Franz-Keldysh oscillations in the photomodulated spectra of an $In_{0.12}Ga_{0.88}As/GaAs$ strained-layer superlattice

F. Cerdeira
Instituto de Física, UNICAMP, Campinas, São Paulo, Brazil

C. Vázquez-López
Instituto de Ciencias, Universidad Autonoma de Puebla, Mexico

E. Ribeiro, P. A. M. Rodrigues, and V. Lemos
Instituto de Física, UNICAMP, Campinas, São Paulo, Brazil

M. A. Sacilotti
CPqD TELEBRÁS, 13 100 Campinas, São Paulo, Brazil

A. P. Roth
Microstructural Sciences, Division of Physics, National Research Council, Ottawa, Canada
(Received 16 May 1990)

The low-temperature photoreflectance and phototransmittance spectra of a 100 Å-period $In_{0.12}Ga_{0.88}As/GaAs$ strained-layer superlattice exhibit oscillatory behavior at photon energies above the absorption edge. This behavior can be consistently interpreted in terms of Franz-Keldysh oscillations induced by a built-in electric field of $\cong 10^4$ V/cm and are made possible by a rather large electron miniband dispersion ($\Delta \cong 38$ meV). The reflectivity spectrum also shows signs of incipient Stark-ladder formation.

The presence of an electric field F profoundly affects the electronic states of a semiconductor.[1,2] In cases where the effective-mass approximation is valid, the eigenstates are represented by Airy functions, which are delocalized and have a continuous-energy spectrum (the Franz-Keldysh effect). The oscillatory nature of the Airy function is reflected in the appearance of oscillations in the frequency dependence of the absorption coefficient and other related quantities.[3,4] These Franz-Keldysh oscillations (FKO's) appear above the energy gap, their period scales with electric field as $F^{2/3}$ and have been widely observed in the modulated absorption and reflectivity spectra of bulk semiconductors.[3–6] The effective-mass approximation breaks down when the electron (hole) is capable of gaining energies from the electric field which is comparable to or greater than the bandwidth Δ of the zero-field energy band. In this high-field regime a tight-binding approach shows that eigenstates become discrete (Stark ladders) and localized on a length scale $\lambda = \Delta / eF$, where e is the electronic charge. These are known as Stark-Wannier states.[1] Although the validity of this result was initially questioned,[2] it is believed now that both approaches are valid in different ranges of electric fields and bandwidths.[7] Thus, in bulk solids, where bandwidths are very large ($\Delta \cong 1$ eV), no clearcut evidence was found of Stark-Wannier states (which would require very large electric fields), while the appearance of Franz-Keldysh oscillations is a common occurrence.[3–6] On the other hand, in a semiconductor superlattice bandwidths can be tailored to a few tens of a meV and Stark ladders could occur at easily attainable fields. These expectations are confirmed by recent experimental observations made in $Al_{1-x}Ga_xAs/GaAs$ superlattices.[8] Theoretical calculations show that two regimes, Franz-Keldysh (FK) and Stark-Wannier (SW), can exist in these superlattices in opposite extremes of electric-field values given approximately by[9]

$$f = eFD/\Delta = D/\lambda \begin{cases} \ll 1 \quad (\text{FK}) & (1a) \\ \geq 1 \quad (\text{SW}) , & (1b) \end{cases}$$

where D is the period of an N-period superlattice. In both cases we must have $Nf = ND/\lambda \gg 1$ in order to avoid edge effects. Excitonic effects are not included in the treatment of Ref. 9. A more recent calculation shows that these effects, and those of possible fluctuations in the superlattice period, do not substantially alter the high-field results.[10] In the low-field limit exciton formation should contribute to localize the carriers and would, thus, inhibit the appearance of Franz-Keldysh oscillations. Hence, these oscillations should be observed only when the field is large enough to ionize the exciton, i.e.,

$$eFa_0 \geq E_{\text{exc}} , \qquad (2)$$

where a_0 and E_{exc} are the Bohr radius and binding energy of the exciton, respectively. Thus, depending upon a delicate balance between F, D, a_0, E_{exc}, and Δ, we could have localized excitons and delocalized Franz-Keldysh states in the low-field limit. On the other hand, the high-field limit always results in Stark-Wannier states. This might be the reason why all the reports so far on $Al_{1-x}Ga_xAs/GaAs$ superlattices show evidence of Stark-ladder formation[8] but not of Franz-Keldysh oscillations. Although some such oscillations are seen in the electroreflectance data of Voisin *et al.*,[11] they are attributed to bulk contributions from the space-charge region in the GaAs buffer layer. This ambiguity in identifying the source of the oscillations would be removed if observations were made in superlattices fabricated in such a way that the material composing the quantum wells did not appear in bulk form in any other part of the sample. This is the case of $In_xGa_{1-x}As/GaAs$ superlattices, where the alloy material appears only in the quantum wells. Thus, any oscillations in the $In_xGa_{1-x}As$ photon-energy region cannot be ascribed to a bulk effect. An additional advantage of this system is that strain decouples heavy- and light-hole states. On the negative side, broadening by random alloy potential and strain relaxation complicates the theoretical analysis. A recent electroreflectance experiment performed in this type of superlattice[12] shows evidence of oscillatory behavior, interpreted by the authors as Franz-Keldysh oscillations. We believe, however, that the displayed line shapes could also be interpreted in terms of Gaussian excitons; also, the authors make no effort to examine the field dependence of the periodicity in their oscillations. In the present work we present photoreflectance and phototransmission spectra of a 100 Å-period $In_{0.12}Ga_{0.88}As/GaAs$ superlattice[13] which exhibits unmistakable oscillatory behavior. These oscillations occur at energies E_n above the lowest heavy-hole to conduction-band transition (E_0) and their period scales as $(E_n - E_0)^{3/2}$. This and the general form of the spectra lead us to identify them as Franz-Keldysh oscillations. The period of the oscillations would be explained by the existence of an electric field (unintentionally built-in during growth) of $F \cong 7$ kV/cm. A bandwidth of $\Delta \cong 38$ meV is obtained from a calculation using the envelope-function formalism,[14,15] yielding $f = 0.18$. Such a field satisfies the inequalities of Eqs. (1b) and (2), thus lending plausibility to our interpretation. Even so the incipient formation of Stark ladders cannot be ruled out. In fact, recent experiments on this type of superlattice revealed clear evidence of Stark-ladder formation in samples with $\Delta \cong 15$ meV (which is small enough to preclude the observation of FKO's).[16] In any case, we believe that our results are the first evidence of a field regime in which Franz-Keldysh states exist.

The sample used in our experiment is a 20-period superlattice of $In_{0.12}Ga_{0.88}As/GaAs$ with equal (50 Å) quantum well and barrier thickness grown by metal-organic chemical-vapor deposition on a (100) GaAs substrate.[13] Previous Raman measurements performed on this sample allowed quantitative determination of the strain present in each type of layer.[15] The quality of the interface and the value of the sample parameters were confirmed by x-ray, TEM, and photoluminescence measurements.[13,14] Experiments were also carried out in a piece of this sample which was subjected to annealing at 850 °C during 30 min in an $AsH_3 + H_2$ atmosphere. This was done in an effort to determine the importance of interface roughness in the interpretation of our results. We measured the transmission and reflectivity spectra modulating the signal with a secondary light beam from a He-Ne laser. Measurements were performed both at 300 and at 77 K, with the sample immersed in liquid nitrogen. The experimental setup is standard in photomodulated spectroscopy,[5] with reflected and transmitted light detected by a Si photodiode whose output goes into a Stanford SR 530 lock-in amplifier. Both ac and dc outputs are digitalized and stored in the memory of a PC-XT microcomputer which also controls the spectrometer scan. No contact was made on the sample, so that the electric field inferred from our spectra is built-in as a consequence of growth conditions.

The modulated reflectivity ($\Delta R/R$) and transmission ($\Delta T/T$) spectra, at 300 and 77 K, of our sample are displayed in Fig. 1. Reflectivity curves show a peak at 1.506 eV (1.426 eV) at 77 K (300 K) which corresponds to the E_0 transition of the GaAs barriers and buffer layer. Another feature that appears in all spectra is a weak line at approximately 40 meV below the GaAs feature. This line appears consistently in a large variety of $In_xGa_{1-x}As/GaAs$ superlattices and some of the substrates, and is attributed to an impurity present in the GaAs portions of our sample.[17] The remaining part of the spectra could, in principle, be interpreted by attributing an intersubband transition to each one of the observed features. Within this framework, we tried to fit this portion of our spectra with a variable number of Aspnes[4] third-derivative line shapes (TDLS).[18] For consistency, at a given temperature, the same number of transitions (at approximately the same energy position) should fit both $\Delta R/R$ and $\Delta T/T$ spectra. It is possible to obtain excellent fits by this procedure using two lines

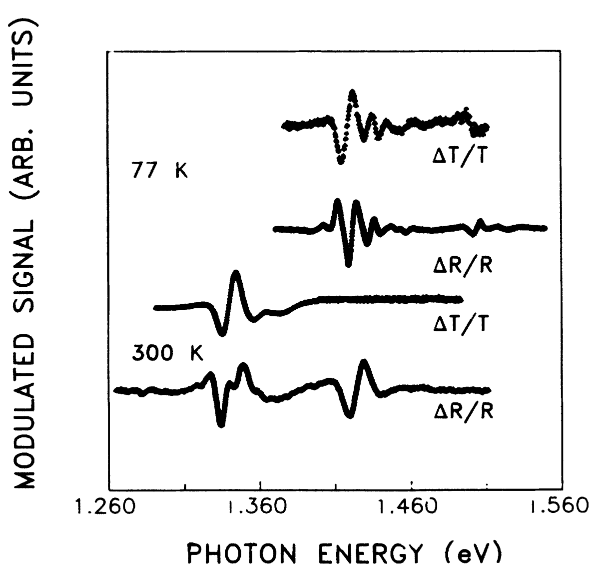

FIG. 1. Photoreflectance and phototransmission spectra of our sample at room and liquid-nitrogen temperature.

TABLE I. Parameters obtained by fitting our spectra with third derivative line shapes. For the annealed sample a Gaussian line shape (see Ref. 20) was used.

T (K)	Sample	n	E_n (eV)	$\Delta T/T$ (Γ_n) (meV)	I_n	E_n (eV)	$\Delta R/R$ (Γ_n) (meV)	I_n
300	As grown	0	1.337	(10)	100	1.333	(7)	100
		1	1.347	(10)	83	1.350	(8)	53
77		−1				1.403	(8)	31
		0	1.415	(8)	100	1.413	(9)	100
	As grown	1	1.423	(7)	73	1.422	(7)	140
		2	1.430	(5)	41	1.434	(5)	58
		3	1.439	(5)	50	1.445	(6)	18
		4	1.455	(11)	22	1.458	(5)	10
77		0	1.414	(4)	100			
	Annealed	1	1.446	(8)	20			

for the room-temperature spectra and five lines for the 77 K transmission spectrum. For the $\Delta R/R$ spectrum at 77 K an additional weak line at 1.403 eV is necessary to obtain a good fit to the data. The energy (E_n), linewidths (Γ_n), and relative intensities (I_n) obtained from these fits are listed in Table I. The transitions are numbered in consecutive order, taking as $n=0$ the lowest-energy strong line, whose energy position (E_0) coincides with the first allowed heavy-hole to conduction-band transition at the center of the minizone (Γ point). In general, we see that there is good agreement in the values of E_n, Γ_n, and I_n obtained by fitting the reflectivity and transmission spectra. The most striking difference occurs at 77 K, where the $\Delta R/R$ spectrum shows a line ($n=-1$) not present in $\Delta T/T$. Even more curious is the fact that while only two lines are needed to fit the room-temperature spectra, five or six are necessary to describe the low-temperature ones, in spite of the fact that the linewidths do not show a substantial decrease upon cooling (see Table I). In contrast, previous absorption measurements[14] in this sample showed only two sharp lines which could be consistently assigned to allowed transitions between heavy- and light-hole states to the only confined conduction-band state for a band-offset parameter $Q=\Delta E_c/\Delta E_g=0.57$. It could be argued that the electric-field modulation produced by the photoinjected carriers brings out forbidden transitions not observed in the unmodulated spectrum. In order to test this hypothesis we compared the energy position of the lines observed in the 77 K spectra with all possible allowed and forbidden transitions (both at the minizone center, Γ point, and edge, π point) in this energy region. These transitions are calculated with an envelope-function formalism[14,15] as a function of the band-offset parameter Q. This comparison is shown in Fig. 2, where straight lines are drawn for each value of E_n in Table I and curved lines represent intersubband transitions calculated for several values of Q. We see that the number of observed lines exceeds the predictions of the theoretical model and that no consistent assignment can be done in this way for any value of Q.

An alternative interpretation can be made by regarding the features observed in the spectrum as oscillations related to the fundamental absorption edge (E_0, transition from the first heavy-hole level to the only conduction-band level at the minizone center) produced by the presence of an electric field built into the sample. In order to consider this possibility we reproduce in Fig. 3 [curves (a) and (b)] the relevant portion of the low-temperature

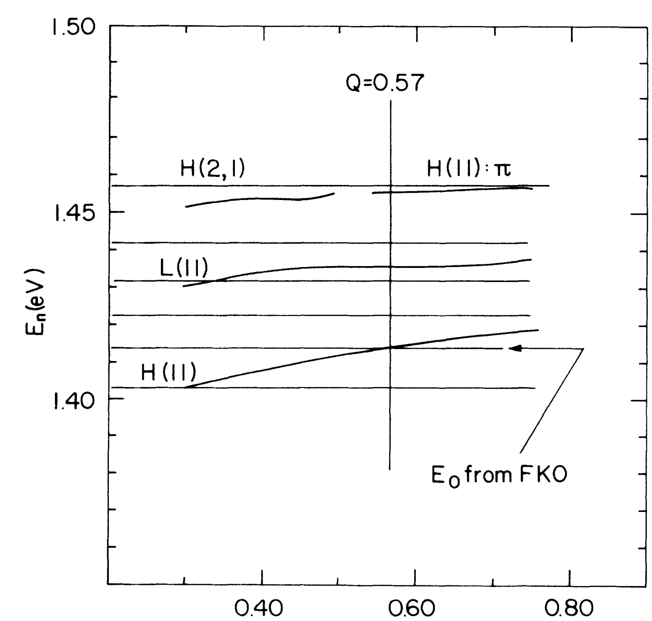

FIG. 2. Comparison between the calculated transition energies for all possible allowed and forbidden transitions (in the energy interval of interest) as a function of the band-offset parameter Q (thick curved lines), and the transition energies (mean value of $\Delta R/R$ and $\Delta T/T$) of Table I for 77 K (straight lines). The arrow indicates the position of the extrapolated value for E_0 from Fig. 4(a).

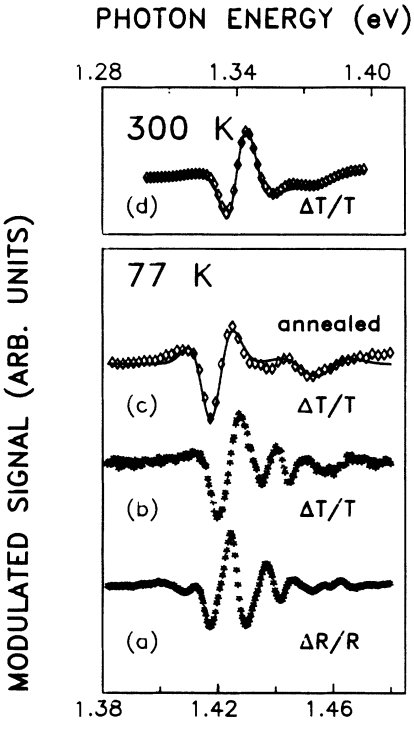

FIG. 3. Detail of $\Delta R/R$ and $\Delta T/T$ spectra. Solid lines in curves (c) and (d) are least-squares fits to standard modulation spectroscopy line shapes with two transitions, whose parameters are listed in Table I.

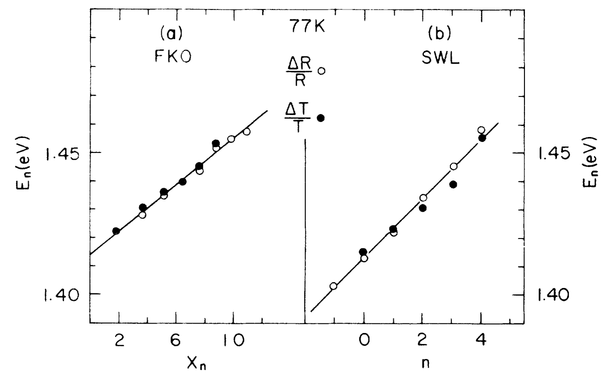

FIG. 4. Plots of the linear relationships of Eqs. (3) and (4).

reflectivity and absorption spectra. First we observe that the shape of the spectra is identical to that obtained for a single transition at a three-dimensional (3D) singular point in bulk GaAs when Franz-Keldysh oscillations are present.[6] With this interpretation, the extrema of the oscillations can be numbered in consecutive order starting from the exponential tail below E_0 and the energies of these extrema (E_n, $n \geq 1$) should scale as[6]

$$E_n = E_0 + \hbar\Omega X_n \quad (n = 1,2,3,\ldots) , \tag{3a}$$

with

$$\hbar\Omega = (e^2F^2\hbar^2/8\mu)^{1/3} \tag{3b}$$

and

$$X_n = [3\pi(n - \tfrac{1}{2})/2]^{2/3} . \tag{3c}$$

In the above equations e is the electronic charge and μ is the reduced effective mass.[19] The linear relationship between E_n and X_n predicted by Eqs. (3) is indeed obeyed by our data, as shown in Fig. 4(a). Here open (solid) circles are extrema obtained from the $\Delta R/R$ ($\Delta T/T$) spectrum, while the straight line is a least-squares fit to the data which gives $F = 6.7$ kV/cm and $E_0 = 1.414$ eV. This value of E_0, when inserted in Fig. 2, gives $Q = 0.57$ in excellent agreement with previous results from photoluminescence and transmission experiments.[14] The value of F, when used in conjunction with the calculated dispersion of the electronic miniband[19] ($\Delta = 38$ meV) gives $f = 0.18$, consistent with the requirements of Eqs. (1b) and (2). Thus, we can consistently interpret the low-temperature reflectivity and transmission spectra (with the exception of the $n = -1$ line in $\Delta R/R$) in terms of FKO's. The room-temperature $\Delta T/T$ spectrum is shown in curve (d) of Fig. 3. When interpreted in the same way, we can distinguish a smaller number of rapidly damped oscillations. This is not surprising, since increasing the temperature increases the lifetime broadening of the electronic state and decreases the spatial coherence length of the wave function. In this sense, increasing the temperature has the effect of localizing the electronic states and thus ultimately destroying the Franz-Keldysh oscillations. In the spectrum of Fig. 3 [curve 3(d)] only three oscillations can be clearly made out. A plot of E_n versus X_n (not shown) gives $E_0 = 1.334$ eV, in perfect agreement with the prediction of the Bastard model for $Q = 0.57$, and $F \cong 11$ kV/cm, consistent with the 77 K result. However, the localization produced by the temperature increase is so sharp that the spectrum could equally well be fitted by two TDLS with the transition energies listed in Table I, which coincide with the model calculation for the first allowed heavy- and light-hole transition to the only conduction-band level ($Q = 0.57$). Hence, the increase in the number of "lines" that is shown in the spectra upon cooling is merely due to an increase of the spatial coherence of the electronic wave function which allows the oscillatory behavior of the states, in the presence of a field $F \cong 7-11$ kV/cm, to manifest itself. An alternative form of localizing the wave function at low temperatures could be achieved by increasing the interface roughness and fluctuations in the superlattice periodicity. In order to produce this effect we subjected a part of our sample to the annealing procedure already described. The modulated transmission spectra at 77 K of this sample is shown in curve (c) of Fig. 3. A comparison of curves (c) and (d) in Fig. 3 shows that increasing the interface roughness through annealing has the same localizing effect as increasing the temperature in the original sample. Thus, the spectrum can be easily fitted by two transitions with Gaussian profiles[18,20] [solid curves in Fig. 3(d)] whose parameters are listed in Table I. These lines are easily interpreted as the first allowed transitions from heavy and light holes to the only conduction-band level and agree with our model calculations for this system.[21] On the other hand, the three identified oscillations in the spectra yield $E_0 = 1.409$ eV and $F \cong 11.6$ kV/cm, when plotting E_n versus X_n (not shown). Hence, all our results are consistent with the presence of a built-in electric field of the order of 10^4 V/cm which produces Franz-Keldysh states. These states can be localized by either raising the temperature or roughening the interfaces.

The above evidence shows that our low-temperature spectra can only be interpreted consistently by assuming the presence of an electric field of $\cong 10^4$ V/cm, which changes the nature of the original 3D-superlattice states. This field is large enough to dissociate the exciton [Eq. (2)] since $E_{exc} \cong 4$ meV. On the other hand, the resultant reduced field ($f \cong 0.2$) is small enough as to be compatible with Franz-Keldysh states [Eq. (1b)]. Also, the general shape of the spectra [curves (a) and (b) of Fig. 3] and the linearity of our E_n versus X_n plot [Fig. 4(a)] lead us to believe that these states are in the Franz-Keldysh regime. Still, how can we be sure that the observed oscillations are FKO's and not the product of Stark-ladder formation? After all, Stark-ladder formation has been reported in $Al_{1-x}Ga_xAs/GaAs$ superlattices even at such low effective fields,[8-11] and in $In_{1-x}Ga_xAs/GaAs$ samples[16] for $f \cong 0.4$. In this case, each of the transitions obtained from the TDLS fit (Table I) would be a member of a ladder given by

$$E_n = E_0 + neFD \quad (n = 0, \pm 1, \pm 2, \ldots) . \tag{4}$$

In Fig. 4(b) the values of E_n from Table I are plotted against n, also resulting in a reasonably linear behavior. The slope of the straight line of Fig. 4(b) yields $F = 10.4$ kV/cm, in consonance with the field values obtained from the previous interpretation. This line extrapolates to $E_0 = 1.413$ eV in agreement with the model calculation for the zero-field superlattice absorption edge for $Q = 0.57$. This apparent agreement is rather a negative argument since the E_0 of Eq. (4) is not the absorption edge of a 50 Å/50 Å superlattice but that of the isolated quantum well.[8-11] The latter is 13 meV higher than the former for $Q = 0.57$, thus making the extrapolated value of Fig. 4(b) too low by this amount. Also on the negative side is the general line shape of curves (c) and (d) of Fig. 3 which are typical of FKO's in 3D material[6] and do not resemble those of previous reports on Stark ladders.[11] In particular, Eq. (4) produces oscillations on both the high- ($n > 0$) and low- ($n < 0$) energy sides of E_0 while FKO's only occur to energies higher than E_0. The transmission spectrum [curve (b) of Fig. 3] shows clear oscillations only for $n > 0$, in agreement with the line-shape interpretation in terms of FKO's. The reflectivity spectrum [curve (a) of Fig. 3], however, shows a weak oscillation for $n = -1$. This oscillation cannot be explained in terms of Franz-Keldysh oscillations, but it is consistent with Stark-ladder formation. On the other hand, it is the only (and weak, at that) oscillation for $n < 0$. In contrast, in the Stark-ladder observations of Voison et al.,[11] the $n < 0$ oscillations are the most prominent features of their electroreflectance spectra.

From the discussion above we conclude that the low-temperature modulated reflectivity and absorption spectra of our sample can only be interpreted consistently by assuming the existence of a built-in field of the order of 10^4 V/cm. This field modifies the 3D electronic states of the superlattices. The observed spectra are best interpreted in terms of Franz-Keldysh oscillations, although the existence of a below the absorption edge oscillation in the $\Delta R/R$ spectrum might indicate the incipient formation of Stark ladders. Such a transition between both regimes could also be present in previously reported modulated reflectance data on $Al_{1-x}Ga_xAs/GaAs$ superlattices.[11] However, the observation of FKO's in these materials can be obscured by similar (and stronger) oscillations from bulk GaAs present in the sample. Such an interference would be impossible in our case, since the well material ($In_{1-x}Ga_xAs$) does not appear in the bulk form anywhere in our sample. In contrast with recent measurements in the same type of materials, the miniband dispersion is sufficiently large to allow observation of FKO's for fields that satisfy Eq. (2). Therefore, we believe that the data shown here are the first clear indication of a Franz-Keldysh regime in superlattice states.

ACKNOWLEDGMENTS

This work was partially supported by the Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP), Consejo Nacional de Ciencia y Tecnología [CONACyT (Mexico)], and Third World Academy of Sciences (TWAS). We are grateful to Professor J. A. Brum, Professor P. Motisuke, and Professor E. A. Meneses as well as Dr. A. A. Bernussi and Dr. M. J. P. Brasil for helpful and illuminating discussions. Dr. Bernussi and Dr. Brasil were also helpful with the experimental setup and software.

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[18]Although the exact line shape appropriate for the description of photomodulated spectra of superlattices depends on a number of factors, TDLS is known to produce good fits and give reliable values for the transition energy. Reference 20 discusses this subject in greater depth.

[19]In the present case the bandwidths for electrons and holes are $\Delta_e \cong 38$ meV and $\Delta_h \cong 1$ meV, respectively. Thus, holes are localized by any field large enough to satisfy Eq. (2). Therefore, instead of the reduced mass we should use the electron effective mass in Eq. (3c).

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