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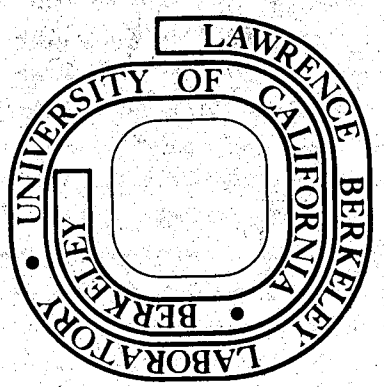

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EFFECTS OF UNIAXIAL STRESS ON THE E_0' PEAKS IN THE DERIVATIVE REFLECTIVITY OF GALLIUM ARSENIDE

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Effects of Uniaxial Stress on the E_o' Peaks in the Derivative Reflectivity of Gallium Arsenide

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ABSTRACT

We have measured the wavelength-modulated reflectivity of GaAs in the E_o' region of the spectrum, under applied uniaxial stresses up to 8 and 9 kilobars in the (111) and (001) directions at 5°K. The results indicate a composite structure, dominated by Δ critical points, with critical points of non-Δ symmetry probably contributing to the E_o' structure. The origin of these critical points is discussed in the light of the current understanding of the band structure of GaAs. Numerical values for the deformation potentials are given, based on our assignment of the main structure at 4.5 eV to Δ transitions.

Nous avons mesuré la réflectivité de GaAs modulée en longueur d'onde dans la région E_o' du spectre sous application d'une tension uniaxiale allant jusqú à 8 et 9 kilobars dans les directions (111) et (001), à 5°K. Les résultats indiquent une structure composée, dominée par les points critiques Δ, avec une probable contribution des points de non-Δ symétrie à la structure E_o'. L'origine de ces points critiques est discutée à l'aide des connaissances actuelles de la structure de bandes de GaAs. Nous donnons les valeurs numériques des potentiels de déformation, basées sur notre attribution de la structure principale à 4.5 eV aux transitions Δ.

I. INTRODUCTION

Recently attention has been focused on some of the smaller structures in the optical spectra of diamond- and zincblende-structure semiconductors. In particular, the effects of uniaxial stress on the E_o' peaks of Si [1,2] and Ge [3] have been studied at helium temperature, with a view to assigning these weak structures to transitions at points in the Brillouin zone. Measurement of the derivative of reflectivity, either directly by wavelength modulation [1,2] or indirectly by numerical calculation from very clean reflectance spectra, [3] was necessary in order to detect the small peak shifts due to stress. In this paper we report the first application of this technique to the E_o' structure of a zincblende crystal, GaAs. Wavelength modulation spectra in the range 4.2 to 4.8 eV, under the influence of large uniaxial compression in the (111) and (001) directions, are presented, along with our interpretation. All curves were taken at 5°K.

The results indicate that the E_o' structure is a composite one, as with Si and Ge, and that its main features at 4.5 and 4.7 eV are most likely caused by critical points in the interband transitions along Δ. Some small structures present exhibit stress and polarization dependence inconsistent with a Δ assignment and could arise from transitions to the higher conduction band at or near Γ. This conclusion, that Δ is the main symmetry, is supported by the recent work of Rehn and Kyser, [4] analyzed the polarization dependence of transverse electroreflectance and found predominantly Δ-behavior in this spectral region. (Their method would not reveal weaker Γ structure.) Also, piezoreflectance measurements of Wells and Handler [5] indicate Δ transitions in this region, although detailed structure in E_o' could not be resolved.

The new Schottky-barrier electroreflectance results of Aspnes and Studna [6] reveal the composite nature of this region. Their assignment of Δ and Γ critical points between 4.5 and 4.7 eV is the same as what we concluded, although firmer.

Various band structure calculations [7,8] have indicated that the main E_o' structure arises from a combination of M_o and M_1 critical points along Δ, there being two of these combinations due to the spin-orbit splitting of the upper Δ valence bands originating from Γ_8. The $k \cdot p$ calculation of Pollak, et al. [7] predicted that these critical points represent transitions to the upper valence band near Γ_7, while the energy of transitions to the lower conduction band increases monotonically from Γ_6 to X_6. Early pseudopotential calculations, [8] on the other hand, showed the major contribution to the E_o' structure to come from transitions to the lower band, with an M_1 point near the "pseudocrossing" and an M_o point about 2/3 of the way toward X.

Recent pseudopotential calculations by Cohen and Chelikowsky,[9] using non-local pseudopotentials and based in part on Aspnes and Studna's data, have apparently resolved this conflict in favor of the former configuration, predicting M_1 points about 5-10% of the way from Γ_7 and weak M_o points near the "pseudocrossing." The shape of their calculated derivative reflectivity agrees well with experimental curves in this region, and the discrepancy between theoretical and experimental critical point energies has been reduced to the order of 0.05 eV.

Using this latest calculation, as well as earlier theory and experiment as a guide, we have analyzed the behavior of the E_o' peaks under uniaxial stress, in order to confirm symmetry assignments and to arrive at the deformation potentials.

II. THEORY

By destroying the tetrahedral symmetry of a zincblende crystal, a uniaxial stress can split degenerate bands and convert the isotropic dielectric function into a tensor, leading to a polarization dependence for normally-incident light. The qualitative effects of stress on the wavelength modulated reflectivity (wmr) spectra can be worked out using group theory, and group theory also provides a framework for defining the deformation potentials. In particular, we can use this approach to arrive at rules for distinguishing the spectra due to Γ, Δ, and X transitions, the principal contributors to the E_o' structure in III-V compounds. Details of the argument for the present case, with graphical illustrations, are given in Ref. 10; we will touch only on the main steps here.

At a critical point away from Γ, two kinds of splittings of symmetry-based degeneracies can be expected: intervalley (between different $\underset{\sim}{k}$) and local (at the same $\underset{\sim}{k}$). (These are often referred to as "interband" and "intraband" splittings, respectively.) For (111) stress all six Δ axes make the same projection (absolute value) on the stress axis, and there is no intervalley splitting between a state at a given $\underset{\sim}{k}$ along Δ and the other five states belonging to the star of $\underset{\sim}{k}$. There may, however, be a local splitting at each of the six $\underset{\sim}{k}$'s, and by symmetry the splitting at each of the $\underset{\sim}{k}$'s in the star will be the same. (All Δ-bands are doubly degenerate at each $\underset{\sim}{k}$ because of time-reversal and two-fold rotational symmetry.) For (001) stress, two Δ axes lie along the stress and four lie perpendicular to it. Thus, of the six degenerate pairs of states associated with $\underset{\sim}{k}$ and its star, four

pairs will split off from the other two; in addition to this inter-valley splitting, the four pairs with $\underset{\sim}{k}$ perpendicular to stress will undergo local splitting, each by the same amount. The two pairs with $\underset{\sim}{k}$ parallel to stress have the same "group of the wave vector" as before stress, and will not undergo local splitting. Thus, where there were once 12 degenerate conduction band levels, (111) stress splits them into two groups of six while (001) stress splits them into three groups of four.

Transitions at X behave in the same manner as Δ transitions except that there is no local splitting because this would violate time-reversal symmetry, a symmetry not destroyed by strain.

At Γ, the four states belonging to a Γ_8 level will split into two pairs, and the doubly-degenerate Γ_7 level will remain unsplit. A minimum degeneracy of two is required at the Γ point, again because of time-reversal symmetry.

In order to understand the actual spectra, we have to combine the above qualitative analysis with a knowledge of the selection rules. For spin-orbit splitting and stress not too large, we can state these for this case approximately [10] as follows: if $\underset{\sim}{E}$ is perpendicular to $\underset{\sim}{k}$, there can be a dipole transition. (Transition probability $\propto 1 - |\hat{E} \cdot \hat{k}|^2$.)

From this analysis we expect the behavior of a peak in the spectrum due to a critical point along Δ to be: (a) for (111) stress, one splitting (local splitting) but no major polarization dependence; (b) for (001) stress, a significant polarization dependence, with the parallel polarization showing one splitting (local), and with the perpendicular polarization being composed of 50% of the parallel spectrum plus a

single unsplit peak representing transitions at $\underset{\sim}{k}$ lying along the stress axis (three peaks in all).

The reader is referred to Ref. [3] for an explanation of the polarization dependence of Γ_7^v, $\Gamma_8^v - \Gamma_7^c$, Γ_8^c transitions in the III-V compounds.

In terms of deformation potentials Ξ_i^j, defined according to the scheme of Kane,[11] the relative energy shifts of Δ transitions as a function of (001) and (111) stress are given by

$$\delta\varepsilon_{\Delta}\,(001) = \frac{1}{\sqrt{3}}(s_{11} + 2s_{12})\sigma\,\Xi_1^1 + \frac{2}{\sqrt{6}}(s_{11} - s_{12})\sigma\,\Xi_1^3$$

$$\delta\varepsilon_{\Delta'}(001) = \frac{1}{\sqrt{3}}(s_{11} + 2s_{12})\sigma\,\Xi_1^1 - \frac{1}{\sqrt{6}}(s_{11} - s_{12})\sigma\,\Xi_1^3 \pm \frac{1}{\sqrt{2}}(s_{11} - s_{12})\sigma\,\Xi_3$$

$$\delta\varepsilon_{\Delta'}(111) = \frac{1}{\sqrt{3}}(s_{11} + 2s_{12})\sigma\,\Xi_1^1 + \frac{s_{44}}{3}\sigma\,\Xi_1^5 \pm \frac{s_{44}}{3}\sigma\,\Xi_4 \qquad (1)$$

where s_{11}, s_{12}, and s_{44} are the elastic compliance constants and σ is the magnitude of the uniaxial stress. Δ and Δ' refer to Δ axes lying parallel to or at an angle to the stress axis, respectively. The expressions for the Ξ_i^j in terms of matrix elements of Bloch functions with the Picus-Bir Hamiltonian are given in Ref. [10].

III. EXPERIMENT

The wavelength modulation spectrometer is that used by Koo, et al. [1] and its operation is described elsewhere. [10,12] Resolution, including modulation amplitude, was 35 Å or 70 meV. A specially-coated quartz plate was used to polarize the light and was good at energies up to around 5 eV, although its transmission characteristic caused a smooth distortion of the spectrum above 4.7 eV. We used the uniaxial stress frame of Koo, et al. [1], with the change that the sample was epoxied between one pair of brass pistons instead of a pair of small stainless steel pistons nested in a pair of larger pistons, in order to minimize misalignment due to residual machining error of the pistons. It was necessary to machine the brass pistons we used to a tolerance of .0003." The preparation and mounting of the sample is described in some detail in another place. [10] The undoped single crystals, obtained from Monsanto, were cut so that the stress was applied along either the (111) or the (001) axis; light was reflected near-normally from the $(1\bar{1}0)$ surface in both cases. Sample dimensions were 2 × 2 × 14mm and 2 × 2 × 9mm respectively. After mechanically polishing on all six sides, they were etched in HNO_3: HF: H_2O (3:1:4) for 90 to 120 sec [13] and glued into the stress frame pistons.

All measurements were made with increasing stress, so as to minimize any effects of plastic (irreversible) deformation. This hysteresis did in fact occur. At the beginning of the run on each sample, observed polarization dependence was negligible, as we would expect for a cubic crystal under near-normal incidence; upon completing the experiment and returning to zero stress, there was a sizable

polarization dependence left in the spectra, about equal to what was produced originally by 2 kbar of stress. Probably the mechanism for this residual polarization dependence includes the formation of dislocations under stress which remain after stress. These could affect the optical spectrum in two ways: (a) through the bulk strain field (or residual stress) associated with the dislocations, and (b) through the formation of surface states where the dislocations terminate at the surface. In this latter case, if these surface states are able to trap holes or electrons and produce a field in the surface layer, there will be an effect on the reflectance. Although the field from these surface charges is normal to the surface, its effect on the optical properties is polarization dependent for all surfaces other than (001) or (111). [14] Since both our samples were measured with light incident on the (110) face in order to get a complete set of data to evaluate deformation potentials, [5] this built in field at the surface might well explain the residual polarization we observed. In light of this, it may be useful to supplement future experiments with data from the faces having isotropic electroreflectance. This surface barrier electroreflectance is the phenomenon exploited in "rotoreflectance" experiments.[14]

IV. RESULTS

The data, taken at stress intervals of approximately 1 kbar (10^9 dynes cm^{-2}), for light polarized parallel (———) and perpendicular (-----) to stress, are shown in Figs. 1 and 2. Two main downward peaks, at 4.5 and 4.7 eV, as well as the double hump at 4.3 - 4.4 eV and the small shoulder at 4.6 eV, characterize the non-stressed spectra (both polarizations). As stress is applied, new peaks appear in both spectra, lineshapes are distorted, and polarization dependence appears. The zero-stress features of the spectra show up clearly in previous wavelength modulation data taken in this laboratory, [8] with the exception of the shoulder at 4.6 eV which we were able to see in the present experiment simply by using higher gain. In another wavelength modulation laboratory, only the two main downward peaks could be seen.[15]

We will discuss the behavior of the two main downward peaks under stress and leave the smaller structures around them for later. Because of the usual interpretation of these two as a spin-orbit-split pair, we expect their behavior to be similar and we will look first at the lower energy one (4.5 eV) because it is larger and more distinct from the rather confusing background of smaller structures.

Briefly, with (111) stress the 4.5 eV dip does not change much in energy or shape, but it increases in magnitude, the parallel polarization a little more so than the perpendicular. In the (001) spectra, however, there is a drastic change in the downward peak. For parallel polarization, it decreases in magnitude and broadens as if it were preparing to split; the perpendicular polarization remains apparently unchanged in height and shape.

The behavior of the 4.7 eV downward peak is consistent with that of its larger partner, but it is harder to interpret because of the jumble of stress-induced peaks occurring in this region.

V. DISCUSSION OF RESULTS

It will be seen from the data that the main downward peak at 4.5 eV follows the expected behavior for Δ transitions as described in the theoretical section. Under (111) stress, this peak retains approximately the same shape for both polarizations, both peaks broadening at a rate of 1.1 ± 0.6 meV kbar^{-1} and both shifting at an average rate of -2.6 ± 1.5 meV kbar^{-1}. (There is some decrease in the magnitude of the peak in the perpendicular spectrum, probably due to a shoulder moving in from the left from 4.4 eV, a shoulder presumed to be Γ transitions.) In the (001) spectra with parallel polarization, which broaden and deform as if they were going to split into two equal peaks half the size of the original, the rate of broadening is 4.3 ± 2.0 meV kbar^{-1}, and the average shift of these two peaks is 0.75 ± 0.5 meV kbar^{-1}. The perpendicular spectra at various stress can be thought of as the sum of two equal peaks each one-fourth the size of the original, representing the Δ' transitions as in the parallel spectra, and a single unsplit peak one-half the size of the original, representing the Δ transitions which do not appear in the parallel spectra. The intervalley splitting is smaller than the local splitting, and so in the perpendicular spectra the Δ peak remains approximately between the two Δ' peaks, the three peaks adding up to produce a deeper, sharper peak

than that in the parallel spectra. The intervalley splitting, as estimated from the shift of the perpendicular peak, is 1.1 ± 0.8 meV $kbar^{-1}$.

We can check these assertions by a graphical analysis. For example, two copies of the zero stress spectrum, each having half the magnitude of the original can be made on tracing paper. These are then graphically added together with a given energy shift between them. With the proper energy shifts, the lineshapes of the parallel (001) spectra should be obtained. Similarly, one can try to reproduce the 25% - 50% - 25% split of the perpendicular spectra.

When this was done, we found good agreement with the overall shapes of the main peak, but for both stresses and polarizations a gradual increase in the magnitude of the structure with increasing stress had to be invoked. This increase could be interpreted as a transition probability (matrix element) increase or as an increase in the joint density of states near the critical point. The interference of other critical points of non-Δ symmetry can also affect the shape of the spectrum.

In terms of the relative energy shifts of the levels,

$$\frac{\delta\varepsilon^{+}_{\Delta},(111) - \delta\varepsilon^{-}_{\Delta},(111)}{\sigma} = 1.1 \pm 0.6 \text{ meV kbar}^{-1}$$

$$\frac{\delta\varepsilon^{+}_{\Delta},(111) + \delta\varepsilon^{-}_{\Delta},(111)}{2\sigma} = 2.6 \pm 1.5$$

$$\frac{\delta\varepsilon^{+}_{\Delta},(001) - \delta\varepsilon^{-}_{\Delta},(001)}{\sigma} = 4.3 \pm 2.0$$

$$\frac{\Delta\varepsilon^{+}_{\Delta},(001) + \delta\varepsilon^{-}_{\Delta},(001)}{2\sigma} \equiv \frac{\overline{\delta\varepsilon_{\Delta},(001)}}{\sigma} = -0.75 \pm 0.50$$

$$\frac{\delta\varepsilon_{\Delta}(001) - \overline{\delta\varepsilon_{\Delta},(001)}}{\sigma} = -1.1 \pm 0.8 \qquad (2)$$

where the values refer to tensile stress. From these we obtain values for the deformation potentials,

$$\begin{aligned} {}^{1}_{1} &= -0.25 \pm 0.13 \text{ eV} \\ {}^{3}_{1} &= -0.07 \pm 0.05 \\ {}^{5}_{1} &= 0.7 \pm 0.3 \\ |{}_{3}| &= 0.5 \pm 0.2 \\ |{}_{4}| &= 0.09 \pm 0.05 \end{aligned} \tag{3}$$

If the main 4.5 eV downward peak were the result of Γ symmetry, it would show signs of splitting for both directions of stress. If X transitions were involved, we would not expect to find the evidence of local splitting that we found, as discussed in the theory section. Λ and Σ transitions, not expected to contribute here, would also give results contradictory to the observed behavior. We conclude that our stress spectra give strong support to the postulate that Δ transitions dominate in this region.

While supporting the assignment of Δ symmetry to the two downward peaks at 4.5 and 4.7 eV, our spectra of deformed GaAs also reveal the presence of transitions from other parts of the Brillouin zone, interspersed among the Δ transitions. In particular, the behavior of the shoulder at 4.43 eV and of a peak at 4.63 eV is not characteristic of Δ symmetry. From electroreflectance data [6] and theoretical results, [9] we expect these two structures to be the $\Gamma_8^v - \Gamma_7^c$ and $\Gamma_8^v - \Gamma_8^c$ transitions.

The behavior of these structures under stress can be inferred from the data of Sell and Kane [3] for Ge and from the observed stress splitting of the Γ_8^v level in GaAs. [16] A careful study of the spectra indicates that the direction and approximate magnitude of the shifts agreed with these authors. (Another small structure, not shown, was observed at 4.95 eV, probably resulting from the third Γ transition, $\Gamma_7^v - \Gamma_8^c$.) However, it is difficult to conclude from our spectra alone that this is the correct interpretation, because of the small magnitude of these structures.

Consideration of the available theoretical and experimental information on the E_o' region leads us also to believe that the hump at 4.32 eV in our spectra is caused by the onset of Δ transitions at the M_o point near the pseudocrossing, its spin-orbit split partner contributing to the rapid rise of the spectrum from 4.51 to 4.57 eV.

CONCLUSION

The low temperature wavelength modulation spectra of the E_o' region of GaAs have been measured, under applied uniaxial stresses in the (111) and (001) directions, revealing the composite nature of the structure and providing evidence that spin-orbit-split Δ transitions are responsible for its two main features. Comparison of the results with predictions of band structure calculations supports this view. It is hoped that this stress data will shed a little more light on the lingering controversy as to the detailed origin of the critical points and spectral structure in this region.

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FIGURE CAPTIONS

Fig. 1. Derivative reflectivity at 5°K of the E_o' peaks of GaAs, for various levels of (111) uniaxial stress. The solid curve is (1/R)dR/dE for light polarized parallel to stress; the dashed curve is for light polarized perpendicular to stress.

Fig. 2. Derivative reflectivity at 5°K of the E_o' peaks of GaAs, for various levels of (001) uniaxial stress. The solid curve is (1/R)dR/dE for light polarized parallel to stress; the dashed curve is for light polarized perpendicular to stress.

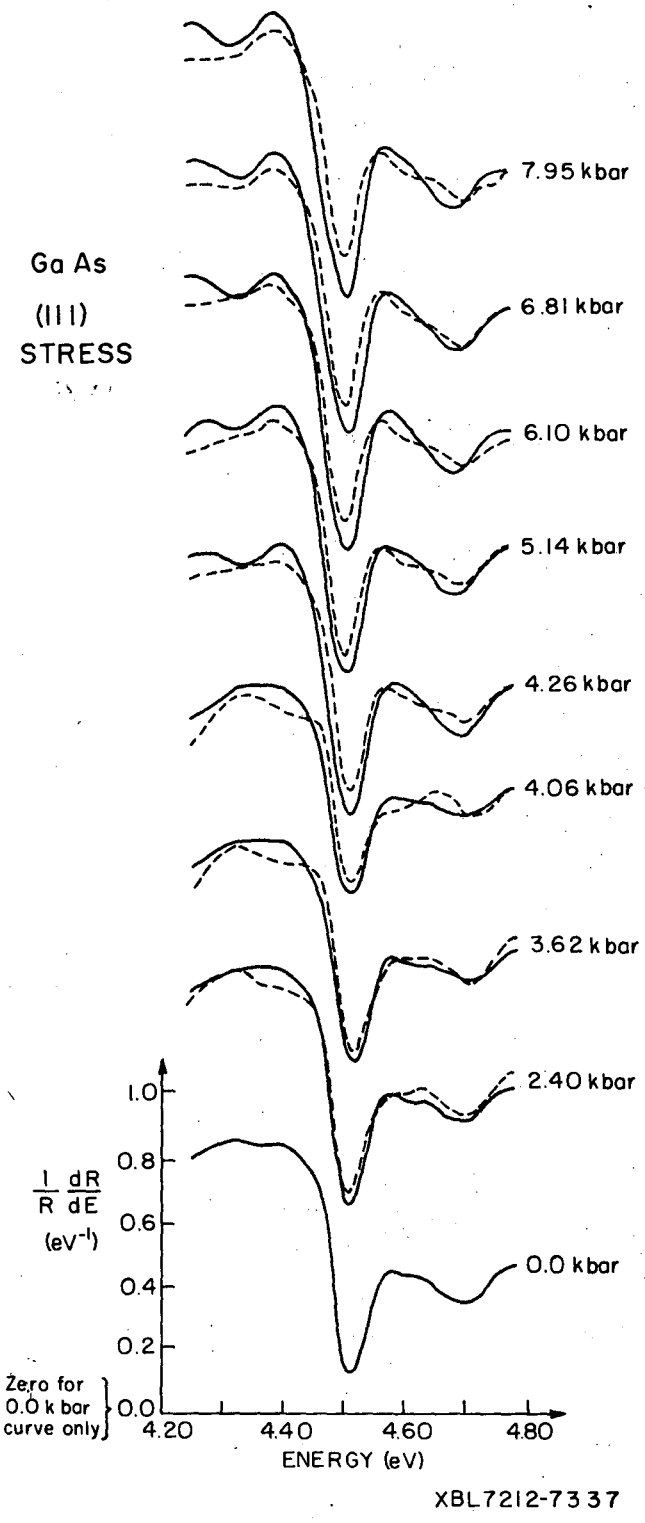
Ga As
(111)
STRESS
7.95 kbar
6.81 kbar
6.10 kbar
5.14 kbar
4.26 kbar
4.06 kbar
3.62 kbar
2.40 kbar
0.0 kbar
1/R dR/dE (eV⁻¹)
1.0
0.8
0.6
0.4
0.2
0.0
Zero for 0.0 k bar curve only
4.20
4.40
4.60
4.80
ENERGY (eV)
XBL7212-7337

Fig. 1

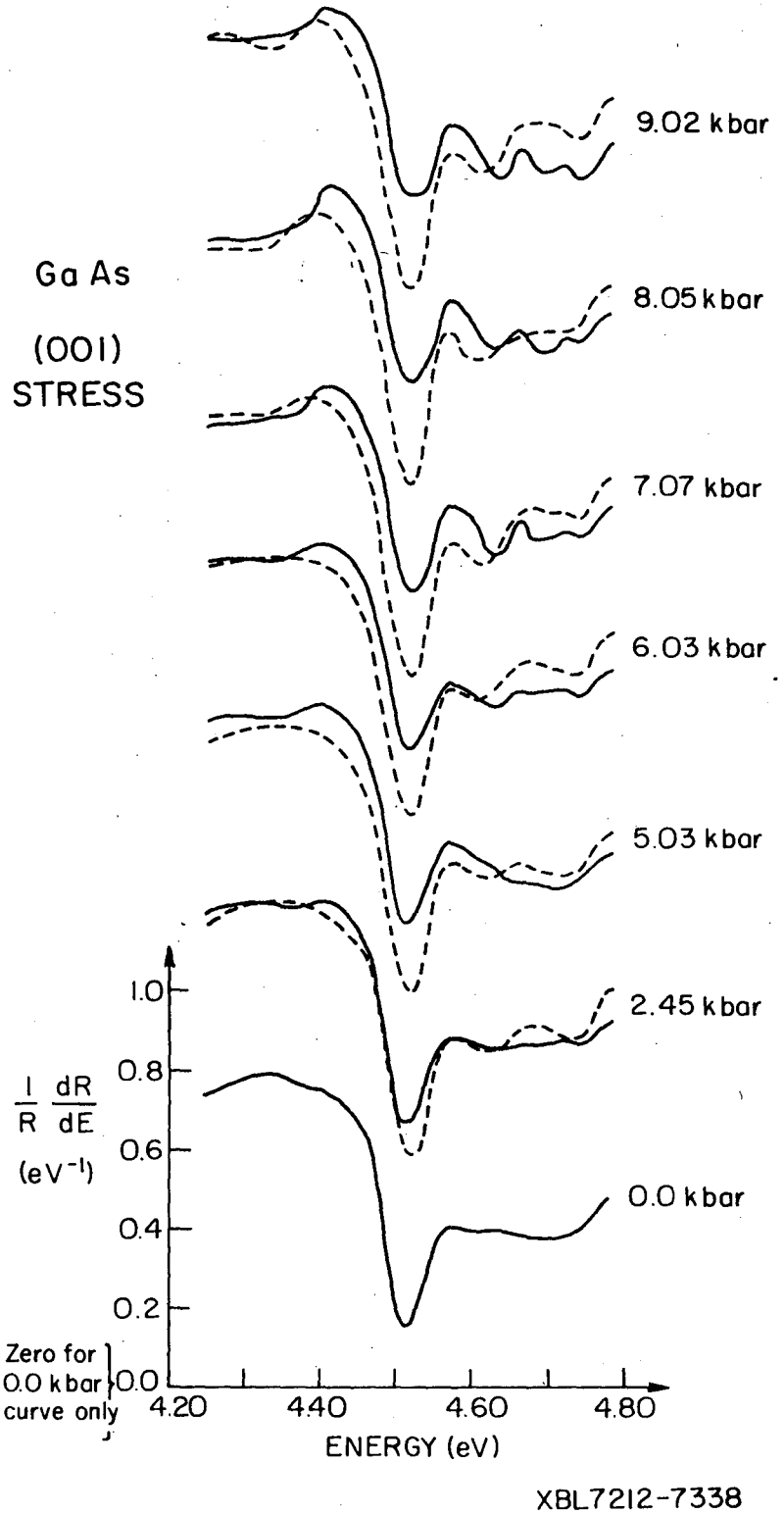

Fig. 2

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