Type II GaAs/AlAs superlattices under high excitation

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Abstract: High intensity photoluminescence data on GaAs/AlAs superlattices and multi quantum wells of type II at low temperature are reported. The variation with density of the renormalized band gap reveals the competition between the Hartree energy and the many-body effects (mainly exchange). Preliminary analysis for a lot of samples with widely different periods provides a satisfactory fit with Hartree-Fock model. The kinetics of luminescence and total decay in the electron-hole plasma regime is measured. In some samples, a phase transition was observed which is tentatively interpreted in term of electron-hole liquid condensation.

1. Introduction.

GaAs/AlAs type II structures grown on (100) substrates have been well studied [1]. Their lowest electron states in the AlAs layers, issued from X valleys, are splitted by the stress from the weak lattice mismatch and also by the mass anisotropy of the X valleys: so the type II region is splitted into two subregions called R_z and R_{xy} (z being the SL growth axis) where the conduction band minimum comes respectively from X_z or X_{xy} valleys [2]. Low level luminescence in type II region has been extensively analyzed [3]. The large lifetime of excitons localized by interface roughness (μs to ms range) makes high excitation easy to achieve, using time-resolved photoluminescence spectra. A specific feature of type II structures under high excitation is the existence of a Hartree energy which comes from the lack of local neutrality, positive and negative electrical charges accumulating respectively in GaAs and AlAs layers. Experimental data [4] as well as theoretical calculations [5] of high density effects in type II SL are still scarce. Half a dozen of samples with quite different period, belonging either to R_z or R_{xy} subregions were studied (they are identified by (i/j) , i and j being the GaAs and AlAs layer widths in $\text{\AA}$). Standard line-fit procedure was used, which allows the determination of the density n and the renormalized band gap $E_g(n)$. A self-consistent calculation of the Hartree energy $E_H(n)$ was performed, to which 2d type exchange energy is added [6]. Experimental results in samples with period shorter than about 10 nm are explained in term of electron-hole plasma (EHP). In samples with larger period, a kind of phase transition was observed: the new phase is tentatively analyzed in term of electron-hole liquid (EHL).

2. EHP in short period samples.

Fig. 1a) Comparison between $\mu(n)$, $E_g(n)$ (full lines) and $E_H(n)$ (dotted line) from the theory and $\mu(n)$ (Δ) and $E_g(n)$ ($\times$) from the line-fit procedure for sample (4.2/14.6) in the R_{xy} region.

Fig. 1b) The same for sample (8.2/13.5) in the R_z region.

Let us remark that the experimental conditions (spectra taken at delay larger than 10 ns) ensure good thermalization of the EHP with the lattice which remains at a temperature close to 2K. Fig. 1 a) to c), give for three samples, one in the R_{xy} region (4.2/14.6) and two in the R_z region (8.2/13.5 and 17/67) the energies $\mu(n)$, $E_g(n)$ and $E_H(n)$ as a function of the density n from the Hartree-Fock calculations; the experimental values of $\mu(n)$ and $E_g(n)$ coming from the line shape analysis are also plotted. The theoretical value of $E_g(0)$ was adjusted in order to get the best fit between theory and experiment for $E_g(n)$. Examination of fig. 1 a) to c) shows at once the competition between the Hartree energy and the exchange energy upon the behaviour of $E_g(n)$: it decreases with n if the SL period is small enough (samples 4.2/14.6 and 8.2/13.5) but increases at large n for larger SL period (sample 17/67). Let us remark that the fit between theory and experiment on fig. 1 is related only to the renormalized band gap $E_g(n)$ since the Fermi energy $E_F(n)$ is, from the procedure, the same in the line fit and the theory. The experimental behaviour of $E_g(n)$ is rather well accounted for by the simple Hartree-Fock model.

The integrated luminescence intensity $I(n)$ was analyzed as a function of the density n . One finds an experimental law $I(n)$ like n^α with α between 3/2 and 2. On the other hand, one finds in the plasma regime a total recombination rate close to linear: EHP lifetimes τ_p (in the 50 ns range) are much smaller than the lifetime τ_{exc} of localized excitons at very low level [3]. The large difference between τ_p and τ_{exc} comes from the fact that electrons and holes in the plasma regime are necessarily mobile, whereas localized excitons are not. So, electrons and holes can find recombination centres and more likely flow towards the substrate (vertical transport). The value $\alpha=2$ is expected under the assumption of complete relaxation of the K-selection rule, which was also assumed in the line-fit procedure.

3. EHL in samples with larger period ?

In a few samples with AlAs layers in the 8-10 nm range and GaAs layer near 2.5 nm, we find at very low excitation level spectra typical of localized excitons in the R_{xy} region: phonon-assisted components dominate the spectrum. Above some threshold, the line shape changes; it is possible to reproduce it as the sum of two contributions: the spectrum of localized excitons, named line Exc and a broad line, named L line, whose shape remains constant for a large range of excitation. From now, we focus our attention on sample 23/97. Fig. 2 shows the spectrum of the two components above threshold. Fig. 3 shows the integrated intensities of these two components as a function of excitation intensity: a very

clean threshold behaviour is found, using uniform excitation. The threshold intensity was about 25 mW cm^{-2} . This behaviour suggests strongly a phase separation as in the case of gas-liquid transition in indirect gap materials like Ge and Si [7]. If we assume that the L line is due to EHL, then its density can be determined by a line-fit procedure. In the present case, a weak no-phonon component and several phonon-assisted components are used; we assume two equivalent conduction valleys X_x and X_y . A rather good fit was obtained for a density $2.6 \cdot 10^{12} \text{ cm}^{-2}$ (see fig. 4). In addition, we can compare carefully the position of Exc and L spectrum (comparing the theoretical component of the L spectrum corresponding to the 48 meV AlAs phonon with the component of Exc spectrum with the same phonon); since the bottom of the free exciton band should be around the high energy cut-off of the localized Exc component, it appears that the binding energy of the liquid could be of the order of 1 to 2 meV. The lifetime of the new phase was also measured: $13 \mu\text{s}$ (as compared to 1.04 ms for localized excitons); such lifetime, much longer than those measured in the EHP regime might indicate that the EHL forms small droplets which do not move. If so, the measured lifetime, $13 \mu\text{s}$, could be due mainly to an Auger process, as in the case of e-h droplets in Si and Ge.

Fig. 1c) The same for sample (17/67) from region R_{Xy}

Fig. 2- Spectra of the two components Exc and L of the luminescence of sample (23/97) above threshold.

Fig. 3- Integrated intensities of lines Exc and L as a function of excitation for sample (23/97).

Fig. 4- Fit of line L using a liquid density of $2.6 \cdot 10^{12} \text{ cm}^{-2}$

On the other hand, using the simplified Hartree-Fock model, we can compute $\langle E(n) \rangle$; as is well known [7], the density n_l of the liquid occurs at the minimum of $\langle E(n) \rangle$, provided $\langle E(n_l) \rangle$ is smaller than the energy of the bottom of the free exciton band E_{X0} : one finds n_l about $2 \cdot 10^{11} \text{ cm}^{-2}$, that is one order of magnitude smaller than the experimental one!

However, this value of n_l corresponds to quite large r_s value, especially for the electrons : this makes our theoretical estimate rather unreliable.

In favour of the existence of EHL, let us summarize the "pro" arguments:

- a) The qualitative behaviour supports this interpretation: sharp threshold to get the new phase, good fit of the line shape, lifetime compatible with an Auger process.
- b) Using higher excitation (under pulsed condition), one can reach a new regime where the L line begins to broaden and to move towards higher energy: this suggests that the L line is of intrinsic origin; in addition, this is reminiscent of a compressed liquid, as observed in Ge when all the available volume is already filled with the EHL.
- c) Analogous results were obtained on several samples with comparable layer widths.

The only argument "against" is our estimate of the e-h liquid density: indeed, a discrepancy by one order of magnitude seems quite strong. More generally, one might ask why stable EHL is found in samples with large period (>10 nm) where the Hartree term is strong? But there is another factor which enters into play: it was shown [8] by variational calculations that the binding energy of free excitons in type II SL decreases with increasing SL period. As a result, the comparison between $\langle E(n_l) \rangle$ and E_{x0} as a function of the period depends upon two competing processes, the result being unknown at the moment. However, let us remark that, if the variation of $\langle E(n_l) \rangle$ with the period is small enough, the region of stable e-h liquid could be on the large period side.

Recently, the observation of EHL was reported [9] in GaAs/AlAs type II SL grown on a (311) GaAs substrate. Its low level luminescence spectrum is like those of (100) samples of the R_z region. In such heterostructures, a strong and periodic corrugation occurs naturally in the layer plane, which should introduce a strong anisotropy of the energy bands in this plane. In the absence of band structure calculation for such a system, it is impossible to make any prediction about EHL properties; and so, comparison with our case is difficult.

To conclude, we believe that the experimental evidences of existence of EHL have a larger weight than the still crude theoretical estimate of the liquid density. In addition, we could not find any alternative model for the L line. So, we hope that refined calculations will explain our findings.

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