Effect of dispersion of ''vertically'' polarized collective plasmon-LO-phonon excitations on Raman scattering of strongly coupled GaAs/AlAs superlattices

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The coupled plasmon-LO phonon collective excitations polarized normal to the layers were studied by Raman scattering in doped GaAs/AlAs superlattices with broad minibands. Different asymmetries of the Raman lines showing a different character (plasmon or phonon) of the coupled modes were found in accordance with theoretical predictions in samples with different electron densities. A strong influence of the superlattice miniband structure on the localization of the collective excitations was observed. Different strengths of the localization were found for the collective modes with different characteristical frequencies.

The physics of the collective plasmon and coupled plasmon-LO phonon excitations in semiconductor superlattices (SL's) attracted much attention in the last years. A rich spectrum of collective excitations has been found theoretically; among them are the quasi-two-dimensional (intrasubband) and the intersubband plasmons, their counterparts arising due to the coupling of plasmons with optical phonons and the superlattice plasmons (polarized normal to the layers), which appear in systems of strongly coupled quantum wells (superlattices).[1–3] Experimental works were devoted to study the energy spectra of the intrasubband[4,5] and intersubband[6] plasmons, and the interaction between them.[7] The superlattice plasmons at the center of the Brillouin zone (BZ) have been investigated in.[8] As a result, the dispersion of the two-dimensional plasmons has been well studied, while, to our knowledge, to date no experimental evidence of the dispersion of the superlattice plasmons was presented.

In this paper, we report on a study of the influence of the dispersion of the collective excitations propagating normal to the layers, which are caused by the coupling of the superlattice plasmons with the optical phonons on Raman spectra of the doped, strongly coupled GaAs/AlAs superlattices. The analysis of the shape of the Raman lines allowed us to identify the character of the coupled plasmon-LO phonon modes—we found that depending on electron density, the coupled modes reveal either phonon or plasmon character in good agreement with the theoretical predictions. This effect was well known in bulk-doped ionic crystals from theoretical considerations for many years (see for instance Ref. 9), however, so far it was not verified experimentally. Finally, we demonstrated the localization of the plasmon-LO phonon excitations caused by impurity scattering.

The samples here studied were strongly coupled $(GaAs)_{17}(AlAs)_2$ superlattices (the numbers indicate the thicknesses of the corresponding layers expressed in monolayers) doped with Si. The total number of 20 periods was grown by molecular beam epitaxy (MBE) on (100) GaAs substrates. The electron energy spectrum of these superlattices was computed by the envelope function approximation[10] with the barriers modified by the segregation. As it has been shown in[11] the nominal 2 ML-wide AlAs barries convert to the $Al_xGa_{1-x}As$ barriers with an average $x \simeq 0.4$ and with an effective thickness about 4 ML. These parameters of the barriers were used in the calculations. The results revealed a broad lowest miniband (with the width about 70 meV) formed by the Γ-Γ electron transfer; the complete occupation of this miniband demands an electron density $N = 4.5 \times 10^{18}$ cm^{-3}.

Backscattering unpolarized Raman spectra were performed at $T = 10$ K with a Jobin-Yvon double grating spectrometer supplied with the usual photocounting system; the 5145 Å line of an Ar^+ laser was used for excitation. In this case, because of the conservation of momenta of the elementary excitations involved in the Raman process, the collective plasmon-LO phonon modes polarized normal to the layers should contribute to the Raman scattering.

As it is well known, the fluctuations of the electronic potential, which occur in doped SL's preferentially because of the impurity random potential, destroy the translational invariance giving rise to a spatial coherence length of the elementary excitations (L) and, as a consequence, cause the breakdown of the Raman selection rules leading to the broadening and asymmetry of the Raman lines.[12,13] The analysis of the shape of the spectral lines allows one to determine the coherence lengths of the elementary excitations involved in the Raman process and thus, to study their localization properties. This effect has been used to study the localization of the optical phonons in semiconductor alloys.[12,13]

According to Ref. 12, the relaxation of the conservation of the crystal momentum caused by a crystal disorder can be taken into account by means of a Gaussian spatial correlation

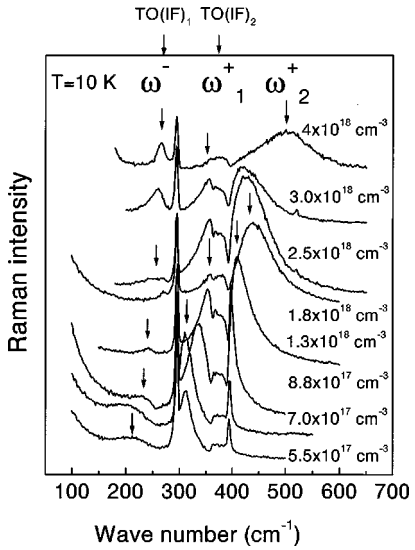

FIG. 1. The Raman spectra of the doped $(GaAs)_{17}(AlAs)_2$ superlattices. The calculated frequencies of the TO GaAs-like and AlAs-like interface modes are shown at the top of the figure.

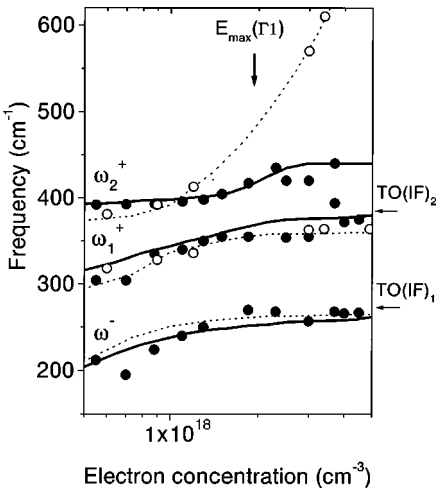

FIG. 2. The frequencies of the Raman lines measured in the $(GaAs)_{17}(AlAs)_2$ superlattices (closed circles) and the frequencies of the collective vertically polarized plasmon-LO phonon modes calculated in the $(GaAs)_{15}(Al_{0.4}Ga_{0.6}As)_4$ superlattices (full lines) with different electron concentrations. The dotted lines and the open circles show the theoretical and experimental data respectively obtained for the $Al_{0.11}Ga_{0.89}As$ alloy.

function $\exp(-4R^2/L^2)$; then the Raman efficiency can be written as

$$I(\omega)\sim\int f_{sc}(\vec{q})\cdot\exp\left(-\frac{q^2L^2}{4}\right)\frac{d^3q}{[\omega-\omega(q)]^2+(\Gamma/2)^2}, \tag{1}$$

where $f_{sc}(\vec{q})=(4\pi/q^2+q_{TF}^2)^2$ is the screening correlation function with the q_{TF} being a Thomas-Fermi wave vector; $\omega(q)$ is the dispersion of the relevant excitations and Γ is their damping constant.

As it is expected from the theoretical considerations,[14] three coupled longitudinal vibrational modes exist in the doped GaAs/AlAs superlattices: that of the low-frequency acousticlike (ω^-) and those of the optical GaAs-like (ω_1^+) and AlAs-like (ω_2^+). All these modes can be found in the Raman spectra of the studied samples, some of which are depicted in Fig. 1. Moreover, the additional modes located at 294 cm^{-1} originated from the depletion surface layer, which correspond to the longitudinal optical (LO) phonons of the wells are seen in the Raman spectra as well.

The frequencies of all the modes determined by Raman scattering are collected in Fig. 2 together with the frequencies of the vertically polarized plasmon-LO phonon modes calculated for wavevectors at the center of the BZ according to the theory presented in Ref. 14 where the finite width of the miniband was taken into account according to Ref. 15. The calculations were performed with the electron effective masses found in the studied superlattices by the envelope function approximation[10] and then averaged over the electron states which form the Fermi surface of the relevant superlattice. The results presented in Fig. 2 show good correspondence between the measured and calculated frequencies of the coupled collective plasmon-LO phonon excitations found for the superlattices under investigation.

The Raman data obtained for the bulk-doped $Al_{0.11}Ga_{0.89}As$ alloy with the contents of Al being equivalent to the SL's here studied are shown in Fig. 2 as well. The same modes as in the spectrum of the collective plasmon-LO phonon excitations of the GaAs/AlAs SL's characterize this alloy due to a two-phonon behavior.[16] The difference is expected in the dispersion of the plasma vibrations influenced by the formation of the miniband structure in SL's, which should modify the dependence of the ω_2^+ mode on electron concentration. This indeed was observed in the SL's, in good agreement with the theory, showing that the collective modes propagating normal to the layers are involved in the Raman scattering and the contributions due to the in-plane components are negligible.

As it is seen in Fig. 1, at low-electron concentrations both the ω_1^+ and ω_2^+ plasmon-LO phonon modes reveal a disorder induced asymmetry opposite to the one observed for the optical phonons in the $Al_xGa_{1-x}As$ alloys;[13] this is explained by the different dispersions of the optical phonons and of the plasmons—a negative dispersion (characterized by a negative group velocity) for the phonons and a positive one (with a positive group velocity) for the plasmons.[17] However, as the electron concentration increases, the observed asymmetry of the GaAs-like ω_1^+ coupled mode becomes less pronounced and finally changes its character at $N>10^{18}$ cm^{-3}. In the samples with such high electron concentrations the observed asymmetry of the ω^- and ω_1^+ modes are definitely caused by the negative phononlike dispersion. At the same time, the AlAs-like ω_2^+ mode reveals the asymmetry caused by the positive plasmon dispersion in all the samples under investigation.

The modification of the character of the ω_1^+ collective mode observed with increase of electron concentration is similar to the one predicted by the theory for the plasmon-LO phonon modes in bulk-doped ionic crystals.[9] According to the theory, the electron screening effects are responsible for this modification. However, the situation is somewhat different in SL's due to their anisotropy. There no longer exist bulk phonons, but instead the confined and in-

terface modes appear.[18] Now, in the presence of electrons the energy spectrum of the collective excitations is given by the zeros of the full dynamical dielectric function tensor of the SL. The component of this tensor related to the electric polarization normal to the layers (parallel to the growth direction z) can be taken in the following form:[9,18]

$$\epsilon_z(q,\omega)=\frac{d_1+d_2}{\dfrac{d_1}{\epsilon_1(q,\omega)}+\dfrac{d_1}{\epsilon_2(q,\omega)}}-v_q^{\infty}P(q,\omega), \qquad (2)$$

where d_1 and d_2 are the thicknesses of the wells and of the barriers respectively; $v_q^{\infty}=4\pi e^2/\epsilon_{\infty}q^2$ is the potential of the electron-electron interaction, $P(q,\omega)$ is the polarization function of the electron gas.

$$\epsilon_{1,2}(q,\omega)=\epsilon_{\infty 1,2}\frac{\omega^2-\omega_{L1,2}^2(q)}{\omega^2-\omega_{T1,2}^2(q)}, \qquad (3)$$

where $\omega_{L1,2}(q)=\omega_{LO1,2}(1-A_{L1,2}q^2)$ and $\omega_{T1,2}(q)=\omega_{TO1,2}(1-A_{T1,2}q^2)$ are the dispersion relations for the longitudinal optical and transverse optical (TO) phonons of the wells and the barriers respectively, with ω_{LO}, ω_{TO} being the frequencies of the LO and TO vibrations taken at the center of the BZ and with the constants $A_{L1,2}$, $A_{T1,2}$ determined according to the data presented in Ref. 19.

The first term in Eq. (2) represents the lattice contribution where the bulk-like LO phonons and the interface TO vibrations, both polarized normal to the layers are included. The second term is due to the electrons.

Two limiting cases were examined. In the low-electron density limit, when $\omega_p \lesssim \omega_{L1},\omega_{L2}$ the electron scattering is weak. Therefore, the plasmon-LO phonon modes in a narrow interval of the wavenumbers close to the center of the BZ, which are characterized by the coherence lengths $L \gg d_{SL}$ (where $d_{SL}=d_1+d_2$), are involved in the Raman process. In this case, the electron term can be taken in the random phase approximation (RPA) in the limit of small q:[9]

$$v_q^{\infty}P(q,\omega)\approx\frac{\omega_p^2}{\omega^2}\left[1+\frac{3}{5}\frac{(qv_F)^2}{\omega^2}\right], \qquad (4)$$

where $\omega_p=(4\pi e^2N/m_z)^{1/2}$ is the plasma frequency and v_F is the Fermi velocity. As a result, all the coupled modes reveal a plasmon character.

At high-electron densities, when $\omega_p > \omega_{L1},\omega_{L2}$ but L is still larger than d_{SL}, the approximation (4) is valid and the theory predicts that free electrons screen out the long-range Coulomb part of the ion-ion interaction, which is responsible for the difference between the LO and TO frequencies and therefore, the dispersions of the low-frequency coupled modes (ω^- and ω_1^+) should approximate the dispersions of the corresponding TO phonons, while the highest-frequency mode (ω_2^+) remains a plasmon character. The analysis of Eq. (2) shows that in this case, in the GaAs/AlAs SL, the ω^- and ω_1^+ coupled modes approximate the TO GaAs-like and AlAs-like inteface modes with vanishing parallel wave vectors.

With further increase of the doping level, when $\omega_p \gg \omega_{L1},\omega_{L2}$ an electron scattering may become so strong that $L \lesssim d_{SL}$; as a consequence, a significant part of the states of the BZ contribute to the Raman scattering and therefore, we need to calculate the energy spectrum of the collective excitations over all the BZ, which presents a rather hard problem. However, in this case theoretical considerations show that electrons cannot screen out the LO phonons with wave vectors close to the edge of the BZ where therefore, the LO-TO splitting appears again. As a consequence, for extremely large wave vectors, beyond the screening limit, the frequencies of the ω^- and ω_1^+ modes should approximate the frequencies of the corresponding unscreened LO phonons. We do not expect that this will cause a drastic modification of the dispersion of the collective excitations in the GaAs/AlAs SL's with ultrathin barriers (when the ratio d_1/d_2 is large) where the dispersion of the LO AlAs-like phonons is located close to the dispersion of the TO AlAs-like interface modes, while the same is true for the LO phonons of the GaAs wells with wave vectors close to the edge of the BZ. The results of the calculation of the energy spectrum of the plasmon-LO phonon modes performed with formulas (2)-(4) in the $(GaAs)_{15}(Al_{0.4}Ga_{0.6}As)_4$ SL are presented in Fig. 3. The obtained modification of the dispersions of the coupled modes is completely consistent with the modification of the Raman spectra observed in the doped GaAs/AlAs SL's with different electron densities.

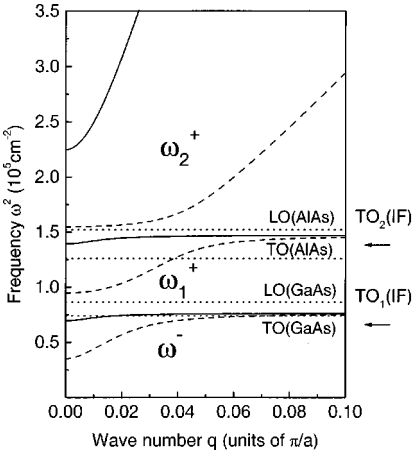

FIG. 3. The dispersions of the collective plasmon-LO phonon excitations polarized normal to the layers, calculated in the $(GaAs)_{15}(Al_{0.4}Ga_{0.6}As)_4$ superlattices with different electron concentrations: the broken lines correspond to the low electron density limit ($\omega_p=200$ cm^{-1}, $N\approx 5\times 10^{17}$ cm^{-3}), while the full lines were obtained in the high electron density limit ($\omega_p=800$ cm^{-1}, $N\approx 9\times 10^{18}$ cm^{-3}). The dotted lines are the dispersions of the corresponding lattice optical vibrations.

It should be mentioned that using the phonon contribution in the form presented in Eq. (2) we neglect the effect of the quantization of the optical phonons. This is certainly true in the case of thick enough layers (GaAs wells in the studied SL's). However, even in the four-monolayer-thick $Al_xGa_{1-x}As$ layers the continuum approximation can be used because due to the alloying the energies of the phonon states are broad and the confined modes are hardly resolved.[11] Thus, indetermination of the phonon energy in thin $Al_xGa_{1-x}As$ layers makes the phonon spectrum a quasicontinual instead of a discrete one.

The dispersions of the coupled modes obtained in this way were used in the calculations of the Raman intensities of

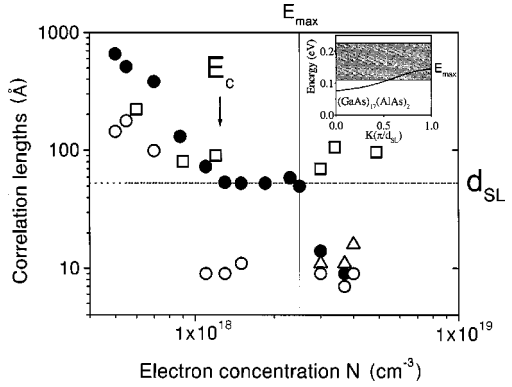

FIG. 4. The coherence lengths L (open triangles), L_1 (open circles), L_2 (closed circles) of the plasmon-LO phonon excitations $\omega^-, \omega_1^+, \omega_2^+$ respectively measured in the doped $(GaAs)_{17}(AlAs)_2$ superlattices with different electron concentrations. E_c is the mobility edge energy, d_{SL} is the superlattice period. The squares present the coherence lengths obtained in the doped $Al_{0.11}Ga_{0.89}As$ alloy.

the doped SL's which were done according to the formula (1) assuming an isotropic spatial correlation. The coherence lengths obtained by the fitting of the calculated spectra to the experimental ones are depicted in Fig. 4. The calculated electron energy spectrum of the superlattice is shown in the insertion to Fig. 4, where the shaded area corresponds to the interval of the Fermi energies relevant to the electron concentrations in the samples studied. We observed a strong decrease of the coherence lengths of the ω_1^+ and ω_2^+ coupled modes associated with a transition from a propagative regime to a localized one, which takes place when the electron density exceeds 10^{18} cm^{-3}; then L_1 and L_2 becomes comparable to or is even smaller than the superlattice period (d_{SL}). The approximate position of the mobility edge corresponding to this transition is shown by an arrow labeled by E_c.

Our results show different values of the coherent lengths associated with different coupled modes. It turned out that the low-frequency modes are localized much stronger than the high-frequency one. This result is evident because the low-frequency modes are stronger subjected to the disorder potential than those with higher frequencies.

For a comparison, the coherence lengths of the AlAs-like plasmon-LO phonon excitations obtained in the $Al_{0.11}Ga_{0.89}As$ alloy are presented in Fig. 4 as well. Significant differences between the coherence lengths measured in the alloy and in the SL's were found when the Fermi level entered a minigap of the SL's (above the energy $E_{\max}$). This shows an influence of the miniband structure of the SL's on the effect of localization of the coupled modes, which probably is caused by a ''reentrant localization'' predicted in Ref. 20.

In conclusion, we found that the asymmetrical form of the Raman lines corresponding to the plasmon-LO phonon modes observed in the doped GaAs/AlAs superlattices is caused by the dispersion of the relevant excitations (phonons or plasmons), depending on electron concentration. This finding was shown to serve as an indicator of the character (plasmon or phonon) of the coupled plasmon-LO phonon modes. We observed the change of the modes character with the increase of electron density in good agreement with the theoretical predictions. In addition, the effect of localization of the collective plasmon-LO phonon excitations was studied. We observed a transition from propagating to localized collective excitations which occurs with the increase of the doping concentrations.

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