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Nonequilibrium luminescence at the $E_0 + \Delta_0$ gap in GaAs with Si- δ doping

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We studied the light scattering spectra of three molecular beam epitaxy GaAs samples with Si- δ doping. A broad feature appears in these spectra which is similar to that attributed by other authors to resonant Raman scattering by electronic intersubband transitions. By studying the dependence of this emission on exciting laser photon energy we believe that this line is really produced by nonequilibrium luminescence at the $E_0 + \Delta_0$ gap.

I. INTRODUCTION

The technique of introducing very thin Si layers (on the monolayer scale) in molecular beam epitaxy (MBE) deposited GaAs leads to a potentially interesting situation where electrons (but not holes) are confined in a narrow V-shaped potential.¹ One of the possible optical techniques to obtain information about the quantized energy levels resulting from this confinement is that of resonant Raman scattering by electronic excitations produced by direct intersubband transitions.² Here, the frequency shift of the scattered light at which a given Raman feature appears gives a direct measure of intersubband level spacing. In this type of experiment the photon energy of the incident laser light is tuned to some optical gap, which for GaAs is usually the $E_0 + \Delta_0$ transition at 1.862 eV ($T = 2$ K). This technique was successfully employed to study intersubband separations in a large variety of semiconductor microstructures.³ Its application to GaAs samples with Si- δ doping,⁴ however, yields results which are not as clear-cut as those previously obtained for GaAs/AlGaAs quantum wells and superlattices.^{2,3} Using laser frequencies slightly above the $E_0 + \Delta_0$ optical gap, Abstreiter *et al.*⁴ observe a very broad structure in the Raman spectrum of δ -doped samples. They assign bumps and shoulders in this broad spectral feature to intersubband transitions, comparing their energy (measured by the Raman shift of these shoulders) to self-consistent model calculations. Under similar circumstances, Maciel *et al.*⁵ report structures at lower frequency shifts which are also attributed to electronic Raman scattering by intersubband transitions, but bear little resemblance to the features reported by the previous authors. Recently, Wagner *et al.*⁶ have reported resonance Raman measurements on δ -doped structures. The features in the spectra that they assign to light scattering by intersubband transitions, are superimposed onto a large background sig-

nal. This background was suggested to be produced by the $E_0 + \Delta_0$ luminescence recombination.

Analysis of the observed spectra is complicated by the presence of a dense electron gas and a high concentration of dopants. Such concentrations have been known to contribute to light emission spectra of uniformly doped bulk semiconductors. Of these contributions we would like to single out, for later discussion, the above-gap luminescence which occurs in n -type materials near $E_0 + \Delta_0$, the spin-orbit split component of the direct gap. The photoluminescence spectrum of n -GaAs crystals contains, along with an intense edge luminescence at about 1.52 eV, a significantly weaker band at about 1.86 eV. It has been shown⁷ that this band results from recombination of photoproduced holes in the split-hole (Γ_7) band with electrons from the conduction band. Luminescence excitation experiments with polarized light on n -type III-V semiconductors can yield interesting information about energy, momentum, and spin relaxation of the hot holes.⁸⁻¹⁰ This phenomenon of nonequilibrium luminescence also occurs at the E_1 and $E_1 + \Delta_1$ optical gaps of heavily doped n -type Ge.¹¹ Resonant light emission in p -type Ge and GaAs was observed in the region of the E_1 and $E_1 + \Delta_1$ optical transitions.¹²

Bearing this in mind we performed a series of light scattering experiments on two types of MBE GaAs samples with Si- δ doping. One sample (A) contains a single δ layer at approximately 150 Å from the surface, while two others contain 15 such layers separated by 400 Å of GaAs from one another. The spectra were taken at low temperatures (10 K) with variable excitation laser frequencies. Some spectra show structures at low Raman shifts similar to those reported by Maciel *et al.*⁵ In our case they were seen to be produced by dye laser luminescence and to bear no relation to emission from the sample. For certain laser frequencies ($\hbar\omega_L$) we observed broad structures similar to

FIG. 1. Light emission spectra observed for sample A (single δ layer) at 10 K with different incident laser wavelengths near the $E_0 + \Delta_0$ GaAs energy gap.

those reported by Abstreiter *et al.*⁴ Careful examination of the dependence of this spectral feature upon polarization and $\hbar\omega_L$ leads us to believe that it originates in nonequilibrium luminescence at the $E_0 + \Delta_0$ gap induced by the high electron concentration and cannot be attributed to electronic Raman scattering by intersubband transitions.

II. EXPERIMENT

Our experiments were performed in the backscattering configurations $z(y'y')\bar{z}$ and $z(x'y')\bar{z}$, where $x' = [110]$, $y' = [\bar{1}\bar{1}0]$, and $z = [001]$. The sample was glued to the cold finger of an Oxford cryostat at a temperature of $T = 10$ K measured by a sensor close to the sample. Laser frequency was varied continuously in the vicinity of the $E_0 + \Delta_0$ optical gap, using a jet-stream dye laser with DCM (Lambda Physik, Göttingen) dye, which was pumped by an Argon ion laser. Some measurements were also performed off resonance, with the 5145 Å line of this ion laser. The scattered radiation was analyzed by a Dilor XY triple spectrometer equipped with an intensified cooled photodiode array detector. The samples were deposited by MBE on semi-insulating (100) GaAs substrates. Sample A contains a single δ layer of Si buried 150 Å below the sample surface with a carrier density of $n_s \approx 8.74 \times 10^{11} \text{ cm}^{-2}$ and a room-temperature mobility of $\mu = 1.8 \times 10^3 \text{ cm}^2/\text{V s}$. Samples B and C contain 15 repetitions of this δ layer separated from one another by 400 Å and with 200 Å GaAs capping layer. Their carrier concentrations (room-temperature mobilities) are $n_s \approx 8.15 \times 10^{12} \text{ cm}^{-2}$ ($\mu = 1.7 \times 10^3 \text{ cm}^2/\text{V s}$) for sample B and $n_s \approx 4.11 \times 10^{12} \text{ cm}^{-2}$ ($\mu = 2.3 \times 10^3 \text{ cm}^2/\text{V s}$) for sample C.

FIG. 2. Light emission observed in sample A (single δ layer) with $\hbar\omega_L = 1.919$ eV for incident and scattered radiation polarized parallel and perpendicular to each other, respectively. Note the factor 3 multiplying the lower spectrum. The arrows mark the structures that have been ascribed to Raman scattering by intersubband excitations.

III. RESULTS AND DISCUSSION

Light scattering spectra from sample A in conditions of near resonance with the $E_0 + \Delta_0$ gap are shown in Fig. 1. The lowest two curves [(a) and (b), respectively] show spectra reminiscent of those reported in Ref. 4, with a broad feature and two sharp Raman lines corresponding to the LO phonon and its overtone (2 LO), which appear prominently only in resonant conditions. The broad feature moves to lower Raman shifts, against the static background of the phonon Raman line, as the laser frequency decreases [curves (a) through (c)]. As $\hbar\omega_L$ continues to decrease [curves (c) and (d) in Fig. 1] the structure gradually disappears until, for $\hbar\omega_L \leq 1.88$ eV, only the phonon Raman lines remain in the spectrum. In Fig. 1 the spectra are displayed in absolute (rather than relative) scattered radiation photon energies to emphasize the fact that the structure does not really fade away, but that it remains centered at constant photon energy ($\hbar\omega_s \approx 1.87$ eV) regardless of the incident laser frequency. In this manner the broad peak behaves as recombination luminescence from the Fermi sea in the conduction band to a nonequilibrium hole at the top of the split-off valence band in a similar process like the $E_0 + \Delta_0$ luminescence in n -type semiconductors.⁷⁻¹⁰ The cut-off frequency ($\hbar\omega_L \approx 1.87$ – 1.88 eV) is consistent with this hypothesis taking into account a Fermi energy of a few hundredths of an eV. Another characteristic of this broad light emission peak is its lack of linear polarization. Figure 2 shows spectra for polarized and depolarized scattering which show no substantial difference in shape or position of this feature, in contrast to the phonon Raman lines which are only allowed in the $z(y'y')\bar{z}$ configuration. Excitation with linearly polarized light produces a linear combination of the two possible

FIG. 3. Luminescence spectra for sample B (15 δ layers separated one another by 400 Å) at 10 K with different incident laser energies. For $\hbar\omega_L = 2.410$ eV due to a very poor signal to noise ratio only the smoothed spectrum is shown. The signal labeled L^- is assigned to the LO-phonon plasmon coupled mode.

spin orientations for holes in the split-off band, which will quickly lose their phase coherence due to linear momentum relaxation.^{13,14} In this case, no angular-momentum alignment is possible for holes in the split-off band, and the degree of linear polarization for the recombination light should vanish as observed in our experiments. In addition we have observed weak signals at 1.871 and 1.891 eV (48 and 28 meV Stokes energy) in the $z(x'y')\bar{z}$ configuration, marked by arrows in the figure. These peaks are similar to those reported by Wagner *et al.*⁶ and have been ascribed to Raman scattering by intersubband transitions.

Figure 3 exhibits the light scattering spectra of sample B, composed of 15 δ layers of Si separated by 400 Å of GaAs from one another and with a much higher areal density of carriers. Curve (b) shows a spectrum for $\hbar\omega_L = 1.979$ eV where a broad structure similar to that observed in sample A (Figs. 1 and 2) is seen. The relative intensity of this feature to that of the one phonon scattering is much larger in sample B than in sample A. This is easily understood in terms of the multiplicity of δ layers in sample B. The spectrum of sample C (not shown) is nearly identical to that of sample B the only difference being that the broad spectral feature in the latter has twice the intensity. Both samples have the same structure, but the carrier concentration in sample B is about twice as large as that of sample C. Thus, we conclude that this spectral feature has an intensity which is approximately proportional to the carrier concentration. Also, when the degree of doping is increased, from sample A to sample B, the luminescence maximum shifts toward higher energies by ≈ 30 meV. This fact is consistent with the corresponding increase in the Fermi level.

Curve (c) in Fig. 3 shows a spectrum taken at low excitation frequency ($\hbar\omega_L = 1.833$ eV), where only the phonon Raman scattering is present. This phonon spectrum is deformed by interferences, and will be discussed

later. Now we focus our attention on the dependence of the broad light emission feature on laser excitation frequency. So far the behavior of this structure for samples B and C is similar to that of the same feature in sample A. The question remains: is this feature a resonant phenomenon or would it appear (although maybe in weaker form) for any excitation frequency larger than a given threshold? Curve (a) in Fig. 3 answers this question. Here we show the smoothed spectrum taken with the 5145 Å line of an argon laser ($\hbar\omega_L = 2.410$ eV) i.e., at energies well above the $E_0 + \Delta_0$ resonance. Although the intensity is very low the same feature at approximately the same absolute emission frequency is observed. This proves that the observed light emission is not a resonant phenomenon as is that which is discussed in Ref. 11, but has all the characteristics of the nonequilibrium luminescence from the large reservoir of electrons in the conduction band to the holes left behind at the top of the split-off valence band, such as those reported in homogeneously doped n -type semiconductors.⁷⁻¹¹ As in this case, the intensity of the luminescence line scales with the carrier concentration and the incident laser power. While the luminescence lineshape may contain information about the electronic subbands, a similar light emission feature would occur in a homogeneously doped sample. Thus, it would be difficult to extract useful information about electronic subband spacing from such spectra.

The spectra of Fig. 3 [curves (b) and (c)] show an additional feature, indicated L^- in the figure, and not present in the spectra of sample A. We ascribe this line to coupled LO-phonon plasmon vibrations. These excitations are possible by the partially three-dimensional (3D) character of the electron gas in the multi- δ layer samples brought about by interwell coupling. In fact, the measured L^- -Raman frequency in Fig. 3 is consistent with a 3D carrier density of $n_s(2D)/d = 2.04 \times 10^{18} \text{ cm}^{-3}$, where d is the spacing between δ layers in sample B.³

In summary, we believe that the broad spectral feature observed in the light scattering spectrum of GaAs samples with Si- δ doping is due to nonequilibrium luminescence at the $E_0 + \Delta_0$ optical gap rather than to Raman scattering by electronic intersubband transitions.

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