[10,11] They are based on the group-theoretical analysis of the direct optical excitations at special points or along symmetry lines in the Brillouin zone. In GaAs the main peaks in the spin-polarization spectrum (SPS) can be identified with the onset of the direct optical transitions from the valence to the conduction band at the Γ and L points. In the alkali metals a comparison of the theoretical predictions with the available experimental results is not possible because the experiments were performed on polycrystalline samples.

In this paper the same theoretical model[10] is used to calculate the SPS for electrons excited along the [100] and [111] directions in tungsten. Electrons excited along the [110] direction are completely unpolarized. For the purpose of comparing theoretical predictions with experimental data tungsten offers definite advantages over the alkali metals: (i) It is well known how to prepare crystallographically oriented clean surfaces,[12] and (ii) fully relativistic band-structure calculations are available.[13]

Recently there has been a growing interest in obtaining new sources of spin-polarized electrons, since they provide a unique tool to probe spin-dependent effects in any scattering process.[7] In this sense tungsten has been described as a promising material to produce spin-polarized electrons by low-energy-electron diffraction .[14] The results obtained here show that the photoemission could be an alternative: 100% polarization is predicted for emission along the [100] and [111] directions in certain energy ranges of the incident light. Furthermore, the polarization of the electron beam could be modulated either by modulating the circular polarization of the light or its energy.

MODEL

We calculate the bulk contribution to the SPS for the following geometry: The circularly polarized light impinges normally on the (100) or the (111) surfaces. The electrons are supposed to be collected in a solid angle around the normal, narrow enough to assure that they have been excited along the corresponding symmetry direction in the Brillouin zone. (This is an energy integrated but angular-resolved spin-polarization measurement.) The electron spin is quantized along the normal, which defines the z axis. We assume the validity of the three-step model[15] and restrict ourselves to the consideration of the first step, that is, the direct optical excitation.[16] The second and third steps, namely, the transport of the excited electron from its production site to the surface and the subsequent transmission through the surface, should not affect appreciably the initial polarization.[7] We try to derive as much information as possible about the SPS without considering explicitly the radial part of the wave functions involved in the transition except for the usual assumption that they lead to transition matrix elements that are slowly varying functions of energy. The calculations are based on the relativistic band structure calculation of Feuerbacher and Christensen.[13]

The SPS is calculated in the photon energy range $1.5 \lesssim \hbar\Omega \lesssim 11.0$ eV. The upper limit is set by the impossibility of producing circularly polarized light of higher energies by conventional means.[17] The lower limit is the work function of the cesiated tungsten surfaces.[18] Predictions are made for both free and cesiated surfaces.

We also discuss the contribution to the SPS coming from the surface state on the (100) surface of tungsten.[19-21]

CALCULATION

The single group representations for the band along the symmetry directions were obtained from the nonrelativistic band structure calculation of Mattheiss.[22] The wave functions that transform according to the irreducible representations contain components of several angular momenta. The angular part of these components can be written as linear combinations of spherical harmonics by the use of the projection operator technique.[23] In the dipole approximation the only allowed transitions are those between components of wave functions whose angular momenta differ by 1. With the introduction of the spin-orbit interaction, and therefore of the spin variables, the wave functions should transform according to the double group representations of the crystallographic point group. The proper linear combinations of products of angular and spin wave functions can be constructed by the use of the coupling coefficients tabulated by Koster *et al.*[24] They are shown in Table I. In this calculation we take into account only the s, p, and d parts of the wave functions. This is consistent with the fact that we are dealing with bands arising from the $5d$ atomic states of tungsten or from the hybridization of one of these bands with the broad band arising from the $6s$ atomic states (s-p band).

TABLE I. Symmetrized linear combinations of products of angular and spin functions transforming according to the double group representations of the crystal point group along the [100] and [111] directions. The irreducible representations are labeled according to BSW[a] notation as follows: The subscript indicates the double group representation, the superscript the single group representation from which they are derived. $\alpha = |\uparrow\rangle$ and $\beta = |\downarrow\rangle$.

s part	p part	d part
	[100] direction	
Δ_6^1 α, β	$z\alpha,\ z\beta$	$(3z^2-r^2)\alpha,\ (3z^2-r^2)\beta$
Δ_6^5 ···	$(x+iy)\beta,\ (x-iy)\alpha$	$z(x+iy)\beta,\ z(x-iy)\alpha$
Δ_7^5 ···	$(x-iy)\beta,\ (x+iy)\alpha$	$z(x-iy)\beta,\ z(x+iy)\alpha$
Δ_7^2 ···	···	$(x^2-y^2)\alpha,\ (x^2-y^2)\beta$
$\Delta_7^{2\prime}$ ···	···	$xy\alpha,\ xy\beta$
	[111] direction	
Λ_6^1 α, β	$z\alpha,\ z\beta$	$(3z^2-r^2)\alpha,\ (3z^2-r^2)\beta$
Λ_6^3 ···	$(x+iy)\beta,\ (x-iy)\alpha$	$z(x+iy)\beta,\ z(x-iy)\alpha$
Λ_4^3 ···	$(x-iy)\beta - i(x+iy)\alpha$	$z(x-iy)\beta - i(x+iy)\alpha$
Λ_5^3 ···	$i(x-iy)\beta - (x+iy)\alpha$	$iz(x-iy)\beta - z(x+iy)\alpha$

[a] L. P. Boukaert, R. Smoluchowsky, and E. P. Wigner, Phys. Rev. 50, 58 (1936).

TABLE II. Electron spin polarization for the transitions of interest. Initial bands are on the left, final bands on the top. The Λ_5^3 bands are taken together because they are degenerate due to time reversal symmetry. $\alpha = |\uparrow\rangle$ and $\beta = |\downarrow\rangle$ indicate the sign of spin produced in a given transition. The dots indicate that there are no transitions for those symmetries.

	Δ_6^1	Δ_7^2	$\Delta_7^{2\prime}$	Δ_6^5	Δ_7^5		Λ_6^1	Λ_6^3	$\Lambda_4^3+\Lambda_5^3$
Δ_6^1	···	···	···	α	β	Λ_6^1	···	α	β
Δ_7^2	···	···	···	β	α	Λ_6^3	β	···	···
Δ_7^2	···	···	···	β	α				
Δ_6^5	β	α	α	···	···	$\Lambda_4^3+\Lambda_5^3$	α	···	···
Δ_7^5	α	β	β	···	···				

For a given transition the expected value of the spin of the excited electron can be obtained using the wave functions of Table I and the angular part of the electron-photon interaction Hamiltonian which is proportional to $x - iy$ ($x + iy$) for right- (left-) circularly-polarized light propagating in the $-z$ direction. The results are shown in Table II. These results show that the only allowed transitions are those coming from or going to spin-orbit split bands for which the radial parts of the wave functions are the same. (One of the transitions produces spin-up electrons and the other spin-down electrons.) Therefore, the partial contribution to the SPS coming from such a pair of transitions can be calculated from the (one-dimensional) joint densities of states

$$\zeta(\hbar\Omega) = \int dk\, \delta(\epsilon_f(k) - \epsilon_i(k) - \hbar\Omega)\, \Theta(e_F - \epsilon_i(k))\Theta(\epsilon_f(k) - e_{\mathrm{VAC}}),$$

as follows:

$$P_p = \frac{\zeta_\uparrow(\hbar\Omega) - \zeta_\downarrow(\hbar\Omega)}{\zeta_\uparrow(\hbar\Omega) + \zeta_\downarrow(\hbar\Omega)}.$$

$\epsilon_f(k)$ and $\epsilon_i(k)$ are the electron energies in the final and initial bands, e_F is the Fermi energy, and e_{VAC} is the energy of the vacuum level. Θ is the step function and the integral is performed on the

wave vector k along a given direction from the center to the boundary of the Brillouin zone. $\zeta_\uparrow(\hbar\Omega)$ and $\zeta_\downarrow(\hbar\Omega)$ are the joint densities of states for transitions producing spin-up and spin-down electrons, respectively.

There are ranges of photon energies where only a single pair of transitions contribute. In these regions the SPS is given directly by P_p.

In those photon energy ranges where two pairs contribute the SPS depends on the radial parts of the wave functions involved. However in two cases it is still possible to make qualitative predictions: (i) When a pair of transitions occurs between an sp-d-like band and a d-like band while the other pair corresponds to a transition occurring between two d-like bands, it is reasonable to guess that the former pair contributes overwhelmingly to the SPS; therefore, the SPS is given approximately by the P_p of the former pair. (ii) When the relative contribution of the two pairs cannot be guessed from symmetry considerations, there are still photon energy ranges where the joint density of states of one pair is much larger than that of the other. In this case the SPS structure would follow that of the P_p of the pair with the larger joint density of states.

Figure 1 shows examples of two of these cases for transitions along the [100] direction. The band structure was taken from Ref. 13. The small arrows show transitions from the spin-orbit-split Δ_6^5 and Δ_7^5 bands to the Δ_6^1 band. In the photon energy range of this pair of transitions there is no other competing pair. The big arrows show two pairs of competing transitions: a pair of transitions from the Δ_6^1 band to the Δ_6^5, Δ_7^5 spin-orbit-split bands and the pair from the Δ_7^2 band to the same spin-orbit-split bands. However, the Δ_7^2 band is mainly d-like as well as the Δ_6^5 and the Δ_7^5, while the Δ_6^1 band, as produced by sp-d hybridization, should be not only d- but also sp-like. Thus the contribution of the transitions going from the Δ_6^1 band should be larger than that for the transitions going from the Δ_7^2 band.

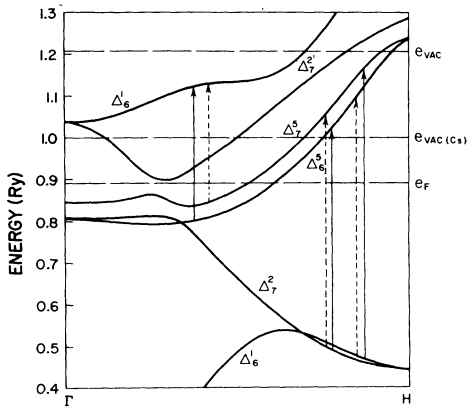

FIG. 1. Band structure of W along the [100] direction (after B. Feuerbacher and N. E. Christensen, Ref. 13), showing examples of transitions where a single pair (small arrows) and two pairs (big arrows) contribute to the SPS. e_{VAC} and $e_{VAC(Cs)}$ are the vacuum levels for the free and cesiated surfaces, respectively. The dashed arrows indicate transitions producing spin-up electrons. The continuous arrows indicate transitions producing spin-down electrons.

RESULTS

Figures 2(a) and 2(b) show the predicted SPS for cesiated surfaces in the directions [100] and [111], respectively. The portions of the curves to the left of the arrows originate from transitions between d-like bands, thus their intensities should be small. The dashed portions of the curves correspond to a region where two pairs of transitions contribute and thus, they represent a best guess based on the above considerations about symmetry [Fig. 2(a)] and joint density of states [Fig. 2(b)]. The main peaks are produced by the indicated transitions.

Figures 3(a) and 3(b) show the predicted SPS for

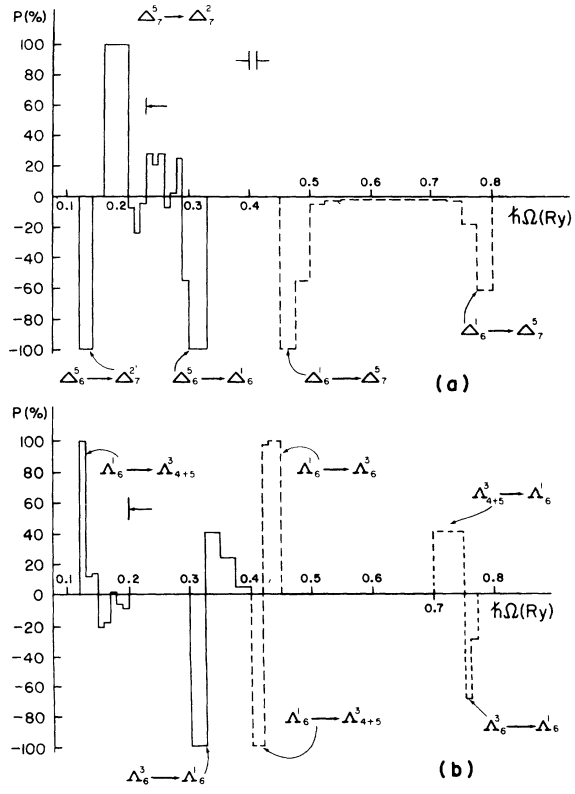

FIG. 2. Spin-polarized spectrum for cesiated W: (a) for (100) surface, (b) for (111) surface.

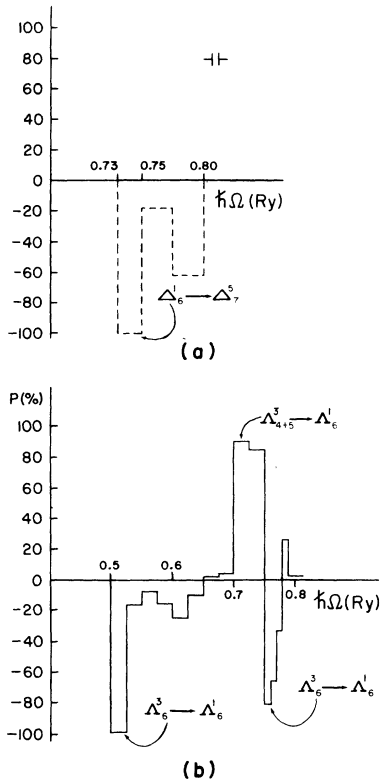

FIG. 3. Spin polarized spectrum of clean W: (a) for (100) surface, (b) for (111) surface.

free surfaces in the directions [100] and [111], respectively. The dotted lines have the same meaning as those in Fig. 2.

DISCUSSION

The predicted SPS may be smoothed by the finite lifetime of the final states and by spin-flip scattering processes during electron escape. A broadening in the final-state bands would produce a broadening in the SPS peaks and a decrease in their heights due to additional overlapping of transitions leading to spin-up and spin-down electrons. A crude estimate based on a typical mean free path of $\lambda \approx 20-50$ Å for hot electrons with energies of 5 eV above E_F gives a broadening of the order of 0.01 Ry. This energy uncertainty is indicated in Figs. 2 and 3. Spin-flip scattering processes may decrease the heights of the peaks. However, this effect should be small even for a cesiated surface according to the results obtained for GaAs.[7]

One of the most prominent features of the photoemission spectrum of tungsten is the peak observed at 0.4 eV below the Fermi level on the (100) face. This has been assigned to a surface state.[19] There is still controversy regarding the origin of this surface state: (i) It may be produced by spin-orbit effects or (ii) it may already exist in the *sp-d* gap, independently of spin-orbit effects. In each case the symmetry of its wave function must be different.[25] In case (i) the symmetry of the wave function must be of the Δ_7 type (since it originates from the Δ_7^5, Δ_7^2, and $\Delta_7^{2'}$ bands). In case (ii), on the other hand, the symmetry must be of the Δ_6 type (since it originates from the hybridized Δ_6^1 bands). The final state is assumed to be a plane wave with wave vector along the z direction and therefore with Δ_6^1 symmetry. The symmetry of the wave functions is enough to conclude that in case (i) the photoemitted electrons will be spin polarized, while in case (ii) they will not. In case (i) only the *s* part of the incident light may produce an electron excitation and in order to have photoemission this process should be assisted by scattering mechanisms providing momentum normal to the surface. In case (ii) only the *p* part of the incident light leads to photoemission. In view of the experimental results of Feuerbacher and Fitton[26] we conclude that the surface state belongs to case (ii) and therefore no spin polarization is expected from this peak.

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