Vol. 103 (2003) ACTA PHYSICA POLONICA A No. 6

Proceedings of the XXXII International School of Semiconducting Compounds, Jaszowiec 2003

Topographical, Magnetic and Optical Studies of (II,Mn)VI Quantum Structures Grown on (Ga,Mn)As

R. Butkutė[a], M. Aleszkiewicz[a], E. Janik[a], G. Cywiński[a], P. Wojnar[a], L. Däweritz[b], J.-L. Primus[c], J. De Boeck[c] and J. Kossut[a]

[a]Institute of Physics, Polish Academy of Sciences
al. Lotników 32/46, 02-668 Warsaw, Poland
[b]Paul Drude Institut für Festkörperelektronik, Berlin, Germany
[c]IMEC, Interuniversity Microelectronics Center (IMEC), Leuven, Belgium

We report on an overgrowth of quantum structures consisting of diluted magnetic semiconductor CdMnTe quantum wells with non-magnetic barriers made of CdMgTe or ZnTe on ferromagnetic MnAs and GaMnAs films by molecular beam epitaxy. Atomic force microscopy images of the quantum structures grown on MnAs demonstrated the existence of two types of regions on the surface: protruded islands with micrometric sizes, surrounded by areas of small-scale roughness. Magnetic force microscopy study of these samples revealed a magnetic domain structure only on the above mentioned islands. The (II,Mn)VI quantum wells grown on GaMnAs films exhibited relatively smooth surface, but no magnetic force microscopy signal was measurable either before or after magnetizing the sample. In the luminescence spectra of all our quantum structures the emission attributed to CdMnTe quantum wells was observed. The influence of magnetization on the luminescence line position was investigated.

PACS numbers: 81.15.Hi, 07.79.Pk, 78.55.–m

1. Introduction

Diluted magnetic semiconductors (DMS) are studied intensively during the last few years, with emphasis on III–V compounds, such as (Ga,Mn)As and (Al,Mn)As, which exhibit carrier-induced ferromagnetism at relatively high temperatures, and thus may be suitable for applications in new devices with magnetic and spin-dependent functions. However, it is difficult to introduce large amounts of Mn ions substituting for Ga or Al. On the other hand, in the case of II–VI DMS, Mn can replace up to 100% of the group II elements. But the ferromagnetic state for classical (II,Mn)VI semiconductors is achieved at extremely low temperatures [1]. There are a few publications describing growth and investigation of non-lattice-matched hybrid II–VI/III–V structures or superlattices, which consist of magnetic ZnMnSe or CdMnTe and non-magnetic GaAs QW [2]. Most of them are devoted to the study of electrical injection of spin-polarized carriers across the II–VI/III–V interface. Recently, the influence of ferromagnetic films on DMS was discussed in terms of an influence of fringing fields on various spin-related properties of such structures: it was demonstrated in hybrid structures composed of a ferromagnetic metallic layer (such as Fe, Ni/Fe, Co) and a semiconductor that such an influence can lead to an additional localization of excitons in II–VI DMS QWs [3].

In this work we present the results of an overgrowth of CdMnTe QW with CdMgTe or ZnTe barriers on ferromagnetic layers of MnAs and GaMnAs (the latter containing intentionally ferromagnetic MnAs precipitates). We investigate here the topography, magnetic force microscopy (MFM) images, and photoluminescence of such structures.

2. Experimental details

The overgrowth of (II,Mn)VI quantum structures was performed in a molecular beam epitaxy (MBE) system on two types of (III,Mn)V films, referred to here as substrates. MnAs substrates consisted of high temperature (HT) GaAs(001) and low temperature (LT) 120 nm thick ferromagnetic MnAs(–1100) layers grown on semi-insulating GaAs(001) [4]. The second type of substrates, the so-called GaMnAs substrates, consisted of HT and LT GaAs buffers covered by 50–62 nm GaMnAs layers with 1.9–14.0% of Mn; subsequent annealing of this layer at a high temperature (about 600–650°C) leads to the formation of ferromagnetic MnAs clusters with a typical size of 3–20 nm [5]. A thin As amorphous layer to protect the surface from aging covered some of the GaMnAs substrates. After the growth of both types of the substrates they were exposed to air and transferred to another MBE apparatus dedicated to II–VI materials. To remove the As layer, or in order to deoxidize the surface of the substrates, they were heated up to 650°C in the deposition chamber before the overgrowth of (II,Mn)VI materials was commenced.

The overgrown structures were composed of a quantum well of CdMnTe (1–4% of Mn), sandwiched between CdMgTe (10–15% of Mg) or ZnTe barriers. The growth was performed at substrate temperatures of 280°C (for CdMgTe/CdMnTe/CdMgTe structures) and 325°C (for ZnTe/CdMnTe/ZnTe) with 0.2 and 0.5 μm/h growth rate. By choosing the width of the first barrier to be only 250 Å, we intended to make the magnitude of the fringe fields (if any) as large as possible in the QW region. On the other hand, by growing a wide (5000 Å) barrier we could improve structural quality of the overgrown material through reducing the number of dislocations. The quantum well and the cap were 25–150 Å and 750 Å, respectively. The overgrowth of CdMgTe barriers was initiated by depositing a very thin (5 monolayers) ZnTe buffer in order to reduce effects related to the large lattice mismatch between (Ga,Mn)As and CdMgTe ($\approx$ 10%); or with ZnTe ($\approx$ 7%). Reflection high-energy electron diffraction (RHEED) was used for *in situ* growth control. During the overgrowth on ferromagnetic MnAs substrates the reflections in the RHEED pattern had a streaky character, with streaks being somewhat shorter and broader at their end points compared to normal II–VI MBE growth in our system. On the other hand, very narrow streaks were observed in the RHEED patterns during the growth on GaMnAs substrates.

After the growth the samples were magnetized in a magnetic field of 5 T for 30 minutes. The field was kept parallel to the growth direction. *Ex situ* topography and magnetic domain patterns were studied at room temperature by means of atomic force microscopy (AFM) and magnetic force microscopy (Digital Instruments MultiMode SPM). Further in the text we use two parameters describing the surface roughness: (i) standard height deviation RMS and (ii) average difference in height between the five highest peaks and five lowest valleys, termed 10pt mean.

Measurements of photo- (PL) and microphotoluminescence (μ-PL) were performed in the temperature range from 4 to 40 K. For spatially resolved μ-PL measurements, an argon laser beam (λ = 477 nm) was focused on the sample as a 1.5 μm diameter spot by an optical microscope. The emitted line was collected by the same microscope and dispersed in a 1 m double monochromator. A sample was mounted on a specially designed cold-finger holder in a continuous-flow helium cryostat.

3. Results and discussion

3.1. AFM and MFM studies

3.1.1. (II,Mn)VI QW on MnAs substrates

The topographical studies of MnAs substrates before the overgrowth revealed very well ordered parallel protrusions about 400 nm wide and 3 nm high, with some sites where two neighbouring protrusions joined each other (Fig. 1a). MFM image taken prior to the magnetization showed strong out-of-plane component of the sample magnetization, which is directly related to the topography (Fig. 1b). When the sample was magnetized, the dark-white contrast of the MFM signal shifted from the steepest part of the topographical slope towards the top of each protrusion (see Fig. 1c, d), showing that the external magnetic field reoriented the domains.

AFM images of the (II,Mn)VI (with the narrow first barrier) grown on MnAs showed two kinds of regions — about one half of the sample area shows homogeneously distributed, relatively small grains of nanometric size, while the other

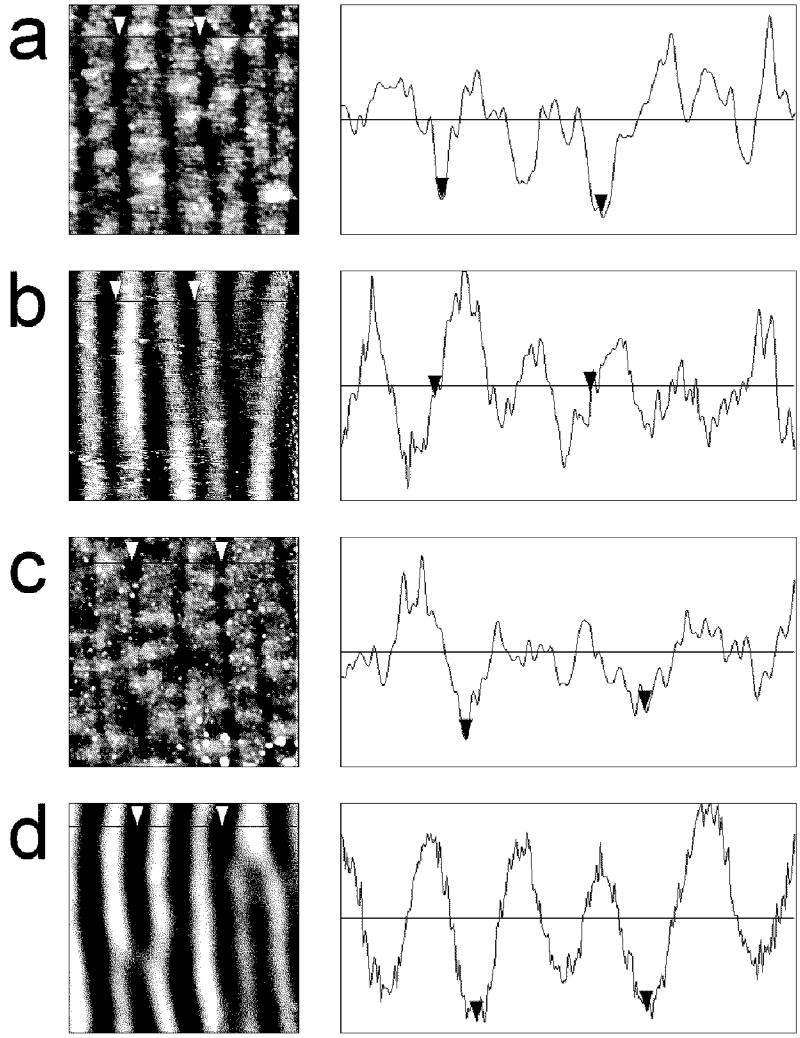

Fig. 1. AFM (a, c) and MFM (b, d) images of MnAs substrate (No. M6332#1) before overgrowth, scan size 2 μm $\times$ 2 μm. The panels on the left show the high profile and the magnetic phase profile, respectively. The scans were made before (a, b) and after (c, d) magnetization procedure (5 T for 30 minutes at 300 K, field perpendicular to the film). Different areas of the same sample are imaged on (a, b) and (c, d).

half forms slightly (a few nanometers) protruded islands of irregular shape. These completely separated islands are laterally tens of micrometers large and easily recognizable even under an optical microscope. On those islands some kind of parallel structures were observed in the case of ZnTe/CdMnTe/ZnTe, but these cannot be directly related to the parallel topographical protrusions of the underlying MnAs substrate (see Fig. 2a,c; RMS = 4 nm, 10pt mean = 30 nm). The MFM signal is measurable only on the above mentioned islands, and no magnetic signal comes from the side (Fig. 2b). The MFM image was checked also after the magnetic field had been applied to the sample. A more pronounced out-of-plane magnetization component was registered from the islands in the case of ZnTe/CdMnTe/ZnTe (Fig. 2d) overlayer compared to the non-magnetized one. There is also a qualitative change in the MFM image: the domains are reduced about twice in size and the weak topography-magnetization correlation — as in Fig. 2a,b — disappears. In contrast, there was no qualitative difference between the MFM images in the case of CdMgTe/CdMnTe/CdMgTe before and after magnetizing the sample.

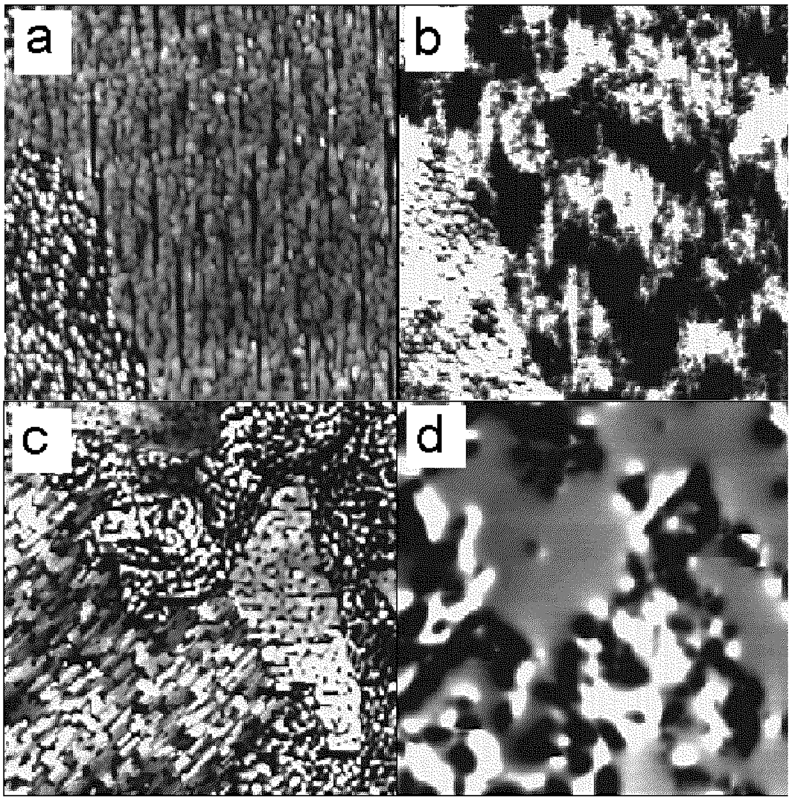

Fig. 2. AFM/MFM images of ZnTe/CdMnTe/ZnTe on a MnAs substrate (sample No. 101002B), scan size 10 μm $\times$ 10 μm: (a) and (b) topography and MFM before magnetizing the sample in an external magnetic field (the same conditions as in Fig. 1); (c) and (d) after magnetizing: topography and MFM. Different areas of the same sample are imaged on (a, b) and (c, d).

The formation of islands may occur due to the Volmer–Weber or the Stransky–Krastanov modes of growth of (II,Mn)VI. An alternative reason of such topography and magnetic properties of the overgrown samples may originate from a chemical destruction of some parts of the surface, which then does not show magnetic properties.

3.1.2. (II,Mn)VI QW on GaMnAs substrates

GaMnAs substrates imaged by AFM revealed remarkably flat surfaces (RMS = 1.2 nm, 10pt mean = 8 nm) consisting of homogeneously distributed crystallites of nanometric size. In particular, we did not observe any MFM signal from the substrate that could be associated with MnAs clusters. Most probably either the size was too small or the magnitude of the effect of their magnetization at the sample surface was too small to lead to observable effects. Alternatively, our substrates did not contain any clusters at all, in spite of their annealing at high temperature, which was reported to lead to formation of such clusters [5].

The topographical AFM images of the overgrown quantum structure with a narrow bottom ZnTe barrier revealed circular islands of micrometric diameters protruding about 1 nm from the surrounding area. On the other hand, topographical images of (II,Mn)VI with a wide barrier demonstrated a relatively smooth homogeneous surface. No MFM signal was measurable for the two types of samples, either before or after magnetization.

3.2. Measurements of the luminescence

Figure 3 demonstrates the PL spectra measured at 6 K for non-magnetized and magnetized CdMgTe/CdMnTe/CdMgTe (with narrow barrier) structures

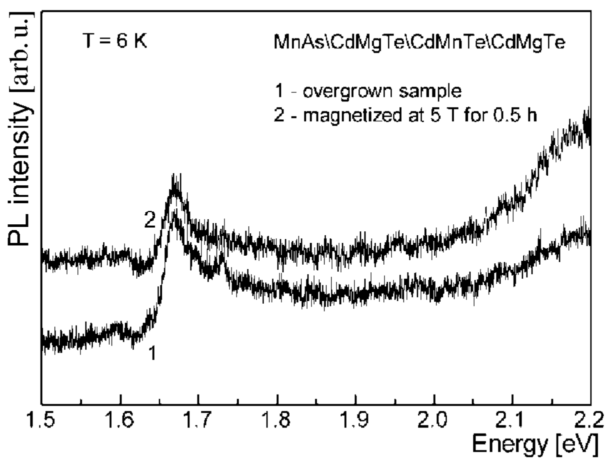

Fig. 3. PL spectra of CdMgTe/CdMnTe/CdMgTe (with thin bottom barrier) on MnAs substrate (sample No. 080902A): (1) as-grown and (2) magnetized at 5 T for 0.5 h.

grown on MnAs. In both spectra we observe an emission line characteristic of CdMnTe (with 4% of Mn) QW at 1.68 eV. The absence of any shift in the spectrum obtained for a magnetized sample may stem from the fact that the average magnetic domain size is considerably smaller than the PL excitation and detection spot. Also, we did not observe any qualitative difference in the CdMgTe/CdMnTe/CdMgTe MFM images before and after magnetizing the samples. The spatially resolved measurements show that the peak position for these samples depends on the position of measurement (magnetic islands emitted the light of higher (1.71 eV) energy), but not on the temperature in the range 4–40 K. This may indicate that the variation is related to, e.g., inhomogeneous strain field rather than magnetization in the DMS QW.

The μ-PL measurements of ZnTe/CdMnTe/ZnTe grown on GaMnAs substrate showed a very wide ($\approx$ 150 meV) line attributed to CdMnTe QW with interdiffused rather than sharp interfaces. This may be due to higher than usual temperature of growth of our CdMnTe QW. The other possibility is that at such elevated growth temperatures the formation of self-assembled quantum dots occurs, which is known to lead to broad luminescence structures. However, as is well known, typical quantum dot μ-PL spectra break into a series of very narrow peaks. Since this does not occur in our case, the second explanation is less probable.

Figure 4 demonstrates the PL spectrum measured for the CdMgTe/CdMnTe/CdMgTe sample with a wide bottom barrier grown on a GaMnAs substrate. In the spectrum we observe two emission lines characteristic of the CdMnTe (with 1% of Mn) QW excitonic recombination at 1.61 eV and CdMgTe barrier at 1.85 eV. The narrow emission line means that we succeeded in growing a quantum structure

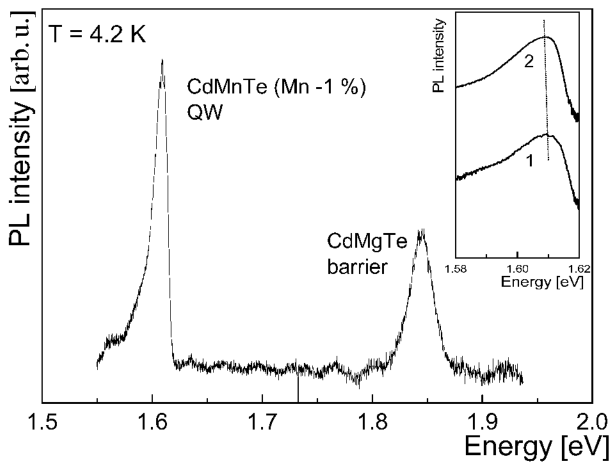

Fig. 4. PL spectrum of quantum structure with thick CdMgTe barrier between CdMnTe QW and GaMnAs substrate (sample No. 040203B). The inset presents the PL peak of the QW measured for (1) as-grown sample and (2) sample magnetized at 5 T for 0.5 h.

with CdMnTe quantum well that confines the carriers. The inset shows spectra of as-grown and magnetized samples. Contrary to the results described for the quantum structures with a narrow bottom barrier, the emission line characteristic of this QW structure is slightly shifted to lower energy after the sample was kept in a magnetic field. To explain the origin of this shift we are planning in the near future to perform PL measurements at lower temperatures (down to 2 K), including spatially resolved μ-PL measurements.

4. Summary

The epitaxial overgrowth of (II,Mn)VI quantum structures was performed on two types of substrates: (i) ferromagnetic MnAs, having a wavy topography, and a magnetic domain structure (as revealed by MFM) directly correlated to the topography of the surface; and (ii) on GaMnAs containing ferromagnetic MnAs clusters, having a smooth surface, but exhibiting no magnetic signal.

The surface topography of the structures grown on MnAs substrates revealed the existence of two kinds of regions: protruded islands consisting of larger grains or possessing higher symmetry, surrounded by areas of small-scale roughness. MFM patterns showed the magnetic domain structure only on the topographical islands. There was no drastic difference in the magnetic MFM image before and after magnetization. In the case of GaMnAs substrates no MFM signal was measurable either before or after depositing the structures, independent of whether the sample was magnetized or not.

In the PL spectra of all (II,Mn)VI quantum structures with narrow bottom barrier grown on both types of substrates we observed the emission line characteristic of the CdMnTe QW. Spatially resolved μ-PL measurements on CdMgTe/CdMnTe/CdMgTe grown on MnAs showed that the peak position depends on where on the sample the measurement was made (magnetic islands emitted light of higher energy), but not on the temperature. The very wide emission line obtained for ZnTe/CdMnTe/ZnTe grown on GaMnAs substrate could be related to the poor crystalline quality of the quantum well or to formation of quantum dots. On the other hand, the narrow emission line attributed to QW excitonic recombination measured for CdMgTe/CdMnTe/CdMgTe sample with a wide bottom barrier grown on GaMnAs indicates that we succeeded to grow a quantum structure with CdMnTe quantum well of good quality on this substrate.

Acknowledgments

This work has been partially supported by the EU within the 5th Framework Programme through QMDS RTN project under the contract HPRN-CT-2000-00134 and through the grant PBZ-KBN-004/P03/2001.

References

[1] F. Matsukura, H. Ohno, T. Dietl, in: *Handbook of Magnetic Materials*, Ed. K.H.J. Buschow, Vol. 14, North-Holland, Amsterdam 2002, p. 1.

[2] M. Ghali, J. Kossut, E. Janik, K. Regiński, Ł. Kłopotowski, *Solid State Commun.* **119,** 371 (2001).

[3] A. Kudelski, K. Fronc, J. Wróbel, S. Maćkowski, G.Cywiński, M. Aleszkiewicz, F. Kyrychenko, T. Wojtowicz, J. Kossut, *Solid State Commun.* **120,** 35 (2001).

[4] L. Däweritz, F. Schippan, A. Trampert, M. Kästner, G. Behme, Z.M. Wang, M. Moreno, P. Schützendübe, K.H. Ploog, *J. Crystal Growth* **227-228,** 843 (2001).

[5] J. De Boeck, R. Oesterholt, A. Van Esch, H. Bender, C. Bruynseraede, C. Van Hoof, G. Borghs, *Appl. Phys. Lett.* **68,** 2744 (1996).