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Vibrational and collective excitations of the Cs/GaAs(110) interface

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We present a high-resolution electron-energy-loss analysis of the interface system Cs/GaAs(110) grown at room temperature. We study the development of the GaAs collective excitation features (Fuchs-Kliewer surface phonon and plasmon of the dopant-induced free carriers) as a function of the alkali-metal deposition. An energy-loss structure at about 8 meV is inferred by a detailed data analysis, that we attribute to a Cs-induced excitation. By using an appropriate dielectric model applied to our data, Cs does not form a uniform metallic layer at saturation coverage, while it cannot be ruled out that it forms metallic clusters of a few tens of Å in size. Cs deposition is known to produce a band bending at the (110) surface of GaAs. Using a semiclassical model for interpreting the energy and intensity modifications of the plasmon and phonon modes, we also deduced the widening of the depletion layer due to the adsorbate. We then obtained a band bending of 0.73 eV for the *n*-type doped substrate, in good agreement with previous results. We remark that this indirect method for determining the band bending is not affected by any surface photovoltage effects.

I. INTRODUCTION

The adsorption of alkali metals on GaAs(110) has been widely studied in the past few years. Among the alkali metals, Cs, forming an unreactive and nondisruptive interface with GaAs,[1] has been the most analyzed one. A lot of work has been carried out to understand the morphology[2,3] and the electronic structure[1,4–7] of this system. In spite of all these efforts, some open questions and controversies remain. In particular, it is still discussed in the literature whether the saturated Cs monolayer (ML) at room temperature (RT) is metallic or not. Scanning tunneling spectroscopy[3] and inverse photoemission[5] results favor a metallization of the overlayer at saturation coverage; meanwhile, direct photoemission[8,9] and previous high-resolution electron-energy-loss spectroscopy (HREELS) (Ref. 6) experiments fail to detect a metallic behavior. A recent first-principles linear augmented-plane-wave calculation on a jellium-slab model of this system[10] confirms a previous self-consistent pseudopotential one,[11] showing that one monolayer of Cs on GaAs(110) does not produce electronic states closing the interface band gap.

Moreover, the growth morphology of alkali metals on III–V semiconductors presents very interesting aspects. Scanning tunneling microscopy (STM) showed that Cs atoms form zigzag chains at very low coverages on the (110) surface of GaAs, that further deposition leads to the creation of double and triple chains, and that the saturation coverage at RT corresponds to the formation of Cs clusters.[2,3] This evolution from one-dimensional to three-dimensional structures is another interesting property of this system. HREELS has been used to gain information on both the vibrational and the collective excitations of several metal/GaAs(110) interfaces. For example, through accurate analysis of the substrate's typical excitations (Fuchs-Kliewer phonon and dopant-induced free-carrier plasmon), it was possible to determine the dielectric nature (semiconducting, semimetallic) of the Sb/GaAs(110) interface at the various coverage steps.[12,13] Moreover, previous HREELS investigations of the GaAs(110) interface with potassium, although not conclusive, showed modifications of the low-energy electronic structure and of the vibrational features.[14,15] Therefore, we decided to deepen the knowledge of the Cs/GaAs(110) interface in the light of these considerations, using the HREELS technique. The dielectric character of the interface—at saturation coverage—will be shown as not being uniformly metallic and an estimate of the band bending on *n*-type doped GaAs, as well as the thickness of the corresponding depletion layer, will be given. The band-bending value is in agreement with previous results[1,4] in the absence of any surface photovoltaic effects. These deductions can be drawn from an appropriate analysis of the dielectric screening of the substrate modes performed by an opportune fit of model calculations to the experimental data.

II. EXPERIMENTAL DETAIL AND DATA ANALYSIS

A. Experimental detail

The experiments have been carried out at the surface physics laboratory Spettroscopia Elettronica Superfici Adsorbati Modena (SESAMO), Dipartimento di Fisica, Università di Modena. The UHV system, constituted by two chambers (one devoted to sample preparation, the other to electron spectroscopy analysis), is equipped with Auger electron spectroscopy, low-energy electron-diffraction (LEED), x-ray photoelectron spectroscopy, ultraviolet photoelectron spectroscopy, and HREELS. HREELS measurements were performed with a Leybold-Heraeus ELS-22 spectrometer. The spectra were taken in the specular direction using an incident angle of 61°. A primary-electron-beam energy of 25 eV was used and a resolution of 6.5 meV [full width at half maximum (FWHM)] on the clean GaAs(110) surface was achieved.

The *n*-type doped [$n_{(\mathrm{Te})}\approx 2.7\times 10^{18}$ cm^{-3}] GaAs(110) bars were cleaved *in situ* at a pressure of 8×10^{-9} Pa, showing a good (1×1) LEED pattern. Cs was evaporated from well-outgassed dispensers, keeping the basis pressure below 2×10^{-8} Pa during deposition.

B. Determination of the Cs coverage

As is well known in the literature, alkali deposition on the surface of metals and semiconductors causes a lowering of their work function. The reproducibility of this property on different surfaces was taken into account to calibrate the Cs depositions. The work-function change ($\Delta\Phi$) was derived by measurement of the cutoff shift of the secondary electrons in ultraviolet photoemission spectra (He I, $\hbar\omega=21.2$ eV). We slightly biased the sample (bias of -4 V) to get rid of any spurious effects due to the $\Delta\Phi$ between the crystal and the analyzer. The work-function change $\Delta\Phi$ as a function of the Cs deposition is shown in Fig. 1. As reported by other authors in analogous experiments at room temperature,[7] there is a saturation in the $\Delta\Phi$ curve that is conventionally indicated as the formation of the first monolayer. Assuming the sticking coefficient to be constant, we performed a linear calibration in terms of fractions of a monolayer ML, where 1 ML is defined as one Cs atom per surface unit cell.

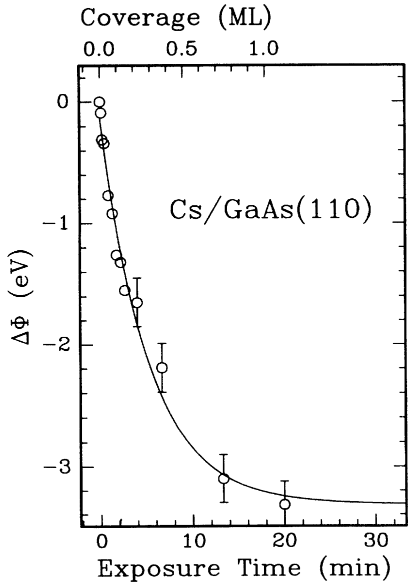

FIG. 1. Work-function change $\Delta\Phi$ of the interface Cs/GaAs(110) as a function of the deposition time. $\Delta\Phi$ has been evaluated by photoemission measurements (He I, $\hbar\omega=21.2$ eV) by measuring the shift of the secondary electrons cutoff (sample bias of -4 V). The Cs monolayer is completed at the beginning of the work function's saturation (~ 20 min).

C. Data analysis

The analysis of the HREELS data has been carried out with a model calculation[16] using a semiclassical model of the interaction of low-energy electrons (1–100 eV) with a surface. We recall that in the very small-angle scattering conditions of an energy-loss experiment, the momentum transferred from the impinging electron to the surface low-energy excitation is very small. Consequently, the main contribution to the excitation cross section is due to a dipole interaction between the electron and the dipole moment of the excitation. Taking into account the "dipole selection rule," a semiclassical model can provide the probability $P_{\mathrm{cl}}(q,w)d\Omega_k d(\hbar\omega)$ that an electron with a wave vector $\mathbf{k}$ is scattered into the solid angle $d\Omega_k$, with a momentum transfer $\mathbf{q}=\mathbf{k}-\mathbf{k}'$ and releasing the kinetic energy $d(\hbar\omega)$:[17–19]

$$P_{\mathrm{cl}}(\mathbf{q},w)=\frac{2\hbar q}{\pi}[1+n(\omega)]\mathrm{Im}\frac{-1}{\epsilon_{\mathrm{eff}}(\mathbf{q},\omega)+1},$$

where $n(\omega)$ is the Boltzmann factor and ϵ_{eff} the dielectric function of the whole system. For a system of n layers, ϵ_{eff} can be expressed in terms of the thickness d_j and the dielectric function ϵ_j of each layer j:[16]

$$\epsilon_{\mathrm{eff}}(q,\omega)=a_1-\cfrac{b_1^2}{a_1+a_2-\cfrac{b_2^2}{a_2+a_3-\cfrac{b_3^2}{a_3+a_4-\cdots}}},$$

with the coefficients

$$a_j=\epsilon_j(\omega)/\tanh(qd_j)$$

and

$$b_j=\epsilon_j(\omega)/\sinh(qd_j).$$

In the particular case of the Cs/GaAs(110) interface, we can define the appropriate dielectric function $\epsilon_j(\omega)$ for each layer. Knowing that the HREEL spectrum of clean GaAs can be fitted by a two-layer model (one representing the surface depleted of free carriers, the second the bulk),[20,21] we added one more layer characterizing the Cs adlayer to fit the data relative to the interface. We therefore chose a model for each layer (adlayer, depletion layer, bulk), using Lorentzian shape for the vibrational features and a drudelike term for the plasmon excitation:

$$\epsilon_{\mathrm{Cs}}(\omega)=\epsilon_{\mathrm{Cs}\infty}+\frac{Q_{\mathrm{Cs}}\omega_{\mathrm{Cs}}^2}{\omega_{\mathrm{Cs}}^2-\omega^2-i\gamma_{\mathrm{Cs}}\omega},$$

$$\epsilon_{\mathrm{depl}}(\omega)=\epsilon_{\mathrm{GaAs}\infty}+\frac{Q_{\mathrm{GaAs}}\omega_{\mathrm{TO}}^2}{\omega_{\mathrm{TO}}^2-\omega^2-i\gamma_{\mathrm{TO}}\omega},$$

$$\epsilon_{\mathrm{bulk}}(\omega)=\epsilon_{\mathrm{GaAs}\infty}+\frac{Q_{\mathrm{GaAs}}\omega_{\mathrm{TO}}^2}{\omega_{\mathrm{TO}}^2-\omega^2-i\gamma_{\mathrm{TO}}\omega}-\frac{\omega_{\mathrm{pl}}^2}{\omega^2-i\gamma_{\mathrm{pl}}\omega},$$

where ω_{pl} and ω_{TO} are the frequencies of the free-carrier plasmon and of the transversal-optical phonon of GaAs, ω_{Cs} is the Cs-atom vibration frequency (see discussion below), $\epsilon_{\mathrm{GaAs}\infty}$ and $\epsilon_{\mathrm{Cs}\infty}$ are the GaAs and Cs dielectric constants for high frequencies in the infrared region, γ_j are damping factors, and Q_j are strength parameters.

III. RESULTS AND DISCUSSION

A. Vibrations of Cs on GaAs

The experimental HREELS data for a freshly cleaved GaAs(110) surface in the energy range −40–100 meV is shown in Fig. 2(a). Apart from the quasielastic peak, the spectrum relative to clean GaAs presents three well-resolved structures. The first one, located at ~35 meV, is due to the excitation of the surface transversal-optical phonon (Fuchs-Kliewer surface phonon).[22] The peak at −35 meV is the correspondent energy gain due to absorption of one Fuchs-Kliewer phonon by the scattered electrons. Of course, the ratio between the gain and loss intensities is a function of the temperature[17] and is very well fitted by the model calculation. The third feature, placed at about 57 meV, is due to the plasmon excitation of the dopant-induced free carriers on the surface and its energy-loss position is a function of the sample doping.

Experimental data in Figs. 2(a)–2(f) (points) show the evolution of the mentioned structures on increasing the Cs thickness. An uptake of Cs on the freshly cleaved GaAs(110) surface causes a weak diminution of the intensity of the Fuchs-Kliewer peak, its energy position remaining fixed. On the other hand, the increase of the alkali-metal coverage does not only cause a strong diminution of the plasmon intensity, but also induces an energy shift of 2.5 meV to lower values. Cs deposition also influences the quasielastic peak as its FWHM increases with respect to that on the clean surface.

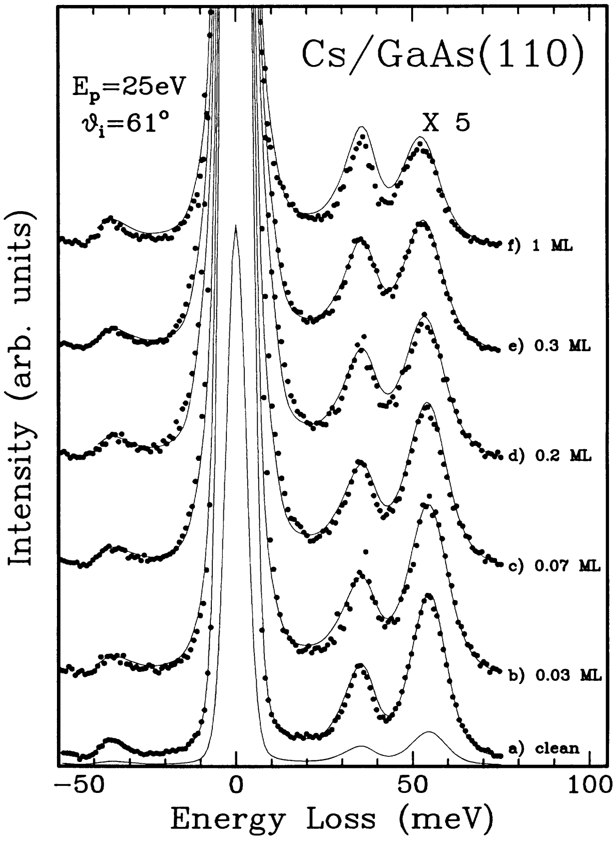

FIG. 2. HREELS data of the Cs/GaAs(110) interface on increasing the Cs deposition. The primary-beam energy was 25 eV, the resolution 6.5 meV, and the incident angle 61°. Continuous lines correspond to the simulation with the semiclassical dielectric model fitting the data (see text).

As mentioned above, the spectrum of clean cleaved GaAs can be quite well described by a two-layer model.[20,21] The reason for defining a "surface" layer different from the bulk has experimental and theoretical motivations, both indicating the absence of free carriers at the surface and in the subsurface region. The cleavage of a semiconductor crystal can give rise to defects on the surface, producing surface states and an initial band bending correlated with a depletion region. Moreover, Ehlers and Mills[23,24] demonstrated that even at an ideal surface, a "dead layer" free of carriers forms at the topmost layers of the crystal. This intrinsic surface space-charge region is caused by the surface boundary conditions of the electron wave functions. These two reasons require the inclusion of a depletion region even in the model of the clean surface. The result of this model calculation for clean GaAs, including a bulk and a surface layer, is shown as a continuous line in Fig. 3(a) superimposed on the experimental data (dots); the values of the fitting parameters are shown in Table I.

Knowing that Cs adsorption leads to the pinning of the Fermi level and a consequent enlargement of the depletion region,[1,4] we tried to extend the fitting procedure to the HREELS spectra taken after Cs deposition. The use of a two-layer model for the spectrum relative to 0.3 ML Cs deposition on GaAs(110) furnishes only a poor fit [Fig. 3(b)]. Even if both the loss features (phonon and plasmon) are well fit, the region close to the quasielastic peak reveals a bad concordance. As a matter of fact, a further layer—with its specific excitations—must be taken into account to reproduce the experimental data. The first attempt we made was to introduce a Cs layer with metallic character [by using the proper Cs surface-plasmon oscillation at 2.1 eV (Ref. 25)]. As it will be shown in the following section, none of the spectra relative to different Cs coverages could be properly reconstructed. Better results [Fig. 3(c)] could be obtained by assuming the presence of a mode at ~8 meV. HREEL measurements of alkali metal on metals[26,27] showed the

TABLE I. Parameters used in the semiclassical three-layer model for each layer (Cs adlayer; GaAs surface; GaAs bulk).

	Cs	GaAs depl.	GaAs bulk
$\epsilon(\infty)$	1.0	10.9	10.9
Q	7.0–0.3	2.03	2.03
$\hbar\omega$	8 meV	33.3 meV	33.3 meV
$\hbar\gamma$	6 meV	0.31 meV	0.31 meV
$\hbar\omega_{\mathrm{pl}}$			214 meV
$\hbar\gamma_{\mathrm{pl}}$			8.4 meV

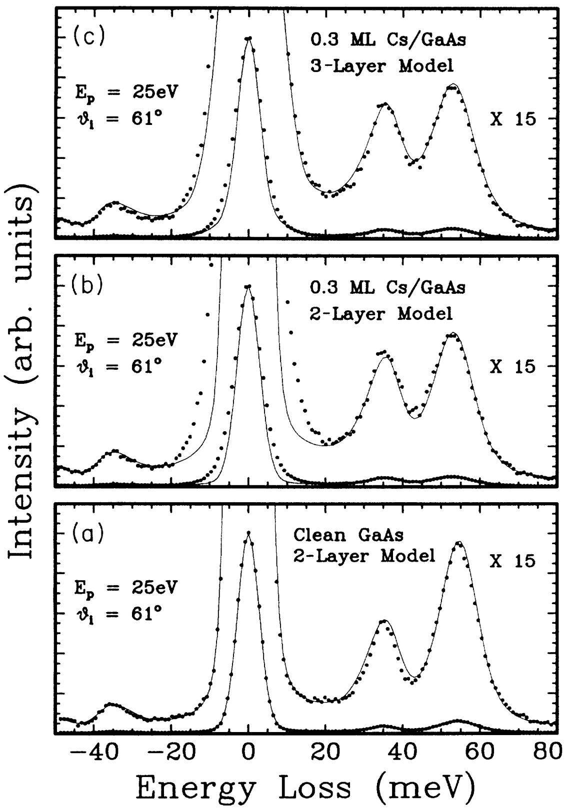

FIG. 3. (a) Experimental HREELS data (points) relative to clean GaAs(110) and fitting simulation obtained using a *two*-layer model. (b) Experimental HREELS data (points) relative to 0.3-ML Cs/GaAs(110) and fitting simulation obtained using a *two*-layer model. (c) Experimental HREELS data (points) relative to 0.3-ML GaAs(110) and fitting simulation obtained using a *three*-layer model.

frequency of the stretching vibration of Li, Na, and K against a copper substrate to be proportional to the square root of the reduced masses of the elements (see Table II). Therefore, within the crude approximation of a spring-strength constant for all the measured alkali metals and independent of the substrate, in a first approximation we can expect the stretching vibration of Cs towards GaAs to be 7.6 meV. Hence, the mode at ~8 meV could be reasonably attributed to a Cs oscillation against the substrate.

TABLE II. Stretching vibration modes between alkali metals and Cu(110) (Ref. 27), Cu(111) (Ref. 26), and GaAs(110) (this work). Energy positions are given in meV.

	Cu(110)	Cu(111)	GaAs(110)
Li	33 meV (Ref. 27)		
Na	18 meV (Ref. 27)		
Na		21 meV (Ref. 26)	
K	14 meV (Ref. 27)		
Cs			8 meV (this work)

B. Metallization of the overlayer

As it was explained in the preceding section, all the spectra could be fitted by using a model system whose topmost layer does not present a metallic character. The scope of this section is to deepen the knowledge of the dielectric nature of the Cs overlayer at saturation coverage. As it was discussed in the Introduction, the metallic character of this interface is still under debate. Hence, in perspective, to gain information about the onset of the metallization, we used the three-layer calculation to simulate a metallic overlayer. We introduced the Cs surface-plasmon excitation at 2.1 eV (Refs. 6 and 28) in the topmost layer and let its thickness increase up to the radius of metallic Cs. The result of the model calculation at different thickness is shown in Fig. 4, along with the HREELS data for the saturation coverage. The thickness varied from 1.75 to 3.5 Å to cover the range from the ionic radius (1.75 Å) to the covalent one (2.3 Å). The simulation considers a *uniform homogeneous* metallic overlayer on top of a semiconductor, screening the dipole field of the collective excitations lying underneath.[29] As a consequence, the HREELS loss features are strongly damped and quenched. Moreover, the coupling of the GaAs plasmon to that of the metallic overlayer causes a wide broadening and asymmetry of the GaAs plasmon loss in the model spectra. As the experimental data show a symmetric and narrow plasmon-related peak and its intensity ratio relative to the phonon is different from that coming out from the simulations, we deduce that the saturated monolayer does not consist in a metallic layer. On the other hand, we recall that Cs has been shown to form metallic clusters by STM.[3] However, photoemission measurements[8,30] showed only a very low density of free carriers at the Fermi level. HREELS data confirm the latter observations, as neither we, nor Di Nardo, Wong, and Plummer[6] in a similar experiment, could

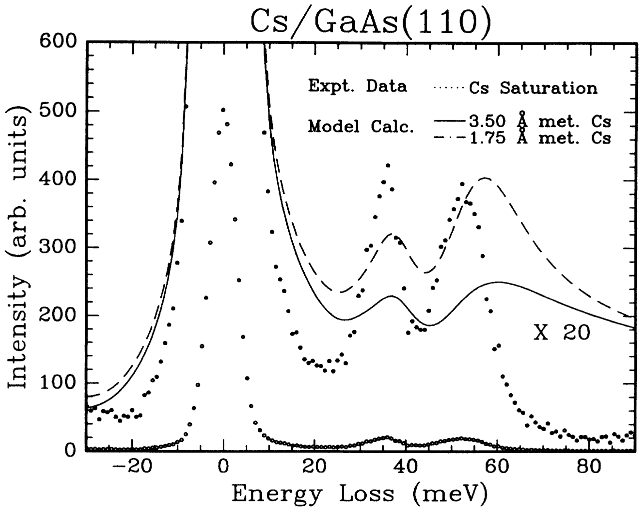

FIG. 4. HREELS data relative to the saturation coverage of Cs on GaAs(110) compared with simulations obtained considering uniform metallic overlayers of different thicknesses (1.75 Å, dashed line and 3.50 Å, continuous line).

detect any feature attributable to a Cs-derived free-electron plasmon. On the other hand, plasmon excitation from the overlayer clusters could only be detected if their size D was of the order of the inverse transferred momentum $q_{\parallel}(D \approx q_{\parallel}^{-1})$.[17] In the present experimental conditions the maximum transferred momentum corresponding to the Cs plasmon (2.1 eV) is 0.096 Å^{-1}, giving $D \sim 10$ Å. Therefore, if metallic clusters are present at the surface, as shown by STM,[3] they cannot be larger than a few tens of Å.

C. Determination of the band bending

As presented in Secs. III A and III B, the analysis of the HREELS data through appropriate dielectric multilayer models leads to the determination of the extension of the space-charge region depleted of free carriers, below the semiconductor surface. Within the framework of the Schottky model[31] for the space-charge region, one can use the simple relation between the depletion layer thickness $d_{\rm depl}$ and the potential difference caused by the band bending:

$$V = -\frac{en}{2\epsilon\epsilon_0} d_{\rm depl}^2 ,$$

where n is the substrate doping, e is the elementary charge, ϵ and ϵ_0 are the dielectric constant and vacuum permittivity. To extract the exact band bending we must consider the net effect of charge depletion caused by the adlayer, i.e., we must substract the dead-layer thickness $d_{\rm DL}$ from the fit $d_{\rm depl}$ value: $d_{\rm DL}$ should be about 30 Å, as given by Ehlers and Mills[23,24] for GaAs with the same doping of our crystal. We obtain for the clean surface $d_{\rm depl} = 120$ Å, indicating an initial band bending V of 290 meV. This last value confirms that the cleavage technique can produce a small density of defects, especially in highly doped samples. On increasing the Cs coverage, the GaAs plasmon peak shifts to lower-loss energies and the phonon structure undergoes a damping. Such effects can be quantified by performing a fit to the data; the former, however, is already visible in the raw data of Fig. 2. Both are caused by an enlargement of the depletion layer. To get a quantitative estimation of V we let the depletion-layer thickness vary for fitting the experimental data and using Schottky's law we estimated the band bending. The results of this procedure are shown in Fig. 5, along with previous data obtained by photoemission.[1,4] With such a doping level of the substrate and having measured at room temperature, the surface photovoltage in the photoemission-derived data should be very small in the present case, hence, there is clearly a very good agreement between the photoemission-derived data and our data. However, we would emphasize that the estimation of the E_F pinning through measurements of surface-core-level shift by photoemission can be affected by surface photovoltage effects (SPV), while HREELS data are SPV free, being measured with typical current densities smaller than 2.5 nA/cm^2.

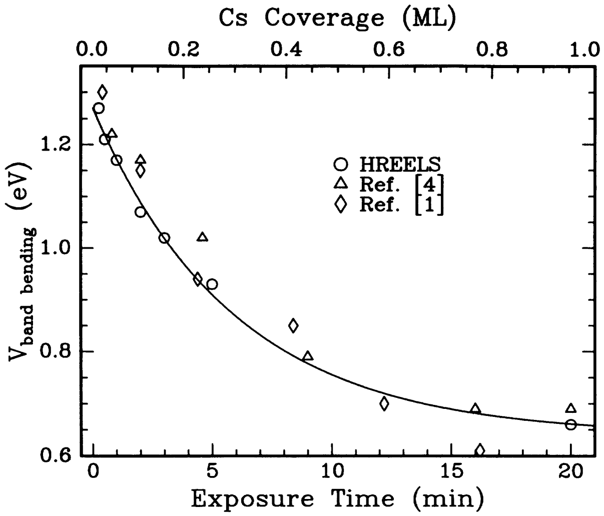

FIG. 5. Band bending at the Cs/[n-type doped GaAs(110)] as a function of the Cs coverage, as derived by HREELS measurement (open circles). Comparison with data obtained by photoemission at the same interface [Prietsch *et al.* (Ref. 1), diamonds and Kendelewicz *et al.* (Ref. 4), triangles]. The continuous line serves only as a guide for the eye.

IV. CONCLUSIONS

HREELS measurements have been performed to characterize the interface system Cs/[n-type doped GaAs(110)] grown at RT. By means of a three-layer semiclassical dielectric model for fitting our data, we estimated the alkali-metal-induced band bending $V_{\rm bb} = 0.73$ eV at 1 ML of Cs, in agreement with previous photoemission result. This indirect method for measuring the Fermi-level pinning at an interface is absolutely free from possible surface photovoltage effects. Moreover, the Cs overlayer at the completion of one monolayer could be identified as not being uniformly metallic, but there can be metallic clusters with a mean size not larger than a few tens of Å. An additional mode could be found at 8 meV, probably due to vibrations of the Cs atoms towards the GaAs substrate.

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