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Observation of surface plasmons by transition radiation from smooth aluminium films

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Résumé. — Cette lettre décrit la première observation expérimentale d'une résonance due aux plasmons de surface, dans le spectre du rayonnement de transition de films d'aluminium évaporés sur des prismes en quartz et bombardés par des électrons de 20 keV. Cette résonance est comparée au minimum caractéristique observé dans des mesures de réflectivité totale atténuée, effectuées sur les mêmes systèmes. La forte corrélation des deux résultats et une analyse théorique du rayonnement de transition permettent d'identifier sans ambiguïté la résonance du rayonnement de transition.

Abstract. — We report the first observation of a strong resonance due to surface plasmons in the transition radiation emitted by films of aluminium deposited on quartz prisms and bombarded with 20 keV electrons. Measurements of the reflectivity of p-polarized light on the same systems show the characteristic attenuated total reflection dip. The correlation of both results and a theoretical analysis allow an unambiguous identification of the resonance phenomenon.

Light is emitted, when charged particles with a constant velocity cross a plane interface between two media of different dielectric constants. This radiation is called the transition radiation [1], it has in general a smooth continuous spectrum and is emitted in almost every directions. Resonances can however be observed in this emission. For instance at a vacuum-metal interface, there is a resonance at the plasma frequency, this resonance is strong for a thin film of a good free electron metal. Resonances associated to the interface electromagnetic modes exist and, they also can be observed, when with a suitable second interface, these modes are allowed to leak radiatively. In this letter, we report the first observation of an interface plasmon resonance in the transition radiation from smooth aluminium surfaces, in suitable configuration.

It is known that charged particles can excite surface plasmons, but in general these modes are not observed radiatively as their phase velocity is greater than that of light in the two media. Non radiative surface plasmons excited by electrons on rough surfaces can however radiate light by interchanging momentum with the surface [2].

Otto [3] has shown that with an appropriate second interface parallel to the interface where plasmons are excited, one can detect optically these interface modes. In this technique of attenuated total reflection (ATR), light propagating in a medium with a refractive index $n > 1$, is incident with an incidence angle greater than the total reflection angle on a vacuum gap between the dielectric and the active interface, and can couple its evanescent component to the plasmons on the active vacuum-metal interface.

Kretschmann [4] offered an alternative scheme in which the vacuum gap is absent and the metal is a film, thin enough, for the totally reflected evanescent wave to cross it and excite plasmons on the vacuum-metal interface. To observe interface modes with transition radiation, Kröger and Raether [5] and Bishop and Maradudin [6] proposed the same schemes. The results reported here are the first successful observation of the resonance due to interface plasmons in the transition radiation.

Consider a beam of electrons impinging normally on an aluminium film deposited on a quartz prism (see insert of figure 1). Interface plasmons are excited at the metal vacuum boundary. For suitable combination of frequency and momentum, that satisfy the dispersion relation, the second interface can make

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Fig. 1. — Calculated transition radiation power, per unit wavelength interval, per unit solid angle, per unit beam current $\frac{1}{i} P_{\lambda,\Omega} = \frac{1}{i} \frac{\partial^2 P}{\partial \lambda \partial \Omega}$ from a 200 Å thick aluminium film, emitted at $\lambda = 3500$ Å, as a function of the observation angle θ in the prism. Energy of the electron is 20 keV. Also shown is the ATR p-polarized reflectivity R_p in Kretschmann configuration (see insert).

them radiate. Following Pafomov [1], we calculate an analytical expression for the transition radiation as a function of the electron velocity, the frequency ω , the angle of observation θ , the different dielectric constants and the film thickness d . This expression exhibits a resonance in the smooth radiation pattern when the parameters satisfy the interface mode dispersion relation [7]. Then, from an observation of this resonance in the radiated pattern, we can determine experimentally the dispersion relation of the interface plasmons. This resonance can be studied either at fixed wavelength as function of the angle θ or at fixed angle as function of the wavelength. In our experimental conditions, we found it easier to study the radiated energy as a function of the observation angle. It is clear that the quartz prism plays here the same role as in an ATR experiment. In figure 1, we give the results of a calculation of the transition radiation power radiated per unit wavelength interval, per unit solid angle, per unit beam current, and of the reflection coefficient R_p for the same Kretschmann configuration, as a function of the observation angle θ in the prism. The different features (width, amplitude, angular position) of the transition radiation resonance

and of the dip of reflectivity are closely related and it is hoped that transition-radiation will provide a tool to study electronic structure of interfaces. This experiment however presents a difficulty, which perhaps explains the past failure to observe the phenomenon : as electrons go through the dielectric an unavoidable strong luminescence is excited. It is only by taking advantage of the sharp resonance that the transition radiation signal can be separated from this bright noise. Bremsstrahlung contributes also to the light emission but for the energy of the electrons, the different materials (aluminium and quartz), it is quite negligible.

The aluminium films, about few hundreds angströms thick, are heat evaporated on fine polished quartz substrates in a vacuum of about 10^{-7} torr. The quartz substrate is then removed from the evaporator and used in the experimental chamber as an optical window (Fig. 2). During this operation, the Al film stays about ten minutes in air before being

Fig. 2. — Schematic drawing of the experimental system.

put again in a vacuum of about 10^{-8} torr. A 60° quartz prism is coupled to the outside of the sample window with an index matching oil. With this experimental arrangement, all the optical components are exterior to the experimental chamber and placed on a rotating table. A 20 keV electron gun is opposite the sample window and the electron beam is incident normally on the film. The light detection system consists of a polarizer, a monochromator and a photon counting system. To investigate the relatively narrow surface plasmon resonances, an angular resolution of 0.64° , corresponding to a solid angle in the prism $d\Omega = 1.7 \times 10^{-4}$ str, has been chosen.

Figure 3 shows the experimental angular dependence of the total radiation detected through the prism at a wavelength of 3500 Å, from a film about 200 Å thick. The wavelength has been chosen to correspond to a spectral region where the fluorescence of the quartz window (without an aluminium film deposited on it) is the weakest. The spectra for both polarization relative to the plane of observation are shown not corrected for the absolute spectral response of

Fig. 3. — Experimental angular dependence of the radiation detected per second through the prism at $\lambda = 3\,500\text{ \AA}$ in the wavelength interval $\Delta\lambda = 26\text{ \AA}$, solid angle $\Delta\Omega = 1.7 \times 10^{-4}\text{ str}$ and for $1\text{ }\mu\text{A}$ beam current. The background radiation is due to the quartz luminescence and a small contribution of p-polarized transition radiation.

Fig. 4. — Angular dependence of the transition radiation deduced from figure 3. The intensity unit incorporates the measured efficiency of the system plus the influence of the prism, as the observation angle θ is measured in the prism. Also shown is the ATR p-polarized reflectivity observed at the same spot of the same film.

the optical system. The s-polarized light intensity is a monotonic increasing function of the observation angle θ in the prism. The p-polarized light intensity shows a peak superimposed to a monotonic increasing background. Calibration with a tungsten standard lamp of the absolute efficiency of the optical detection system for both polarizations indicates that the light emitted through the prism is almost unpolarized except for the p-polarized emission peak. As the absolute intensity of this unpolarized background is also of the same order as the measured fluorescence of a quartz window without an aluminium film, we can interpret the background as fluorescence noise from the prism. Figure 4 shows the angular dependence of the difference between the p-polarized intensity of figure 3 and the p-component of the background luminescence. This spectrum accounts for the efficiency of the optical system. The process of data reduction leaves an uncertainty of about 20 W/cm.str.A on the background p-polarized radiation. This is of the order of the transition radiation power, calculated off resonance. On the same figure is shown the experimental ATR reflectivity from the very same spot of the same film and for p-polarized light.

Comparison of these two spectra with the calcu-

lated ones (Fig. 1), using the experimental values of the parameters and a plausible value of the aluminium dielectric constant [8], shows that the experimental reflectivity dip is shifted relative to the experimental transition radiation resonance in the same direction and with about the same amplitude as predicted from theory. The angular position and the half width of the experimental reflectivity dip are seen to be greater than the calculated ones. Several reasons can explain this fact : presence of an oxide film due to the air exposition of the aluminium film, approximate value of the aluminium dielectric constant. The same reasons and the presence of an oil film between the quartz window and the prism is quite sufficient to explain the smaller experimental ATR reflectivity observed for observation angles approaching the total reflection critical angle. In consequence, only a good qualitative comparison can be made here between the experimental and calculated transition radiation angular spectra. But it remains that experimentally resonances in the ATR and transition radiation spectra are the same. We have verified in particular that transition radiation resonance is not observed when the strength of the ATR resonance is too weak. This is the case for example when intentionally too thin films are studied. This strong correlation allows an unambi-

guous identification of the experimental transition radiation resonance as due to interface plasmons.

Experiments are now going on to determine of surface plasmon dispersion curve and probe the sensitivity of the technique to modification of the vacuum-aluminium interface.

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