Absorption spectra of GaAs/Al_xGa_{1-x}As random superlattices at 2 K

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Transmission spectra of ordered and disordered GaAs/$Al_{0.3}Ga_{0.7}$As superlattices (SL) were measured at 2 K. The experimental results are compared with numerical simulations obtained by means of a transfer matrix method. The numerical results indicate that a multiple-quantum-well model accounts for the main features of random SL, and that a disorder-induced fine structure is present. The agreement between the measured spectra and the numerical calculations supports the predicted features.

The study of the optical properties of disordered superlattices (DSL) optical properties has been attracting growing interest in recent years.[1-3] Since the first theoretical calculations,[1] intended to encourage and stimulate experimental efforts to study SL with controlled randomness, many different works on DSL have been performed. In particular, DSL have been extensively used to probe to the existence of disorder-induced localized states.[2] Recently, new experimental results renewed the interest in DSL: the photoluminescence (PL) of DSL was found to be more efficient than that of bulk material and that of ordered SL (OSL).[3] Such a result indicates that the study of DSL with tailored properties can greatly help the understanding of quantum-confinement-induced effects in disordered materials.

These arguments strongly stimulated theoretical efforts[4,5] in order to predict electronic and optical properties of DSL. In particular, the study of one-dimensional (1D) DSL within the framework of the effective-mass approximation in the Wannier-Bloch mixing representation[4,5] accounted for the observed redshift of absorption edge.[3] These calculations also predicted that the features of optical absorption spectra of DSL should become richer as the disorder is increased.[5] Moreover, by considering the coupling between the motion of the holes along the z axis and in the xy plane, it has been predicted[5] that the mixing of the heavy-hole (hh) and light-hole (lh) subbands results in a large nonparabolicity, which will become more and more apparent by increasing the degree of randomness and will produce a few weak structures, corresponding to forbidden transitions in single quantum wells (SQW).

In this paper we report low-temperature absorption spectra and their numerical calculations in GaAs/$Al_{0.3}Ga_{0.7}$As OSL and DSL. The disorder is introduced by varying the thickness of GaAs layers. The numerical evaluation of the energy levels of DSL is carried out by using the transfer matrix methods.[6] We show that the minibands of extended states of all the carrier species essentially split into N_W narrow subminibands of localized states centered on the SQW eigenvalues of energy (N_W is the number of different well widths present in the DSL). A DSL is thus expected to behave similarly to a multiple quantum well (MQW), especially in the strong disorder limit, where the simulations predict the complete localization of the carriers within the wells. Moreover, the numerical analysis shows the presence of a disorder-induced fine structure in the subminibands of localized states. This fine structure is more evident in the weak disorder limit, where the coupling between adjacent wells is stronger. Our experimental results confirm the expected optical features of DSL (Refs. 4 and 5) (e.g., the redshift of the absorption), and give first evidence that absorption spectra of DSL are richer.[4,5] The joint density of states (JDOS), computed by supposing a MQW recombination model of DSL, accounts quite well for the main features present in the absorption spectra, thus permitting a detailed physical interpretation of the observed

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absorption structures. Further, the experimental results give a quantitative confirmation of the disorder-induced fine structure predicted by the numerical analysis.

In a DSL the thickness L of every layer is a random variable with probability distribution $P(L)$. In order to simplify the problem, we consider random SL with constant barrier width $L_B = L_{Bo}$ and stochastic well width L_W. The stochastic variables L_B and L_W, expressed in monolayers (ML), can only be integers. Then, $P(L_W)$ can be expressed as follows:[4]

$$P(L_W) = \sum_{i=1}^{N_W} P(L_{Wi})\delta(L_W - L_{Wi}) . \quad (1)$$

Here L_{Wi} are the possible values of the well thickness with probability $P(L_{Wi})$. In order to study the influence of disorder, we suppose that L_{Wi} can be expressed by $L_{Wi} = L_{Wo} + (i-1)\Delta L_W$, where L_{Wo} is the minimum width of the well. In this case, the degree of disorder, represented by the width of the probability distribution of the energy in the SL sites (as in the case of the Anderson model[7]), goes to 0 as $\Delta L_W \to 0$ (OSL) and increases with ΔL_W. We choose to study a simple class of DSL with few different well widths ($N_W = 4$) having the same probability of occurrence [$P(L_{Wi}) = 1/4 \forall i \in \{0,1,2,3,\}$] and with $L_{Wo} = L_{Bo} = 9$ ML. To this purpose, one OSL (sample A, with $\Delta L_W = 0$ ML) and four DSL with different disorder strength (samples B, C, D, and E, with $\Delta L_W = 5$, 18, 20, and 23 ML, respectively) were designed.

The samples were grown by molecular beam epitaxy at a temperature of 890 K on (001)-oriented GaAs substrates. They consist of a GaAs substrate upon which there are a buffer layer of GaAs (0.3 μm thick), the SL (1 μm) cladded between two $Al_{0.3}Ga_{0.7}As$ layers (1 μm), and a GaAs cap layer (2 nm). The SL have about 200 periods. The growth rates were 0.6 μm/h. The error on the layer thickness is about 1 ML. The samples were prepared for transmission electron microscopy (TEM) by means of the wedge cleaving technique. TEM was carried out at 300 keV, with a resolution of 0.2 nm. The TEM analysis confirmed the morphological design of the produced samples. The Al concentration x of the barriers, deduced from the PL spectral position of the heavy-hole exciton line of $Al_xGa_{1-x}As$ cladding layers at 2 K, was found to be $x = 0.30 + 0.01$.[8] The GaAs substrate was etched away by using a selective etching[9] on a region of $\simeq 0.5$ mm diameter for transmission measurements. The etched samples were anodized to avoid multiple reflections of light.[10] The transmission measurements of samples immersed in superfluid He were performed by using a tungsten lamp. The light propagated along the growth axis of samples and the illuminated spot was of about 50 μm diameter. The transmitted light was analyzed with a double spectrometer and detected by a cooled GaAs photomultiplier tube. The spectra were recorded by using standard lock-in techniques.

The transfer matrix method has been used in order to obtain quantitative information on the energy levels of DSL.[6] The wave functions Ψ of the particle in the jth SL layer will depend on the coefficients A_j and B_j, representing the amplitudes of the traveling waves in opposite directions.[6] The transfer matrix $\mathcal{T}_{j\to j+1}$, which joins the coefficients of the jth layer with the ones of the $(j+1)$th layer is defined as

$$\begin{bmatrix} A_{j+1} \\ B_{j+1} \end{bmatrix} = \mathcal{T}_{j\to j+1} \begin{bmatrix} A_j \\ B_j \end{bmatrix} . \quad (2)$$

$\mathcal{T}_{j\to j+1}$ depends on the boundary conditions imposed at each interface between the different layers. Its explicit expressions can be obtained by using the particle current conserving boundary conditions.[6] The transfer matrix method permits the calculation of the SL densities of states $D(E)$ and of the Lyapunov exponent $\gamma(E)$, which is proportional to the reciprocal of the localization length $\lambda(E)$ of states.[11] In the case of localized states the amplitude of the wave function near some point z_0 in which it has a maximum will on average decay as[11]

$$\overline{|\Psi(|z-z_0|,E)|} \simeq \exp[-|z-z_0|\gamma(E)] . \quad (3)$$

Here the overbar indicates the average behavior of the probability amplitude in the SL. In our calculations we used the following material parameters:[12] band gap $E_g = (1.5194 + 1.48x)$ eV, electron effective mass $m_e = (0.0665 + 0.0835x)m_0$, hh effective mass $m_{hh} = (0.51 + 0.20x)m_0$, lh effective mass $m_{lh} = (0.082 + 0.078x)m_0$, where m_0 is the free electron mass. A band-offset ratio of $\Delta E_c/\Delta E_v = 0.65/0.35$ is used, corresponding to a conduction (valence) band discontinuity of 288 meV (155 meV) between GaAs and $Al_{0.3}Ga_{0.7}As$.

In order to clarify the effective of the disorder on the SL properties, we show $\gamma(E)$ [Fig. 1(a)] and $D(E)$ [Fig. 1(b)] predicted by the transfer matrix method for electrons in an OSL (A) and in a DSL (B). In the case of OSL, $\gamma(E)$ goes to zero, clearly indicating the existence of the extended states of the electron miniband, and $D(E)$ assumes the expected behavior. In the case of DSL the electron miniband splits in N_W subminibands of localized states [$\gamma(E) > 0$]. As previously reported,[2] the subminibands are centered on the eigenvalues of energy $e_n^{(i)}$ of SQW of widths L_{Wi} (triangles in Fig. 1), thus indicating that the carriers in the ith subminiband are mainly localized in the ith kind of well (n is the number of the subbands in the SQW). Our results show a strong disorder-induced splitting of e and lh minibands, as well as of hh minibands. This fact is different from previous results, which show a disorder-induced splitting that could be spectrally resolved for the hh miniband only.[2]

The inset in Fig. 1(a) shows the average probabilities $\overline{|\Psi|^2}$ for electrons of energy $e_1^{(4)}$ in correspondence of wells of width L_{W4} for a weakly DSL (sample B) and for a strongly DSL (sample E). They are computed by means of Eq. (3). The complete localization of carriers in the strong disorder limit is evident, and even in the weak disorder limit the carriers are considerably localized. This implies that optical transitions in DSL will mainly occur between energetic states of the same well. However, the numerical results also predict fine structure in $D(E)$, which is shown in the inset of Fig. 1(b) by the plot of $dD(E)/dE$ (sample B): the presence of states at lower and

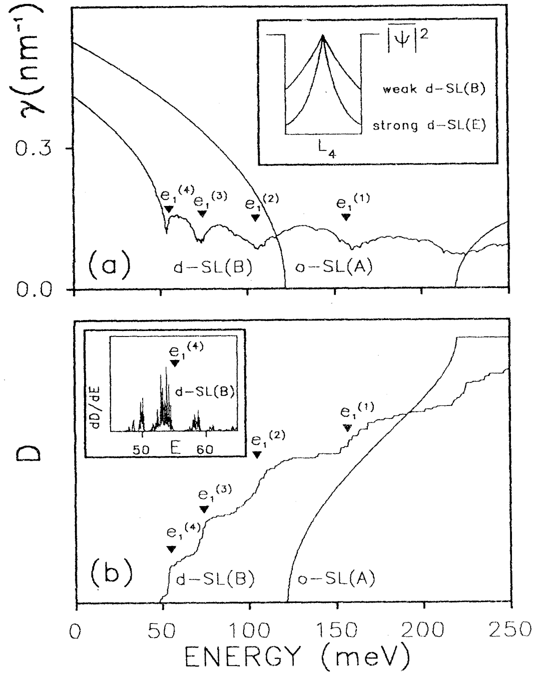

FIG. 1. (a) Lyapunov exponent for electrons $\gamma(E)$ and (b) electron density of states $D(E)$ for ordered and for disordered $GaAs/Al_{0.3}Ga_{0.7}As$ SL. The extended states miniband splits into subminibands of localized states centered on the eigenvalues of energy $e_1^{(i)}$ of isolated well of width L_{Wi}. The inset in (a) shows the average probability to find an electron of energy $e_1^{(4)}$ in a well of width L_{W4} in the cases of weakly and of strongly disordered SL: the complete localization in the strong disorder limit is evident. The inset in (b) shows detail of the derivative of $D(E)$ near $e_1^{(4)}$: a disorder-induced fine structure is evident. See the text for further details.

higher energies with respect to the SQW eigenvalue is evident. As in the case of field-induced localization in OSL,[13] the disorder-induced fine structure is evident in the weak localization limit and disappears in the strong localization limit, where SQW transitions dominate. A similar disorder-induced fine structure was also observed in numerical works on one-dimensional (1D) disordered systems[11] and in experimental results on layered disordered semiconductors.[14] The disorder-induced fine structure can be attributed to the existence of sequences of adjacent wells having the same width in the DSL. In fact, a sequence of two adjacent wells with the same width has two different eigenvalues, at lower and higher energies with respect to the SQW eigenvalue; a sequence of three adjacent wells shows three different eigenvalues, and so on. As the number of adjacent wells increases, the subminiband should approach the miniband of extended states of an OSL. However, the probability to have m adjacent wells with the same width in the investigated DSL decreases as $(1/N_W)^m$, and therefore the existence of long sequences of similar wells in the DSL is highly improbable. This implies that the subminibands of the investigated DSL cannot be continuous, as in the case of OSL, and that they must have a maximum on the SQW eigenvalue, as confirmed by the numerical simulation [inset of Fig. 1(b)]. The two main side bands visible over and below $e_1^{(4)}$ in the inset of Fig. 1(b) are related to eigenvalues of a system of two adjacent wells.

In order to obtain quantitative information about the absorption spectra of our DSL, the JDOS have been computed. In the following discussion we will thus assume a MQW model of recombination in DSL. The transitions between hh (or lh) and e subminibands will therefore be considered forbidden if e and h are localized in different wells, and allowed if they are localized in the same well, as in the MQW structures. In fact, the absorption coefficient for interband transitions can be written as[4]

$$\alpha(\hbar w)=(\hbar w)^{-1}\sum_{n,n'}|\hat{\epsilon}\cdot P_{n,n'}(k)|^2\rho_{n,n'}(\hbar w)\ , \qquad (4)$$

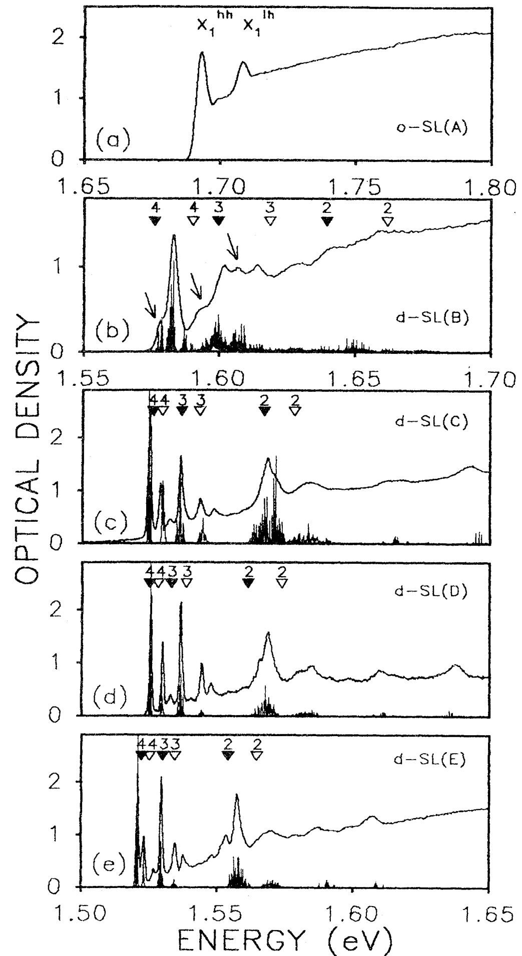

FIG. 2. Absorption spectra at 2 K of (a) ordered and (b)–(e) disordered SL. The disorder degree increases from (a) to (e). The experimental results for disordered SL (upper curves) are compared with the one-dimensional joint density of states (lower curves). In the ordered case (a) X_1^{hh} and X_1^{lh} indicate heavy-hole and light-hole ground-state excitonic transitions, respectively. In the disordered case (b)–(e) the numbered filled and hollow triangles indicate the energies of the excitons X_1^{hh} and X_1^{lh} in isolated wells of width L_{Wi}, respectively. The arrows in (b) indicate the disorder-induced fine structure in the weak disorder limit. See the text for further details. The spectral resolution is 1 meV.

where k is the e-h pair momentum in the xy plane, $P_{n,n'}$ denotes the momentum matrix element, and $\rho_{n,n'}$ is the JDOS between the eigenstate of the nth conduction subband with energy E_n and the eigenstate of the n'th valence subband with energy $E_{n'}$.

In Fig. 2 we show the measured absorption spectra (upper curves) and the computed 1D JDOS (lower curves) for the OSL (sample A) and for the four DSL (samples B–E). In fact, in our theoretical calculations excitonic effects have been neglected. Thus, the JDOS obtained in this approximation is expected to be a series of steps. To make the comparison between our approximate theoretical results and the measured spectra easier, we do two things: (i) We derive the calculated JDOS, to obtain the 1D JDOS, i.e., a series of peaks whose amplitudes are proportional to the step height and thus also approximately to the exciton oscillator strengths. (ii) We shift the obtained peaks by the approximate exciton binding energy obtained from the literature.[15] Therefore, we can compare only the calculated spectral position of the peaks, and their relative heights, with those observed in the absorption spectra. We do not pretend to compare line shapes, which would be completely inadequate. In Fig. 2, we report the computed energies of excitonic transitions in MQW (triangles), where SQW energy eigenvalues were determined by means of transfer matrix method, while the corresponding excitonic binding energies were taken from Ref. 15.

In the ordered case [Fig. 2(a)], the absorption spectrum shows the well-known features:[16] the two resolved peaks correspond to the ground state of the hh and lh ground-state excitonic transitions, respectively.

The absorption spectra of DSL [Figs. 2(b)–2(e)] show the expected redshift of the absorption edge.[4,5] This effect can be considered as a consequence of the introduction of wide wells in the DSL, rather than as a proper effect of the disorder. Further, as predicted by Ref. 5, the absorption spectra become richer in structures by increasing the disorder. By comparing the absorption lines of DSL and the energies of excitonic transitions in MQW, it results that DSL practically behave as a MQW in the strong disorder limit [Fig. 2(e)]. The numerical results for the JDOS of DSL agree fairly well with the experimental features of the absorption spectra, thus giving a further confirmation of the MQW model of recombination in DSL. A few weak structures that are not predicted by our 1D numerical simulation could be ascribed to hh-lh subband mixing.[5] The agreement shows that the excitonic binding energies are those of the SQW,[15] thus giving a further confirmation of the complete localization of carriers and of the MQW model in the strong disorder limit.

The absorption spectra also confirm the prediction of the numerical analysis on disorder-induced fine structure: in the weak disorder limit [Fig. 2(b)], where the lines are relatively wide, several shoulders and additional peaks appear (arrows), while in the strong disorder limit [Fig. 2(e)] the observed structures are quite narrow and the disorder-induced fine structure cannot be resolved. Finally, our results account for the observed high-temperature PL enhancement in DSL.[3] In fact, the observed optical features are attributed to disorder-induced localization of excitons along the growth axis of the DSL, that prevents the motion of *eh* pairs through the sample and thus reduces the nonradiative recombination probability.

In conclusion, our results confirm the previous predictions on DSL,[4,5] the validity of MQW model of recombinations and the existence of a disorder-induced fine structure. Our results yield a deeper understanding on the general properties of disordered semiconductor, which might be used to design optoelectronics devices by means of controlled randomness.

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