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COHERENT AND INCOHERENT RADIATION FROM OPTICALLY EXCITED SURFACE PLASMONS ON A METAL GRATING*

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Excitation of surface charge waves (surface plasmons) produces intensity anomalies in the light reflected from a metal grating. We demonstrate that coupling occurs for both s- and p-polarized light if the grating lines are oblique to the plane of incidence, provided there is a component of the incident electric field along the direction of surface-wave propagation. We have measured the magnitude, phase, and direction of the plasmon electric field, and find Fano's theory confirmed. Underlying the coherent radiation we observe a small incoherent component resulting from spontaneous emission by the surface plasmons.

That electromagnetic waves can run along a metal surface whose dielectric constant ϵ is less than -1 is well known. Many years ago Wood[1] observed anomalies in the intensity of p-polarized light diffracted from a metal grating. Fano[2] explained the anomalies as being due to the resonant excitation of surface waves. He pointed out that this occurs when the incident light, phase modulated by the periodic grating structure, contains a component whose wavelength just matches that of the surface waves. To satisfy the boundary conditions at the surface it is then necessary for there to be an additional reflected field, which interferes with the background reflected field and produces the observed anomalies. p polarization is required to give an incident electric field varying in the direction of surface-wave propagation.

Phase modulation of the incident beam builds up coherent fields at the surface whose momenta along the surface are given by the relation $\vec{k}_s' = \vec{k}_s + n\vec{K}$, where if λ is the incident wavelength, θ the angle of incidence, and d the grating spacing, then $k_s = 2\pi \sin\theta/\lambda$ and $K = 2\pi/d$, and n is an integer. Surface waves are excited whenever $\vec{k}_s' = \vec{k}_p$, the surface-wave momentum. Since surface waves have a momentum only a little greater than $2\pi/\lambda$, $k_p^2-(2\pi/\lambda)^2 = 1/(|\epsilon|-1)$, the anomalies occur, as Fano explained, where a higher order diffracted beam has just disappeared into the surface.

In the last few years there has been a renewed interest in the properties of surface-charge waves (or surface plasmons). Teng and Stern[3] excited these waves on a grating by the electric fields associated with a beam of electrons, and observed spontaneous emission of light. They pointed out that waves running perpendicular to the grating lines would not propagate when their wavelength just matched the grating spacing, and indeed this property had already been observed of the grating anomalies by Stewart and Gallaway.[4] Recently more data on several metals have been presented by Ritchie et al.[5] Teng and Stern and Ritchie et al. have shown that the plasmons follow roughly the dispersion relation above.

It is surprising, however, that the resonant electric fields have not before been explicitly measured. In this Letter we demonstrate that when the waves run at an angle to the plane of incidence, they may be stimulated by either s- or p-polarized light. We separate the fields associated with the charge waves from those of the background, and measure the magnitude, direction, and phase of the plasmon fields directly. Underlying the coherent plasmon fields we observe an incoherent radiation, which is the spontaneous emission by the charge waves which Teng and Stern observed using electron excitation.

We use a geometry where the grating lines are oblique to the plane of incidence. Keeping $\vec{k}_s$ fixed we rotate the grating about its normal, sweeping the vector $\vec{K}$ in a circle. In this way $\vec{k}_s'$ is varied between the extremes $k_s - nK$ and $k_s + nK$. With a suitable choice of θ and λ, k_s' will match k_p at some intermediate value. The vectors are shown in Fig. 1. This is the geometry used by Teng and Stern in their experiment. The grating here has 606 lines/mm, is blazed at 15°, and is overcoated with 1500 Å of aluminum. Monochromatic parallel light is incident onto the grating. The grating is attached directly to an infinite resolution potentiometer so that the x axis of a recorder reads φ, the angle between K and the plane of incidence, directly. The intensity of the specularly reflected beam is displayed on the y axis.

As the angle φ is reduced from 90° by anticlockwise rotation, k_s' increases. At an angle φ_d k_s' just equals $2\pi/\lambda$. At this point the nth order diffracted beam just grazes the surface and vanishes. A sharp discontinuity occurs in the slope of the curves. As φ is reduced still further, k_s' becomes larger, equaling k_p at φ_p, and a peak or dip occurs in the reflected intensity. k_p is in the plane of incidence only if $\varphi_p = 0°$. Notice that if a plasmon occurs at the angle φ_p, it will also occur at $-\varphi_p$.

In Fig. 2(a) we show typical curves for three angles of incidence for s- and p-polarized light. Peaks are found for $n = 2, 3, \cdots$, a dip for $n = 1$.

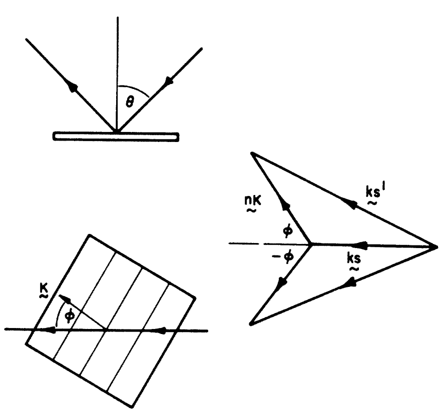

FIG. 1. The angles θ, φ, and the vectors $\vec{k}_s$, $\vec{k}_s'$, and $\vec{K}$.

At $\theta = 50°$ the $n = 1$ dips are rather broad, and the lowest point is at different values of φ_p for the two polarizations. Such line-shape effects, and indeed the occurrence of peak or dip, result from the phase difference between the plasmon field and the background field, as we discuss further below.

It is seen in Fig. 2(a) that the plasmons at φ_p and $-\varphi_p$ are equally strongly excited—the curves are symmetric about $\varphi = 0$ and 90°. If the incident polarization is rotated to a position between s and p, then equal coupling no longer occurs. Figure 2(b) presents an example. We have found that maximum strength occurs when the component of the incident electric field in the surface along the direction of k_p is greatest.

To analyze the interference between plasmon

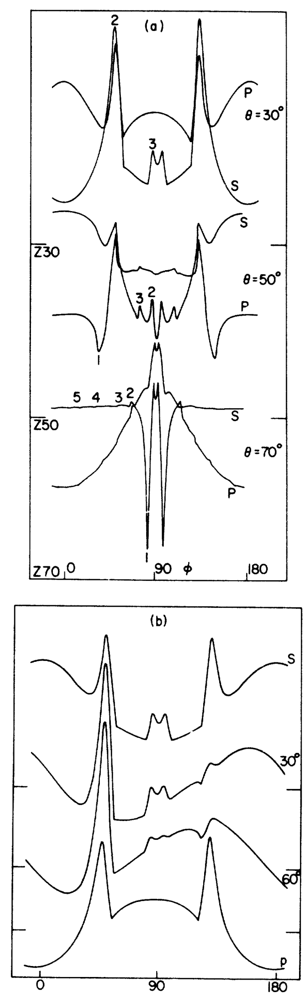

FIG. 2. (a) Typical rotation curves of reflected intensity versus φ for s and p polarized light. $\lambda = 0.526\ \mu$ and $\theta = 30°, 50°, 70°$. (b) Rotation of the incident polarization from s to p in 30° steps. $\lambda = 0.526\ \mu$.

and background fields we have used combinations of analyzer and quarter-wave plate in the reflected beam. The reflected intensity may in general be elliptically polarized. Using an analyzer alone, its setting at minimum intensity tells one the direction of the minor axis, and the value of the minimum intensity is just that of the minor axis. If a quarter-wave plate is placed in front of the analyzer, and both then adjusted for minimum intensity, then the direction of the quarter-wave plate is along the major axis, and the angle of the analyzer with respect to the quarter-wave plate provides the ratio of minor-axis to major-axis amplitudes and the direction of rotation of the ellipse. A nonzero intensity reading occurs now only if incoherent radiation is present.

In regions where there are no anomalies we have found that the reflected light is linearly polarized. The grating acts as though it has amplitude reflectivities r_i and r_j parallel and perpendicular to the grating lines, respectively.[6] For s polarized light the intensity of the background light follows the relation $E_r^2 = E_0^2(r_i^2 \times \cos^2\varphi + r_j^2 \sin^2\varphi)$. It is polarized at an angle $\varphi - \beta$ to the incident polarization, where β is given by $\tan\beta = (r_i/r_j)\tan\varphi$. E_0^2 is the incident intensity. A similar relation may be derived for p polarized light. The reflected light rotates away from the direction of E_0 and back again as φ varies between 0 and 90°.

In the region of the anomaly we have found the reflected light to be elliptically polarized. The direction of the major axis swings away from and then back to E_r as we pass through the anomaly. Part way through the anomaly the light is linearly polarized, indicating that the plasmon field is exactly in phase (or 180° out of phase) with the background light. The reflected light may be described by the relation $\vec{E} = \vec{E}_r + \vec{E}_p e^{i\delta}$. $\vec{E}_p$ is the plasmon reflected field, δ the phase difference between $\vec{E}_r$ and $\vec{E}_p$. $\vec{E}_p$ and δ vary through the resonance; $\vec{E}_r$ and $\vec{E}_p$ are not, in general, collinear.

We illustrate these effects in Fig. 3(a). Here is shown the total intensity, the minor-axis intensity, the residual incoherent intensity, and the directions of the quarter-wave plate (the major axis) and analyzer when used in combination for the $\theta = 30°$, s-polarization data of Fig. 2(a). The magnitude and direction of E_r can be found from the regions of linear polarization away from the resonance, as outlined above. Its direction is shown dotted in the lower curve. Notice that the analyzer and quarter-wave plate

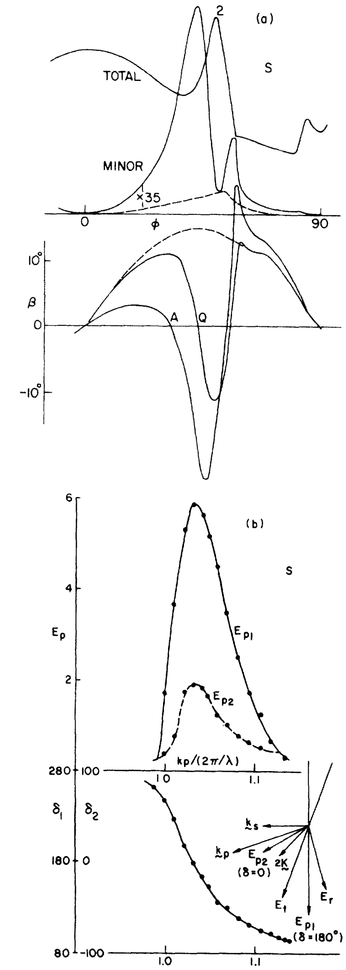

FIG. 3. (a) Top, the total intensity, minor axis, and uncompensated signal for the $n = 2$ anomaly at $\lambda = 0.526$ μ, $\theta = 30°$. Bottom, the direction of the major axis (Q) of the elliptically reflected light, and the position of the analyzer (A). If ξ is the difference in angle between Q and A, $\tan\xi = b/a$, where b and a are the minor and major axis amplitudes, respectively. (b) The amplitude of the plasmon field E_p and the phase difference δ plotted versus $k_p/(2\pi/\lambda)$ for $n = 2$, $\lambda = 0.526$ μ, and $\theta = 30°$. The directions of the vectors are shown at maximum E_p in the inset. The two possibilities are discussed in the text.

point in the same direction when $\delta = 0°$ (or 180°); the swing of the analyzer in passing through resonance indicates a change in sign of δ. Since we know $\vec{E}_r$ it has been possible to analyze the ellipse completely, finding E_p, δ, and the direction of E_p. The results are shown in Fig. 3(b), where the resonance variation of E_p and δ is displayed.

In fact, there are two possible solutions for E_p. This happens because one knows only the line along which the major axis points. The two solutions are shown in Fig. 3(b), and the direction of the vectors in the inset. There appears to be no a priori way of choosing between these. At the peak of the resonance one solution is closely parallel to E_0, with a phase shift $\delta = 180°$ at the in-phase point. It remains within 5° of E_0 over the whole resonance region. The other solution at the resonance peak points between k_p and K, with a phase shift $\delta = 0°$ at the in-phase point. Off resonance E_p varies from this direction but seemingly in no way related to φ or the direction of k_p. The first solution seems the more realistic.

Analyzing the $\theta = 50°$, $n = 1$ data of Fig. 2(a), we have found that the direction of the major axis swings further away from E_0 as one goes through the resonance. One solution for E_p is about 30° off E_0; the other lies between k_p and K. δ in both cases varies through 180°. In this case it is not evident which is the most likely solution. For the $\theta = 70°$ data of Fig. 2 the point of $\delta = 180°$ occurs near the center of the resonance. For the $\theta = 50°$ data, δ equals 180° on one side of the resonance with s-polarized light, on the other with p-polarized light. This produces the line-shape distortion which has been referred to already above. Such line-shape distortion may account for the deviations of k_p from the predicted values found by Teng and Stern and Ritchie et al.

The resonance anomalies may be considered as arising from virtual excitation of surface plasmons. The residual incoherent signal must come from spontaneous emission by real plasmons. After excitation these propagate for a short while before scattering from the grating into outgoing waves. The emission process is separate from the excitation process, and the emitted light is therefore incoherent with the incident light. For the $n = 1$ plasmons the incoherent light is sharply peaked at φ_p, but for the $n = 2$ it is peaked more towards φ_d. At present it is not clear whether this is an angle of incidence effect, since the $n = 2$ anomalies tend to occur at smaller angles of incidence, or an order effect.

We are continuing studies of the line shape of the spontaneous emission and of the interference effects in the diffracted beams as well as the specular beam. We have enjoyed conversations with Dr. Michael Greene and many others in our laboratory.

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NATURE OF OPTICAL TRANSITIONS IN COPPER AND Cu:Ni ALLOYS*

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The direct $L_{2'} \rightarrow L_1$ transition is very apparent in photoemission data from pure Cu; however, as might be expected, there is no evidence for this transition in 87% Cu–13% Ni alloys. In contrast, structure in ϵ_2 for $h\nu \geq 2.1$ eV for the alloy and pure Cu is qualitatively similar. These observations support the assignment of major structure in ϵ_2 of pure Cu to nondirect transitions from the d states and argue against the 5.0 eV peak being due principally to the $L_{2'} \rightarrow L_1$ transition.

In their pioneering work on the optical properties of metallic copper, Ehrenreich and Philipp[1] attributed structure in the optical constants characterizing the interband absorption to direct, or $\vec{k}$-conserving, transitions. Berglund and Spicer[2] and Krolikowski and Spicer[3] on the basis of photoemission data have emphasized the dominance of nondirect transitions, or transitions in which conservation of $\vec{k}$ does not provide an important selection rule in the usual one-electron sense, in determining the optical absorption of copper. In particular, in transitions from the d-derived states they found no evidence that conservation of $\vec{k}$ provided an important selection rule; however, a well-defined direct transition was found between s- and p-derived states near the L sym-