Coupled Ge Quantum Dot Crystals

Growth, Microstructures and Optical Properties

Coupled three-dimensional Ge quantum dot crystals (QDCs) are realized by multilayer growth of quantum dots (QDs) on patterned Si (001) substrates. With increasing the vertical periodic number of the QDCs, the photoluminescence (PL) spectral linewidth decreased exponentially, and so did the peak energy blueshift caused by increasing excitation power, which are attributed to the electronic coupling and thus the formation of miniband.

Introduction

Self-assembled SiGe quantum dots (QDs) on Si (001) substrates has been investigated broadly not only to learn inherent mechanism during heteroepitaxial growth but also for their potential applications in devices. To realize QD-integrated opto-electronic devices, it is demanded to control QDs positions and size, as well as size distribution [1]. Considerable effort has been devoted to control the growth of self-assembled GeSi QDs in recent years. Ordered SiGe QDs with various spatial alignments have been achieved via different strain engineering [2] or lithography [3] techniques. However, thoroughly understanding of the physical properties induced by the ordered QD alignment and high size uniformity are still needed.

High density and three-dimensionally (3D) ordered QD arrays have been referred to as quantum dot crystals (QDCs) [3]. The strong coupling among densely ordered QDs in QDCs may provide a path for the engineering of band structure and functionalities. Various physical phenomena, including electrostatic interactions [4], localized states splitting and associated quasimolecular states [5] have been studied in coupled GeSi QD systems. Generally, in 3D artificial QDCs, the QDs play a role similar to that of atoms in real crystals. The band offsets at the interfaces between the QD and the matrix in the QDC also play a role analogous to the periodic potential in real crystals. When QDs are 3D ordered, uniform and in close proximity to one another, significant carrier wave function overlap occurs. Thus, the discrete energy levels of a single QD will split into extended states and emerge as 3D minibands. Considering the compatibility with the sophisticated Si integration technology, there is a substantial motivation to investigate both the fundamental properties and the device applications of miniband formation in GeSi QDC.
In this paper, high density and 3D ordered GeSi QDCs are realized by multilayer growth of QDs on pit-patterned Si (001) substrates via molecular beam epitaxy (MBE). A systematic investigation of the photoluminescence (PL) spectra of this QDC is performed. Anomalous PL features were observed and are explained in terms of miniband formation due to the strong coupling of closely adjacent dots and the emergence of quasioptical phonon modes due to periodic scatters in the QDC.

**MBE Growth of 3D Coupled QDCs**

We developed a feasible Si patterning technology via nanosphere lithography (NSL), by which the size and period of the nanopattern on the Si surface can be easily controlled and modulated [6]. The Ge QDCs samples are grown in a Riber EVA-32 MBE system on pre-patterned Si (001) substrates via NSL. The growth processes for 3D ordered GeSi QDs were studied systematically [7]. The pre-patterned Si substrates were chemically cleaned, outgassed in vacuum at 780°C for 5 min in the MBE chamber. And then the Si buffer layer was deposited. The first Ge QD layer was grown by depositing Ge of nominal coverage of 8 MLs with increasing the substrate temperature from 450 to 550°C. After that, layer stacking of QDs started with 5.5 nm thick Si spacer layers. It was found that the deposition of the first Ge QD layer is very crucial. The first layer of Ge QDs was deposited with the growth temperature ramping form 450 to 550°C in order to ensure the nucleation of QDs in the pits and the subsequent QDs Ostwald ripening, thus to obtain unimodal ordered QDs with high size uniformity. The Si spacer layer was deposited with the growth temperature ramping form 400 to 550°C, the initial low temperature is to suppress the atomic intermixing while the subsequent high temperature is to ensure a flat Si surfaces after the spacer layer growth. To investigate coupling effect in 3D QDCs, Ge QDCs with single, 5, 10, 15 and 20 stacked dot layers were fabricated and characterized.
Figure 2(a) shows the surface morphology of the pyramid shaped Ge islands that nucleated in the ordered nanopits on a pit-patterned Si (001) substrate. The uniform Ge islands are arranged in a hexagonal lattice with a periodicity of 100 nm. Lateral ordering of the QDs are maintained after 15 layer stacking, as shown in figure 2(c). Based on statistical analyses of the QD height, shown in figure 2(d), the mean height of the Ge QDs in the 15 layer (topmost) was found to be 7.8 nm with a standard dispersion of 10%. The vertical ordering can be verified by the XTEM image shown in figure 3, which is obtained from a sample of ten-layer uncapped QDs grown on a patterned substrate with the same growth parameters. From figure 3(a), the in-plane interdot spacing can be estimated to be ~10 nm. From the high resolution XTEM image shown in figure 3(b), the vertical interdot spacing and the QD height in the center-line of a QD column are found to be ~2.5 and ~4.5 nm, respectively. A schematic side view illustration of the QDC structure is illustrated in figure 3(c).

**PL Features of the Coupled 3D Ge QDCs**

Figure 4(a) shows the PL spectrum of a capped 15 layer Ge QDC and a random Ge QDs sample that was grown under the same conditions measured at 16 K and under excitation power of 0.3 W. The intensity of the PL peak for the QDC is much stronger than that for the random QDs. This result demonstrates that the optical properties of the QDC can be significantly improved compared to the random QDs. The power dependent PL spectra of the capped 15 layer Gei QDC sample measured at 16 K are shown in figure 4(c). It is evident that the NP and TO peak energies maintain a nearly constant value of 872 (±3 meV) and 818 meV (±3 meV), respectively, as shown in figure 5(a). Such features are significantly different from previously reported PL spectra obtained from Ge QDs with the type-II band alignment, which show a large blueshift with increasing excitation power. We attribute such anomalous PL features to the formation of minibands in our GeSi QDC samples due to the strong interdot coupling [8]. The FWHM of the NP peak is slightly increased due to the filling effect. The FWHM of the TO replica shows a much different power dependence, as shown in figure 5(b). We attribute the decrease of the FWHM of the TO replica with excitation power to be the effect of quasioptical phonon emergence in our QDC sample [8].

The linewidth and the peak energy blueshift (at the excitation powers of 0.07 and 1.2 W respectively) as a function of the periodic number are plotted in figure 6. Both can be modeled by an exponential fit based on tight-binding formalism. Those PL features are also attributed to the electronic coupling and thus the formation of miniband [9].

**Conclusion**
In summary, we realized the growth of coupled 3D Ge QDCs on patterned Si substrates, and investigated their optical properties with various periodic numbers along vertical direction via excitation power dependent PL measurements. The PL spectral linewidth, as well as the peak energy blueshift caused by increasing excitation power, decreases exponentially with the periodic number. Those observed optical properties suggest that the hole ground state miniband is essentially formed in our QDCs when the vertical periodic number is over 15.

References

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