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Franz-Keldysh oscillations and Wannier-Stark localization in GaAs/AlAs superlattices with single-monolayer AlAs barriers

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Using photocurrent spectroscopy, we have studied the absorption properties of GaAs/AlAs superlattices with only 1-monolayer (ML) and 2-ML-wide AlAs barriers in an electric field F parallel to the growth direction. Wannier-Stark localization of the carrier states takes place at large electric fields, even for the case of 1-ML AlAs barriers. In the low-field regime, Franz-Keldysh oscillations are observed at fields up to 60 kV/cm. The oscillation period shows an $F^{2/3}$ behavior, as theoretically predicted, and is larger than expected from a one-electron model.

If the barrier width in a semiconductor superlattice (SL) is kept small enough, the coupling between different wells of the SL gives rise to miniband formation and, in an electric field F along the direction of the SL, Wannier-Stark localization is observed.[1,2] This effect is due to the splitting of the minibands of delocalized states into a set of localized subbands at equidistant discrete energies, as the translational symmetry of the SL with period d is destroyed by the electric field F. Experimentally, the phenomenon manifests itself by the appearance of excitonic optical transitions with a field-dependent energy shift approximately given by $neFd$. The index n ($n = \ldots, -2, -1, 0, 1, 2, \ldots$) assigned to these Stark ladder transitions gives the number of SL periods between the centers of the participating conduction and valence subbands, the energy slope $neFd$ corresponds to the potential drop along this distance.

The appearance of this effect is a signature of the *coherence* between the different wells of the SL (Ref. 3) which improves if the barrier width is further reduced. In bulk semiconductors in an electric field, on the other hand, coherence manifests itself by the occurrence of Franz-Keldysh oscillations[4] (FKO) in electroabsorption experiments. Although there has been some evidence of FKO reported for the case of $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ SLs,[5] it is still an open question what precisely happens to the absorption properties if the barrier width goes to zero, i.e., for the transition from SL material to bulk material.

We have therefore studied the electroabsorption properties of GaAs/AlAs SLs with 1- and 2-monolayers (ML, 1 ML=0.283 nm) -wide AlAs barriers by photocurrent (PC) spectroscopy. Our experiments reveal that Wannier-Stark localization still takes place at 1 ML barrier thickness. There is also a relatively large field regime (up to 60 kV/cm) where only FKO are observed. This enables us to perform the first analysis of the field dependence of FKO in a SL.

Before going into the experiments, it is important to discuss a few theoretical results which are plotted in Fig. 1. The calculations were carried out using the transfer-matrix method in the two-band $\mathbf{k}\cdot\mathbf{p}$ approximation.[6] First, as shown in Fig. 1, the introduction of single-ML (and even sub-ML) AlAs barriers produces a clear confinement effect on the lower edge of the first miniband. In combination with similar calculations for the valence minibands, we can therefore determine the AlAs barrier thickness from the experimental absorption edge. Second, these confinement energies are affected only very weakly if the AlAs contained in the barriers is smeared out across a few monolayers which might happen for nonoptimized growth conditions. An example of this fact is given in the inset in Fig. 1 where we have plotted the energy-momentum dispersion of the conduction minibands for SLs with 1-ML-wide AlAs (solid lines) and 3-ML-wide $Al_{0.33}Ga_{0.66}As$ (dotted lines) barriers and identical periods of 13 ML. We note that this result also provides an *a posteriori* justification of using envelope functions. Third, not only the miniband energies but even the entire miniband dispersion are nearly identical, thus suggesting the same

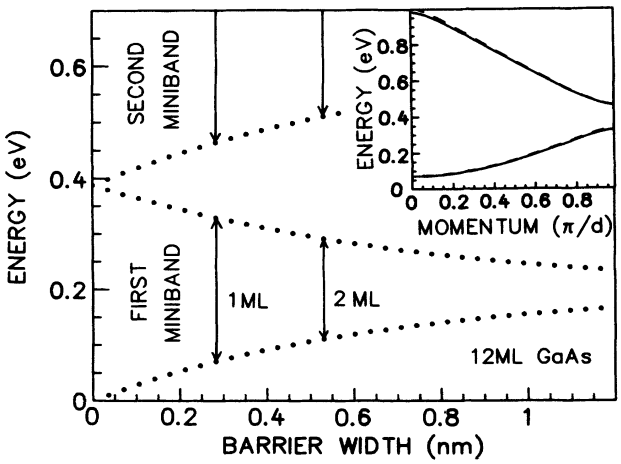

FIG. 1. Energies of the conduction minibands of a GaAs/AlAs SL with 12-ML GaAs wells vs barrier width. The inset shows the conduction miniband dispersion of a SL of period $d=13$ ML with 1-ML AlAs (solid lines) and 3-ML $Al_{0.33}Ga_{0.66}As$ (dashed lines) barriers.

electrical and optical properties in both cases. This means that we do not have to worry about a *small* spatial broadening of the AlAs barrier layers in our experiments. We note, however, that such a difference is known to produce clear effects on the phonon modes confined in the barriers and, consequently, on the vibronic Raman signal of these structures.[7] Fourth, because of the strong coupling, the dispersion of the first miniband resembles a parabola rather than a cosine function, i.e., the center of the miniband (in momentum space) is not located at the energy center but at a significantly smaller energy. Thus, the tight-binding approximation[8] breaks down. The effective mass obtained from this calculation is only 8% larger than for bulk GaAs.

The samples used in this study were grown by molecular-beam epitaxy on a (100)-oriented n^+-type GaAs substrate. The active region consists of an 80 period SL with nominally 1- and 2-ML AlAs barriers, respectively, and 3.2-nm GaAs wells. The undoped SL is grown on top of a 0.2-μm-wide Si-doped n^+-type GaAs buffer followed by 0.6-μm n^+-type $Al_{0.45}Ga_{0.55}As$ (both 10^{18} cm^{-3}). On top of the SL there are Be-doped layers of 0.1-μm $Al_{0.45}Ga_{0.55}As$ (10^{19} cm^{-3}), 0.5-μm $Al_{0.45}Ga_{0.55}As$ (2×10^{18} cm^{-3}), and 30-nm GaAs (2×10^{18} cm^{-3}). These *n-i-p* diode structures were processed into 300×400 μm^2 rectangular mesas with small Ohmic contacts of Ti-Au on the top side and Au-Ge/Ni on the substrate side.

PC spectra were obtained at a temperature of 77 K using standard lock-in techniques. The illumination system consists of a halogen lamp and an 85-cm double monochromator. The spectral resolution was set to 0.5 nm. The PC spectra are corrected for the spectral dependence of the illumination system as determined from a calibrated Si photodiode.

Figure 2(a) shows a series of PC spectra under different applied voltages. At large fields (reverse bias regime), spatially indirect Stark-ladder transitions are observed. Unlike in more weakly coupled SL structures where these transitions show strong excitonic enhancement,[1,9] they appear as shoulders in the spectra without any excitonlike peaks. We attribute this feature to the very strong interwell coupling giving rise to efficient exciton dissociation in an electric field. Another set of structures observed at small electric fields are due to FKO as will be shown later. The high quality of the samples manifests itself from the absence of PC below 1.6 eV at these small fields, thus indicating that there is no significant formation of growth islands in the SL region.

The voltage dependence of the transition energies as obtained from these spectra is plotted in Fig. 2(b). Transitions between electron and heavy-hole Stark-ladder states are seen down to a displacement index of $n=-7$, as identified through their linear dependence on the applied bias. The slopes are well described by n/N, with the number of SL periods $N=80$, indicating a well-defined electric field F. As already mentioned, the two lowest-energy structures in the forward bias regime (positive applied voltage) are associated with FKO.

At energies above 1.7 eV, the features become more difficult to understand, however, which is due to the oscillatory field dependence of the oscillator strengths of the

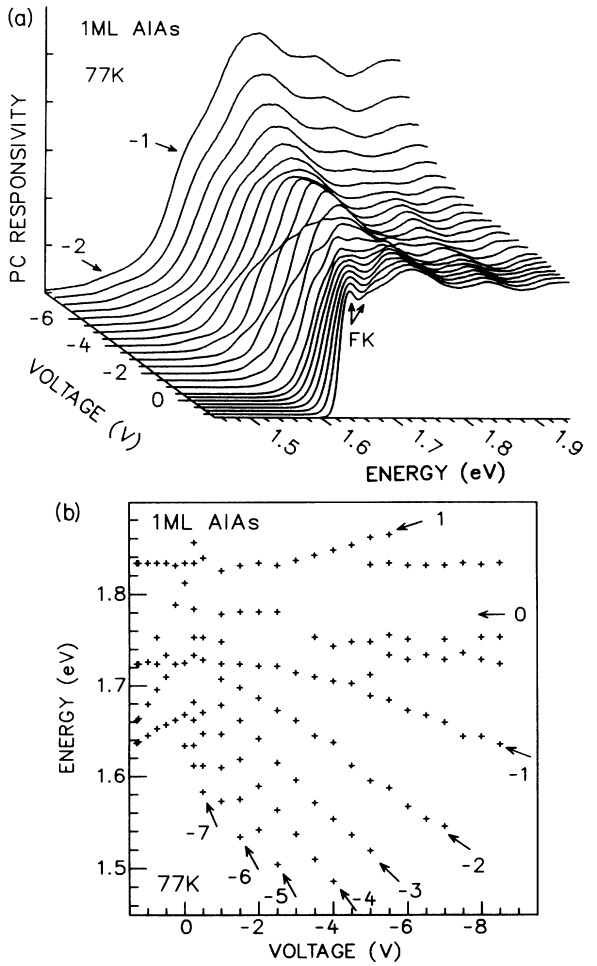

FIG. 2. (a) PC spectra of the 1-ML sample at different applied voltages between +1.2 V (forward bias) and −8.0 V. Structures due to the Franz-Keldysh effect (FK) and due to the $n=-1$ and -2 Stark-ladder transitions are indicated. (b) Experimental exciton transition energies, as obtained from the spectra of (a), vs applied voltage.

individual transitions at moderate electric fields,[10,11] and due to the appearance of additional transitions, like light-hole transitions, with similarly field-dependent oscillator strengths. Already the $n=-1$ transition deviates from its linear high-field behavior at bias values larger than −5 V [see Fig. 2(b)], probably because of some low-field mixing effects of the type discussed in Ref. 10.

Moreover, an experimental analysis of the transition energies suffers from the large (lifetime) broadening. The $n=0$ transition, for example, expected at about 1.78 eV, shows up at the correct energy at bias voltages between −1 and −2.5 V. At larger reverse bias, however, the apparent transition energy is only 1.75 eV. This discrepancy is too large to be explained by an increase of the exciton binding energy with increasing electric field.[11] It is caused by the $n=-1$ light-hole transition occurring in the same energy regime, thus overlapping with the $n=0$ heavy-hole transition. This point becomes evident by comparing Fig. 2(b) with similar data obtained from the

2-ML barrier sample (not shown here) where these two transitions can still be distinguished.

In order to investigate the low-energy structures in the forward bias regime of Figs. 2(a) and 2(b) further, we generated electroabsorption spectra by numerical subtraction of PC data recorded under different bias voltages. To this end, the PC at 1.3-V bias (roughly 5 kV/cm), which is just above the exciton dissociation limit, was taken as a reference. Figure 3(a) shows some results of this procedure. In fact, the spectra at 1- and 0-V bias [solid and dashed curves in Fig. 3(a), respectively] show the usual shape which is characteristic for FKO (see, e.g., Ref. 4).

The dotted line in Fig. 3(a) shows the electroabsorption at a bias of -4 V, corresponding to $F=190$ kV/cm. The shape still resembles the low-field behavior. However, there are additional features which are caused by Wannier-Stark localization. Comparing these results with the energies plotted in Fig. 2(b) we conclude that the shoulders of the dotted line in Fig. 3(a) at 1.48, 1.55, and 1.64 eV, respectively, are associated with the $n=-4$, -3, and -2 Stark-ladder transitions. Additional data obtained at similar fields reveal that the peak at 1.70 eV is a superposition of a Franz-Keldysh peak and the $n=-1$ Stark-ladder transition. We note that these Stark-ladder transitions are much more pronounced in *differential* electroabsorption spectra, i.e., when subtracting PC spectra obtained at almost identical electric fields.

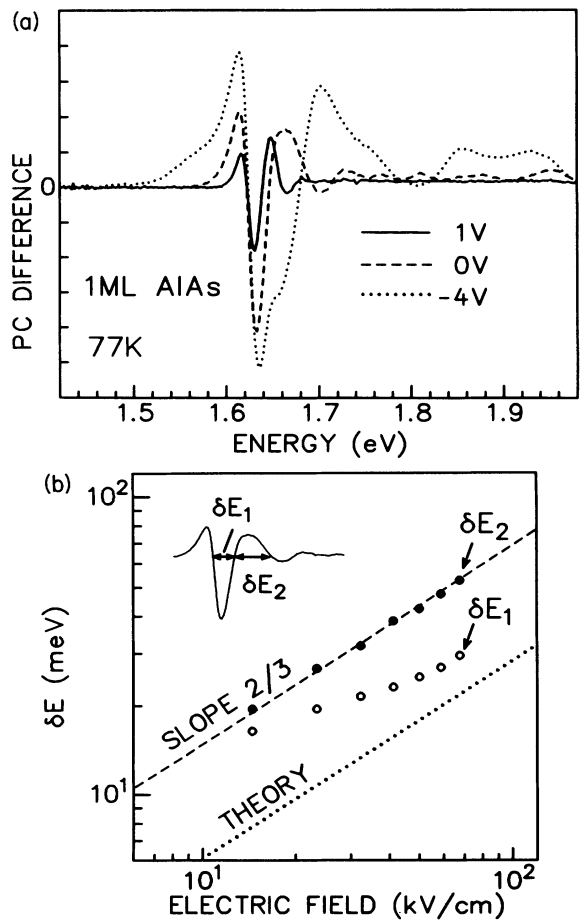

FIG. 3. (a) PC difference vs energy. The solid, dashed, and dotted lines correspond to a bias of $+1$, 0, and -4 V, respectively. The subtracted reference spectrum was recorded at $+1.3$ V. (b) Energy differences δE_1 and δE_2 vs electric field. The inset shows the definition of δE_i.

For a more detailed analysis of the FKO, we adopted the method of Hamakawa, Germano, and Handler[4] as follows. In a one-electron model, the imaginary part of the dielectric function $\Delta\epsilon_2(E,F)=\epsilon_2(E,F)-\epsilon_2(E,0)$ at the energy E and the electric field F can approximately be expressed as a function of the dimensionless variable

$$\eta=(E-E_g)(8\mu)^{1/3}(eF\hbar)^{-2/3}. \quad (1)$$

Here μ is the reduced effective mass in the field direction.[12] Furthermore, the zeros of the change $\Delta\alpha$ in the absorption coefficient coincide with the zeros of $\Delta\epsilon_2$ because of the relation[4] $\Delta\alpha=\omega\Delta\epsilon_2/nc$, with the refractive index n and the light velocity c. Similarly, the zeros of the absorption (or PC) difference itself coincide with those of $\Delta\alpha$. Thus, using Eq. (1), the difference $\delta E_i=E_{i+1}-E_i$ between two subsequent zeros E_i $(i=1,2,\ldots)$ of $\Delta\alpha$, as sketched in the inset in Fig. 3(b), obey the relation

$$\delta E_i=(e^2\hbar^2/8\mu)^{1/3}F^{2/3}\delta\eta_i, \quad (2)$$

with η_i $(i=1,2,\ldots)$ being the zeros of $\Delta\epsilon_2$ and $\delta\eta_i=\eta_{i+1}-\eta_i$. Numerically, we obtain $\delta\eta_1\approx\delta\eta_2\approx 1.15$ from Ref. 4.

The result of this evaluation is plotted in Fig. 3(b). Here the built-in voltage V_{bi} was considered as a free parameter and adjusted such that the solid circles in Fig. 3(b) were on a straight line, which was the case at $V_{bi}=1.42$ V. The dotted line shows the theoretical result obtained for a reduced effective mass of $\mu=0.06$ free-electron masses. δE_2 shows the correct $F^{2/3}$ behavior. As in bulk material,[4] the slope of δE_1 does not agree very well with theory. This discrepancy is attributed to excitonic effects which are important close to the band gap E_g and which are not contained in the simple theoretical model.

The striking difference from the calculated behavior and also from the case of bulk GaAs,[13] however, consists in the fact that the oscillation periods δE_1 and δE_2 are significantly larger than theoretically predicted, δE_2 being more than twice as large as expected. This effect, which is not yet fully understood, must therefore be associated with specific SL features, like the energy splitting between the light- and heavy-hole states, the large period as compared to the host lattice, or the anisotropy of the SL material. We point out that the discrepancy cannot be related to the effective-mass value μ since it influences the oscillation period only weakly [see Eq. (2)]. Also it cannot be due to collision broadening[14] because of the $F^{2/3}$ behavior.

Theoretically, one would, of course, expect to see several sets of FKO, namely, for the lower and upper (van Hove singularities of type M_0 and M_1) edges of the heavy- and light-hole minibands. However, only one set of oscillations is observed. The reason probably arises from weakness of the M_1 singularities and from the small splitting between the two M_0 singularities. Indeed, simple one-band Kronig-Penney calculations yield a splitting of

only 10 meV between the two M_0 singularities. In addition, the miniband widths (36 and 255 meV, respectively) differ by a factor of about 7. Thus, via the respective densities of states, the amplitude of the FKO should be 7 times larger for the heavy-hole case than for the light-hole case. The study of these weaker contributions would therefore require an experimental discrimination between heavy-hole and light-hole transitions, e.g., by applying polarization selection rules.[15]

In conclusion, we have investigated the electric-field dependence of the absorption properties of GaAs/AlAs SLs with single-monolayer AlAs barriers. At small fields, we performed the first analysis of the field dependence of FKO in a SL, which was possible because of the extremely strong interwell coupling. The oscillation period shows an $F^{2/3}$ behavior as theoretically expected. The period itself, however, is unexpectedly large, causing a highly sensitive field dependence of the absorption properties which might be exploited in novel absorption modulators. With increasing field ($F > 60$ kV/cm), we found a transition from FKO to Wannier-Stark localization. Our results also show that the electric-field-induced localization of carriers can still be achieved by barriers which are only one atomic monolayer wide.

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