EXCITON STUDY IN AlGaAs

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1) MEASUREMENTS

The first exciton observed within this project and by using the photoluminescence technique (PL) was in the undoped (UD) $Al_xGa_{1-x}As$ sample n.er M417 @ 14 K.
With a laser power density of 50 mW/cm^2 and a monochromator resolution of 6 Å, what is shown in Fig. 1 was observed. The spectrum obtained by an higher laser power density of 250 W/cm^2 normalized to the intensity of the carbon peak is shown too. A big increase of the exciton peak is noted with the latter laser intensity. This fact comes out from a characteristic of the exciton recombination that depends on the square of the excitation intensity; on the contrary the recombination involving the carbon acceptor follows a linear dependence.

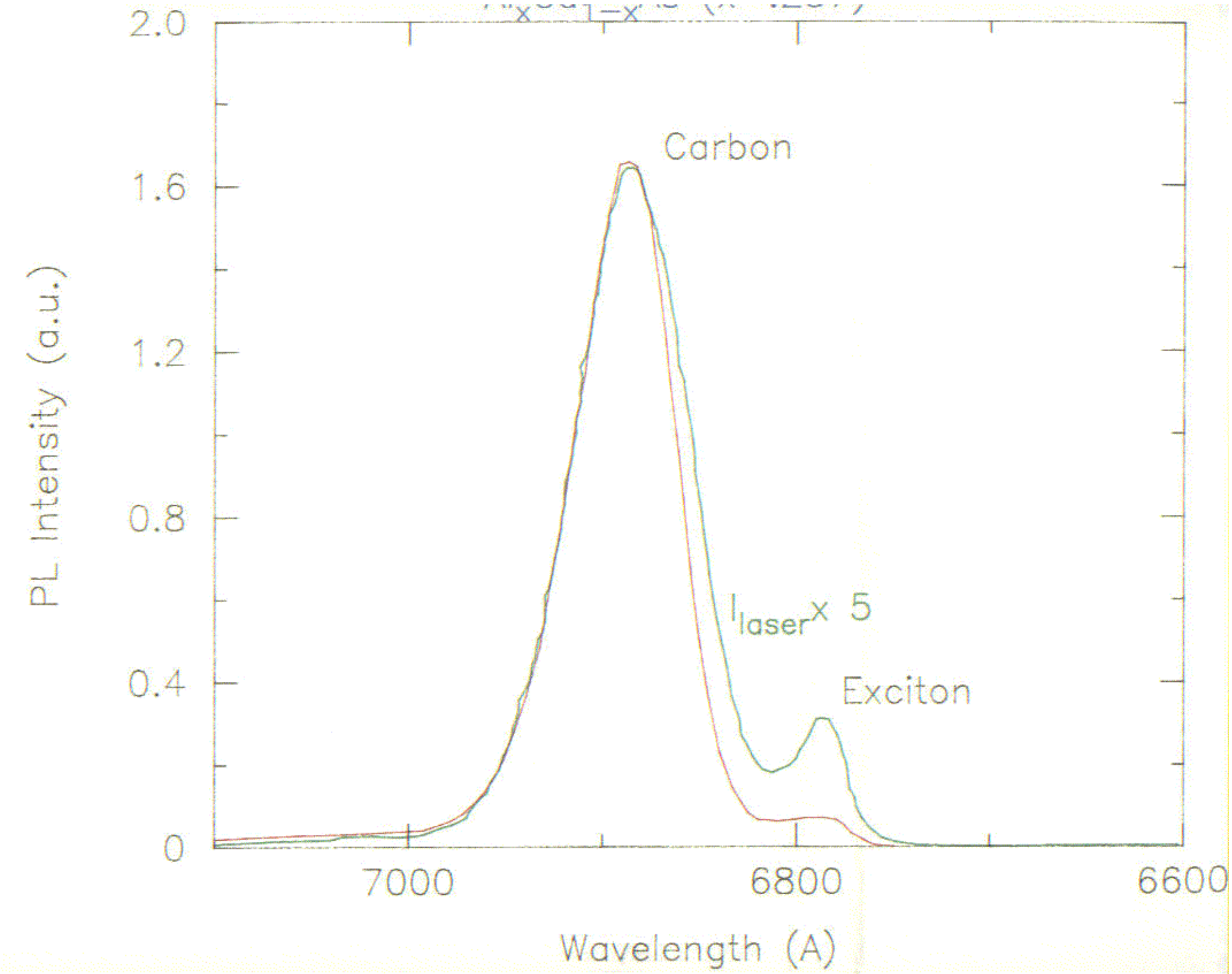

Fig. 1. PL spectrum of the M17 AlGaAs sample at 14 K.

Referring to the GaAs, this type of exciton should be the one bound to a donor [1]; its peak energy is:

$$(1.8281 \pm 0.0005)\ \text{eV}$$

The exciton full width at half the maximum (FWHM) is: 6.5 meV, little higher than the theoretical one [2]. The carbon peak energy is: 1.802 eV. The carbon FWHM is: 18 meV.
In the same figure, an evolution of a shoulder at higher energy can be noted in the carbon peak ought to the "free-to-bound" electron transition.

Other peaks have been seen in this sample and Fig. 2 shows their energy as a function of the temperature of the measurement. The energy gap (E_g) of the GaAs, shifted to the value of the E_g of our AlGaAs sample, is shown for comparison. For this sample the Al concentration, as indirectly measured by the X-ray Double Crystal Diffractometer (DCD), is 25.71%.

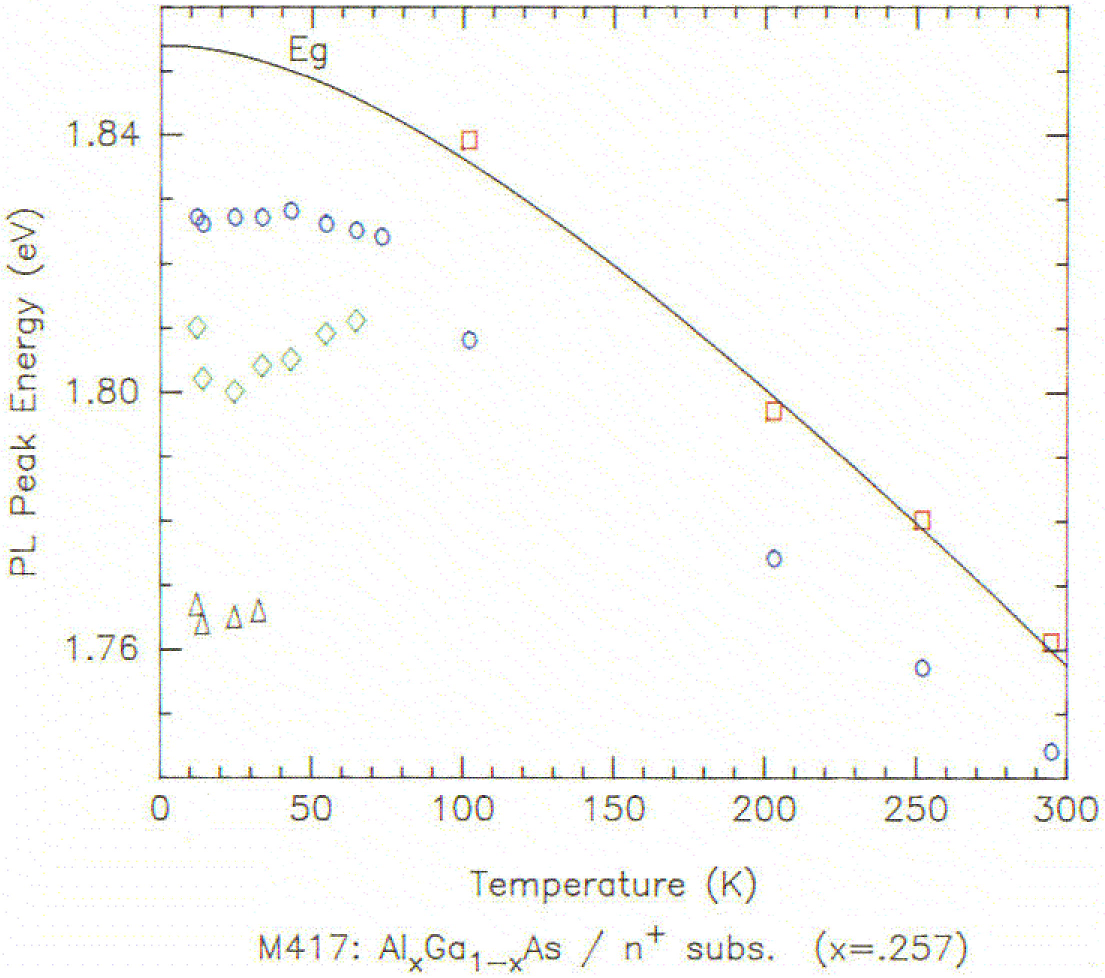

Fig. 2. Excitons energy peaks as a function of the temperature.

The second sample where the bound exciton (BE) peak has been observed is the UD AlGaAs n.er M420, of which the aluminum concentration measured by DCD is 15.00%.
Fig. 3 shows the PL spectrum at 11 K along with the Gaussian fit of the exciton peak; the monochromator resolution is 1.2 Å. In AlGaAs the shape of the PL exciton peak is a Gaussian [2], instead of a Lorentzian as for the GaAs; this is due to the statistical distribution of the aluminum atoms in the alloy.

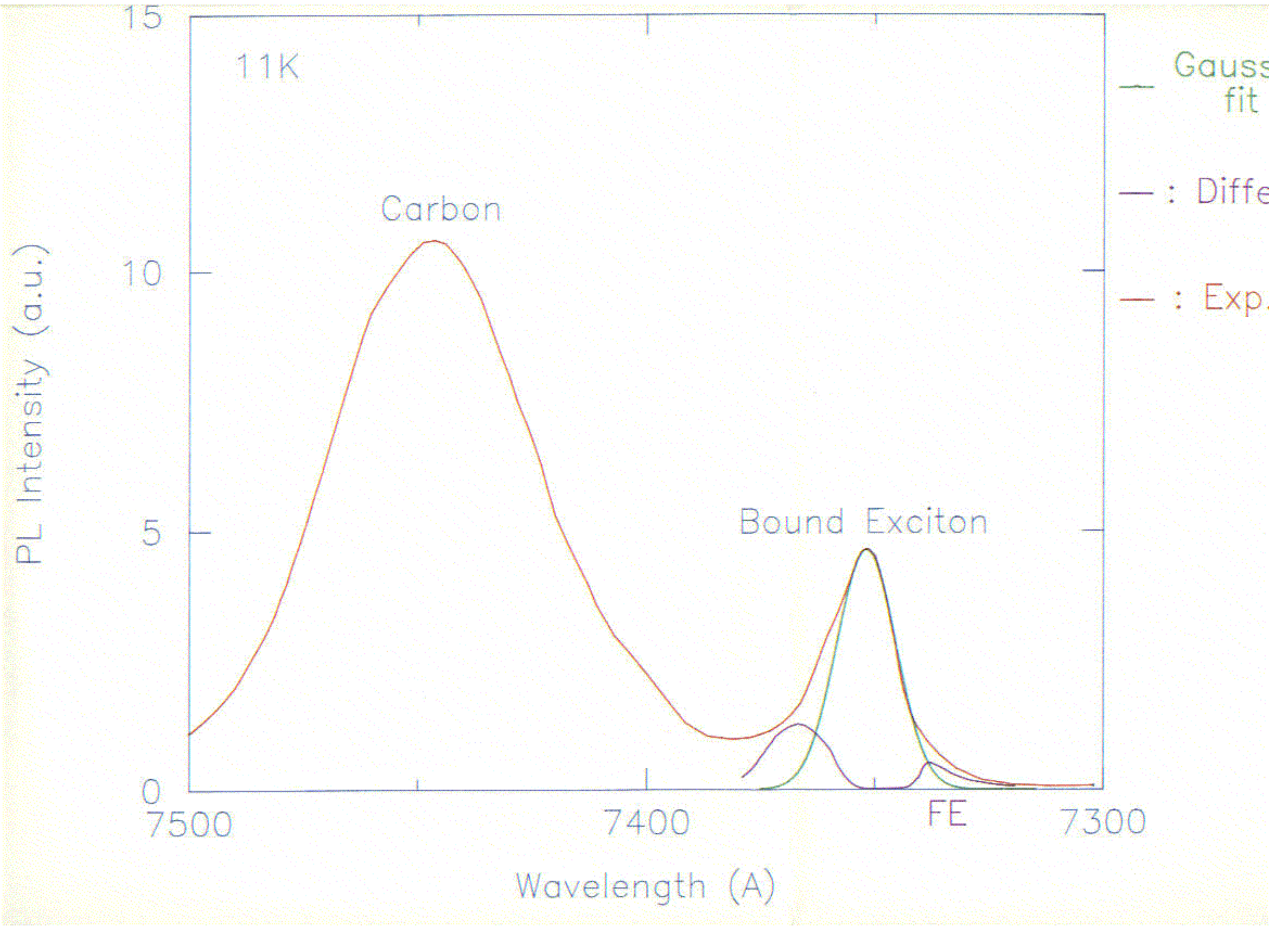

Fig. 3. PL spectrum of the M420 AlGaAs sample at 11 K.

By the difference between the experimental and the fitting curve two other peaks are resolved, of which the higher energy one, located at 3.4 meV above the BE, corresponds to the free exciton recombination peak [4] and in analogy with the GaAs exciton peak disposition [11], the other should belong to the exciton bound to the carbon acceptor.
The exciton peak position is:

$$(1.6857 \pm 0.0005)\ \text{eV}$$

and its FWHM is: 4.2 meV, again little higher than the theoretical one [2]. The carbon peak is located 21.2 meV below the BE, and its FWHM is 17 meV.

In Fig. 4 the PL peak energy vs. temperature plot is shown for this sample: the blue circles are the energy peaks of the exciton, probably till 60-70 K where they merge in something not well understood so far and that lasts till RT at least. The peaks located roughly at 1.67 eV presumably belong to the electron-to-carbon and donor-to-carbon recombination transitions. Again here the GaAs energy gap corrected to the AlGaAs E_g is shown for a comparison.

The BE recombination peak has been seen in the UD AlGaAs sample M373 too.
Fig. 5 shows this peak at 13 K with two different laser excitation intensities: 40 W/cm^2 and 80 W/cm^2; again the monochromator resolution is 1.2 Å. Indeed the FWHM is very large for an exciton (~ 23 meV), but its behavior with the laser intensity is the right one. What is remarkable now is the aluminum concentration, measured by DCD to be 42.12%.
The BE energy peak position is:

$$(2.044 \pm 0.001)\ \text{eV}$$

The carbon energy peak position is: 1.985 eV and its FWHM is: 36 meV.

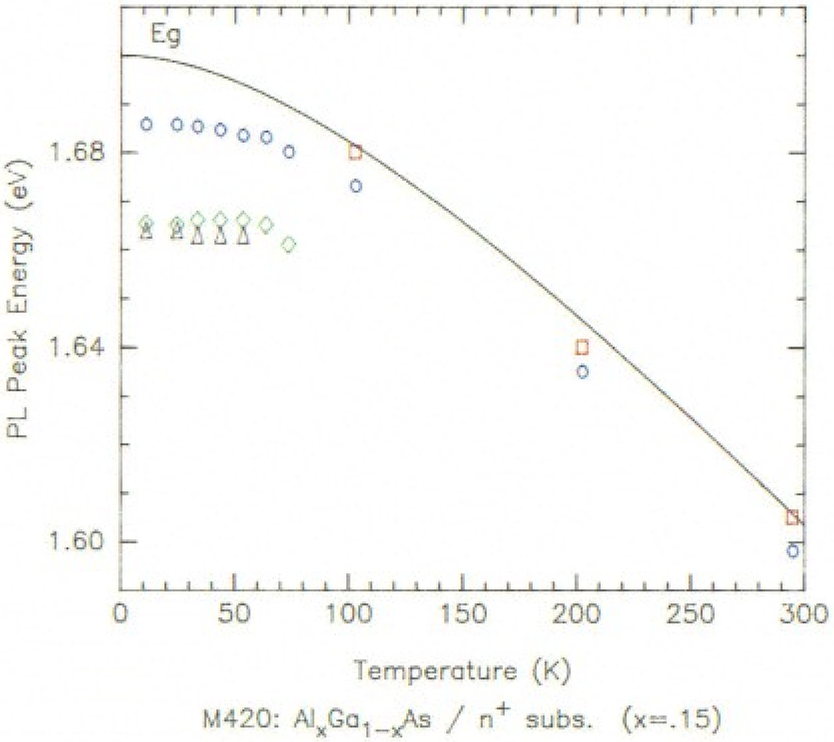

Fig.4. PL peak energy as varying the temperature of the sample M420.

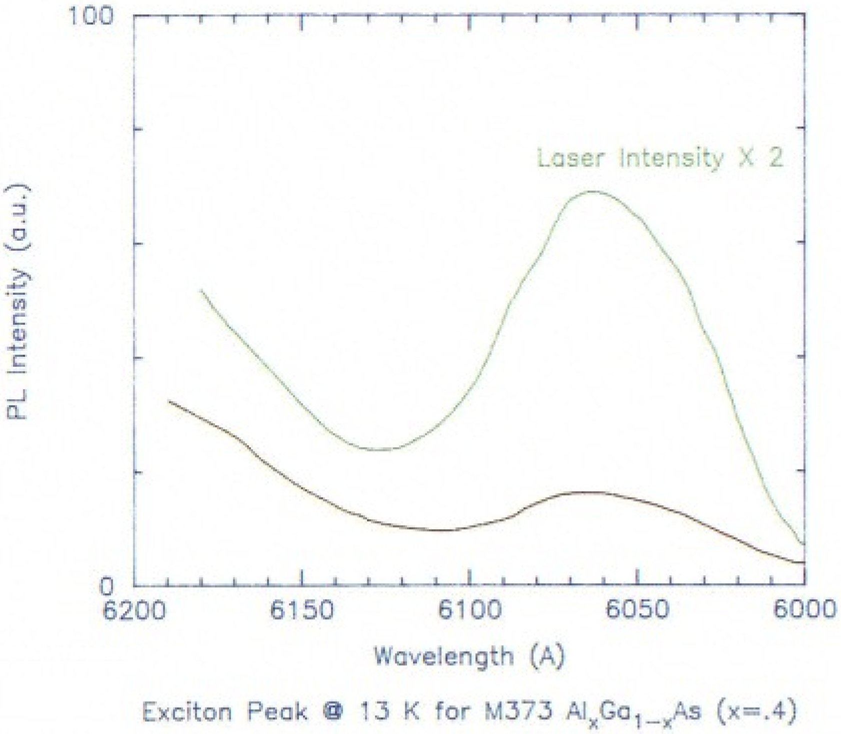

Fig.5. Exciton peak @ 13 K for the M373 sample at two intensities of the laser.

2) THEORY

To estimate how much the energy distance of the level of an exciton bound to a neutral donor is from the bottom of the conduction band (E_g) at different value of aluminum concentration in AlGaAs, I tried to calculate both the dissociation energies of the free exciton (FE) and of the complex BE; then from the sum of the two it should be possible to obtain an indication of how deep the BE level is located in the forbidden gap.

In the covalent crystals the FE is well described by the Mott & Wannier exciton that is derived by an hydrogen like model [5], i.e. the electron-hole system is considered like an hydrogen atom, but taking into account the dielectric constant of the material (ε) and the effective mass (m^*).

The theory is well known and so it is not worthy to spend time on it, but it is important to know the distance of the energy levels of this system below the conduction band, in particular:

$$E_n = \mu e^4/(2\hbar^2\varepsilon^2 n^2)$$

where $\mu = 1/m^*_{\mathrm{e}} + 1/m^*_{\mathrm{h}}$, e is the electron charge and n is the quantum number (for the ground state $n = 0$). If we consider the dielectric constant [6,7] and the electron and hole effective masses [8,9] of the AlGaAs having a linear dependence on the aluminum content we obtain what is shown in Fig. 6. In the same figure the experimental values collected by Reynolds (1986) [10] and Pearah (1985) [1] are drawn. They are far from the theoretical curve and the reason is not clear: they look like BE more than FE energy transitions.

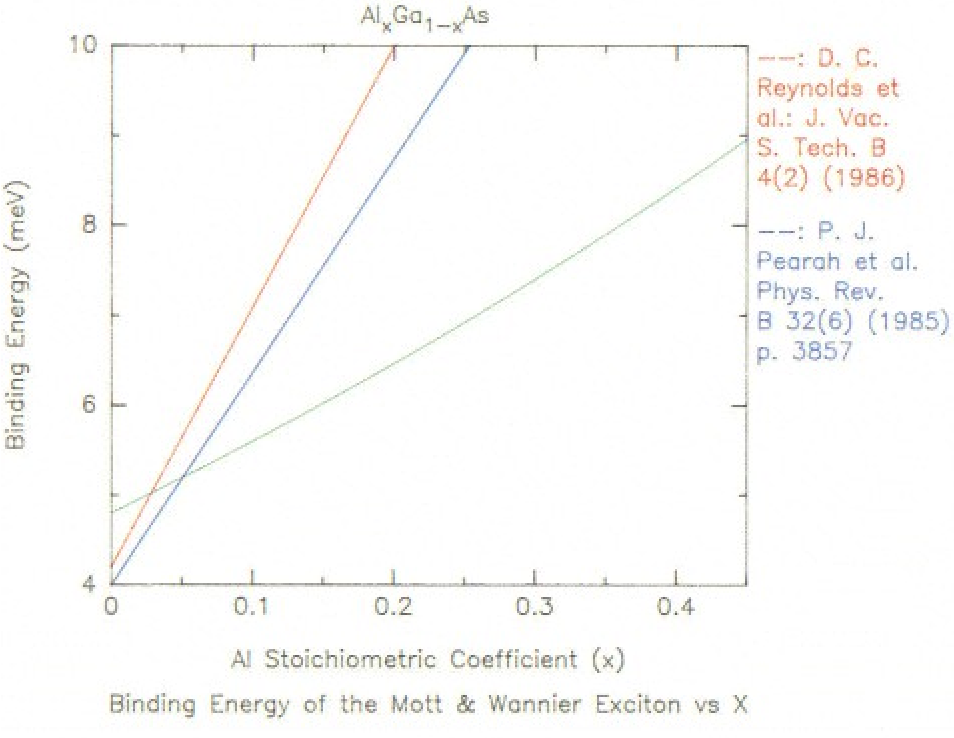

Fig. 6. BE of the M&V exciton as varying the x coefficient. The green line refers to the FE calculated one.

For what the dissociation energy (D_0) of an exciton bound to a neutral donor concerns, we shall refer to a paper wrote by Sharma (1967) [12]. For GaAs and AlAs we have $m^*_{\mathrm{e}}/m^*_{\mathrm{h}} = 0.14$ and 0.17 respectively and from the calculation made in it (see Fig. 1 of that paper) it comes out that $D_0/E_D \sim 0.13$ for both the type of semiconductors, where E_D is the ionization energy of the involved donor, therefore in our case the binding energy of the bound exciton should be the 13% of the value of the binding energy of the donor owner. In this range of the aluminum content the energy of the donor has been observed to be between 6 meV and roughly 30 meV [13] and so, in a first approximation, E_D should be between 0.8 meV and 3 meV.

In our experimental data, if we consider the BE and FE value added together, we have not a good agreement with our samples, especially in the case of the M373 where:

$$E_g(\mathrm{RT}) = 1.959 \text{ eV} \quad (\text{from PL})$$

and by using the same temperature dependence of the band gap of the GaAs:

$$E_g(13 \text{ K}) = 2.053 \text{ eV}$$

now the observed BE peak energy at this temperature is:

$$E_{BE}(13 \text{ K}) = (2.044 \pm 0.001) \text{ eV}$$

this means that the exciton binding energy should be only:

$$E_g(13 \text{ K}) - E_{BE}(13 \text{ K}) = 9 \text{ meV}$$

but this number is too low in comparison to what calculated for this high aluminum concentration.

Regarding to the binding energy of the exciton bound to an ionized donor, the work of Sharma (1967) [11] is quite clear:

for $Al_xGa_{1-x}As$ with $0 < x < 0.4$ there is $0.14 < (m^*_e/m^*_h) < 0.17$;

in Fig. 1 of that paper it comes out that in this range of σ

$$1.04 < W/E_D < 1.02$$

where W is the binding energy of the exciton bound to an ionized donor.

From this picture, in this particular case we can find out that the binding energy of the BE bound to an ionized donor is very close to the donor one and so we do not make a big error if we take directly the latter for the former.

In the sample M420 we have a reasonable agreement:

$$E_g(11 \text{ K}) \sim 1.7 \text{ eV}$$

$$E_{BE}(11 \text{ K}) = 1.6857 \text{ eV}$$

and from the difference between these two energy the binding energy of BE will be:

$$W \sim 14 \text{ meV}$$

On the contrary, for the other two samples we have no agreement:

for the M417 it comes out $W \sim 27$ meV, too high for its aluminum concentration;

for the M373, as it has been seen, W is too low.

These disagreements could be related to a inexact extrapolation to low temperature of the energy gap of the studied materials; in any case this picture with the question related to the big FWHM observed in the BE peak of the sample M373 necessitate more speculations.

3) EXCITON PEAK vs. $(\Delta a/a)_{\mathrm{relaxed}}$

With the aid of the lattice parameter measurements made by Stefano Lagomarsino (IESS - CNR Italy) by using the X-ray DCD, and the PL measurements of the excitons in our AlGaAs samples, it has been possible to draw a graph of the exciton energy peaks as a function of the relative difference in relaxed lattice constant $(\Delta a/a)_{\mathrm{relaxed}}$ between the AlGaAs and the GaAs substrate obtained at different aluminum concentrations.

In Fig. 7 this diagram is shown with the best linear fit of our three data, taking as a sure point the exciton peak energy of GaAs [14]; the error in the peak energy can be as far as ± 1 meV. In my knowledge it is the first time that such a study has been made so far.

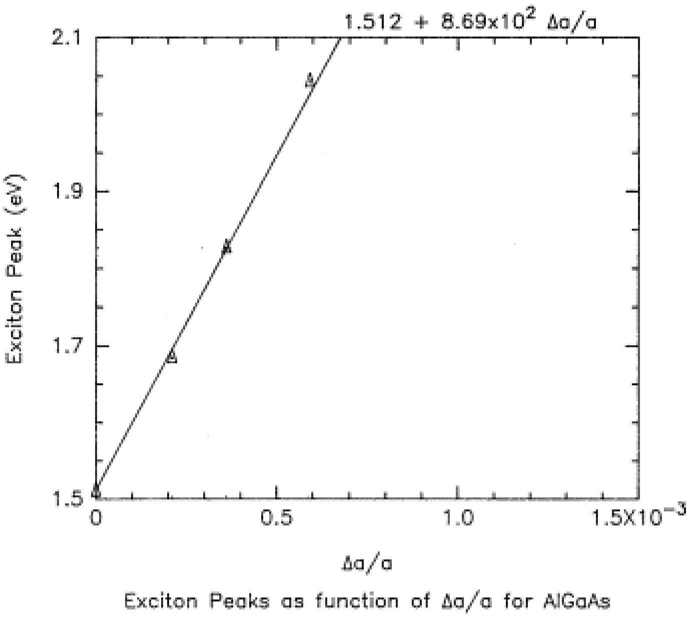

Fig. 7. Excitons peaks as varying the relaxed lattice constant.

If we make the assumption that the relation between the aluminum content and $\Delta a/a$ is linear in AlGaAs growth over GaAs substrates [15], that is:

$$x = 714.285(\Delta a/a)_{\text{relaxed}}$$

then it is possible to draw the pictures of Fig. 8 and 9, which take as a certain point the 1.424 eV for the GaAs E_g. From the best linear interpolation of the data (Fig. 8) the following equation valid for our samples comes out:

$$E_g(\text{RT}) = 1.424 + 1.273x, \text{ with } r^2 = 0.9996,$$

which is close to the one obtained by Casey (1978) [8]

$$E_g(\text{RT}) = 1.424 + 1.247x\ .$$

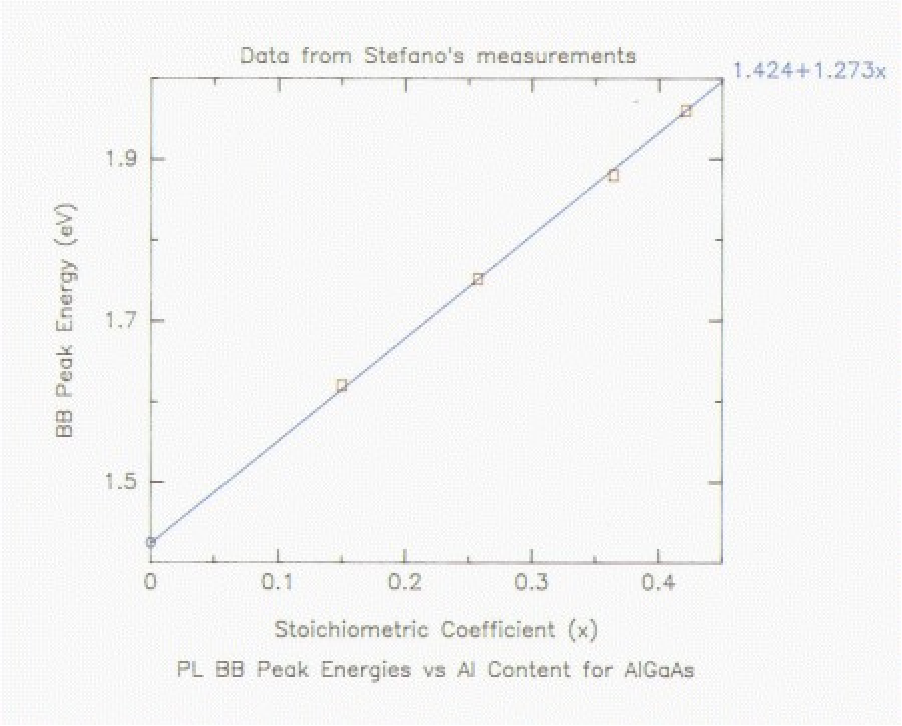

Fig. 8. PL band to band peak energy as a function of Al content with the linear interpolation.

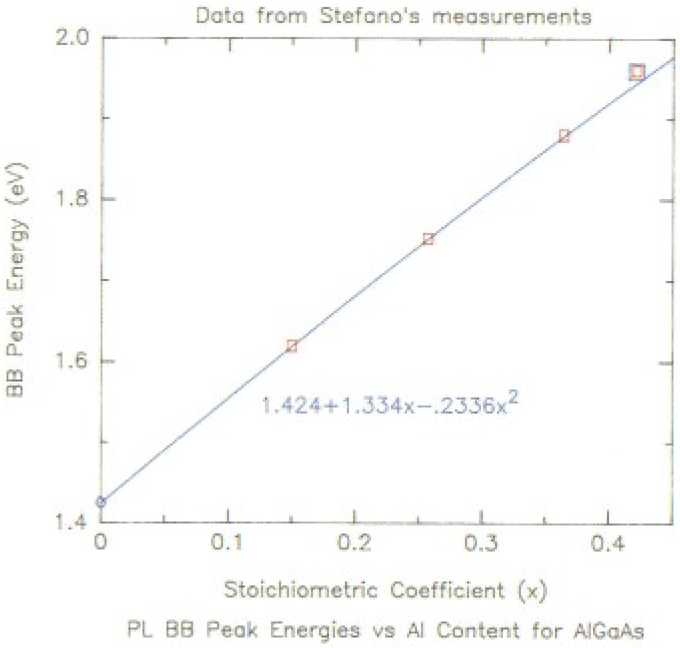

Fig. 9. **Fig. 8.** PL band to band peak energy as a function of Al content with the quadratic interpolation.

Instead, by using the best quadratic interpolation of the data, excluding the M373 one and again taking as a certain point 1.424 eV, we obtain what is shown in Fig. 9, that is:

$$1.424 + 1.334x - 0.2336x^2, \text{ with } r^2 = 0.9997,$$

which can be compared with the one of Miller (1985) [16]:

$$1.42 + 1.45x - 0.25x^2$$

Anyhow, from Fig. 10, it can be seen that our data are matched better by the Casey's equation than by the Miller's one. Indeed the PL band-to-band transition energy peak is not considered to be peaked right at the energy gap of the semiconductor, but a few meV below it [3].

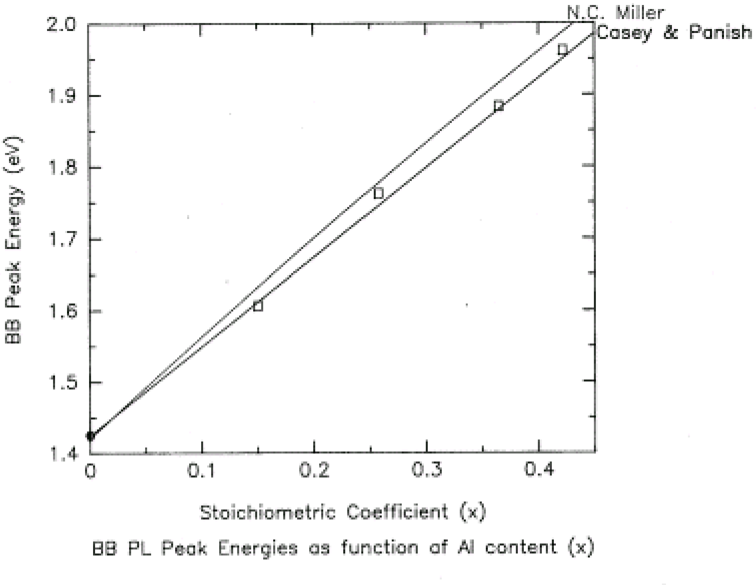

Fig. 10. Comparison of our data with the different matching equations.

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