Change in dimensionality of superlattice excitons induced by an electric field

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(Received 11 August 1989)

We have observed an electric-field-induced change in the dimensionality of excitons in superlattices, from three to quasi-two dimensions. The exciton binding energy of a (40 Å)/(40 Å) GaAs/(GaAl)As superlattice, determined from low-temperature photocurrent experiments, increases more than 6 meV by the action of an electric field perpendicular to the superlattice layers. This sharp increase, from nearly the bulk value of GaAs at very low fields to the isolated-quantum-well value at high fields, is a direct consequence of the Stark localization of electrons and holes in superlattices.

The Coulombic interaction between electrons and holes leads to a bound state, the exciton, whose binding energy depends on the dimensionality of the system. Thus, in three dimensions that energy equals the effective Rydberg, whereas in two dimensions it is four times larger.

In bulk GaAs the exciton Bohr radius is 140 Å and the binding energy is 4.2 meV.[1] In artificial structures the exciton binding energy E_b can be modified by changing the average electron-hole separation. In a GaAs/(GaAl)As quantum well, where both carriers are strongly confined in one direction, E_b depends on the well thickness.[2-4] For thicknesses larger than the exciton Bohr radius it is close to the GaAs bulk value, but when the thickness decreases below the Bohr radius the exciton becomes quasi-two dimensional, and E_b increases several times. Finally, for very small thicknesses E_b decreases down to the (GaAl)As bulk value, due to the penetration of the electronic wave functions into the barriers.

In a superlattice the confinement of electrons and holes depends on the electronic coupling between wells. If the coupling is small, the carriers are confined in individual wells and the exciton resembles that of an isolated well (quasi-two-dimensional exciton). If the coupling is strong, then the carriers can move over several wells and the exciton is similar to the bulk exciton (three-dimensional exciton). In practice, the interwell coupling can be reduced, for example, by increasing the superlattice period. The dependence of the exciton binding energy on the superlattice period has been studied by Chomette *et al.*[5] In a given superlattice, the interwell coupling can be reduced by an electric field normal to the layers. In this paper we show that, for a fixed period, an electric field produces a transition from three-dimensional to quasi-two-dimensional excitons, with a sharp increase of the exciton binding energy.

When an electric field is applied perpendicular to the layers of a superlattice, its minibands are split into Stark ladders of levels.[6,7] Each level corresponds to an electronic state centered around a different well but extended through neighboring wells. As the field increases, these states localize, and at very high fields they are completely confined in single wells. (Note, however, that strictly speaking these are not bound states because the barriers are finite and the electrons can tunnel out of the wells.) Because of their small miniband width, heavy holes reach this stage at very low fields, compared with electrons and light holes. At moderate fields the optical spectra show excitonic transitions from a hole state, fully localized in a well, to an electron state centered in the same well (intrawell transition) or in a different well (interwell transitions).[6,7] At high fields, when all carriers are localized, the overlap between electron and hole states centered in different wells vanishes, and the intrawell transitions dominate the spectra.

The effect of the electric field on the exciton binding energy depends on whether the exciton is "intrawell" or "interwell." At zero field, when the electronic states are extended over many wells, E_b will be close to the bulk value for both kinds of excitons, but as the field localizes the states, E_b will increase for intrawell excitons and will decrease for interwell excitons. Here we focus on the intrawell excitons, whose binding energy was measured as a function of the electric field.

The exciton binding energy in high-quality heterostructures can be measured in absorption-related experiments from the energy difference between the 1*s* excitonic transition and the onset of the continuum. This method has been applied to study E_b as a function of the well thickness in quantum wells and of the period in superlattices.[2-4] When studying the effect of the electric field on E_b in superlattices, one is constrained in practice to a limited range of period lengths. When the period is very short, the large number of Stark-ladder transitions masks the onset of the continuum. On the other hand, for long-period superlattices the electronic states are completely localized even at zero field.[7] In this paper we report on a 20-period (40 Å)/(40 Å) $GaAs/Ga_{0.65}Al_{0.35}As$ superlattice, which is free of these limitations.

The superlattice, grown by molecular-beam epitaxy, was in the intrinsic region of a p^+-i-n^+ diode structure, which guaranteed a very uniform electric field, $\mathcal{E}$, related to the reverse bias voltage V by $\mathcal{E} \simeq (V - V_b)/W$, where V_b is the built-in voltage ($V_b = 1.62$ V) and W the total intrinsic-region thickness. Photocurrent (PC) spectra in the range of the fundamental band gap of the superlattice were measured at 5 K using low-power excitation (<0.2 mW/cm^2) from a dye laser pumped by a Krypton ion laser. Photoluminescence excitation spectra were less clear than PC spectra and will not be considered here.

In Fig. 1 we plot the PC spectra for two representative values of the electric field. At high fields [Fig. 1(b)] the spectrum is dominated by the intrawell transitions (noted here as $0h$ and $0l$ for heavy- and light-hole transitions, respectively) and it resembles the spectrum of an isolated quantum well. The strong peaks correspond to the fundamental 1*s* state of the excitons. The steplike features correspond to the 2*s* or other excited states of the excitons. We will assume that their position marks the onset of the continuum (band-to-band transitions). By doing so, the error introduced in E_b is at most the energy difference between the 2*s* level and the continuum, which is of the order of 1 meV.

At low fields [Fig. 1(a)] the PC spectrum becomes weaker and the intrawell transitions shift slightly to higher energies. For reasons that are not well understood, the onset of the continuum transforms from a steplike shape into a well-defined peak when the field is decreased [see Fig. 1(a)]. Moreover, there are two additional peaks which correspond to the 1*s* state of the interwell exciton, in which the electron and the hole wave functions are centered in two adjacent wells (following the notation of previous works[6,7] we label them as $-1h$ and $-1l$ for the heavy- and light-hole exciton, respectively). The interwell peaks shift strongly to lower energies as the field increases and disappear at high fields, when the coupling between wells becomes very small. (The symmetric transitions on the high-energy side cannot be seen here, as they are usually much weaker.[7])

The transition energies as a function of the electric field, obtained from a complete series of spectra similar to the ones shown in Fig. 1, are represented in Fig. 2. The intrawell transitions shift weakly with the electric field. The small red shift of the onset of the continuum is due to the Stark shift inside a quantum well.[8] The shift of the 1*s* transitions has an additional contribution from the change in the exciton binding energy, which is given by the energy separation to the onset of the continuum. The -1 interwell transitions shift strongly with the field, the shift being proportional, at moderate fields, to $\mathcal{E}$ and the superlattice period.[7]

The exciton binding energy, obtained from the energy separation between the 1*s* peak and the onset of the band-to-band transitions, has been plotted in Fig. 3 for heavy and light holes (solid and open circles, respectively). The binding energy increases sharply for fields up to about 20 kV/cm and then stays almost constant within the experimental error. The error in the exciton binding energy, estimated to be ± 1 meV, originates in the uncertainty in the position of the spectral features, mainly the onset of the continuum, which becomes difficult to recognize as the features broaden at high fields.

The field-induced increase of the exciton binding energy in Fig. 3 reflects the change in its dimensionality, from three dimensions at $\mathcal{E}=0$ to quasi-two dimensions in the high-field limit. In Fig. 3 we also show (shaded area) the

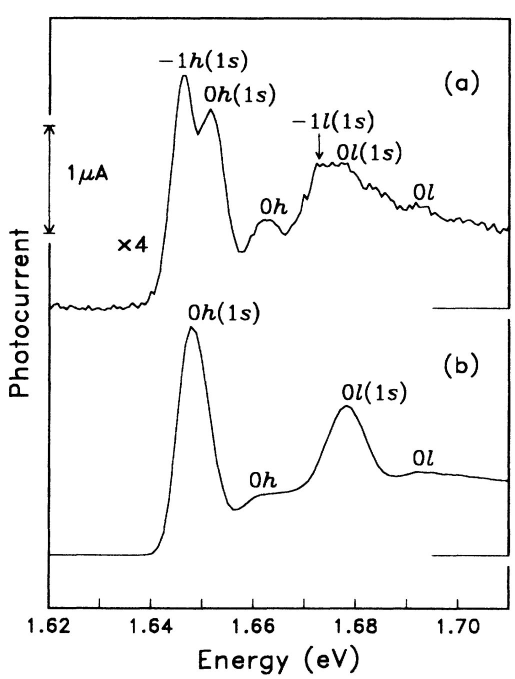

FIG. 1. Photocurrent spectra of a (40 Å/40 Å) $GaAs/Ga_{0.65}Al_{0.35}As$ superlattice at 5 K for two representative values of the electric field applied normal to the layers. (a) 10 kV/cm and (b) 40 kV/cm.

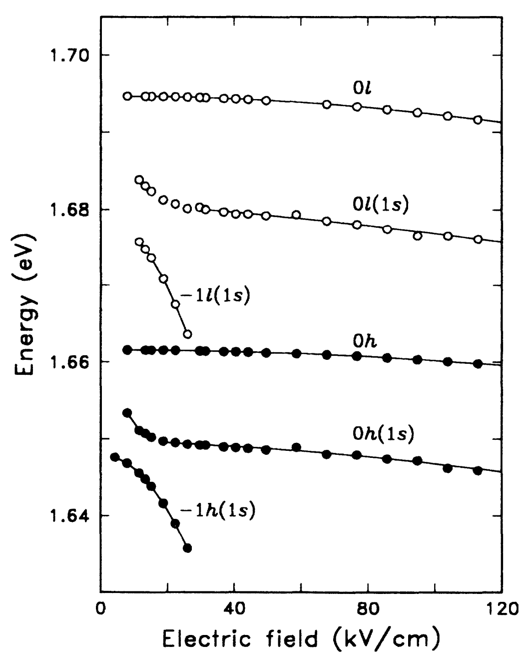

FIG. 2. Electric-field dependence of the transition energies for heavy holes (solid circles) and for light holes (open circles) obtained from photocurrent spectra. The separation between $0h(1s)$ [$0l(1s)$] and $0h$ [$0l$] peaks gives the binding energy for heavy- and (light-) hole excitons.

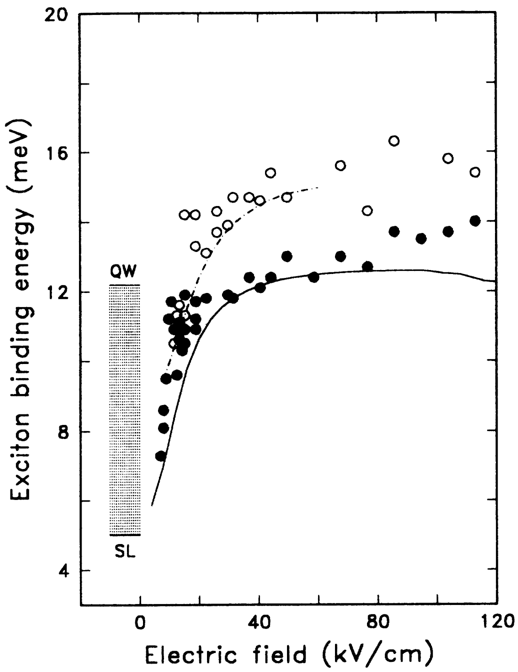

FIG. 3. Exciton binding energy as a function of the electric field. Solid (open) circles stand for heavy- (light-) hole excitons. The solid (dashed) curve represents the calculated values for the heavy- (light-) hole exciton shifted upwards by 3 meV (5 meV) in order to compare directly with the experimental results (see text). The shaded rectangle depicts the expected limits for the heavy-hole exciton binding energy: the superlattice (Ref. 5) (SL) and the isolated–quantum-well (Ref. 4) (QW) values at zero electric field.

region of expected values for the heavy-hole exciton binding energy: the range limited by the heavy-hole exciton binding energy for (40 Å/40 Å) superlattice[5] and for an isolated 40-Å-wide quantum well,[4] both at zero electric field. At very high fields, the exciton binding energy should reach a maximum and then decrease because of the electric-field-induced polarization of the exciton (Stark effect),[9] but in the considered range the effect is smaller than the experimental uncertainty.

The experimental results of Fig. 3 have been compared with theoretical values of the intrawell exciton binding energy, calculated within the envelope-function approximation, as a function of the electric field. First, electron and hole wave functions were obtained in the absence of Coulomb interaction as linear combinations of Airy functions in each layer. Then, the boundary conditions at the interfaces were introduced with the transfer-matrix method. For simplicity, the difference in effective mass between the wells and the barriers was neglected, and the GaAs values were considered throughout. The heavy- and light-hole states were calculated independently, without considering valence-band mixing effects. For numerical reasons, the superlattice was modeled as a finite number of quantum wells, large enough so that the central electronic states at low fields were unaffected by the presence of both ends. The other states were then obtained from the central one by the proper transformation. In order to guarantee the existence of bound states, the superlattice was limited by infinite barriers near both ends.

The exciton states were approximated by one-parameter variational wave functions, written as a product of the calculated electron and hole wave functions for the superlattice direction, and an exponentially decaying function for the in-plane direction. Finally, the coupling between different excitons was considered. Here we will discuss only the results directly related to the experiments and will leave other details, like the interwell excitons, for a further publication. At zero electric field, electrons and holes are extended over many periods, and the exciton, although anisotropic, is very similar to the bulk one. However, for fields of a few kV/cm, the heavy-hole exciton becomes very peculiar, in the sense that the electron is delocalized over a few periods while the hole is localized in a single well. The exciton is then forced to move close to the well, and its binding energy decreases slightly with respect to the zero-field value. Unfortunately, this range of fields is below the experimental values. For stronger fields, the theoretical binding energies have the same field dependence as observed experimentally, although their values are smaller. A comparison between the two is shown in Fig. 3, where the theoretical values for heavy and light holes, shifted upwards by 3 and 5 meV, respectively, have been represented by solid and dashed lines. As can be seen, the calculations account very well for the change in exciton binding energy once the rigid shift is introduced. The smallness of the theoretical values is due in part to the simplifications used in the calculations: the use of only one variational parameter for the exciton wave function, and the omission of the valence-band mixing. Since the latter affects mainly the in-plane light-hole mass, its exclusion from the calculation explains the larger rigid shift in Fig. 3 for the light-hole exciton. (For example, in a single quantum well, when valence-band mixing is included, the light-hole exciton binding energy increases by about 2.5 meV while the heavy-hole exciton binding energy increases by less than 0.5 meV.[10])

In summary, we have shown that an electric field reduces the coupling between the superlattice quantum wells, localizing the carriers into individual wells and producing a change of dimensionality for intrawell excitons from three dimensional to quasi-two dimensional. Consequently, the intrawell exciton binding energy increases from nearly the bulk value at low fields to the isolated-well value at high fields.

This work has been sponsored in part by the U.S. Army Research Office. One of us (F.A.-R.) acknowledges the support of the Institute of Materials Science of Madrid (Spain). We thank L. F. Alexander for sample preparation.

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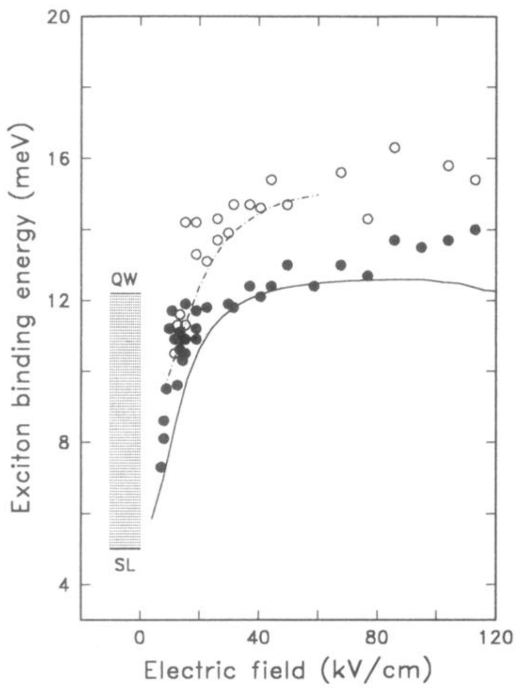

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