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Triply resonant Raman scattering by LO phonons in a Wannier-Stark ladder

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We report on Raman scattering by longitudinal-optical phonons in a strongly coupled GaAs/AlAs superlattice. Applying an electric field along the growth direction, the Stark-ladder energy is adjusted to multiples of the longitudinal-optical-phonon energy in order to achieve multiple resonance conditions for the scattering processes. Double and triple resonances are investigated by excitation at suitable laser wavelengths. The triply resonant process is observed both for the case when the second intermediate state is at $k \neq 0$ in the same Stark-ladder level as the third intermediate state, as well as when the second intermediate state is located at $k=0$ in a separate Stark-ladder level.

Semiconductor superlattices (SL's) have proven to be interesting systems both for the investigation of transport and band-structure phenomena[1-5] as well as for possible device applications.[6-8] One important topic is related to the enhancement of the electron-phonon interaction for the case when the energy spacing between different quantum states of the SL coincides with optical-phonon energies. In a weakly coupled SL with period d in a longitudinal electric field F, this effect should lead to a resonant enhancement of the tunneling probability if the potential drop eFd per SL period is swept across the phonon energy. In a strongly coupled SL with period d, the minibands are broken up by the electric field into Stark-ladder states[1,2] with an energy spacing of eFd and a localization length of several SL periods.[4] To our knowledge, phonon-related structures in the transport properties have not yet been observed in the latter case. We note, however, that bulk CdS shows some conduction anomalies[9] at very strong electric fields. These anomalies might be associated with the situation that the optical-phonon energy is an integer multiple of the potential drop per CdS lattice period.[10]

Another method of investigating the electron-phonon interaction is to study the efficiency of the Raman-scattering process. Resonant Raman scattering has been used successfully to characterize the subband structure of GaAs quantum wells.[11] Double-resonance conditions can be realized if the spacing between two subbands equals an optical-phonon energy.[12] This effect has also been demonstrated by Agulló-Rueda, Mendez, and Hong[5] using the Wannier-Stark ladder of a GaAs/$Al_{0.35}Ga_{0.65}As$ SL in an electric field. Doubly and triply resonant Raman scattering by two longitudinal-optical (LO) phonons has been reported by Alexandrou, Cardona, and Ploog[13] in a 2.8 nm/2.86 nm GaAs/AlAs multiple quantum well.

In this paper we report on multiply resonant Raman scattering involving the Stark-ladder states of a strongly coupled GaAs/AlAs SL in an electric field. In the case when the Stark-ladder energy eFd equals the energy of two GaAs LO phonons, we realize triply resonant conditions where the second intermediate state is formed by a Stark-ladder state at finite momentum k. Then the second-order Raman signal is much stronger than the one-LO-phonon line. For the situation that there are three Stark-ladder states separated by one GaAs-LO-phonon energy each, we observe a resonant enhancement in both the one-phonon and two-phonon scattering intensities. For this exceptional situation all the participating states are located at $k=0$, leading simultaneously to a double resonance of the one-phonon and a triple resonance of the two-phonon Raman-scattering process.

For our experiments we use an undoped SL consisting of 40 periods of 3.2-nm GaAs and 0.9-nm AlAs. The SL is sandwiched between 800-nm-wide $Al_{0.33}Ga_{0.67}As$ window layers which are n-doped at the substrate side to 10^{18} cm^{-3} and p-doped at the top side to 10^{19} cm^{-3}. The whole n-i-p diode structure was grown by molecular-beam epitaxy on a (100)-oriented n^+-GaAs substrate and processed into mesas of 450 μm diameter. Ohmic contacts were formed by Cr/Au metallization at the top side and Au-Ge/Ni at the substrate side. The photocurrent spectra were excited using a halogen lamp and a double monochromator, and recorded by a lock-in amplifier. The Raman experiments were performed using a Ti-sapphire laser pumped by an Ar^+ laser. The polarizations of both the incident and the scattered light were parallel to the same (110) crystallographic direction. The Raman spectra were recorded by a triple monochromator equipped with an intensified Si-diode array detector. The spectral resolution was set to 6 cm^{-1}.

Figure 1 shows the exciton peak energies which were extracted from a series of photocurrent spectra obtained at different bias voltages. The transition from the miniband regime towards completely localized carrier states has been described, e.g., in Refs. 1-3. The important

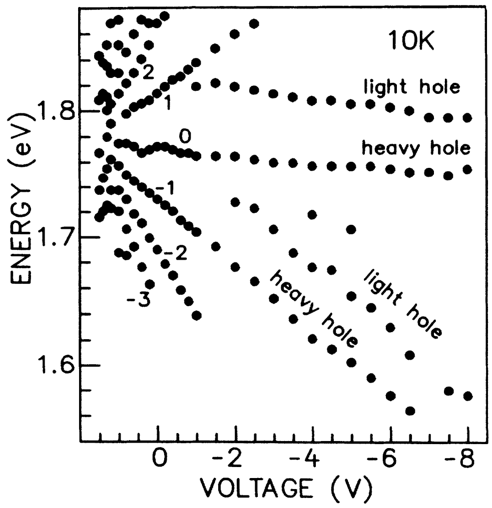

FIG. 1. Exciton peak energies as obtained from a series of photocurrent spectra vs applied voltage. Negative voltages refer to reverse direction of the *n-i-p* diode.

features with respect to the Raman spectra discussed below are as follows. The spatially direct ($n=0$) heavy-hole exciton has its peak at around 1.76 eV and undergoes a slight voltage-dependent energy shift due to a field-dependent increase of the exciton binding energy and due to the quantum-confined Stark effect. The spatially indirect transitions between $n=-3$ and $n=+2$, which are indicated in Fig. 1, depend almost linearly on the applied voltage. The energy shift relative to the $n=0$ transition is approximately given by $neFd$.[1,2] Deviations are due to the significantly larger exciton binding energy of the $n=0$ exciton as compared to the $n=\pm 1$ transitions. The light-hole transition lies about 50 meV above the energy of the respective heavy-hole exciton and is clearly resolved for $n=0$ and -1.

In our Raman experiments we use the incoming resonance of the $n=1$ heavy-hole transition and measure the enhancement of the LO-phonon Raman signal due to multiple resonances associated with the $n=0$ or -1 transitions. The schematics of the resonant configurations under study are shown in Fig. 2. Figure 2(a) illustrates doubly resonant Raman scattering as reported in Ref. 5. In this case, ingoing and outgoing resonance conditions (at the frequencies of the incident light $\hbar\omega_L$ and the scattered light $\hbar\omega_S$) are simultaneously achieved for a Stark-ladder separation equal to one LO-phonon energy $\hbar\omega_{LO}$. As indicated in Fig. 2(b), triple-resonance conditions are realized involving a second scattering event into the $n=-1$ state. On the other hand, triply resonant Raman scattering is also possible if the $n=1$ and 0 states are separated by two LO-phonon energies [see Fig. 2(c)]. In this case, the second intermediate state is represented by the $n=0$ level with an appropriate kinetic energy (at finite momentum $\mathbf{k}$). Referring to the respective $\mathbf{k}$ values of the second intermediate state, the situation shown in Fig. 2(b) will be called in the following "direct" whereas the triple resonance displayed in Fig. 2(c) is called "indirect." Doubly resonant and *indirect* triply resonant Raman scattering only requires two intermediate states with a certain energy separation and can therefore be studied, e.g., in quantum wells of appropriate well width,[12,13] provided that the region of the energy $\hbar\omega_s$ of the scattered light is not masked by photoluminescence. *Direct*-triple-resonance conditions, however, can only be achieved with three equidistant energy states. We note that, obviously, direct-triple-resonance conditions [Fig. 2(b)] also give rise to contributions from the indirect triple resonance [Fig. 2(c)] occurring simultaneously.

Experimental evidence of direct triply resonant Raman scattering by LO phonons is shown in Fig. 3 where we have plotted a series of Raman spectra recorded at different electric fields. In these experiments, the laser power was 10 mW focused to a spot size of $\approx 50\ \mu$m, resulting in a photocurrent of about 0.6 mA. The electric field is calculated assuming a built-in voltage of 1.65 V and a thickness of the whole SL of 164 nm. We also take into account a voltage drop of 0.7 V at some series resistance as determined from a comparison between peak positions of photocurrent-voltage curves at different illumination intensities. The field dependence of the Ra-

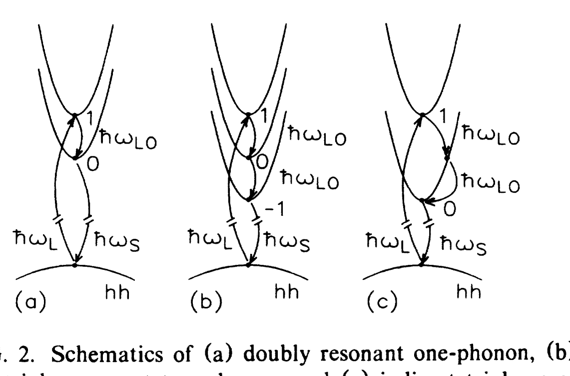

FIG. 2. Schematics of (a) doubly resonant one-phonon, (b) direct triply resonant two-phonon, and (c) indirect triply resonant two-phonon Raman scattering.

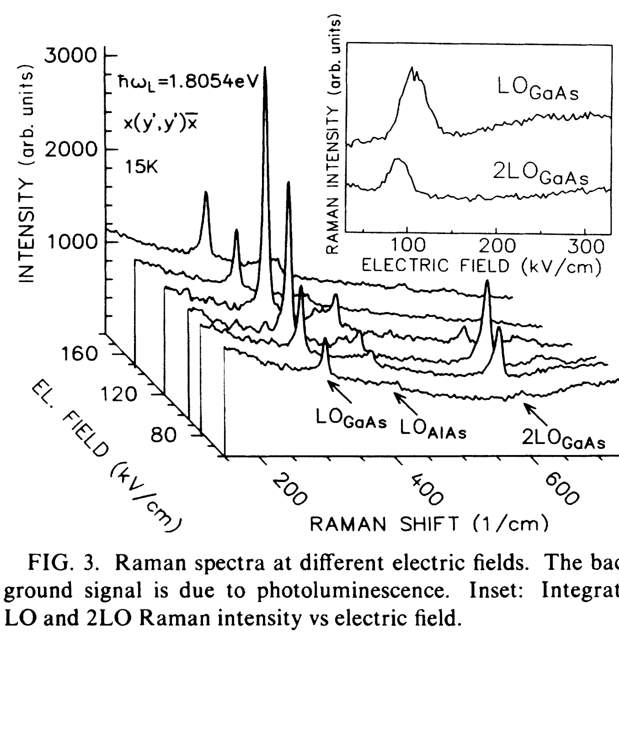

FIG. 3. Raman spectra at different electric fields. The background signal is due to photoluminescence. Inset: Integrated LO and 2LO Raman intensity vs electric field.

man intensity (inset of Fig. 3) reveals a resonant enhancement of the one-phonon (LO_{GaAs}) and the two-phonon ($2LO_{GaAs}$) signals which are peaked at around 110 and 90 kV/cm, respectively. The apparent shift between these two resonance peaks depends on the specific laser energy. A discussion of the Raman intensity and of this shift is given below.

Indirect triply resonant Raman scattering by LO phonons is studied using electric fields where eFd approaches two phonon energies. An example is plotted in Fig. 4. Under these conditions, the triply resonant $2LO_{GaAs}$ signal is much stronger than the one LO_{GaAs} signal which only shows an incoming resonance. We point out that the present labeling of the phonon modes is in analogy to the LO-phonon modes in the corresponding bulk materials, ignoring effects of Brillouin-zone backfolding.

In the energy range of the AlAs LO phonon, in addition to the first even backfolded LO-phonon mode, scattering by interface modes is observed at somewhat lower energies.[14] Figure 3 reveals that these AlAs phonon modes can equally be resonantly enhanced like the GaAs modes. Moreover, the data of Fig. 4 also demonstrate the strong enhancement of the GaAs+AlAs two-phonon structure due to the nearby indirect triple resonance.

The enhancement of the two-phonon signal under triple

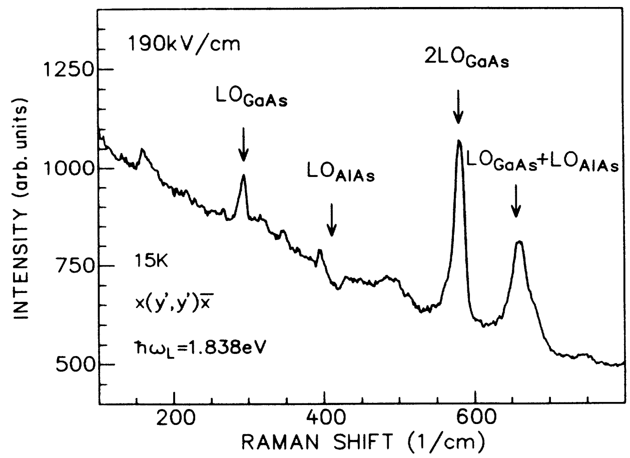

FIG. 4. Raman intensity vs energy shift for indirect triple resonance conditions.

resonance conditions is attributed to the Fröhlich interaction.[13] The scattering amplitude of the iterated electron–one-phonon process has been calculated in perturbation theory.[13,15] For the direct and indirect triple resonances, respectively, the result is

$$\frac{\langle g|H_{e\text{-}r}|-1,0\rangle\langle -1,0|H_F|0,0\rangle\langle 0,0|H_F|1,0\rangle\langle 1,0|H_{e\text{-}r}|g\rangle}{(\hbar\omega_S-E_{-1}+i\Gamma_{-1})(\hbar\omega_L-\hbar\omega_{LO}-E_0+i\Gamma_0)(\hbar\omega_L-E_1+i\Gamma_1)} \tag{1}$$

and

$$\frac{\langle g|H_{e\text{-}r}|0,0\rangle\langle 0,0|H_F|0,k\rangle\langle 0,k|H_F|1,0\rangle\langle 1,0|H_{e\text{-}r}|g\rangle}{[\hbar\omega_S-E_0+i\Gamma_0][\hbar\omega_L-\hbar\omega_{LO}-E_0-\Delta E_0(k)+i\Gamma_0][\hbar\omega_L-E_1+i\Gamma_1]} \tag{2}$$

with the electron-radiation Hamiltonian $H_{e\text{-}r}$, and the Fröhlich Hamiltonian H_F. $|g\rangle$ and $|n,k\rangle$ are the wave functions of the ground state and of the Stark-ladder state with $n=1,0,-1$ and momentum **k**, respectively, and $E_n(n=1,0,-1)$ are the energies of the respective transitions involving the first heavy-hole subband. $\Delta E_0(k)$ is the momentum dispersion of the $n=0$ electron state. Γ_n stands for a phenomenological energy broadening of the resonance denominators. For simplicity, we omitted the influence of the light-hole transitions which should be included in a more precise model.

Concerning the experiments presented here, the following conclusions can be drawn from the above expressions and their one-phonon counterparts. First, for the direct triple resonance, expression (1) allows the possibility that the doubly resonant one-phonon signal has its maximum at a slightly different electric field and/or laser energy than the two-phonon process. This is the case if the laser energy is not exactly at the resonance or if the subbands are not exactly equidistant (see Fig. 3). Second, the multiplicative structure of the resonance denominator in expression (1) explains the observation that the measured width of the two-phonon resonance profile as plotted in the inset of Fig. 3 is somewhat smaller than the width of the one-phonon resonance.

In principle, Wannier-Stark ladders may be well suited to generate higher-order multiple resonance conditions. However, one limitation is related to the fact that the oscillator strengths of the $n\neq 0$ transitions drastically decrease if $neFd$ becomes too large.[3] Moreover, the Raman efficiency for multiple-phonon scattering involving more than three LO phonons scales with the third power of the Fröhlich coupling constant[16] resulting in an additional reduction of the signal. The higher-order Raman signals might be strongly enhanced by applying a magnetic field along the axis of the SL, as has been demonstrated recently for bulk GaAs.[17]

In summary, we have reported the first study of triply resonant Raman scattering by LO phonons in a Wannier-Stark ladder. For the GaAs/AlAs SL studied, we were able to produce two, conceptually different, triple-resonance conditions involving two or three Stark-ladder states, respectively, where the second intermediate state is (a) at zero momentum **k**, parallel to the SL layers or (b) at finite **k**, away from the center of the Brillouin zone. In principle, this difference can be exploited to measure the **k** dependence of the electron-phonon interaction in SLs.

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