

# Effect of reconstruction-induced strain on the reflectance difference spectroscopy of GaAs (001) around $E_1$ and $E_1 + \Delta_1$ transitions

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We report measurements of reflectance difference (RD) spectra for  $c(4 \times 4)$  and  $(2 \times 4)$  reconstructed (001) surfaces of GaAs. Surface dimers induce an inhomogeneous orthorhombic strain field that penetrates several monolayers into the crystal. The thickness  $d$  of this strained region is smaller than the penetration depth  $L$  of the probing light, and an inhomogeneous anisotropy formalism can be used. The dielectric function of GaAs at room temperature can be described by a discrete (Lorentzian) excitonic line shape. For the contribution of the region near the surface to the optical response, the excitonic response should be at least partly quenched because of the electric field present near the surface. Thus, a one-electron contribution (logarithmic line shape) may have to be used to fit the RD spectra. By using an inhomogeneous perturbation formalism and a logarithmic line shape, we have been able to isolate the strain component induced by the dimers around  $E_1$  and  $E_1 + \Delta_1$  critical points. We also inferred that the RD spectra include a component related to surface roughness.

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## I. INTRODUCTION

Reflectance difference spectroscopy (RDS) is emerging as a powerful technique to characterize the surface of diamond<sup>1</sup> and zinc-blende-type semiconductors.<sup>2-6</sup> However, understanding RD spectra is a difficult task due to the multiplicity of physical mechanisms that contribute to optical surface anisotropy. RDS line shapes of GaAs (001) surfaces in ultra-high vacuum (UHV) are closely related to surface reconstruction, as well as to surface morphology.<sup>7-12</sup> RDS measurements of the (001) surface of GaAs crystals have shown that the relaxation of the position of surface atoms induces an anisotropic strain which penetrates several atomic layers into the bulk, leading to optical surface anisotropy.<sup>8,13-21</sup>

In the energy region around the  $E_1$  and  $E_1 + \Delta_1$  transitions, the RD spectra of  $c(4 \times 4)$  and  $(2 \times 4)$  GaAs surfaces are predominantly determined by the reconstruction-induced component of the strain. Although this strain has the same orthorhombic symmetry as that resulting from the application of a uniaxial external stress along a [110] crystal direction, we expect substantial differences in RD line shapes between surface reconstruction and external stress perturbations. We reach this conclusion on the basis of the following arguments:

(1) Reconstruction-induced strain penetrates into a region of thickness  $d$ , a depth of a few monolayers. In the energy region around  $E_1$  and  $E_1 + \Delta_1$ , the light penetration depth  $L$  is larger than  $d$  and the perturbation strain is thus inhomogeneous. In contrast, for an externally applied stress,  $d \gg L$  and the perturbation induced by the strain becomes homogeneous. On the other hand, it is known that in the limit  $d \ll L$  an inhomogeneous perturbation introduces a  $\pi/2$  phase difference that mixes the real and imaginary parts of the

sample anisotropic response, with the consequent change in RD line shape.<sup>22,23</sup>

(2) The line shape of the GaAs dielectric function at room temperature is dominated by discrete excitons rather than by one-electron two-dimensional (2D) band-to-band transitions.<sup>24,25</sup> Consequently, the RD line shape of GaAs crystals under an external stress perturbation has been found to be mainly excitonic.<sup>3</sup> In reconstructed surfaces, however, the piezoelectric field, induced by the surface strain, could ionize the excitons, thus quenching its contribution to the RD spectrum and rendering the 2D contribution relatively more important.

In the present paper, we report RD measurements carried out in order to explore the two points mentioned above. RD spectra for GaAs (001) with  $c(4 \times 4)$  and  $(2 \times 4)$  surface reconstructions were measured in the energy range from 2.4 to 3.6 eV, bracketing the  $E_1$  and  $E_1 + \Delta_1$  transitions. By following the RD spectrum evolution as reconstruction changes gradually from  $c(4 \times 4)$  to  $(2 \times 4)$ , the contribution of the surface strain to the line shape was identified. We found that this contribution could be fitted with a 2D inhomogeneous-strain model, as expected.

## II. EXPERIMENTAL SETUP

RDS and spectroscopic ellipsometry (SE) measurements were carried out in UHV on GaAs (001) crystal surfaces. RD (Ref. 26) and SE (Ref. 27) spectrometers were attached to the UHV chamber through a strain-free optical viewport. The spectra were obtained in the energy range from 2.4 to 3.6 eV. A 75 W short-arc Xe lamp was employed as the light source and an end-on GaAs (multialkali photocathode) photomultiplier tube as the photodetector.

The sample studied was a 0.5- $\mu\text{m}$ -thick homoepitaxial GaAs layer, not intentionally doped, grown by molecular-beam epitaxy (MBE) on a heavily  $n$ -doped (001) substrate. The sample surface was protected with an  $\text{As}_2$  film in order to be transferred uncontaminated from the growth chamber to the UHV chamber.<sup>28</sup> Once inside the UHV chamber, a  $c(4 \times 4)$  surface reconstruction was obtained after thermal desorption of the capping layer at  $\sim 640$  K. By subsequent thermal treatments at increasing temperatures, the surface was gradually modified until a  $\beta(2 \times 4)$  reconstruction was reached at  $\sim 760$  K. The surface reconstruction was confirmed by reflection high-energy electron diffraction.<sup>7</sup> The base pressure of the chamber was kept below  $5 \times 10^{-10}$  Torr during the experiment. RD and SE spectra were measured at both room temperature and at 150 K.

### III. THEORY

Let us consider a GaAs crystal with a (001) surface. The RD line shape for light impinging perpendicularly on this surface may be written as<sup>29</sup>

$$\frac{\Delta R}{R} = \text{Re} \left[ \frac{2}{\sqrt{\varepsilon(\varepsilon - 1)}} \Delta\varepsilon \right], \quad (1)$$

where  $\varepsilon$  is the dielectric function of the unperturbed crystal and  $\Delta\varepsilon$  is the anisotropy of the dielectric function defined as  $\Delta\varepsilon = \varepsilon_{[110]} - \varepsilon_{[1\bar{1}0]}$ , where  $\varepsilon_{[110]}$  ( $\varepsilon_{[1\bar{1}0]}$ ) represents the dielectric response for  $[110]$  ( $[1\bar{1}0]$ ) light polarization. The energy dependence of  $\Delta\varepsilon$  and of the RD line shape is determined by the nature of the perturbation that renders the crystal anisotropic.  $\Delta\varepsilon$  also depends on the symmetry of the critical point being considered. In this paper, we will limit our discussion to  $E_1$  and  $E_1 + \Delta_1$  transitions, known to take place along the eight equivalent  $\Lambda$  axes  $\langle 111 \rangle$ .

Two regimes will be considered,  $d \gg L$  and  $d \ll L$ , which respectively correspond, as discussed above, to homogeneous and inhomogeneous perturbations. We will assume that in the first case the perturbation is due to an externally applied stress, while in the second case it is related to surface reconstruction.

#### A. $d \gg L$ regime

Consider a uniaxial stress applied along  $[110]$ . This stress induces an orthorhombic bulk strain with nonzero components given by<sup>30</sup>  $e_{xx} = e_{yy} = (S_{11} + S_{12})X/2$ ,  $e_{zz} = S_{12}X$ , and  $e_{xy} = e_{yx} = S_{44}X/4$ , where  $X$  is the magnitude of the stress and  $S_{ij}$  are the elastic compliance moduli. The change in dielectric functions for a single critical point of  $\Lambda$  symmetry may be written as<sup>3,31</sup>

$$\begin{aligned} \Delta\varepsilon' &= \frac{1}{E^2} \frac{\partial [E^2 \varepsilon'(E, E_1 + \delta E_{so} + \delta E'_h)]}{\partial E} \delta E'_s \\ &\pm \frac{4r\gamma'}{\Delta_1} \varepsilon'(E, E_1 + \delta E_{so} + \delta E'_h), \end{aligned} \quad (2)$$

where  $\varepsilon'$  stands for the contribution of either the  $E_1$  or the  $E_1 + \Delta_1$  critical points to the overall dielectric function  $\varepsilon$ , +

(−) refers to  $E_1$  ( $E_1 + \Delta_1$ ),  $\Delta_1$  is the spin-orbit splitting, and the coefficients  $\gamma'$  and  $\delta E'_s$  are proportional to  $e_{xy}$  and depend on the corresponding band deformation potentials<sup>30,32–34</sup> and the elastic compliance moduli.<sup>32</sup> The overall change  $\Delta\varepsilon$  is obtained by superposing the  $E_1$  and  $E_1 + \Delta_1$  contributions.

To model  $\Delta\varepsilon'$ , the  $\varepsilon'$  in Eq. (2) is obtained from a fit to the experimental dielectric function using the discrete excitonic line shape

$$\varepsilon' = A e^{i\theta} (E - E_g + i\Gamma)^{-1}, \quad (3)$$

with a phase angle  $\theta$  close to zero. The phase angle  $\theta$  is usually interpreted as a coupling parameter between the discrete exciton interaction with an overlapping continuum of interband transitions.<sup>25</sup>

#### B. $d \ll L$ regime

Let us consider now the surface strain induced by reconstruction. Under such strain the surface possesses only two perpendicular symmetry planes (orthorhombic symmetry) and the changes  $\Delta\varepsilon'$  are also given by an expression similar to Eq. (2). However, the thickness  $d$  of the anisotropic region induced by the surface strain is lower than the penetration depth  $L$  of the probing light around the  $E_1$  transition.  $\Delta\varepsilon(z)'$  now becomes dependent on the depth along  $z$ , perpendicular to the surface, and the total change  $\Delta\varepsilon'$  is obtained by averaging  $\Delta\varepsilon(z)'$  from  $z=0$  (surface) to  $z=d$ . The total change can be obtained by setting

$$\Delta\varepsilon = -2ikd \overline{\Delta\varepsilon} \quad (4)$$

in Eq. (1),<sup>22,23</sup> where  $\overline{\Delta\varepsilon}$  is the averaged change in dielectric function over the perturbed region and  $k$  the wave number of the probing light. The factor  $i$  in Eq. (4) interchanges the real and imaginary parts of the anisotropy response function.

As we have mentioned, the surface electric field induced by the strain may partially ionize the excitons in a region of thickness  $d$  below the surface. For fully ionized excitons, the surface dielectric function is described by the 2D line shape given by

$$\varepsilon' = A e^{i\theta} \ln(E - E_g + i\Gamma). \quad (5)$$

The phase angle  $\theta$  in Eq. (5) describes the metamorphism of 2D critical-point line shapes due to excitonic effects;<sup>25</sup> an angle  $\theta=0$  corresponds to one-electron transitions. The excitonic metamorphism increases the value of  $\theta$ .<sup>25</sup> The RD line shape for the  $d \ll L$  regime is thus calculated from Eqs. (1), (2), and (4), modeling the  $\varepsilon'$  spectrum in Eq. (2) with the 2D line shape of Eq. (5).

### IV. EXPERIMENTAL RESULTS AND DISCUSSION

Figures 1(a)–1(f) show the evolution of the RD spectrum in the range of the  $E_1$  and  $E_1 + \Delta_1$  transitions, as reconstruction gradually changes from  $c(4 \times 4)$  to  $(2 \times 4)$  by increasing annealing temperature. The change in reflectivity is defined as  $\Delta R/R = (R_{[110]} - R_{[1\bar{1}0]})/R$ . Starting with a  $c(4 \times 4)$  reconstruction [spectrum (a)], we observe the spectrum amplitude

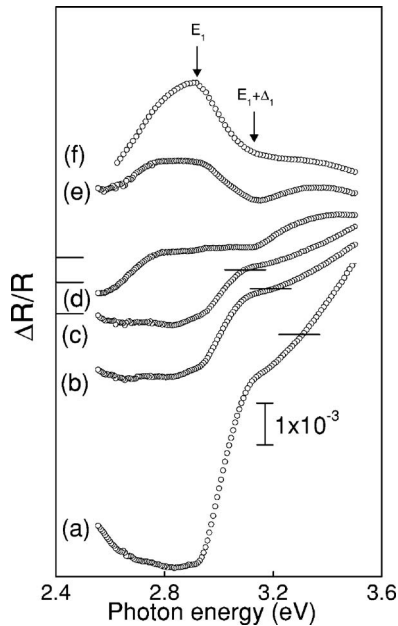


FIG. 1. Sequence of RD spectra of the GaAs(001) surface reconstructions obtained by thermal desorption. (a)–(c) correspond to  $c(4 \times 4)$ , (d) seems to correspond to an average zero strain, and (e)–(f) to  $(2 \times 4)$ . The energies of the  $\Gamma$  critical points were taken from the literature (Ref. 38). The annealing temperatures were (a) 370 °C, (b) 390 °C, (c) 400 °C, (d) 410 °C, (e) 440 °C, and (f) 490 °C.

to first decrease monotonically [Figs. 1(a)–1(c)], becoming almost quenched in Figs. 1(d) and 1(e). Thus, the condition of average zero strain<sup>35–37</sup> seems to lie between curves (d) and (e). Beyond this point, the amplitude increases monotonically with a sign opposite to that of the initial spectrum [Figs. 1(e) and 1(f)]. We note that in the energy range from 2.55 to 2.8 eV, we observe an additional weak structure superimposed on a smooth background whose slope changes sign as the transition takes place. This structure has been assigned to transitions between occupied bulk valence-band states and empty surface states.<sup>6,12,39</sup> In the energy range from 2.8 to 3.6 eV, RD structures for  $c(4 \times 4)$  and  $(2 \times 4)$  have been attributed to transitions between bulk and bulk-modified-by-the-surface electronic states.<sup>6,39,40</sup> No contributions of surface states are considered in the present discussion.

Figure 2(a) shows the RD spectrum of Fig. 1(a) (hollow circles) along with the corresponding fit (solid line) performed on the basis of an inhomogeneous perturbation ( $d \ll L$ ). As discussed in the previous section, in order to fit the RD line shape we first modeled the experimental GaAs dielectric function with the 2D line shape of Eq. (5). The dielectric function fit was optimized with a phase angle  $\theta = 70^\circ$ , rather close to that employed in Ref. 25. The RD line shape was then calculated by means of Eqs. (1), (2), (4), and (5), using the parameters obtained from the dielectric function fit, with the exception of the phase angle  $\theta$  which was regarded as a fitting parameter. For the  $E_1$  and  $E_1 + \Delta_1$  components of the fit shown in Fig. 2(a), we employed  $\theta$  values equal to zero. This zero  $\theta$  values are indicative of exciton

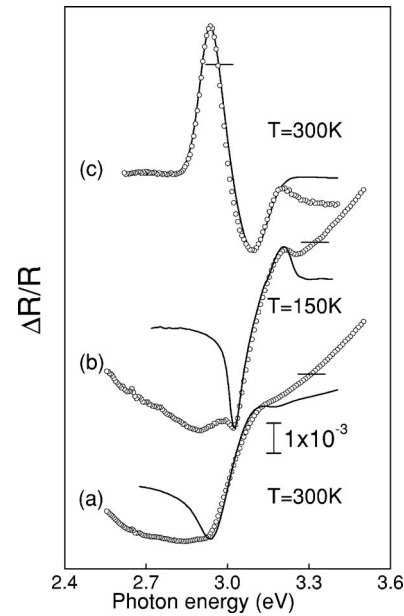


FIG. 2. Homogeneous and inhomogeneous strain fields. (a) Spectrum (a) of Fig. 1 (hollow circles) and its theoretical line shape (solid lines) obtained by using Eq. (2) and 2D line shapes. (b) Spectrum (a) of Fig. 1 at  $T = 150$  K and the corresponding line shape obtained by using the same approach as for spectrum (a). (c) RD spectra taken from literature (Ref. 3) for  $[1\bar{1}0]$  uniaxial applied stress (hollow circles) and its theoretical model (solid lines) obtained by using Eq. (2) and excitonic line shapes.

quenching near the surface due to the surface electric field.

The RD (experimental) spectrum represented by the hollow circles in Fig. 2(b) corresponds to the same sample as the spectrum of Fig. 2(a) but measured at a lower temperature ( $T = 150$  K). The small structure around 2.95 eV has been reported to have bulk and surface origins.<sup>41</sup> The solid line in Fig. 2(b) displays the best RD line shape fit, obtained following the same procedure outlined in the paragraph above. In this case, the GaAs dielectric function was fitted with a 2D critical-point model, with  $\theta = 85^\circ$ . The increment in  $\theta$  with respect to the value obtained at room temperature indicates that excitonic effects become more important at lower temperatures, as expected.<sup>25</sup> It is important to note that although bulk exciton effects increase when lowering the temperature, as indicated by the increase in  $\theta$  required for the dielectric function fit, the phase angle  $\theta$  used to fit the RD line shape is relatively independent of temperature. We may expect this result by considering that the mechanism that ionizes excitons near the surface is temperature independent.

For the sake of comparison, we show as hollow circles in Fig. 2(c) the RD spectrum reported in the literature for a homogeneous perturbation ( $d \gg L$ ), together with the corresponding fit.<sup>3</sup> This fit was obtained by using an excitonic line shape for the dielectric function with a phase angle  $\theta$  approximately equal to zero, in agreement with similar fits reported in the literature.<sup>25</sup>

The binding energy  $E_B$  for the excitons related to  $E_1$  and  $E_1 + \Delta_1$  transitions should be between 0.05 and 0.1 eV, and the exciton radius about  $1 \times 10^{-7}$  cm.<sup>42</sup> The ionization field given by  $E_{\text{ion}} = E_B / ea_0$  has a minimum value

$E_{ion} \approx 5 \times 10^5$  V/cm. On the other hand, the order of magnitude of the electric field near the surface can be estimated by the fit of Fig. 2(a). From the fit, we obtain a value of  $e_{xy}d \approx 7.0 \times 10^{-3}$  nm. For a region of thickness  $d=1.0-0.5$  nm, the surface strain gives  $e_{xy} \approx (7.0-14.0) \times 10^{-3}$ . This value is of the same order of magnitude as the reported surface strain calculated for the first five monolayers just below the dimer for Si and Ge.<sup>13</sup> The corresponding piezoelectric field has a value  $F=e_{xy}\sqrt{3}e_{14}/\epsilon_0\epsilon_v$ . By using  $e_{14}=0.21$  C/m<sup>2</sup>,<sup>43</sup>  $\epsilon_0=8.85 \times 10^{-12}$  F/m, and  $\epsilon_v=12.5$ , we estimate  $F \approx (2.25-4.5) \times 10^5$  V/cm, a very close value to the estimated ionization field. Thus, the excitons should be at least partially ionized near the surface.

The strain induced by dimers on GaAs (001) surfaces has been measured by using photoreflectance difference (PR-D).<sup>20</sup> In this case, the PR signal has its origin in a region of thickness  $L \approx 20$  nm because the surface electric field is modulated in the whole region of the penetration depth of the light. Thus, the value of  $e_{xy}^s$  reported by using PR-D corresponds to the mean value averaged in the region of thickness  $L$ . On the other hand, the strain  $e_{xy}$  determined by RDS is the mean value averaged in the region of thickness  $d$ , and it is expected to be larger than  $e_{xy}^s$ . We also mention that the surface electric field induced by the space charge that plays an important role in PR-D is not important in RD. The samples used in this work were undoped MBE layers ( $10^{15}-10^{16}$  cm<sup>-3</sup>). For this carrier concentration, the electric field should be  $F \approx (1.0-5.0) \times 10^4$  V/cm. The converse piezoelectric effect  $e_{xy}=d_{14}F$  gives a strain of  $e_{xy}=(2.7-13.0) \times 10^{-6}$ . This strain is 3 orders of magnitude lower than the strain induced by the dimers and its influence on the RD spectra was neglected.

In order to improve the RD line shape, we have included surface roughness effects. The RD line shape for surface roughness has been reported to be proportional to  $1/\epsilon$ .<sup>44</sup> This spectral component, together with that of surface strain, is shown in the inset of Fig. 3 for  $T=150$  K. It can be seen that roughness contribute a smoother component with a positive slope, in contrasts to the richer optical structure related to the strain. Figure 3(b) shows the fit obtained by the superposition of the strain and roughness components. Figure 3(a) shows the RD spectrum at room temperature and the fit obtained by superposing the strain and roughness components. The improvement of the fit with the inclusion of the roughness component is evident for energies both above and below the  $E_1$  and  $E_1+\Delta_1$  transitions.

Additional evidence of the 2D nature of the RD spectra associated with surface reconstruction is obtained as follows. First, we note that the RD discrete exciton line shape [Eq. (3)] corresponds to the energy derivative of the RD 2D line shape [Eq. (5)]. Moreover, according to Eq. (4), an inhomogeneous perturbation exchanges the real and imaginary parts of the anisotropy response, and we may model the first energy-derivative of the 2D RD spectrum by performing a Kramers-Kronig analysis<sup>45</sup> of the spectrum of Fig. 2(c).

We show with hollow circles in Fig. 4 the energy derivative of the spectrum obtained by taking the difference of spectra (b) and (c) of Fig. 1. We note that this difference

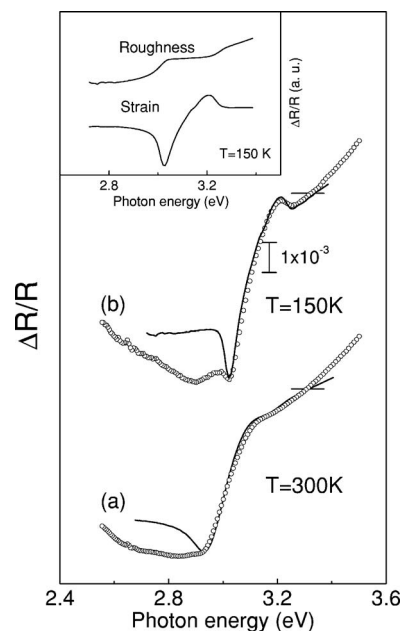


FIG. 3. RD spectra of Fig. 1 and its theoretical modeling (solid lines) obtained by adding the strain and surface roughness components. (a) Taken at  $T=300$  K and (b) taken at  $T=150$  K. The inset displays the components used to fit spectrum (b).

spectrum minimizes the surface roughness RD component. We may thus expect the difference spectrum to consist only of the surface reconstruction component. The solid line in Fig. 4 was obtained from a Kramers-Kronig analysis of Fig. 2(c); we note the excellent agreement between the model and the experimental data. In order to show that the experimental

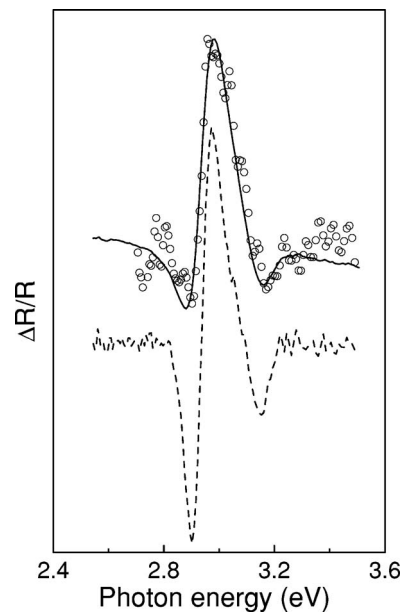


FIG. 4. Hollow circles show the first derivative with respect to photon energy of the spectrum obtained by taking the numerical difference between spectra (b) and (c) of Fig. 1. The solid line represents the Kramers-Kronig analysis of the spectrum in Fig. 2(c) (see text). The dashed line corresponds to the first energy derivative of Fig. 2(c).

spectrum cannot be fitted well by the discrete exciton line shape, we plot as a dashed line in Fig. 4 the energy derivative of the spectrum of Fig. 2(c). Note that the dashed line spectrum is narrower than the experimental one and results in a poor fit at lower energies. We thus conclude that the RD spectrum induced by surface reconstruction corresponds to 2D transitions.

We have been able, with the procedure just described in this paper, to fit the RD spectra measured for GaAs around the  $E_1$  and  $E_1 + \Delta_1$  structures reported in the literature. The results are acceptable for the surfaces prepared with the same arsenic capping and desorption method used in the paper.<sup>9-11</sup> For the MBE prepared surfaces,<sup>6,40</sup> these fits are not perfect because true surface state components must be taken into account.<sup>7,40</sup>

## V. CONCLUSIONS

We have measured RD spectra of GaAs (001) in the region around the  $E_1$  and  $E_1 + \Delta_1$  transitions for different surface reconstructions. By analyzing the evolution of these spectra with the surface geometry, we have isolated the RD component associated with the surface strain induced by the reconstruction. In order to fit the measured RD spectra, we

have used the line shape for the bulk strain component while considering that near the surface the strain is inhomogeneous along the direction perpendicular to the surface and that the strain-induced electric field ionizes the excitons (at least in part), thus quenching the excitonic effect that is present in the bulk of semiconductor samples. We have also included in our model a component induced by surface roughness. The resulting fits are in rather good agreement with the experimental data.

We believe that the surface strain sensitivity accomplished with the model presented in this work renders RD spectroscopy as a useful optical probe that can be further extended to more complicated systems, such as the *in situ* detection of quantum dot formation and the fabrication of highly strained heteroepitaxial interfaces, among others.

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