magnitude to that when the hexagonal array is completely formed. In this case, an alternative explanation is called for and the Mott-transition hypothesis remains as a possibility. The problem can be settled by further experiments to clarify the nature of the (2×2) layer.

This work was begun while one of us (K.L.N.) was at the University of Chicago. He wishes to thank J. J. Lander, A. U. MacRae, J. Morrison, and J. C. Phillips for useful conversations.

*The Lincoln Laboratory portion of this work was sponsored by the U. S. Air Force, and the University of Chicago portion by the National Aeronautics and Space Administration and the Advanced Research Projects Agency.

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SURFACE-PLASMON–ONE-ELECTRON DECAY AND ITS OBSERVATION IN PHOTOEMISSION*

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Evidence for the decay of surface plasmons into one-electron excitations has been obtained in photoemission measurements from a slightly roughened surface of aluminum. A simple theory has been developed which explains the resultant photoyield in terms of both the drop in reflectance resulting from roughness-aided plasmon coupling, and the relative penetration depth of the fields associated with these excited plasmons.

Optical excitations of volume plasma oscillations have long been observed. Ives and Briggs[1] first noted anomalies in the photoemission of metals near the volume plasma frequency, and Steinman[2] has since shown that these anomalies may be interpreted as the decay of volume plasmons into one-electron excitations. This plasmon-electron coupling occurs through the macroscopic Coulombic fields associated with the optically excited volume plasmons.

Surface plasma oscillations were first noted by Ritchie,[3] while Stern and Ferrell[4] later described the macroscopic Coulombic fields associated with these oscillations. The existence of such oscillating macroscopic fields implies that a surface-plasmon–one-electron coupling should occur which is analogous to the volume-plasmon–one-electron decay mechanism described above. The analogy breaks down in one respect, however. Optically excited volume plasmons are coherently excited, and their associated fields screen the excitation field to yield a unique optical-field decay length. Surface plasma oscillations, on the other hand, are most easily optically excited through the intermediary of momentum-conserving charge inhomogeneities such as surface roughness.[5] These surface excitations are not coherent with the incident optical field, and are characterized by macroscopic-field decay lengths which are independent of the

penetration depth of the optical field.[4,5]

In this paper we predict the effect which the decay of these optically excited surface oscillations should have on the photoemissive yield of a nearly-free-electron metal. Optical coupling to the surface plasmons is accomplished via surface roughness and the strength of the coupling is expressed in terms of the reflectance from that roughened surface.[6] The relative effect of this coupling on the photoemissive yield is derived in terms of the coupling strength modified by the relative penetration depths of the optical excitation field and macroscopic fields of the surface plasmon. Preliminary experimental results of reflectance and yield measurements on roughened films of aluminum are described and found to be in excellent agreement with the predictions of the reported theory. These results are believed to be the first reported observations and explanation of the decay of surface plasmons into one-electron excitations.

Surface oscillations, or surface plasmons, consist of periodic charge densities traveling along the surface of a medium with phase velocity $\omega(k)/k$. For $k \gg k_p$ (ck_p is the volume plasma frequency of the medium) the electric fields associated with these oscillations have the form[4]

$$\vec{E} = E_0 e^{-k|z|} \times \{\sin[kx-\omega(k)t]\vec{i}_x - \cos[kx-\omega(k)t]\vec{i}_z\} \quad (1)$$

for a wave traveling in the x direction along a surface whose normal is in the z direction. The penetration depth of these plasmon fields would thus vary as $k^{-1}(\omega)$. In the region of k space $k \leq k_p$, these fields are no longer valid although the penetration depth of the fields still tends to increase with decreasing k. It is crucial to note that the frequency dependence of this penetration depth is related to the plasmon dispersion relation, and that in general these decay lengths may be either less than or greater than the optical decay length in the material, depending on the frequency of the exciting surface oscillation.

A calculation of the dispersion relation $\omega(k)$ for surface plasmons in a free-electron metal having the electron density of aluminum is shown in Fig. 1.[7] This calculation includes the effects of retardation.[8] Retardation causes the dispersion curve to approach the electromagnetic dispersion curve asymptomatically at low frequencies, and prevents momentum-conserving coupling of the optical excitation to the surface oscillations at all frequencies.

Momentum-conserving mechanisms have been described which allow the coherent excitation of surface plasmons.[9,10] However, the most convenient method of allowing optical coupling to surface plasmons, and the method used in the discussion which follows, is through the incoherent interaction with surface roughness. Ritchie[5] and Ritchie and Crowell[6] have calculated the total probability for the excitation of a surface plasmon by a photon of energy $\hbar\omega$ incident upon a roughened surface. The calculation assumes a surface characterized by a roughness distribution having a Gaussian autocorrelation function with correlation length σ and rms height variation $\bar{\xi}$. Ritchie and Crowell have expressed this probability in terms of a drop in the reflectance of the metal and this result is given by

$$\Delta R_S(\hbar\omega) = \frac{hb\alpha^5(1-\alpha^2)^2}{(1-2\alpha^2)^{5/2}} \exp\left[-\frac{b\alpha^2(1-\alpha^2)}{4(1-2\alpha^2)}\right], \quad (2)$$

where $\alpha = \omega/\omega_p$, $b = \omega_p{}^2\sigma^2/c^2$, $h = \pi\omega_p{}^2\bar{\xi}^2/c^2$. This result was calculated in the small-perturbation approximation and is valid only in the limit of small $\bar{\xi}$ so that $\Delta R_S \ll R$. The energy dependence of the reflectance drop given in Eq. (2) includes the effect of the momentum spectrum of the roughened surface. It also includes a factor which is related to the density of plasmon states which exist at a given energy. The density-of-states function experiences a sharp singularity as ω approaches $\omega_p/\sqrt{2}$, and thus the reflectance function of Eq. (2) will show a sharp peak near $\omega_p/\sqrt{2}$ for surfaces which allow coupling to high-k plasmons. If, as will be assumed in the remainder of this paper, the momentum spectrum is such that $\sigma k_p \geq 1$, then coupling to the plasmons in the high-k region of Fig. 1 is weak, and the peak in the coupling may be substantially broad-

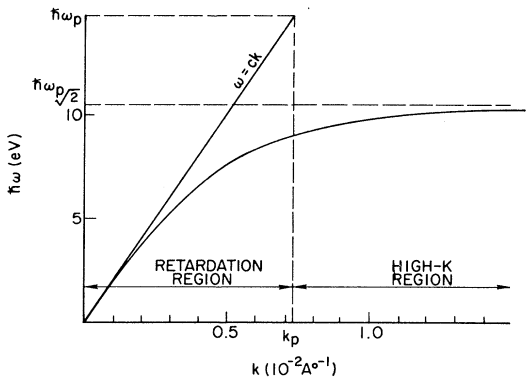

FIG. 1. The surface-plasmon dispersion curve for a free-electron metal having the electronic density of aluminum. ω_p is the volume plasma frequency.

ened and shifted to frequencies below $\omega = \omega_p/\sqrt{2}$.

The discussion to this point has ignored lifetime broadening of the plasmons. This assumption remains justified when dealing with surfaces for which $\sigma k_p > 1$ such that the structure in $\Delta R_S(\hbar\omega)$ is noticeably broader than the energy width of the function $\mathrm{Im}\{1/[\epsilon(\omega)+1]\}$, an approximate expression for the exact surface loss function, which includes the effects of lifetime broadening.[3,11] Within this assumption, one may interpret a drop in reflectance at energy $\hbar\omega$ as having resulted from the excitation of a surface plasmon of energy $\hbar\omega$ and momentum $\hbar k(\omega)$.

In the preceding presentation, the theory of roughness-aided optical coupling to surface plasmons has been reviewed and discussed in terms of the experimental determination of the energy and momentum spectra of plasmons excited on surfaces for which coupling is predominantly in the retardation region ($\sigma k_p \geq 1$). It has been shown that these spectra may be expressed in terms of a drop in the reflectance of the roughened surface. One may now proceed from a knowledge of the energy and momentum spectra of the excited plasmons to a prediction of how the photoyield from this roughened surface might be affected by the decay of these plasmons into one-electron excitations.

The photoyield per incident photon from an unroughened surface may be expressed in its most simple form as[12]

$$Y(\hbar\omega) = [1-R(\hbar\omega)]\alpha(\hbar\omega)\beta(\hbar\omega), \tag{3}$$

where $[1-R(\hbar\omega)] \equiv \Delta R(\hbar\omega)$ is the fractional number of photons absorbed, $\alpha(\hbar\omega)$ is the optical absorption coefficient, and $[1-R(\hbar\omega)]\alpha(\hbar\omega)$ is proportional to the density of absorbed photons within an electron escape length of the surface. The final parameter, $\beta(\hbar\omega)$, includes the effect of the electron inelastic scattering length (escape length), electron escape probabilities, and electron excitation probabilities averaged over the entire internal energy distribution of all electrons excited by photons of energy $\hbar\omega$. This simple expression for the photoyield is valid in the very common case for which the electron escape depth l is much less than the optical decay length, $1/\alpha$.

This expression for the photoyield may be extended to predict the additional photoyield resulting from the decay of surface plasmons if we make two well justified assumptions. First, we note that photoexcitation of interband transitions is independent of the polarization of the exciting fields for polycrystalline materials or materials of cubic symmetry.[13] Thus, it is reasonable to assume that excitation of the medium by surface-plasmon fields of Eq. (1) will have the same effect on the medium as an optical excitation field characterized by an absorption coefficient $\alpha(\hbar\omega) = \kappa(\hbar\omega) \equiv 2k(\hbar\omega)$, where $k(\hbar\omega)$ is the plasmon momentum vector of Eq. (1). If we further assume that there are no additional loss mechanisms, such as reradiative decay, peculiar to the surface oscillations, then we may conclude that both the number and internal distribution of electrons excited through the decay of a photon-excited surface plasmon is the same as for the excitation of the medium by a photon absorbed in a normal optical excitation. Thus, $\beta(\hbar\omega)$ is also the same as for an optical excitation and the expression for the additional yield due to surface plasmon decay is given by

$$\Delta Y_S(\hbar\omega) = \Delta R_S(\hbar\omega)\kappa(\hbar\omega)\beta(\hbar\omega). \tag{4}$$

Equations (3) and (4) may be summarized in the following concise form:

$$\frac{\Delta Y_S(\hbar\omega)}{Y(\hbar\omega)} = \left(\frac{\Delta R_S(\hbar\omega)}{\Delta R(\hbar\omega)}\right)\left(\frac{\kappa(\hbar\omega)}{\alpha(\hbar\omega)}\right). \tag{5}$$

For highly reflecting metals, the first factor in Eq. (5) may be greater than 1 for even small surface roughness, while the second factor may be significantly greater than 1 if coupling is possible to high-k plasmons. It is thus possible easily to obtain increases in photoyield, near $\omega = \omega_p/\sqrt{2}$, of several hundred percent from slightly roughened films of such metals.

Photoemission and reflectance studies of slightly roughened films of aluminum have been carried out, and these experimental results tend to confirm strongly the anomalies in photoyield predicted by Eq. (5). The highly free-electron nature of Al justifies use of the plasmon dispersion relation of Fig. 1, while the high reflectance of this material allows one to simultaneously to satisfy the assumption that $\Delta R_S \ll R$ and to allow $\Delta R_S \approx 1-R$. This means that significant increases in yield may be observed for even small roughness values.

The reflectance of a roughened surface of Al was measured by the author by evaporating a 1000-Å film on a substrate of known roughness[14] at a pressure of 1×10^{-8} Torr. Measurements were taken in situ at 10^{-10} Torr. The Al reflectance published by Ehrenreich and Phillips[15] was used as the smooth-sample reflectance in calculating the ratio $\Delta R_S(\hbar\omega)/\Delta R(\hbar\omega)$ shown in Fig. 2.

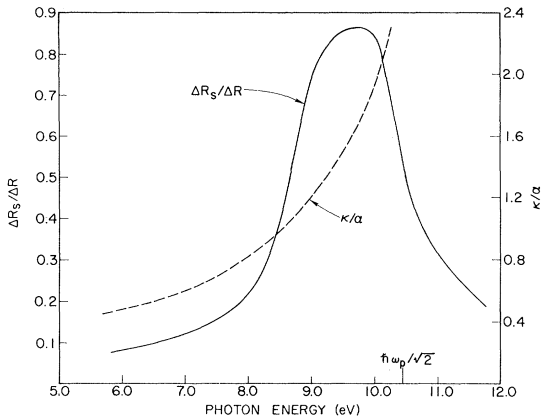

FIG. 2. The experimentally observed relative drop in reflectance, $\Delta R_S/\Delta R$, from a roughened surface of aluminum. ΔR_S, the difference between the rough- and smooth-surface reflectance, is assumed to result primarily from coupling to surface plasmons. ΔR is merely 1 minus the smooth-surface reflectance. Also shown is the ratio of the plasmon energy-decay coefficient κ to the optical absorption coefficient α.

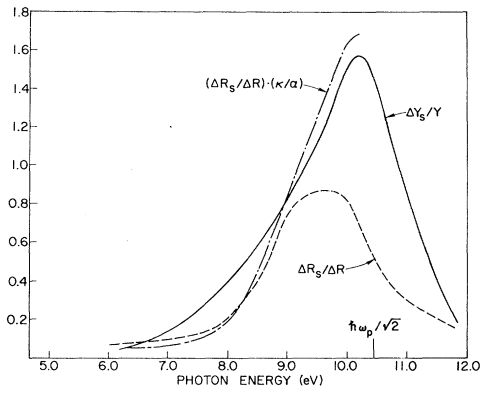

FIG. 3. A comparison of the experimentally determined relative increase in yield, $\Delta Y_S/Y$, from a roughened surface of aluminum, with the increase predicted from reflectance measurements on a similarly roughened aluminum surface. The predicted increase in yield is calculated according to a theory which assumes that the decay of excited surface plasmons is the dominant source of additional photoyield. Y in the above calculation is the smooth-surface photoyield for aluminum.

It can be seen from the broadening of this curve and the peak shift to $\omega < \omega_p/\sqrt{2}$ that coupling via the surface studied was in the retardation region consistent with the assumption of our theory that $k_p\sigma > 1$. Also shown in Fig. 2 is the ratio $\kappa(\hbar\omega)/\alpha(\hbar\omega)$. The optical constants calculated for Al by Ehrenreich and Phillips[15] were used to determine $\alpha(\hbar\omega)$, while the dispersion relation of Fig. 1 was used to find $\kappa(\hbar\omega)$.

Photoyield measurements on a roughened film of Al have been carried out by R. Koyama and N. Smith of this laboratory. Preparation techniques were virtually the same as those used in the reflectance measurement, while roughness measurements on polished metal substrates similar to the one used in the photoemission measurement showed roughness comparable to that of the reflectance substrate. The smooth-surface photoyield for Al, $Y(\hbar\omega)$, was estimated from the rough-surface yield by linearly extrapolating the rough-surface yield from its value at frequencies below 6.0 V to its value at 11.8 V. It was assumed that coupling to surface plasmons could be ignored at frequencies so far removed from $\hbar\omega = \hbar\omega_p/\sqrt{2}$. The values estimated for $Y(\hbar\omega)$ were in good agreement with the values measured by Wooten, Huen, and Stuart[16] on Al samples of unknown roughness. The ratio $\Delta Y_S(\hbar\omega)/Y(\hbar\omega)$ could thus be calculated and is plotted in Fig. 3.

In Fig. 3 we see the qualitative confirmation of the results of Eq. (5). Shown in addition to the experimentally determined ratio $\Delta Y_S(\hbar\omega)/Y(\hbar\omega)$ is the value of this ratio as predicted from the experimentally determined ratio of absorbed photons, $\Delta R_S(\hbar\omega)/\Delta R(\hbar\omega)$, and the calculated ratio, $\kappa(\hbar\omega)/\alpha(\hbar\omega)$, of the relative densities of these photons. The predicted yield ratio is in good agreement with the measured yield. Magnitudes are comparable, and the predicted enhancement of the photoyield in the short plasmon-field decay-length region near $\hbar\omega = \hbar\omega_p/\sqrt{2}$ is in strong evidence. Almost all of the assumptions which we have made break down as the calculated $[\Delta R_S/\Delta R] \times (\kappa/\alpha)$ approaches $\hbar\omega = \hbar\omega_p/\sqrt{2}$. Thus this expression is terminated on Fig. 3 at $\hbar\omega < \hbar\omega_p/\sqrt{2}$.

Reflectance and photoemission measurements are continuing on substrates of varying, known roughness. Studies are being extended to metals such as silver for which the assumptions of free-electron surface oscillations and high reflectance are no longer valid.

The authors would like to express their deep appreciation for helpful suggestions and information from Dr. H. E. Bennett and Dr. R. H. Ritchie, and particularly for valuable conversations with Dr. J. L. Stanford.

*Work supported by the Advanced Research Projects Agency through the Center for Materials Research at Stanford University and by the National Science Foundation.

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LIQUID-VAPOR-LIKE CRITICAL POINTS IN ANISOTROPIC FERRIMAGNETS*

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A magnetic critical phase transition which involves no change in symmetry is described theoretically. In the example, $Yb_6Fe_{10}O_{24}$ with applied field along [100], a critical point is predicted at $T^c = 7.51°K$ and $H^c = 43\,640$ Oe. There is a large associated anomaly in C_H: $C_H(H=H^c) \approx 0.3(T-T^c)^{1/\delta-1} R$ mole^{-1}, where δ should realize the classical value 3.

Traditional phase transitions in magnetic systems are characterized by broken symmetry. In a space of external variables which do not themselves change the symmetry, the phases must be completely separated by phase transition boundaries. We describe here a magnetic transition between phases of like symmetry. The boundary need not be complete, and in general it should end in a critical point reminiscent of that in liquid-vapor systems. The critical point occurs for some substances in experimentally accessible magnetic fields, but no measurements of the predicted anomalies have yet been reported.

We illustrate the specific-heat anomaly at a critical point in ytterbium iron garnet (YbIG, $Yb_6Fe_{10}O_{24}$). Remarkably, true classical critical behavior is expected for this case. In other systems there should be some effect due to fluctuations. In any case the critical exponents expressing the nature of the singularities in thermodynamic functions should be different from any which have heretofore been observed.

As is well known[1] a ferrimagnet near its compensation temperature likes to assume a canted configuration in an applied field. The angle which a given sublattice makes with the field depends on the field magnitude and on the size of the various (temperature-sensitive) sublattice moments. Treating one particular sublattice angle as a free variable, we may say that the free energy (from exchange and Zeeman interactions) has a minimum at the equilibrium canting angle, and that this exchange minimum moves in angle as field and temperature vary. We add anisotropy by superimposing a background with wells at the sublattice easy directions.[2] In high fields the exchange minimum can be steeper than the wells and as it sweeps along (as, for example, when the temperature passes a compensation point) it can carry the sublattice continuously between them. In low fields the minimum is not so steep and the system will only be able to follow by jumping in first-order transitions to the an-