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Dispersion of Surface Plasmons in InSp

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Infrared-reflectivity measurements on InSb surfaces having inscribed line gratings yield the first complete dispersion relation for surface plasmons.

Surface plasmons (SP) are collective oscillations at the boundary between a free-carrier plasma and a dielectric medium.[1] The SP dispersion for a plasma-vacuum interface is given by[2,3]

$$k = \frac{\omega}{c}\left(\frac{\epsilon_1(\omega)}{\epsilon_1(\omega)+1}\right)^{1/2}, \quad (1)$$

where k is the SP wave vector and $\epsilon_1(\omega)$ is the real part of the dielectric function of the conducting medium. The asymptotic frequency ω_s for large k,

$$\omega_s = \omega_p(1+\epsilon_L^{-1})^{-1/2}, \quad (2)$$

depends on the optical dielectric constant ϵ_L of the lattice and the plasma frequency ω_p. Direct optical excitation of SP is impossible[4] because for each frequency ω, the SP wave vector is greater than the photon wave vector ω/c. Larger wave vectors for the exciting light can be achieved at the interface by a surface having a periodic grating structure with spacing d. Thus SP can be excited by light at[5]

$$k = (\omega/c)\sin\theta + 2n\pi/d, \quad (3)$$

where θ is the angle of incidence, and n is an integer. Excitation occurs only for the polarization of light with $\vec{E}$ perpendicular to the grooves.[6] Such experiments have been performed on metal gratings[7,8,5] where the spacings d are large compared to the plasma wavelengths λ_p in the uv region. Thus, only SP with relatively small k could be excited, and only a restricted portion of the dispersion curve was accessible.

In this paper we present the first complete SP dispersion curve where experimental points reach up to the asymptote at $\omega = \omega_s$. This result is achieved by using the free-carrier plasma of a semiconductor. The material, InSb, and its doping-dependent plasma frequency have been chosen so that complications by plasmon-phonon interaction can be avoided. We attain the large relative k since λ_p is now in the infrared region and is comparable to d. This research is a continuation of preliminary work on GaAs.[9]

We used n-type samples [$(1-7)\times 10^{18}$ electrons/cm^3] with areas of about 1 cm^2. Line gratings were scribed into optically polished surfaces using a diamond ruling machine. The grating spacings ($d = 10$, 20, and 30 μm) were controlled

by a scanning microscope to an accuracy better than 1%. Replica electron microscopy showed that the groove depth and the extent of surface destruction were small compared to d. Reflectivity spectra were taken in the zeroth order of diffraction for reasons of intensity. The angle of incidence, θ, could be varied. Beam divergence was reduced to 5 deg. Results with the polarization $\vec{E}$ parallel to the grooves are omitted here; however, they confirm for all doping levels the presence of the reflectivity maximum obtained on GaAs.[9]

The solid line of Fig. 1 shows the spectrum of a 30-μm grating surface. The dips are caused by SP excitation at various orders n. A series of those spectra was obtained by varying d, θ, and doping level. The construction of a dispersion curve $\omega(k)$ involves the problem of determining the eigenfrequencies ω_r of the excited SP from these dips. Complicated calculations for some specific metal grating profiles[10] based on exact grating theory yielded ω_r values at slightly higher frequencies than the reflectivity minima. We got the same result for our semiconductor surfaces by a much simpler approach, describing SP excitation by Lorentz-oscillator contributions at ω_r to the complex $\epsilon(\omega)$. Oscillator strength, damping, and ω_r were used as fitting parameters. The plasma frequency ω_p was determined from the position of the plasma edge. The values of ω_r resulting from our calculations are indicated by arrows for the spectrum in Fig. 1. We found SP damping to be only slightly higher than single-particle damping of the free carriers (obtained from the smooth surface[11]).

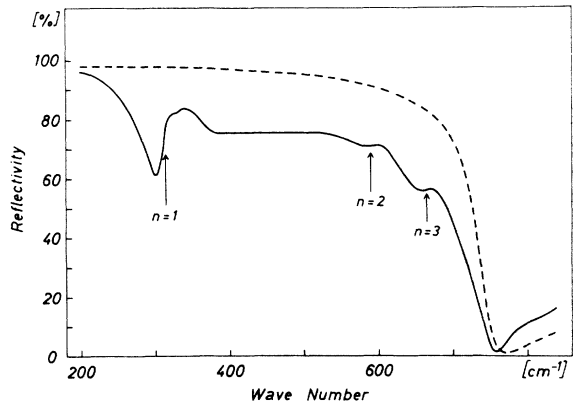

FIG. 1. Solid line, infrared reflectivity, at room temperature and normal incidence, of InSb sample with 7×10^{18} electrons/cm^3 having a 30-μm grating inscribed. Arrows indicate the fitted SP resonance frequencies at orders n. Dashed line, results from smooth surface for comparison.

The experimental dispersion curve is shown in Fig. 2. In comparison, the solid line shows the theoretical dispersion of Eq. (1) with the $\epsilon_1(\omega)$ data from reflectivity of the smooth surface. Frequencies and wave vectors are normalized to ω_p and $k_p \equiv \omega_p/c$ for each doping. Our results enable us for the first time to compare experimental values with the prediction of Eq. (1) for a semiconductor. At low k there is good agreement; with increasing k, however, experimental frequencies lie below the theoretical values by a few percent. This deviation cannot be caused by plasmon-phonon interaction because the shift of ω_p relative to the plasma edge is too small for our samples. We tend also to exclude influence of the grating itself[12] towards reducing the eigenfrequencies at higher k.[13] We rather suppose that the assumption of an abrupt interface underlying Eq. (1) is not fully justified. Even for highly doped semiconductors, surface layers with optical properties different from the bulk must be taken into account.[14] We suppose them to be responsible for the observed deviations; this assumption is supported by investigations on metal surfaces where dielectric layers give an important influence on SP dispersion.[4,15]

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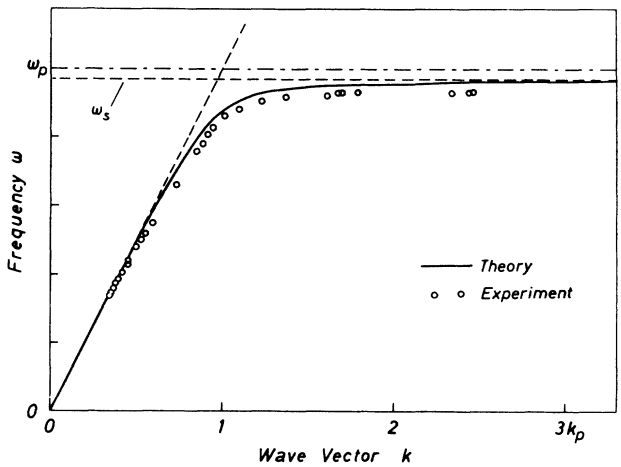

FIG. 2. Dispersion of surface plasmons in InSb. Experimental results for different doping and grating spacings, normalized to ω_p and k_p. Theoretical curve calculated from Eq. (1).

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Location of the Zn 3*d* States in ZnO†

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We have carried out uv photoemission measurements on hexagonal ZnO cleaved in vacuum. The results of this study have placed the Zn 3*d* core level at 7.5 ± 0.2 eV below the valence-band maximum, some 3 eV lower than predicted by Rössler's Green's-function (Korringa-Kohn-Rostoker) band calculation. This assignment is in excellent agreement with recent x-ray photoemission measurements and is shown to be consistent with the low value of the effective number of free electrons calculated from the reflectivity.

It has proven difficult to use band theoretical calculations to locate absolutely in energy tightly bound states in solids such as filled *d* and *f* states. For example, considerable problems in this respect were found with the *d* bands in Cu as calculated[1] with augmented plane waves and with the 4*f* levels in the rare earths.[2] Recently, Rössler[3] has calculated the location of the Zn 3*d* states in ZnO using the Korringa-Kohn-Rostoker (KKR) method.[4] In view of the difficulty of such calculations, it is important to obtain an early comparison of these results with experiment. Unfortunately, the several experiments that have been done on ZnO do not agree on the location of the Zn 3*d* states.[5-7] It was the purpose of the work reported here to provide additional experimental evidence so that this question could be resolved. Once determined, the absolute energy of the *d* bands will have to be taken into account by any subsequent band calculations.

The first published band structure for ZnO, calculated by Rössler using the KKR method, exhibits broad (1.6-eV) *d* bands lying closely below the upper valence bands.[3] For comparison, the *d* bands of the sulfides, sellenides, and tellurides of Zn and Cd are relatively narrow and lie some 6 and 10 eV below the upper valence bands.[3]

The Green's-function (KKR) approach appears to have been reasonably successful in locating *d* bands in such cubic II-VI semiconductors as ZnS, ZnSe, and ZnTe.[8] This method involves the use of an *ad hoc* potential with only a single adjustable parameter which is adjusted to obtain the correct band gap. Such a calculation, then, does not require the input of a great deal of experimental data. Indeed, it is significant that for ZnO, the first calculated band structure actually precedes much of the relevant optical data. In this case, experimental results will be particularly valuable in confirming or correcting the