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QUANTUM WIRE SPECTROSCOPY AND EPITAXIAL GROWTH VELOCITIES IN InGaAs-InP HETEROSTRUCTURES

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ABSTRACT

We study excitons bound to quantum wires of InGaAs embedded in an InP matrix, where the wires vary from 2.93Å to 11.72Å (one to four monolayers) thick and from 25Å to 250Å wide. We combine spectroscopic data from measurements of photoluminescence with variational calculations of the binding energies of excitons to the wires to deduce the wire widths and thicknesses. The widths are then related to the growth times to deduce lateral growth velocities in the vapor levitation epitaxial technique. Monolayer growth rates, at ~80Å/sec, are significantly faster than growth rates for the multilayer wires.

It seems natural, following the striking success of two-dimensional carriers and excitons in semiconductor heterostructures in generating not only new physics but new device opportunities, to push that effort one or two dimensions lower, and investigate the properties of what have come to be called quantum wires and quantum dots. Morais, Cox, and coworkers have previously reported spectroscopic evidence for well defined, extremely thin (down to monolayer thickness) quantum wells[1,2] of InGaAs, grown lattice-matched to InP by vapor levitation epitaxy (VLE)[3]. The spectroscopy and their understanding of the VLE process suggested strongly that the photoluminescence peaks obtained from these layers corresponded to thickness fluctuations in exact monolayer increments (2.93Å). Later, using extremely short growth cycles, they were able to grow uniform quantum wires of sufficiently narrow lateral width that the recombination luminescence of excitons bound to them was substantially modified[4]. At that time, they were unable to secure a quantitative relation between the exciton-quantum wire binding energy and the lateral width. Other investigators were nevertheless able to verify the dimensionality of similar structures using submillimeter spectroscopy and transmission electron microscopy (TEM)[5].

In this work, we present a simple new variational technique (within the effective mass approximation) for obtaining the binding energy of a particle to a finite potential well of rectangular dimensions. We then use the results of such calculations for both electrons and holes to estimate the widths of the VLE-grown wires, and thus deduce the lateral rates of growth for several different thicknesses of quantum wells. We expect that these conclusions will be useful in perfecting the understanding of the growth process

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We assume a two-dimensional rectangular quantum box with smooth sides of thickness L_x, width L_y, and depth V_o, as depicted in Figure 1. In the *z-direction, mercifully not depicted,* there is translational symmetry. Inside the box is the InGaAs quantum wire and outside is InP. Different effective masses inside and outside the wire are assumed for both electrons and holes. V_o is the band offset. Band non-parabolicity has not been taken into account, since the electron binding energies are small and their wave functions have only mild curvature, and the heavy hole band is parabolic in the three-band Kane theory. It is conceivable that the mixing of the heavy and light hole bands for finite wave vector in confined situations[6] should be invoked, but we have not done so. Parameters used in the calculations are: conduction (valence) band discontinuity 265 (366) meV, electron (hole) mass for InP .077 (.50), and electron (hole) mass for InGaAs .041 (.47). Effective masses are given as fractions of the free electron mass.

In the *Variational Method,* we write the wave function as a product of two one-dimensional square-well potential solutions, where the well widths accurately reflect the dimensions of the quantum wire but the potential depths V_x and V_y are variational parameters. This method gives a definite lower bound to the binding energy, and has the advantages of simplicity and speed, with only a two-dimensional minimization. It would be possible to improve the accuracy with more complicated wave functions.

The variational calculation was supplemented with three other approaches for comparison. We discuss here their relative merits and drawbacks. Using *Separation of Variables* one decouples the x and y motion, and solves two one-dimensional problems. This is strictly appropriate only in the limit $V_o \to \infty$. The resulting binding energy is a lower bound to the exact value. In fact for narrow quantum wires, the calculations can return a negative binding energy, i.e. no bound state at all, a result of the fact that the potential implied in this approach is $2V_o$ in the corner regions outside the quantum wire. In the *Adiabatic Approach* one solves first the one-dimensional problem in the narrow direction, and uses the energy level found there as the position of the potential floor for a second one-dimensional solution in the other direction. This approach yields neither an upper nor a lower bound and is difficult to assess, but it has been successfully used in estimating optical confinement[7], and recently for obtaining electron energy levels in a quantum wire laser structure[8]. The *Plane-wave Expansion* method was employed by Gershoni, et al[9] to calculate the confinement energies in etched InGaAs-InP quantum wires. Our numerical explorations of this method have borne out our initial guess - that it is unsuitable for cases like ours where the quantum wire is too small or too weak to contain a large fraction of the probability density.

In Figure 2, we compare the results of three different calculations (variational, adiabatic, and decoupled), showing separately the hole and electron binding energies to a quantum wire with L_x = 8.79Å, (three monolayers), as a function of L_y. For large L_y, all binding energies approach the 2D limit. The heavier holes reach this limit before the lighter electrons. We see the erroneous result predicted above for the decoupled calculation, that the binding energies disappear for small L_y. As expected, the adiabatic approach gives a larger binding energy, remaining well above that of the variational calculation, and especially so at small L_y, where it is bound to fail as $L_y \to L_x$. Note also that most of the binding is that of the hole, because it is heavier and its potential is deeper. For the remainder of the discussion, we will use exclusively the results of the variational calculation.

Figure 3 shows the total energy (electron + hole) binding the pair to the InGaAs quantum wire as a function of wire width, with thicknesses of n = 1, 2, 3, and 4 monolayers. We shall use these total binding energy calculations to determine L_y from the recombination energies measured.

The entity whose energy we measure in luminescence we take to be a Coulomb-bound state of the electron-hole pair, i.e. the ground state exciton. The energy of the exciton in such a structure will be different from the free pair energy calculated. For very weak single-particle binding (small L_y) both the electron and the hole are predominantly in the outer InP region, so we are inclined to take the Coulomb-binding energy as that of a 3D InP exciton, or 5 meV. We know this is wrong, since the wave functions are not those used in the construction of a 3D exciton. Indeed, for relatively large L_y the electron binding to the hole will be stronger than to the quantum wire. This, then, is a separate problem to be addressed elsewhere[10], and we proceed, noting that a few meV error will not affect our conclusions substantially.

Spectroscopic data from Cox, Morais, et al[2,4] giving photoluminescence peak energies for a variety of InGaAs quantum wires are shown in Figure 4. For shortened growth times, on the order of seconds, the recombination energies rise, indicating diminution of the energy binding the excitons to the wires. For zero growth time, all will approach the limit of 1.42 eV, representing the energy of an exciton in InP. We determine for each experimental point the exciton-wire binding energy by subtracting its energy from 1.42 eV. Finally, from a family of curves representing the binding energies calculated for L_x equal to 1, 2, 3, and 4 atomic monolayer thicknesses (2.93Å), we can read off the appropriate lateral dimension L_y.

Values of L_y so determined are plotted against the experimental growth times in Figure 5. We see immediately that except for the monolayer wires (n = 1), the growth velocities range between 12 and 20Å/sec, with the bilayer wire (n = 2) on the faster fringe. These velocities compare well with the average lateral growth velocity of 16Å/sec obtained from TEM measurements of an InGaAs layer 11 Å thick grown on a terraced InP substrate[4]. The single monolayer grows 5 - 6 times faster, with an estimated velocity of 80Å/sec. Two caveats are important here. One is that the monolayer spectroscopy is the most strongly affected by the accuracy of the assumption about exciton binding energy. The second is that coupling between wires is ignored although two of the monolayer wires represented in Figure 5, with widths determined by this analysis to be 51Å and 76Å, were grown on a substrate misoriented from (100) by 2°, for which the average terrace width is 84Å. Nevertheless, we emphasize our conclusion that monolayers grow faster than multilayers, since it has important implications for controlling the bunching of steps on misoriented and patterned substrates[11].

The quantum wells and quantum wires we have analyzed here may be compared with the extremely thin layers grown and studied by Sato and Horikoshi[12]. Using flow-rate modulation of metalorganic chemical vapor deposition epitaxy, and rather low growth temperatures, they were able to produce monomolecular and even submonomolecular (~25% coverage) planes of InAs in GaAs with intense and extremely sharp exciton luminescence. The sharpness of the luminescence indicates a high degree of uniformity, which suggests to us that the submonolayer planes contain many islands of InAs on a scale smaller than the exciton radius. In contrast, we believe that the VLE process takes place near equilibrium, so that growth occurs at terrace edges with the characteristic velocities that we have shown here.

Summarizing, we have made use of a variational calculation to estimate the dimensions of quantum wires grown by Vapor Levitation Epitaxy. Our analysis indicates that the first monolayer of InGaAs on InP grows much more rapidly than subsequent layers. This fact can be used to advantage in growing novel heterostructures on patterned or vicinal substrates.

January 15, 1990

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FIGURE CAPTIONS

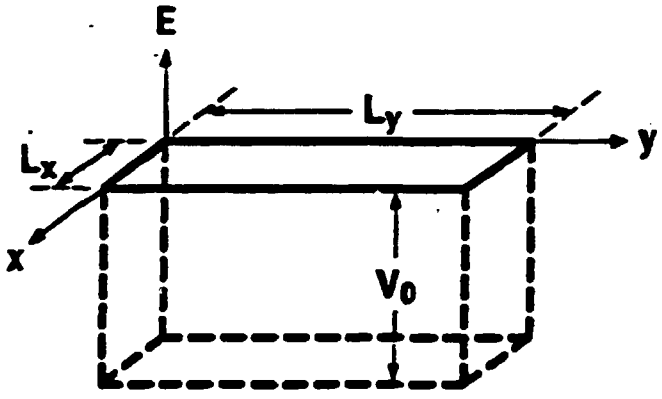
Figure 1. Two-dimensional rectangular quantum box for a particle whose effective mass is different inside and outside. The variational calculation gives a lower bound to the energy by which the particle is bound to the box.

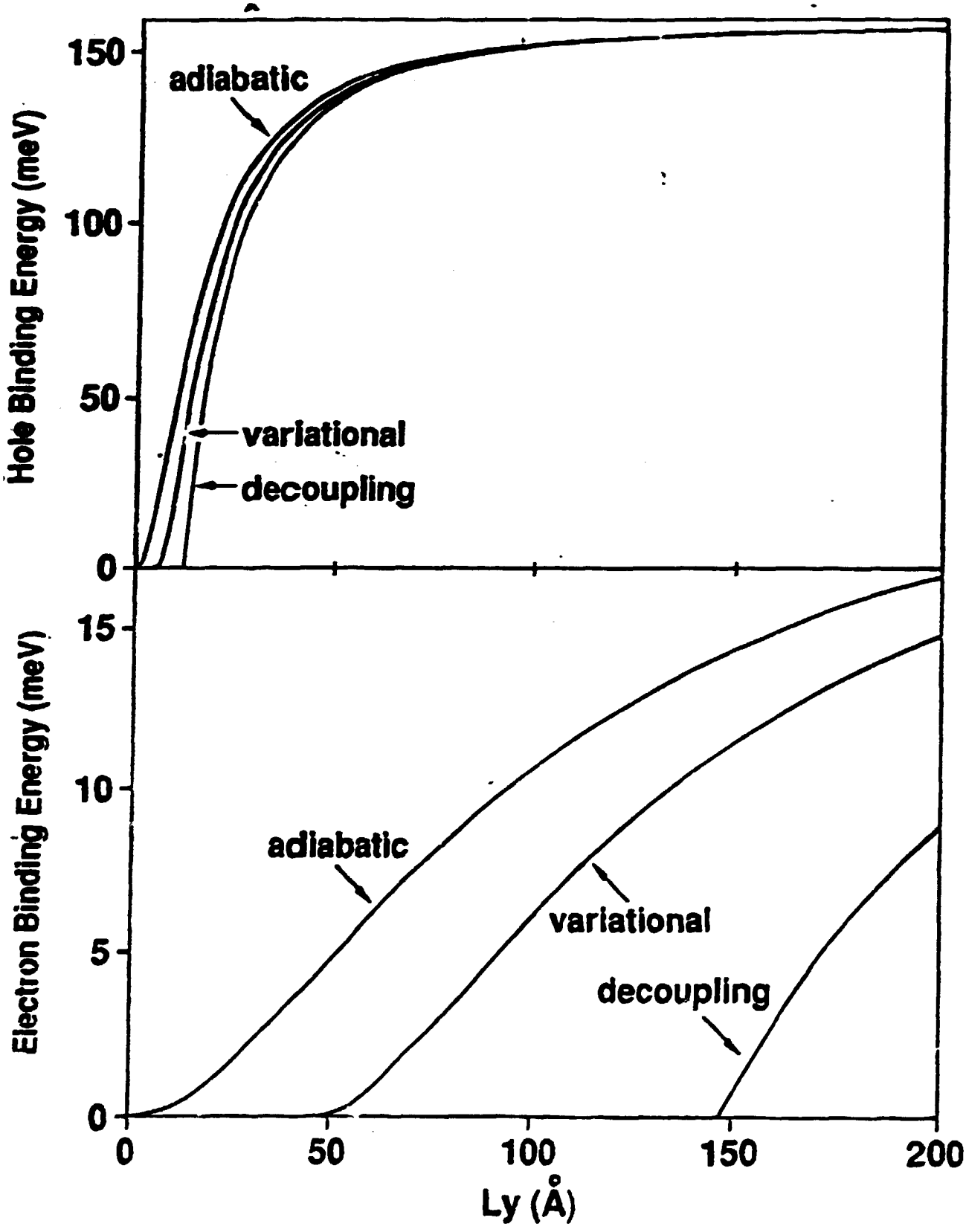
Figure 2. Binding energies of electrons and holes to a quantum wire 8.79Å (three monolayers) thick as a function of wire width, L_y. Three calculations are depicted: variational, adiabatic, and decoupled.

Figure 3. Binding energy of electron, hole, and the pair to a quantum wire 8.79Å (three monolayers) thick as a function of wire width, L_y, using the variational calculation.

Figure 4. Photoluminescence peak energies as a function of growth times. The two open circles represent data from a sample grown on a substrate oriented 2° from (100). The lines are guides to the eye: all curves should reach 1.42 eV as time → 0.

Figure 5. Wire width versus growth time for quantum wires whose thicknesses vary from one to four monolayers. Lateral growth velocities are the slopes of the lines.




Hole Binding Energy (meV)
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100
50
0
adiabatic
variational
decoupling
Electron Binding Energy (meV)
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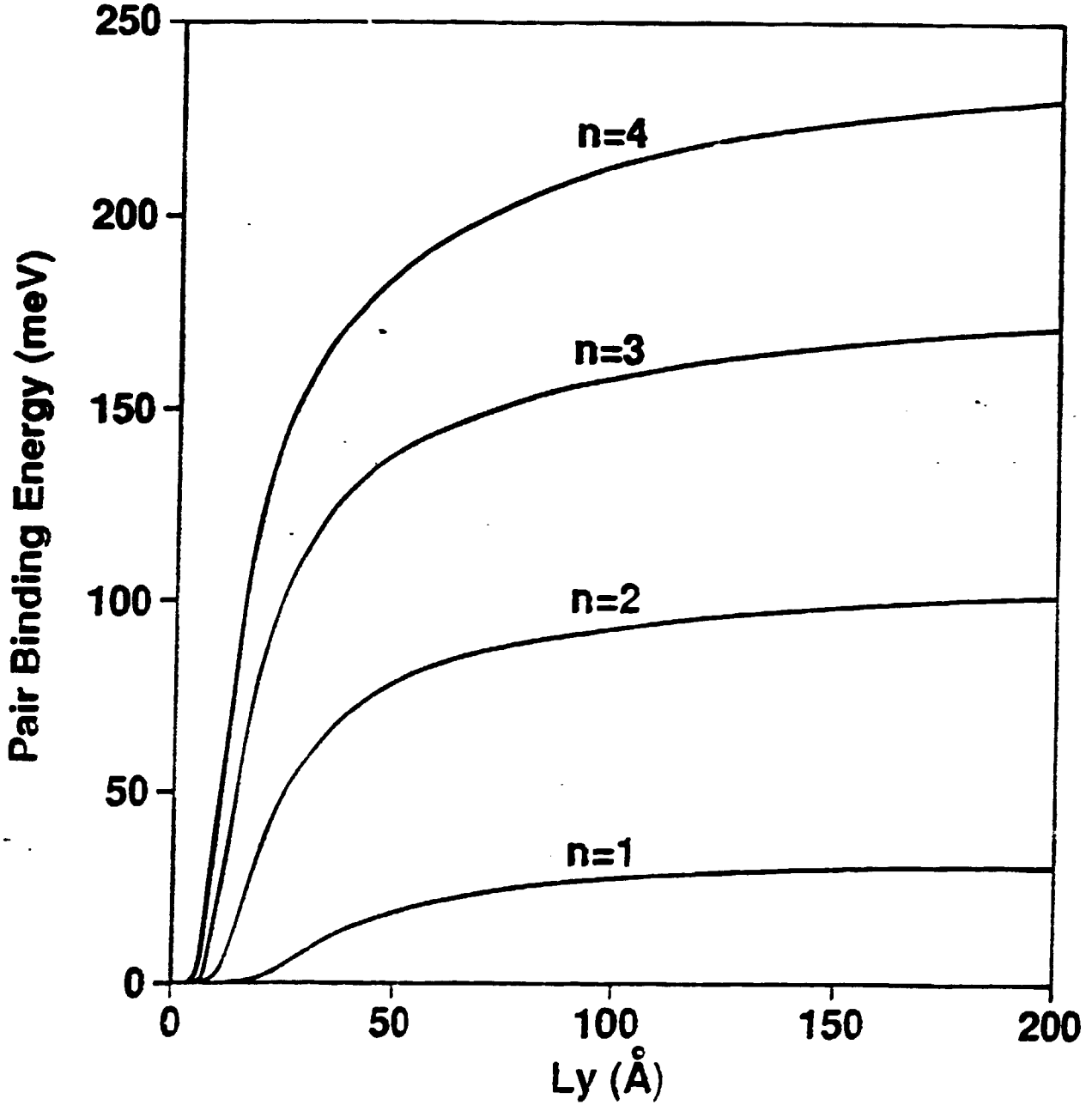
10
5
0
adiabatic
variational
decoupling
0
50
100
150
200
Ly (Å)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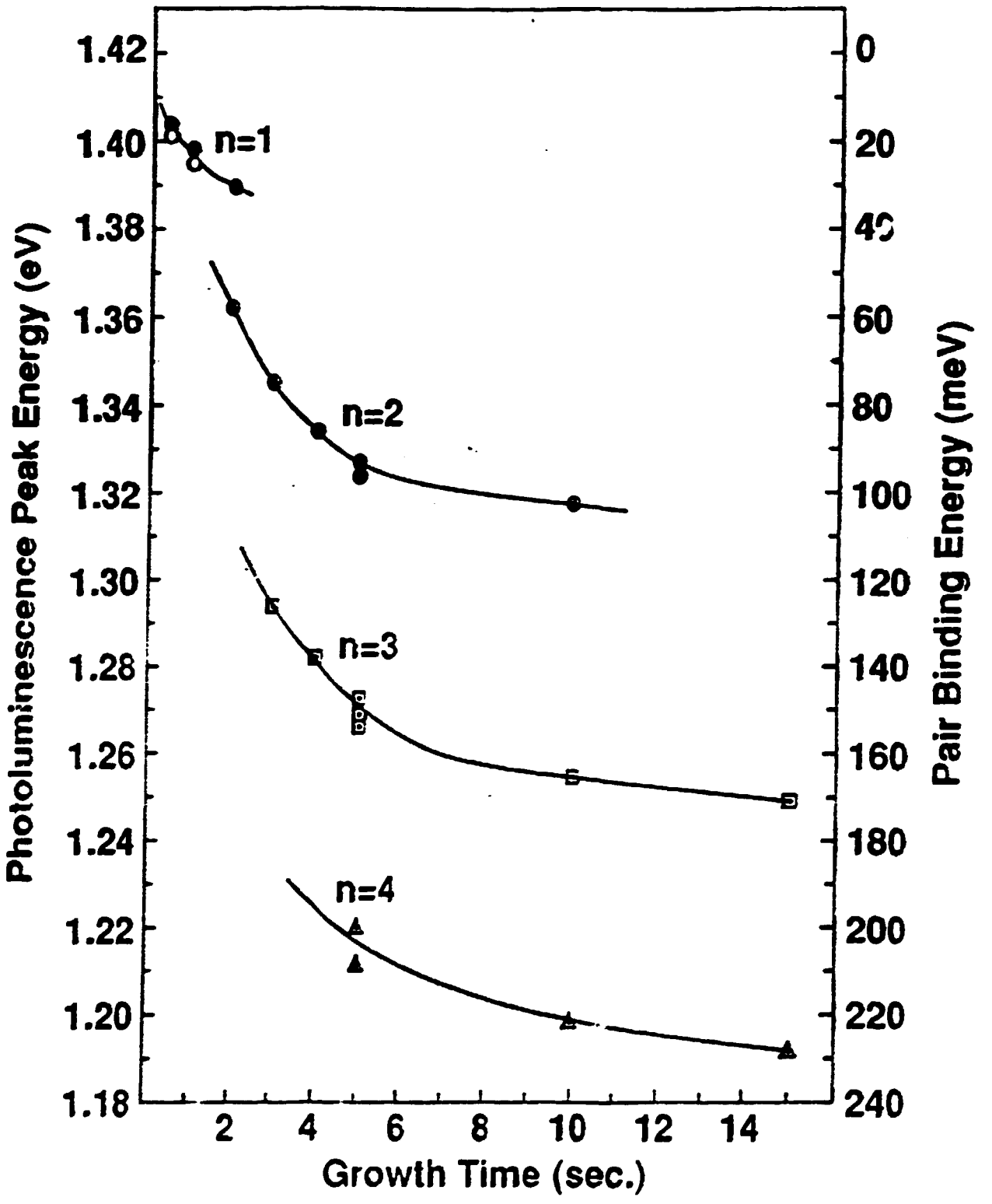
Ly (Å)



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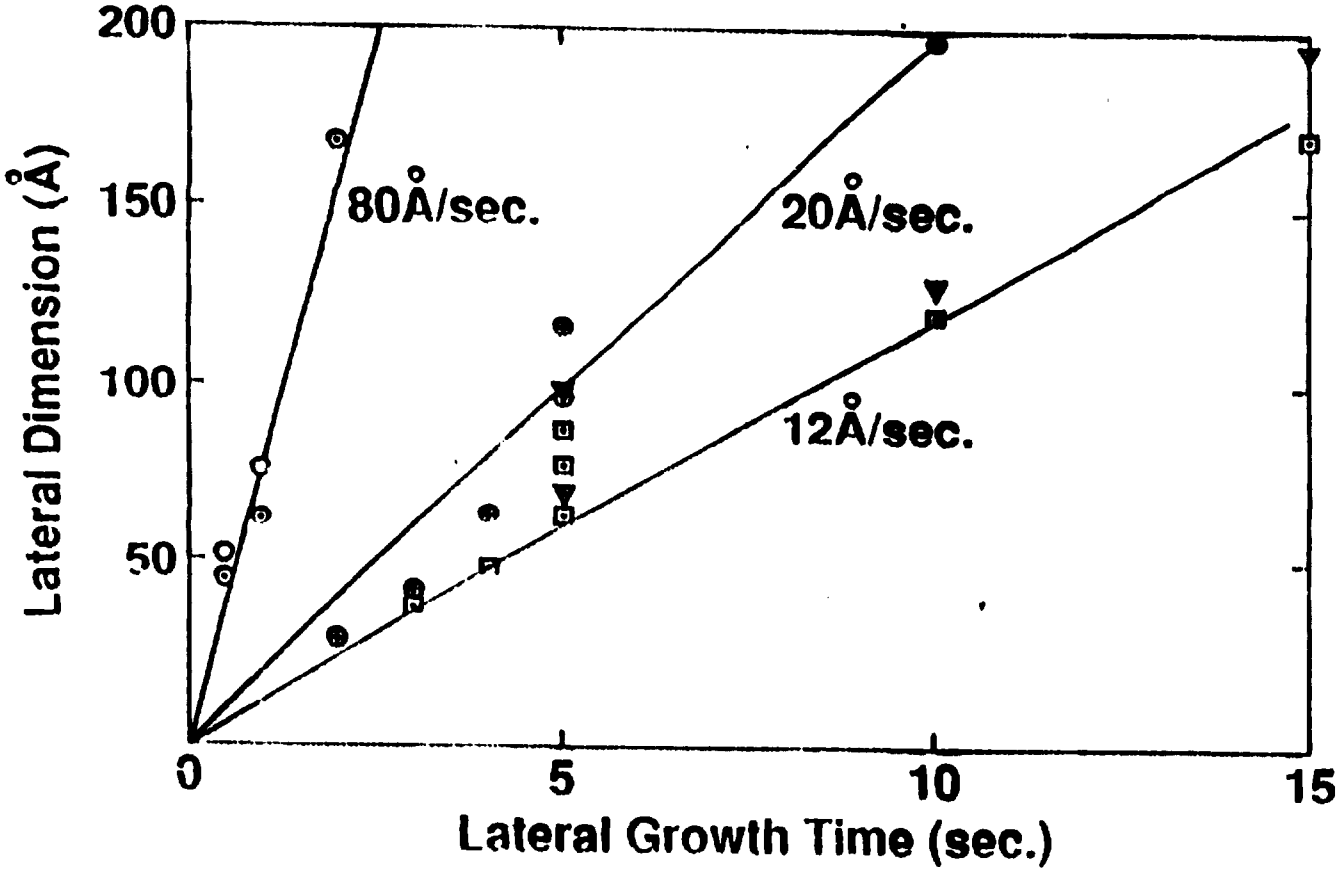
200
150
100
50
0
5
10
15
Lateral Dimension (Å)
Lateral Growth Time (sec.)
80Å/sec.
20Å/sec.
12Å/sec.