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Dispersion anomalies of surface plasmons on corrugated metal-insulator interfaces

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Experiments on the anomalies in surface-plasmon dispersion on periodically modulated single- and multilayered structures are analyzed. These anomalies arise if the dispersion curves intersect in the reduced Brillouin zone. Experiments on different types of gratings and for different excitation methods, e.g., excitation by light or by electrons are discussed. It is shown that the type of anomaly depends strongly on the periodicity, depth, and profile of the surface corrugations and also on the excitation mechanism and the method of detection. Three types of anomalies were found: a momentum gap which does not depend on the type of the excitation and detection, an energy gap showing the same features, and a situation in which, depending on the applied method of detection, both momentum and energy gaps were found.

I. INTRODUCTION

In the last few years numerous publications, both theoretical and experimental, have been devoted to the anomalies appearing at the intersection of surface-plasmon (SP) dispersion branches on a periodically modulated metal-insulator (e.g., vacuum) interface.[1] The surface electromagnetic waves, propagating on a periodically modulated surface (as, e.g., a metallized holographic grating) can be scattered elastically with a momentum transfer of $\kappa = \pm nG$, where $n = 1,2,3,\ldots$ $G = 2\pi/a$ is the reciprocal grating vector and a is the grating period. This scattering results in a crossing of SP dispersion branches in the reduced Brillouin zone $-\pi/a < k_x \leq \pi/a$. k_x is the wave-vector component parallel to the surface and perpendicular to the grating rules. The SP dispersion on periodically corrugated metal surfaces and the process of coupling on a grating are sketched in Figs. 1(a) and 1(b). The interference of SP's in the crossing region has been investigated experimentally in different arrangements, e.g., reflection, transmission, and emission. It has been studied for different excitation processes,[1] e.g., excitation by light [Fig. 1(c)], by tunneling electrons in metal-oxide-metal (MOM) diodes,[2] and with fast (50—80 keV) electrons[3] [see Fig. 1(d)]. In the crossing region a complex behavior is observed which depends strongly on the experimental arrangement, excitation mechanism, and grating profile. In some experiments the "dispersion"—we will specify this expression below—shows gaps in energy, whereas in other ones, in momentum. This is analogous to the propagation of guided light waves in media with a periodically modulated effective refractive index or gain, where the interaction between modes can be characterized by a complex coupling constant. In this case both energy or momentum gaps may occur at the intersection of the dispersion curves of the two Bragg-scattered plasma waves, depending on the ratio of the real and imaginary part of the coupling constant.[4] In recent theoretical calculations, performed both with Rayleigh and extinction-theorem methods, it was shown that the interaction of elastically scattered SP waves produced only energy gaps.[5] Consequently, based on these calculations, serious doubts were expressed on the reliability of experiments showing momentum gaps and it was stated that experiments using momentum scans at fixed photon energy lead to "unphysical results."

In a more recent theoretical work[6] based on the extinction-theorem method, the reflected intensity was calculated for a silver grating with a sawtooth profile of a period $a = 1.1$ μm and amplitude $h = 30$ nm. A well-pronounced energy gap was calculated at the intersection of the $n = +3$ and -1 dispersion branches, and the distribution of the reflection coefficient (which is inversely proportional to the SP density of states) in the ω-θ plane shows a saddle-point structure around the intersection of the two branches [Fig. 2(a)]. The angle θ is connected via $k_x = (\omega/c)\sin\theta$ with the momentum k_x of the SP. It was declared that only θ=const scans can truly reproduce the gap structure, whereas ω (or λ)=const scans give an "unphysical," gapless, continuous dispersion relation as a consequence of the saddle structure shown in Fig. 2(b).

In the following sections we prove, using our earlier and some recent experimental results, that the above-mentioned and similar theoretical results are not of general validity. Rather, we show that both situations, namely a gap in momentum transfer as well as in energy may be observed in particular arrangements. We describe the influence of the type of SP excitation process and the grating profile on the formation of the gap.

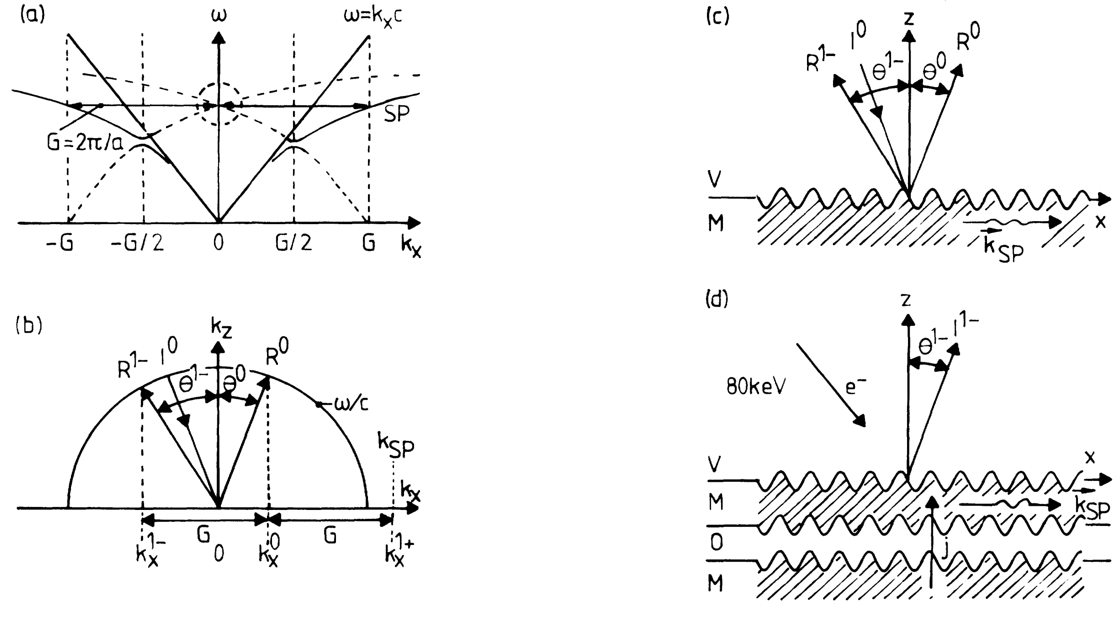

FIG. 1. (a) Surface plasmon dispersion on a periodically corrugated metal surface. Surface plasmons (SP) propagating both in x and $-x$ directions on the surface with wave vector k_x and $-k_x$, respectively, are Bragg scattered on the corrugations of periodicity a. The momentum transfer will be $\kappa=\pm nG$, where $G=2\pi/a$ is the reciprocal-lattice vector. Bragg scattered SP waves interact strongly with each other at the Brillouin-zone boundaries at $k_x=\pm G/2$. In this paper we are interested in the crossing regime at the center of the Brillouin zone, indicated by the dotted circle. Here beside direct plasmon interaction we have interaction with the free-photon field, too. (b) Vector diagram and (c) experimental configuration for a plane light-wave incident on a periodically modulated surface with angle of incidence θ^0. The wave vector of the plane wave $\mathbf{k}$ has a magnitude $k=\omega/c$. The plane wave is partly reflected (R^0) and partly diffracted on the grating (R^{1-})with an angle θ^{1-}. In this latter process the diffracted wave has a wave-vector component $k_x^{1-}=k\sin\theta^{1-}=k\sin\theta^0-G$ parallel to the surface. If G is large enough, also an evanescent wave is induced with $k_x^{1+}=k\sin\theta^0+G$ decaying exponentially both into vacuum and metal. If, for this situation, the momentum conservation law for the surface plasmons is fulfilled, i.e., $k_x^{1+}=k_{\mathrm{SP}}$, SP's are excited and a resonant minimum is observed in the reflectivity R^0. (d) In other experiments SP's are directly excited with fast electrons transversing the vacuum (V) -metal (M) boundary or by inelastically scattered electrons in a biased MOM structure. Via the periodical corrugation, SP can decay leading to a resonant light emission (I^{1-}) at an angle θ_M^{1-} when the condition $k_{\mathrm{SP}}-G=k\sin\theta_M^{1-}$ is fulfilled.

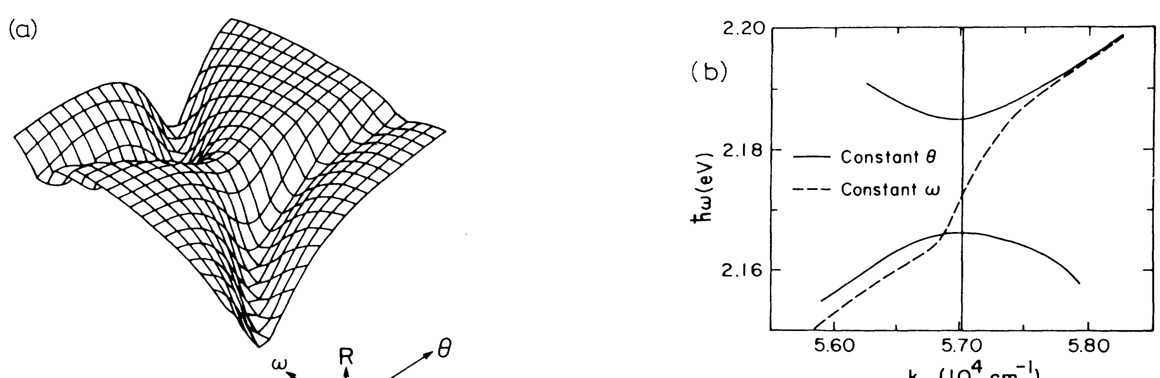

FIG. 2. (a) The reflectivity surface $R(\omega,\theta)$ as calculated in Ref. 6 for silver on a sawtooth grating with a period $a=1.1$ μm and amplitude $h=30$ nm. A gap is found in energy. (b) Dispersion curves deduced from the reflectivity distribution of (a) with $\theta=$const scans (solid line) and $\omega=$const scans (dashed line). No gaps are found in the latter case. (From Ref. 6.)

II. SOME REMARKS ON THE DISPERSION OF SP IN A PERIODICALLY CORRUGATED SYSTEM

Before we discuss our experimental results, the meaning of dispersion is briefly given from the point of view of experiments and applications. The dispersion of an electromagnetic mode is defined in a strict sense only for a system without damping, i.e., with a δ-function-type spectrum in the ω-k plane. For the systems of interest here, the coupling effect of the periodical corrugation between plasmons and the radiative electromagnetic waves results in an inherent damping (radiative damping) and in reactive processes which shift the plasmon dispersion. It is clear, that if one is interested in the SP dispersion itself, one should made the modulation of the grating coupler as small as possible; thus its influence on the dispersion can be neglected. Here, however, we are interested in structures which strongly couple plasmons and photons. Such couplers are of interest for technical applications, e.g., plasmon-resonance-enhanced light-emitting metal-oxide-metal diodes,[2] or for experiments where one wants to use a plasmon-resonance-enhanced excitation process, e.g., for plasmon-resonance-enhanced Raman spectroscopy[7] and other investigations.[1]

For strong couplers one has inherently broad resonances. The experimentally observed quantity, e.g., the reflectivity $R(\omega,k_x)$ or the intensity of the emitted light $I(\omega,k_x)$, forms a three-dimensional pattern above the ω-k_x plane and extrema (minima for R, maxima for I) are interpreted as points of the SP dispersion. Because of the inherent broadening different ω-k_x pairs may be obtained depending on the way the experiments are performed, i.e., ω is scanned at fixed k_x, or k_x (or θ) at fixed ω. This is even more pronounced for the complicated crossing regime. Moreover, in different experimental arrangements (excitation with light or electrons) the interfering plasmons are excited with different relative phases, resulting in a different response and consequently in a different dispersion.

Concerning theories, this means that for this situation of strong couplers it is not enough to calculate solutions of the homogeneous system, but one has to calculate the full response, including the exciting field if it is aimed to describe the experimentally observed extrema in the spectra.

An example of calculated results for an optical experiment is reproduced from Ref. 6 in Fig. 2. The reflectivity $R(\omega,\theta)$ has been calculated for a silver surface of a symmetrical sawtooth profile with a period $a=1.1$ μm and an amplitude $h=30$ nm. If this reflectivity pattern is scanned in energy at fixed angle θ with corresponding wave vectors k_x, two minima are found. The dispersion of these minima are shown in Fig. 2(b) by solid lines. If, however, for certain fixed frequency ω the wave vector k_x is varied, only one relative minimum was found, as shown by the dashed line in Fig. 2(b). [Note that here not the crossing regime in the center of the Brillouin zone has been calculated but at $k_x=G$, where one has the interaction of the $(3+)$ order of the left SP branch with the $(1-)$ order of the right SP branch.] This calculated result is one example for a situation which may be called a "real" energy gap. However, we will show from experimental results that this is not the general situation. Also, real momentum gaps can be observed in different configurations.

Our experiments also indicate that beside other parameters it is important to take into account the "external" damping of the system, i.e., the radiation losses caused by the scattering of plasmons into the $-\omega/c<k_x<\omega/c$ radiative region.

The interaction of electromagnetic modes via a grating coupler is a general problem which is also important for other systems, e.g., guided light waves. In this case, interaction between Bragg scattered plane waves of frequency ω was characterized with a complex coupling constant. When its real part was dominant, a splitting in ω (frequency gap) was found while with dominant imaginary part a small momentum gap (angular splitting) was calculated.[4] This is in formal analogy with the SP momentum gaps observed by us at the crossing point [Fig. 1(a)], i.e., inside the radiative region, where the imaginary part of SP wave vectors (which is proportional to the sum of external and internal dampings) increases strongly. This strong coupling of the surface plasmon field to the photon field is often omitted in the theories.

III. EXPERIMENTS ON THE PLASMON DISPERSION IN MOM STRUCTURES

Experiments were performed on metal-oxide-metal thin-film sandwich structures with gold or silver as a top electrode. When a dc bias of a few volts is applied to these structures, electrons tunnel through the barrier (oxide). The hot electrons are scattered inelastically inside the layers and they may excite surface plasma waves. Radiative decay of plasmons results in light emission. If the MOM diode is deposited onto a periodically modulated (i.e., holographic grating) substrate, the experimental determination of the SP dispersion relation is possible by measuring the angular distribution of emitted light. Our experiments revealed that, independently of the direction of the current, the top metal-vacuum interface surface plasmons emit light anisotropically from corrugated substrates of period $a=0.3$–1 μm. Ion-beam-etched sapphire holographic-grating substrates were used in the experiments. The grating profile was typically of slightly rounded-off rectangular shape having, consequently, a lot of higher-order Fourier components.

In the case of shallow gratings ($h<10$ nm), no gaps were observed at the intersection of the $n=-1$ and $+1$ branches at $\theta=0$. Moreover, the light intensity in this direction was exactly twice as large as that at neighboring angles, because of the constructive addition of light coming from different dispersion branches.[8] We have to note, however, that the observation of a possibly 10–100-meV-wide energy gap cannot be expected in this experiment as a consequence of the very low light intensity (typically 10^{-10} W) and the necessarily poor spectral resolution of the spectrometer.

On identically prepared deep ($h>10$ nm) gratings definite momentum gaps were observed. The gap width (Δk) increased in proportion to the amplitude.[8] The SP

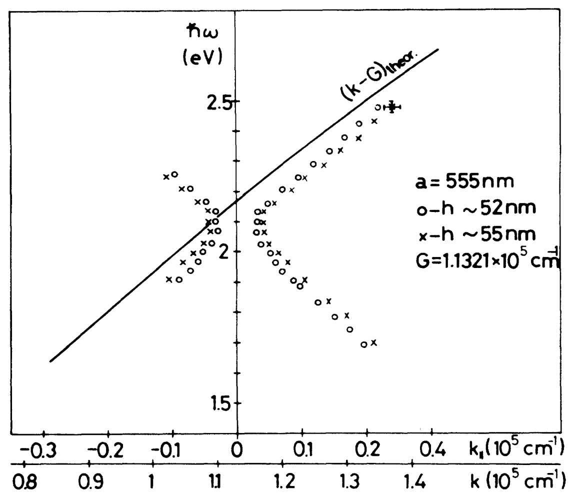

FIG. 3. Dispersion relation of SP for silver topped MOM diodes with period $a=555$ nm and with different amplitudes h. Tunnel emission intensity was measured in ω=const scans at $U=+2.5$-V bias and at room temperature (Ref. 8).

dispersion relation as measured in Al-AlO$_x$-Ag ($d=33$ nm) structures is shown in Fig. 3. Diodes were deposited on a grating substrate with a period of $a=550$ nm and with groove amplitudes $h=52$ and 55 nm. Constant-λ scans were used to determine the dispersion curves and the experimental points mark the positions of peaks in the intensity-angle scans. In order to check the validity of the conclusions of Ref. 6 concerning the experimental methods for the determination of dispersion relations, we have repeated the above-mentioned measurements with λ=const and θ=const scans successively. The results are shown in Figs. 4 and 5, respectively.[9] In Fig. 4 one can clearly see that emission maxima are grouped into two

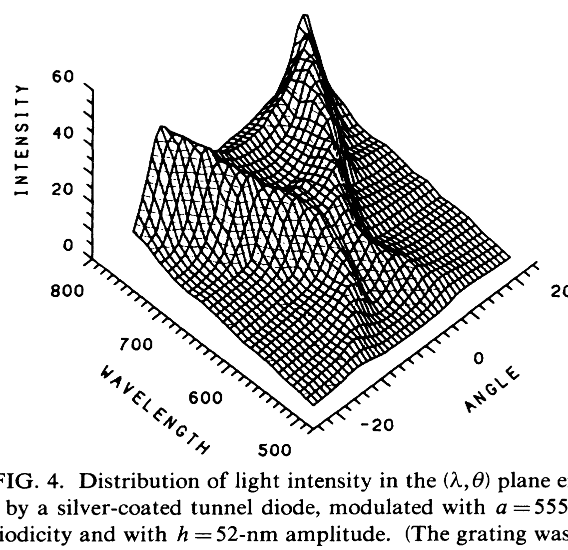

FIG. 4. Distribution of light intensity in the (λ,θ) plane emitted by a silver-coated tunnel diode, modulated with $a=555$-nm periodicity and with $h=52$-nm amplitude. (The grating was the same as in Fig. 3.) Measurements were made with λ=const scans at $U=+2.5$ V and at $T=-106\,^{\circ}\mathrm{C}$. A well-defined momentum gap is observed. (Wavelength in nanometers, angle in degrees, intensity in arbitrary units.)

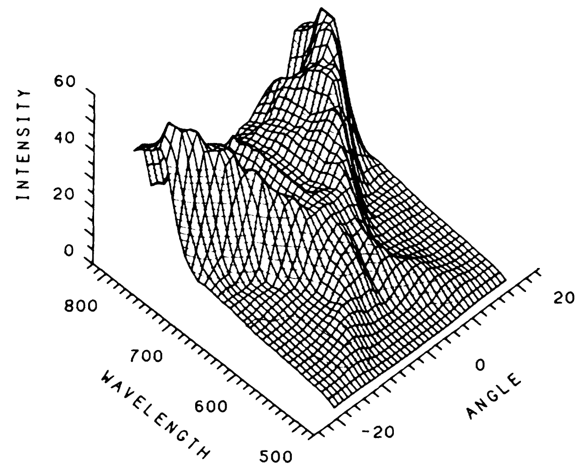

FIG. 5. Intensity distribution of light from the same sample as in Fig. 4, but the measurements were made by θ=const scans. (For units, see Fig. 4.)

folds which get close to each other around the Bragg point ($\theta=0$, $\lambda\sim630$ nm) but still remain separated by a valley. This figure can be regarded as the visualization of the dispersion relations in Fig. 3.

Figure 5, which was constructed from θ=const scans, giving practically the same picture but with the difference that at the long-wavelength side of the gap region there is a small plateau giving rise to a dispersion relation similar to that plotted with dotted lines in Fig. 2(b). The physical picture, consequently, will be similar to that given in Ref. 6, but in the opposite sense, i.e., with a definite momentum gap in λ=const scans and with a gapless dispersion in θ=const scans. This means, in other words, that for the particular grating here the SP dispersion shows a true momentum gap and θ=const scans result in an unphysical picture.

Another experiment was made to check the role of the grating profile. We have prepared, similarly as in the former case, an Al-AlO$_x$-Ag diode on a holographic grating substrate, produced using photoresist over a flat sapphire plate. The grating period was $a=549$ nm, the amplitude $h\sim31$ nm and the scanning-electron-microscope picture showed a quasisinusoidal profile.[10] The Fourier analysis of the (model) profile gave essential components only up to $n=5$, being much less than in the case of the etched grating ($n>60$). The experimental setup and techniques were the same as before. The result of λ=const scans is plotted in Fig. 6. No gaps could be found in the intensity distribution, but at the Bragg point ($\theta=0$, $\lambda\sim620$ nm) a definite peak appeared. This is due to the constructive addition of the intensity of $n=-1$ and $+1$ dispersion branches. The dispersion relation in Fig. 7 has been determined from the position of the intensity maxima. It is continuous, in good agreement with the dotted curve in Fig. 2(b). These results lead to the conclusion that for a particular experiment the shape and amplitude of grating profiles are important parameters which determine whether energy or momentum gaps appear around the intersection of SP dispersion branches. These experi-

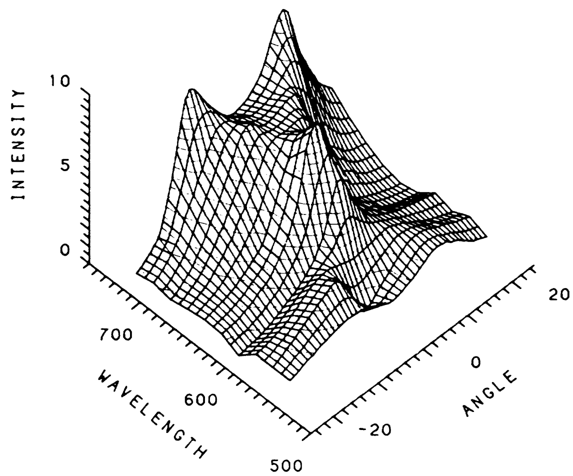

FIG. 6. Distribution of light intensity in the (λ, θ) plane emitted by a silver-coated MOM diode, sinusoidally modulated with $a = 549$-nm periodicity and with $h = 31$-nm amplitude. Measurements were made with λ=const scans at $U = +3.2$ V and $T = -112\,^{\circ}\mathrm{C}$. No gaps can be seen. (For units, see Fig. 4.)

ments have proved that both energy and momentum gaps are realistic and not only artifacts, caused by an improper performance of the experiments.

MOM diodes may possibly be applied as light-emitting devices, therefore the angular and spectral distribution of emitted light is of practical importance. The experimental observation of a gap in k_x demonstrates that these devices will not emit with maximal intensity in the forward direction ($\theta = 0$). However, it has been demonstrated that it is possible to realize devices that show no frequency gaps in the emission spectra. This is of direct technical importance and by no means an unphysical result.

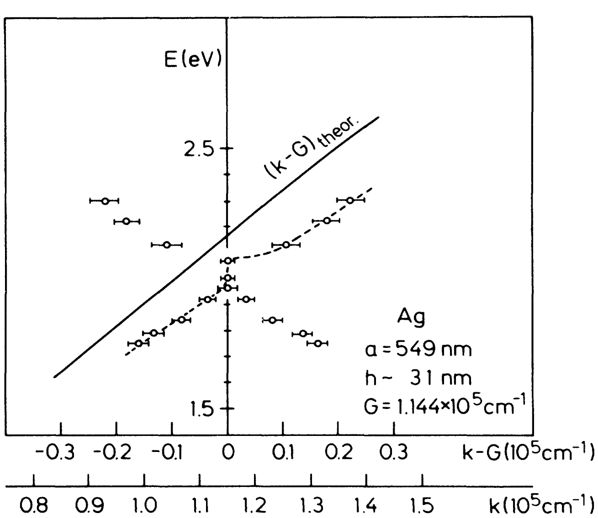

FIG. 7. SP dispersion relation determined from scans shown in Fig. 6.

IV. EXPERIMENTAL PLASMON DISPERSION FROM EXCITATION WITH FAST ELECTRONS

Similar observations, namely gaps both in frequency as well as in momentum space, have also been observed for the following experiments. A well-defined beam of electrons of 80 keV kinetic energy transverses the boundary between vacuum and an optically nontransparent, thick silver film with a sinusoidal surface profile. The angle of incidence is $\alpha = 45^{\circ}$. These electrons excite a spectrum of electromagnetic waves which is in a very wide range continuous both in ω and $\mathbf{k}_{\parallel} = (k_x, k_y)$. Surface plasmons of different propagation direction are excited with a very high efficiency. The radiative decay of these SP's via the periodically modulated surface results in resonant structures in the emitted radiation. They can be analyzed as a function of the wavelength λ and the angles of emission θ and ϕ (ϕ is the azimuthal angle). Experiments for electron-induced surface plasmon excitation and the radiative decay via periodical surface corrugations, which was first observed in Ref. 11, are described in detail in Ref. 3.

In further experiments[12] the behavior in the crossing region of surface plasmon branches has been studied. A complicated intensity pattern $I(\omega, \theta)$ with a saddle point for the emitted intensity was observed having a strong asymmetry in the excitation strength for emission below and above the gap. In particular, the emission peaks have

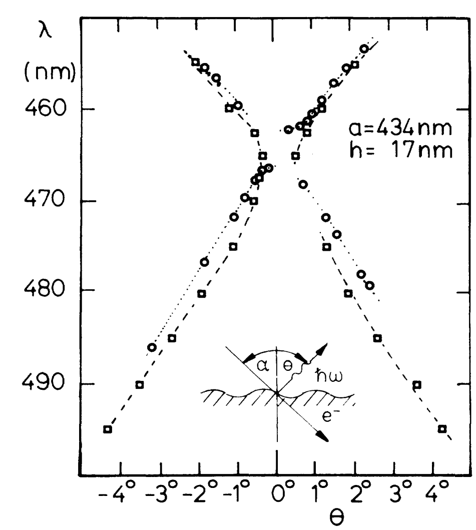

FIG. 8. Dispersion of the emission maxima for the radiative decay of surface plasmons, which were excited by 80 keV electrons on a sinusoidally modulated Ag surface with periodicity $a = 433.6$ nm and modulation amplitude $h = 17$ nm. Electrons are incident at an angle $\alpha = 45^{\circ}$ with respect to the surface normal. The projection v_x of the electron velocity is perpendicular to the grating grooves. The radiative decay of the plasmons is observed in the plane of electron incidence. θ is measured with respect to the surface normal with positive θ in the direction of v_x. Maxima measured at fixed λ in a θ scan are marked by squares, maxima measured at fixed θ in λ scans are depicted by circles. Lines are guides for the eye. The spectral and angular resolution of the experimental setup is $\Delta\lambda = 2.6$ nm and $\Delta\theta = 0.16^{\circ}$, respectively.

a higher intensity for energies above the gaps as compared with those below. The dispersion of the intensity maxima in the emitted radiation measured at fixed wavelengths as a function of the angle of emission is plotted in Fig. 8 with squares. The position of maxima measured at fixed angles for a variation in λ are given by circles. θ scans at fixed λ show again a momentum gap. Scans of λ at fixed θ point in the direction of the formation of a gap on the energy scale. However, exactly for $\theta=0$ only one low intensity peak was measured. Thus the situation is similar as in Fig. 7. We also like to note that for the dispersion branch at positive θ below the gap in Fig. 8, the emission peaks are only weakly pronounced and thus slightly different positions are measured in θ or in λ scans. The reason for the weak intensity is firstly the strong asymmetry of the emission below and above the gap, which is discussed above, and secondly, that due to the non-normal incidence of the electrons plasmons of positive and negative k_x are excited with different probabilities, resulting in different emission intensities from both branches.

V. EXPERIMENTS USING OPTICAL EXCITATION OF PLASMONS

On samples, which were prepared in a similar way and with similar parameters as those for the experiments described in Sec. IV, reflection measurements were performed.[13] Figure 9 shows experimental spectra for the reflection of monochromatic light on a sinusoidally modulated Ag surface with periodicity $a=426.5$ nm and modulation amplitude $h=14.6$ nm. In these optical experiments we find that the dispersion of the reflection minima in the crossing region shows for wavelength scans at fixed θ energy gaps. This is similar to the observation in optical measurements in Ref. 14. For θ scans at fixed energies in the energy-gap region we find weakly pronounced structures. A strict evaluation of the reflection minima dispersion gives similar results as shown by the dotted line in Fig. 2(b). The behavior found for the optical experiments is thus in contrast to the dispersion that has been observed for excitation with fast electrons on similar gratings in Sec. IV.

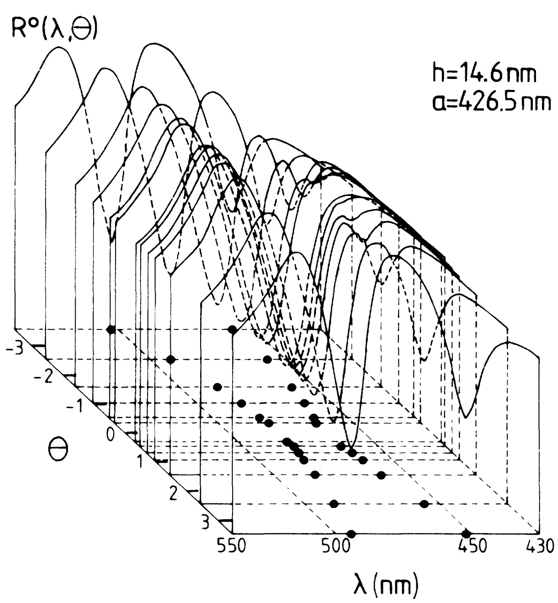

FIG. 9. Reflection of monochromatic light from a sinusoidally modulated Ag surface with periodicity $a=426.5$ nm and modulation amplitude $h=14.6$ nm. The spectra are measured at a fixed angle of incidence θ in a sweep of the wavelength λ. The spectra have not been corrected for the smooth-spectral sensitivity of the experimental setup. The position of the reflection minima is projected onto the (θ,λ) plane.

We attribute the strong asymmetry in the plasmon response, which was observed for both electron excited SP (Sec. IV) and optically excited plasmon resonances (Fig. 9), to the fact that plasmons in a superlattice have the character of standing waves, where both branches have a different symmetry and thus couple differently to exciting and radiative fields. The different formation of the gap structure that is observed for electron excitation (Sec. IV) on the one hand and for optical excitation on the other, may also be attributed to the interference of the plasmons. With electrons plasmons both with $-k_x$ and $+k_x$ momentum are excited simultaneously with a well-defined phase relation. This phase relation is different for optical and for electron excitation leading to different interference and thus emission. The difference in the phase relation of the exciting fields is also reflected in the observation that for electron excitation the plasmon response is strong for energies above the gap (see Sec. IV), whereas for optical excitation the response is stronger below it (Fig. 9).

Using optical excitation in a slightly different arrangement, Pockrand found gaps in the k_x-k_y plane when he studied the coupling of plasmons at different interfaces of a thin corrugated film.[15] Optical excitation of SP has also been used to study the internal photoeffect in MOM structures[16] where the exciting source was an Ar-ion laser. The same type of grating substrates and MOM diodes were used as in Sec. III. The dispersion curves determined from these studies agree within the experimental error with those obtained from the light emission measurements of Sec. III, including the existence and the width of the momentum gap.

We would like to note that a splitting of the plasmon dispersion in energy has also been found for two-dimensional (2D) plasmons in metal-oxide-semiconductor structures (MOS), where through a modulated oxide thickness a "2D" electron gas with a lateral periodic modulation of the surface charge-density distribution $N_s=N_s(x)$ is induced.[17] Here the experimental situation is such that the frequency of the plasmon excitation ω is small (in the far-infrared region), therefore the plasmon wave vector k_x is much larger than ω/c. It was shown in Ref. 18 that in this case the electromagnetic coupling, having been discussed so far in this paper, is small. The experimentally observed splitting arises directly from the superlattice effect of the periodical charge-density modulation.

VI. CONCLUSIONS

Having surveyed the existing experimental data, we have shown that the anomalies of the dispersion curves of

SP on periodically modulated metal surfaces may be different depending on the experimental conditions. In some of the experiments well-defined momentum gaps occur, while others exhibit energy gaps. It may happen that different formations of the gap are obtained for the same grating if different types of excitations are used.

The character of the anomaly in the gap region depends on the period of the grating, on the depth of the grooves, and on their profile (i.e., on the number of its dominant Fourier coefficients), as well as on the type of excitation. In the special case of the three thin-film structure of the MOM diodes, however, we have shown that the momentum gap is really existing, independently of the experimental method used, as seen from the comparison of light emission and internal photoeffect data.

The above observations lead to the conclusion that existing theoretical calculations are so far valid only for particular, simple experiments. In order to be able to explain all existing experiments, the described theoretical models have to be further refined.

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