Changes in Luminescence Emission Induced by Proton Irradiation: InGaAs/GaAs Quantum Wells and Quantum Dots

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The photoluminescence (PL) emission from equivalent InGaAs/GaAs quantum well (QW) and quantum dot (QD) structures are compared after controlled irradiation with 1.5 MeV proton fluxes. Results presented here show a significant enhancement in radiation tolerance with three-dimensional quantum confinement. Some additional radiation induced changes in photo-carrier recombination from QDs, which include a slight increase in PL emission with low and intermediate proton doses, are also examined.

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Semiconductor Quantum Dot (QD) lasers with low threshold currents and high gain [1, 2], and QD Infrared Photodetectors [3] capable of incident photon absorption are already showing successful technological implementations of the unique optical properties of self-forming Semiconductor Quantum Dots (QDs). Future device applications include the use of coupled QDs as the basic structures in the fabrication of cellular automata in novel computing architectures [4] and frequency domain optical storage devices [5] based on self-assembled QDs.

Minimizing the impact of radiation induced degradation in optoelectronic devices is important for several applications. In space, protons pose a particularly severe threat to both planetary and Earth-orbiting spacecraft because they produce damage effects by several mechanisms. Because of their mass, protons can cause significant displacement damage in the semiconductor lattice, which is the primary cause of performance degradation and failure in several types of semiconductor devices. The effects of proton irradiation are also of interest in the use of ion beam modification or "defect engineering" in electronic materials. Proton implantation is often used for device isolation in compound semiconductors [6], and can also be used to induce interfacial compositional disordering in both quantum wells [7] and quantum dots [8], which in turn, results in blue shifted photoluminescence emission from both types of quantum structures [9].

Some of the fundamental properties of QDs suggest that optoelectronic devices incorporating QDs could tolerate greater radiation damage than other heterostructures. One of them is based on a simple geometrical argument: the total volume percentage of the active QD region is very small. Specifically, in self-forming InGaAs/GaAs QDs surface coverage range from 5% to 25%, depending on growth conditions [10]. Therefore, the chance of finding radiation-induced defects in the active region is reduced. Also, exciton localization in the quantum dots due to three-dimensional confinement (the InGaAs dots used here average 5 nm height and 25 nm diameter) will also reduce the probability of carrier non-radiative recombination at radiation induced defect centers. Here we compare the optical emission from InGaAs Quantum Well (QW) and QD structures after controlled irradiation with 1 MeV protons and show improved radiation hardness from the QDs.

Details of the growth conditions of InGaAs/GaAs QDs by metalorganic chemical vapor deposition have been described in previous work [10]. After deposition of GaAs buffer layers at 650°C, the temperature was lowered to 550°C and nanometer sized InGaAs islands were grown by depositing ~ 5 ML of $In_{0.6}Ga_{0.4}As$. QW samples were obtained by stopping the growth of InGaAs before the onset of the Stranski-Krastanow transformation, giving thin (1 nm) QWs. Ternary compositions between the samples were identical, and so was the capping layer thickness (100 nm for both QDs and QWs), therefore these results are not dependent on material or proton energy loss differences. Force microscopy and transmission electron microscopy [10-12] have been used extensively on these samples to give information on island sizes and surface densities in capped and uncapped InGaAs QDs. Proton irradiations were carried out using a Van De Graaff accelerator. Samples were irradiated at room temperature using 1.5 MeV protons at doses ranging from 7×10^{11} to $2 \times 10^{15}/cm^2$, with a dose rate of 6×10^{12} protons/sec. Dose uniformity was monitored using radiochromic film at low doses. Variable temperature photoluminescence (PL) measurements (from 4 K) were done using the 514 nm line of an Argon ion laser for excitation and a cooled Ge detector with lock-in techniques for signal detection.

Fig. 1 shows the effects of different proton fluences on the measured PL emission from both types of samples, InGaAs/GaAs QDs and QWs. The differences in the un-irradiated (as-grown) PL emission are apparent and have been discussed in previous work [9, 12-14]. Due to increased excitonic oscillator strength in the structures with three dimensional confinement [15], the integrated emission intensity is greater, even though only a fraction of the surface area is covered by QDs. As shown in Fig. 1 (a), the QD peak is also much broader. This inhomogeneous PL broadening originates from slight size non-uniformities and from the effects of varying lateral strain in disordered dense dot ensembles [11]. In QD laser applications, this problem is addressed by growing a multiple stack of dots, with a narrow separation between the dot to increase gain [16]. It can also be seen from Fig. 1 (a) that the emission energy from the QW is at a higher energy than from the QDs. This is because in order to obtain a strained QW of the same ternary composition as the QDs but without dislocations, very thin QWs (1 nm) are used. This approach has been succesful in a recent study that compared the effects of compositional disordering in QDs and QWs [9]. Fig. 1 (a) shows that despite the degradation in optical emission from both QD and QW structures with high proton doses, the emission wavelength was not affected.

Fig. 1 (b) shows the measured integrated PL intensities as a function of dose for 1 MeV proton irradiation in QWs and QD structures. InGaAs QDs are seen to be more radiation tolerant than QWs of the same composition. This increase in radiation hardness is significant, because QW based devices already represent a vast improvement in radiation hardness over bulk devices like optocouplers, which show significant degradation with proton irradiation [17]; and light emitting diodes (LEDs) based on QWs have shown an order of magnitude greater tolerance to proton induced damage when compared to LEDs based on pn junction geometries [18].

Fig. 2 shows the effects of proton irradiation in in QD structures with a low dot density, which show a strong PL peak from the wetting layer (WL). The WL is a very thin QW which forms prior to the dots in Stranski-Krastanow growth. If the average QD separation is greater than the photocarrier diffusion lenghts, recombination from WL states will ocurr for photocarriers generated in the WL. Fig. 2 shows that proton irradiation has different effects in the 2-D structures (WL peak at 1.3 eV) than in the 0-D structures (QD peak showing excited states emission, 1.1 eV for ground state).

Fig. 1 (and Fig. 2) show a slight increase in QD integrated PL (from ~ 10% to 70%) with low to intermediate proton doses (from 7×10^{11} to $7 \times 10^{12}/cm^2$). The fact that no such increase is observed in the QW structures leads us to conclude that this PL enhancement is an effect of three-dimensional confinement. Reduction of the phonon bottleneck by defect assisted phonon emission has been proposed [19] as a mechanism to explain the bright PL emission in QDs. Perhaps in dots with defect free interfaces, introduction of deep level defects as those originated from displacement damage might provide additional relaxation paths [20] for thermalization of carriers and therefore increase the luminescence emission. The mechanisms responsible for the small degradation observed in the optical emission from QD structures ($> 10^{13}/cm^2$) also remain to be fully investigated. From what is known about carrier generation, capture, transfer and recombination in InGaAs QDs [12-14] the degradation in minority carrier diffusion lengths expected in the barrier and wetting layer materials will contribute first to any observed degradation in QD PL emission, by limiting carrier capture into the dots. Reduction in diffusion lengths in the barrier material (GaAs) and in the InGaAs WL is the most probable cause for the initial degradation observed in QD PL with higher proton doses.

Fig. 3 shows some subtle but interesting differences induced by proton radiation damage in the temperature dependence of the QD luminescence signal (all normalized over the degraded signal at low temperature). The temperature dependence of the integrated PL signal from dense QD ensembles is closely related to their confining potential [21] just as in QWs [22], hovewer, this excludes effects from mid-gap deep levels or non-radiative recombination, which would have the effect of lowering the values for this activation energy. Fig. 3 (a) shows a slightly lower activation energy, with lower normalized PL at temperatures ~ 100 K. This latter effect can be explained in light of recent results, which show that for low QD densities, the temperature dependence of minority carrier mobility in the GaAs barrier (which peaks at 80 – 100 K) and InGaAs wetting layer can enhance PL intensity in quantum dots at intermediate temperatures [13]. Mobility degradation due to proton damage in the barrier and WL would then affect carrier capture and transfer into the dots. Fig. 3 (b) shows a more pronounced decrease in the inhomogenous PL broadening with temperature after radiation damage. It is also observed, that there is no subsequent rise in FWHM above 150 K, but this could be due to the signal being too small for reliable measurements. This decrease in the FWHM of the PL band has been attributed to carrier thermal emission from the smaller dots in the ensemble [13]. With radiation damage, the onset of thermionic emission will also be acompanied by defect assisted non-radiative recombination, making this effect even stronger, which might explain the stronger decrease in inhomogeneous PL broadening seen in Fig. 3 (b).

In summary, the results presented here show that QDs structures are inherently more radiation tolerant due to the effects of three dimensional quantum confinement. An increase in radiation hardness of as much as two orders of magnitude has been obtained by comparisons with quantum wells of the same composition and placed at the same depth in the structure. Additionally, we show that a slight increase in PL emission from InGaAs/GaAs QDs can be observed with low to moderate 1 MeV proton doses, and that radiation also induces subtle changes in the temperature dependence of the luminescence emission from InGaAs quantum dots.

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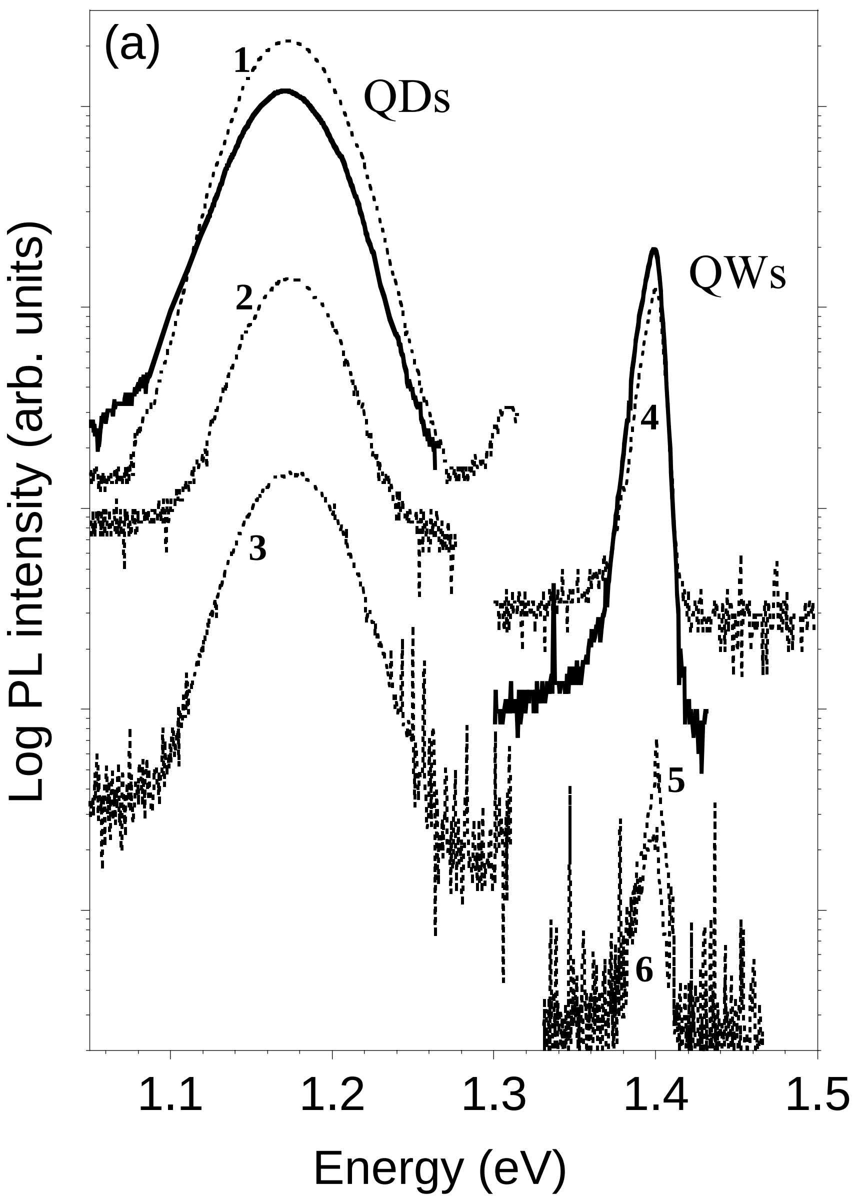
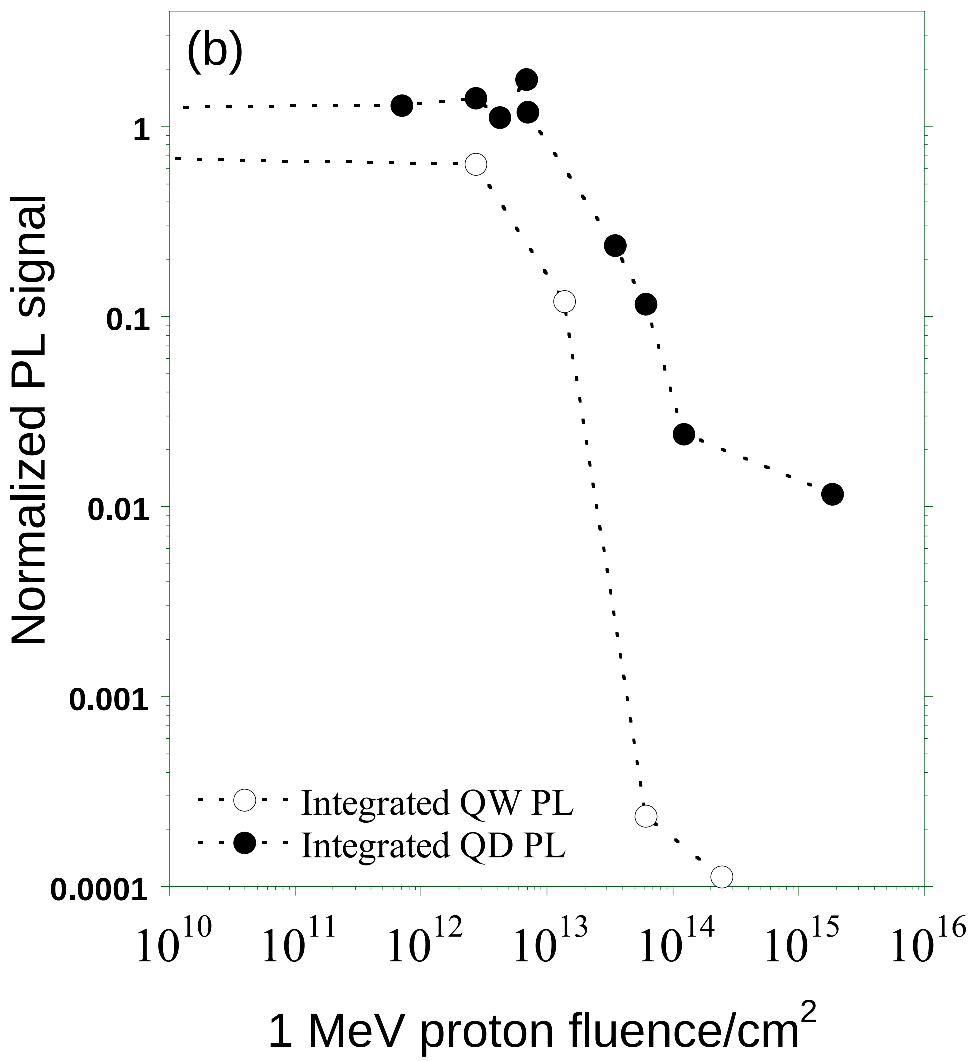
Figure Captions:

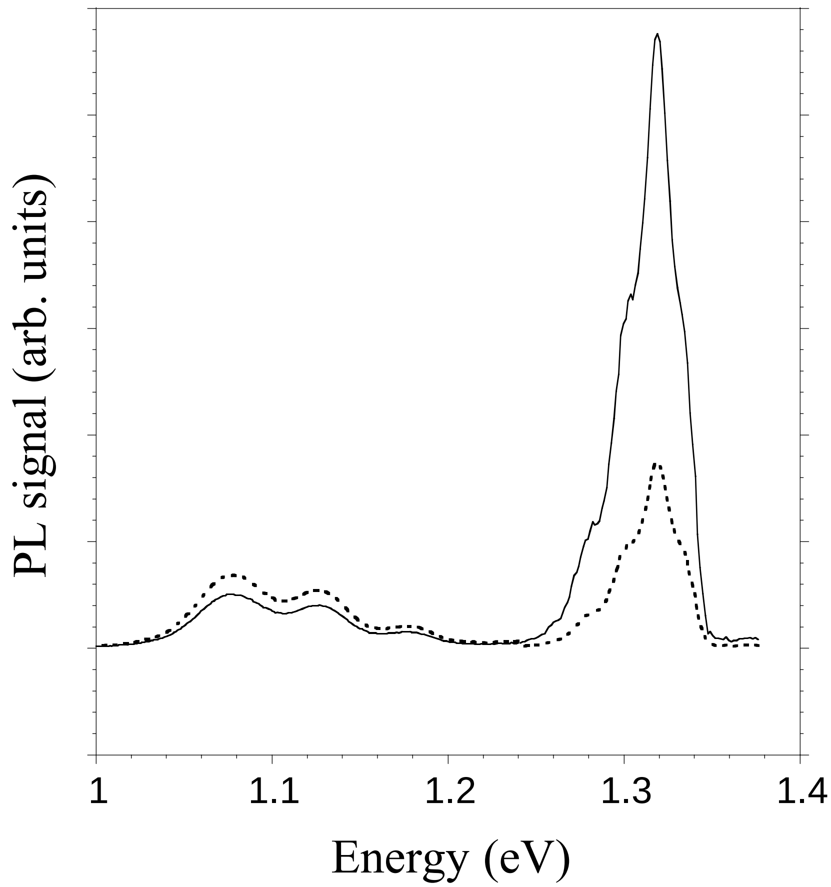
Fig. 1. (a) Comparison of PL spectra from InGaAs/GaAs quantum wells and from quantum dots in high surface densities (2.4 x 10^{10} dots per cm^2) after selected proton fluxes. The solid lines show spectra before irradiation. The dotted lines show spectra after 1.5 MeV proton irradiation in doses (per cm^2) of 1) 7 x 10^{12}, 2) 6 x 10^{13}, 3) 2 x 10^{15}, 4) 3 x 10^{12}, 5) 6 x 10^{13}, and 6) 2 x 10^{14}. (b) Integrated PL emission normalized to the as-grown samples for QW and QDs as a function of proton dose.

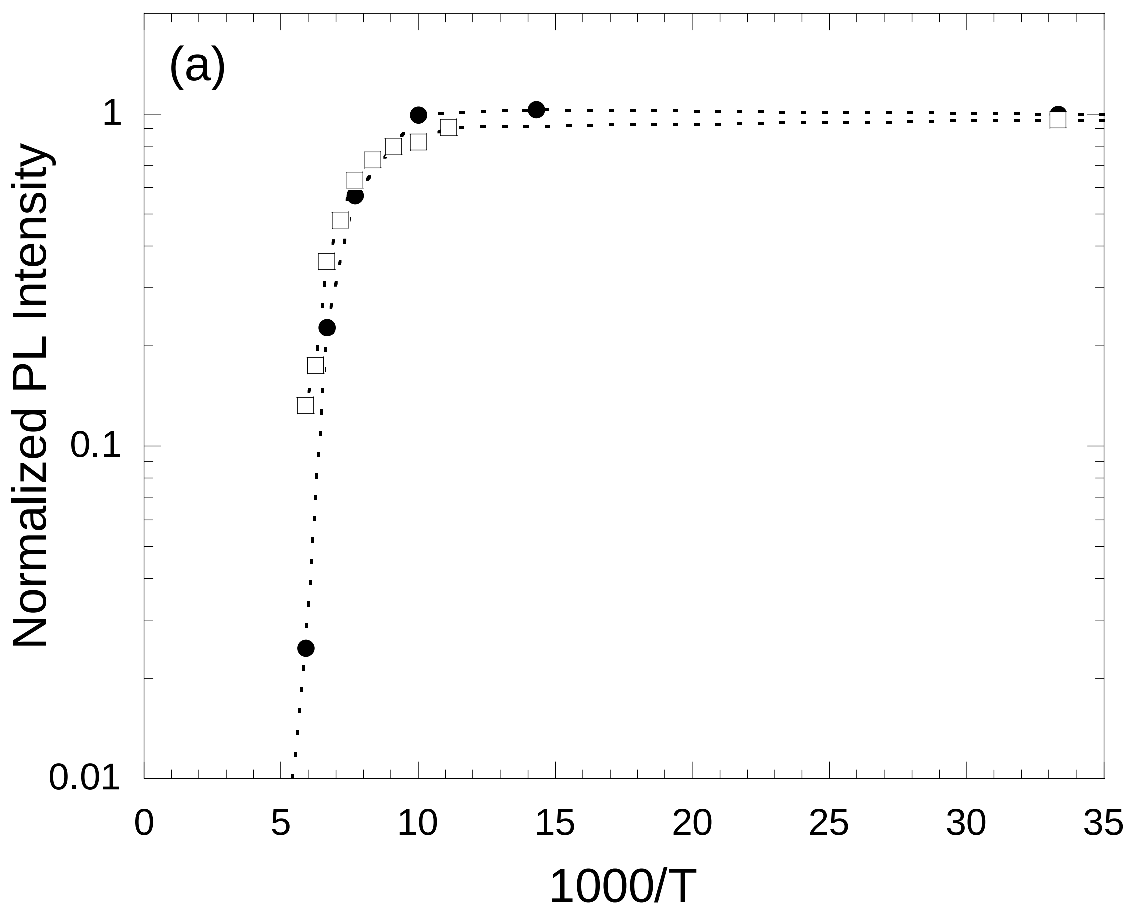
Fig. 2. Comparison of initial (solid line) and post irradiation (dotted line) PL spectra at a proton dose 2.7 x 10^{12}/ cm^2 of low density InGaAs/GaAs QDs (3.5 x 10^8 dots per cm^2). The spectra were obtained at constant excitation and show simultaneous emission form QD and wetting layer states.

Fig. 3. Radiation induced changes (with 1.5 MeV protons at a dose of 3.5 x 10^{13}/ cm^2) in the QD PL temperature dependence. (a) Total integrated PL emission from QD structures, filled circles show signal before proton irradiation, hollow squares indicate signal after irradiation. (b) Temperature dependence of the inhomogeneous broadening or full width at half maxima (FWHM) of the PL emission from QDs before (filled circles) and after irradiation (hollow squares).

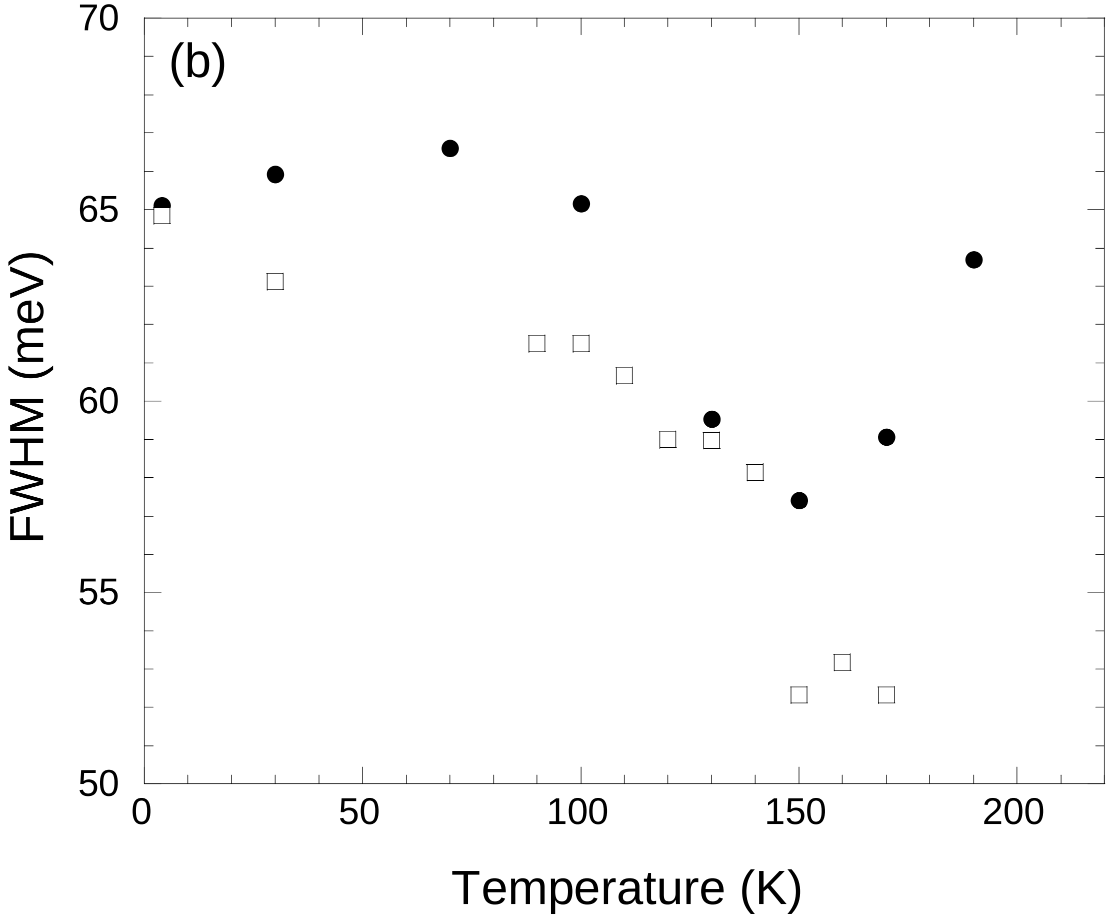

(a)
1
QDs
2
3
QWs
4
5
6
Log PL intensity (arb. units)
1.1
1.2
1.3
1.4
1.5
Energy (eV)


(b)
Normalized PL signal
1
0.1
0.01
0.001
0.0001
10^{10} 10^{11} 10^{12} 10^{13} 10^{14} 10^{15} 10^{16}
1 MeV proton fluence/cm^2
Integrated QW PL
Integrated QD PL


PL signal (arb. units)
1
1.1
1.2
1.3
1.4
Energy (eV)



(a)
Normalized PL Intensity
1
0.1
0.01
0
5
10
15
20
25
30
35
1000/T


(b)
FWHM (meV)
Temperature (K)
70
65
60
55
50
0
50
100
150
200