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# Energy-distribution studies of photoexcitation in photofield emission

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Total energy distributions of the photocurrent in photofield emission from tungsten have been measured for *s*- and *p*-polarized light. The results show that for photon energies below 3.5 eV the dominant mechanisms of photoexcitation are the surface photoeffect and the indirect bulk photoeffect. In the photofield-emission configuration the surface and indirect bulk distributions can be separated by suitably choosing the polarization and angle of incidence of the illumination.

Recent experimental advances have focused attention on the spectroscopy of electronic states of a metal in the energy range between the Fermi level and the vacuum level. Photofield emission has been used by several groups to study emission from electronic states in this energy range,[1-3] and there has been a growing interest in $\vec{k}$-resolved inverse photoemission spectroscopy.[4-6] The purpose of this Communication is to report the first observation of a dependence of the total energy distribution in photofield emission on the polarization of the incident light. The results show that, for the range of photon energies used in photofield emission, the indirect bulk photoeffect and the surface photoeffect are the predominant excitation mechanisms in tungsten.

The sample is a field emitter on which the major crystallographic planes form distinct facets, making it possible to study emission from many different planes with a single sample. A large static potential difference is applied between the field emitter and a fluorescent screen, producing a strong electric field at the surface of the metal, which lowers the surface potential barrier by the Schottky effect. This allows electrons to tunnel from the Fermi distribution to the vacuum, where they form a field emission pattern on the fluorescent screen. The field emitter is illuminated by a focused laser beam with a photon energy less than the work function of the metal. Electrons emitted from any desired crystal plane pass through a probe hole in the screen and enter a retardation energy analyzer, which selects the photoexcited electrons from the total emission current and measures the photocurrent and its total energy distribution.[3] Because the transmission coefficient of the Schottky barrier decreases exponentially with increasing transverse electron momentum, photofield emission from a given single-crystal plane involves electron states in a small region of $\vec{k}$ space close to the surface normal. Photofield emission is thus equivalent to a $\vec{k}$-resolved technique in which $\vec{k}$ is confined to directions normal to the crystal planes developed on the field emitter.

Recent experiments[7] have shown that the vectorial photoeffect observed in photoemission[8,9] is also present in photofield emission. It was found that in photofield emission the polarization dependence of the total emission current $J$ from any crystal plane of tungsten can be expressed as

$$J = A \sin^2(\gamma - \gamma_s) + B \ , \tag{1}$$

where $A$ and $B$ are nonzero and vary from crystal plane to crystal plane, and $\gamma$ is the polarizaton angle of the incident light ($\gamma_s$ corresponds to a *s*-polarized light). To interpret the polarization dependence, the electromagnetic field configuration at the surface of each facet of the field emitter was modeled by the reflection of a plane wave at an infinitely extended plane interface. Prompted by the experimental polarization dependence, Eq. (1), the photocurrent was assumed to contain a component $J_1 \propto (\vec{\epsilon}^{\,t} \cdot \hat{z})^2$ dependent on the projection of the polarization vector inside the metal $\vec{\epsilon}^{\,t}$ along the surface normal $\hat{z}$, and a component $J_2 \propto |\vec{\epsilon}^{\,t}|^2$ independent of the direction of polarizaton inside the metal. The predictions of the plane interface model were found to agree with the observed dependence of the photocurrent on the polarization angles $\gamma$ and $\gamma_s$, and with the dependence of $A$ and $B$ on the angle of light incidence.

The polarization dependence of $J_1$ is that expected for surface photoexcitation[10] associated with the potential gradient in the Schottky barrier. It is also consistent with bulk photoexcitation due to direct transitions involving a reciprocal-lattice vector parallel to the surface normal.[11] The polarization dependence of $J_2$ is consistent with bulk photoexcitation due to indirect transitions.[7] Thus the experimental confirmation of the plane interface model suggests that at least two distinct photoexcitation mechanisms contribute to the photocurrent. This can be tested by measuring the total energy distributions of $J_1$ and $J_2$. The distributions $j_1(E)$ and $j_2(E)$ (with the normalization appropriate to *p*-polarized light) can be deduced by measuring the total energy distributions for *s*- and *p*-polarized light and combining them according to[12]

$$j_1(E) = j_p(E) - |T_p/T_s|^2 j_s(E) \ , \tag{2}$$

$$j_2(E) = |T_p/T_s|^2 j_s(E) \ , \tag{3}$$

where $T_s$ and $T_p$ are the transmission coefficients of the electric field for *s*- and *p*-polarized light at a plane interface. The ratio $|T_p/T_s|^2$, determined from Fresnel theory of re-

 

fraction, is a function of the angle of light incidence $\Psi$ and the complex index of refraction $\tilde{n}$.[7] The observation of significantly different total energy distributions would be strong evidence that these two terms do indeed arise from distinct mechanisms of photoexcitation.

The total energy distributions for *s*- and *p*-polarized light were recorded at small angles of incidence on the (111) crystal plane of tungsten, for surface electric field strengths of 1.35, 1.43, 2.12, and $2.51\times10^{9}$ V m$^{-1}$. Ultraviolet light (a closely spaced doublet with photon energies 3.48 and 3.53 eV) was focused onto the field emitter. The luminous intensity was typically 2 MW m$^{-2}$. The principal experimental difficulty was to minimize any differences between the distributions that might be caused by gradual tip contamination and slow drifts in the tip alignment, or by polarization-dependent heating of the tip by the laser beam. To compensate for drifts, the plane of linear polarization of the laser beam was switched rapidly between the *s* and *p* states by means of a Pockels cell, and the total energy distributions were accumulated in alternate channels of a multichannel analyzer. The resolution function of the energy analyzer (whose full width at half maximum is approximately 200 mV) was determined in a separate experiment, and was deconvolved from the observed distributions.[3] A small correction was required to allow for the greater tip heating that occurs when the light is polarized parallel to the shank of the field emitter.[13] The resulting distributions were combined according to Eqs. (2) and (3) to yield the distributions $j_1(E)$ and $j_2(E)$.

Figure 1(a) shows a representative pair of total energy distributions $j_1(E)$ and $j_2(E)$. As a consequence of the overriding influence of the barrier transmission function, the two distributions have the same general shape. To facilitate comparison, they were scaled to the same area and then subtracted. This yields the difference distribution

$$\Delta(E)=j_2(E)-Cj_1(E)\ , \tag{4}$$

where

$$C=\int j_2(E)dE \Big/ \int j_1(E)dE\ . \tag{5}$$

If $j_1(E)$ and $j_2(E)$ arise from the same mechanism, then $\Delta(E)$ will be zero within experimental uncertainty. Figure 1(b) shows the difference distribution for the data of Fig. 1(a). It is negative at high energy (near $E_F+\hbar\omega$) and positive at lower energy. This characteristic energy dependence is reproduced at the other field strengths.

The field dependences of $j_1(E)$ and $j_2(E)$ were also compared. Experimental values of the scaling factor $C$ obtained from the total energy distributions measured at four different field strengths are given in Table I. The fifth value is derived from measurements of the total photocurrent. The systematic field dependence of $C$ offers additional evidence that $j_1(E)$ and $j_2(E)$ have different shapes. Since the probability of transmission through the surface barrier is more sensitive to field at low energy than at high energy, low-energy electrons make the major contribution to the field dependence of $C$. The observed increase in $C$ with increasing field strength indicates that $j_2(E)$ is peaked at a lower energy than $j_1(E)$. Taken together, the experimental energy and field dependences of $j_1(E)$ and $j_2(E)$ provide strong evidence that these two components of the photocurrent arise from different mechanisms of photoexcitation, and that $j_2(E)$ is more heavily weighted to low energy than $j_1(E)$.

It is possible to identify the excitation mechanisms responsible for the two components of the photocurrent by combining the polarization and total energy distribution results. The polarization dependence of $J_1$ is consistent either with the surface photoeffect or with the direct bulk photoeffect arising from transitions involving the reciprocal-lattice vector parallel to the surface normal. In surface photoexcitation only the component of the crystal momentum of the electron parallel to the surface is conserved. This makes for an abundance of final states, many of them evanescent. The total energy distribution is expected to be a broad triangular distribution, peaked near the energy $E_F+\hbar\omega$, falling sharply at higher energy because of the Fermi-Dirac cutoff in the occupation of initial states, and falling gradually at lower energy because of the increasing thickness of the Schottky barrier.[10] These are just the characteristics of $j_1(E)$. Transitions caused by direct bulk photoexcitation are severely restricted by the conservation of energy and momentum, especially at the small energies

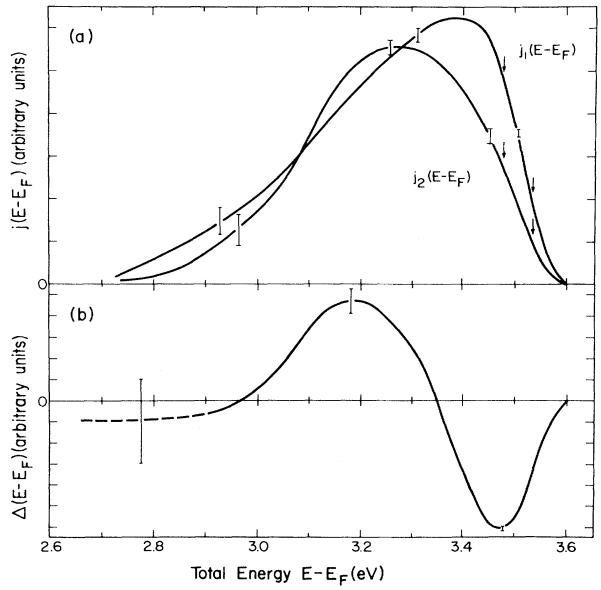


FIG. 1. (a) Total energy distributions $j_1(E)$ and $j_2(E)$ measured at a field strength of $2.12\times10^{9}$ V m$^{-1}$. The arrows mark the energies $E_F+\hbar\omega$. The error bars indicate the uncertainty due to shot noise in various regions of the curves. (b) Difference distribution $\Delta(E)$ for the two distributions shown in (a).

TABLE I. Scaling factor for a range of static surface electric field strengths.

| Electric field (units of $10^{9}$ V m$^{-1}$) | $C$ |
|---|---|
| 1.35 | 0.70(2) |
| 1.74 | 0.80(2) |
| 2.12 | 0.84(2) |
| 2.51 | 0.90(3) |
| 2.70 | 1.00(3) |

involved in photofield emission. Therefore the total energy distribution consists of, at most, a few narrow peaks at those energies at which vertical transitions can occur.[11] The broad asymmetrical peak in $j_1(E)$ is not of this form. Furthermore, direct bulk transitions consistent with the observed peak at 3.40 eV must occur between initial and final states in a narrow region of $k$ space close to the surface normal. The band structure of tungsten along the ⟨111⟩ direction, calculated from the potential $V_2$ of Christensen and Feuerbacher,[14] is shown in Fig. 2. The band structure indicates that for a photon energy of 3.5 eV there are no initial states along ⟨111⟩ from which one-photon transitions to final states near $E_F+\hbar\omega$ can occur. Small uncertainties in the relative positions of the energy bands do not alter this conclusion. Therefore the total energy distribution measurements are inconsistent with direct bulk photoexcitation and indicate that the polarization-dependent component of the photocurrent is caused by the surface photoeffect.

The polarization dependence of $J_2$ is consistent with the indirect bulk photoeffect. The total energy distribution $j_2(E)$ is broad, as would be expected from the relaxed requirements of $\vec{k}$ conservation for indirect transitions, and it is peaked below $E_F+\hbar\omega$. The band structure of tungsten in Fig. 2 shows that indirect bulk transitions to final states in the (111) direction close to $E_F+\hbar\omega$ can occur only to one relatively steep band. At lower energy, however, two more bands can provide final states. These bands have a Van Hove singularity at the symmetry point $P$ at energy $E-E_F=3.15$ eV, which is consistent with the experimental peak of $j_2(E)$ observed at $E-E_F=3.25$ eV. Electrons in states near $P$ have a large probability of emission since their transverse momenta are very small and their energies are close to the peak of the Schottky barrier. These critical points represent a high density of final states for indirect bulk transitions, and are believed to be the cause of the prominent peak in $J_2(E)$.

The present results demonstrate that, in the energy range investigated here, photofield emission from tungsten is dominated by surface and indirect bulk excitations. A striking feature is the absence of significant direct bulk photoemission, which is attributable to the very small joint density of states for direct transitions from just below the Fermi level for photon energies below 3.5 eV.[7] This is in contrast to studies based on the inverse photoelectric effect which involve much larger photon energies, and in which direct bulk transitions and surface transitions to states just above the Fermi level dominate.[5,6] In this sense, photofield emission and the inverse photoelectric effect are complementary techniques. The present work demonstrates that, in the photofield-emission configuration, indirect bulk transitions and surface transitions can be distinguished by selecting the polarization and angle of incidence of the incident light. This opens up the possibility of studying separately indirect bulk transitions and surface transitions involving electron states between the Fermi level and the vacuum level.

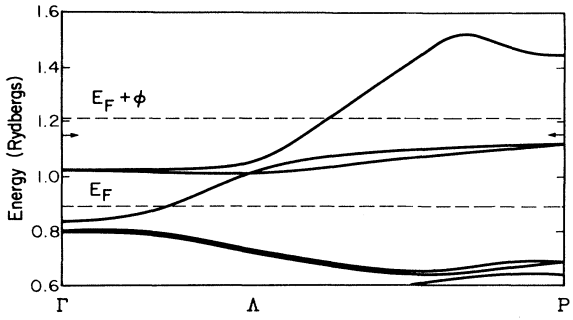


FIG. 2. Band structure of tungsten in the ⟨111⟩ direction. The dotted lines mark the Fermi energy and the vacuum level. The arrow indicates the energy $E_F+\hbar\omega$.

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