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GaAs quantum dot molecules filled into droplet etched nanoholes

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ABSTRACT

We fabricate self-aligned vertically stacked GaAs quantum dot molecules (QDMs) by filling of self-assembled nanoholes in AlGaAs. The tunable nanoholes are created using local droplet etching (LDE) combining conventional molecular beam epitaxy with self-assembled, lithography-free patterning. The optical emission from single, strain-free QDMs shows clear excitonic features with linewidths below 150 μeV after optimizations of the fabrication process. This allows investigations of the coupling among the individual dots forming a QDM. In electric fields oriented along the axis of the QDM, luminescence emission from direct and indirect transitions can be clearly distinguished. Furthermore, an anti-crossing behaviour demonstrates inter-dot coupling in the QDM.

1. Introduction

Closely spaced semiconductor quantum dot (QD) pairs are highly interesting objects for studies of coupling between quantized electronic systems. In case of significant coupling they are called quantum-dot molecules (QDM) [1–4]. In addition to fundamental research related topics [2,3], QDMs are also promising for potential applications in quantum information processing e.g. as quantum gates [4–6] controlling the entanglement between solid-state qubits. Several approaches have been demonstrated for the self-assembled formation of laterally [7] or vertically aligned QDMs. An established method for vertical alignment is the utilization of strain allowing the creation of QDMs composed of two stacked InAs QDs [3–6]. We discuss here a novel approach for the self-aligned fabrication of strain-free GaAs QDMs in AlGaAs by filling two QDs and a separating tunnel barrier into a droplet etched nanohole.

Local droplet etching (LDE) allows the self-assembled creation of nanoholes in several arsenide based compound-semiconductor surfaces [8,9]. An example of an AlGaAs surface with low-density nanoholes after LDE with Al droplets is shown in Fig. 1a. The whole process is performed in a conventional solid-source molecular beam epitaxy (MBE) growth chamber. LDE works with various droplet (Ga, Al, In, AlGa, InGa) and substrate (GaAs, AlAs, AlGaAs) materials. The nanohole openings are surrounded by walls composed of arsenides of the droplet material [10]. Here, we use Al droplets for drilling of low-density holes with optically inactive AlAs walls into AlGaAs surfaces.

Filling of LDE nanoholes in AlGaAs with GaAs allows the fabrication of uniform GaAs quantum dots (QDs) with ground-state emission energy tuneable from 700 to 800 nm by the filling level [10]. Single LDE QDs exhibit sharp excitonic lines with linewidth down to 25 μeV [11], a low neutral exciton fine-structure splitting (FSS) down to less than 5 μeV [11,12], and single-photon emission with a pronounced minimum of the autocorrelation function of 0.01 [11].

In a previous publication [13], we have already demonstrated the fabrication of stacked QD pairs filled into a droplet-etched nanohole. However, there, the optical emission from a QD pair shows lines that are rather broad with linewidths above 1 meV. Such broad lines do not allow the separation of the different excitonic features, with, e.g., a typical splitting of 1–2 meV between the exciton and biexciton peaks [14]. In the present paper, we describe optimizations of the QDM fabrication procedure yielding optical linewidths below 150 μeV. Gate-voltage dependent experiments demonstrate resonant coupling of QDM states.

2. Sample fabrication

The samples are fabricated using solid-source MBE on (001) GaAs substrates. After a buffer layer for surface smoothening, a 50 nm thick Si-doped (1×10^{18} cm^{-3}) GaAs layer is grown as a back gate. This is followed by a 120 nm thick $Al_xGa_{1-x}As$ ($x = 0.33$) layer, in which the nanoholes are drilled via droplet etching. For LDE, the As_4 flux is minimized by closing the valve and shutter of the Arsenic valved-cracker cell as well as the main shutter in front of the sample surface. This yields an As flux of less than 1×10^{-7} Torr, which is at least hundred times lower than typical

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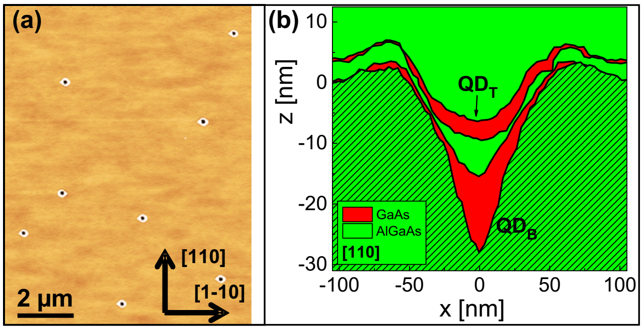

Fig. 1. (a) AFM image of an AlGaAs surface with uniform, low-density nanoholes after local droplet etching (LDE) with Al droplets at T = 640 °C. QDMs are fabricated by hole filling. (b) AFM linescans along [1 1 0] direction from a sample series illustrating the different interfaces during QDM fabrication, i.e., the initial nanohole in the AlGaAs substrate (patterned green), the hole filled with the bottom dot QD_B (red), the AlGaAs or AlAs tunnel barrier (green), the top dot QD_T (red), and the AlGaAs cap layer (green). We note that unintentional fluctuations of the process conditions during fabrication of the samples of this series do not allow a reliable determination of the respective dot sizes and of the tunnel barrier thickness from the AFM data. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

GaAs growth conditions. Now, Al is deposited as droplet material at a temperature T, with amount corresponding to 1.0 monolayers (ML) of AlAs, and with flux corresponding to 0.4 ML/s AlAs growth. The liquid Al droplets are formed in a self-assembled fashion in Volmer-Weber growth mode [15]. The droplets are transformed into nanoholes during a 180 s post-growth annealing step at unchanged temperature [9]. The central processes for nanohole drilling are the diffusion of As from the substrate into the liquid droplet material driven by the concentration gradient and the concomitant material transport from the metal droplet onto the whole surface. The mechanism is discussed in more detail in Ref. [16]. The depth of droplet-etched nanoholes can be tuned by the process parameters from 1 to more than 100 nm [17].

For QDM fabrication, about 28 nm deep nanoholes are filled with 0.45 nm GaAs in a pulsed deposition mode (0.5 s deposition/10 s growth interruption) to form the bottom quantum dot QD_B, followed by 3 nm AlGaAs or AlAs for the tunnel barrier, and 0.9 nm GaAs for the top dot QD_T again using pulsed deposition. Finally, a 73 nm thick AlGaAs cap layer is grown. The Ga flux corresponds to 0.8 ML/s growth speed, the Al flux to 0.4 ML/s, and the growth temperature is 600 °C.

To allow gate-voltage dependent measurements, the QDMs are embedded into a Schottky-diode like geometry with an 18 nm thick evaporated Ti-layer as a top gate. A schematic of the final structure is shown in Fig. 2.

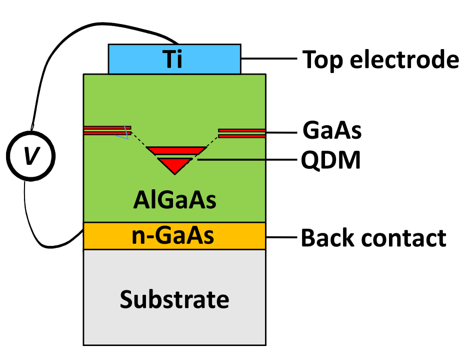

Fig. 2. Schematic of a self-aligned GaAs QDM being integrated in a Schottky-diode for field-dependent measurements. The QDMs are created by filling of droplet etched nanoholes in AlGaAs.

3. Structural characterization

Atomic force microscopy (AFM) linescans from a sample series where the fabrication process has been stopped at the different interfaces is shown in Fig. 1b. Obviously, due to the shape of the nanohole template, bottom and top dot have significantly different shapes. The bottom dot is roughly shaped like an inverted cone whereas the top dot is rather disc-like. Furthermore, the thickness of the tunnel barrier is not uniform with a maximal thickness at the QDM axis.

In order to model the hole filling we assume that a nanohole is shaped like an inverted cone. Furthermore, we assume that all material arriving through the hole opening will migrate to the hole bottom driven by capillarity and form the dot there. This yields a filling material volume per nanohole of $V_F = \pi d_F W^2/4$, with filling layer thickness d_F, the diameter $W = 2d/\tan\alpha$ of the approximately circular hole opening, the hole depth d, and the angle $\alpha \cong 0.8d[\mathrm{nm}]^{0.4}$ [16] between the hole side-facets and the plane surface. An inverted cone-shaped QD has a volume $V_{QD} = \pi h_{QD} W_{QD}^2/12$, with the QD height h_{QD} and top-area radius $W_{QD} = 2h_{QD}/\tan\alpha$. Setting $V_{QD} = V_F$ yields the quantum dot height $h_{QD} = (3d^2 d_F)^{1/3}$. For the bottom QD with d_F = 0.45 nm we calculate a height of 10.2 nm. In the next step we calculate filling with QD_B plus tunnel barrier yielding now d_F = 3.45 nm. Here, a total height of 20 nm is calculated. Filling in addition with the top dot (d_F = 4.35 nm) yields a total height of 22 nm. The QDM structural properties calculated with this simple model mostly agree with trend indicated by the AFM data of Fig. 1b.

4. Photoluminescence linewidth optimization

The optical properties of the LDE QDMs are investigated using micro photoluminescence (PL) spectroscopy at low temperature (T = 6 K). Due to their low density, single QDMs are selected using a focussed laser with energy of 2.33 eV for excitation.

For the fabrication of single GaAs QDs we use about 15 nm deep holes created at a LDE process temperature T = 600 °C as template [11]. However, for filling with a QDM the holes must be significantly deeper. An increase of the temperature T during droplet etching from 600 up to 660 °C yields an increase of the hole depth from 15 up to about 50 nm [11]. However, we observe an abrupt broadening of the single-dot PL linewidths from less than 100 µeV for $T \leq 640$ °C to several meV for $T > 640$ °C [11]. The same crucial line broadening is observed for filling with a QDM. This identifies for the present process technique an upper limit of 40 nm for the hole depth if the linewidth has to remain small.

Under consideration of the above limit we perform the LDE processes for the fabrication of the present QDM samples at T = 630 °C. This yields a nanohole template with depth of about 28 nm and density of about 1.5×10^7 cm^{-2}. By filling of these nanoholes we have fabricated QDM samples using both, AlGaAs and AlAs tunnel barriers. Examples of the respective low-temperature PL emission are shown in Fig. 3. The emission from the top dot at higher energy is clearly separated from the bottom dot PL due to the strong quantization of QD_T along growth direction. Variations of the respective layer thicknesses used for hole filling allow to independently tune the bottom and top dot energy levels as well as the thickness of the tunnel barrier. Interestingly, the material of the tunnel barrier crucially influences the linewidth of the PL emission. QDM samples with AlGaAs tunnel barrier show broad PL lines with width of several meV (Fig. 3a) whereas those with AlAs barrier demonstrate a narrow linewidth below 150 µeV (Fig. 3b). We attribute the dot PL peak broadening to a fluctuating Stark-shift [18,19] caused by fluctuating charges in crystal traps or on neighbored surfaces

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