Angle-of-Incidence Dependence of Photoemission of a Localized Electron from a Jellium Solid*

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We show via a calculation for a localized electron in a jellium solid that an escape-cone mechanism accounts for a significant component of the peak seen by Rowe and Christman in photoyield data as a function of angle of incidence for Ar embedded in Ge(111) and Si(111) surfaces.

In this Comment we present a calculation of the angle-of-incidence dependence of photoemission of localized electrons from a jellium solid, and compare the results to the experimental angle-of-incidence dependence of photoemission from Ar atoms embedded in Ge(111) and Si(111) reported by Rowe and Christman.[1,2] This angular dependence, in which the photoyield appears to vary approximately in proportion with a power somewhat greater than 2 of the electric field normal to the surface, has until now been supposed to be largely due to strong local electric fields.[2] While local field effects in general should play a role in photoemission, we show here that an escape-cone mechanism can explain a large component of the angle-of-incidence dependence observed by Rowe and Christman.[1,2] Similar mechanisms have recently been discussed by Schaich,[3] Sass,[4] and Munz[5] for several other somewhat different systems (e.g., interband transitions in Ag,[3] internal photoemission at a Au-electrolyte interface,[4] and near-threshold emission from EuO[5]).

The basic idea is that for an electron to be ejected from a solid by a relatively low-energy photon, the excited electron must have a large proportion of its momentum normal to the surface. Thus, if the photoemission maxtrix element contains a term of the form $\vec{\mathcal{E}}\cdot\vec{p}$, where $\vec{\mathcal{E}}$ is the electric field and $\vec{p}$ is the electron momentum, the component of $\vec{\mathcal{E}}$ normal to the surface ($\mathcal{E}_\perp$) will be most effective in ejecting an electron, and it would not be surprising to find the photoyield proportional, approximately, to the square of $\mathcal{E}_\perp$.

In order to obtain a quantitative estimate of this escape-cone effect, we consider a three-step model[6,7] of photoemission from a localized level of an atom embedded in a jellium solid. (We later specialize to the case of the $3p$ level of Ar embedded in Ge.) The basic idea of the three-step model is that the photoexcitation matrix element can be calculated as if the sample had no surface. Thus for photoemission from an atomic level, we assume the square of the matrix element (summed over magnetic levels) to be of the form dictated by rotational invariance,[8]

$$a|\vec{\mathcal{E}}|^2+b(\vec{\mathcal{E}}\cdot\vec{p})^2/|\vec{p}|^2, \tag{1}$$

where a and b are constants (which depend on the photon energy, $\hbar\omega$), and where $\vec{p}\equiv(\vec{p}_\parallel, p_\perp)$ is here taken to be the electron momentum *inside* the sample. Note that $p_\perp$ is given by $p_\perp\equiv[2m(E+V_0)/\hbar^2-p_\parallel^2]^{1/2}$, where E is the electron energy (referred to the vacuum level) and V_0 is the sample's inner potential.

Under the assumption of a homogeneous distribution of Ar atoms within a few times the escape depth, λ, of the surface,[1] the three-step model then yields the following formula[7] for the angle-integrated photocurrent, J:

$$J\propto\int_0^\infty dD\int d^2p_\parallel\,\frac{\theta(2mE/\hbar^2-p_\parallel^2)}{(2mE/\hbar^2-p_\parallel^2)^{1/2}}\exp\left(\frac{-2Dp}{\lambda p_\perp}\right)\left(a|\vec{\mathcal{E}}|^2+\frac{b(\vec{\mathcal{E}}\cdot\vec{p})^2}{|\vec{p}|^2}\right). \tag{2}$$

The step function in Eq. (2), $\theta(2mE/\hbar^2-p_\parallel^2)$, represents the escape-cone effect; it requires that the photoelectron have a normal component of momentum outside the sample which is greater than or equal to zero. The exponential factor represents the inelastic damping of the photoelectrons in transport to the surface from depth D.

The integral of Eq. (2) can be carried out straightforwardly, leading to a final formula of the form

$$J \propto |\vec{\mathcal{E}}|^2 + \gamma(b/a, E/V_0)\mathcal{E}_\perp^2, \quad (3)$$

where the function $\gamma(\alpha,\epsilon)$ is given by

$$\gamma(\alpha,\epsilon) = \frac{\alpha}{2}\,\frac{\frac{1}{2}[\epsilon(1+\epsilon)]^{1/2}(\epsilon+\frac{11}{2}) + (\frac{5}{4}-\epsilon)\ln[(1+\epsilon)^{1/2}+\sqrt{\epsilon}]}{[\epsilon(1+\epsilon)]^{1/2}[1+\epsilon+\alpha(\epsilon-\frac{1}{2})/4] + [1+\epsilon+\alpha(\epsilon+\frac{1}{4})/2]\ln[(1+\epsilon)^{1/2}+\sqrt{\epsilon}}\,. \quad (4)$$

Thus in order to compare the three-step model to the data in Refs. 1 and 2, we need only specify the appropriate values of $\alpha \equiv b/a$ and $\epsilon \equiv E/V_0$. Note that our anisotropy parameter α is related in a simple way to the anisotropy parameter β commonly referred to in gas-phase photoemission work,[8] i.e., $\beta = 2\alpha/(\alpha+3)$.

In general, α can assume any value between -1 and ∞ (corresponding to the limitations[8] on β of $-1<\beta<2$). The maximum value of α, i.e., $\alpha = \infty$, is achieved for photoemission of an s electron; for higher values of orbital angular momentum l, α is large if the final electron wave function is nearly a plane wave, i.e., if its orthogonalization to core states is a small effect.

For the sake of an estimate, it is presumably reasonable to use the experimental value of α for gas-phase Ar, measured at the same photoelectron kinetic energy as in the experiment.[1,2] This energy, which is just what is called E above, is equal to the photon energy, 21.2 eV, minus the embedded Ar's $3p$ ionization potential, found for Ar embedded in Ge by Rowe[2] to equal 13.4 eV. Thus E is equal to 7.8 eV. Experimental as well as theoretical values of b/a have been tabulated by Manson.[8] For $E = 7.8$ eV, and for the $3p$ level of gas-phase Ar, the general consensus is that $\alpha \equiv b/a \approx 3.7$ (i.e., $\beta \simeq 1.1$). At the same time, assuming an inner potential for Ge of 17.5 eV, one has that $\epsilon \equiv E/V_0 \approx 0.45$ and, using these values of α and ϵ, one finds that $\gamma(3.7, 0.45) = 1.9$. A plot of photoyield versus angle of incidence, using this value of γ,[9] is compared to the data of Ref. 2 in Fig. 1. Curves for maximal anisotropy, i.e., for $\gamma(\infty, 0.45) = 6.9$ ($\beta = 2$), and for an anisotropy somewhat lower than the gas-phase anisotropy, i.e., for $\gamma(1.6, 0.45) = 0.95$ ($\beta \simeq 0.7$), are also shown, in order to give an idea of how sensitive the escape-cone mechanism is to the choice of the anisotropy parameter $\alpha = b/a$.

The curves of Fig. 1 make it quite clear that the escape-cone mechanism must play an important role in explaining the approximate proportionality of photoyield and $\mathcal{E}_\perp^2$ reported in Refs. 1 and 2. For all three values of the anisotropy parameter, this mechanism yields a peak in the predicted intensity versus θ_i at a value of $\theta_i \sim 50$–$55°$, i.e., roughly the experimental value. For the anisotropy corresponding to the gas-phase value of b/a (curve 2 in Fig. 1), the intensity at the maximum is about 50% higher than that at $\theta_i = 0°$, and for the maximal anisotropy it is about 3 times the $\theta_i = 0°$ value.

On the low-θ_i side of the maximum, the experimental points do seem to be fitted best by the maximal-anisotropy ($b/a = \infty$) curve; and the

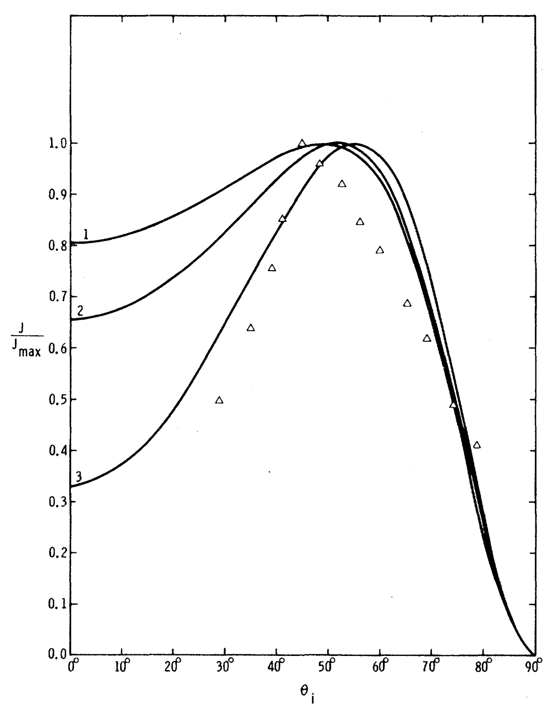

FIG. 1. Comparison of experimental and theoretical curves for angle-of-incidence dependence of photocurrent for photoemission of $3p$ electrons from Ar embedded in a Ge(111) surface. The experimental points (triangles) are from Ref. 2. The theoretical curves 1–3 were calculated for anisotropy parameters (b/a) equal to 1.6, 3.7 (the gas-phase value), and ∞, respectively, i.e., gas-phase β parameters of $\beta = 0.7$, 1.2, and 2.0. The Ge inner potential V_0 was taken to be 17.5 eV, the ionization potential of the Ar atom was 13.4 eV (Ref. 2), the photon energy $\hbar\omega$ was 21.2 eV, and the dielectric constant of Ge was $0.85 + 0.40i$ (Ref. 9).

width of this curve also seems closest to the data. However, the experimental points clearly fall off more rapidly than the theoretical curves (even for $b/a=\infty$) as θ_i becomes small.[10] This effect may in part be an artifact of uncertainties associated with background subtraction in computing the area under the Ar resonance in the experimental spectra; these uncertainties are, of course, greater at those values of θ_i for which the Ar resonance is less pronounced.[11] On the other hand, this rapid falloff of J at smaller values of θ_i may be due to local field effects as suggested in Refs. 1 and 2.

In summary, we have quantitatively estimated the angle-of-incidence dependence caused by the escape-cone mechanism in photoemission of localized electrons from a jellium solid, and have shown that this mechanism (although not the only one) plays a significant role in explaining data for Ar embedded in Ge and Si. In future related work it would be useful to see experimental measurements extended down to smaller values of θ_i and over a wider range of photon energies $\hbar\omega$. It would be particularly useful to gain an understanding of the difference between $\beta(\hbar\omega)$ for a gas-phase atom versus an atom embedded in a solid. Having such an understanding one could test our idea that the escape-cone mechanism is an important factor in the θ_i dependence of $J(\theta_i)$. If our picture is correct then the height of the peak in $J(\theta_i)$ should diminish considerably and then saturate to a constant value [in a manner given by Eq. (4)] as $\hbar\omega$ increases, apart from changes in β with $\hbar\omega$.

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[9]$\mathcal{E}_\perp^2$ and $|\mathcal{E}|^2$ were calculated classically by solving for the fields which arise when an electromagnetic wave impinges from the vacuum upon a flat semi-infinite dielectric of dielectric constant $0.85+0.40i$. This the experimental value of the dielectric constant for Ge at 21.2 eV. [See E. T. Arakawa, R. N. Hamm, and M. W. Williams, J. Opt. Soc. Am. 63, 1131 (1973)].

[10]This effect is even more pronounced in the Si data reported in Ref. 1.

[11]In Ref. 1, in fact, it has already been suggested that experimental uncertainties may be responsible for the narrowness of the observed peaks compared to the shape of $\mathcal{E}_\perp^2$ as a function of θ_i.