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**LETTER TO THE EDITOR**

# The collective response of interacting localized electrons

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**Abstract.** The collective excitations in doped short-period GaAs/AlAs superlattices were studied. An extremely small broadening of the plasmon–LO phonon collective modes caused by the coherent collective response of the electron plasma was detected in an unexpectedly high range of doping levels. The observed spectral narrowing accompanied by a pronounced red-shift of the relevant Raman lines was found, in good qualitative agreement with the theoretical prediction that the cooperative behaviour of the interacting localized electrons is responsible for the established effects. The behaviour of the parameters associated with the collective excitations indicates the formation of coherent dynamic clusters similar to those known to exist in ferroelectrics. The coherent clusters were found to disappear both with the increase of doping (abruptly) and the increase of temperature (gradually).

The problem of interacting electrons subjected to a random potential, though still unsolved, is fundamental in solid-state physics. This problem involves an analysis of the interplay between disorder and interaction. Each of these two aspects considered separately presents a complicated problem, which becomes extremely hard to resolve when they are combined. However, an understanding of some features of the behaviour of the systems of interacting electrons in a disordered potential has been achieved recently thanks to the attention given to the problem of the metal-to-insulator transition discovered in 2D electron gas [1] where, as has been shown, disorder and interaction play determinative roles. Strongly interacting disordered electron systems are currently the focus of active theoretical interest [2–4].

So far, experimental studies of the localization effects of the interacting electrons have been carried out by means of measurements of the electrical conductivity [1, 5–7]. However, the collective effects discussed here determine the plasma oscillations of the electron system, which take place at the resonance frequency $\omega_p$. Therefore, it is clear that the reaction of the disordered electron system to a field of electromagnetic radiation with a frequency close to $\omega_p$ provides a straightforward way to probe the effects of interaction. From this point of view, the behaviour of the interacting localized electrons resembles the response of the bound (valence) electrons localized on the atoms forming a crystal. In spite of their strong localization, due to the strong Coulomb interaction, they reveal plasma oscillations at the resonance frequency, which depends only on the total electron density in the valence band. This is because the plasma energy is rather higher than the excitation energy of the valence electrons ($E_g$). In this way the collective interaction completely 'screens out' the influence of the crystal periodic potential, and, therefore, the valence electrons, though being strongly localized, behave collectively like free electrons. On the other hand, electrons in the cores of atoms do not contribute to the plasma vibrations because they are too tightly bound; their excitation energies are much larger than the plasma energy.

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A similar behaviour can be expected in the case of electrons localized in a random potential. In this case, a system of localized electrons can be represented by a system of randomly positioned oscillators where the contribution of each localized electron to the crystal polarization is characterized by the electron polarizability. The collective behaviour of the two-dimensional array of randomly distributed localized oscillators which are mutually coupled by the electron–electron Coulomb interaction has been considered in [8]. It was shown that neighbouring electrons mutually adopt their phases, forming coherently oscillating spatial clusters; as a result, the natural inhomogeneous broadening of a disordered system can be completely screened out by the dynamic many-particle interaction. This causes a narrowing of the absorption lines associated with the intersubband electron transitions.

Recently, the spectral properties of strongly correlated disordered 3D systems have been considered in [9] where results similar to those presented in [8] were obtained; namely, it was shown that the strong interaction causes band narrowing and a red-shift.

It is worth adding that, like the bound core electrons, localized electrons with polarization frequencies too different from the collective plasma frequency $\omega_p$ (which are strongly localized) will not contribute to the plasma oscillations. This gives rise to the effective electron density—the number of electrons contributing to the plasma vibrations.

In order to obtain these features of the collective behaviour of the localized electrons, we explored Raman scattering of the plasma oscillations of the electrons localized in the random potential of the semiconductor superlattices, where both the localization and the strength of the electron–electron interaction can be controlled by the growth. It should be noticed that the collective coupled excitations in semiconductor superlattices have been widely studied by means of Raman scattering [10]. However, to the best of our knowledge, a study of the behaviour of the collective plasma-like excitations in the presence of disorder has not yet been performed.

In [11] we showed that Raman scattering can serve as a tool to probe both spatial and energy characteristics of the collective excitations in heavily doped semiconductors. In this case of a strong disorder, plasmons can be represented as a superposition of plane waves with the wave vectors distributed in a finite interval $\Delta \boldsymbol{q}$, which gives rise to their finite spatial extent (with coherence or localization length $L_c \sim \Delta \boldsymbol{q}^{-1}$). As a consequence, the observed Raman line is actually caused by a superposition of a number of narrow Lorentz-type lines weighted with respect to the density of states of plasmons. Therefore, the shape of the resulting Raman line is determined by the homogeneous broadening (damping) of the contributing individual Lorentz lines and by the spatial correlation of plasmons. Thus, the Raman line acquires a specific asymmetry clearly seen in the spectra of disordered semiconductors when the spatial correlation dominates [11]. The formation of the Raman line shape for the disordered materials is shown in figure 1; the shaded areas represent the plasmon states contributing to the Raman process (the left-hand panel) and the resulting Raman intensities (the right-hand panel).

In this letter, an analysis of the individual dampings together with the frequencies and localization lengths of the collective plasmon–LO phonon excitations in the doped GaAs/AlAs superlattices is presented. Different dopings allowed us to vary the strength of the electron–electron interaction. It is shown that in qualitative accordance with the theory [8, 9], the individual broadenings and the frequencies reveal complex behaviours with the increase of the electron density, which is believed to be associated with the formation of the strongly correlated states of the electron plasma in disordered matter.

$(GaAs)_{17}(AlAs)_2$ superlattices (where the numbers denote the thicknesses of the corresponding layers expressed in monolayers) doped with Si were studied. The superlattices were designed so as to achieve a strong localization of electrons. According to the calculations made using the envelope function approximation, these superlattices reveal a lowest broad

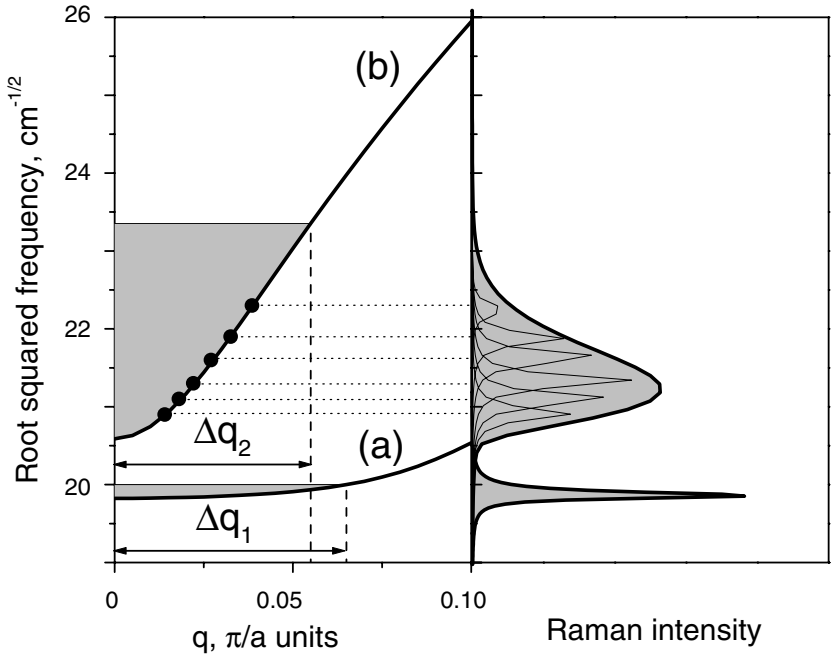


**Figure 1.** The left-hand panel presents the RPA dispersions of the plasmon–LO phonon excitations in $(GaAs)_{17}(AlAs)_2$ superlattices calculated with the electron densities $5 \times 10^{17}$ cm$^{-3}$ (a) and $3 \times 10^{18}$ cm$^{-3}$ (b) where the shaded areas show the collective states contributing to the Raman process, while in the right-hand panel the corresponding Raman intensities are plotted.

miniband with a width 65 meV, while the monolayer fluctuations produce a random distribution of the electron energy of the order of 60 meV, which should cause the expected localization [12]. Indeed, these superlattices revealed an extremely high resistance (some MΩ) even at extremely high doping levels (close to $10^{18}$ cm$^{-3}$). Moreover, the presence of the localized electrons was confirmed by the measurements of the capacitance [16] and the thermostimulated current [19].

As a result of the Coulomb coupling between the plasmons and the LO phonons, the following coupled plasmon–LO phonon modes appear in the superlattices under investigation: a low-frequency acoustic-like mode ($L^-$) and two high-frequency optic GaAs-like ($L_1^+$) and AlAs-like ($L_2^+$) ones. As has been shown in [13], at appropriate electron concentrations the AlAs-like mode reveals a mostly plasmon character and it is well separated from the other modes, which simplifies the analysis. Therefore, we present a qualitative analysis of the broadenings of the Raman lines corresponding to the high-frequency AlAs-like coupled mode ($L_2^+$) in differently doped $(GaAs)_{17}(AlAs)_2$ superlattices.

In order to obtain the characteristic parameters of the coupled modes, the Raman intensities were calculated according to the formula [11]

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

where

$$f_{sc}(\vec{q}) = \left(\frac{4\pi}{q^2 + q_{TF}^2}\right)^2$$

is the screening correlation function with $q_{TF}$ being a Thomas–Fermi wave vector; $\omega(q)$ is the dispersion of the relevant excitations contributing to the Raman scattering, calculated in the

random-phase approximation (RPA) as in [13], and $L_c$ and $\Gamma$ are their localization length and damping constant respectively.

The selected Raman spectra of the $(GaAs)_{17}(AlAs)_2$ superlattices with different doping levels taken in the frequency range of the AlAs optical vibrations are depicted in figure 2 where the Raman lines associated with the contributions of the individual collective excitations are shown by broken lines. The broad Raman lines caused by the well known interface modes [14] were detected around 375 $cm^{-1}$ for all the samples. The values of the broadenings $\Gamma$, the positions of the AlAs-like plasmon–LO phonon Raman lines $\omega_2^+$, and the values of the coherence lengths $L_c$ associated with the spatial localization of the collective excitations, obtained by the fitting of the intensities calculated from expression (1) to the experimental spectra, are plotted in figures 3(a), 3(b), 3(c). The calculated spectra are shown by solid lines in figure 2. For comparison, the dependence of the frequency of the AlAs-like plasmon–LO phonon excitations at the centre of the Brillouin zone on the electron concentration calculated in the case where the line shift is determined by the occupation of the miniband of the perfectly ordered superlattice is shown in figure 3(b), by the solid line. These calculations were made according to the theory presented in [15], where the finite width of the miniband was taken into account.

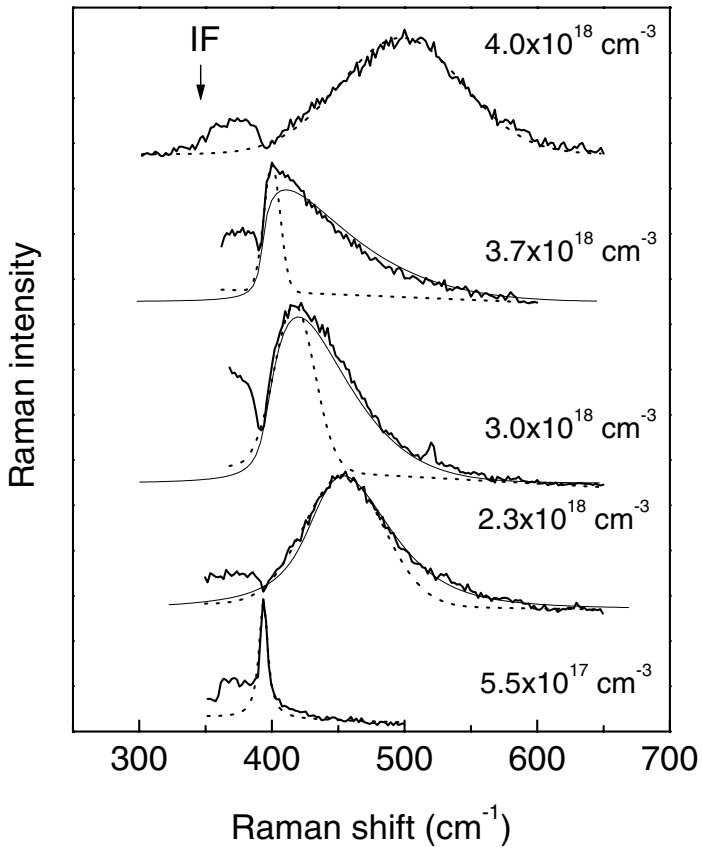


**Figure 2.** Raman spectra of the AlAs-like plasmon–LO phonon modes in the $(GaAs)_{17}(AlAs)_2$ superlattices with different electron concentrations measured at the temperature $T = 10$ K. The contributions of the individual collective excitations are shown by broken lines, while the full lines represent the spectra calculated using expression (1).

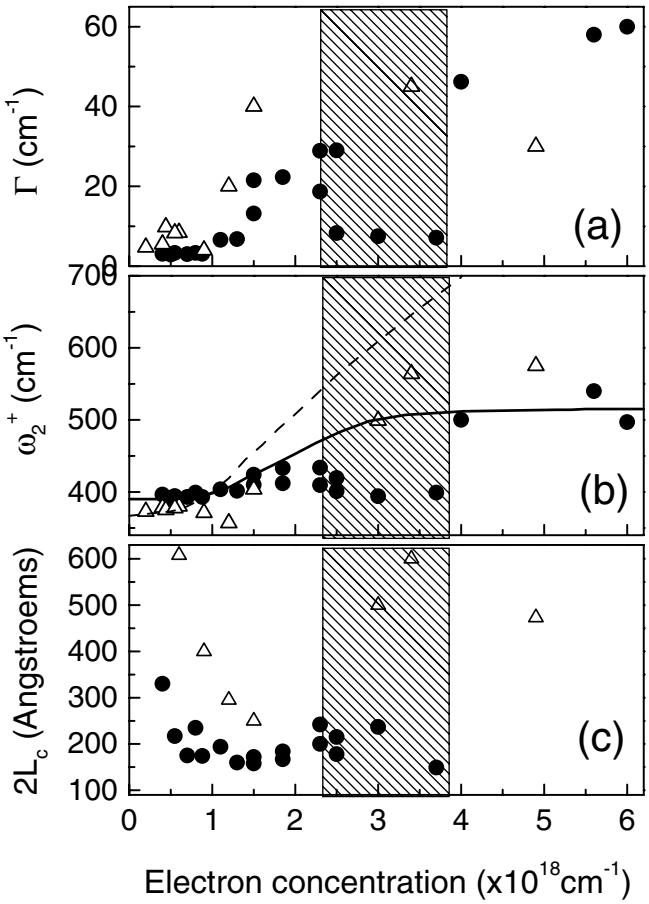


**Figure 3.** Parameters of the collective excitations measured in the $(GaAs)_{17}(AlAs)_2$ superlattices (full circles) and in the equivalent $Al_{0.11}Ga_{0.89}As$ alloys (open triangles) with different electron concentrations: (a) the damping constants ($\Gamma$); (b) the frequencies ($\omega_2^+$); and (c) the localization lengths ($2L_c$ for the superlattices and $L_c$ for the alloys). The hatched areas show the interval of electron densities where the effects of the strong correlation in the electron plasma were observed. The dependencies of the frequencies on the electron density calculated as explained in the text are shown by full and broken lines for the superlattices and for the alloys respectively.

In the sample with the lowest doping ($5 \times 10^{17}$ cm$^{-3}$), the AlAs-like optical mode is a mostly phonon-like mode and it reveals a Lorentzian-type shape characterized by homogeneous broadening ($\Gamma = 3.5$ cm$^{-1}$). With the increase of the doping level, as a consequence of the plasmon–LO phonon interaction, this line shifts to high frequencies and increases in intensity, and its shape acquires a strong asymmetry. Meanwhile, the broadening $\Gamma$ of the individual lines reveals a rise with the increase of the electron density, caused by the increase of the disorder induced by the random impurity potential. However, a strong line narrowing accompanied by a pronounced red-shift was found beginning at the electron concentration $N = 2.5 \times 10^{18}$ cm$^{-3}$.

According to the theory presented in [8,9], the coherent collective response of interacting electrons localized by the superlattice random potential is responsible for the observed spectral narrowing. At low electron densities, the long distances between electrons prevent an effective interaction between them. With increasing electron densities, the cooperative behaviour of the interacting electrons becomes dominating, resulting in the formation of a strongly

correlated electron plasma in the presence of disorder, when the dynamical many-particle interaction almost completely screens out the effects of the random potential fluctuations. As a consequence, the broadenings $\Gamma$ acquire values very close to those found for the low-doped samples.

With a further increase of the doping, an abrupt increase of both the line broadening and the spectral line shift was observed at $N = 4.0 \times 10^{18}$ cm$^{-3}$. At such high electron concentrations the impurity disorder becomes very strong; this breaks down the correlation responsible for the line narrowing, and the collective excitations turn out to be strongly damped. As a result, the broadening $\Gamma$ dominates, resulting in a symmetrical shape of the plasmon–LO phonon line, and its value and the value of the characteristic frequency $\omega_2^+$ return to those values appropriate to a perfectly ordered superlattice (see figures 3(a), 3(b)).

Figure 3(c) shows the dependence of the coherence length $L_c$ on the electron density. It should be mentioned that these data were obtained using the RPA dispersion of the coupled collective modes $\omega(q)$ calculated numerically, while in [13] we used the parabolic approximation. As is shown in figure 1, the parabolic approximation is not valid for wave-numbers very close to the centre of the Brillouin zone where the frequency approximates to its value at $q = 0$. Therefore, very different widths of the Raman lines can result in close values of $\Delta q$ and, as a consequence, similar values of $L_c$ can be obtained for samples with different electron densities. A decrease of the coherence length $L_c$ with the rise of the electron concentration was observed in the samples with low doping, where the electron–electron interaction is still weak and the impurities are responsible for the spatial limitation of the collective excitations. However, in the range where the correlation effects were detected (the hatched areas in figure 3), $L_c$ reveals the increase predicted in [2,3] for the interacting localized electrons.

The double data shown in figure 3 for some of the samples indicate that the Raman lines associated with the collective excitations could not be fitted well with a single line calculated from formula (1). This shows that the inhomogeneity of the correlated clusters increases in the superlattices with the electron densities prior to the critical value being reached.

Thus, we can state that qualitative agreements with the theories [2, 3, 8, 9] were found; however, abrupt alterations of the damping and the line shift, which were not predicted theoretically, were observed in the experiment. These abrupt changes are probably evidence for phase transitions in the state of the electron plasma in the disordered matter, caused by the formation of a strongly correlated plasma state in a form of coherently polarized dynamic clusters. The formation of such dynamic clusters is clearly seen in the temperature behaviour of the integrated intensity of the Raman line associated with the AlAs-like coupled mode, which is proportional to the number of coherent excitations contributing to the Raman process. The data obtained for the superlattice with $N = 2.5 \times 10^{18}$ cm$^{-3}$, where the line narrowing was already observed, are plotted in figure 4. This shows that, beginning at the temperature $T \simeq 50$ K, the coherent clusters gradually disappear.

Meanwhile, no evidence of the presence of coherently polarized clusters was detected for the equivalent bulk $Al_{0.11}Ga_{0.89}As$ alloys (the alloys with concentration of Al equal to that in the superlattices studied)—we did not find abrupt changes in $\Gamma$, $L_c$, and in the frequency of the collective excitations; on the contrary, the latter was found to be in good agreement with the calculations taking into account the occupation of the nonparabolic conduction band only. Also, the integrated intensities were found to be independent of the temperature. The reason for such a difference between the behaviours of the collective modes in the superlattices and in the alloys is the origin of the disorder. The monolayer fluctuations provide a relatively long-range disorder in the superlattices, which enables them to cause electron localization, while the short-range disorder of the alloys does not strongly localize electrons, mostly acting

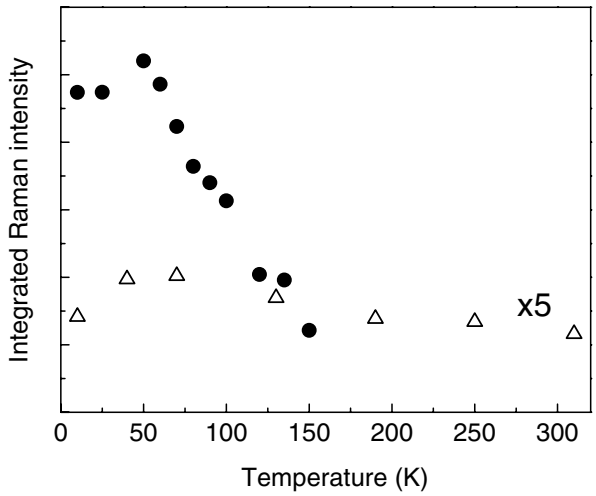


**Figure 4.** The temperature dependence of the integrated intensity of the Raman line associated with the AlAs-like plasmon–LO phonon coupled mode measured for the $(GaAs)_{17}(AlAs)_2$ superlattice with the electron concentration $N = 2.5 \times 10^{18}$ cm$^{-3}$ (full circles) and for the $Al_{0.11}Ga_{0.89}As$ alloy with the electron concentration $N = 6.0 \times 10^{17}$ cm$^{-3}$ (open triangles).

as a cause for electron scattering. Thus, the system of the localized interacting electrons in the superlattices reveals different properties in comparison with the scattering electrons in the alloys.

It is worth mentioning that the system of electrons localized by a random potential considered here has close similarities with dipoles in spatially random ferroelectrics, where the existence of ferroelectric order has been recently established [17]. It was shown theoretically that despite the strong frustration present in random systems, long-range ferroelectric order is possible above a critical density [18]. From this point of view, the observed abrupt alterations of the frequency and damping associated with the collective excitations could indicate spontaneous formation of strongly correlated dynamic clusters, which takes place at a critical electron density; with further increase of the doping, the clusters of the coherent dynamic polarization are destroyed by a random impurity potential. In this case, the localization lengths obtained here could be associated with the size of the coherent clusters.

To conclude, we have presented an experimental study of the collective excitations in heavily doped GaAs/AlAs superlattices. Our results testify to the spectral narrowing caused by the coherent collective response of the interacting localized electrons predicted theoretically in [8, 9]. An analogy of the behaviour of the dynamic polarization of the localized electrons with the properties of ferroelectrics was discussed. In addition, the important role of the dispersion of the collective excitations in the formation of the shape of the Raman lines was established.

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