Radiative Decay of Non Radiative Surface Plasmons Excited by Light

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(Z. Naturforsch. **23 a**, 2135—2136 [1968]; received 15 November 1968)

There are two modes of surface plasma waves:

1) Non-radiative modes with phase velocities ω/k smaller than the velocity of light c. They cannot decay into photons in general.

2) Radiative modes with $\omega/k > c$ which couple directly with photons [1].

The following paper is concerned with the excitation of these modes by light and their decay into photons. It has been shown that the radiative mode on thin silver- and potassium-films can be excited by light and that the mode reradiates light almost into all directions with an intensity maximum at the plasma frequency ω_p [2]. It had been further observed that the non-radiative modes radiate under certain conditions if they are excited by electrons (grazing incidence of electrons on a rough surface [3] or at normal incidence on a grating [4]).

The mechanism of this emission is in these cases always the same: The "wave vector" of the roughness of the surface or its irregularity changes the plasmon wave vector k so that

a) in the case of the radiative mode light emission is found in directions in addition to that of reflexion and transmission,

b) in the case of the non-radiative mode its wave vector is reduced so that the condition $k < \omega/c$ is fulfilled and the mode is able to radiate.

The theoretical considerations [5] are in good agreement with the observations.

The experiments described here shall demonstrate that the non-radiative mode excited by light can also radiate. The non-radiative mode is produced by the inhomogeneous light wave obtained by total reflexion inside a quartz prism as proposed in [6]. If the angle of incidence inside the prism is Θ_0, the wave vector of the inhomogeneous wave is $(\omega/c) \cdot \sqrt{\varepsilon_q} \cdot \sin \Theta_0$ ($\varepsilon_q =$ 2.16 for quartz) and thus can excite a non radiative mode on the boundary of the prism for $\sqrt{\varepsilon_q} \sin \Theta_0 > 1$ or $90° > \Theta_0 > 43°$. If one vaporises a silver film *directly* on the quartz surface the inhomogeneous light wave penetrates into the silver film and excites a non-radiative mode on the boundary silver/air. The excitation will be highest for those frequencies which fulfill the dispersion relation of these surface plasmons.

The dispersion relations at the boundary of (bulk) silver/air and silver/quartz are [7]

$$k^2 = \left(\frac{\omega}{c}\right)^2 \frac{\varepsilon}{\varepsilon+1} \quad \text{and} \quad k^2 = \left(\frac{\omega}{c}\right)^2 \frac{\varepsilon\,\varepsilon_q}{\varepsilon+\varepsilon_q}$$

with ε, ε_q the real parts of the dielectric constants of silver and quartz. These relations are little modified by the fact that the silver has a finite thickness. At a thickness of about 1000 Å Ag the difference between the exact formula and the formulas given above is not important.

Fig. 1 demonstrates the dispersion relations and shows that those surface oscillations on the silver-air-boundary can be excited which lie between $\omega/k = c$ and $\omega/k = c/\sqrt{\varepsilon_q}$ whereas those on the quartz-silver-boundary cannot be excited by light. The light emitted by these non-radiative surface plasmons by means of the roughness wave vector from the silver surface should be observable.

In the experiments one side of the quartz prism (60°) is covered with a 800 Å Ag-film. A light beam of a Xenon lamp polarised parallel to the plane of incidence penetrates the prism and is totally reflected at the quartz-silver-boundary. Outside the prism on the other side of the silver film in air a monochromator together with a multiplier can be turned around the normal of the irradiated foil to detect the radiation, so that its angular distribution and the dependence on the wave length of the incoming light could be measured.

If $\Theta_0 > 43°$, and light polarised parallel to the plane of incidence is used, the λ-dependence shows two maxima (Fig. 2). The first at 3200 Å is independent of Θ_0 and is also observed with light polarised perpendicular to the plane of incidence. This comes from light scattered at the boundary silver/quartz. Silver has a maximum transparency at 3200 Å.

The second higher maximum is only observed with parallel polarised light; it is dependent on Θ and lies at frequencies correlated with the wave vector

$$k_0 = \sqrt{\varepsilon_q} \cdot \sin \Theta_0 \cdot (\omega/c)$$

by the dispersion relation of the non radiative modes. It is remarkable that the intensity of the emitted light is higher at low frequencies which are near to the light

[1] e. g.: K. L. KLIEWER and R. FUCHS, Phys. Rev. **153**, 498 [1967].

[2] J. BRAMBRING and H. RAETHER, Phys. Rev. Letters **15**, 882 [1965]. — J. BÖSENBERG and H. RAETHER, Phys. Rev. Letters **18**, 397 [1967].

[3] H. BOERSCH and G. SAUERBREY, Optical Properties and Electronic Structure of Metals and Alloys, Ed. F. ABELÈS, North-Holland Publ. Comp., Amsterdam 1966, p. 386.

[4] YE-YUNG TENG and E. A. STERN, Phys. Rev. Letters **19**, 511 [1967].

[5] E. A. STERN, Phys. Rev. Letters **19**, 1321 [1967]. — E. KRETSCHMANN and H. RAETHER, Z. Naturforsch. **22 a**, 1623 [1967]. — R. E. WILLEAMS and R. H. RITCHIE, Phys. Rev. Letters **19**, 1325 [1967].

[6] A. OTTO, Z. Physik **216**, 398 [1968]. His measurements are analogous to the plasma resonance absorption whereas ours correspond to the plasma resonance emission of the radiative modes.

[7] taken from T. KLOOS, Z. Physik **208**, 77 [1968].

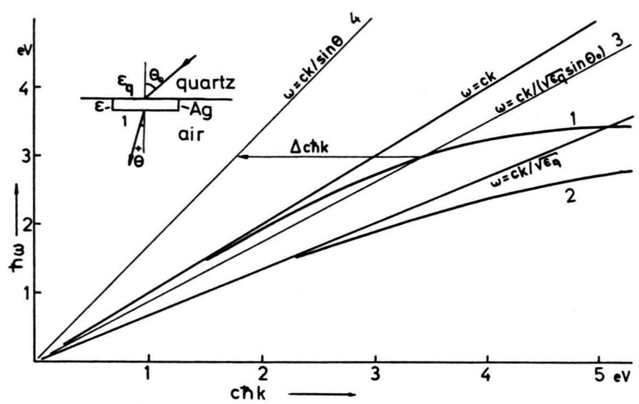

Fig. 1. Dispersion relations of the non-radiative surface oscillations at the boundaries silver/air (1) and silver/quartz (2) calculated from the optical constants [8]. The light which is incident in quartz at the angle Θ_0 is indicated by the light line (3). The maximum excitation of the surface mode takes place where line (3) cuts the curve (1). Line (4) represents the light measured at the angle Θ.

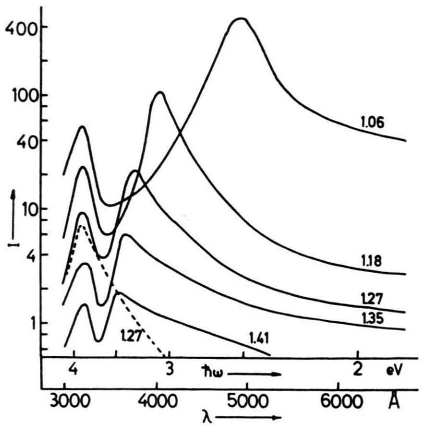

Fig. 2. Relative intensity I of the emitted light excited by parallel polarised incident light as a function of the wave length in the optimum of Θ. The dotted line corresponds to the perpendicular polarisation. The parameter of the curves is $\sqrt{\varepsilon_q}\cdot\sin\Theta_0$.

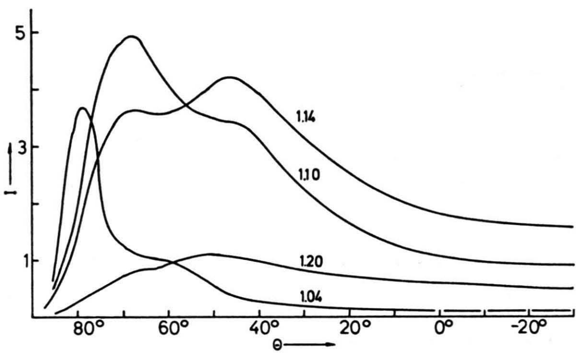

Fig. 3. The relative intensity I of the emitted light at the wave length 4500 Å and various $\sqrt{\varepsilon_q}\cdot\sin\Theta_0$ as a function of Θ.

line corresponding to a small change of the wave vector *.

If the angular distribution of this plasma radiation is measured with Θ_0 as parameter one obtains Fig. 3.

With $\Theta_0 \sim 43°$ a sharp maximum at $\Theta \sim 90°$ is observed. This indicates again that a small change of the wave vector $\Delta k = (\omega/c)\,(\sqrt{\varepsilon_q}\sin\Theta_0 - \sin\Theta)$ yields high radiation intensity. At larger Θ_0 a second less sharp maximum is observed for $\Theta \sim 50°$.

This behaviour can be understood by using the equation for the emission intensity I of the radiative modes (PREL) [5]

$$I(k) \sim \left|\frac{1+r(k)}{\varepsilon}\right|^2 k^2 \exp\{-(\tfrac{1}{2}\,\sigma\cdot\Delta k)^2\}$$

(r = Fresnel coefficient for the reflection at the boundary air-silver, σ = roughness parameter, $k = (\omega/c)\sin\Theta$).

It shows a maximum for $\Delta k = 0$ ($\Theta = 90°$) and another one for such values of Θ which make $k^2\,|1+r(k)|^2$ a maximum ($\Theta \sim 50°$).

More exact investigations of the radiation, taking into consideration the theory of Prel, can give information on the roughness of metal foils. These investigations are still worked on.

* It is also possible to reverse the light path: If the silver foil is irradiated by a source, situated in air (in Fig. 1 below the prism), light is scattered into the quartz prism with the λ dependence of Fig. 2.

[8] H. Ehrenreich and H. R. Philipp, Phys. Rev. **128**, 1622 [1962].

Nachdruck — auch auszugsweise — nur mit schriftlicher Genehmigung des Verlages gestattet
Verantwortlich für den Inhalt: A. Klemm
Satz und Druck: Konrad Triltsch, Würzburg