

OPTICAL DETECTION OF SURFACE STATES IN GaAs(110)  
AT ENERGIES ABOVE THE GAP

P.Chiaradia, G.Chiarotti, F.Ciccacci, R.Memeo  
S.Nannarone, P.Sassaroli, S.Selci

Department of Physics, University of Rome, Italy and GNSM-CNR

Optical transitions between As-derived and Ga-derived surface states are detected in GaAs(110) by differential reflection spectroscopy at energies around 3 eV. Hydrogen on top of GaAs(110) seems to introduce a broad absorption band reminiscent of the continuous spectrum of the diatomic molecules. A discussion of the experimental results in the frame of the semiclassical theory of McIntyre and Aspnes is presented.

Optical detection of surface states in semiconductors at energies below the gap has proved very useful for studying the structure of electronic states on reconstructed surfaces [1]. The method has been recently extended to energies above the gap, greatly increasing its potentiality [2,3].

The method essentially consists in a measurement of normal differential reflectivity in presence and absence of surface states (for example for a clean and oxidized surface). At energies above the gap, however, where the substrate is itself absorbing a change of reflectivity is expected also when a dielectric (non absorbing) film (for example the oxide layer) is present on top of the surface. Moreover, at relatively large energies the adsorbate itself may be optically absorbing, so that a careful discussion is necessary before attributing any change of reflectivity to surface states.

We present here results for GaAs(110) exposed to H<sub>2</sub> and O<sub>2</sub> that illustrate in detail the various cases that might occur.

The experimental apparatus and methods for surface differential reflectivity have been described elsewhere [2]. The fractional change of reflectivity, defined as:

$$\frac{\Delta R}{R} \equiv \frac{R_{\text{clean}} - R_{\text{ads}}}{R_{\text{ads}}}, \quad (1)$$

where  $R_{\text{ads}}$  is the reflectivity of the sample when covered with an adsorbate layer, is plotted as a function of energy in Fig.(1), for GaAs(110) exposed to oxygen, and in Fig.(2), for GaAs(110) exposed to hydrogen.  $R_{\text{ads}}$  has been taken as the asymptotic value of reflectivity after exposure to oxygen and hydrogen respectively. It is seen

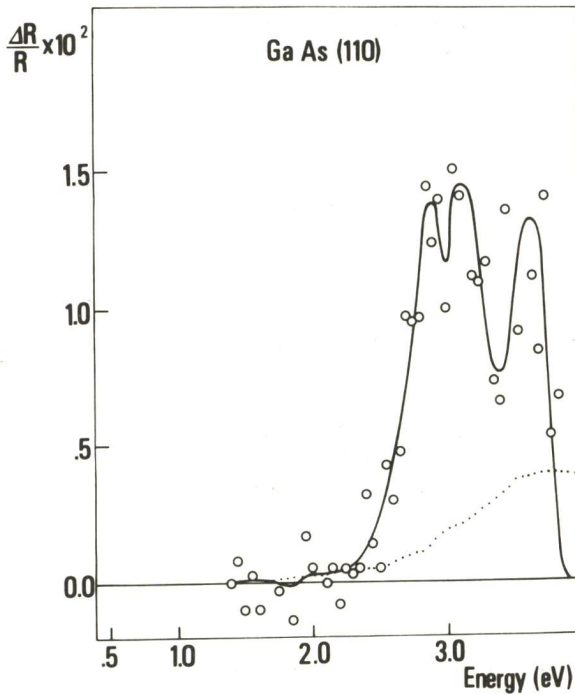


Fig.1 Fractional change of reflectivity as a function of energy for GaAs(110), taking as a reference the reflectivity of the surface after exposure to oxygen: The dotted line represents the mixed term of eq. (3) evaluated for  $d=5\text{\AA}$

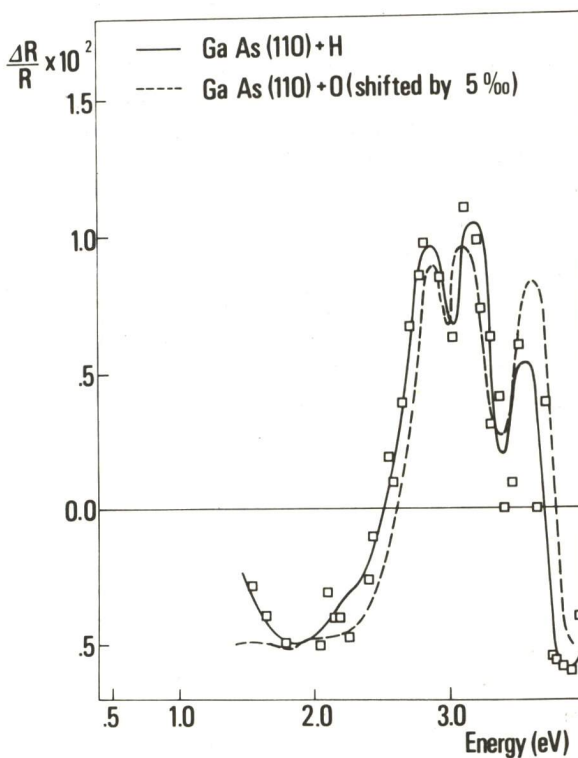


Fig.2 Fractional change of reflectivity as a function of energy for GaAs(110) taking as a reference the reflectivity of the surface after exposure to hydrogen: The dashed curve is the (solid-line) curve of Fig. (1) displaced by -0.5%

that, in both cases, the main structure consists of two peaks at approximately 3 and 3.6 eV with the first peak split as well. The curve for the hydrogenated surface seems merely displaced by  $\sim 0.5\%$  towards the negative values of  $\Delta R/R$ . The dashed curve of Fig. (2) is in fact the solid-line curve of Fig. (1) displaced by  $-0.5\%$ . Though some of the features of the two curves are at the limit of the experimental accuracy, their similarity is striking and supports the conclusion that hydrogen on the surface of GaAs(110) introduces an absorption band rather flat in the range between 1.3 and 4 eV.

In order to discuss quantitatively the experimental results, a theory of the optical response of a surface is required. A simple classical theory that, for energies below the gap, yields results in qualitative agreement with experiments [4] has been developed by McIntyre and Aspnes [5]. Such a theory assumes that the surface is a homogeneous film of thickness  $d$  on top of a substrate and that the three media (external, surface, substrate) have definite dielectric functions  $\hat{\epsilon}_j = \epsilon'_j - i\epsilon''_j$  ( $j=1,2,3$ ).

When the external medium is the vacuum, it is found:

$$\frac{R(d)-R(0)}{R(0)} = -\frac{8\pi d}{\lambda} \operatorname{Im} \left( \frac{1-\hat{\epsilon}_2}{1-\hat{\epsilon}_3} \right) \quad (2)$$

For energies above the gap ( $\hat{\epsilon}_j$  complex) eq. (2) can be considerably simplified under the assumption that  $d$  is the same for the surface and the adsorbate films. One easily obtains:

$$\begin{aligned} \frac{\Delta R}{R} &\equiv \frac{R_{\text{clean}} - R_{\text{ads}}}{R_{\text{ads}}} \approx \frac{R_{\text{clean}} - R(0)}{R(0)} - \frac{R_{\text{ads}} - R(0)}{R(0)} \approx \\ &- \frac{8\pi d}{\lambda} \left\{ \frac{(1-\epsilon'_3)\epsilon''_2}{(1-\epsilon'_2)^2 + (\epsilon''_3)^2} + \frac{(\epsilon'_2 - \epsilon'_{\text{ads}})\epsilon''_3}{(1-\epsilon'_3)^2 + (\epsilon''_3)^2} - \frac{(1-\epsilon'_3)\epsilon''_{\text{ads}}}{(1-\epsilon'_3)^2 + (\epsilon''_3)^2} \right\} \end{aligned} \quad (3)$$

where  $\hat{\epsilon}_{\text{ads}}$  is the dielectric function of the adsorbate layer.

The first term represents the contribution of the surface states: it is the only one present when the substrate and the adsorbate are transparent (like for Ge and Si(111)2x1 at energies below the gap). The third term represents the absorptive contribution of the adsorbate itself. The second term represents a mixed contribution and is present even when there are no surface states and the adsorbate is transparent. However, in absence of surface states the second term can be calculated from bulk data and atomic polarizabilities [2], and it turns out that it is generally small. The dotted line of Fig. (1) represents such a contribution evaluated for  $d = 5\text{\AA}$ .

It is seen from eq. (3) that the sign of the last term is opposite to that of the first one, i.e. that the last term gives rise to a negative contribution to  $\Delta R/R$ .

From the previous analysis and the curves of Figs. (1) and (2), it seems then plausible to conclude i) that  $\epsilon''_{\text{oxygen}} = 0$  while

$\epsilon''_{\text{hydrogen}} \neq 0$ ,  $\epsilon''_{\text{oxygen}}$ ,  $\epsilon''_{\text{hydrogen}}$  being the imaginary parts of the

dielectric functions of the oxide and hydrogen layer; ii) the contribution of the surface states is the same in the two cases; iii) hydrogen on top of GaAs(110) gives rise to a broad absorption band rather flat in the explored range.

In the frame of the rotation-relaxation model of the surface of GaAs(110), the optical absorption due to surface states is associated to transitions from a filled As-derived state "localized" on the outwardly displaced As to an empty Ga-derived state "localized" on the inwardly displaced Ga. As such, it was predicted theoretically at energies around 3 eV [6].

The broad band related to hydrogen ( $\Delta R/R < 0$ ) is reminiscent of the continuous spectrum of the diatomic molecules for which the electron-nuclei interaction is strong. Theoretical studies of the properties of the As-H bond could clarify this suggestive hypothesis.

#### References

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