**Abstract.** In this paper we report on the growth and characterisation of CdSe quantum dots grown on a MgS-rich ZnMgSSe barrier and characterized by 77K PL and 4K µ-PL. The dot density was estimated to be approximately $10^{10}$ cm$^{-2}$ however some emission lines were sufficiently well resolved to investigate their behaviour individually; their emission was observed to change both in intensity and energy, showing a high frequency random jittering. Correlation of the dot intensity over much longer time periods (~200s) was also observed. These results will be compared with previous results from CdSe dots grown on ZnSe barriers.

1. Introduction

Wide bandgap II-VI semiconductor devices are typically grown on commercially available III-V substrates, which have bandgaps far smaller than the epitaxial II-VI layers. This is inconvenient for many experiments and applications; including any where absorption by the substrate prohibits the transmission of light through the epitaxial layers.

For ZnSe based structures grown on GaAs, we have recently developed an epitaxial lift-off process utilizing MgS as a sacrificial layer [1] which allows the epitaxial layers to be removed easily and without the use of any mechanical or chemical processes that might damage them. The lift-off process also allows for the use of a range of new functional substrates; this is particularly useful for experiments that require a micro-cavity [2].

MgS is also an excellent barrier material with a bandgap of ~5eV [3, 4], but cannot be used as such in any structures where lift off is also required. Consequently, we have developed a wide bandgap quaternary alloy Zn$_{0.2}$Mg$_{0.8}$S$_{0.64}$Se$_{0.36}$ which provides both the required confinement and resistance to the etching solution [5]. Our aim is to use this material as a barrier for quantum dot containing layers and as a first step toward this we report in this paper what we believe to be the first µ-PL study of CdSe dots grown on a MgS-rich ZnMgSSe quaternary alloy.

2. Growth

All of the samples examined in this paper were grown in a VG V80H MBE system using 6N elemental sources of Zn, Cd, Mg and Se, as well as a 6N ZnS source as the only source of sulphur. All layers, except the QDs, were grown by conventional MBE [2] at 300°C. The QDs were grown by atomic layer epitaxy (ALE) using alternate deposition cycles of Cd and Se at 300°C, and were then annealed for 4 minutes at 350°C to thermally activate the dot formation process [7]. The structures were monitored with RHEED throughout.

A series of samples were grown with the same basic structure – GaAs/34nm ZnSe (buffer)/10-15nm ZnMgSSe (barrier) /CdSe QD layer/10-15nm ZnMgSSe (barrier). All of the samples received no additional ZnSe capping layer, as the ZnMgSSe alloy has been found to resist oxidation sufficiently to protect the samples [4].

Two samples were selected for µ-PL study with 9 and 11 ALE cycles giving 4.5 and 5.5 ML of CdSe respectively. During the growth of both samples RHEED showed that they did not relax and are fully pseudomorphic.
3. Ensemble PL Characterisation

Ensemble PL measurements were made at 77K. These show (see Fig.1) a broad peak centred around ~2.5eV (496nm), from the CdSe QDs similar to those grown on ZnSe or MgS barriers [6, 9] and a smaller peak at ~2.79eV (444nm) representing emission from the ZnSe buffer layer.

![Fig. 1 – Ensemble 77K PL measurements of CdSe/ZnMgSSe QD samples](image)

The intensity of the PL emission from these samples was quite weak, ~500 times less intense than samples grown on ZnSe, as the ZnMgSSe barrier has a bandgap far larger than the incident photon energy and hence only the CdSe QD layer absorbs incident photons. This results in a small absorption cross section and relatively low emission intensity.

4. µ-PL Characterisation

The µ-PL measurements were made at 4K using a diffraction limited confocal microscope system utilizing either a 0.85 NA microscope objective or a combination aspheric and solid-immersion lens, with an effective NA of 1.3. The samples were excited using a 60mW 405nm laser diode mounted to a TEC cooler and driven at roughly 90% of its maximum drive current, so as to ensure it produce a highly stable output. The beam was then collimated and fibre coupled to the microscope head; mounted directly about the liquid helium cryostat in which the samples were placed [8].The excitation density for all our measurements was kept constant at ~ 220 kW cm$^{-2}$. The emitted light was dispersed using either 300 or 1800 line gratings onto a LN2 cooled CCD giving energy resolutions of 0.95meV and 90µeV respectively.

Within the microscopes ~0.2 µm diameter resolution, ~15 dots were visible giving an estimated density of ~10$^{10}$ cm$^{-2}$ similar to the densities seen with CdSe dots grown on both ZnSe and MgS [6, 9].

Figure 2 shows a µ-PL spectrum for both samples which was obtained by averaging the emission of the samples over 30s, due to the low emission intensity. Both samples show a series of closely spaced, relatively narrow features at 4K centered around 2.5 eV representing emission from individual dots. An additional smaller feature is seen at ~2.8eV again represents the emission from the ZnSe buffer layer, as can be seen in Fig. 2A. By translating the sample using sub-nm positioning stages, it is possible to find spatially isolated individual dots that are sufficiently separated from the main ensemble in emission energy to investigate their behavior. An example of this can be seen in fig. 2B. The sharpest features observed with these samples have FWHM in the region of 3±0.95 meV.
Figure 3 shows the time evolution of the spectra observed from the dot shown in fig. 2B. Over a period of time both the intensity and peak emission energy of the dot fluctuate. It should be noted that although it appears in the figure that the emission intensity periodically falls to zero (for example at times 500-550s and around 750s) this is not actually the case; in reality the intensity has just dropped to a level where it is not well resolved from the background in this colour map. It is also apparent that the large FWHM of the lines obtained in the integrated traces arises from the fluctuations in the peak position.

The graph also shows a second feature whose emission energy is roughly centered on 2.475 eV. The fluctuations in intensity and energy of this feature are highly correlated to that of the lower energy line, indicating that they arise from the same dot, as has previously been observed in ZnSe/CdSe QD emission [10].

Fluctuations in both the energy and intensity of QD emission have been observed for CdSe dots grown on ZnSe buffer layers [11] and it is believed that these arise from the Quantum Confined Stark Effect produced by fluctuating charge close to the dots; either located at the sample surface [10] or at dislocations originating at stacking faults [11]. We observe a typical fluctuation (spectral diffusion) in the dot energy of 2-3meV. Assuming our dots are roughly similar in size and polarizability to CdSe
dots on ZnSe [12], this could arise from single charges located approximately 10nm from the dot, which is compatible with the charge being located at the surface of the sample.

QCSD produced by a fluctuating electric field will cause the energy of the emission line to be reduced together with the emission intensity. Consequently, there should be a strong correlation between these two effects produced by the fluctuating electric field, as has been previously observed for ZnSe/CdSe dots [11-13]. However, the weaker emission from the ZnMgSSe/CdSe samples means that although we are able to identify both energy and intensity fluctuations from all dots we have examined so far, we have thus far not been able to find any statistical correlation between the two quantities.

If the quantities are indeed correlated, then our inability to resolve them signifies that the fluctuations are occurring on timescales shorter than our smallest integration time, < 25ms.

For the brightest isolated dots observed, as per fig. 3, the emission intensity shows a characteristic long period oscillation, with a strong correlation in the intensity occurring on a timescale of 200s. This effect has also been observed on ZnSe/CdSe dots by other groups where correlation times of 160s [11] and 220s [13] have been obtained.

5. Conclusion
We have investigated the behavior of CdSe QDs grown on a MgS-rich ZnMgSSe alloy barrier and found the alloy to provide good carrier confinement, however many of the individual lines observed showed significant spectral diffusion, giving peak FWHM of ~3meV. The energy and intensity of all the QDs within the field of view was also observed to fluctuate on a time scale of <25ms which is compatible with QCSD arising from isolated charges located close to individual dots.

A characteristic timescale of ~200s has been determined by statistical analysis for the variation in emission intensity and energy for some of the dots examined.

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