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Resonant Raman spectra of spin-density transitions in periodically δ-doped GaAs

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Theoretical calculations for the resonant Raman spectra of spin-density transitions elucidating the influence of hole states participating in the resonance scattering process as well as a continuum of intersubband transitions are reported for periodically δ-doped GaAs. Raman cross sections calculated in conditions of extreme resonance with the spin-split edge show remarkable coincidence with the experimental line shapes we have also measured for the resonance spectra of these transitions.

The inhomogeneous distribution of electron screening charge around a sheet of donor ions in an epitaxially grown semiconductor leads to an interesting situation where electrons are confined in a purely space-charge self-consistent potential.[1] For a surface areal density $n_D > 0.3 \times 10^{12}$ cm^{-2} of Si atoms in GaAs, for instance, it is known from magnetotransport measurements that this so-called δ layer represents a metallic system of quantized subbands each of which has a different mobility for transport along the doping layer as the result of the distinct spatial configurations of the subbands relative to the donor sheet.[2] In periodic multilayer δ-doped systems, on the other hand, the overlap of carrier wave functions from adjacent layers is predicted to give rise to the formation of a quasicontinuum of subband states (minibands) whose nature is expected to be very dependent on the doping parameters used in the design of these structures. Recent investigations in these systems emphasize the relatively low doping metallic regime of the individual layers such that electronic states in the δ-doping superlattice may be described by a single nondispersive miniband where most of the electrons reside, and a partially filled nearly-free-electron (NFE) quasicontinuum of subbands extending above the top of the self-consistent potential.[3,4]

Inelastic light scattering is an ideal standard tool to investigate the energy level structure of free carriers in low-dimensional semiconductor systems.[5] In resonant scattering experiments, incident light whose energy is generally close to the spin-split ($E_0 + \Delta_0$) forbidden gap of bulk GaAs causes transitions from the valence band to excited subband states in the conduction band and scattered light is emitted by the recombination of the remaining holes in the valence band and electrons in occupied subband states of the conduction band. Two types of spectra are measured: polarized spectra, interpreted as due to intersubband charge-density excitations and depolarized spectra as arising from spin-density transitions. While the energy spectrum of the former excitations is known to be affected by the macroscopic polarization fields that follow the charge-density oscillations, the latter differ from the energy spectrum of single-particle transitions by excitonic interactions. In extended GaAs systems where such excitonic effects are small, the spectrum of spin-density excitations may be identified with that of intersubband transitions.[5,6]

Application of the resonant light-scattering technique to GaAs δ-doping systems, however, yields line shapes exhibiting a complex resonance behavior that finds no analogue in the corresponding spectra of modulation doping GaAs-$Al_xGa_{1-x}As$ multiple quantum well structures.[6] Using laser frequencies in resonance with the $E_0 + \Delta_0$ edge, Abstreiter *et al.*[5] observe broad spectral features in the depolarized Raman spectra of single δ layers which were attributed to electronic Raman scattering by intersubband transitions by comparing their energy positions with self-consistent calculations of the subband structure. Under similar circumstances, Maciel *et al.*[7] report a lower energy structure in a periodic multilayer δ-doped system which was also attributed to electronic Raman scattering by intersubband transitions. In both cases, spectral line shapes were found to be strongly dependent on the frequency of the exciting radiation and characterized by the appearance of broad bands in the high-energy side of the Raman spectra which more recent experimental work interpret as due to nonequilibrium luminescence at the $E_0 + \Delta_0$ edge.[8]

In this paper we present a theoretical investigation of the Raman spectra of spin-density transitions in periodically δ-doped GaAs based on an energy level structure calculated self-consistently. Calculations of the subband structure of both isolated δ layers and superlattices have been performed by various authors,[1,9,10] but the calculation of the electronic Raman spectra based on such level structure calculations has not been performed so far. Here, Raman cross sections corresponding to the spectrum of spin-density transitions were numerically computed for various exciting laser energies in conditions of extreme resonance with the $E_0 + \Delta_0$ edge. When compared with experiment, the theoretically computed cross sections show remarkable agreement with the line shapes we have also measured for the spectrum of these transitions or with those previously reported in Ref. 7 for a sample of similar characteristics.

As a consequence of the spatial separation of electron and hole wave functions implied by the presence of

a purely space-charge potential, different intersubband transitions are expected to resonate at different exciting laser frequencies. In particular, the broad bands that appear in the high-energy side of the Raman spectra are interpreted, according to the results of this paper, as due to electronic Raman scattering by a continuum of intersubband transitions which is made resonant when incident photon energies closely match those of a group of intermediate hole states having substantial overlap with the lowest subband states. Additional support for this interpretation is found in the recent observation of Fano antiresonances in the region of the longitudinal-optical (LO) phonon in periodically δ-doped GaAs samples as consequence of the Fröhlich coupling between LO phonons and an unscreened continuum of intersubband transitions.[4]

The inset of Fig. 1 shows the discrete spectrum of subband levels $E_c(n,k_z)$ using periodic boundary conditions and density-functional theory in the local density approximation for a 11-period δ-doping superlattice with spacing $d = 500$ Å and sheet carrier density $n_s = 1.0 \times 10^{12}$ cm^{-2} at zero temperature. In these calculations, the effective mass approximation was used and the discrete distribution of donor charge of each layer replaced by a continuous Gaussian distribution of positive charge of width $D = 50$ Å (full width at half maximum) spread along the perpendicular direction to the doping sheets along with a uniform distribution of negatively charged acceptors with density $N_a = 1 \times 10^{15}$ cm^{-3}. To calculate the cross section by spin-density excitations in backscattering configuration, the random phase approximation formula of Hamilton and McWhorter[11] was extended for the present situation. Based on this extension, the differential cross section for electronic Raman scattering in crossed polarizations is expressed in terms of the Fourier spectrum of spin-density fluctuations as

$$\frac{\partial^2\sigma_{sd}}{\partial\omega\partial\Omega} = \left(\frac{\omega_S}{\omega_L}\right) r_0^2 \int_{-\infty}^{\infty} \frac{dt}{2\pi} \langle N(t)N^\dagger(0)\rangle e^{i\omega t}, \qquad (1)$$

where r_0 is the classical electron radius, ω_L (ω_S) the frequency of the incident (scattered) radiation field with $\omega = \omega_L - \omega_S$, and

$$N^\dagger = \sum_{\nu'\nu} \Gamma_{\nu'\nu} \left[c^\dagger_{\nu'\uparrow} c_{\nu\uparrow} - c^\dagger_{\nu'\downarrow} c_{\nu\downarrow} \right] \qquad (2)$$

with

$$\Gamma_{\nu'\nu} = \frac{P_{cv}^2}{3m_0} \sum_h \frac{\langle \nu' | e^{\frac{1}{2}iqz} | h \rangle \langle h | e^{\frac{1}{2}iqz} | \nu \rangle}{E_g + i\gamma_g - \hbar\omega_L + \epsilon_{\nu'} + \epsilon_h}. \qquad (3)$$

Here, the subscript ν (h) stands for conduction (valence) -band envelope states with subband energies $\epsilon_\nu = E_c(n,k_z) + \hbar^2k^2/2m_c$ [$\epsilon_h = E_h(n,k_z) + \hbar^2k^2/2m_{so}$] with $\vec{k}$ being the two-dimensional wave vector common to valence- and conduction-band states, P_{cv} is the interband momentum matrix element for transitions across the spin-split gap of energy E_g (broadened by a phenomenological damping constant γ_g), and $q/2$ the light wave number which we assume equal for the incident and scattered radiation fields.

Figure 1 shows the spectral distribution of oscillator strength for spin-density transitions calculated in conditions of weak resonance from the discrete spectrum of miniband states shown in the inset. The scattering intensities of this figure were obtained from Eqs. (1)–(3) assuming a light wave-number transfer $q = 0.685 \times 10^6$ cm^{-1} deliberately chosen to be comensurate with the proposed discretization of the minibands. For reasons of clarity, a large number of transitions with very small oscillator strength was supressed from the graph. For comparison, the corresponding spectral distribution for a uniform electron gas with the same three-dimensional (3D) density and satisfying the same boundary conditions is indicated by the empty squares. By conditions of weak resonance, we understand a limit imposed on the frequency of the incoming radiation such that the energy difference between the spin-split edge and the energy of the light is large in comparison to typical energy differences among subband states. In these circumstances, one may replace all energy denominators in Eq. (3) by an average enhancement factor with the consequent elimination of the intermediate sum through the completeness of the intermediate states.

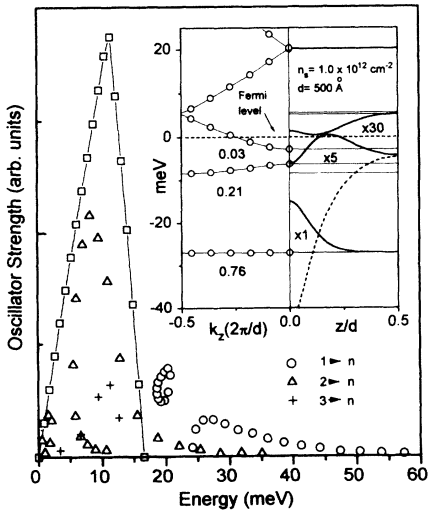

FIG. 1. Distribution of oscillator strength for spin-density transitions calculated in conditions of weak resonance as described in the text. The various symbols indicate the minibands from which the transitions originate. The empty squares are the corresponding results for a uniform system. The numbers on the left side of the inset indicate the relative populations of the minibands while the solid curves on the right side are the corresponding densities. The effective potential is indicated by the dashed line. All energies are measured with respect to the Fermi level at zero.

One of the interesting aspects revealed by the results of Fig. 1 is that in spite of the strong periodical potential which confines the majority of the electrons in the ground subband states, a considerable fraction of the total oscillator strength for spin-density transitions still remains in spectral region where such transitions are expected for the corresponding uniform electron system. Clearly, they correspond to transitions within NFE bands with a spectral density distorted by umklapp processes. As

the sum over oscillator strength weighted by the respective transition frequency amounts to the same sum in the uniform system through the f-sum rule, the remaining part of the total oscillator strength originates from transitions from the ground subband states. Because the photon wave-number transfer q is large on the scale of the Brillouin zone of the minibands, a substantial part of this oscillator strength is transferred to the high-energy side of the Raman spectrum corresponding to transitions from the ground subband states to a quasicontinuum of NFE states. We remark that 99.5% of the total oscillator strength is exausted by the transitions in the energy range of Fig. 1.

Implicit in the results of Fig. 1 is the assumption of complete translational invariance along directions parallel to the doping sheets so that each spin-density transition represents a coherent superposition of vertical single-pair excitations. Of course, an excitation of this kind cannot be stable in the real system since it may be scattered to an overlapping continuum of nonvertical single-pair excitations via random potential fluctuations originated in the planes of the impurities. As has been suggested in previous studies of low-mobility GaAs-$Al_xGa_{1-x}As$ quantum wells,[12] the decay mechanisms implied by this coupling is likely to be dominated by one-electron relaxations similar to those that limit electron mobilities for transport along the layers. The decay time due to these relaxations, however, is to be distinguished from the relaxation time that can be deduced from a transport mobility.[13] While the former represents the probability per unit time of a carrier being scattered to any of the energy conserving momentum states, the latter weights this probability by a factor which supresses scattering in the forward direction. While large differences between these two scattering times is expected to exist in high-mobility quantum well structures,[13] this is not the case for the low-mobility structures of this paper for which Shubnikov–de Haas measurements indicate transport relaxation times about 2.5 times larger than the one-electron relaxation times.[14] Of course, the correct description for the decay of a spin-density excitation would involve the knowledge of a corresponding broadening spectral function, a quantity whose precise form is not known. Therefore, for the purpose of this paper, we represent these spectral functions by Lorentzians with broadening factors characterized by the minibands from which the various spin-density transitions originate. For transitions originating in the lowest subband states we assume a broadening factor $\gamma_1 = 11$ meV consistent with a transport mobility of the order of 2000 $cm^2/V\,s$ that was observed in the mobility spectrum of samples with sheet doping densities comparable to that of this paper.[2] For transitions originating in the higher miniband states for which no transport mobilities are avaliable, a common broadening factor γ_2 was assumed for all NFE transitions and taken as an adjustable parameter to be fitted to the experimental data.

In conditions of extreme resonance, when $\hbar\omega_L \approx E_g$, the previous calculations become clearly invalid and the details of the energy level structure of spin-split holes become important. Assuming that holes are subjected to the electrostatic (Hartree) part of the self-consistent potential, we have also calculated within the effective mass approximation the energy level structure and wave functions for states in the spin-split band using the same boundary conditions adopted for the conduction-band electrons. As can be judged from the results shown in the inset of Fig. 2, the presence of the space-charge potential exerts a profound influence on important low-lying intermediate states contributing to the resonant light scattering process for which exchange and correlation effects tend to reduce the interband optical gap. Note that hole energies are plotted in the upward direction with the zero of the energy scale set at the Fermi level. From these results, the oscillator strength associated to each transition frequency of Fig. 1 was recalculated through Eqs. (1)–(3) and the results convoluted with Lorentzian spectral line shapes as discussed in the previous paragraph. In these calculations, the completeness of the intermediate states was verified to be better than one part in 10^4 for transitions in a Raman frequency interval of 70 meV, by taking 13 hole minibands spaning a range of about 200 meV below the top of the spin split-off band. In total, more than 2000 matrix elements were calculated. The continuous lines of Fig. 2 indicate the result of such computations, while the discrete points indicate the results of our measurements on a 10-period GaAs δ-doped sample with nominal doping parameters identical to those used in the calculations.[15] The present Raman measurements were taken in a nearly perfect $z(x', y')\bar{z}$ backscattering configuration with the sample mounted on the cold finger of a He cryostat at 6 K, using the variable frequency output of dye laser with DCM as a dye pumped by all lines

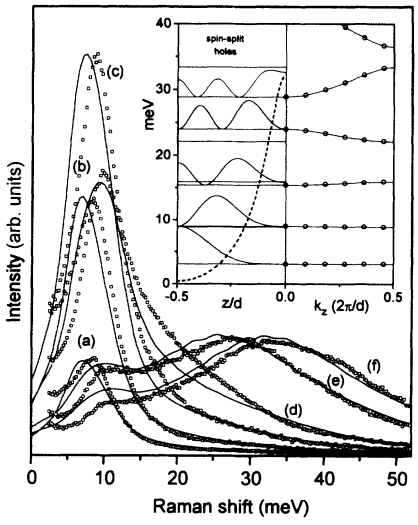

FIG. 2. Scattering cross sections for spin-density transitions for various laser energies obtained under conditions of extreme resonance with the $E_0 + \Delta_0$ edge: (a) 1856.7 meV, (b) 1865.0 meV, (c) 1873.2 meV, (d) 1882.1 meV, (e) 1899.1 meV, (f) 1907.8 meV. The discrete points indicate experimental data while the solid lines indicate the corresponding theoretical results. The energy level structure of spin-split holes are shown in the right side of the inset. The left side shows the probability densities at $k_z = 0$. The effective (Hartree) potential for holes is indicated by the dashed line.

of an Ar-ion laser. A double monochromator equipped with standard photocounting electronics was used to detect the scattered radiation. The theoretical results were fitted to the experimental data by adjusting both the width and the peak intensity of the curve labeled (*c*) of Fig. 2. The remaining input parameters were kept fixed and given by those compiled in Ref. 16 for bulk GaAs. The broadening factor resulting from this fitting, e.g., $\gamma_2 = 2.3$ meV, is physically reasonable to the extent it suggests a transport mobility of about 9300 cm^2/Vs for the second and higher miniband states.[2] The remaining theoretical curves of Fig. 2 follow as consequence of this fitting without further adjustings on the parameters. Concerning the discrepancies of about 1.5 meV observed in the peak positions of the lower energy structures, we remark that much better agreement with experiment is obtained through the use of an increased sheet carrier density, consistent with an estimated 20% increase in electron density due to charged acceptor neutralization in the depletion layers during laser illumination.[14] The same is found for an increased width in the distribution of donor charge provided it remains less than the quantum-mechanical width of the electron charge distribution, i.e., between 0 and 120 Å.

In conclusion, we have investigated the resonant Raman spectra of spin-density transitions in a system where the coexistence of two- and three-dimensional electron gases is explicltly manifested in the spectral line shapes—notably, a lower energy structure due to a NFE gas similar to that found in the resonant spectra of free electrons in bulk GaAs (Ref. 17) and broad bands centered at higher energies due to a continuum of intersubband transitions originating in a layered 2D gas which resonates at higher laser energies due to the spatial separation of electrons and holes. Owing to the low mobility of electrons in the ground miniband states, no discernible structure is predicted to exist in the region of the $1 \rightarrow 2$ miniband transitions contrasting with the conclusions of Ref. 7 where a resonance structure at 17 meV seen in the polarized Raman spectra was attributed to these transitions. In our view, such a structure arises from a collective plasmalike excitation of the 3D gas whose existence we have demonstrated in this paper.

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[17] See, e.g., G. Abstreiter, M. Cardona, and A. Pinczuk, in *Light Scattering in Solids IV*, edited by M. Cardona and G. Guntherodt (Springer, Berlin, 1984), p. 5.