Self-assembled ErAs islands in GaAs: Growth and subpicosecond carrier dynamics

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We report the growth of self-assembled ErAs islands embedded in GaAs by molecular beam epitaxy. The nucleation of ErAs on GaAs occurs in an island growth mode leading to spontaneous formation of nanometer-sized islands. Several layers of ErAs islands separated by GaAs can be stacked on top of each other to form a superlattice. X-ray diffraction shows superlattice fringes from such samples. Pump–probe measurements indicate carrier capture times as short as 120 fs. These capture times are strongly correlated with the period of the superlattice. © *1999 American Institute of Physics.* [S0003-6951(99)02848-X]

Materials with subpicosecond response times allow the fabrication of sources of THz radiation such as photomixers[1] and photodetector operation up to 560 GHz.[2] The response time of a material is a crucial parameter for device design and performance, and engineering control over this time is desirable. The most widely used material for these types of applications is low-temperature-grown GaAs (LTG GaAs). The control over the response time is achieved by changing the growth and the postgrowth anneal temperatures, which in turn determine the properties and concentrations of point defects and As precipitates. Doping of semiconductors with transition or rare-earth metals can also yield material with fast response times. In particular, response times as short as 1.5 ps were reported for Er-doped GaAs.[3,4] Usually, the response time of these materials is understood in terms of impurity recombination according to Shockley–Reed–Hall theory (SRH theory). Here, we report on self-assembled ErAs islands in GaAs. We focus on the growth by molecular beam epitaxy (MBE) and on the response times, which are investigated on a subpicosecond time scale by pump–probe measurements. The study of self-assembled ErAs islands in GaAs also has several other motivations: Samples similar to the ones discussed in this letter show interesting properties in magnetotransport at low temperatures.[5] Furthermore, the ErAs islands are nanometer sized and form spontaneously during growth similar to self-assembled quantum dots (SAQD).[6]

Recent reports describe a way to fabricate ErAs particles that is based on doping GaAs with Er beyond the solubility limit.[4,7] The excess Er that cannot be incorporated as dopants forms nanometer-sized ErAs precipitates. We have taken a different approach based on the fact that the nucleation of ErAs on GaAs occurs in an island growth mode[8] driven by surface chemistry rather than strain, as in the case of InAs SAQDs.

Our samples were grown by MBE on semi-insulating (100) GaAs substrates using a Varian Gen II MBE machine. The Er source is a solid-source effusion cell operated at a temperature of 1050 °C resulting in an ErAs growth rate of 0.039 ML/s. We measure the Er flux with the optical flux monitor (OFM).[9] As expected for a solid-source effusion cell, the Er flux shows little variation over time periods of several months. The variation of the Er flux with temperature agrees well with the vapor pressure over solid Er. An absolute flux calibration with an accuracy of approximately 10% was obtained by Rutherford backscattering measurements on samples with continuous ErAs films.

Due to the fabrication process, the ErAs islands in our samples are arranged in layers. As shown schematically in
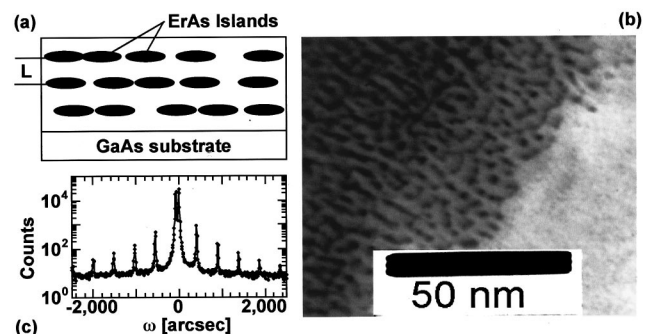

FIG. 1. Sample structure: (a) schematic cross section showing the superlattice structure, (b) plan-view TEM showing the ErAs islands, and (c) x-ray rocking curve.

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Fig. 1, several of these layers can be stacked on top of each other to form a superlattice with period L. The GaAs was grown under standard growth conditions. The ErAs was deposited at the same temperature of 535 °C as the GaAs matrix. For each layer of ErAs islands we used the following procedure: After closing the Ga shutter, the As shutter is closed as well and the Er shutter is opened for the appropriate time. The As to form ErAs is supplied only by the background As pressure in the growth chamber. From the reading of the beam flux monitor we estimate an As:Er ratio of 25:1 while the As shutter is closed. Before opening the Er shutter the GaAs surface shows a 2×4 reconstruction in reflection high-energy electron diffraction (RHEED). Upon opening the Er shutter, the reconstruction changes and a weak 1×1 RHEED pattern is observed.

The density and size of the islands can be controlled by the growth parameters. A higher growth temperature leads to larger islands and a higher ErAs deposition results in a higher density of the islands. Figure 1 also shows a plan-view transmission electron micrograph (TEM) of a sample grown at $T_{growth} = 535$ °C and an ErAs deposition of 0.6 ML. From the TEM we estimate an island density of 7×10^{12} cm^{-2} and an island diameter between 1 and 2 nm.

During overgrowth of the ErAs with GaAs, the RHEED pattern turns spotty initially. After deposition of a few nanometer of GaAs, the RHEED pattern becomes streaky. Before the next layer of ErAs islands is deposited, the GaAs surface again shows the 2×4 reconstruction. For ErAs depositions thicker than 3 ML the overgrowth of ErAs films with GaAs becomes difficult.[10,11] We believe that these problems are circumvented in the case of ErAs islands because the GaAs overgrowth can seed from the underlying GaAs crystal.[11]

As shown in Fig. 1, x-ray rocking curves around the (004) diffraction peak of the GaAs substrate clearly show superlattice fringes. The superlattice period calculated from these fringes is within 2% of the design. The zeroth-order superlattice peak is split from the GaAs substrate peak indicating compressive strain in the superlattice. This is expected because the ErAs lattice constant exceeds the GaAs lattice constant by 1.6%. Similar results were reported earlier by Yamaguchi and Horikoshi.[12] At the very least these data show that the period L of the superlattice is well defined.

Time-resolved differential reflectance measurements were performed on samples grown under several different growth conditions. The experimental setup is a pump–probe arrangement using short laser pulses generated by a mode-locked Ti:sapphire laser. The laser pulses are centered at wavelengths ranging from 800 to 860 nm. They are between 80 fs and 140 fs long. The pump and the probe pulses are polarized orthogonally to each other. The $\Delta R/R$ signals are on the order of 10^{-4}. For their measurement, lock-in detection employing a rf front end is used. All wavelengths used are shorter than the wavelength corresponding to the GaAs band gap of 870 nm at room temperature. Linear absorption measurements on similar samples show that the position of the band gap is not affected by the presence of the ErAs islands.[13] The absorption coefficient for the wavelength used in our experiments is approximately 25% higher than for a GaAs reference sample. We estimate that each pump pulse
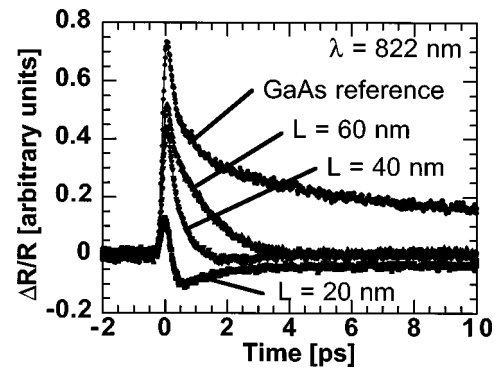

FIG. 2. Time-resolved differential reflection traces on ErAs containing samples with different superlattice periods L.

injects approximately 5×10^{17} cm^{-3} carriers into the GaAs matrix.

In Fig. 2 results from three samples that contain ErAs islands and a GaAs reference sample are compared. The GaAs reference sample is a 1.2-μm-thick GaAs film on a GaAs substrate grown under the same conditions as the ErAs-containing films. All four samples are grown on the same day. The ErAs-containing samples differ from each other by the period of the superlattice L ($L = 20$ nm, $L = 40$ nm, and $L = 60$ nm). The three samples have the following structures: $20 \times (1.2$ ML ErAs, 60 nm GaAs), $40 \times (1.2$ ML ErAs, 40 nm GaAs), $60 \times (1.2$ ML ErAs, 20 nm GaAs). The data show that first, having the ErAs islands changes the observed signal significantly. After an initial positive transient the signal returns to a value close to the base line within a few picoseconds in contrast to the GaAs reference. Second, the different periods L of the superlattice structures change both the magnitude and the decay time of this initial transient response seen in all three ErAs containing samples. Third, the $\Delta R/R$ trace for the sample with the 20 nm period in Fig. 2 contains an additional negative term.

The $\Delta R/R$ traces taken at several different wavelengths are fit with either a single-exponential function or the sum of two exponential terms to obtain the different time constants involved. The fits are started approximately 50 fs after the pump pulse has arrived at the sample. The time constant of the initial decay τ_1 shows little dependence on the wavelengths used and also on ErAs depositions between 0.6 and 2.3 ML. However, it does depend strongly on the period of the superlattice. Figure 3 shows this dependence of the time constant τ_1 on the superlattice period L. The data are from
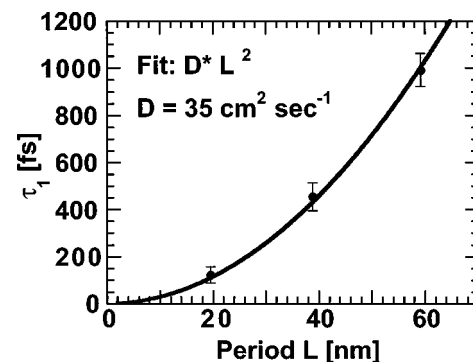

FIG. 3. The time constant τ_1 of the initial transient decay as a function of the superlattice period L.

the three samples described before. The error bars indicate the variation of τ_1 with wavelength. It can be seen that τ_1 increases in a superlinear fashion with L. A fit with a quadratic function is shown in Fig. 3.

The GaAs response is well understood and results from several processes. During the first ps the carrier distribution may be nonthermal. Afterwards the distribution is thermal and the signal is dominated by cooling of hot carriers to the band edges and recombination.[14] We believe that the curves for the ErAs-containing samples show some of these characteristics initially. After a few hundred fs, however, the carriers created in the GaAs matrix are captured by the ErAs islands. We believe this to be the origin of the initial transient decay and interpret τ_1 as the capture time.

The monotonic increase of τ_1 with increasing superlattice period L seen in Fig. 3 can be expected for two reasons. First, the number of capture sites per unit length N is decreasing with increasing superlattice period L. According to SRH theory, we expect $\tau_1 \propto 1/N \propto L$. Second, as the superlattice period L becomes longer, the carriers will need more time to travel to the ErAs islands. These two possible explanations are different because SRH theory assumes that the occupation of the capture sites is the limiting factor while the second explanation assumes that the travel time is the limiting factor. The superlinear dependence of τ on L seems to indicate that the travel time is important. The quadratic fit in Fig. 3 would be consistent with a diffusion process with diffusion constant $D = 35\ \mathrm{cm^2\,s^{-1}}$, which is feasible for carriers in GaAs. However, a diffusion picture is too simple considering the short time scale and that electrons and holes are involved in the process. A more accurate description will have to include several refinements: First, one type of carrier is probably captured first, creating an attractive potential for the oppositely charged carrier. Second, the subpicosecond time scale implies that at least the electrons will not experience many scattering events before they are captured. It is interesting to note that Gupta and co-workers[3,4] find that also for GaAs doped with Er the response time τ is not inversely proportional to the doping concentration N_{Er} as expected from SRH theory but rather $\tau \propto 1/\sqrt{N_{\mathrm{Er}}}$.

In summary, we have demonstrated the MBE growth of superlattices of self-assembled ErAs islands and GaAs. It is evident from x-ray diffraction that the superlattice periods are well defined. Pump–probe experiments show that the response times of these samples are in the subpicosecond range and controlled by the superlattice period. We attribute this to carrier capture by the ErAs islands. This should allow engineering of the response time of the material, which is very desirable for device applications such as THz sources or high-speed optoelectronic circuits.

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