Experimental Evidence for Surface Quenching of the Surface Plasmon on InSb(110)

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(Received 6 December 1983)

In high-resolution electron-energy-loss spectroscopy on clean cleaved InSb(100) surfaces, coupled surface plasmon–phonon losses are studied as a function of primary energy E_0. The E_0 dependence of the relative intensities of the phononlike and plasmonlike losses shows the existence of a dead layer for surface plasmons of free electrons. Comparison of the space-charge electron density with the bulk concentration indicates that the dead layer arises from a dynamical quenching of the plasmon amplitude rather than from a depletion of carriers due to band bending.

PACS numbers: 71.45.Gm, 79.20.Kz

In this paper we present experimental evidence for the existence of a dynamical quenching effect for surface plasma waves near semiconductor surfaces. Such surface plasma excitations of free carriers in the conduction band and their coupling to Fuchs-Kliewer surface phonons have recently been detected in high-resolution electron-energy-loss spectroscopy (HRELS) on GaAs(110).[1] In the conventional description[2] both collective excitations exhibit the same spatial structure, an exponentially decaying vibrational amplitude (of lattice polarization and of electron density, respectively) with a decay length equal to the inverse wave vector $q_{\parallel}$ parallel to the surface. From a more detailed consideration of the bilocal structure of plasmons, i.e., the correlation between electron-hole pairs that constitute the plasmon, Egri *et al.*[3] were led to the conclusion that a "plasmon free' surface layer might exist on the surface of a semiconductor. Meanwhile Stahl[4] has solved a boundary-value problem for surface plasmons at semiconductor surfaces and has calculated the spatial dependence of the plasmon amplitude normal to the surface. This theory confirms the surface quenching of plasmons, but experimental evidence for the effect is missing so far.

The present experimental evidence for such a "dead layer" for surface plasmons is derived from a variation of the probing depth in HRELS by a change of the wave-vector transfer $q_{\parallel}$. A variation of the experimentally controlled $q_{\parallel}$ transfer changes the decay length of both the surface phononlike and plasmonlike excitations. For large $q_{\parallel}$ transfers, i.e., a small penetration depth of the fields, the presence of a plasmon dead layer should significantly decrease the plasmon loss intensity as compared with the phonon loss. This effect is indeed found on InSb(110) surfaces.

For the present study n-type material of InSb with different bulk doping is used. Clean (110) surfaces with an area of about 5×8 mm^2 were prepared by cleavage in ultrahigh vacuum (UHV, $p < 10^{-8}$ Pa). The loss spectra were recorded by an electron spectrometer with two electrostatic 127° deflectors as monochromator and analyzer. The energy resolution was 8-meV full width at half maximum (FWHM) current in the direct beam without sample. After reflection on the clean cleaved surface the resolution usually deteriorates up to a FWHM of about 12 meV.

The bulk free-electron concentration n_b of the samples used was determined from the plasma edge measured by infrared (IR) reflection. Measurements on different spots of a sample surface showed that the doping was quite homogeneous over the surface. Maximum deviations of the energetic position of the plasma edge were found to be within about 1%. The carrier concentration was evaluated from a fit of Drude dielectric functions to the experimental reflectivity data; the effective mass used in these calculations was $m^* = 0.013 m_0$.[5]

Figure 1 shows loss spectra measured on a clean cleaved InSb(110) surface of n-type material with a bulk electron concentration of $n_b = 4.9\times 10^{17}$ cm^{-3}. The loss spectra at 20 eV primary energy are characterized by two losses and corresponding gains at ± 24 and ± 62.5 meV. With decreasing primary energy E_0 the loss near 62 meV shifts slightly to higher energy (dispersion). The most dramatic effect, however, is the strong dependence of the two loss intensities on primary energy. At $E_0 = 20$ eV the high-energy loss is the prominent one, whereas at 4 eV primary energy it is strongly suppressed and dominated by the loss near 24 meV. The energetic position of the high-energy loss is strongly dependent on the carrier concentration. On material with

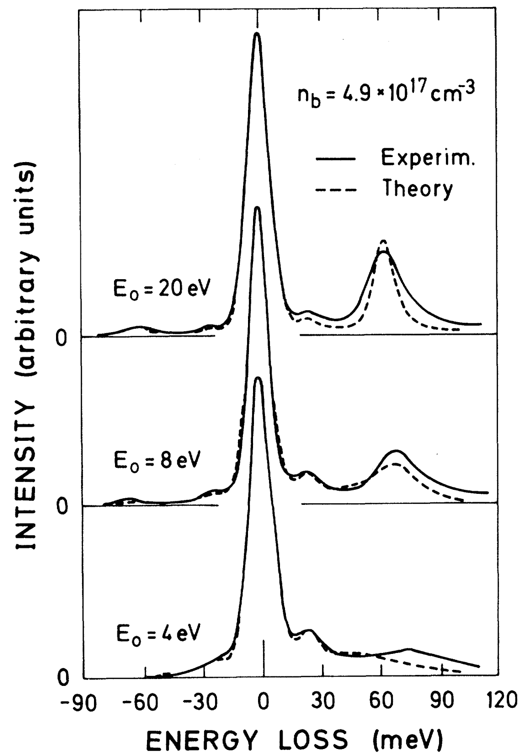

FIG. 1. Loss spectra measured on a clean cleaved InSb(110) surface with several primary energies E_0 (solid curves). The bulk electron density n_b has been determined by IR-reflection spectroscopy. For the theoretical spectra (broken curves) a "plasmon free" surface layer with a thickness of 40 Å has been assumed. The fit is calculated with a carrier concentration of $n = 5.0 \times 10^{17}$ cm^{-3}.

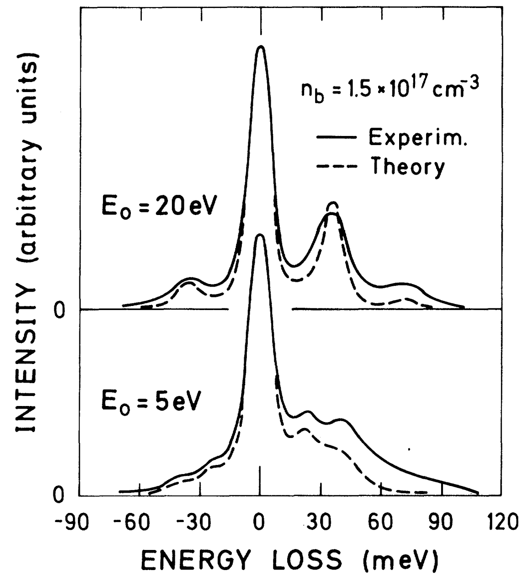

FIG. 2. Loss spectra measured on a clean cleaved InSb(110) surface with two different primary energies E_0 (solid curves). The theoretical spectra (broken curves) are calculated with a "plasmon free' surface layer of 50 Å and an electron density of $n = 1.5 \times 10^{17}$ cm^3 (equal to the bulk density n_b).

$n_b = 1.5 \times 10^{17}$ cm^{-3} (Fig. 2) it appears at 38.5 meV. As for the highly n-doped material (Fig. 1) the intensity of this loss is significantly suppressed at lower primary energies.

From a comparison of Figs. 1 and 2 it is evident that the high-energy loss is related to the free electrons in the conduction band of InSb. As in previous work on GaAs(110),[1,2] a quantitative description of the loss peak positions is possible in terms of coupled surface phonon–plasmon excitations. The loss peak positions can be calculated from the maxima of the surface loss function $\mathrm{Im}[-1/(\epsilon+1)]$, in which the dielectric function

$$\epsilon(\omega) = \epsilon(\infty) + \chi(\mathrm{TO\ phonon}) + \chi(\mathrm{plasma}) \quad (1)$$

is assumed to be composed of susceptibility contributions due to the lattice IR oscillator (long-wavelength TO phonon) and due to the plasma of the free electrons. In the simplest approximation χ(plasma) is a Drude susceptibility. Concerning loss peak positions such a Drude term gives nearly the same results as a Lindhard dielectric function (see below). With the electron concentration n as an adjustable parameter in Eq. (1), the best fits to the experimentally determined loss peak positions are given in Figs. 1 and 2 for concentrations of 5.0×10^{17} cm^{-3} and 1.5×10^{17} cm^{-3}, respectively. These values do not necessarily coincide with the bulk concentrations n_b, since for the surface plasmon the carrier density in the space-charge layer is relevant. The low-energy loss near 24 meV is more surface phononlike (close to the LO bulk phonon) whereas the high-energy loss has more surface-plasmon character, which arises from the χ(plasma) term in Eq. (1).

In the classical standard description both the surface phonon and plasmon exhibit a vibrational amplitude that decays exponentially from the surface into the bulk with a decay length equal to $1/q_{\parallel}$. For both excitations the wave vector, i.e., the $q_{\parallel}$ transfer

$$q_{\parallel} \simeq k_0(\hbar\omega/2E_0)\sin\theta_i, \quad (2)$$

is determined by energy and wave-vector conservation (parallel to the surface).[2] k_0 and E_0 are primary wave vector and energy, respectively, $\hbar\omega$ is the loss energy, and θ_i the angle of incidence. A smaller primary energy enhances the wave vector $q_{\parallel}$ of both coupled collective modes and, therefore, decreases the "penetration depth" of the excitations; the probing depth of HRELS is diminished by decreased E_0. A suppression of the plasmon-type loss intensity as compared with that of the phonon of lower E_0 can therefore be explained by the presence of a spatial region just below the surface where the surface-plasmon amplitude is quenched in contrast to the phonon amplitude. This effect can quantitatively explain the experimental data in Figs. 1 and 2.

We have used a simple layer model to simulate such a situation: The crystal is represented by a semi-infinite half space in which lattice vibrations are allowed up to the very surface. A surface quenching effect of the plasmon is formally taken into account by assuming a layer of thickness d and dielectric function ϵ_L, which contains only this lattice contribution: $\epsilon_L(\omega)=\epsilon(\infty)+\chi(\text{TO phonon})$. Below this layer the remaining half space is described by a bulk dielectric function ϵ_b which contains both lattice and plasma contributions according to Eq. (1). As in previous work $\chi(\text{plasma})$ is modeled by a Lindhard dielectric function in the nondegenerate Boltzmann limit.[3] Ohmic damping is thus neglected and boundary conditions at the dead-layer inteface are not considered. The dielectric scattering cross section according to Mills's theory[6] is then calculated by means of a surface loss function in which the dielectric function is an effective $\tilde{\epsilon}$ containing bulk (ϵ_b) and layer (ϵ_L) contributions:

$$\tilde{\epsilon}=\epsilon_L\frac{1+\Delta\exp(-q_{\parallel}d)}{1-\Delta\exp(-q_{\parallel}d)}, \qquad (3)$$

with $\Delta=(\epsilon_b-\epsilon_L)/(\epsilon_b+\epsilon_L)$. Integrating the differential inelastic scattering cross section over the entrance aperture of the analyzer, simulating multiple energy gains and losses by a Poisson distribution,[6] and finally convoluting with the transmission function of the analyzer, we can compare the calculated loss spectra (Figs. 1 and 2, broken curves) directly with the experimental data. Fitting parameters were the thickness d of the dead layer and the carrier concentration n. For the samples of Fig. 1 the best theoretical fit is achieved with a thickness of the dead layer of 40 Å and a concentration n of 5.0×10^{17} cm^{-3}. In his theoretical calculation Stahl obtained a similar estimate for the dead-layer thickness d at a corresponding carrier concentration.[4]

To obtain an impression of the quality of the fit we have varied simultaneously both the thickness of the dead layer d and the concentration n such that the calculated loss peak positions fit the experimental data of Fig. 1. Reasonable fits are possible with d ranging between 50 and 30 Å and n between 5.5×10^{17} cm^{-3} and 4.9×10^{17} cm^{-3}. For lower carrier concentrations and thinner dead layers the intensities of the plasmonlike and phononlike losses are not described correctly. It should be mentioned further that in the calculation the effect of Landau damping also contributes to the suppression of the plasmonlike loss at lower E_0 (higher $q_{\parallel}$).

One might argue that the dead layer obtained could be due to a depletion of free carriers near the surface (depletion space-charge layer) rather than due to a dynamical quenching effect of the plasmon amplitude. To rule out this possibility one has to discuss carefully the comparison of bulk (n_b) and surface carrier concentration (n). The absolute errors in both values might be considerable, e.g., due to a wrong effective mass m^*. Since, however, in both types of measurements (IR and HRELS) the concentrations are determined from a plasma frequency measurement, the ratio n/m^* enters in essentially the same way. Therefore the comparison of n_b with n is accurate to the extent to which the plasma frequencies can be determined experimentally. For n_b (from IR) this accuracy is 1% [$n_b=(4.9\pm0.05)\times10^{17}$ cm^{-3} for the sample in Fig. 1]. The surface concentration n (from HRELS) depends to some extent on the choice of the dead-layer thickness d. Reasonable fits are only possible with n varying between 5.5×10^{17} and 4.9×10^{17} cm^{-3}. We must therefore conclude that only a flat-band situation or a slight accumulation (but unlikely) of carriers in the space-charge layer is compatible with the data.

The dead layer for the surface plasmon found here must therefore be attributed to a dynamical quenching of the plasmon amplitude near the surface rather than to a static depletion of carriers due to band bending. Finally it should be emphasized that on metal surfaces, because of the high electron density, such dynamical dead layers have thicknesses on the order of an angstrom.[4] They can therefore hardly be detected experimentally.

For the IR-reflection measurements and the careful analysis of the IR data we would like to thank J. Geurts of the 1. Physikalishes Institut der Rheinisch-Westfälischen Technischen Hochsuchule Aachen. We are also thankful for some crystal material supplied by that laboratory. This work was financially supported by the Deutsche Forschungsgemeinschaft (Sonderforschungsbereich 202).

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