MBE growth of AlGaAs/Ge/AlGaAs core-shell nanowire

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Abstract. Heterostructured AlGaAs/Ge/AlGaAs core-multishell nanowires having hexagonal crystal structure were synthesized by molecular beam epitaxy. Formation of 2-3 nm Ge quantum well structure was demonstrated. Raman characterization revealed a 200 cm⁻¹ peak corresponded to hexagonal phases of germanium.

1. Introduction

While germanium (Ge) based heterostructures are one of the most successful for creating sensitive near-infrared detectors [1], their implementation in active photonic devices remains one of the greatest challenges due to the indirect bandgap of Ge. To date, various strategies to facilitate the emissivity ability of Ge have been proposed, including synthesis of SiGe quantum dots [2] and tensile strained layer [3], as well as Ge/GaAs heterostructures [4].

At the same time, Ge-based nanostructures synthesized in the hexagonal phase can demonstrate sufficiently effective emissivity due to the modified direct bandgap [5]. Unfortunately, the methods commonly used typically result in the formation of hexagonal Ge clusters in amorphous matrices. [6-9]. In turn, alternative approach based on the growth of radial layers on the surfaces of wurtzite A₃B₅ nanowires (NWs) was proposed [10]. Furthermore, light emission in the range 1.8-3.5 µm of was achieved in hexagonal Si₁₋ₓGe, bulk layers has been demonstrated recently [11]. On the other hand, the growth of Ge hexagonal quantum wells of 1-10 nm thick can be used to tune the emission range up to 1 µm.

Here we report on the growth of hexagonal AlGaAs/Ge/AlGaAs core-multishell nanowires by molecular-beam epitaxy (MBE).

2. Experiment

Heterostructured NWs were grown step-by-step in the same growth session using Riber Compact EB200 MBE set up equipped with A₃B₅ sources, as well as Ge, Si electron-beam evaporators.

The synthesis of Al₀.₃Ga₀.₇As-core NWs was carried out via "vapor-liquid-solid" mechanism using gold as a catalyst at a substrate temperature of 500°C. Ga and Al deposition rate was equal to 1.7 Å/s and 0.56 Å/s, respectively. It should be noted that AlGaAs NWs grown in such way applying the same growth recipes were used as references in all subsequent experiments. After the NW formation, a technological pause to pump out the residual As species was taken (until the pressure in the growth chamber reached 10⁻⁹ Torr). Afterwards, the deposition of Ge at a rate of 0.2 Å/s and temperature of 320°C was performed. The growth temperature chosen typically leads to smooth radial Ge layers.
forming, as previously shown [12]. After the Ge deposition, the growth of AlGaAs cap layer was conducted. At the end of the synthesis, the samples were cooled to room temperature and unloaded from the growth chamber.

The samples obtained were studied by scanning electron microscopy (SEM) using a Zeiss Supra 25 microscope. Structural analysis of NWs was performed by transmission electron microscopy (TEM) using a Jeol JEM-2100F microscope equipped with an EDX spectroscope (Bruker QUANTAX XFlash 6/60). For TEM analysis NWs were mechanically separated from the substrates and transferred onto a carbon-film-coated copper mesh. Raman spectra were acquired using a Witec Alpha 300R microscope. A 532 nm Nd:YAG laser was used for excitation.

3. Results

SEM images of samples obtained are shown in figure 1. As it can be seen, deposition of Ge and AlGaAs layers at the same temperature of 320°C resulted in the formation of NWs with flag-like terminal (figure 1(a)). The average length of such NWs was 250 nm, and the diameter was about 50 nm. At the same time, the flag had a length of about 400 nm and diameter, which gradually decreased from 40 to 10 nm at the tip.

![Figure 1. SEM images of the obtained samples: (a) – growth temperature 320°C of AlGaAs cap layer; (b) – growth temperature 500°C of AlGaAs cap layer.](image)

The formation of flags was apparently caused by a migration of a catalyst drop onto the NW side facet during the growth of shells. It should be noted that deposition of Ge at temperatures less than 360°C typically leads to the formation of straight NWs, as it was shown previously [12]. Thus, the droplet migration was occurred in the process of AlGaAs cap layer growth. In this regard, the temperature of cap layer growth was varied in the range of 360-500°C. It was found that the ratio of vertical NWs in the arrays linearly magnifies with increasing temperature. In fact, arrays of straight NWs with a diameter of 70 nm and length of 2.5 μm were obtained at the AlGaAs growth temperature of about 500°C (figure 1(b)).

Combined with EDX map scanning TEM images of straight NWs are presented in figure 2. It is interesting that Au droplet was overgrown. Apparently, Ge droplet formed on the top of the NW could play the role of the catalyst (see figure 2). Unquestionably, it requires more detailed investigations.

TEM images of the NW oriented in the direction of [1100] are shown in figure 3. The formation of about 2 nm thick radial Ge layer was observed. High-resolution TEM images showed that the Ge layer and the covering AlGaAs layers exhibited hexagonal crystal structure of AlGaAs core. This fact was also confirmed by the microdiffraction pattern obtained from the near-surface region of the NW.
Figure 2. STEM (a) and EDX (b-e) images of a 500°C AlGaAs cap layer growth temperature sample.

Figure 3. TEM image of NW with a 500°C AlGaAs cap layer growth temperature.

The hexagonal structure of Ge was additionally confirmed by Raman spectroscopy. Several peaks belonging to Ge can be distinguished in the obtained spectra (figure 4). The peak in the range of 300 cm$^{-1}$ is associated with various Ge polytypes [13], while, 200 cm$^{-1}$ peak is attributed to hexagonal phases [13].

Figure 4. Raman spectra of a sample with a 320°C Ge growth temperature.

4. Conclusion
As a result, heterostructured AlGaAs/Ge/AlGaAs core-multishell NWs were synthesized by MBE. Thin Ge quantum well in AlGaAs barrier layers having radial geometry was demonstrated by TEM. A 200 cm$^{-1}$ peak corresponded to the hexagonal phase of germanium was detected on the Raman spectra. The possibility to growth flag-like NWs by tuning the AlGaAs cap layer growth temperature was shown.
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References
[1] Aliane A et al 2020 Semicond. Sci. Technol. 35 035013
[2] Wan J, Luo Y H, Jin G L, Jiang Z M, Liu J L, Liao X Z, Zou J and Wang K L 2001 Mat. Res. Soc. Symp. Proc. 667 G6.4.1-G6.4.6
[3] Gatti E et al 2014 J. Appl. Phys. 116 043518
[4] Aleshkin V Ya et al 2014 J. Appl. Phys. 115 043512
[5] Rödl C, Furthmüller J, Suckert J R, Armuzza V, Bechstedt F and Botti S 2019 Phys. Rev. Mater. 3 034602
[6] Parsons J R and Hoelke C W 1985 Philosophical Magazine A 50 (3) 329-337
[7] Zhang Y, Iqbal Z, Vijayalakshmi S, Qadri S and Grebel H 2000 Solid State Commun 115 657-660
[8] Vincent L, Patriarche G, Hallais G, Renard C, Gardes C, Troadee D and Bouchier D 2014 Nano Lett. 14 (8) 4828-4836
[9] Okugawa M, Nakamura R, Ishimaru M, Yasuda H and Numakura H 2016 J. Appl. Phys. 120 134308
[10] Hauge H I T et al 2015 Nano Lett. 15 (9) 5855-5860
[11] Fadaly E M T et al 2020 Nature 580 205-209
[12] Ilkiv I V, Kotlyar K P, Kirilenko D A, Osipov A V, Soshnikov I P, Terpitsky A N and Cirlin G E 2021 Semiconductors 55 (8) 621-624
[13] Fasolato C et al 2018 Nano Lett. 18 (11) 7075-7084