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Raman probing of spatial extents of collective excitations in AlGaAs alloys

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Abstract

The spatial extents of the wave functions of the collective plasmon–longitudinal optical (LO) phonon excitations have been studied as a function of the doping concentration and the temperature in the AlGaAs alloy, where the short-range alloy potential fluctuations, together with the impurity random potential, are responsible for the disorder. A strong increase of the localization length was found with increase of the doping, where the plasmons substitute for phonons as collective excitations. A further decrease of the localization length of the plasmon-like excitations detected with increase of the doping level at high electron densities was attributed to the interaction of the plasmons with the impurities. A theoretical analysis based on the variational principle showed that the ionized impurities are responsible for the observed modification of the localization length.

1. Introduction

The problem of the interaction of carriers with crystal imperfections such as impurity centres, native defects, and phonons is of great importance in solid-state physics. In the case of a weak interaction, this problem is usually treated in a semiclassical approach, where an electron can be represented as a point particle in a well defined Bloch state $\psi(\boldsymbol{q})$. The Born approximation is then used to determine the scattering rate $W(\boldsymbol{q}, \boldsymbol{q}')$, which defines the probability of the electron in a state $\boldsymbol{q}$ being scattered into a state $\boldsymbol{q}'$ [1]. This approximation is basically valid when the potential energy of the interaction with scatterers is small in comparison with the particle kinetic energy. In this case one supposes that far away from the scattering centre the wave function can be represented as a superposition of a plane wave (before the scattering) and a spherical one (after the scattering). An additional consequence of the scattering process is the decay of the wave function of the scattered particle. This decay is negligible when

the interaction is weak; however, for a strong interaction the uncertainty in the determination of the particle wavenumber δq—associated with the spatial localization of the particle L_c ($\delta q \sim L_c^{-1}$)—can be large enough compared with the Brillouin zone size. In this case, the Born approximation fails and a more general approach for treating the scattering process, which takes into account the modification of the wave function, should be used. An extremely strong interaction presents the opposite limit, when the particle becomes localized with the corresponding wave function. The process of the transformation of the extended states to localized ones is known as the problem of the metal-to-insulator transition (MIT).

One of the fundamental but yet not fully resolved questions concerning the physics of the MIT is that of the behaviour of the electron wave functions at energies close to the boundary energy E_c separating the localized and delocalized states. It follows from general principles of quantum mechanics that when the MIT takes place, the wave function must change in a gradual way [2]. In particular, the localization length, which reflects the modification of the wave function in space, must also gradually change when the energy passes the value of E_c.

An exhaustive theoretical analysis of the behaviour of the electron wave function across the MIT was made more than two decades ago (see, for instance, [3–8]). However, to our knowledge, until now there have been no attempts to probe experimentally the electron wave function close to the MIT, although the critical behaviour of the electron system undergoing a MIT has been broadly studied and the behaviour of the dc conductivity (and as a consequence, the diffusion and relaxation of carriers) has been established.

On the other hand, the possibility of utilizing Raman scattering as a tool to study the spatial extent of collective excitations such as phonons, plasmons and coupled plasmon–phonon modes in disordered solids has been demonstrated in our recent articles [9, 10]. It should be emphasized that plasmons determine the electrical response of the electron system to a high-frequency external electric field, while a single electron is mostly responsible for the dc response. In many respects the dynamic properties of the single-particle excitations and of the collective excitations are similar. In particular, like the single electron, the plasmons reveal a localization when exposed to a random potential [11]. Therefore, the investigation of the spatial extent of the localized plasmon can shed some light on the behaviour of the carrier wave function near the MIT.

Using results obtained in references [9, 10], the plasmon-like excitations were studied in superlattices with an intentionally introduced disorder where the formation of spatially coherent dynamic clusters caused by the electron–electron interaction of the localized electrons was found [12, 13]. The long-range random potential of the disordered SL (which varies on a scale comparable to the well thicknesses) causes a strong localization of the electrons in the wells. It turned out that a complicated interplay between the interaction and the disorder defines the localization properties of the collective excitations in the disordered superlattices, resulting in the formation of coherent clusters. On the other hand, a relatively short-range alloy potential gives rise to the scattering of electrons without a significant localization. Therefore, the coherent dynamic clusters are not expected to dominate the collective electronic response in an alloy with a relatively low concentration of Al, where the DX centres still do not cause a strong electron localization.

The absence of a strong localization and, as a consequence, relatively weak correlation effects (resulting in a formation of the coherent clusters) allow us to carry out an analysis of the localized collective excitations in an alloy in a rather simple approach where the plasmon is represented by a wave function in the form of a Gaussian with the spatial extent influenced by a scattering mechanism.

In this paper we present a systematic study of the influence of imperfections on the propagation and localization of collective plasmon-like excitations in the AlGaAs alloys. We

show that at rather moderate doping levels the impurities already strongly modify the wave functions of the plasmons, giving rise to their localization. A simple theoretical model was developed to account for the interaction of the plasmons with the impurities. In addition, a significant increase of the plasmon localization caused by the interaction with phonons was found with the increase of the temperature.

2. Experimental procedure

$Al_{0.11}Ga_{0.89}As$ alloys doped with Si were grown by molecular beam epitaxy on (100)-oriented GaAs substrates. The content of Al was chosen such as to provide alloys equivalent to the $(GaAs)_{17}(AlAs)_2$ superlattices studied in [10]. The electron concentrations were obtained both by Hall and capacitance measurements. Unpolarized back-scattered Raman spectra were taken at $T = 10$ K with an 'Instruments S.A. T64000' triple-grating spectrometer supplied with a CCD detector cooled by nitrogen; the 5145 Å line of an Ar^+ laser was used for excitation.

Some selected Raman spectra of the $Al_{0.11}Ga_{0.89}As$ alloy are shown in figure 1. The lines due to the GaAs-like optical phonons (TO_1 and LO_1) and the AlAs-like ones (TO_2) together with the GaAs-like (ω_1^+) and the AlAs-like (ω_2^+) coupled plasmon–LO phonon collective modes were clearly detected. As was shown in [10,12], the dispersion $\omega(k)$ of the collective excitations determines the shape of the corresponding Raman lines. The different characters of the coupled modes, either phonon-like or plasmon-like, are responsible for their different asymmetries; the AlAs-like coupled modes were shown to already have an essentially plasmon-like character at rather low electron concentrations, which allowed them to be easily analysed [10]. Therefore, we present further data concerning the AlAs-like mode. The Raman spectra of some of the samples measured in the range of the AlAs optical phonons are shown in figure 2. The positions

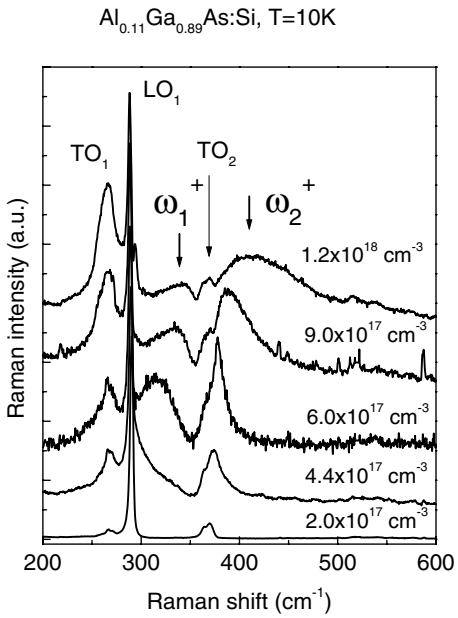

Figure 1. Raman spectra of the $Al_{0.11}Ga_{0.89}As$ alloys with different electron concentrations measured at $T = 10$ K. The intensities were normalized with respect to the intensity of the GaAs-like phonon (LO_1).

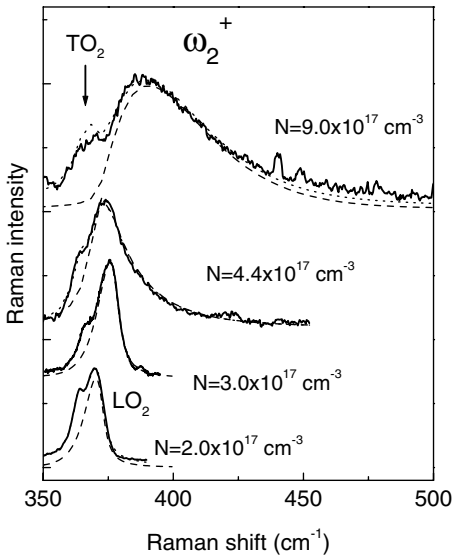

Figure 2. Raman spectra of the $Al_{0.11}Ga_{0.89}As$ alloys with different electron concentrations measured at $T = 10$ K in the frequency range of the AlAs optical phonons (solid lines). The dotted lines show the full calculated spectra, while the separate contributions of the collective plasmon–LO phonon excitations are plotted as dashed lines.

and the linewidths of the Raman lines are determined by the characteristic frequencies and by the processes of the dissipation of energy (damping constants) of the collective excitations, while the strengths of the line asymmetry are governed by the spatial localization of the relevant excitations [9]. The values of the parameters characterizing the AlAs-like coupled modes (the mode frequencies which determine the positions of the relevant Raman lines, the damping constants and the localization lengths) were determined by the fitting of the experimental Raman spectra as in [10]. The dispersions of these modes, indispensable for the fitting, were calculated at different electron densities in the random-phase approximation (RPA) and they are given in figure 3.

The parameters of the collective excitations of interest measured for the alloys with different electron concentrations are shown in figure 4. The whole interval of the electron densities studied here can be divided into the three following ranges, where different behaviours of the collective excitations were found:

(I) The low-electron-densities range. Here, with increase of the doping level, the LO phonons are transformed into coupled plasmon–LO phonon excitations. This process is accompanied by an increase in the intensities of the relevant Raman lines which takes place because the active volume of the excitations contributing to the Raman process rapidly increases when changing from phonons to plasmons. This increase of the active volume of the excitations contributing to the Raman scattering is directly seen in the increase of the localization length (L_c) presented in figure 4(c).

(II) The intermediate-electron-densities range. In this range the collective modes mostly acquire a plasmon character, which is revealed in the corresponding asymmetry of the Raman lines [9] and in the blue-shift of the mode frequency. The dependence of the frequency ω_2^+ on the calculated electron density in the $Al_{0.11}Ga_{0.89}As$ alloy is shown in figure 4(a) by a solid line. At such doping levels, the scattering of the plasmons by the

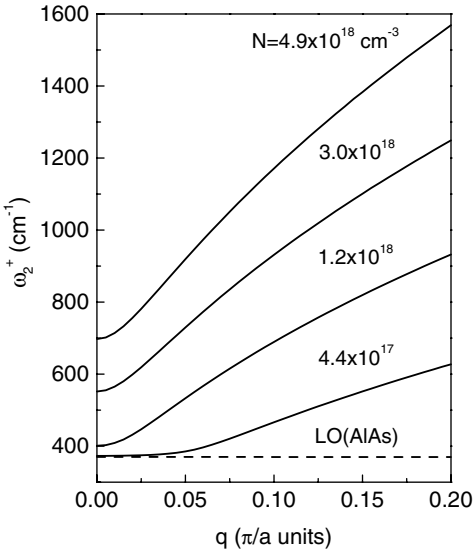

Figure 3. Dispersions of the AlAs-like collective coupled plasmon–LO phonon excitations calculated using the RPA for the $Al_{0.11}Ga_{0.89}As$ alloys with different electron densities. The dispersion of the LO phonon is shown by the broken line.

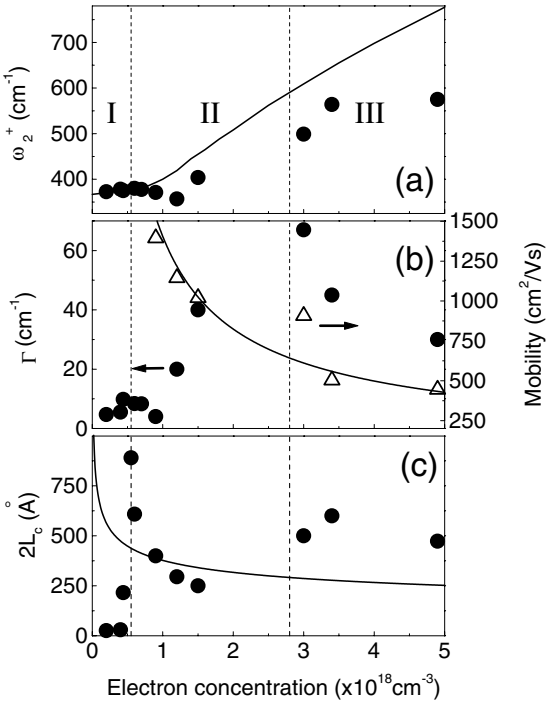

Figure 4. Dependencies of the frequency (ω_2^+), the damping constant (Γ) and the localization length ($2L_c$) of the AlAs-like collective plasmon–LO phonon excitations on the electron density measured in the $Al_{0.11}Ga_{0.89}As$ alloy at $T = 10$ K. The values of the Hall mobilities (μ) are shown in (b) by triangles with the corresponding scale on the left-hand side of the figure. The solid lines are the dependencies $\omega_2^+(N)$ (a), $\mu(N)$ (b) and $2L_c(N)$ (c) calculated as explained in the text.

impurity silicon atoms dominates, resulting in an increase of the damping constant (Γ) and in a decrease of the localization length (L_c) of the collective excitations studied.

(III) The high-electron-densities range, where the effects of the correlation caused by the electron–electron interaction—which have already been found in disordered GaAs/AlGaAs superlattices—become significant, resulting in the formation of coherent spatial clusters [12, 13] and, as a consequence, in narrowing of the Raman lines (decrease of the damping constant Γ) and in increase of the localization length with increase of the doping levels.

It ought to be pointed out that the relatively small values of the localization lengths of the collective coupled plasmon–LO phonon excitations (comparable with the interelectron distances) obtained in the highly doped samples mean that in this case the collective excitations are mostly represented by the LO phonon-like vibrations, probably strongly coupled only with the neighbour electrons.

The effects of the temperature measured in the doped $Al_{0.11}Ga_{0.89}As$ alloys are shown in figures 5, 6. A weak decrease of the AlAs-like coupled-mode frequency with the increase of the temperature caused by the decrease of the frequency of the optical phonon was observed. Meanwhile, an increase of the damping constant with the temperature accompanied by a decrease of the localization length caused by the scattering of the plasmons by phonons was

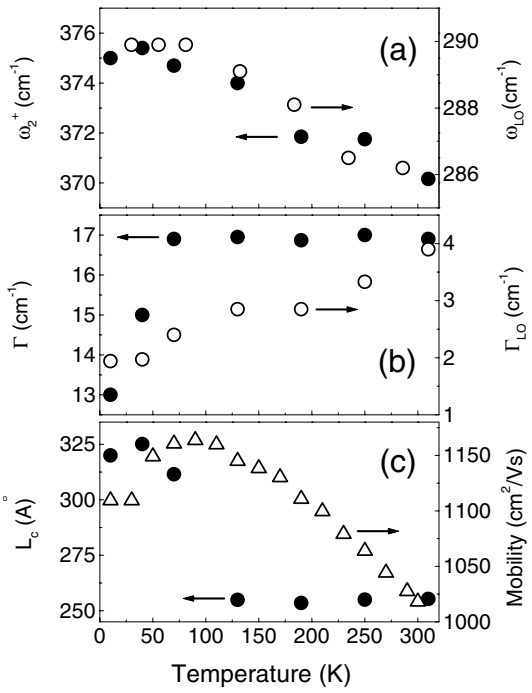

Figure 5. Temperature dependencies of the frequency (ω_2^+), the damping constant (Γ) and the localization length ($2L_c$) of the AlAs-like collective plasmon–LO phonon excitations measured in the $Al_{0.11}Ga_{0.89}As$ alloys with the electron concentration $N = 6.0 \times 10^{17}$ cm^{-3} (closed circles). The temperature dependencies of the frequency and the damping constant of the GaAs-like LO phonon are given as a reference by open circles in panels (a) and (b) respectively. The Hall mobility measured as a function of the temperature in the alloy with $N = 9.0 \times 10^{17}$ cm^{-3} is given by triangles in panel (c). The scales corresponding to the open symbols are shown on the left-hand side of the figure.

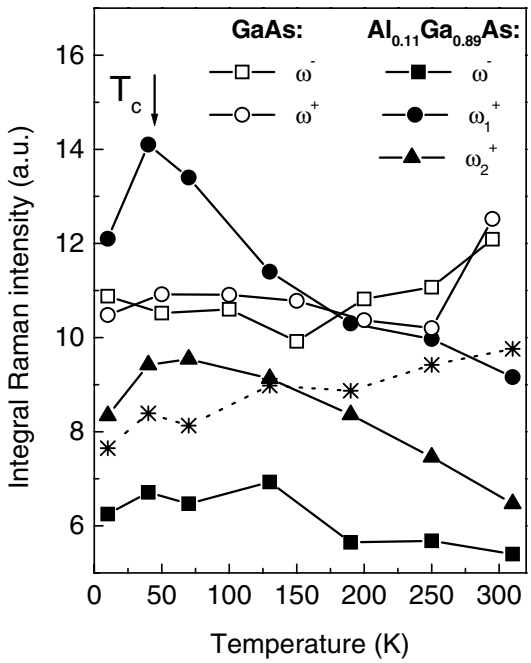

Figure 6. Temperature dependencies of the integral intensities of the Raman lines associated with the coupled modes in the $Al_{0.11}Ga_{0.89}As$ alloy with the electron concentration $N = 6.0\times10^{17}$ cm^{-3} (closed symbols) and in GaAs with $N = 1.0 \times 10^{18}$ cm^{-3} (open symbols). The integral intensity of the GaAs-like LO phonon line of the alloy is given as a reference (stars).

found at low temperatures ($T < 150$ K). The dependencies of the integral Raman intensities associated with the number of modes contributing to Raman scattering are shown in figure 6. The decrease of the integral Raman intensities of the coupled plasmon–LO phonon excitations with increase of the temperature, indicating the destruction of the coherently oscillating clusters at high temperatures (higher than the critical temperature T_c), although not so clearly pronounced as in the superlattices [13], was also observed in the $Al_{0.11}Ga_{0.89}As$ alloys. In both disordered systems the electron–electron correlation observed at high electron densities resulted in a partial delocalization which was revealed in the increase of the localization length with the increase of the electron concentration—a theoretically predicted effect (see references in [13]). As expected, no evidence of the coherent clusters was detected for the doped GaAs (the data are shown in figure 6) where no signs of electron localization were found. Thus the measured localization lengths of the plasmon-like excitations are attributed to the sizes of the coherently oscillating clusters.

The influence of the correlation effects on the collective plasmon-like excitations is directly seen from a comparison between the results obtained by means of Raman scattering and the data obtained by electrical dc measurements. In contrast to the Raman data indicating the influence of the correlation effects in the alloys with high electron densities (in the range III in figure 4), the values of the Hall mobilities plotted in figure 4(b) reveal a monotonic decrease with increase of the doping, found in good agreement with the mobilities calculated for the ionized impurity scattering (shown in figure 4(b) by a solid line) [14], without any indication of a contribution from the collective effects. This is an expected result, because the electron–electron interaction and the corresponding correlation play determining roles in the collective

plasma oscillations, while the single-particle excitations are principally responsible for the dc transport.

The temperature dependence of the Hall mobility measured in the alloy with the electron concentration $N = 9 \times 10^{17}$ cm^{-3} is shown in figure 5(c) by open triangles. In the range of high temperatures it shows a dominant contribution from the scattering of electrons by phonons.

3. Theoretical analysis and discussion

In the range of the intermediate electron densities (range II in figure 4), where the scattering dominates in the origin of the spatial extents of the coupled plasmon-like excitations and the above-discussed electron–electron correlations are still not significant, the modification of the wave function of the plasmons can be calculated taking into account the interaction of the plasmons with the impurities.

As was mentioned in the introduction, in the presence of disorder, collective excitations can be treated as a superposition of plane waves with the wave vectors distributed in a finite interval $\delta \boldsymbol{q}$. In this case, according to the model successfully used for the optical phonons in microcrystalline silicon [15], one can present the trial wave function in the form of a Gaussian:

$$\psi(\boldsymbol{r}) = \frac{2\sqrt{2}}{\pi^{3/4} L_c^{3/2}} \exp\left(-\frac{2\boldsymbol{r}^2}{L_c^2}\right) \tag{1}$$

meaning effective localization at $|\boldsymbol{r}| \lesssim L_c$. This means that we treat the collective excitations as non-interacting quasiparticles with an effective charge e^* and a mass M^*, spatially localized in a sphere of characteristic radius L_c. This wave function, coinciding with a spherically symmetric ground-state wave function of a 3D harmonic oscillator, allows us to write the effective Hamiltonian of the collective excitations as

$$\hat{H}_{\text{eff}} = -\frac{\hbar^2}{2M^*} \frac{1}{r^2} \frac{\partial}{\partial r}\left(r^2 \frac{\partial}{\partial r}\right) + \frac{8\hbar^2}{M^* L_0^4} r^2 \tag{2}$$

where the wave function $\psi(\vec{r})$ is the eigenfunction of $\hat{H}_{\text{eff}}$ for $L_c = L_0 = 2\sqrt{\hbar/M^*\omega_0}$, which is the localization length of the harmonic oscillator with a resonance frequency ω_0. One should note that L_0 has the sense of a localization length of the plasmon-like excitations in the absence of impurities (in an undoped alloy). The interaction of the plasmon with impurities transforms $\hat{H}_{\text{eff}}$ into

$$\hat{H}_{\text{pl}} = \hat{H}_{\text{eff}} + U_{\text{imp}}(\boldsymbol{r}) \tag{3}$$

and one should calculate the variation of the localization length caused by the interaction with the scattering potential of the ionized impurities:

$$U_{\text{imp}}(\boldsymbol{r}) = U_{\text{imp}}^{\text{ion}}(\boldsymbol{r}) + U_{\text{imp}}^{\text{neutral}}(\boldsymbol{r}) = \frac{Zee^*}{\epsilon_\infty} \sum_j \frac{\mathrm{e}^{-q_0|\boldsymbol{r}-\boldsymbol{r}_j|}}{|\boldsymbol{r}-\boldsymbol{r}_j|} + U_0\theta(R_0 - r) \tag{4}$$

where two contributions to the interaction are included: one of them is caused by the interaction of plasmons with ionized impurities (the first term in equation (4)) whereas the second one is due to the interaction with neutral impurities (the second term in equation (4)). Here Ze is the effective charge of the ionized impurities, e is the electron charge, ϵ_∞ is the dielectric constant of the medium, U_0 and R_0 are the interaction potential and the effective radius of neutral impurities respectively and $\theta(R_0 - r)$ is the step function [1]. The exponential dependence of $U_{\text{imp}}^{\text{ion}}(\boldsymbol{r})$ is associated with the screening action of the electrons located between the plasmon and the charge centre interacting with it. The screening parameter is $q_0^2 = 6\pi n_{el} e^2/\epsilon_\infty E_F$ where n_{el} is the volume electron concentration, and E_F is the Fermi energy.

The wave function given by equation (1) is not the eigenfunction of the operator $\hat{H}_{\rm pl}$. We can however treat it now as a trial function of the Schrödinger equation for the plasmon depending on the Hamiltonian $\hat{H}_{\rm pl}$ and containing L_c as a variational parameter. Then, the ground-state energy can be written as

$$E_0 = \langle\psi(\boldsymbol{r})|\hat{H}_{\rm pl}|\psi(\boldsymbol{r})\rangle = \frac{3}{4}\hbar\omega_0\left(\frac{L_0^2}{L_c^2}+\frac{L_c^2}{L_0^2}\right) + \int \psi(\boldsymbol{r})\left[U_{\rm imp}(\boldsymbol{r})\right]\psi(\boldsymbol{r})\,{\rm d}^3\boldsymbol{r} \tag{5}$$

or

$$E_0 = \frac{3}{4}\hbar\omega_0\left(\frac{L_0^2}{L_c^2}+\frac{L_c^2}{L_0^2}\right) + E_i + E_n \tag{6}$$

where the first term corresponds to the contribution of the ground energy of an unperturbed plasmon (with the localization length L_0), while the second and the third ones are the contributions due to the interaction of the plasmon with neutral and ionized impurities respectively. Using the interaction in the form presented by equation (4) and estimating the effective number of impurities corresponding to the interaction with the plasmons as $N_i q_0^{-3}$ and $N_0 L_c^3$ for the ionized and neutral impurities respectively, we calculated

$$E_i = \frac{N_i e^2 L_c^3}{4\epsilon_\infty}\left[\exp\left(-\frac{q_0L_c-1}{4}\right)\operatorname{erfc}\left(\frac{q_0L_c-1}{4}\right) - \exp\left(-\frac{q_0L_c+1}{4}\right)\operatorname{erfc}\left(\frac{q_0L_c+1}{4}\right)\right] \tag{7}$$

$$E_n = \frac{U_0 L_c^3 N_0}{8}\left[\operatorname{erf}\left(\frac{2R_0}{L_c}\right) - \frac{R_0}{\sqrt{\pi}L_c}\exp\left(-\frac{4R_0^2}{L_c^2}\right)\right] \tag{8}$$

where N_i and N_0 are the concentrations of the ionized and neutral impurities respectively.

The value L_c of interest can be found from the condition for the minimum of E_0 as a function of L_c:

$$\partial E_0/\partial L_c = 0. \tag{9}$$

As is seen from equations (7), (8) the value E_0 depends on the impurity and on the electron concentrations causing the corresponding dependence of the localization length calculated from equation (9). This mainly explains the experimental results shown in figure 4(c). It should be mentioned that the interaction of the plasmon with the neutral impurities reveals a negligible variation with the impurity concentration, obtained with the reasonable parameter values $U_0 = 1$ eV and $R_0 = 10$ Å. This is an expected result because the effective radius of the impurity is much smaller than the localization length of the plasmons, which implies a weak interaction of the neutral impurities with the plasmon. Therefore, the main contribution to the dependence of the plasmon localization length on the concentration of impurities is due to the interaction with the ionized impurities. The results of numerical calculation of L_c from equation (9) are represented in figure 4(c) by a solid line. A reasonable agreement between our calculations and the experimental data obtained for the $Al_{0.11}Ga_{0.89}As$ alloys is observed. This means that the modification of the wave function of the collective excitations observed in the alloys with not-too-high electron concentrations can be understood in terms of their interaction with the ionized impurities. It should be emphasized that no fitting parameters except the localization length of the plasmon-like excitation in the undoped alloy (L_0) were used in the calculation of the localization lengths presented in figure 4(c); the best correspondence with the experimental data was obtained for $L_0 = 4000$ Å.

4. Conclusions

The dynamic localization properties of the collective coupled plasmon–LO phonon excitations were studied in the doped AlGaAs alloys, where the electrons are subjected to an impurity random potential together with the fluctuations of the crystal alloy potential. The behaviour of the collective excitations was found to be different from that observed in the disordered short-period GaAs/AlGaAs superlattices. In the presence of the strong electron localization found in the superlattices by the measurements of the thermostimulated current [16], clear evidence of the formation of coherently oscillating spatial clusters was obtained [12, 13], while only a tendency to form such clusters was observed in the alloys studied here, where no significant symptoms of localization were detected. A fundamental difference between the superlattices and the alloys responsible for the observed difference is in the origin of the disorder. In the case of alloys with low concentrations of Al (where the influence of the DX centres is still negligible), the disorder is caused by the relatively weak short-range alloy potential fluctuations acting as a cause of the electron scattering, while in superlattices the fluctuations of the periodicity cause strong long-range disorder, which effectively localizes the electrons. Therefore, coherent clusters which are principally provoked by the localized electrons, arise in the superlattices, while only weak consequences of their presence were found in the heavily doped alloys.

Thus, the doped alloy presents a disordered electron system where the localization is weak and the scattering processes dominate in the origin of the spatial extents of the wave functions of the collective excitations. Therefore, the plasmon-like excitations are strongly influenced by the ionized impurities revealing a decrease of the localization length with increase of the doping level. Our theoretical model presented here, based on the representation of the plasmon in the form of a Gaussian wave function, qualitatively reproduced the essential features of the experimental results obtained on the highly doped alloys before any influence of the correlation effects occurs.

Acknowledgments

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