SURFACE-PLASMON RESONANCE EFFECT IN GRATING DIFFRACTION*

R. H. Ritchie, E. T. Arakawa, J. J. Cowan,† and R. N. Hamm
Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
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Anomalies found in the intensity of p-polarized light from concave diffraction gratings have been analyzed in terms of an interaction between the incoming photon and a surface-plasmon resonance in the grating surface. There is clear evidence for zone gaps in the dispersion curves which we have constructed for surface plasmons in Al and Au. The phenomena reported here may be regarded as manifestations of second- and possibly higher-order plasmon-grating interactions.

Anomalies in the intensity of light diffracted from a grating were first observed by Wood[1] in 1902. These anomalies, which are most important for light polarized with its electric vector perpendicular to the grating rulings, have been studied both experimentally and theoretically by many workers. Recent treatments have been given by Hessel and Oliner[2] and Hägglund and Sellberg.[3] A classical theoretical analysis was given some time ago by Fano.[4]

We have studied the polarization of light by concave diffraction gratings in the wavelength region 3000 to 16 000 Å and present an explanation in terms of a surface-plasmon resonance.

Measurements were made of the intensity of the diffracted light for polarization with the electric vector parallel and perpendicular to the plane of incidence which, in turn, was perpendicular to the grating rulings. These measurements were made on a Bausch and Lomb replica grating ruled with 600 lines/mm and having a blaze angle of 2°35′. The grating was coated with a vacuum-evaporated layer of aluminum. For the other measurements an additional layer of gold was evaporated over the aluminum. Measurements were made using a Polaroid HN-22 sheet polarizer and a tungsten lamp. The angle between the source and detector (i.e., between the incident and diffracted beam) was varied from 80° to 2°. Data taken in the wavelength region 5000 to 15 000 Å using a PbS detector are shown in Fig. 1. The angle θ shown is half the angle between the incident and diffracted beam. The intensity of p-polarized light (electric vector in the plane of incidence) exhibits peaks that shift in wavelength as the angle θ is varied. The s component varies smoothly with wavelength for all angles of incidence. These data, along with data taken in the region 3000 to 8000 Å using an EMI 9558 BQ photomultiplier, were used to plot Fig. 2 for both the Al and Au coatings. The photon energy at which the peaks occurred is plotted as a function of the angle θ.

Teng and Stern[5] have been able to construct a dispersion curve for the surface plasmon in aluminum by observing dips in the spectrum of light specularly reflected from an aluminum-coated grating[6] and from peaks in the light emitted from the same grating bombarded by fast electrons. Both of these effects may be regarded as occurring in first order in the plasmon-grating interaction. The phenomena which we report here, on the other hand, may be regarded as manifestation of second-order and possibly higher order processes.

We assume that the surface plasmon is only slightly perturbed by the presence of density variations due to the grating structure. For ex-

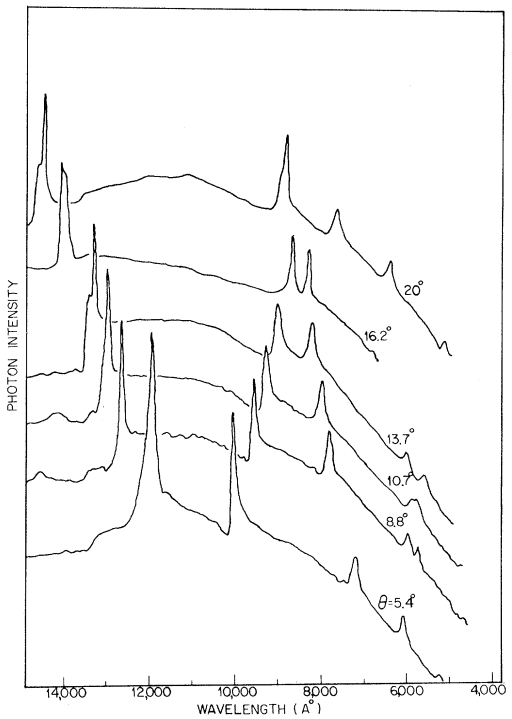

FIG. 1. p-polarized spectra of tungsten lamp diffracted by concave grating for varying angles between entrance and exit slits.

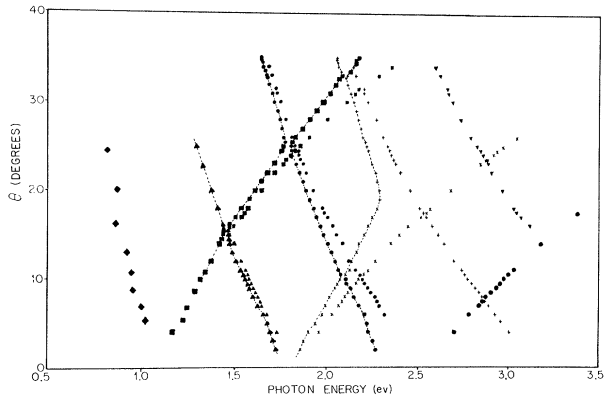

FIG. 2. Position of peaks in the p-polarized spectra as a function of angle θ. Dashed line, Au.

ample, the dispersion curve of the plasmon on a grating surface does not seem to be greatly different from that of a plasmon on a plane surface of the same material, at least in the long-wavelength region. In this view, the resonances observed in first-order diffraction may be discussed in terms of the graph shown in Fig. 3(a) in which we depict a second-order process. Here a photon of wave vector $\vec{k}$ in the region outside of the grating is incident upon the grating surface, which if perfectly plane would only specularly reflect the photon; a surface plasmon could not be excited under these conditions. Because there is a periodic density variation in the region of the surface, the grating may absorb momentum in a direction normal to the rulings and in multiples of $2\pi\hbar/\delta$, where δ is the distance between rulings. We shall assume that the average grating surface lies in the x-y plane, that the gratings are parallel to the y axis, and that the wave vector $\vec{k}$ lies in the x-z plane. Thus at the lower vertex in the graph on Fig. 3(a) surface plasmon with x momentum equal to $2\pi\hbar n/\delta$ plus the x component of the momentum of the incident photon may be created. Here n is a positive or negative integer. For a free-electron-like metal, the interaction Hamiltonian giving rise to this process may be written as an integral over the half-space containing the grating,

$$H' = -(\omega_p^2/4\pi c^2)\int \vec{A}_{ph} \cdot \vec{A}_{sp} f(\vec{r}) d\tau,$$

where $\vec{A}_{ph}$ is the vector potential operator of the photon, $\vec{A}_{sp}$ is the vector potential operator of the surface plasmon, and $f(\vec{r})$ is the fractional variation due to the grating structure of the conduction-electron density from its value n_0 deep in the metal. The volume plasma frequency is given by $\omega_p = [4\pi n_0 e^2/m]^{1/2}$. In order for its effect to be observed in a first-order diffraction direc-

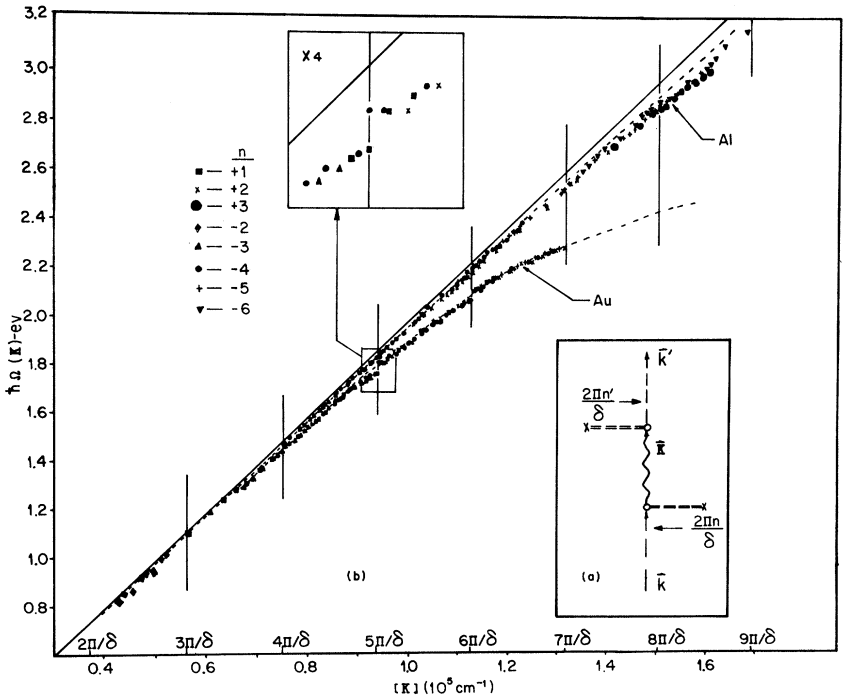

FIG. 3. (a) Feynman diagram of photon–surface-plasmon interaction. (b) Dispersion curve of surface plasmons in Al and Au.

tion, the plasmon must, in general, get rid of some of the x momentum which it acquired from the grating by making a second encounter, indicated by the top vertex, in which a photon of wave vector $\vec{k}'$ is emitted. The compound process proceeds through an intermediate surface-plasmon state; it is well known that when the energy of an intermediate quantum state is nearly the same as that of the initial state, a resonance in the cross section for the compound process will occur. Thus, from the theory of quantum resonant intermediate states one could expect that P_n, the probability for the process under discussion, might have the typical Lorentzian form

$$P_n \sim \frac{|F_n F_{n\pm 1}|^2}{\{\omega_k - \Omega(\kappa_n)\}^2 + \{\frac{1}{2}\gamma(\kappa_n)\}^2} \tag{1}$$

valid in the neighborhood of the frequency for which the denominator has its minimum.[7] In this expression $\kappa_n = k_x + 2\pi n/\delta$, $\omega_k = c|\vec{k}|$, $\Omega(|\vec{k}|)$ and $\gamma(|\vec{k}|)$ are the eigenfrequency and damping rate, respectively, of a surface plasmon with wave vector $\vec{k}$, and F_n is proportional to a certain average of the nth Fourier coefficient of the electronic density variation in the neighborhood of the grating surface. Possible higher order processes giving rise to a probability function containing a product of several resonance denominators may also be important in this connection.[8]

The condition for intermediate-state resonance is that the energy of the virtual surface plasmon should be equal to the energy of the incoming photon, i.e., $\omega_k = \Omega(\kappa_n)$; dropping the subscript on ω_k,

$$\omega = \Omega(|(\omega/c)\sin\alpha + (2\pi n/\delta)|), \tag{2}$$

where α is the angle between $\vec{k}$ and the normal to the grating surface. Thus if the wavelength corresponding to the center of a resonance of given order n is measured as a function of the angle of incidence α, one may construct the $\Omega(\kappa)$ curve by graphical methods.

The angle of incidence α is given as a function of the angle θ by the grating equation

$$\alpha = \theta + \sin^{-1}\left\{\frac{\pi n c}{\delta\omega\cos\theta}\right\}. \tag{3}$$

Using this relation in Eq. (2), the dispersion curve $\Omega(\kappa)$ was constructed and is shown in Fig. 3(b). Note that the different symbols correspond to specified values on n in the equation for κ_n, and that each specified value of n, in turn, corresponds to a branch in the experimental data (Fig. 2). For comparison, theoretical dispersion curves for the surface plasmon on a smooth plane surface were calculated from[9]

$$\kappa = \frac{\Omega}{c}\left\{\frac{\epsilon_1(\Omega)}{1+\epsilon_1(\Omega)}\right\}^{1/2} \tag{4}$$

using $\epsilon_1(\omega)$, the real part of the dielectric constants of aluminum and gold, from the data of Ehrenreich, Philipp, and Segall[10] and of Schultz,[11] and are shown as the dashed curves in Fig. 3(b). There is good agreement for Au, and for Al the agreement is good for quantum energies up to ~2 eV. For energies larger than 2 eV in Al, the experimental points lie below the theoretical curve defined by Eq. (4). These deviations may be due to (a) the effect of the grating structure, (b) depolarization effects due to oxidation of the surface, or (c) possible differences between the optical properties of the aluminum of the grating surface and those reported in Ref. 10 for aluminum.

The second-order self-energy of a surface plasmon involves the excitation of virtual plasmons with momenta differing from the momentum of the plasmon under consideration by multiples of $2\pi\hbar/\delta$. Thus discontinuities or zone gaps in the self-energy and, consequently, in the dispersion relation, are to be expected at values of κ equal to integer multiples of π/δ. Inspection of Fig. 3(b) shows clear indication of discontinuities at $5\pi/\delta$ and $6\pi/\delta$ for Au, but for Al these indications are less clear.

We plan to attempt a detailed perturbation theoretic analysis of the data and to investigate the dispersion curves of other metals.

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†Radiological Health Physics Fellow, University of Tennessee, Knoxville, Tenn.

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[6]Similar results have been obtained earlier by J. E. Stewart and W. S. Gallaway, Appl. Opt. 1, 421 (1962).

[7]Interference with the ordinary first-order diffracted beam has been neglected here. Such interference effects are responsible for the structure shown in Fig. 1 in which shallow dips appear on the long-wavelength

side of the peaks.

[8]Analogous compound resonances involving the radiative surface plasmon have been considered theoretically by R. E. Wilems and R. H. Ritchie, Phys. Rev. Letters 19, 1325 (1967) in the case of a thin grating foil.

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SPECTROSCOPIC FACTORS FROM (d,p) REACTIONS

S. T. Butler and B. H. J. McKellar
Department of Theoretical Physics, University of Sydney, New South Wales, Australia
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The recent communication by Garcia and Pearson, of the same title as this note, is examined and shown to contain an error. The conclusions drawn by Garcia and Pearson are therefore incorrect.

We refer to a recent Letter by Garcia and Pearson[1] in which the authors comment on the Butler-Hewitt-McKellar-May (BHMM) analysis of (d,p) reactions.[2-4] They purport to show that the factor $(1-S)^{-2}$ multiplying the cross section in this method does not in fact exist and is the result of an error. It is, however, the Garcia-Pearson conclusion that can be reached only through a mathematical error; it appears necessary to point this out explicitly.

We use the same notation as Ref. 4 and, as in Ref. 1, consider the case of one bound state only; extension to several bound states is straightforward.

The direct matrix element for the stripping reaction $X(d,p)Y$ is written

$$M(0,\vec{k}_p)=\langle\psi_p^{-}\Lambda_c\Psi_0(X,n)|V_{np}|\Lambda_c\Psi_d^{+}\rangle, \quad (1)$$

where ψ_p^{-} describes proton elastic scattering, V_{np} is the neutron-proton interaction, Ψ_d^{+} is the full many-body deuteron wave function, and $\Psi_0(X,n)$ is the wave function of the final state. The operator Λ_c projects onto the core-state wave function $u_0(X)$. One then has

$$M(0,\vec{k}_p)=S^{1/2}M(0,\vec{k}_p),$$

where S is the spectroscopic factor for the final state and

$$M(0,\vec{k}_p)=\langle\psi_p^{-}u_0(X)F_0(n)|V_{np}|\Lambda_c\Psi_d^{+}\rangle.$$

After expansion in terms of the complete set of states $\Psi_i(X,n)$ of nucleus Y it is found[4] that

$$(1-S)M(0,\vec{k}_p)=\int d\vec{k}_n\,\langle 0|\vec{k}_n\rangle M(\vec{k}_n,\vec{k}_p), \quad (2)$$

where

$$M(\vec{k}_n,\vec{k}_p)=\langle\psi_p^{-}\Lambda_c\Psi(\vec{k}_n,X,n)|V_{np}|\Psi_d^{+}\rangle. \quad (3)$$

We now briefly reproduce the Garcia-Pearson argument. The wave equation for Ψ_d^{+} is used to rewrite (3) in the form

$$M(\vec{k}_n,\vec{k}_p)=\langle\psi_p^{-}\Lambda_c\Psi(\vec{k}_n,X,n)|(H-E)|\Psi_d^{+}\rangle, \quad (4)$$

where H is the full Hamiltonian without the V_{np} interaction and E is the total energy. The sudden approximation[2,3] is now substituted for Ψ_d^{+} so that the right-hand side of (2) becomes

$$[(4\pi)^{1/2}\hbar^2N/(2\pi)^{3/2}m]\int d\vec{k}_n\,d\vec{k}_p'\,g(\vec{k}_p',\vec{k}_d)\langle 0|\vec{k}_n\rangle\langle\psi_p^{-}(\vec{k}_p)|\psi_p^{+}(\vec{k}_p')\rangle\langle\Lambda_c\Psi(\vec{k}_n,X,n)|\Lambda_c\Psi(\vec{Q}',X,n)\rangle. \quad (5)$$

However,

$$\langle 0|\vec{Q}'\rangle=S\langle 0|\vec{Q}'\rangle+\int d\vec{k}_n\,\langle 0|\vec{k}_n\rangle\langle\Lambda_c\Psi(\vec{k}_n,X,n)|\Lambda_c\Psi(\vec{Q}',X,n)\rangle$$

so that (5) reduces to

$$(1-S)M_S \quad (6)$$

with

$$M_S=[(4\pi)^{1/2}\hbar^2N/(2\pi)^{3/2}m]\int d\vec{k}_p'\,\langle 0|\vec{Q}'\rangle\langle\psi_p^{-}(\vec{k}_p)|\psi_p^{+}(\vec{k}_p')\rangle. \quad (7)$$