Acoustic-phonon Raman scattering from Wannier-Stark levels in GaAs/AlAs superlattices

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We study the electronic structure of a GaAs/AlAs superlattice in an electric field by acoustic-phonon Raman scattering. Resonantly enhanced disorder-induced processes (continuous emission) occur whenever the exciting laser energy coincides with interband transitions between heavy hole and electron Stark levels with $\Delta n = 0, \pm 1$. We also observe oscillations of the scattering intensity due to the delocalization of localized excitons induced by resonant interaction either with emerging Stark states of neighboring quantum wells in the same miniband or with Stark states from the next higher electron miniband. [S0163-1829(97)07328-1]

I. INTRODUCTION

The electric-field-induced localization of extended superlattice (SL) wave functions, i.e., the Wannier-Stark (WS) effect, has been intensively studied theoretically[1,2] and experimentally by photocurrent and photoluminescence excitation spectroscopy,[3] as well as electroreflectance.[4] The influence of electric fields on Raman scattering by *optic* phonons in GaAs/AlAs SL's has been investigated before.[5] Changes in the intensities of the different confined modes,[6,7] as well as effects related to resonant tunneling, have been observed.[8]

In this paper we report on continuous emission, i.e., disorder-induced Raman scattering by *acoustic* phonons[9–11] in an electric field. For resonant excitation at interband transitions between WS levels one expects Raman spectra similar to those that have been recently observed in multiple quantum wells (MQW's) in high magnetic fields and in indirect-gap short-period SL's even without additional Landau quantization.[9–11] In these studies it has been shown that disorder causes a breakdown of crystal-momentum conservation for resonant Raman processes involving acoustic phonons. Thus, in addition to the usually observed sharp folded-phonon doublets, a continuous emission background occurs with features of the SL acoustic phonon density of states, such as dispersion gaps, superimposed. While crystal-momentum nonconservation in the studies performed so far was introduced by sample imperfections, i.e., in a random way, electric fields offer the possibility to change the localization of the wave function in a controlled manner. The continuous emission associated with acoustic phonons should therefore allow one to obtain detailed information on the SL electronic structure. The advantage of resonant continuous emission involving acoustic phonons, as compared to optic-phonon Raman scattering, is that resonances can be studied at arbitrarily small Raman shifts. Thus, individual resonances can be investigated in great detail. Also, for phonon energies comparable to the homogeneous linewidth of an electronic transition, conditions for double resonance are almost fulfilled. This leads to stronger resonances and more pronounced spectral features than for optic phonons with their much larger frequencies.

II. EXPERIMENT

Experiments were performed on the same SL as in Ref. 12. Sixty periods of (70/9) Å GaAs/AlAs are sandwiched in a pin-diode configuration between 300 Å GaAs and 5000 Å $Al_{0.5}Ga_{0.5}As$. The $Al_{0.5}Ga_{0.5}As$ layers are *n* type on the substrate side and *p* doped on the top side of the device, respectively (for other details see Ref. 12). Measurements were performed in a closed-cycle refrigerator cryostat at 15 K. Resonant Raman spectra were excited by a Ti-sapphire laser pumped by an Ar^+ laser. The excitation power did not exceed 1–2.5 W cm^{-2}. Spectra were recorded with a Spex 1404 double monochromator using single-photon counting and a cooled GaAs photomultiplier.

III. RESULTS AND DISCUSSION

In the flat band regime where the built-in electric field is just compensated by an external applied field *F* the photoluminescence spectrum of the sample shows a peak at 1.603 eV [''LE'' in the inset of Fig. 1(b)] with a full width at half maximum (FWHM) of 6 meV. It is due to luminescence of quasi-two-dimensional excitons localized (LE) by interface roughness and layer thickness fluctuations. In previous photocurrent measurements transitions between the lowest hole and electron minibands were found between 1.610 and 1.636 eV.[12] In order to observe continuous emission resonances with WS states we proceeded in the same way as in our previous study using Landau levels in high magnetic fields.[9] For each laser energy the spectrometer was set as a spectral bandpass to a fixed Stokes Raman shift of 6 cm^{-1}. Under these conditions we measured the scattered light intensity versus electric field. Such electro-Raman profiles are sensitive to all electronic states for which incoming or outgoing resonance conditions of Raman processes are fulfilled.

A. Wannier-Stark ladder and high-field resonances

Figure 1 shows typical electro-Raman profiles measured at different excitation energies with respect to the lowest miniband transition. The peak labeled $n = +1$ in Fig. 1(a) moves towards higher fields with increasing excitation en-

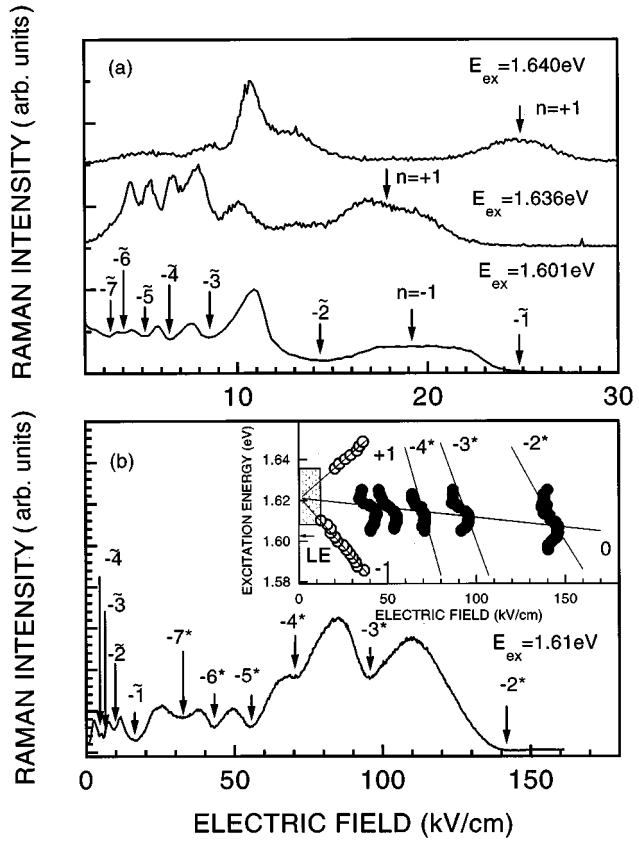

FIG. 1. Electro-Raman profiles of a GaAs/AlAs SL for different laser energies E_{ex}. The inset in (b) is a fan plot of the various spectral features; the dashed region shows the miniband range. See text for details.

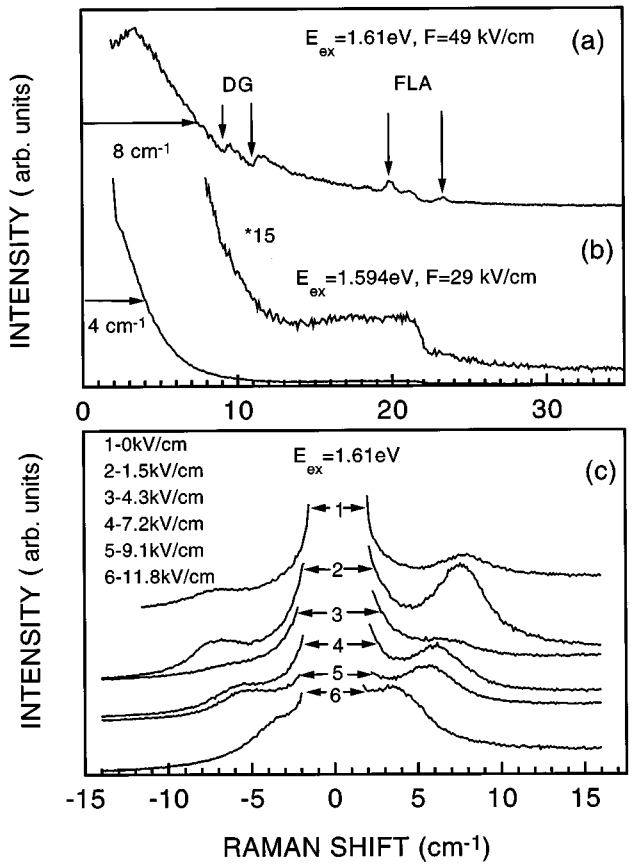

FIG. 2. Raman spectra excited in resonance with (a) the $(n=0)$ and (b) $(n=-1)$ WS level transitions. (c) Stokes/anti-Stokes (positive/negative shifts) Raman spectra in the range where the resonant delocalization of localized excitons by hole states is important. See text for details.

ergy E_{ex}. A similar peak $(n=-1)$ is also observed for excitation below 1.610 eV. However, with increasing field it moves towards smaller energies. The open circles in the inset of Fig. 1(b) give the positions of these peaks for a range of laser energies. They change linearly with the field strength. Their slopes are $+0.76$ meV/(kV/cm) and -0.94 meV/(kV/cm), respectively. The solid lines through these resonances are fits to the data. We attribute these resonances to transitions between heavy-hole and electron WS states located in adjacent QW's. Resonances with $n>1$ are not observed, presumably due to decreasing wave-function overlap, which in Raman scattering enters to a higher power than in photocurrent measurements. Another reason could be the decreasing FWHM of continuous emission Raman spectra, which, in analogy to a single-slit experiment, is approximately inversely proportional to the localization length of the wave function.[9] Therefore the signal intensity decreases at a fixed detection energy from the laser if the wave function becomes more spatially extended.

As shown in Fig. 1(b), the high-field region exhibits a series of pronounced intensity dips (indices marked by asterisks) superimposed on a broad resonance. The filled circles in the inset of Fig. 1(b) show these intensity minima versus excitation energy. They have a rather steep negative average slope and some characteristic nonlinearities that for higher fields occur at increasingly lower energies. This effect only occurs for excitation around the $(n=0)$-WS transition, which moves towards lower energies by an amount of -0.09 meV/(kV/cm) [taken from the average of the $n=\pm 1$ Stark states in the inset of Fig. 1(b)] due to excitonic effects and changes in the effective confining potential. By comparison with theoretical calculations and Ref. 12 we find that these dips occur when the $(n=0)$ state is delocalized due to anticrossings with WS levels for $n=-7--2$ (marked by asterisks) originating from the next higher electron miniband. Three of the observed resonances [$n=-2^*$, -3^*, -4^* in Fig. 1(b)] coincide with those obtained from photocurrent measurements in Ref. 12. The solid lines through these resonances in the inset of Fig. 1(b) are calculated transition energies of the higher miniband states involved. The coupling of the $(n=0)$ state with WS levels from the higher miniband facilitates electron tunneling between the wells. Consequently the lifetime decreases. This has an influence on the Raman intensity, which is also reduced.[13]

We now discuss Raman spectra measured in these regimes. For strong electric fields ($F\geqslant 20$ kV/cm) the energy separation between WS states exceeds the localization energy of electrons and the SL can be considered as a set of single QW's that are regularly spaced along the growth direction. For laser energies corresponding to the $(n=0)$ WS-level transition we observe spectra that are very similar to those obtained for MQW's under resonance excitation of excitonic states.[9] The spectrum of Fig. 2(a) consists of a broad

continuous emission background with pronounced intensity anomalies at folded-phonon dispersion gaps (DG, indicated by arrows).[10] It also shows sharp lines that correspond to folded-acoustic-phonon doublets (FLA, indicated by arrows). For excitation with laser energies corresponding to the $(n=\pm 1)$ WS state transitions, such as the $(n=-1)$ spectrum of Fig. 2(b), we also observe a continuous emission background but no folded-phonon doublets. Only a broad structure with a kink occurs in this range. This feature does not change with electric field. Its origin and possible relation to the phonon density of states or to coherence effects of the wave functions from neighboring QW's involved in the $(n=-1)$ resonance remains to be clarified. The width of the continuous emission background in the two spectra of Figs. 2(a) and 2(b) is quite different. While a FWHM of 8 cm^{-1} is observed in Fig. 2(a), that of the spectrum in Fig. 2(b) is only 4 cm^{-1}. Due to the strongly increasing laser background at small Raman shifts the maximum of the spectrum in Fig. 2(b), which needs to be known in order to determine the FWHM, only appears as a kink around 2.5 cm^{-1}. The smaller FWHM of the $(n=-1)$ spectrum reflects an effective increase of the QW width at $(n=-1)$ resonances due to the larger wave-function delocalization as compared to the $(n=0)$ case. In this case $(n=0)$ the dominant contribution to the scattering comes from wave functions that extend over one well only.

B. Low-field resonances

Characteristic Raman intensity oscillations, such as those below 15 kV/cm in Fig. 1, are observed for excitation energies of up to 20 meV above as well as below the lowest heavy-hole and electron miniband gap. The electric fields at which these resonances occur depend only slightly on the excitation energy.

In bulk GaAs it has been shown that excitons with a binding energy of $E_{ex}=4.2$ meV are easily ionized by rather small electric fields of about $F=1$ kV/cm.[14] In an electric field the probability of exciton ionization is determined by the asymmetric lowering of its Coulomb potential and varies monotonically with increasing field strength. In a SL the situation is more complicated because of the excitonic continuum in the miniband, which consists of a large set of extended wave functions. An electric field splits this quasicontinuum into a discrete set of WS states centered in the constituent single QW's of the SL. These WS states nevertheless extend over a few periods and can thus couple with other states located away from a particular well. This coupling becomes very important for levels with energies close to each other.[15] The interaction of the electron or hole components of localized excitons with WS states of different single QW's leads to resonant tunneling effects. We conjecture that such resonances reveal themselves in oscillations of the Raman efficiency as well as in the photoluminescence intensity. We interpret the low-field behavior of the spectra in Fig. 1 as resonant delocalization of excitons localized by interface roughness due to their coupling to miniband exciton states. These effects have been studied theoretically for SL excitons in Ref. 16.

In the electro-Raman spectra of Fig. 1 we observe a series of minima between 3 and 25 kV/cm (indices marked by tildes), which we attribute to the coupling of localized excitons with states in up to seven neighboring QW's. In contrast to the direct continuous emission resonances discussed above, the influence of higher-index states is observed here since they only modulate the strong $(n=0)$ excitonic WS state. The dips labeled $n=-\tilde{1}-\tilde{7}$ in the 1.601-eV spectrum of Fig. 1(a) occur at 25, 14.5, 8.5, 6.5, 5.1, 4.0, and 3.2 kV/cm, respectively. With the center of the miniband located at 1.623 eV, WS level resonances at this excitation energy are calculated [from $E=neFd$, using experimental slopes of -0.94 and -0.09 meV/(kV cm) for the $n=-1$ and $n=0$ Stark states] to occur at 25.9, 12.9, 8.6, 6.5, 5.2, 4.3, and 3.7 kV/cm, in good agreement with the experiment. Note that the intensity dip due to interaction of the localized exciton with the $(n=-\tilde{1})$ WS state occurs at a larger field than the continuous emission maximum $(n=-1)$ originating from the mechanism involving only miniband WS states discussed in Sec. II A. As can be seen from Fig. 1(a) this resonance occurs for 1.601 eV at about 18 kV/cm while the $(n=-\tilde{1})$ minimum is located at 25 kV/cm. We attribute this difference to changes of the LE energy with electric field due to the transition from the quasi-three-dimensional SL to the two-dimensional QW regime.[17]

A series of Stokes and anti-Stokes spectra for different electric fields is shown in Fig. 2(c). These spectra were excited at 1.610 eV, i.e., in the range where localized excitons are important as intermediate states of Raman processes. The flat-band Raman spectrum consists of a weak background and a broad line at 8.5 cm^{-1} with FWHM of 3 cm^{-1}. With increasing electric field the latter moves to smaller Raman shifts. While the Raman shift decreases almost linearly with increasing field [slope: -0.3 cm^{-1}/(kV/cm)], the scattering intensity shows the same oscillations as the electro-Raman profile of Fig. 1(b). We therefore attribute these resonances to the influence of delocalization on the electron part of localized excitons and their lifetimes as discussed above. At zero field the Raman shift of the line corresponds to the localization energy of heavy holes due to layer thickness fluctuations which for 1 ML amounts to about 8 cm^{-1}. We therefore propose that this Raman line is due to doubly resonant acoustic-phonon scattering between coupled localized and miniband heavy-hole states. Due to the small phonon energies involved, crystal-momentum nonconserving Raman processes are resonantly enhanced and can be experimentally observed. For zero electric field the Raman shift of the line is largest and its width reflects the inhomogeneous broadening of the heavy-hole localization energy due to interface roughness. An electric field tilts the miniband with respect to localized hole states and the energy separation decreases. Consequently, phonons with smaller energies are involved in the Raman process and the Raman shift is reduced. Oscillations of the Raman efficiency for this process occur in the same range of low electric fields as in Fig. 1. Another feature of Raman spectra measured in this range of excitation energies is the absence of folded-acoustic-phonon doublet scattering for low electric fields, which reflects the localized character of the states.[11]

IV. CONCLUSIONS

Disorder-induced acoustic-phonon Raman scattering (continuous emission) can be used to study the electronic

structure of SL's in an electric field. In strong fields we find resonantly enhanced continuous emission whenever the exciting laser energy coincides with an interband transition between heavy-hole and electron WS levels. The scattering intensity oscillates due to wave-function delocalization induced by resonant interaction with WS states from the next higher electron miniband. In the low-field regime we observe intensity oscillations due to the resonant delocalization of excitons which are localized by well width fluctuations and interface roughness. This delocalization occurs when localized excitonic states are in resonance with emerging WS states of neighboring QW's in the same miniband.

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