The chalcopyrite compound Cu(In,Ga)Se₂ (CIGS) has the potential to be used as a semiconductor material in thin-film photovoltaic devices with high conversion efficiencies [1], and CIGS solar cells have recently shown a record conversion efficiency over 22% using CIGS layers with 1.1 eV band gap energy. According to the so-called Schockley-Queisor limit for the conversion efficiency, the optimum band gap energy should be around 1.4 eV. Therefore, the conversion efficiency can be improved further by adjusting the Ga content in CIGS. However, further increase in Ga content deteriorates the conversion efficiency owing to the problem caused by the CIGS/CdS heterostructure. In addition, most of CIGS solar cells have been based on polycrystalline films on glass substrates. Therefore, if high-quality single-crystal thin films are available, higher efficiency could be achieved. In this study, we try to fabricate single-crystal CIGS thin films on single crystalline substrates by a three-stage process [2].

CIGS layers were fabricated on p-type GaAs (001) and Ge (001) substrates by a MBE method. Native oxide layers on GaAs and Ge surfaces are removed by a thermal flash annealing at 600 °C. A 2.0-μm-thick CIGS layer was deposited using the three-stage process. The growth temperature of the first stage was 350 °C, and that of the second and third stages was 550 °C. The surface structures of the substrates and grown films were investigated by RHEED. After growth, the surface conditions were observed by SEM, and CIGS films were characterized by X-ray diffraction (XRD) with Cu-Kα radiation. The minority carrier lifetime was evaluated by time-resolved photoluminescence (TRPL) using a near-infrared fluorescence lifetime spectrometer (Hamamatsu Photonics C12132).

During the 1st stage of CIGS deposition, RHEED patterns showed 4-fold symmetry as with GaAs (001) substrates. The RHEED patterns had chevron patterns, indicating that {111} facets were formed on InGaSe surfaces. Figure 1 shows a SEM image of a CIGS layer on a GaAs (001) substrate. Uniform facets appeared on the surface, and the facets were identified to be {112}B of CIGS crystals. Only a (008) diffraction peak was confirmed from a CIGS layer on a GaAs (001) substrate by XRD 2θ/ω scan. Therefore, CIGS layers on GaAs (001) substrates had a single-crystalline structure. On the other hand, the CIGS layers on Ge (001) substrates had a twin structures owing to anti-phase domains. PL decay profiles for the CIGS layers on GaAs, Ge, and sapphire substrates with CdS layers are shown in Fig. 2. For a reference, we fabricated CIGS layers on sapphire substrates, and the CIGS layers had a polycrystalline structure. The lifetime of CIGS on GaAs and Ge substrates was longer than that of the polycrystalline CIGS layer. Therefore, single-crystalline CIGS layers should have the potential of high-conversion efficiency solar cells.

Fig. 1 SEM image of CIGS/GaAs(001).
Fig. 2. PL decay profiles for CIGS layers.

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