Hole-plasmon damping on heavily doped p-type GaAs(110)

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The free-carrier-induced plasma excitation in heavily doped p-type GaAs(110) is studied by means of high-resolution electron-energy-loss spectroscopy. This surface hole-plasmon feature is very broad and approximately centered at 60 meV. The large broadening is due to the low hole mobility and its value depends on the energy of the incident-electron beam. Two causes can concur to determine this behavior: the momentum dependence of the Landau damping of the plasmon (due to the nonlocal nature of the free-carrier response) and the dependence of the hole mobility on the distance from the surface in the crystal. Measurements performed after increasing hydrogen exposures, which modify the subsurface charge region, allowed us to conclude that the momentum dependence of the Landau damping is the cause of the observed variation of the plasmon damping with the primary beam energy.

INTRODUCTION

In the last few years much work has been devoted to the study of the vibrational and collective properties of the clean GaAs(110) surface, mostly using the high-resolution electron-energy-loss spectroscopy (HREELS) technique.[1-8] The major part of these studies dealt with n-type-doped GaAs(110). In fact, HREELS analysis of such semiconductor surfaces showed clear structures in the 0–100-meV energy-loss range, which could be unambiguously identified. In particular, these systems are characterized by the presence of the polar optical phonon (Fuchs-Kliewer phonon) and of the dopant-induced free-carrier plasmon. In the low-doped crystals, the free carriers induce an energy broadening in the quasielastic peak and do not produce distinct structures,[9] while in more highly doped samples ($n > 10^{17}$ cm^{-3}) they give rise to a well-defined structure, whose proper energy width is much lower than the instrumental broadening. Analysis of HREELS data on p-type-doped GaAs is not so straightforward. In fact, as was shown in previous works on the (100) (Ref. 7) and (110) (Refs. 10 and 11) surface of p-type GaAs, the spectra relative to the heavily p-type doped semiconductor are characterized by a wide hole-induced plasmon, much broader than the instrumental resolution and of the corresponding electron-induced plasmon on n-type crystals. Hence, only after a careful data reduction and analysis through appropriate dielectric models[12,13] could the nature of this excitation be explained.[10,11] The broad plasmon feature was attributed to the optical branch of the collective excitation of the carrier gas constituted by both light and heavy holes.[10,11] This attribution was justified by the very good agreement between the measured and expected energy position of the plasmon (as deduced from the nominal doping), and by the good fit with a suitable dielectric model.[13] Most of the very large broadening of the plasmon can be ascribed to the low mobility of the holes in the valence band, as deduced by Hall effect measurements quoted by the sample supplier (that corresponds to ~ 50 meV).

In the present work we show that the plasmon damping depends on the energy (E_0) of the exciting electrons impinging on the surface, from ~ 58 meV (close to the Hall-measurements-derived value) at $E_0 = 50$ eV to ~ 79 meV at $E_0 = 2$ eV. Such behavior could be ascribed either to an intrinsic dependence of the plasmon damping on the exchanged momentum ($q_{\parallel}$)—originating from the nonlocal nature of the free-carriers response (Landau damping), as expected for n-type GaAs (Ref. 14)—or to a dependence of the holes' mobility on the position (z) below the surface, given the relation between the primary beam energy (E_0) and the sampling depth. In order to answer this intriguing question, we performed again measurements as a function of E_0 to study the plasmon damping, refining the fit of the experimental data by means of a dielectric model, and modified the space charge region below the surface by exposing the surface itself to atomic hydrogen, while maintaining E_0 constant. The main result of this investigation is that the q dependence of the plasmon Landau damping is the only cause of the dependence of the plasmon damping on E_0, ruling out variations of the hole mobility on going from the surface into the bulk.

EXPERIMENTAL

Experiments were carried out at the surface physics laboratory Spettroscopia Elettronica Superfici e Adsorbati (SESAMO), Dipartimento di Fisica, Università di Modena. The HREELS spectrometer (Leybold-Heraus ELS-22) is contained in an UHV system also equipped with low-energy electron diffraction (LEED), photoemission, and other ancillary facilities for sample preparation. Base pressure was below 7×10^{-11} mbar (6.8×10^{-9} Pa). The GaAs (110) bar was cleaved with the single-wedge technique. The doping level was $p \simeq 2 \times 10^{19}$ cm^{-3}. Crystalline order of the surface was observed by LEED. Cleanness was monitored by HREELS. HREELS measurements were performed in the specular direction, with an angle of incidence of 65° and primary beam energies

E_0 of 2–50 eV. The energy resolution was about 8 meV, as derived by the full width at half maximum of the elastic peak in the fitting model. The hydrogen exposure was made by placing the surface at approximately 3 cm from a hot (~1700 °C) tantalum filament in line of sight with the surface. The dose was recorded as molecular-gas exposure [1 langmuir (L) $\equiv 10^{-6}$ Torr s] since the conversion rate is unknown.

RESULTS AND DISCUSSION

The HREELS measurements performed on *p*-type GaAs(110) ($p \simeq 2\times10^{19}$ cm^{-3}) at room temperature and at several primary beam energies are shown in Fig. 1, along with their respective best fits obtained by using an appropriate dielectric model[13] (as discussed below). The data show a very broad and almost structureless bump, extending between 20 and 100 meV. Only the spectrum taken at $E_0 = 2$ eV presents a weak structure at ~36 meV which could be the reminiscence of the polar-optical Fuchs-Kliewer phonon. At the same loss energy a dip develops as E_0 increases. There is clearly a high degree of arbitrarity in the attribution of such loss features without an appropriate data reduction. The fit to the data was obtained by using a model calculation within the dielectric theory of the energy loss.[15,16] We recall that, in the dielectric theory, the differential cross section of the dipolar scattering is proportional to the loss function through a kinematic Factor depending on the kinematic of the experiment. The loss function can be written

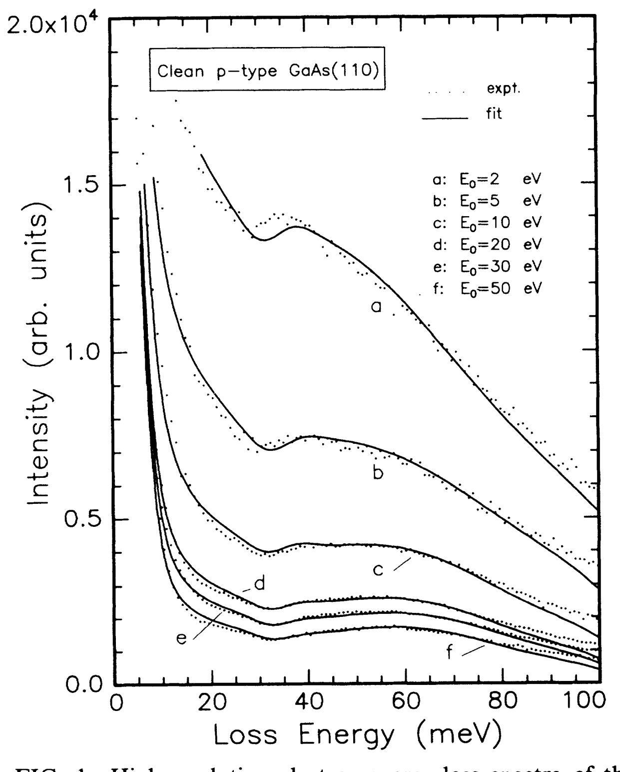

FIG. 1. High-resolution electron-energy-loss spectra of the clean *p*-type GaAs(110) [$p(\mathrm{Zn}) \simeq 2\times10^{19}$ cm^{-3}], taken at various primary beam energy E_0 and with angle of incidence $\theta_i \simeq 65°$. Experimental data (dotted lines) and results of a three-layer dielectric model (solid line) are shown. Note that the data are not shifted along the vertical axis with respect to each other.

in terms of an effective dielectric function (ϵ_{eff}) of the system as[12,15]

$$\mathrm{Im}\frac{-1}{1+\epsilon_{\mathrm{eff}}(\omega,q_{\parallel})} \ . \tag{1}$$

We modeled the physical system as constituted by three layers, characterized by their respective dielectric responses: vacuum, surface, and bulk. In the surface layer we assumed only the Fuchs-Kliewer phonon while in the bulk we considered also the presence of the free-carrier gas. At the surface is the so-called "dead layer," a surface region depleted of free carriers, because of the boundary condition imposed to the carrier wave function to be zero just out of the surface.[17,18] Actually this layer is equivalent to a depletion layer, although it exists for basic reasons and it is not caused by the presence of charge at the surface (either induced by defects or by surface states). We assumed a step profile for the charge density at the interface between the dead layer and the bulk. This is a crude approximation, because the charge density profile varies within about a Thomas-Fermi length in a degenerate gas of free carriers,[14] but it gives a reasonable estimation of the depletion region. We used a Lorentz oscillator for the Fuchs-Kliewer phonon

$$\epsilon_{\mathrm{ph}}(\omega)=\epsilon_{\infty}+\frac{(\epsilon_0-\epsilon_{\infty})\omega_{\mathrm{TO}}^2}{\omega_{\mathrm{TO}}^2-\omega^2-i\omega\gamma_{\mathrm{ph}}} \tag{2}$$

with $\omega_{\mathrm{TO}}=33.41$ meV, $\gamma_{\mathrm{ph}}/\omega_{\mathrm{TO}}=0.009$, and $\epsilon_{\infty}=10.91$,[4] while for the hole plasmon gas we used the Drude model

$$\epsilon_{\mathrm{pl}}(\omega)=-\frac{\omega_{\mathrm{pl}}^2}{\omega(\omega+i\gamma_{\mathrm{pl}})} \tag{3}$$

with ω_{pl} and γ_{pl} the plasmon energy and damping parameters.

It is known that *p*-type-doped GaAs presents dopant-induced holes with two different masses, given the presence of two degenerate subbands with different curvature at the Γ point of the Brillouin zone: heavy ($m_h = 0.45m_0$) and light ($m_l = 0.082m_0$),[19] where m_0 is the free-electron mass. If n_h and n_l are the densities of the heavy and light holes, respectively, we get $n_h/n_l = (m_h/m_l)^{3/2}$, and if N is the dopant concentration, $n_h = 0.93N$ and $n_l = 0.07N$. Although $n_l \ll n_h$ the unscreened plasma frequencies of the two gases within the Drude model ($\omega_b^2 = 4\pi n/m^*$) are comparable; at the present doping level ($N = 2\times10^{19}$ cm^{-3}) we have $\omega_{bh} = 238.9$ meV and $\omega_{bl} = 155.3$ meV. Hence, n_l cannot be neglected with respect to n_h. The exact theory, however, predicts two plasmons [$\omega_+(q), \omega_-(q)$], which are not due to the two gases taken separately, but are two excitations deriving from their mutual interactions.[20–22] The first excitation is caused by the "in-phase" oscillation of the two gases, with a frequency $\omega_+ = \sqrt{\omega_h^2+\omega_l^2}$ and "optical" character [$\lim_{q\to0}\omega_+(q)\neq0$], while the other mode is "acoustic" [$\lim_{q\to0}\omega_-(q)=0$]. This last, however, cannot be taken into account, being unexcitable in a HREELS experiment. Thus, we calculated the model $\epsilon_{\mathrm{pl}}(\omega)$ by considering only the ω_+ excitation.

The fit to the experimental data by means of the model calculation was obtained by letting ω_+, γ_{pl}, and the dead-layer thickness (d) vary. The best-fit curves are plotted in Fig. 1 and the corresponding parameters are shown in Table I. The large value of γ_{pl} is consistent with the average γ_{pl} estimated by infrared measurements performed on the same sample.[23] The agreement between experiment and calculation is very good, revealing how a simple dielectric model can account for the intrinsic physical properties of such a system. In particular, the vibrational mode not coupled to the plasmon is found at ~36 meV, while the two plasmarons (i.e., coupled plasmon-phonon excitations) are found at about 25 and 60 meV in the calculated spectra. The latter value corresponds to the ω_+ frequency, once it is screened by the semiconductor ϵ_∞ and is strongly influenced by the extremely large damping parameter γ_{pl}.[11] The slight disagreement between experimental and theoretical spectra at high loss energy (~100 meV) could be attributed to the presence of low-energy electronic transitions among and within the valence subbands.[24] In fact, the Fermi-level position for such a doping level (2×10^{19} cm^{-3}) is below the valence-band maximum, and this allows intraband electronic transitions to take place in the absence of band bending.[24] We tried to model these excitations with a simple broad oscillator, but with little success, as such an approximation to the spectral characteristic of this electronic transitions is very crude.

We recall that the component of the momentum parallel to the surface, exchanged in a HREELS experiment, is $q_\parallel=[(2m)^{1/2}/\hbar](\sqrt{E_0}-\sqrt{E_0-\hbar\omega_{loss}})\sin\theta_i$, with θ_i the angle of incidence. Moreover, from the dipole scattering theory,[16] $1/q_\parallel$ can be assumed to be a rough estimate of the probing depth in HREELS. Thus, the dependence of a physical parameter on E_0 can be interpreted as a direct dependence on $q_\parallel$ or an indirect dependence on the probing depth.

We notice (Table 1) that the dead-layer thickness (d) is found to be almost independent on E_0 (hence on $q_\parallel$), while ω_+ presents only a slight modification and γ_{pl} shows a strong increase with E_0. Once we assume that there are neither defects nor surface states, d must be almost constant and rather small, while ω_+ can be a weak function of $q_\parallel$, given its plasmaron nature. The variation of γ_{pl} as a function of E_0 (and $q_\parallel$) is shown in Fig. 2. Also accounting for the error bar, there is a distinct growth of the plasmon damping on increasing E_0 (about 50%).

On the basis of our previous considerations, assuming the γ_{pl} values entirely determined by the hole mobility and $q_\parallel^{-1}$ only affecting the sampling depth, it could be argued that the hole mobility decreases on going from the bulk to the surface. Based on the classic theory, in our earlier paper[10] we excluded the Landau damping from the possible causes of the measured hole-plasmon width, as this excitation corresponds to the optical branch of the coupled modes of heavy and light electron gases. In a first approximation, this mode could be assimilated to the plasmon of a gas constituted by only one kind of carrier. In this case, in a standard local treatment, the Landau damping becomes relevant only when $q_\parallel$ is larger than a critical value q_c,[25] much higher than the typical values involved in HREELS measurements. However, Streight and Mills,[14] using a self-consistent treatment including the nonlocal response of the electron gas to the perturbing field, demonstrated that even in the very low $q_\parallel$ range (covered by HREELS) the Landau damping can assume large values. These authors showed, in particular, that in n-type-doped crystal,[14] there is a clear growth of the damping γ_{pl} on increasing $q_\parallel$ in the typical range of HREELS experiments. In p-type materials, besides the intraband processes, interband transitions—between the heavy- and light-hole subbands—can also occur, giving very large contribution to the damping. Therefore, in heavily p-type-doped GaAs the damping can be expected to be much stronger than in corresponding n-type crystals.[26]

Summarizing, there could be two possible causes for the E_0 dependence of the hole-plasmon damping: i) the change of sampling depth in the presence of a hole mobility lower at the surface than in the bulk; or ii) a large Landau damping with constant mobility.

TABLE I. Results from the best fit of the three-layer dielectric model calculation (Ref. 13) to the experimental data on the clean p-type GaAs(110) surface shown in Fig. 1. The primary beam energy (E_0) and the inverse of the transferred parallel momentum ($q_\parallel$) depend only on the scattering conditions. The hole-plasmon energy ω_+, the damping parameter γ_{pl}, and the dead-layer thickness d are obtained from the best fit.

E_0 (eV)	$q_\parallel^{-1}$ (Å)	ω_+ (meV)	γ_{pl} (meV)	Dead layer (Å)
2	135	217±5	78.6±3	20±5
5	180	290±5	84.1±3	16±5
10	215	280±5	67.2±3	25±5
20	288	271±5	62.3±3	25±5
30	350	271±5	62.1±3	22±5
50	450	262±5	57.6±3	25±5

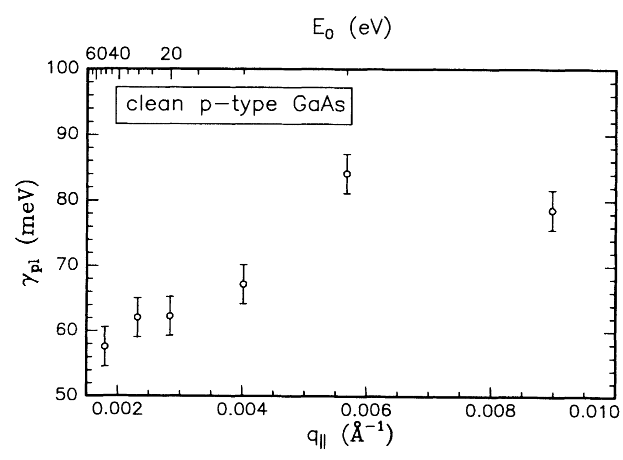

FIG. 2. Behavior of the hole-plasmon damping γ_{pl} as a function of the transferred parallel momentum $q_\parallel$, as evaluated through the fit of the model calculation to the experimental data.

There is, however, an experimental way to discriminate between these two origins, namely, increasing the depletion-layer thickness while maintaining a constant sampling depth (that is, $q_{\parallel}$). Since the energy-loss spectrum results from the integration of all the contributions to the dipolar field originating within the sampling depth, the measured plasmon width is a weighted average of $\gamma_{\rm pl}(z)$, with z normal to the surface. We can roughly write

$$\gamma_{\rm pl}^{\rm meas} = \int_0^{\lambda_s} \gamma_{\rm pl}(z) W(z) dz \ , \tag{4}$$

where λ_s is the sampling depth and $W(z)$ is an appropriate weighting function decreasing with z ($\propto e^{-q_{\parallel}z}$).[16] Assuming that $\gamma_{\rm pl}(z)$ changes on going from the bulk to the surface—while keeping constant both the sampling depth and the weighting function—data taken on samples with a lower depletion layer should show a plasmon generated in a thinner and deeper layer of free carriers, therefore on average it should show a larger $\gamma_{\rm pl}$.

In order to establish whether $\gamma_{\rm pl}$ depends on z, we performed several measurements at fixed E_0 (i.e., constant sampling depth) and modifying the depletion-layer thickness as sketched in Fig. 3. This latter effect was accomplished by chemisorption of activated atomic hydrogen, whose effect is the generation of a depletion layer[6] which adds up to the dead layer typical of the clean surface.

HREELS data relative to different H_2^* exposures—all taken at $E_0 = 5$ eV—are shown in Fig. 4, along with the respective best fits obtained by using the dielectric model previously described. A general good agreement between experiments and model calculations is found in the whole exposure range. Only the spectrum relative to the clean surface displays a discrepancy at high loss energies, again probably due to the electronic transitions within the valence-band (VB) subbands,[24] which could not be easily accounted for in the calculation. We remark that such excitations are no longer allowed when the Fermi level

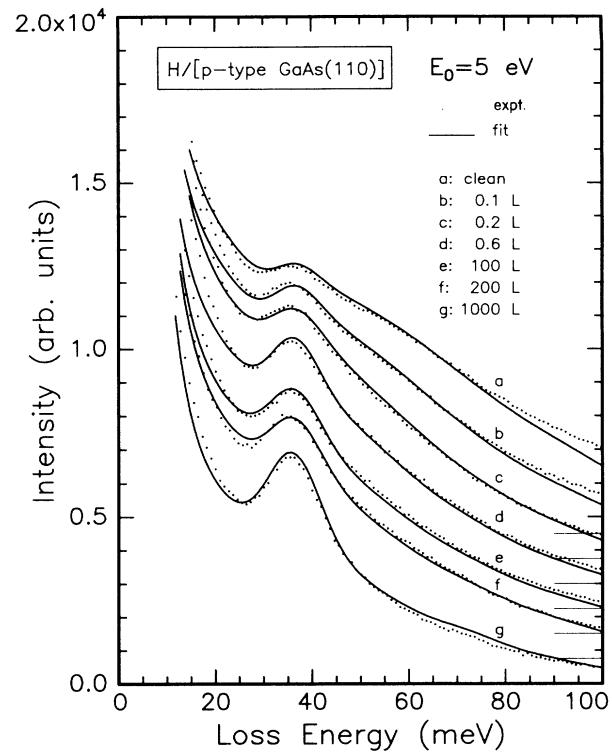

FIG. 4. High-resolution electron-energy-loss of the clean (a) and hydrogenated (b)–(g) p-type GaAs(110) surface, taken at $E_0 = 5$ eV and with angle of incidence $\theta_i = 65°$. Experimental data (dotted lines) and results of a three-layer dielectric model (solid lines) are shown. The data are displaced along the vertical axis for the sake of clarity.

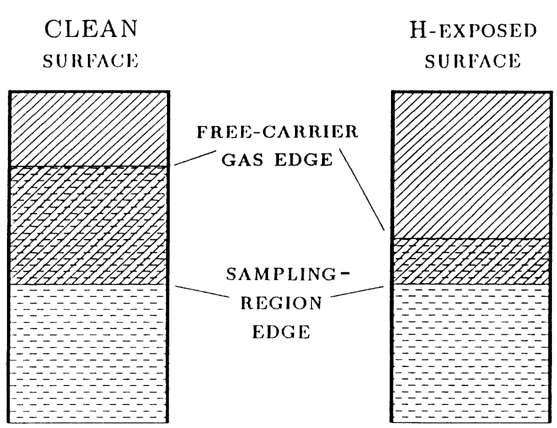

FIG. 3. Scheme of the procedure followed to evaluate the dependence of the hole mobility on the distance of the free carriers from the surface. The spectra are taken at the same primary energy (i.e., the same sampling depth). On increasing the exposure to activated hydrogen one increases the depletion-layer thickness, increasing the weight of the surface contribution to the hole mobility (plasmon damping) detected in the HREELS measurements.

shifts out of the VB into the gap region, which happens after the lowest hydrogen chemisorption, giving Fermi-level pinning. In fact, the agreement between the experimental and model data improves for the exposed samples. On increasing the H_2^* exposure, the phononlike mode at ~36 meV becomes more evident, since the carrier-free region increases and the plasmon is detected only

TABLE II. Results from the best fit of the three-layer dielectric model calculation (Ref. 13) to the experimental data on the hydrogenated p-type GaAs(110) surface, taken at $E_0 = 5$ eV (shown in Fig. 3). The inverse of the transferred parallel momentum ($q_{\parallel}^{-1}$) depends only on the scattering conditions. The hole-plasmon energy ω_+, the damping parameter $\gamma_{\rm pl}$, and the depletion-layer thickness d are obtained from the best fit.

Exposed (L)	$q_{\parallel}^{-1}$ (Å)	ω_+ (meV)	$\gamma_{\rm pl}$ (meV)	Depletion layer (Å)
clean	180	286±5	77.2±3	38±5
0.1	180	271±5	73.2±3	48±5
0.2	180	265±5	74.2±3	55±5
0.6	180	265±5	70.2±3	70±5
1	180	265±5	70.2±3	70±5
2	180	265±5	71.6±3	70±5
20	180	265±5	68.9±3	70±5
100	188	275±5	74.2±3	74±5
200	180	296±5	88.8±3	71±5
1000	180	310±5	86.8±3	100±5

through a carrier-depleted region.

The parameters resulting from the bestfits are shown in Table II and the behavior of two of them (γ_{pl} and d) as a function of exposure is shown in Fig. 5. Let us discuss first the exposure region most relevant to our problem (up to ~100 L).

As expected,[6] at very low exposures there is already an increase of the depletion region thickness, which varies from ~38 to ~74 Å, consistently with the larger carrier-free region below the surface. At variance to the increase of d there is no increase in γ_{pl} with the exposure; on the contrary, there seems to be a slight reduction (~9%, within the estimated error bar). Hence, the hole-plasmon damping (i.e., the mobility) surely does not increase (decrease) at the surface with respect to the subsurface region, as it might be argued by simply looking at Fig. 6, where the γ_{pl} is reported as a function of the depletion-layer thickness d. Moreover, according to the small reduction observed at large depletion layer, while keeping the sampling depth constant, there might be a very weak increase of the hole surface mobility with respect to the bulk. However, the main result of this analysis is that the γ_{pl} dependence on $q_{\parallel}$ must be mainly

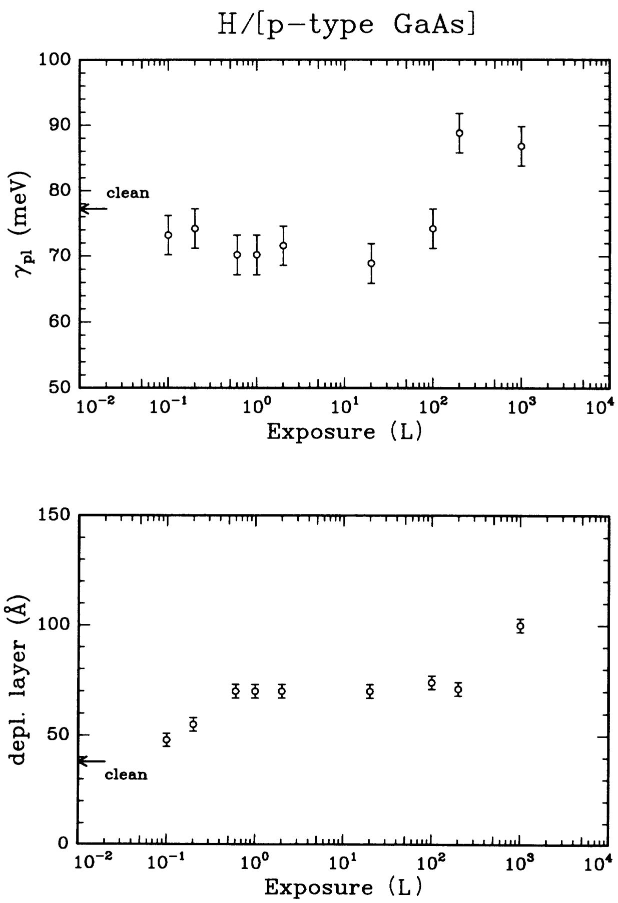

FIG. 5. Behavior of the hole-plasmon damping γ_{pl} (above) and of the depletion layer (below) as a function of the exposure to activated hydrogen. Evaluation is by means of a fit of the model calculation to the experimental data.

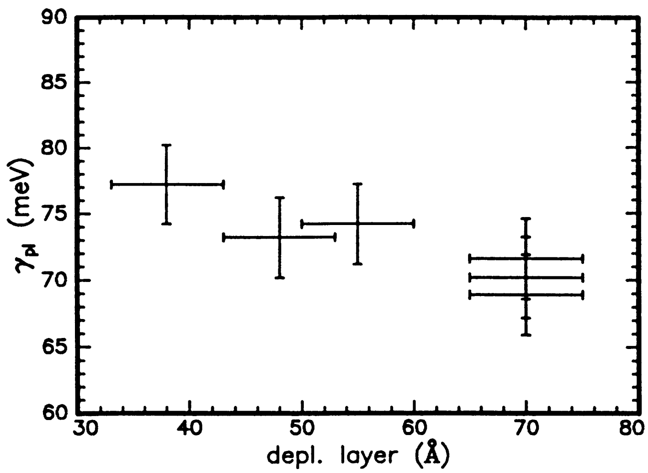

FIG. 6. Hole-plasmon damping γ_{pl} reported as a function of the depletion layer thickness d as derived from the best fit of the experimental data obtained by exposing the surface to different doses of activated hydrogen.

ascribed to the intrinsic Landau damping of the hole-plasmon excitation.

The situation at the crystal surface changes drastically when it is exposed to high doses of hydrogen. As can be observed in Fig. 5, both d and γ_{pl} show a large increase above 100 L. This modification takes place in a coverage region where, presumably, the effect of hydrogen cannot be described simply as the generation of a depletion layer. In fact, charge transfer and chemical processes that modify the physical system can take place.[27] We have no explanation for this effect in the H/GaAs(110) interface. It will be investigated in the future.

CONCLUSIONS

We have presented a high-resolution electron-energy-loss investigation of the hole-induced plasmon on the surface of heavily doped *p*-type GaAs(110). By means of an appropriate three-layer dielectric model we could fit the data taken at different primary beam energies, obtaining the values of the dead-layer thickness and of the plasmon damping as a function of the transferred parallel momentum $q_{\parallel}$. A possible dependence of the large plasmon damping on the probing depth, hence a dependence of the hole mobility on the z direction normal to the surface, was ruled out after a series of measurements. By chemisorbing hydrogen on the surface we could modify the depletion-layer thickness while maintaining a constant probing depth and $q_{\parallel}$. The plasmon damping was found to be essentially independent of the hydrogen dose, in the low exposure range, i.e., of the carrier population below the surface. Hence, the mentioned dependence of the plasmon damping on $q_{\parallel}$ must be ascribed to a strong Landau damping present at very low $q_{\parallel}$ values, as it was expected by Streight and Mills[14] for *n*-type carriers on the basis of the nonlocal nature of the free-carrier response, but not yet measured or calculated on *p*-type GaAs. A theoretical effort in explaining our data would be welcomed.

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