CuInSe2 薄膜でのスピン緩和の観測

Observation of exciton spin relaxation in CuInSe₂ thin films

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CuInSe₂ (CIS) and related materials are promising candidates for low cost, high efficiency photo-absorber layers for solar cells.¹ These materials can be also applied to a large variety of electro-optic devices.¹ There are attempts to use carrier spin as a new degree of freedom in order to apply for spin dependent devices. However, the spin relaxation in these materials has not yet been investigated. In this study, we report the spin relaxation of CIS thin films obtained by spin dependent time-resolved pump and probe measurement.

A 1 µm thick-CIS layer was grown on (001) substrate by employing the GaAs deposition sequence of migration enhanced epitaxy using molecular beam epitaxy system.² In the spin dependent time-resolved pump and probe measurement, a femtoseconds Ti-sapphire laser with an optical parametric oscillator was used as an optical source. The pump beam was chopped by an electro-optic modulator at 1.9 MHz to avoid optical noise in the low-frequency region. After right circularly polarized photoexcitation, the population of carriers with a down (up) spin along the direction of light propagation was probed using a right (left) circularly polarized probe pulse. The population change of the spin polarized carriers is measured through the change of intensity of the reflected probe light. The excitation laser wavelength was tuned to 1192 nm at 300 K. In this experimental setup, the time resolution was 200 fs. The excitation power intensities were 30 mW.

The time evolutions of spin-dependent reflection intensity at 300 K for 30 mW is shown in Fig. 1, where I_+ indicates a right circularly polarized excitation with a right circularly polarized probe, while I_- indicates a right circularly polarized excitation with a left circularly polarized probe. The difference between I_+ and I_- , corresponding to the spin polarization, is clearly observed. The time evolution of spin polarization ($I_+ - I_-$)/($I_+ + I_-$) is

shown in Fig. 2. The spin relaxation time, which is twice the relaxation times of the spin polarization, is evaluated to be 339 ps at 300 K by single exponential fitting. This is the first observation of the exciton spin relaxation in CIS, to our knowledge. This result opens up the possibility for CIS to be applied as a spin dependent device.



Fig.1 Time evolutions of reflection intensity at 300 K.



Fig.2 Time evolution of spin relaxation at 300 K. ¹ K. Puech et al., Appl. Phys. Lett. **69**, 22 (1996). ² S. Thiru et al., J. Crys Growth. **425**, 203 (2015).