However, within its limitations it is quite illuminating. We have been able to show that (i) the second moment of $\chi''(\vec{q}, \omega)/\omega$ gives a measure of the mixing energy, allowing us, in principle, to establish a connection between inelastic neutron scattering and cohesive-property data; (ii) the $\vec{q}$ dependence of $\chi''(\vec{q}, \omega)$ is weak, even for a fully "coherent" intermediate valence paramagnet; (iii) the success of Lorentzian fittings implies very fast ($\tau < 10^{-15}$ s) correlation times for the torques $\dot{\sigma}^z$; and (iv) there can be no diffusive modes in the paramagnetic phase. It seems, in addition, that only very precise measurements, from which the fourth moment of $\chi''(\vec{q}, \omega)/\omega$ could be reliably determined, would allow us to distinguish incoherent ($\vec{q}$-independent) from coherent ($\vec{q}$-dependent) intermediate valence. We suggest that both TmSe and $Ce_{1-x}Th_x$ fall into the former category, but for different reasons: thermal disorder in the case of TmSe and static potential disorder in the case of $Ce_{1-x}Th_x$.

The author would like to acknowledge useful discussions with Professor Erdös, Dr. G. Solt, and Mr. H. Razafimandimby.

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Inelastic Light Scattering from a Quasi-Two-Dimensional Electron System in GaAs-Al_xGa_{1-x}As Heterojunctions

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(Received 21 December 1978; revised manuscript received 28 March 1979)

Resonance enhanced inelastic light scattering from a quasi-two-dimensional electron gas confined at the interface of abruptly doped GaAs/n-Al_xGa_{1-x}As heterojunctions has been measured. The samples were fabricated using molecular-beam epitaxy with a high-contrast doping technique. The results show strong evidence for intersubband excitations in a two-dimensional electron system.

We report the observation of resonant Raman scattering from a quasi-two-dimensional electron system which is confined at the interface of GaAs/n-Al_xGa_{1-x}As heterojunctions. The experiments were performed in backscattering geometry using an exciting laser frequency at resonance with the $E_0+\Delta_0$ energy gap of GaAs. Recently it has been shown that at this resonance one can observe free-carrier excitations with electron densities as low as about 1×10^{11} cm^{-2}.[1] It has also been suggested that the resonance enhancement of the carrier-density light-scattering mechanism should enable one to observe electron-gas excitations at semiconductor surfaces and interfaces.[2]

The existence of two-dimensional electron-gas systems at certain GaAs/$Al_xGa_{1-x}As$ heterostructures has been demonstrated in a recent publication.[3] High mobility of electrons in heterojunction superlattices has been achieved using a modulation doping technique during growth with molecular-beam epitaxy.[4] The heterojunctions studied in the present work are fabricated by a similar technique. The samples consist of a thick $Al_xGa_{1-x}As$ layer covered by a thin GaAs layer grown successively on a nominally undoped (100) GaAs substrate. The $Al_xGa_{1-x}As$ layers, with typical values $x \simeq 0.20$, are intentionally doped either with Sn or with Ge. Doping levels of the order of $n \simeq 1\times10^{18}$ cm^{-3} are easily achieved. The doping source is terminated abruptly synchronous with the Al source.

Using Sn impurities, a segregation at the $Al_xGa_{1-x}As$ surface is observed,[5] which causes a smearing out of the dopant into the GaAs layer yielding heavily doped n-GaAs. Abrupt profiles are obtained with Ge impurities. However, because of a "memory effect" some Ge is incorporated in the GaAs layer. Therefore, although diffusion can be neglected at the growth temperature of 550°C, the GaAs layers of Ge-doped samples have a carrier concentration of $n \simeq 10^{17}$ cm^{-3}.

The Al concentration of the order of 20% results in a band-edge discontinuity of about 200 meV. The donor binding energy in $Al_xGa_{1-x}As$ is assumed to be much smaller than this value. The conduction-band edge of GaAs therefore lies lower than the impurity states in $Al_xGa_{1-x}As$. The electrons from the donors will move into the GaAs layer and form an accumulation layer close to the interface. The $Al_xGa_{1-x}As$ layer will be depleted towards the interface. This behavior of the conduction and valence bands is shown schematically in Fig. 1(a). At the surface of n-GaAs there exists a depletion layer which is caused by a Fermi-level pinning about 1 eV below the conduction-band edge. The width of this depletion layer depends on the carrier concentration.

At the GaAs/$Al_xGa_{1-x}As$ interface the charge carriers are confined in a one-dimensional potential well, similar to an accumulation layer in metal-oxide-semiconductor structures. The electrons are bound perpendicular to the interface. For an isotropic conduction band the energy eigenstates are given by

$$\epsilon_i(k) = \epsilon_i + \hbar^2 k_{\parallel}^2/2m^* \quad (i = 0, 1, 2, \ldots),$$

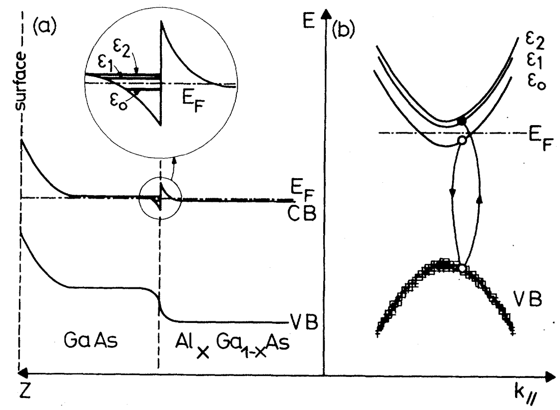

FIG. 1. (a) Energy-band disgram for abrupt GaAs/n-$Al_xGa_{1-x}As$ heterojunction. The conduction-band behavior at the interface is enlarged. ϵ_0, ϵ_1, and ϵ_2 denote the bottom of the electric subbands in the one-dimensional potential well. (b) Possible intersubband excitation process in a two-dimensional electron system. The valence band is shaded indicating the band bending towards the interface.

where the first term denotes the bottom of the electric subbands and the second one the free energy parallel to the interface. m^* is the effective mass of the electrons and $k_{\parallel}$ the wave vector parallel to the interface. For accumulation layers in silicon the energy values ϵ_i have been calculated self-consistently, for example, by Stern[6] and Ando.[7] Spectroscopic studies at far infrared frequencies[8] are in reasonable agreement with such calculations and demonstrate that many-body interactions play an important role.

Self-consistent calculations of the subband splitting in GaAs accumulation layers are not yet available. The energy separation $\epsilon_1 - \epsilon_0$ depends on various properties of the heterojunctions as there are the band-gap discontinuity (Al concentration), the acceptor and donor concentration in the GaAs layer, the density of the interface states, and the number of free carriers in the accumulation channel. We want to demonstrate that inelastic light scattering can be used to determine some of these parameters as well as the subband splitting in the accumulation layer itself.

Raman scattering of free carriers in GaAs has been proven to be a useful technique to study single-particle and collective excitations.[1] The selection rules and scattering mechanisms have been discussed by Mooradian.[9] The present work concentrates on single-particle excitations which involve spin flip. Spectra of this type from bulk carriers in n-GaAs have been reported, for example, in Ref. 1. The spin-flip single-particle excitations are antisymmetric. The polarization of the scattered light is perpendicular to the polarization of the incident light. The scattering cross section is strongly enhanced at the $E_0+\Delta_0$ energy gap.[1]

Burstein, Pinczuk, and Buchner[2] have discussed the possibility of resonant inelastic light scattering by charge carriers in quasi-two-dimensional systems. The scattering process for a two-step single-particle excitation involving spin flip is shown schematically in Fig. 1(b). The valence band is smeared out indicating a band bending towards the interface. The conduction band is split into the subbands ϵ_i. In true backscattering geometry the scattering wave vector parallel to the interface is zero. The three-dimensional single-particle excitation spectrum transforms into intersubband excitations. The frequency shifts of the observed peaks directly correspond to the energy separation of the subbands.

Figure 2 displays a set of Raman spectra obtained of a GaAs/n-$Al_xGa_{1-x}As$ heterojunction. The sample consists of a highly doped n-$Al_xGa_{1-x}As$ layer and a 1500-Å-thin GaAs layer grown subsequently on a nomially undoped GaAs substrate. A semitransparent gate electrode (Ni) has been evaporated onto the thin GaAs layer. The exciting laser line was chosen to be close to the $E_0+\Delta_0$ energy gap of GaAs. At this wavelength the penetration depth in GaAs is about 3000 Å. Spectrum a is obtained in the $z(yy)\bar{z}$ backscattering configuration; the polarization of the incident and scattered light are parallel to each other. In this configuration spin-flip, single-particle excitations are not allowed. Instead we observe resonance-enhanced collective excitations (coupled plasmon–LO-phonon modes L_- and L_+ from bulk GaAs) and the pure LO-phonon modes of GaAs (LO) and $Al_xGa_{1-x}As$ (LO_1 and LO_2). This spectrum is used to characterize the sample. From the position of LO_1 and LO_2 we determine $x=0.20$.[10] The L_- and L_+ modes are sensitive to the bulk carrier concentration of the GaAs layer.[10] In the presently studied sample we find $n=2\times10^{17}$ cm^{-3}. The LO mode originates from the depletion layer at the GaAs surface, which is of the order of 800 Å wide. The results obtained in $z(xy)\bar{z}$ configuration are shown in Fig. 2, spectra b. In this configuration we observe resonance-enhanced single-particle excitations similar to the work of Ref. 1. The single-particle excitation band of bulk carriers cuts off

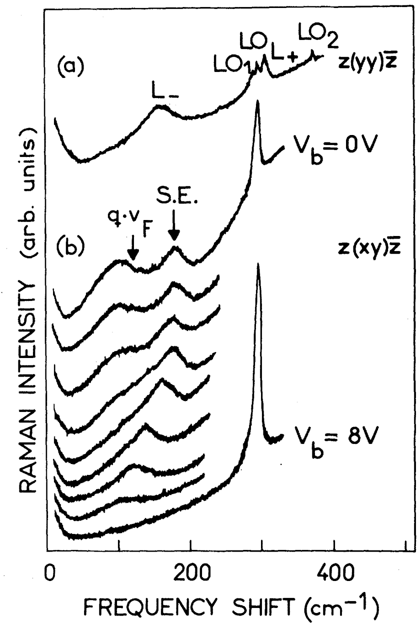

FIG. 2. Raman spectra in two scattering configurations of a GaAs/n-$Al_{0.20}Ga_{0.80}As$ heterostructure at $T=2$ K. The exciting laser frequency is $\lambda_0=6471$ Å (1.92 eV). V_b is the voltage applied across a Ni Schottky barrier in backward direction. The voltage steps in the spectra shown in b is $\Delta V=1$ V. The peak labeled S.E. is interpreted as subband excitation of the electron accumulation layer at the interface (see discussion in the text).

around $\vec{q}\cdot\vec{v}_F\simeq120$ cm^{-1}, where q is the scattering wave vector ($\simeq0.7\times10^6$ cm^{-1}) and v_F the Fermi velocity. In addition to the bulk single-particle scattering we observe the LO-phonon mode of the GaAs depletion layer (295 cm^{-1}) and a third peak at 180 cm^{-1} (labeled S.E.). We believe that this peak is caused by single-particle subband excitations (S.E.) of the two-dimensional electron gas confined at the GaAs/$Al_{0.20}Ga_{0.80}As$ interface.

This interpretation becomes more evident when one studies the voltage dependence of the spectrum (Fig. 2, spectra b). A voltage V_b applied in a backward direction across the Schottky barrier increases the depletion width. Therefore the bulk single-particle excitation peak decreases in intensity and vanishes at $V_b>3$ V. The intensity of the LO-phonon mode of the surface depletion region increases, while the subband excitation peak remains unchanged. A further increase of V_b, however, results in a shift of the band at 180 cm^{-1} to smaller wave numbers and to a decrease in intensity. It disappears completely when $V_b>7$ V.

Then the accumulation layer at the interface is also depleted. The voltage which is necessary to empty the accumulation channel depends on the power of the incident laser radiation. This is due to the screening of the depletion field by the excited photocarriers in the GaAs layer. Extrapolating the voltage to zero laser power we determine the carrier concentration in the accumulation layer $n_s \simeq 1.2\times10^{12}$ cm^{-2} at $V_b = 0$ V. This is in reasonable agreement with the value estimated from the doping level of the n-$Al_{0.20}Ga_{0.80}As$ layer and the barrier height at the interface ($n_s \simeq 1.5\times10^{12}$ cm^{-2}).

We have also studied samples with thinner GaAs layers. For $d_{GaAs} < 1000$ Å we do not observe any scattered light from bulk carriers because the natural depletion layer eliminates all except the accumulation layer electrons. The subband excitation peak disappears when $d_{GaAs} < 400$ Å. These results exclude the $Al_xGa_{1-x}As$ layer and the GaAs substrate as possible sources of the scattered light observed in thicker GaAs layers. In samples with higher Al concentration ($x = 0.36$ and 0.56) the subband excitation peak is shifted to smaller wave numbers (80–110 cm^{-1}). The lattice mismatch between GaAs and $Al_xGa_{1-x}As$ gets larger when x is increased. Therefore probably more carriers are trapped in interface states, which results in a smaller subband splitting.

Further evidence that the observed peaks are caused by the two-dimensional electron system is the resonance behavior of the scattering intensity. Plotting this intensity versus laser excitation energy, we find that the resonance enhancement peak of the subband excitations is about 10 times wider than the resonance of the single-particle excitations of bulk carriers. This is explained by the band bending of the valence band towards the interface (see Fig. 1). The half-width of the resonant subband excitation peak of the sample studied most extensively (Fig. 2) is 120 meV and the peak position is 1.91 eV.

In summary, we have demonstrated that under resonance conditions it is possible to observe inelastic light scattering from a quasi-two-dimensional electron system. This is the first observation of subband excitations using Raman spectroscopy. For a quantitative analysis of the experimental data one needs self-consistent calculations of the subband splittings which are not available so far. Effects on the line shape due to impurity-induced subband excitations with finite $k_\parallel$ should also be considered in such an analysis. The measurements reported here promise to be a useful tool in studying both single-particle and collective excitations of various space-charge layers and heterojunction superlattices which are of technological and fundamental interest.

We would like to acknowledge the expert help in sample preparation of A. Fischer and the assistance in performing the Raman scattering experiments of M. Berger and A. Köhler. One of us (G.A.) wants to mention useful and stimulating discussions with M. Cardona, G. Döhler, and A. Pinczuk. Part of the work was supported by the Bundesministerium für Forschung und Technologie of the Federal Republic of Germany.

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