Ellipsometric characterization of random and random-dimer GaAs-Al$_x$Ga$_{1-x}$As superlattices

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We studied the optical properties of ordered and intentionally disordered GaAs-Al$_x$Ga$_{1-x}$As superlattices, with and without dimer-type correlations in the disorder, by means of spectroscopic ellipsometry. The electronic structure of the superlattices has been calculated and compared with the experiments. The optical transitions in ordered, correlated, and uncorrelated disordered superlattices show specific features that we relate to their different electronic structures.

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Latest advances in nanotechnology make it possible to grow artificial semiconductor superlattices (SLs) with tailored physical properties. This feature opened the possibility to verify experimentally former theoretical predictions, such as Stark-Wannier ladders, Bloch oscillations, Anderson localization by uncorrelated disorder, and electron delocalization by correlations in the disorder. Concerning this last finding, photoluminescence (PL) and electron-transport experiments in intentionally disordered SLs showed that spatial correlations of the disorder lead to electron delocalization of states in low-dimensional systems, as previous theoretical calculations suggested, in contrast to the earlier belief that all eigenstates might be localized. This is a validation of basic physical phenomena, i.e., the inhibition of partial correlations of the disorder lead to electron delocalization of states in low-dimensional systems, as previous theoretical calculations suggested, in contrast to the earlier belief that all eigenstates might be localized. This is a validation of basic physical phenomena, i.e., the inhibition of Anderson localization in quasi-one-dimensional systems with correlated disorder, predicted theoretically at the beginning of the 1990s.

Spectroscopic ellipsometry (EL) is an optical technique that measures the complex ratio $\rho = \chi_r/\chi_i$ between the polarization states of the reflected and incident waves $\chi_r$ and $\chi_i$, respectively. In the case of ambient-bulk material interfaces described by the two-phase model, this ratio is directly related to the dielectric function of the material. In GaAs-Al$_x$Ga$_{1-x}$As SLs, being multilayered structures, the two-phase model no longer applies. Nevertheless, it is useful and very common to represent the complex reflectance ratio as a pseudodielectric function in a two-phase model since this allows a direct comparison with the data for bulk GaAs and AlAs. This procedure has been successfully used by other authors to study dielectric and optical properties of ordered GaAs-AlAs SLs.

In the present work, we report on EL measurements performed in ordered and intentionally—uncorrelated and correlated—disordered GaAs-Al$_{0.35}$Ga$_{0.65}$As SLs. We present a physical interpretation of the EL spectra in the energy region close to the near-band-edge optical transitions on the basis of their different electronic states. In particular, the EL experiments provide further support of previous theoretical claims that correlated and uncorrelated disordered SLs exhibit a remarkably different electronic structure.

We characterized three undoped SLs grown by molecular beam epitaxy. All the SLs have 200 periods and Al$_{0.35}$Ga$_{0.65}$As barriers 3.2 nm thick. In the ordered SL (OSL) all the 200 wells are identical with thickness 3.2 nm (hereafter referred to as A wells). In the random SL (RSL), 58 A wells are replaced by wells of thickness 2.6 nm (hereafter referred to as B wells) and this replacement is done randomly. The so-called random-dimer SL (DSL) is similar to the RSL with the additional constraint that the B wells appear only in pairs. In the latter sample the disorder exhibits the desired short-range spatial correlations. Every SL is clad on each side by 100 nm of n-Al$_{0.3}$Ga$_{0.7}$As, Si doped to 4 x 10$^{18}$ cm$^{-3}$, with a 50-nm n-GaAs buffer layer (doped to 4 × 10$^{18}$ cm$^{-3}$) on the substrate and a 3-nm n-GaAs cap layer (doped to 6 × 10$^{18}$ cm$^{-3}$). Figure 1 shows the schematic view of the conduction-band-edge profile along the growth direction for the three SLs.

The SLs have been characterized by x-ray diffraction in order to verify their structural parameters and quality. Room

FIG. 1. Schematic diagram of the conduction-band-edge profiles of the three SLs.
temperature EL spectra have been recorded with a rotating-polarizer spectroscopic ellipsometer, with variable incidence angle, in the spectral range between 1.4 and 5.0 eV. The spectra reported in this work were measured with an incidence angle of 75°, i.e., close to the Brewster angle of the Al$_{0.3}$Ga$_{0.7}$As alloy. PL at room temperature have been measured in order to facilitate the interpretation of the EL spectra.

PL spectra at low temperature were presented in a previous paper \cite{4}. The PL spectra were recorded with a Jobin-Yvon Labram system, allowing different laser spot diameters from 1 mm down to 30 μm, power intensities, and at different positions on the samples. The spectra reported in this work have been measured with a low-excitation laser power density (0.3 mW/cm$^2$).

Figure 2 shows the pseudodielectric function of the DSL from 1.4 eV (an energy well below the optical absorption gap of all the layers forming the SL) up to 5.0 eV (an energy for which its absorption coefficient is fairly large). The oscillations between 1.4 and 1.6 eV correspond to interference due to multiple reflections within the SL, which is transparent in this low-energy region. When the absorption coefficient of the SL increases, the amplitude of the oscillation decreases accordingly. The occurrence of these oscillations indicates that the two-phase model does not hold and a three-phase model or, even better, a multilayer model should be used to analyze the complex reflectance ratio. The sharp structure around 1.6 eV is due to near-band-edge optical transition of the SL. For energies around the Al$_{0.3}$Ga$_{0.7}$As energy gap (1.85 eV),\textsuperscript{15} the Al$_{0.3}$Ga$_{0.7}$As thick layers of the samples start to absorb light and the EL spectra of the OSL, RSL, and DSL samples show the same energy dependence. In the energy range 2.8–3.4 eV, the features labeled $E_1$ and $E_1 + \Delta_1$ are clearly observed while those referred to as $E_0'$ and $E_2$ arise in the range 4.5–5.0 eV.\textsuperscript{10,13,14}

Let us now comment on the EL spectra in the region of the near-band-edge optical transition. In the upper panels of Fig. 3 we show the real part of the pseudodielectric function in this energy range. Data represented by solid lines (circles) have been recorded with an energy step of 0.001 eV (0.002 eV). We can see that the solid lines and circles superpose very well, indicating that the measurements are repeatable. Well-resolved transitions, not masked by noise, are detected around 1.6 eV. In the lower panel of Fig. 3 we report on the PL spectra of the same samples, showing that the energy of the radiative transitions match the EL features mentioned above.

In the low-energy region ($\leq$1.55 eV), the EL spectra of the SL’s show oscillations corresponding to multiple reflections. Note that the position and period of the oscillations in the RSL and DSL are identical. This agreement confirms that the thick layers embedding the RSL and DSL samples have exactly the same thickness and also that the SL’s have the same total thickness. The period and position of the oscillations in the OSL is different since this SL is thicker (narrow $B$ wells are absent). The interband optical transitions of the SL’s are only slightly resolved in the $\varepsilon_2$ pseudodielectric function. Therefore, the pseudodielectric function $\varepsilon_1$ is found to be much more sensitive to the interminiband transitions.

In Fig. 4 we show on the same plot the $\varepsilon_1$ spectra of the three SL’s (right axis corresponds to the OSL while the left one corresponds to both random SL’s). For energies around the near band edge. In upper panel, solid lines and circles refer to measurements with 0.001 meV and 0.002 meV energy steps, respectively.
be attributed to the wide A wells while that denoted by hh(B) at 1.64 eV to the narrow B wells. In this SL the carriers are mainly localized due to the intentional uncorrelated disorder and we can detect transitions from both A and B wells separately. These features are separated by 20 meV, in agreement with the calculated energy difference between the ground-state levels of isolated A and B wells, which is found to be 23 meV. The PL spectrum is peaked nearly at the same energy of the hh(A) transition, indicating that the PL is mainly due to recombination from this level. A small shoulder is observed in the PL spectra at 1.64 eV, corresponding to the hh(B) feature.

The EL spectrum of the DSL shows a peak at 1.60 eV, which is redshifted with respect to the transition in the RSL at 1.62 eV. As it has been shown by Fujiwara, the redshift of the near-band-edge optical transition in semiconductor SL’s is due to the formation of a miniband due to coherent tunneling processes between the quantum wells. This redshift is also revealed in the PL spectra at room temperature (see Fig. 3) as well as at 4 K (see Ref. 4), and provides further support to our previous theoretical calculations and experiments about the occurrence of a band of delocalized states in correlated disordered SL’s.4,6

The $\varepsilon_1$ lineshapes of the three SL are clearly different in this region of the fundamental interminiband transition. The lineshape is closely related to the oscillator strength and, consequently, depends on the particular electronic structure of each SL.4–6 The OSL and DSL present band of extended states while in the RSL all the states are localized, as theory6 and experiments showed.4,5 Therefore, the arrangement of electronic levels responsible for the optical transitions depends on the SL considered (OSL, RSL, or DSL).

Zheng and co-workers showed that the reflectance lineshape of an interband optical transition in a low-dimensional heterostructure depends not only on the nature of the transition but also on the thickness of the cap layer (i.e., on a geometrical parameter of the heterostructure).16 Since the EL is a reflectivity-based technique, also its lineshape will depend on the cap layer thickness, and not only on the oscillator strength lineshape of the transition. Therefore any realistic analysis of the EL lineshape must be performed by taking into account the multilayered structure of the sample. The numerical simulation of the lineshape around $E_0$ from first principles is a nontrivial task in random SL’s and goes beyond the aim of this work.

In conclusion, we have studied the fundamental optical transition of ordered, random, and random-dimer SL’s by means of EL and PL spectroscopies. These near-band-edge optical transitions have different EL lineshapes rising from and evidencing the different electronic structure of the SL’s.

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