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Effects of growth temperature and arsenic pressure on size distribution and density of InAs quantum dots on Si (001)

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Abstract

InAs self-assembled quantum dots (QDs) were grown on Si (001) substrates via molecular beam epitaxy. The size distribution and density of InAs QDs grown under different conditions were studied using plan-view transmission electron microscopy. Dot density was shown to strongly depend on arsenic beam equivalent pressure (BEP) ranging from 2.8×10^{-5} to 1.2×10^{-3} Pa. In contrast, dot density was nearly independent of substrate temperature from 295 to 410 °C under constant arsenic BEP, while broadening of size distribution was observed with increasing temperature. The mechanism accounting for some of the main features of the experimental observations is discussed. Finally, InAs quantum dots with optimized narrow size distribution and high density were grown at low arsenic BEP of 7.2×10^{-5} Pa and low temperature of 250 °C followed by annealing at arsenic BEP of 1.9×10^{-4} Pa and temperature of 410 °C.

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1. Introduction

Epitaxial InAs self-assembled quantum dots (QDs) on GaAs (001) substrate is one of the most extensively studied hetero-structure in terms of its optoelectronic properties, such as type-I direct energy band alignment and discrete energy levels for both holes and electrons [1–5]. The interest in InAs quantum dots has been extended to those fabricated on Si substrates because of the potential application in Si-based optoelectronic devices [6–8]. From the standpoint of materials physics, InAs quantum dots on Si represents a substantially more complex subject. In comparison to InAs quantum dots on GaAs (001), InAs/Si(001) is associated with a much higher lattice mismatch (11%) as well as a polar–nonpolar interface. Currently, the understanding of the formation mechanism of InAs quantum dots on Si is lacking [7,9,10]. There is very little knowledge in the literature with regard to controlling the dot size distribution and density, two important concerns for optoelectronic applications. Therefore, a systematical study of size distribution and density as a function of growth conditions is necessary.

In this paper, we report experimental studies of the dependence of InAs quantum dot size distribution and density on various growth conditions, including substrate temperature and arsenic beam equivalent pressure (BEP). Quantum dot size distribution and density were measured using plan-view transmission electron microscopy (TEM). The experimental results indicate that dot density is strongly affected by arsenic BEP and depends only slightly on substrate temperature. A clear broadening of dot size distribution was observed with increasing growth temperature indicating coarsening (Ostwald ripening). A mechanism is discussed to account for the observed dependence.

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InAs QDs with narrow size distribution and high density have been achieved via growths at low temperature and BEP followed by annealing.

2. Experimental details

InAs dots were grown on n-type (001) orientated Si substrate in a Perkin-Elmer 430 MBE with an arsenic cracker cell. Si substrates were cleaned using the following recipe: 1) H_2O_2:H_2SO_4 (3:5) 1 min, 2) HF:H_2O (1:10) 1 min. This procedure was repeated 3 times with the final step being the HF dip, rendering H-terminated Si surface. The substrates were blown dry with nitrogen and immediately loaded into the MBE chamber to preserve the clean H-passivated surface. The background vacuum in MBE chamber was kept below 10^{-7} Pa with liquid nitrogen cryo-shield. Before growth, substrates were degassed for 50 min at 250 °C to desorb carbon-containing species. H desorption was carried out at 780 °C until the appearance of a clear 2×1 RHEED pattern. At this point, substrate temperature was ramped down to the growth temperature and InAs was deposited directly on Si, without buffer layer deposition. InAs growths were initiated by opening the arsenic shutter 2 min before opening the indium shutter. In all growths, we use InAs growth rate of 0.02 ML/s with indium BEP of 7.6×10^{-6} Pa. Nominal InAs coverage of 0.7 ML was deposited as determined by InAs growth rate. For growth rate calibration, InGaAs was grown on GaAs. Composition of InGaAs was determined by analyzing lattice constant using reciprocal space maps of X-ray diffraction (XRD). Thickness of InGaAs epilayer was obtained from both XRD fringes and cross-section TEM. Substrate temperature was varied from 295 to 410 °C for growth temperature dependence studies with constant arsenic BEP of 1.9×10^{-4} Pa. For arsenic BEP dependence studies, arsenic BEP was varied from 2.8×10^{-5} to 1.2×10^{-3} Pa, while the growth temperature was held constant at 320 °C. After growths, samples were removed from the MBE chamber and exposed to the atmosphere without capping. Dot density and size distribution were determined using plan-view TEM in a 200 keV JEOL 2000 FX machine. The micrographs were collected under [001] zone axis conditions, i.e. with the electron beam oriented parallel to the growth direction of the sample.

3. Results and discussion

The appearance of well defined dots at coverage below 1 ML indicates Volmer–Weber growth mode, as shown in Fig. 1. The left side of Fig. 1 shows bright-field plan-view [001] TEM micrographs of samples grown at different substrate temperatures increased from 295 °C for (a) to 410 °C for (e). The right side of Fig. 1 is the histogram of size distribution based on the plan-view TEM. Gaussian curves were fitted on the size distribution histogram of Fig. 1. It is clear that increasing substrate temperature results in broadening of dot size distribution. Gaussian peak width, referred to as 0.849 full-width at half maximum (FWHM), increases from 8 to 14 nm when substrate temperature increases from 295 to 410 °C. Coarsening is likely to be responsible for the broadening of dot size distribution. Higher substrate temperature leads to more rapid surface diffusion and subsequently more significant coarsening (Ostwald ripening). This is because large dots grow at the expense of small ones via migration of adatoms. Therefore, more large dots will form at higher temperature growths. Fig. 2 shows the dependence of dot density on substrate temperature. Dot density is nearly independent of substrate temperatures. The dot density at the onset of nucleation can be written for the well-studied quantum dot systems such as Ge/Si and InAs/GaAs [11,12] as follows[13]:

$$n \propto \left(\frac{F}{D_s}\right)^{\alpha} \tag{1}$$

where F is the growth flux coming from source, D_s is the surface diffusion coefficient which increases exponentially ($D_s=D_0\exp(-E_a/k_BT)$) with increasing temperature. α is a constant associated mainly to the critical cluster size. α increases with increasing cluster size and saturates at unity for critical cluster size larger than 10 atoms [14]. For epitaxial growth, α can be treated as a constant [11,12]. n in Eq. (1) is the initial dot density. This simple model cannot explain the experimental observation that the dot density is nearly independent of temperature within the range of 295 to 410 °C under a constant arsenic BEP. This suggests that the nucleation model as in Eq. (1) that has been shown to be valid for Ge/Si and InAs/GaAs systems, does not apply to InAs growth on Si under our growth conditions.

Fig. 3 shows the plot of InAs dot density dependence on arsenic BEP. The dot density was counted from [001] TEM images, covering total area of 4 μm^2. It turns out that dot density increases by two orders of magnitude in a near exponential fashion with decreasing arsenic BEP from 2.8×10^{-5} to 1.2×10^{-3} Pa. This behavior is quite different from that in InAs/GaAs. For InAs/GaAs, both As_4 and As_2 (cracked As_4) have been investigated in the scientific literature. It seems that no consistent trend emerges from the numerous studies on InAs/GaAs QDs growth dependence on arsenic BEP [15–20]. In later work, effects of arsenic BEP on dot density was shown to be temperature related where dot density increases with decreasing As_4 BEP at low temperature of 420 °C [17,18]. Authors suggest that As_4 dissociation would be the limiting step of InAs QDs growth at low temperature. Therefore, increasing As_4 BEP enhances the incorporation rate of indium in InAs islands which would reduces dot density. However, the situation becomes different at high temperature (above 480 °C) when As_4 dissociative reaction is no longer the rate limiting step for InAs deposition, whereby dot density becomes nearly