Standing Optical Phonons in Finite Semiconductor Superlattices Studied by Resonant Raman Scattering in a Double Microcavity

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We report optical double resonant enhancement of Raman scattering in a new double microcavity geometry. The design allows almost backscattering geometries, providing easy access to the excitations' in-plane dispersion. The cavity is used to study the phonon spectra of a finite GaAs/AlAs superlattice. A new type of "standing optical vibration" is demonstrated involving the GaAs confined phonons with a standing wave envelope determined by the superlattice thickness. A strong dispersion of the first order standing wave mode is observed, as well as its anticrossing with higher order confined modes of the same symmetry.

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The optical phonons in the polar semiconductors GaAs and AlAs form narrow bands which do not overlap. Thus, in GaAs/AlAs heterostructures the vibrations propagate in one of the constituents and not in the other. This leads to phonon confinement and strong anisotropies which arise due to the layering induced modification of the long-range electrostatic fields [1–3]. In recent years novel experimental developments in Raman spectroscopy, including micro-Raman techniques [4] and waveguide geometries [5], have provided detailed information on these excitations in superlattices (SL's). The challenge is to disclose new physics by extending the sensitivity with new techniques down to ever smaller nanostructures.

We have recently shown that 4 orders of magnitude enhanced Raman scattering efficiencies can be obtained through double optical resonances (DOR) in semiconductor microcavities [6]. These DOR are attained by tuning both the laser and scattered photons with microcavity modes at different angles. In order to study optical phonons, however, incidence angles typically larger than $\sim 50°$ have to be used. This condition limits the access to the excitations' in-plane dispersion.

In this Letter, we demonstrate a novel *double* microcavity for DOR Raman scattering enhancement at backscattering. This geometry is used to study the *finite-size* effects on the optical vibrations of GaAs/AlAs SL's made by a few periods. The SL's are limited by an AlGaAs alloy where the GaAs-like optical modes cannot propagate. We show that this confinement on the full thickness of the finite SL's results in a new type of "standing optical" vibrations. A strong dispersion and anticrossing of these standing waves with higher order confined modes of the same symmetry are observed.

Optical microcavities have received much attention recently in semiconductor science [7]. The optical field E is confined and its intensity enhanced within these structures, leading to a variety of phenomena including a modified emission rate and exciton-cavity-photon polaritons [7]. We are interested in using these modified photon spectral and spatial density of states to enhance the inelastic light scattering efficiency. Raman scattering is a second order process in light-matter interaction [8], and thus the photon-confinement efficiency enhancement is $\sigma \propto E^4$ [6]. Standard semiconductor microcavities made by enclosing a $\lambda/2$ spacer between distributed Bragg reflectors (DBR's) possess one optical mode for which E is amplified $\sim(20-30)$ times at the cavity center. Thus, amplification factors of $\sim 10^4-10^5$ can be achieved [6].

We specifically designed and fabricated a double-cavity structure for optical-phonon DOR Raman scattering experiments in a backscattering geometry. Double microcavities were previously proposed for dual-wavelength laser emission [9]. A coupled double cavity consists of two $\lambda/2$ spacers enclosed by top and bottom DBR's, and separated by a lower reflectivity mirror. Two degenerate cavity optical modes exist which are split by the mutual coupling. This splitting can be tuned with an excitation of the active material (e.g., a GaAs-like optical phonon) by selecting appropriately the middle DBR. The Stokes scattering DOR is attained by exciting and collecting the scattered light at the upper and lower cavity modes, respectively. It is important that the optical modes of the two cavities be degenerate, so that the coupled eigenmodes are distributed equivalently in the two cavities, giving the necessary overlap between the laser and Stokes fields. The double cavity described here has two $\lambda/2$ spacers made by 10.5 period GaAs(8.5 nm)/AlAs(4.4 nm) SL's, with 20 DBR $Ga_{0.8}Al_{0.2}As$/AlAs pairs on the bottom, 16 on top, and 7.5 in between. The longitudinal optical (LO) GaAs-like phonons of $Ga_{0.8}Al_{0.2}As$ are spectrally well separated from the SL GaAs-like vibrations. The structure was purposely grown with a slight taper so that the cavity modes could be tuned by displacing the laser spot on the sample surface.

The optical experiments were performed using a triple Jobin-Yvon T64000 spectrometer equipped with a N_2-cooled charge-coupled device. A Ti-sapphire laser was used as the excitation source. In Fig. 1 we show a typical reflectivity spectrum of the double cavity and the cavity modes as a function of spot position obtained from luminescence measurements at 77 K. Two optical modes (1 and 2) are clearly observed. Their splitting across the structure is almost constant and about 32–33 meV, slightly less than the GaAs LO phonon energy. The average energy can be tuned over a large energy range. The DOR Raman scattering experiments we report were performed well within the transparency region (~80 meV below the exciton energy X), where the modes are purely optical. The amplification of specific features in the spectral region of the GaAs-like optical phonons (34–36 meV) is accomplished by slightly adjusting the incidence angle ($\theta_0 \sim 2°–5°$) [6]. Once the DOR is attained, the in-plane dispersion of the excitations can be followed by turning the sample with respect to the incidence (and collection) angle, and slightly displacing the laser spot on the sample for retuning. Because of the backscattering geometry and the huge DOR enhancement (which for this cavity is $\sim 10^4$, and independent of angle), the spectra can be followed without any difficulty up to $\theta_0 \sim 80°$.

Spectra for varying sample angles (that is, transferred in-plane wave vector $k_\parallel$) are displayed in Fig. 2. The main observations that can be drawn are as follows: (i) For the smallest $k_\parallel$ (topmost spectrum) basically one split Raman peak is observed at the GaAs-bulk LO energy ~295 cm^{-1}, plus a broad and asymmetric feature at ~286.5 cm^{-1}. (ii) As $k_\parallel$ increases the split mode develops into two intense separate peaks. One component remains basically unaltered, while the second displays a strongly dispersive behavior. (iii) As a function of increasing $k_\parallel$ the dispersive component decreases in energy and generates a complex anticrossing behavior with a series of smaller peaks appearing at lower energies. The transfer of intensity between the two peaks involved in each anticrossing is a clear signature of mode-coupling. The energies of the peaks displayed in Fig. 2 are shown in Fig. 3.

In order to analyze these figures we recall established results on optical phonons in GaAs/AlAs SL's [10]. These phonons are confined to individual layers, this confinement being characterized by the integer mode order m [1–3]. The energies of the confined modes, plotted against their effective wave vector $k_m = m\pi/(n+1)a$, map the bulk dispersion. Here n is the number of GaAs monolayers of thickness a. The $m = 1$ mode carries a macroscopic polarization and thus disperses as a function of $k_\parallel$. This results in the anticrossing with the other odd order confined modes, which are almost dispersionless [1–5]. In an infinite SL the $m = 1$ modes form a continuous band corresponding, for a given $k_\parallel$, to Bloch-like states with k_z in the range $0 < k_z < \pi/d$ (d is the SL period) [1]. The bottom (top) edge of this band corresponds to the most (less) dispersive mode with $k_z = 0(\pi/d)$.

The backscattering Raman spectra of standard GaAs/AlAs SL's grown on [001] oriented GaAs substrates display a series of confined phonon peaks with intensity falling as $1/m^2$ [1–3]. These peaks are basically insensitive to the incidence angle. Because of the large refractive index $n \sim 3.5$, for this geometry $k_z \gg k_\parallel$ and thus the

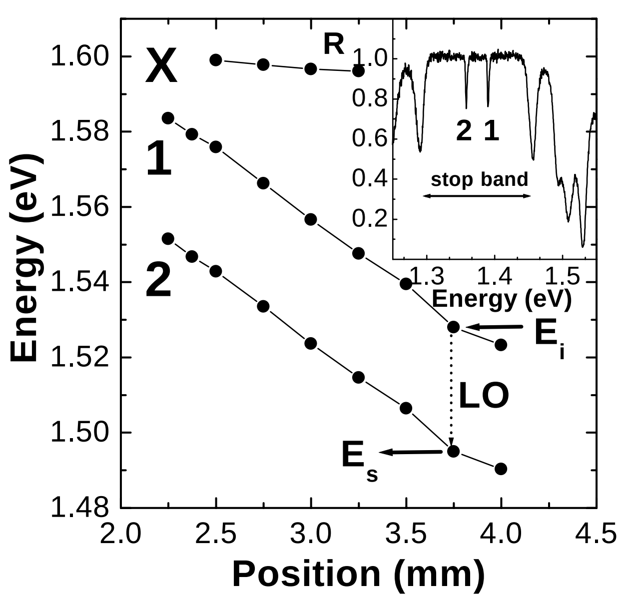

FIG. 1. Luminescence measurements at 77 K. X, 1, and 2 label the exciton, and the upper and lower cavity modes, respectively. The DOR Raman scattering by an LO phonon is also schematized. Inset: room temperature reflectivity.

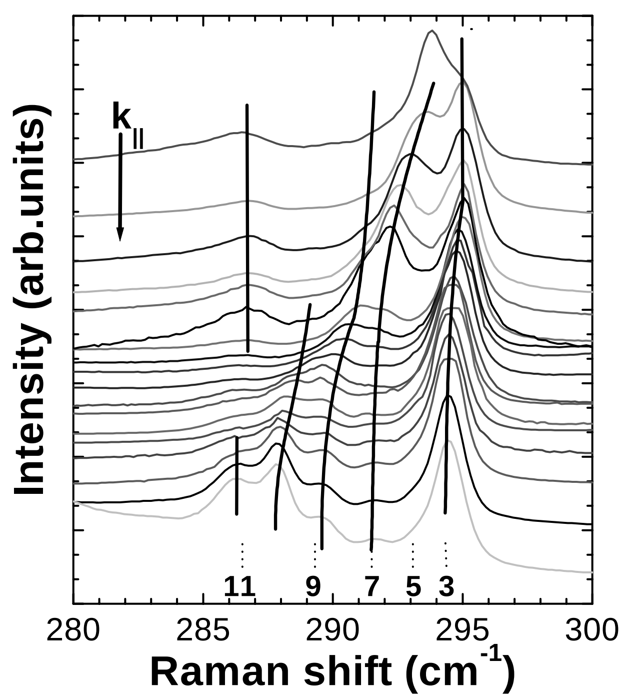

FIG. 2. Raman spectra taken under DOR conditions for varying $k_\parallel$. The solid lines are guides to the eye. The numbers 3 to 11 indicate the energy of the m confined phonons.

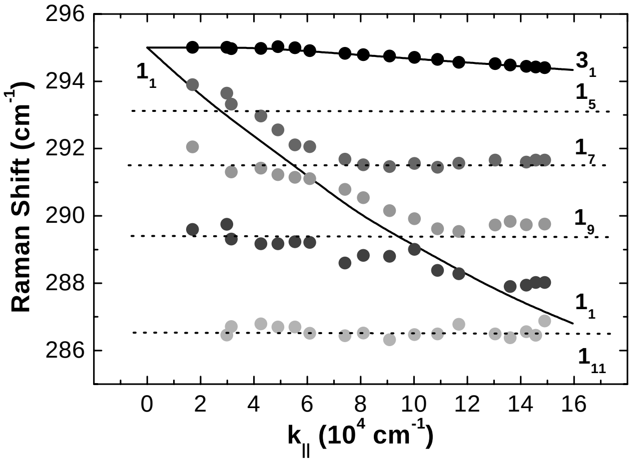

FIG. 3. Phonon $k_{\|}$ dispersion. The dashed lines indicate the m confined phonon energies. The solid lines were obtained from the dielectric model discussed in the text. N_m labels the standing wave N, confined mode m.

internal incidence angle is always smaller than $\sim 15°$. In order to observe any dispersion of the $m = 1$ mode, relatively large $k_{\|}$'s have to be transferred. This has been best accomplished previously through in-plane backscattering micro-Raman geometries [4]. In these experiments the in-plane wave vector was varied by carefully polishing the SL side face at different angles. The spectra show that, as predicted by theory, with increasing $k_{\|}$ the most intense $m = 1$ mode disperses and anticrosses with the other odd order confined modes. For large $k_{\|}$'s only a very weak $m = 3$ mode remains close to ~ 295 cm^{-1} [4].

The general picture described above, i.e., a dispersive mode and anticrossings with almost $k_{\|}$-independent vibrations, resembles the behavior displayed by the peaks in Figs. 2 and 3. In fact, the position of the small peaks that anticross with the dispersive mode matches closely the energies of the $m = 5$, 7, 9, and 11 confined GaAs-like vibrations calculated for our SL. However, notwithstanding the overall resemblance, two important facts indicate that such direct interpretation is inappropriate. First, considering that our experiments were performed from the SL [001] face the measured dispersion is huge. Second, the line which for large $k_{\|}$'s lies close to ~ 295 cm^{-1} is too intense to be assigned to the $m = 3$ confined vibration.

In order to shine light on these issues we have performed calculations of the phonon spectra of the double microcavity. For this purpose we have used a matrix-method-based dielectric continuum model specially adapted to complex multilayer structures [6]. It is important to keep in mind that this model *does not* include the mechanical boundary conditions and thus fails to account for the phonon confinement [1,2]. Only the $m = 1$ vibrations come out from the calculations. Notwithstanding this limitation, the model provides a rather good description of the macroscopic field effects on these modes because the related atomic displacements are nonzero until very close to the interfaces [1]. In addition, the displacement patterns of the modes can be easily obtained. We show in Fig. 4 the calculated double cavity $k_{\|}$ dispersion. We also present the displacement patterns along z for some selected vibrations. For presentation clarity, the patterns shown were obtained for a simplified double cavity with the same number of quantum wells but reduced DBR layers. The GaAs-like DBR alloy vibrations constitute the dense discretized modes occupying the spectral region between 272 and 287 cm^{-1}. The air-DBR and sub-DBR modes correspond to vibrations localized at the air-DBR and DBR-substrate interfaces, respectively. The cavity-mode field in these regions is small, and thus no sizable scattering from such modes is to be expected. In the spectral region corresponding to GaAs-like LO SL phonons, on the other hand, several modes are observed which we have labeled with the integer number $N = 1, 2, 3$. N defines the order of these "standing optical phonons" (SOP), vibrations characterized by a standing wave envelope of displacements which are confined to the GaAs regions of the SL. Note that three quantum numbers are required to identify these new type of finite-size SL optical vibrations: $N_m(k_{\|})$. As expected from their macroscopic polarization, the SOP dispersion is most important for $N = 1$ and is reduced with increasing N.

Within the framework given by the above model we can understand most of the experimental results in Figs. 2 and 3. In fact the most intense peaks (one strongly dispersive, the other at ~ 295 cm^{-1}) can be assigned to the 1_1 and 3_1 SOP, respectively. Their dependence with $k_{\|}$ is well accounted for by the model (see the solid curves in Fig. 3) [11]. With increasing $k_{\|}$ the 1_1 mode softens and anticrosses with the high order confined vibrations. These modes are not included in the dielectric model. We have independently derived their frequencies using the corresponding effective wave vector k_m and the bulk GaAs phonon dispersion. The thickness of the sample was estimated from the energy of the cavity optical modes. The

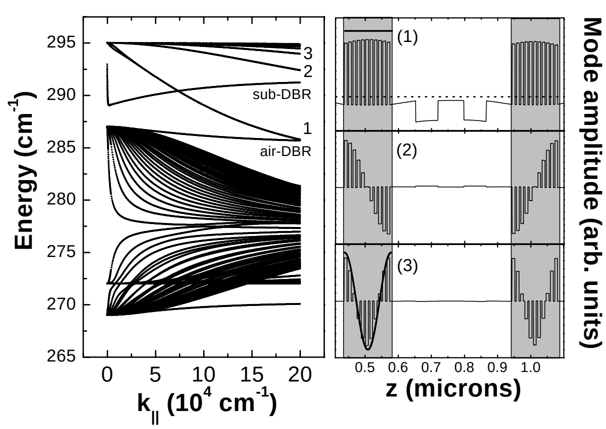

FIG. 4. Left: calculated double cavity phonon dispersion. The numbers correspond to the labels in the right panel. Right: SOP $N = 1$, 2, and 3 displacement pattern in the central region of the double cavity. The horizontal dashed line in (1) indicates zero displacement. The grey zones correspond to the SL's spacers. The thick solid lines in (1) and (3) indicate the forward and backscattering photon field distributions, respectively.

$k_{\parallel}$'s, where the anticrossings occur, are nicely reproduced by the calculations. These anticrossings with the odd-parity confined modes are expected from symmetry grounds [1–3]. The broad asymmetric feature below $\sim$287 cm^{-1} is clearly due to the GaAs-like DBR alloy vibrations. On the other hand, we believe that the peak that develops for large $k_{\parallel}$'s close to $\sim$286.5 cm^{-1} is the confined 1_{11} phonon. This assignment could explain its line shape, similar to the other confined phonons, and the increase of intensity which follows from the mixing with the 1_1 mode. The observed strong and similar intensity of the 1_1 and 3_1 modes and the nonobservation of any other SOP need to be addressed. As we will show next, this "selection rule" follows from the fact that, in a microcavity, forward and backscattering components are simultaneously detected.

The Raman efficiency can be written as [1]

$$\sigma \propto \left| \int (e^{ik_i z} + e^{-ik_i z})^* \phi_{\mathrm{ph}} (e^{ik_s z} + e^{-ik_s z}) \right|^2, \quad (1)$$

where the first and third terms describe the incident and scattered fields, respectively, and ϕ_{ph} is the displacement pattern associated with a specific phonon. Note that the photon fields are themselves standing optical waves constituted by the incident and reflected components. By construction, for the cavity optical modes $k_i = k_s = \pi/D$. Thus, Eq. (1) can be separated in the backscattering and forward scattering components $\sigma_b \propto |2 \int \cos(2\pi z/D) \phi_{\mathrm{ph}}^b|^2$ and $\sigma_f \propto |2 \int \phi_{\mathrm{ph}}^f|^2$, respectively. It follows that the phonon displacement pattern of the 3_1 mode overlaps exactly with the field distributions associated with the backscattering component $[\cos(2\pi z/D)]$. The 1_1 dispersive mode, on the other hand, presents an almost uniform displacement that matches with the *constant* forward scattering field distribution (see Fig. 4). Based on the same arguments, scattering by all other SOP is expected to display a much weaker intensity.

As a final consideration we discuss the Raman scattering in standard GaAs/AlAs SL's [1–3] in the frame of the results reported here. First, the scattering by the 3_1 SOP, which corresponds to a wave vector transfer $k_z = 2k_i$, is the standing wave counterpart of the [001]-face backscattering in standard SL's. The conceptual difference is that the SOP is a standing wave limited in space to a single Bloch wave period. Second, the "infinite" SL counterpart of the 1_1 SOP is a $k_z \sim 0$ interface phonon. The latter could be observed by forward scattering experiments along [001]. Such experiments are seldom performed, because they imply the complete etching of the GaAs substrate. To the best of our knowledge, the only such experiment has been reported in Ref. [12]. Interestingly, in the GaAs-like optical phonon region a strong line which remained unexplained was observed at $\sim$5 cm^{-1} below the LO $m = 1$ confined mode. As follows from our results here, and the calculations performed for that structure, it can be precisely assigned to the 1_1 SOP of the 590-nm-thick GaAs(1.425 nm)/AlAs(1.425 nm) SL.

In conclusion, we have observed standing optical phonons in finite GaAs/AlAs SL's. The $N = 1$ order standing wave displays a giant dispersion with the in-plane wave vector, and successively anticrosses with the odd symmetry higher order $m = 5$, 7, 9, and 11 confined phonons. The SOP dispersion is well accounted for by the dielectric model for the multilayer structure. The intensity of the observed lines, in addition, can be understood as being due to the separate contribution of backscattering and forward scattering processes. A more detailed model that takes into account the mechanical boundary conditions is, however, required to fully describe the vibrations in finite SL's. For the experiments we used the optical double resonant Raman scattering enhancement of a novel double microcavity geometry. Besides the $\sim 10^4$ efficiency amplification, this structure allows backscattering geometries and the mapping-out of the excitations in-plane wave vector dispersion. In addition, in a natural way both forward and backscattering simultaneously contribute to the collected spectra.

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[10] Note that the observed phonons are solely a property of the finite SL's located at the cavity spacers.

[11] The observed softening of the 3_1 phonon amounts to $\sim$0.5 cm^{-1}. This is within the experimental indetermination of our system, resulting in unwanted scatter in the mode dispersion data. We have thus slightly shifted in energy the data to coincide with the calculated 3_1 curve. The soundness of this correction is demonstrated by the resulting reduced scatter and zero dispersion of all confined modes.

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