PLASMA RADIATION FROM METAL GRATING SURFACES*†

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Surface plasma oscillations at optical frequencies have been detected by their radiation from an optical grating surface permitting a measurement of their dispersion relation.

A type of surface-plasma oscillation (SPO) in thin metal films which radiates electromagnetic waves at the bulk plasma frequency was first predicted by Ferrell.¹ and subsequently was experimentally verified.² However, there are other SPO which do not normally couple to the radiating electromagnetic field but are guided along the film surface. Wavelengths of the order of 10 Å of these types of SPO, which occur on metal films of all thicknesses, are responsible for some of the discrete energy losses suffered by electrons interacting with metal films.³ At these short wavelengths the frequency of the SPO are practically independent of wavelength. However, at longer wavelengths the frequency starts decreasing in such a way that the phase velocity of the SPO along the surface remains slightly less than that of light. Such SPO are not detected in electron-energy-loss measurements since they do not produce any discrete losses. The expression for the dispersion relation for these SPO traveling along a plane boundary between a semi-infinite metal and free space is^4

$$k^{2} = \left(\frac{\omega}{c}\right)^{2} \left(\frac{\epsilon_{1}(\omega)}{1 + \epsilon_{1}(\omega)}\right), \tag{1}$$

where ω is the angular frequency of the SPO, k is its wave number, c is the velocity of light in vacuum, and $\epsilon_1(\omega)$ is the real part of the dielectric function of the metal. In order for the SPO to exist, $\epsilon_1(\omega) \leq -1$. This type of SPO was first studied theoretically by Sommerfeld⁵ in regard to radio-wave propagation along the earth's surface and is also called the Sommerfeld surface wave. It's presence has been required to explain some of the Wood's anomalies on optical gratings.⁶ There therefore exists much theoretical and indirect experimental evidence to support the existence of these SPO at optical frequencies, but no direct experimental evidence. In this Letter such direct experimental evidence is presented which gives a rather detailed description of their properties.

On a flat surface the SPO does not radiate because its phase velocity is less than that of light. However, any surface roughness permits the surface to impart some additional momentum to the SPO so that it can couple to the radiating electromagnetic field.⁷ Then the SPO can be detected by observing this radiation. In order to obtain any quantitative results, that surface roughness must be known. We were therefore led to perform the experiment on a grating surface. The experiment consisted of bombarding an optical grating of 1200 lines/mm with normally incident 10-keV electrons, and observing the emitted light. The stainless-steel grating surface is usually coated by vacuum evaporation with aluminum metal, though a silver coating was also used. The results were similar for both coatings. The coating thickness varied between about 0.35 and 1.5 μ . Even though such coatings are much thicker than the depth of the grating grooves, the profile of the grating surface should not be much changed. The coating adds thickness uniformly to all parts of the grating profile, to the peaks and to the valleys. A typical result is shown in Fig. 1(a) where the intensity of 5400-Å light is plotted as a function of emission angle from the grating normal. The plane of emission of the light determined by the normal to the grating and the direction of observation makes an angle φ of 6° with the grating rulings. The intensities of the two polarizations are plotted separately, where || and \perp mean parallel and perpendicular to the emission plane, respectively. Two peaks appear in the \perp polarization and no appreciable peaks are discernable in the || polarization.



FIG. 1. (a) Angular distribution of the emitted radiation of 5400 Å in the electron-beam experiment. The ruling of the grating makes an angle of 6° with respect to the emission plane. $\|$ and \perp mean that the polarization of the radiations are parallel and perpendicular to the emission plane, respectively. The emission plane is the plane determined by the normal to the grating surface and the direction of emitted radiation. There are two peaks in the \perp polarization. (b) Angular distribution of the reflected light of 5400 Å in the specular-reflectance experiment. The ruling of the grating makes an angle of 5° with respect to the incident plane. There are two dips in the \perp polarization.

The angular position of both peaks shifts to the normal of the grating as the wavelength of the radiation increases. As the angle φ between the grating rulings and the emission plane is increased, the splitting between the peaks increases: One peak shifts toward, while the other shifts away from the normal of the grating. As the angle between the rulings and the emission plane reaches 90°, i.e., the ruling of the grating is perpendicular to the incident plane, the peaks in the \perp polarization have disappeared and reappeared in the || polarization. Further study indicates that the radiation is always polarized with its plane perpendicular to the grating rulings. The line shape of the peak is influenced by the surface condition of the metal grating and by the place on the grating surface⁸ where the electron beam bombards. The metal grating surface deteriorates after an extended electron bombardment. However, the angular positions of the peaks are still the same despite the deterioration of the grating surface or exposure to the atmosphere. The position of the peaks also did not change as the bombarding electron energy was varied from 7 to 12 KeV. There was no attempt to monitor intensity changes as the electron energy varied.

It is suggested that the cause for this anomalous radiation from the metal grating is the SPO. A verifying experiment suggests itself. If the bombarding electrons excite the SPO which subsequently radiate, then the inverse process of exciting the SPO should be present. Electromagnetic radiation incident at the emission angle of the anomalous radiation of the same frequency should be anomalously absorbed as it excites the SPO. Such is the case. One of the results from the specular-reflectance experiment for 5400-Å incident light is shown in Fig. 1(b). The ruling of the grating is at 5° with respect to the incident plane, which is defined by the incident direction and the normal of the grating. The x axis of the graph is the angular position of the reflected light with respect to the normal of the grating. The y axis represents the intensity of the reflected light in arbitrary units. There are also two dips in the angular distribution curve for the reflected light of one polarization. The angular positions of the dips are practically the same as the emission peaks in the electron beam experiment for the same wavelength of radiation and the same orientation of the grating. Moreover, the dips occur only when the light has an electric field component normal to the rulings, in agreement with the polarization of the emitted light. The line shape of the dips is also greatly influenced by the grating-surface condition and by the places on the grating surface where the incident light falls.

From the data of these experiments we were able to determine the wave number k_{\parallel} of the SPO responsible for the emitted radiation by calculating the component of the wave number of the emitted radiation parallel to the surface. A SPO with a wave vector \vec{k} on a flat surface would, on a grating surface, consist of some linear combination of wave vectors $\vec{k} + n\vec{g}$, where *n* can have any positive or negative integer value including 0 and $g = 2\pi/d$ with the ruling spacing equal to *d*. \vec{g} is directed normal to the grating rulings. When $|\vec{\mathbf{k}}+n_0\vec{\mathbf{g}}| \leq \omega/c$ for some n_0 , the SPO can radiate in a direction θ from the grating surface normally given by

$$\sin\theta = \frac{|\vec{k} + n_0\vec{g}|}{\omega/c} = \frac{k}{\omega/c}$$

In Fig. 2(a) the data are analyzed in this manner. The dashed curve is obtained directly from $k_{\parallel} = (\omega/c) \sin\theta$ for light of 5000-Å wavelength. (The direction and magnitude of k_{\parallel} is given by the tangential component of the wave vector of the emitted or absorbed radiation.) The $\mathbf{\tilde{k}}$ of the SPO plotted as the solid curve is determined by either adding or subtracting $\mathbf{\tilde{g}}$ to $\mathbf{\tilde{k}}_{\parallel}$, whichever is more appropriate. The observed double peaks in the emitted and absorbed radiation can be understood from Fig. 2(a). Based on the experimental data the figure illustrates the \vec{k} of the SPO versus its angle with the ruling direction for constant frequency. Also illustrated are the corresponding curves for $\vec{k} \pm \vec{g}$. The dotted lines are those parts of the curves which were not seen directly experimentally but are reasonable extrapolations. Radiation is emitted along a given direction of the grating if the intersection of a line in that direction with the curves occurs at a magnitude of wave number less than ω/c . For $\varphi = 0$ this occurs for only one angle in the range of observations. (The apparatus was not able to observe angles θ less than 35° from the grating normal or greater than about 87°.) For a range of $\varphi \neq 0$ this occurs at two angles. Such an analysis can explain all of the observed data.

The polarization of the emitted and absorbed radiation indicates that the electric field of the observed SPO is in the plane determined by the normal to the grating surface and \vec{g} . On a flat $surface^{4,5}$ the electric field for the SPO is in the plane determined by the surface normal and \vec{k} , and is thus modified by the grating surface. Yet, the plot of \vec{k} versus angle shown in Fig. 2(a) is surprisingly circular, indicating that the effect of the grating rulings on that property is small. For example, no gaps at the Brillouin zone boundaries were detected. In Fig. 2(b) the ω -vs-k characteristics of the SPO at 40° to the rulings are plotted. On the same curve the corresponding curves for light and SPO on a flat aluminum surface calculated from Eq. (1) are also plotted. The measured curves indicate that the



FIG. 2. (a) Plot of the wave number \vec{k} of the surface plasma oscillation that propagates along the grating surface versus its direction with respect to the rulings. Zero degrees is defined for \vec{k} parallel to the ruling. Shown are the curves for constant frequency corresponding to the wavelength of light of 5000 Å. The curves are three circles. One is centered about k = 0and the others are centered at $k = \pm g$. (b) A plot of the \vec{k} of surface plasma oscillations determined by this experiment versus their angular frequency ω on a grating surface. The points are for \vec{k} directed at 40°. Curve A is the corresponding plot for light in free space and the Curve B is the theoretical expression for surface-plasma oscillations on the flat surface of aluminum.

grating slightly slows up the SPO compared with a flat surface, but again confirms that the ω -vs-k characteristics of the SPO are not much affected by the grating rulings.

In conclusion, it is important to distinguish this experiment from one performed by Smith and Purcell.⁹ In their experiment light radition was observed when a grating was bombarded by an electron beam traveling at a glancing angle (almost 90° to the grating normal) and at right angles to the rulings. In that case, the radiation is produced by the induced image charges of the electrons oscillating normal to the surface as the electron beam passes by the rulings. In our experiment, the bombarding electrons are directed normally to the grating surfaces exciting SPO which then radiate. The dispersion relations of these SPO can be obtained in this fashion as illustrated in Fig. 2.

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FERMI-LIQUID EFFECTS ON PLASMA WAVE PROPAGATION IN ALKALI METALS

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The dispersion of plasma-wave propagation in the alkali metals sodium and potassium has been observed to deviate significantly from that expected for a noninteracting electron gas. Modifications of the dispersion have been calculated using the Landau Fermiliquid theory and a quantitative evaluation of the first two moments of the interaction function has been made for the case of potassium.

Recently we reported the observation of a new class of wavelike excitations in potassium which propagate perpendicular to a magnetic field near the Azbel'-Kaner cyclotron resonances $n\omega_c = \omega^{1}$. The clearest data were obtained with microwave surface currents J flowing parallel to H ($J \parallel H$, "ordinary mode") at field values above the fundamental resonance. The measured dispersion relation (wavelength versus H) for these waves proved to be in quite good agreement with that calculated for a degenerate free-electron gas having an effective mass appropriate to potassium. Later experiments have revealed similar propagation "windows" at each of the first few subharmonics in both the $J \parallel H$ and the $J \perp H$ ("extraordinary mode") polarizations.² While the free-electron model is generally successful in accounting for the qualitative existence of all the waves

near resonances,³ it has become apparent that it fails to account quantitatively for the experimental data. The discrepancy is particularly severe for $J \perp H$ near $\omega = \omega_C$. In this Letter we show that the discrepancy is removed if electron correlations are included in the model.

Using the Landau Fermi-liquid theory, we account for the experimental dispersion relation and directly evaluate the first two moments of the Landau interaction function. In principle, precise measurements of the dispersion relation of all the modes would provide a complete quantitative evaluation of all moments of the spin-independent interaction function. The latter is independent of and complementary to the spin-dependent interaction which gives rise to the spin waves recently observed in sodium and potassium.^{4,5}

The experimental details remain essential-