Resonant Inverse Photoemission via Plasmons

W. Drube and F. J. Himpsel
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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A large enhancement, up to a factor of 30, of the photon intensity is observed in inverse photoemission when the emitted photon energy equals the plasmon energy. The effect is explained as a resonance between a discrete channel, i.e., radiative decay of a plasmon, and the inverse photoemission continuum. Examples are shown for various structures of antimony films.

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Resonant photoemission [1] has generated considerable interest for the study of multielectron phenomena in solids.[2] In general, such resonant phenomena can be described as an interaction between a discrete channel and a continuum.[3] Analogous effects[4] have been observed in the time-reversed photoemission process, i.e., inverse photoemission[5] or bremsstrahlung isochromat spectroscopy. So far, these resonant phenomena have involved a core-to-valence transition as the discrete channel. Therefore, they are limited to the high-energy regime where core levels are accessible. We see a new type of resonance which is based on a plasmon excitation as the discrete channel. The observed resonant enhancement is dramatic (a factor of 30 in the example shown below). An analogous effect is predicted to exist in photoemission.

An overview of the resonance effect is given in Fig. 1. The data were taken for a thin Sb film evaporated onto a cleaved InP(110) crystal. It shows a series of spectra for the photons that are produced by electrons arriving with energy E_i at the Sb surface. The electron energies range from $E_i = 11.6$ eV above the Fermi level E_F (top curve) to $E_i = 28.6$ eV (bottom curve) in steps of 0.5 eV. The spectrum of emitted photons is recorded by a spectrograph with a position-sensitive detector as described previously.[7] The two channels that lead to emission of photons can be identified by their energy dependence. The discrete channel emits photons of a fixed photon energy which equals the plasmon energy of the Sb film $\hbar\omega_p = 16.4$ eV.[8] Thus, it corresponds to a vertical line in Fig. 1. The inverse photoemission continuum is characterized by a photon spectrum with an upper limit at $\hbar\omega = E_i - E_F$ corresponding to a diagonal line in Fig. 1 (arrow labeled E_F). This upper limit corresponds to radiative transitions into empty electronic states just above E_F. For the Sb film on InP(110) several empty electronic (surface) states are observed at constant final-state energies. The lowest state is located at $E_F + 0.25$ eV. Transitions into higher-lying final states (e.g., at $E_F + 0.95$ eV and $E_F + 3.9$ eV) correspond to diagonal lines displaced downwards from the E_F arrow by the energy of the final states. Whenever one of these diagonal ridges crosses the vertical plasmon line there is a resonant enhancement that by far exceeds the mere sum of the two channels. For the state at $E_F + 0.25$ eV the intensity increases by a factor of 30 at resonance and leads to signal levels that are at least an order of magnitude higher than those we have encountered with any material stud-

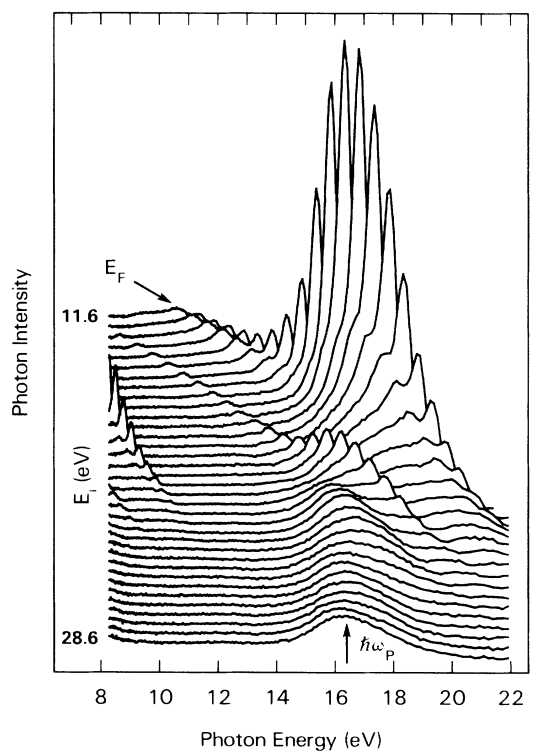

FIG. 1. Inverse photoemission spectra (raw data) for a thin film (20 Å) of Sb on cleaved InP(110) for various energies E_i of the incoming electrons at $k_{\parallel} = 0$. A resonant enhancement is observed where the inverse photoemission and the plasmon channels coincide (Ref. 6). The initial-state energy relative to E_F varies from $E_i = 11.6$ eV for the top curve in steps of 0.5 eV to $E_i = 28.6$ eV for the bottom curve. Their vertical displacement is proportional to E_i; the abscissa defines the intensity zero of the bottom curve. The diagonal arrow labeled E_F points at the onset of the bremsstrahlung spectra due to states at E_F; the vertical arrow marks the photons of energy $\hbar\omega_p$ emitted by decaying plasmons.

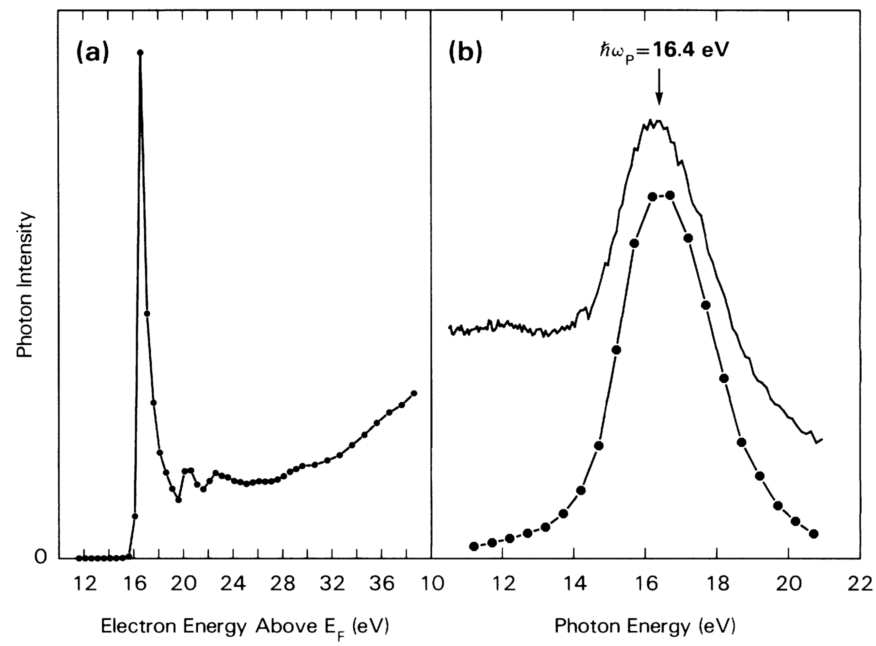

FIG. 2. Three different cuts through Fig. 1. (a) Vertical cut along the discrete plasmon decay channel at $h\nu = \hbar\omega_p = 16.4$ eV (isochromat at resonance). (b) Lower curve: diagonal cut (parallel to the E_F arrow in Fig. 1 but displaced downwards by 0.25 eV) along the inverse photoemission from the state at $E_F + 0.25$ eV (resonance curve for the inverse photoemission channel). Upper curve: horizontal cut at $E_i = 28.6$ eV showing the line shape of the radiative plasmon decay. The curves in (b) have the same center and width providing strong evidence for a plasmon decay as the origin of the observed resonance.

ied so far in nonresonant inverse photoemission.

Three different cuts through the graph in Fig. 1 are given in Fig. 2. The first cut (a) is vertical, a so-called isochromat, and covers the discrete channel with the photon energy $h\nu$ fixed at $\hbar\omega_p$. The second cut [(b) lower curve] follows the inverse photoemission channel for the Fermi-level state, i.e., $h\nu = E_i - E_f$ with the final-state energy $E_f = E_F + 0.25$ eV. The third cut [(b) upper curve] is horizontal with a high initial-state energy $E_i = 28.6$ eV and shows the photon spectrum caused by the plasmon decay (plus some structureless inelastic bremsstrahlung background). From cut (a) we learn that the plasmon decay channel has slowly varying intensity until it becomes resonantly enhanced at threshold. The resonant fine structure near threshold corresponds to the density of unoccupied states with the Fermi level displaced upwards by the chosen photon energy $h\nu = 16.4$ eV. For example, the states at $E_F + 0.25$ eV ($E_F + 3.9$ eV) show up at electron energies of 16.65 eV (20.3 eV). Cut (b), lower curve represents the resonance in the inverse photoemission channel from the state at $E_F + 0.25$ eV. It shows the intensity enhancement by a factor of 30 with a full width at half maximum of 3.0 eV. The position and the width of the resonance curve are the same as observed for the photon spectrum of the decaying plasmon [(b) upper curve]. This similarity makes a strong case for assigning the plasmon decay as discrete channel for the inverse photoemission resonance.

In order to test how ubiquitous a phenomenon this is we looked for the effect in other materials (Al, Si, W) and for various structures of the Sb film. For classic free-electron-like materials such as Al the resonant effect described above cannot be detected. Instead, we observe an intensity minimum at the bulk-plasmon energy due to the (inverse) photoelectric effect.[9] For Sb films the resonance effect is found to be strongly dependent on the crystal structure (Fig. 3). Metastable quasicubic phases exhibit a large resonant enhancement [×30 for Sb/ InP(110), ×22 for Sb/W(100)], whereas the normal hexagonal structure of Sb has only traces of the resonance left. We notice that the (nonresonant) radiative plasmon decay also becomes weak when the resonance disappears (Fig. 3 right panels). This is one more piece of evidence for the connection between the plasmon and the resonance. Why, then, does the interaction between plasmons and radiation vary so much, even for the same material? The answer is not completely clear but is very likely connected with the coupling of the longitudinal plasma oscillation with the transverse photon fields.[10] The coupling of plasmons with the incoming electrons has already been worked out.[11] It is possible that the photon-plasmon interaction is mediated by interband transitions which have ΔE and Δk matched to the plasmon E and k. Interband transitions do not cou-

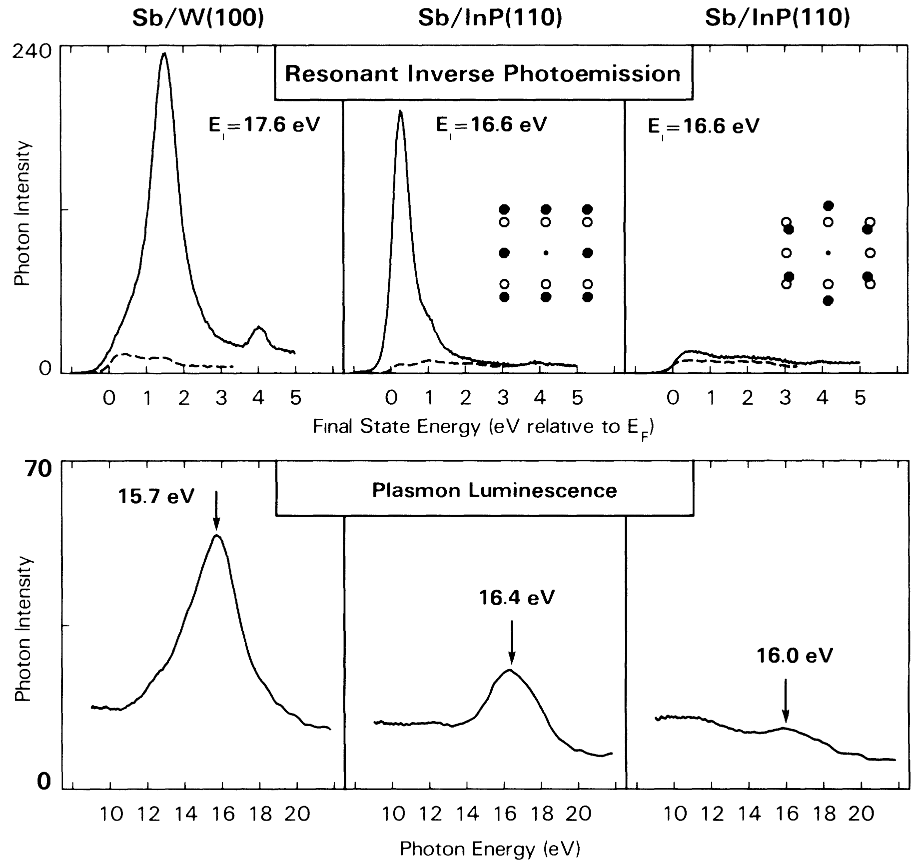

FIG. 3. Dependence of the inverse photoemission resonance on the structure of the Sb film (about 20 Å thick in all cases). For metastable quasicubic phases (left and middle panels) a strong resonance is seen, which is almost unobservable for the normal hexagonal phase of Sb (right panel). As the discrete plasmon decay channel vanishes the resonance also disappears. A sketch of the low-energy electron diffraction patterns for Sb/InP(110) (solid symbols) is shown in the upper panels [open symbols represent the pattern seen for clean InP(110)]. The pattern for Sb/W(100) is the same as for the clean substrate (not shown). The dashed lines in the upper panels are off-resonance spectra ($E_i = 11.6$ eV) drawn to scale. The photon spectra in the lower panels were taken at $E_i = 27.6$ eV.

ple with bulk plasmons for $k \simeq 0$ in free-electron-like materials such as aluminum. Therefore a deviation from the free-electron behavior might be responsible for the resonance's occurrence in the quasicubic structured Sb film.

We have observed a coupling of surface plasmons (as opposed to bulk plasmons) to light in inverse photoemission from the free-electron-like materials Al and Si and also from W. The light emission from the surface plasmons is found to become stronger for rough surfaces. The enhancement of the inverse photoemission intensity at the surface-plasmon energy, however, is much weaker than the resonance effect seen with bulk plasmons.

In summary, we have observed a novel resonance effect which is caused by the coupling of bulk-plasmon excitation and inverse photoemission. A similar resonance effect is predicted for photoemission.

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