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Influence of surface plasma waves on the surface and volume photoemission yields from Al thin films in the photon energy domain 1.5 to 4 eV

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Résumé. — L'excitation d'ondes de plasma de surface (OPS) par la radiation incidente peut renforcer énormément l'émission photoélectrique de surfaces métalliques. Les contributions respectives des effets de volume et de surface ont été déterminées quantitativement en présence d'OPS pour des films minces d'aluminium activés au césium, en excellent accord avec les valeurs expérimentales, dans tout le domaine d'énergie $1.5 < \hbar\omega < 4$ eV.

Abstract. — Excitation by the incident light of surface plasma waves (SPW) may increase dramatically the electron emission from metal surfaces. Respective enhancements of volume and surface photoeffects are quantitatively determined for smooth thin cesiated aluminium films, in excellent agreement with measured values of the photoyield in the whole photon energy domain $1.5 < \hbar\omega < 4$ eV.

Electron photoemission from a metal surface illuminated by low energy photons may be analysed by using a theory based upon a description of the metal by its local dielectric function [1, 2, 3], and the consideration of two distinct origins for the emitted electrons : a part of them come from the volume of the metal (volume effect) and the other part from an infinitely thin layer at the surface (surface effect).

Using these theory, we have tried to calculate the respective contributions of volume and surface effects to the electron emission from aluminium thin films surfaces on which surface plasma waves are excited by p-polarized light through a prism coupler.

For a thin metal film of thickness d , that we consider as perfectly smooth, illuminated with a monochromatic light, the volume photoemission yield can be calculated by the following expression [1, 2] derived from the three-step model :

$$Y(\varphi) = \int_0^d C_0 \exp \frac{-z}{L} \cdot \eta(z, \varphi) dz = C_0 F(\varphi) \quad (1)$$

$C_0 \exp(-z/L)$ may be interpreted as the probability for a photon absorbed at the distance z below the surface to produce photoemitted electrons. L is the escape depth of the electrons — and $\eta(z, \varphi)$ the volume

density of absorbed energy reported to the incident flux density on the surface — φ is the angle of incidence of the radiation. Expressions giving $\eta(z, \varphi)$ may be found in [1] and [4].

The factor C_0 , which is assumed to be independent of the direction of the electric field, does not depend on the polarization of the light. L and d being known, $F(\varphi)$ may be easily calculated, but an accurate value of C_0 cannot be obtained from theoretical expressions.

Many authors have published the values of the optical constants of Al, but we have preferred to measure these constants for each metal film [5] in order to calculate with a good precision the absorbed energy density $\eta(z, \varphi)$ and the function $F(\varphi)$. The length L was also obtained from the measurement of the photoemission yields respectively for a front illumination and a back (through the prism) illumination of the film, the radiation being s-polarized [5].

For a metal film having a perfectly smooth surface illuminated with s-light, the volume photoeffect is the only source of electrons. But the existence of microroughnesses may be the origin of a surface effect as a component of the wave electric field normal to the metal surface is then present.

A proof of the smoothness of the films was found in the good agreement between theoretical and experimental results concerning the variations of the volume effect as a function of the incidence angle [5].

(*) ERA 660.

In these conditions, $\sigma_s(\varphi)$ represents with a good accuracy the experimental volume photoemission yield for an s-polarized light and we are able to calculate C_0 from the relation

$$Y_s(\varphi) = C_0 F_s(\varphi) \equiv \sigma_s(\varphi). \quad (2)$$

Using p-polarized light at oblique incidence, the experimental yield $\sigma_p(\varphi)$ is always greater than $\sigma_s(\varphi)$, the difference originating essentially in the surface effect. The surface photoemission yield $S(\varphi)$ is assumed to be proportional to the square of the electrical field normal component $E_z(0)$ existing on the metal side of the surface ($z = 0$) and we may write [3]

$$S(\varphi) = K_0 \eta_z(0, \varphi) \quad (3)$$

where $\eta_z(0, \varphi)$ is the density of absorbed energy at $z = 0$ corresponding to the z component of the electric field inside the metal. K_0 as C_0 is a function of the photon energy $\hbar\omega$, not of the incidence angle φ .

For the calculation of $\eta_z(0, \varphi)$ we assume that the film has a perfectly clean and smooth surface, neglecting a possible photoemission from surface states. K_0 can be obtained from the experimental surface photoemission yield $S_{\text{exp}}(\varphi)$, and the calculated value of $\eta_z(0, \varphi)$:

$$K_0 = S_{\text{exp}}(\varphi) / \eta_z(0, \varphi) \quad (4)$$

$S_{\text{exp}}(\varphi)$ may be given by the expression

$$S_{\text{exp}}(\varphi) = \sigma_p(\varphi) - Y_p(\varphi) = \sigma_p(\varphi) - \sigma_s(\varphi) \frac{F_p(\varphi)}{F_s(\varphi)}. \quad (5)$$

The second factor in the right member corresponds to the volume effect $Y_p(\varphi)$ in p-polarized light, which differs from $\sigma_s(\varphi)$ as the absorbed energy density depends on the light polarization.

Our experimental arrangement to study volume and surface photoemission has been described elsewhere [6].

In order to measure a photoelectric emission current throughout the visible domain, the work function of Al is lowered from about 4 eV to 1.5 eV by evaporation on the Al surface of a very thin layer of cesium. This Cs film, of thickness less than a monolayer, remains stable for several weeks only if the pressure is maintained under 10^{-10} torr. The presence of Cs — probably under a ionized state — does not appreciably disturb neither the surface smoothness, nor the plasmon resonance curves and consequently the optical constants of the Al film [5]. In these conditions a simple three media model (silica-Al-vacuum) with plane interfaces may be used to calculate $\eta(z, \varphi)$ and $\eta_z(0, \varphi)$.

The film being illuminated on its front face, we measure for a fixed value of φ ($\varphi = 45$ or 70 degrees) the values of $\sigma_s^+(\lambda)$ and $\sigma_p^+(\lambda)$. From these data, and from (2) and (5), we obtain the photoemission yields of the volume and surface effects, respectively $Y_p^+(\lambda)$

Fig. 1. — Surface S_{exp}^+ and volume (Y_p^+) spectral photoemission yields in mA/watt of incident light for a cesiated Al film illuminated with p-polarized radiation on its front face, at two angles of incidence ($\varphi = 45$ and 70 degrees).

and $S_{\text{exp}}^+(\lambda)$, represented on the figure 1 and expressed in $\mu\text{A/watt}$. The incident power is measured with a calibrated silicon photocell.

The surface effect is more important than the volume effect for photon energies near the emission threshold. The volume effect increases with photon energies and prevails in the UV domain. In addition, from the figure 1 it seems that the energy threshold for the surface effect could be slightly lower (a few 10^{-2} eV) than the volume effect threshold, in agreement with the rules governing the wave-vector variations in the two types of electron photon interactions [7].

In order to check the validity of these results, we have used the preceding method and the value of K_0 obtained by (4) from our front illumination experiments to calculate the respective enhancements of the volume and surface photoemission yields which can be observed by excitation of SPW on the metal interface, the film being illuminated through the prism (ATR method) [8].

We have measured $\sigma_s^-(\varphi)$ and $\sigma_p^-(\varphi)$ for several values of λ and various values of φ ranging from 42 to 46° . Making again the hypothesis that

$$Y_s^-(\varphi) \equiv \sigma_s^-(\varphi),$$

we determine $Y_p^-(\varphi) = \sigma_s^-(\varphi) \cdot F_p^-(\varphi) / F_s^-(\varphi)$.

The spatial repartition of the energy absorbed in the Al film varies dramatically when SPW are excited by p-polarized light. Far from the SPW resonance, the absorbed energy decreases continuously from the prism to the emitting surface. On the contrary, at the resonance angle the absorbed energy density attains its maximum along the emitting surface, and the $E_z(0)$ field component into the metal is highly increased, both phenomena inducing an important enhancement of the photoemission current.

$E_z(0)$ and $\eta_z(0)$ have been calculated as functions of φ . Using the values of K_0 obtained as a function of $\hbar\omega$ for $\varphi^+ = 45^\circ$, we are now able to calculate the theoretical value of the surface photoemission yield $S_{\text{th}}(\varphi)$. Both effects, $Y_p^-(\varphi)$ and $S_{\text{th}}^-(\varphi)$ are represented

Fig. 2. — Enhancement by a SPW excited near the photoelectric threshold (at $\lambda = 765$ nm) of the surface and the volume photo-yields for a cesiated Al film 17 nm thick evaporated on a silica prism and illuminated through the prism. (—·—) calculated volume effect Y_p^- ; (---) surface effect S_{th}^- ; (—) total calculated value of the photoyield; ... experimental values. ϕ_L is the limit angle of reflexion in the prism.

on the figure 2 for $\lambda = 765$ nm, a wavelength near the energy threshold, and on the figure 3 for $\lambda = 500$ nm. The total theoretical photoemission yield

$$T_{th}^-(\phi) = \{ Y_p^-(\phi) + S_{th}^-(\phi) \}$$

is also represented, as well as experimental points ($\sigma_p^-(\phi)$).

The surface effect prevails for photon energies near the energy threshold and is responsible for about 80 % of the total photoyield. The absolute value of the surface effect increases continuously with the photon energy in the range $1.5 < \hbar\omega < 4$ eV, but less rapidly than the volume effect. At $\lambda = 500$ nm both effects are equivalent, with a maximum quantum photoyield $\phi_{max} \simeq 2 \times 10^{-3}$ el/photon. At shorter wavelengths, the surface effect increases again and remains slightly above the volume effect.

A comparison between the curves show that neither the volume effect, nor the surface effect is able to explain by itself the exact amplitude and position of the photoemission peak and the shape of the experimental curve $\sigma_p^-(\phi)$.

Fig. 3. — Results obtained with the same film at $\lambda = 500$ nm. See figure 2 for the meaning of the curves. Note the change in scale for the photoields.

Theoretical curves $Y_p^-(\phi)$ and $S_{th}^-(\phi)$ attain their respective maximum for two distinct values of ϕ , which are both different from the angle corresponding to the maximum of the experimental curve $\sigma_p^-(\phi)$. The angular separation between them (a few tenths of a degree) originates in the relatively large value of the imaginary part of the complex dielectric constant of Al, and could not be observed with a very low absorption metal as Ag [9].

However, the theoretical values $T_{th}^-(\phi)$ are in excellent agreement with the experimental results $\sigma_p^-(\phi)$, as well for the exact position of the peak, than for the absolute values of the yield in the whole angular domain of surface plasma wave excitation, and for all photon energies ranging from 1.5 to 4 eV.

These results show at the same time the good optical quality of our Al films, and the possibility to analyse experimental data by using a theory for the photoelectric yield based upon a description of the photoemitter by a local isotropic dielectric function [1, 3] and in which volume and surface photoeffects are considered as distinct phenomena.

In conclusion, for photons frequencies very low compared to the plasma frequency of the photoemitter, it is not necessary to have recourse to the recent non-local theories [10, 11] which attempt to describe photoemission as a unique physical phenomenon. A description of the experimental methods and a more detailed discussion about the results presented here will be published in a near future.

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