## Band structure of GaMnAs near the Fermi level studied by time-resolved light-induced reflectivity measurements

## <sup>°</sup>Tomoaki Ishii<sup>1</sup>, Tadashi Kawazoe<sup>1</sup>, Yusuke Hashimoto<sup>2</sup>, Hiroshi Terada<sup>1</sup>, Iriya Muneta<sup>1</sup>, Motoichi Ohtsu<sup>1</sup>, Masaaki Tanaka<sup>1</sup>, Shinobu Ohya<sup>1</sup> *Graduate School of Engineering, The Univ. of Tokyo<sup>1</sup>, Radboud Univ. Nijmegen<sup>2</sup>* E-mail: ishii@cryst.t.u-tokyo.ac.jp

There have been intense discussions over the Fermi level ( $E_F$ ) position in GaMnAs [1-4]. So far a number of studies have indicated that  $E_F$  exists in the impurity band (IB) in the band gap ( $E_g$ ) [3-4], while recent time-resolved light-induced reflectivity measurements have shown conflicting results indicating that  $E_F$  exists in the valence band (VB) [2]. This conflict may be due to a high fluence pump, giving a quite dense photo-excited carrier density  $4 \times 10^{19}$  cm<sup>-3</sup> and significant change in the band structure [5], and thus the definition of both  $E_g$  and  $E_F$  are obscure. In this study, we carefully carry out time-resolved reflectivity measurements with low fluence pump to suppress the accumulation of photo-carriers.

We grew 100-nm-thick Ga<sub>1-x</sub>Mn<sub>x</sub>As films with x=1%, 3%, and 6% after growing a 20-nm-thick GaAs buffer layer on semi-insulating (S.I.) GaAs (001) substrates by low-temperature molecular beam epitaxy. The Ga<sub>0.94</sub>Mn<sub>0.06</sub>As film was annealed at 240°C for 89 h. The Curie temperature of the Ga<sub>0.99</sub>Mn<sub>0.01</sub>As, Ga<sub>0.97</sub>Mn<sub>0.03</sub>As, and Ga<sub>0.94</sub>Mn<sub>0.06</sub>As films is 13 K, 38 K, and 120 K, respectively. Time-resolved reflectivity measurements were performed by using pump-probe procedure and a pulsed light source with time-duration of 3 ps and a reputation rate of 80 MHz. At time *t* after the irradiation of the pump-pulse with the fluence of 0.16  $\mu$ J/cm<sup>2</sup>, the low-power probe-pulse with 1 nJ/cm<sup>2</sup> and the same photon energy as that of the pump light was irradiated to the sample, probing the reflectivity change  $\Delta R/R$ . Photo-excited carrier density is estimated to be below  $3 \times 10^{17}$  cm<sup>-3</sup> by assuming that the pump-pulse light is absorbed only in the GaMnAs layer. This is two orders magnitude smaller than the carrier density for the previous study [2]. We used the pico-second pulses to achieve the high energy resolution of ~1 meV.

Figure 1(a) shows  $\Delta R/R$  measured at 5 K for the S.I. GaAs substrate and for the Ga<sub>1-x</sub>Mn<sub>x</sub>As films at t=166 ps. Generally, as shown in Fig. 1(b), positive $\Delta R/R$  peaks are observed above  $E_g$  due to band filling (BF) and near  $E_g$  due to band-gap renormalization (BGR). Figure 1(c) shows possible photo-induced electron transitions in GaMnAs. In Fig. 1(a), a clear peak is observed at ~1.51 eV (pointed by the green arrow) for all the samples. This can be attributed to BGR and thus assigned to  $E_g$ . In the data for the GaMnAs films, we see clear structures in the region having lower energy than  $E_g$ , which means that electrons (and thus  $E_F$ ) exist in the band gap (Fig. 1(c)). The peaks pointed by red arrows in Fig. 1(a) are attributed to BGR due to photo-carriers excited from  $E_F$  to CB. Also, the positive peak observed between the red and green arrows when x=3% and 6% is attributed to BF induced by electrons excited from IB to CB. The BGR peaks pointed by the red arrow shifts to the lower energy with increasing x from 1%, which is consistent with the previous resonant tunneling experiments indicating that  $E_F$  moves away from VB with increasing x from 1% [4]. The energy difference from  $E_F$  to the top of VB in Ga<sub>0.94</sub>Mn<sub>0.06</sub>As in our study is ~40 meV smaller than that in the previous report [3]. This may imply that there is still a small

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Fig. 1 (a) Energy dependence of  $\Delta R/R$  measured at 5 K for the S.I. GaAs substrate (red) and the Ga<sub>1-x</sub>Mn<sub>x</sub>As films with x=1% (brown), 3% (green), and 6% (blue) at t=166 ps. The red and green arrows are the positive  $\Delta R$  peaks caused by BGR induced by the electrons excited from  $E_F$  to CB and from VB to CB, respectively. (b) Schematic  $\Delta R/R$  components due to BF (red curve) and BGR (blue curve) [5]. If the peak of the positive  $\Delta R/R$  component due to BGR is larger than the peak of the negative  $\Delta R/R$  component due to BGR can be observed. (c) Photo-induced electron transitions from  $E_F$  to the CB (red arrow) and from VB to CB (green arrow) in GaMnAs.