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Structural, electric, and optical properties of MgGa₂Se₄ epilayers grown by hot wall epitaxy method

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ABSTRACT

The epilayer growth of the MgGa₂Se₄ compounds was successfully achieved through the hot wall epitaxy method. The grown layer was accumulated along the <116 > direction onto the GaAs(100) substrate. From the Hall effect measurement, the mobility was determined to be 264 cm²/Vs at 293 K. At a high temperature range (T > 150 K), it tended to decrease as a function of $T^{-3/2}$ by increasing the temperature and increase as a function of $T^{3/2}$ at the low-temperature range (T < 100 K). In the photocurrent (PC) measurement, we observed the A, B, and C peaks corresponding to 529.9 (2.3398 eV), 495.2 (2.5037 eV), and 477.6 nm (2.5960 eV) at 10 K, respectively. Three peaks of A, B, and C were caused by the band-to-band transitions from the valence band state of $\Gamma_4(z)$, $\Gamma_5(x)$, and $\Gamma_5(y)$ to the conduction band state of $\Gamma_1(s)$, respectively. By comparing the results of PC and absorption, the temperature dependence of the optical bandgap energy was well interpreted using Varshni's relation $E_r(T)=2.3412-8.87 \times 10^{-4} T^2/(T+251)$.

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1. Introduction

The $A^{II}\text{--}B^{III}_2\text{--}C^{VI}_4$ materials are attractive materials because of their applicability for solar cells, light emitting diodes, and photoconductive devices [1-3]. Magnesium gallium selenide (MgGa₂Se₄) is a new material satisfying such a requirement. Thus, in order to realize these applications, it is of primary importance to grow high quality epilayers and characterize the fundamental properties. But, it has been reported that the bulk of MgGa₂Se₄ crystals grew through the Bridgman method, and its characterization investigated through absorption and photoluminescence [4,5]. Therefore, the information on MgGa₂Se₄ is still very limited. MgGa₂Se₄ is a photoconductive material having the bandgap energy of 2.2 eV at room temperature. Therefore, it is important to investigate the conductivity change of photoconductive MgGa₂Se₄ caused by incident radiation. Thus, the photocurrent (PC) spectroscopic measurement had been studied for applications in photodetection and radiation measurements [6]. But, the PC-measurement analysis of MgGa₂Se₄ has not yet been reported. In PC measurements, absorbed photons with higher energy than the bandgap energy create electron and hole carriers in the conduction and valence bands, respectively. These carriers instantly flow out through the electrodes. Thereby, the obtained PC peak corresponds to the direct bandgap energy.

In this study, photoconductive MgGa₂Se₄ epilayers were first grown by the hot wall epitaxy (HWE) method. The grown MgGa₂Se₄ layers were investigated by high-resolution X-ray diffraction (HRXRD), atomic force microscopy (AFM), Hall effect, PC, and absorption spectroscopy. From these results, we discussed the structural, electric, and optical properties of the MgGa₂Se₄ epilayers. In addition, the bandgap energy obtained from the PC peak position was discussed as a function of temperature together with that of the absorption experiment.

2. Experimental procedures

Prior to the MgGa₂Se₄ layer, MgGa₂Se₄ polycrystalline was formed. The starting materials were 6 N purity shot-types of Mg, Ga, and Se. After the materials were weighed in stoichiometric proportions, they were sealed in a quartz tube for maintaining the vacuum atmosphere. At this time, the horizontal synthesis furnace was used for the formation of the MgGa₂Se₄ polycrystalline. The sealed ampoule was placed in the synthesis furnace and was continually rotated at a rate of 1 rpm. In order to avoid an explosion of the ampoule due to the selenium vapor pressure, the temperature of the ampoule was increased gradually to $1000 \,^{\circ}$ C, which was then maintained for 48 h. Fig. 1 presents a schematic diagram of the HWE apparatus used for the MgGa₂Se₄ layer. The MgGa₂Se₄ layers were grown on semi-insulating GaAs (100). The GaAs substrate was cleaned ultrasonically for 1 min in successive baths of trichloroethylene, acetone, methanol, and

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Fig. 1. Schematic diagram of the HWE apparatus used for the MgGa₂Se₄ layer.

2-propanol and etched for 1 min in a solution of H₂SO₄:H₂O₂:H₂O (5:1:1). The substrate was degreased in organic solvents and rinsed with deionized water (18.2 M Ω). After the substrate was dried off, the substrate was immediately loaded onto the substrate holder in Fig. 1 and was annealed at 580 °C for 20 min to remove the residual oxide on the surface of the substrate. To find the optimum growth conditions, the grown MgGa₂Se₄ layers were analyzed by HRXRD measurements. The thickness and stoichiometric composition of the MgGa₂Se₄ layers were measured by using an α -step profilometer and an energy dispersive X-ray spectrometer (EDS), respectively. Also, the electric properties were achieved by Hall effect measurement of the van der Pauw method with various temperatures. In order to take PC measurements, two Au electrodes with a coplanar geometry on MgGa₂Se₄ were fabricated by an e-beam evaporator and an Ohmic contact of electrodes was confirmed by the current-voltage measurement. After the electrodes were connected to a wire, the sample was mounted on the holder of a low-temperature cryostat. The PC spectrum measurement was done while monochromatic light emitted from a halogen lamp passed through a chopper to illuminate the sample. Thus, the optical absorption measurement was performed with a UV-vis-NIR spectrophotometer (Hitachi, U-3501) for a range of 400 to 800 nm. At this time, the PC and absorption experiments were measured while varying the temperature from 10 to 293 K.

3. Results and discussion

3.1. Growth and structural property

In order to grow the MgGa₂Se₄ layer, an ingot of synthesized MgGa₂Se₄ polycrystalline was used as a source material of HWE. At this time, the source temperature was fixed at 610 °C, which was obtained through experimental repetition. Thus, the MgGa₂Se₄ layers were grown by changing the substrate temperature from 360 to 420 °C. Fig. 2 presents the HRXRD rocking curves on the MgGa₂Se₄ layers grown as a function of the substrate temperature. The observation of this HRXRD rocking curves indicate that the grown layers have a high crystalline quality. As shown in Fig. 2, the HRXRD intensity of the layer on the 400 °C substrate temperature had the highest than that of the measured other samples. Fig. 3 displays the intensity and FWHM value of



Fig. 2. HRXRD rocking curves on the $MgGa_2Se_4$ layers grown as a function of the substrate temperature. Here, the subfigure shows the XRD curves on the specimen grown at the substrate of 400 °C.



Fig. 3. Intensity and FWHM value of the HRXRD rocking curves on the MgGa₂Se₄ layers grown as a function of the substrate temperature.

the HRXRD rocking curves on the MgGa₂Se₄ layers grown as a function of the substrate temperature. As shown in Fig. 3, the intensity of the HRXRD curves increased with an increase in the substrate temperature. However, its intensity tended to decrease after an increase in the substrate temperature of 400 °C. On the contrary, the FWHM on the HRXRD curves decreased with an increasing substrate temperature. Then, it again increased after having a minimum value of 117 arcsec at 400 °C. These results indicate that the optimum temperature of the substrate is 400 °C. With a source temperature of 610 °C, the most suitable substrate temperature for growth turned out to be 400 °C. Thus, the MgGa₂Se₄ layers grown under these optimum conditions were obtained with a thickness of 2.8 µm and a growth rate of 1.39 Å/sec. On the other hand, to confirm the orientation of the MgGa₂Se₄ layer, XRD analysis was used. The subfigure in Fig. 2 shows the XRD curves on the specimen grown on the substrate of 400 °C. As shown in subfigure of Fig. 2, two dominant peaks were observed. These correspond to the diffraction peaks of the MgGa₂Se₄ (116) and the GaAs (400) plane. The intensity of the Download English Version:

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