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Structural and optical properties of ultrananocrystalline diamond / InGaAs/GaAs quantum dot structures

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ABSTRACT

The combination of the unique properties of ultrananocrystalline diamond (UNCD) films and of semiconductor quantum dot (QD) structures could significantly improve the performance of different electronic and optoelectronic devices, where e.g. good thermal management and advanced mechanical parameters are required. In the current work quantum dot InGaAs/GaAs heterostructures have been grown by molecular beam epitaxy (MBE) with different densities between $1.6\times10^{10}\,cm^{-2}$ and $1.6\times10^{11}\,cm^{-2}$ controlled by the substrate temperature in the range between 490 and 515 °C. These structures were overgrown with UNCD by microwave plasma chemical vapor deposition (MWCVD) using methane/nitrogen mixtures at 570 °C. Scanning electron microscopy (SEM) reveals that without ultrasonic pretreatment the diamond nucleation density on QD structures is low and only separate islands of UNCD are deposited, while after pretreatment thin closed films are formed. From the cross-section SEM images a growth rate of ca. 3 nm/min is estimated which is very close to that on silicon at the same deposition conditions. The UNCD coatings exhibit a morphology consisting of two types of structures as shown by atomic force microscopy (AFM). The first one includes nodules with diameters between 180 and 350 nm varying with the density of the underlying QDs; the second is formed by a kind of granular substructure of these nodules with diameters of about 40 nm for all QD densities. The optical properties were investigated by photoluminescence (PL) spectroscopy before and after the deposition of UNCD. The PL signals of QD structures overgrown with UNCD, although with decreased intensity, remain almost unchanged with respect to the peak positions and widths, revealing that the UNCD/QD structures retain the optical properties of uncoated InGaAs/GaAs quantum dots.

1. Introduction

Quantum dots (QDs) of III–V semiconductors have been intensively investigated in the last decade because of their unique physical properties. These structures attract great scientific interest due to their potential applications for fabrication of different electronic and optoelectronic devices, such as superluminescent diodes for optical sensors [1], quantum dot lasers [2–4], single electron transistors [5], and single photon emitters [6]. The electronic and optical properties of the QD structures can be tailored by their composition, size and spatial arrangements, controlled by the growth conditions. Another material of interest for preparation of solid-state quantum devices is diamond [7]. In addition to its superior mechanical, tribological and chemical properties, diamond possesses very wide optical bandgap, low electron concentrations and low phonon scattering at room temperature [8]. This allows the fabrication of high quality factor two-dimensional photonic crystal microcavities, e.g. in nanocrystalline diamond (NCD) films, enhancing the light–matter interaction for quantum electrodynamic applications [9]. The combination of the unique properties of diamond and of semiconductor quantum dot structures could lead to the preparation of novel quantum devices.

Another common application of semiconductor QDs and diamond is in high-power devices, e.g. lasers, which require good thermal management in order to overcome the production of heat in very small areas during their operation. A possible solution is the deposition of a thin layer of material with high thermal conductivity between the device and the cooling system. Polycrystalline diamond (PCD), prepared by chemical vapor deposition, possesses thermal conductivity on the order of 2000 W/mK [10], superior to copper over a wide temperature range and has already found application in the thermal management of ICs [11] and high-power laser diodes [12]. Nanocrystalline diamond (NCD) films deposited at lower temperatures retain to a great extent the superior properties of PCD, being at the same time much smoother. The thermal conductivity of such films depends on the thickness and is about half of that of bulk diamond due to scattering by the grain boundaries, but still reaching values up to ca. 1400 W/mK [13], which makes them suitable for thermal management in high-power devices.

In the present work we have studied the possibility for deposition of ultrananocrystalline diamond films on InGaAs QDs and investigated the morphology and optical properties of the obtained structures.

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Table 1
QD densities and heights as well as PL signal parameters before and after the overgrowth with UNCD.

Sample	Substrate temperature	QD density	Height of QD	PL signal	
				Position	FWHM
	[°C]	[cm^{-2}]	[nm]	[eV]	[meV]
LD-QD	515	1.6×10^{10}	3.5÷5.5	1.320	53
UNCD/LD-QD				1.316	50
HD-QD	490	1.6×10^{11}	3.0÷7.7	1.211	71
UNCD/HD-QD				1.220	67

2. Experimental

Quantum dot $In_{0.6}Ga_{0.4}As$ heterostructures have been grown on (100) GaAs substrates using GEN II molecular beam epitaxy (MBE) system. All structures included 200 nm GaAs buffer layer deposited at a substrate temperature of 590 °C, which was lowered to the desired one for the subsequent formation of the QDs without interruption of the growth process. For the quantum dot formation a thin $In_{0.6}Ga_{0.4}As$ layer with a nominal thickness of 1.4 nm was deposited on the buffer layer. The as-grown QDs were covered with a 50 nm GaAs barrier layer and a second QD uncapped layer for morphology investigations, both grown at the same substrate temperature like the first QD layer. The density of the dots was controlled by variation of the substrate temperature [14] as shown in Table 1.

The QD structures were coated with ultrananocrystalline diamond (UNCD) film by microwave plasma chemical vapor deposition (MWCVD) using 17% CH_4/N_2 gas mixtures in the set-up described elsewhere [15]. The substrate temperature was 570 °C, the working pressure 2.3 kPa, the forwarded MW power 800 W, the deposition time 30 min. Under similar growth conditions films composed of diamond nanocrystallites (3–5 nm in diameter) embedded in an amorphous carbon matrix have been deposited on different substrates, like silicon, polycrystalline diamond, c-BN, TiN, etc [16,17]. In the current work bare GaAs substrates were also coated with UNCD film for comparison. In order to enhance the diamond nucleation, part of the samples were pretreated ultrasonically for 60 min in a suspension of 80 mg ultra-disperse diamond powder (3–5 nm grain size) and 50 mg diamond powder (250 nm grain size) in n-pentane prior the deposition.

The topography of the QD structures before and after coating with UNCD film was investigated by atomic force microscopy (AFM, NanoScope III) in a tapping mode and the analysis of the images was performed with Gwyddion software. The morphology and the thickness of the top UNCD layer were studied by scanning electron microscopy (SEM, Hitachi 3000). The optical properties of the uncoated and coated samples were measured with a standard photoluminescence (PL) set-up. A DPSS-laser with a wavelength of 532 nm and a spot of ca. 400 µm with Gaussian intensity distribution was used for the optical excitation, and the emitted PL light in reflection was focused into a 1/4 m monochromator. The resulting signals were detected by a thermoelectrically cooled silicon photodiode in combination with a Lock-In amplifier.

3. Results and discussion

The density and height of the QDs before the overgrowth with UNCD were determined from the AFM images, as presented in Fig. 1, and the results are summarized in Table 1. Varying only the substrate temperature QDs with different densities were prepared: 1.6×10^{11} cm^{-2} at 490 °C (HD-QD in the following) and 1.6×10^{10} cm^{-2} at 515 °C (LD-QD). With increasing the substrate temperature the density of the dots decreases, while their height increases, due to the increased surface migration length of In atoms at higher temperatures [14].

The topography of the prepared UNCD/QD structures was also studied by AFM. From the images the existence of two types of structures in the UNCD morphology is observed. The first one consists of nodules with diameters of 180–350 nm, decreasing in the order bare GaAs substrate — LD-QD–HD-QD, and accompanied by a decrease of the height difference (Fig. 1 (d)–(f)). The second type of structures is represented by a kind of granular substructure of these nodules with diameters of about 40 nm irrespective of the size of the main structures. It can be supposed that the size and density of the main structures are determined by the initial nucleation density on the substrates overgrown with UNCD, where the density of the QDs plays an additional role to the nucleation process. This is the reason for the smaller size of the main UNCD structures grown on the QDs with higher density. The size of the substructures is determined to a

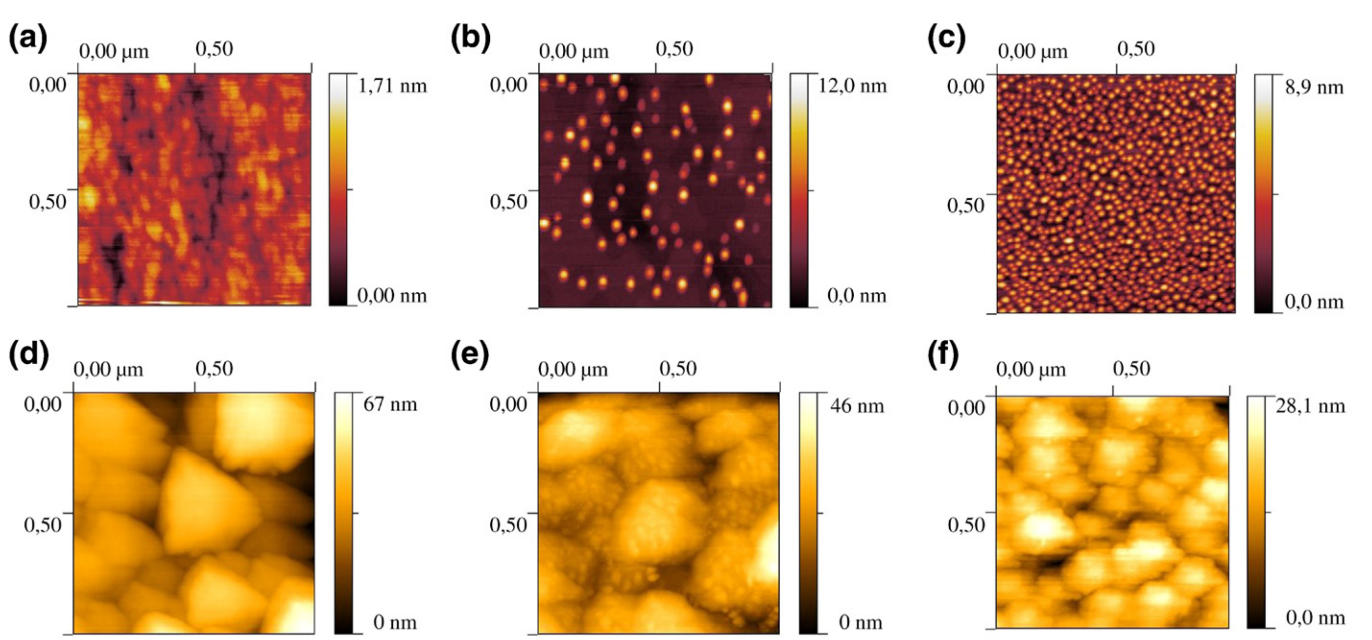

Fig. 1. AFM images of (a) bare GaAs substrate, (b) QDs with low density (LD-QD), (c) QDs with high density (HD-QD), (d) GaAs substrate coated with UNCD layer, (e) LD-QD coated with UNCD layer, (f) HD-QD coated with UNCD layer.