Infrared Magneto-Optical Imaging on Phase Separation of Solids

In first-order transitions such as the magnetic transition in magnetic materials and the Mott transition in strongly correlated electron systems, physical properties drastically change. Around the transition point, the phase separation in which both phases coexist sometimes appears. Anomalous physical properties often originate from the phase separation, for instance, the colossal magnetoresistance of Manganese oxides [1] and the magnetic-field-induced superconductor-insulator transition of organic superconductors [2] are believed to originate from the phase separation. The domain size of the phase separation is typically on the nanometer scale. Owing to the lattice distortion or trapped magnetic polaron, however, micrometer-sized domains sometimes appear. Such large domains and the electronic structure of each domain can be identified by infrared imaging. The target material dealt with in this study, namely, electron-doped europium monoxide (EuO), has a large magnetoresistance [3], whose origin is still debated to be trapped magnetic polarons. Therefore, the detecting of the large domain size as well as the inhomogeneity of the sample surface near the ordering temperature/magnetic field is important for studying the magnetic polaron scenario.

EuO is a ferromagnetic semiconductor with a Curie temperature (T_C) of around 70 K. With excess Eu electron doping or the substitution of Gd^{3+} or La^{3+} for Eu^{2+} ions, T_C increases up to 150 K and electrical resistivity drops by twelve orders of magnitude below T_C. Since the magnetic moment originates from local Eu^{2+} $4f^7$ electrons, electron-doped EuO has larger magnetic moments than colossal magnetoresistance manganites, which exhibit a similar insulator-to-metal transition at T_C. Therefore, electron-doped EuO is attracting attention as a next generation functional material for spintronics devices.

Infrared reflectivity spectra [$R(\omega)$] and spatial images with unpolarized light were obtained at the infrared magneto-optical station of beamline **BL43IR** [4]. Infrared $R(\omega)$ imaging was performed in the 6000 to 12000 cm^{-1} wave number range at 10 cm^{-1} resolution at different temperatures from 40 to 80 K in magnetic fields of up to 5 T. To acquire spatial imaging data, a total of 1681 spectra were obtained in a 200 × 200 μm^2 region in steps of 5 μm with a spatial

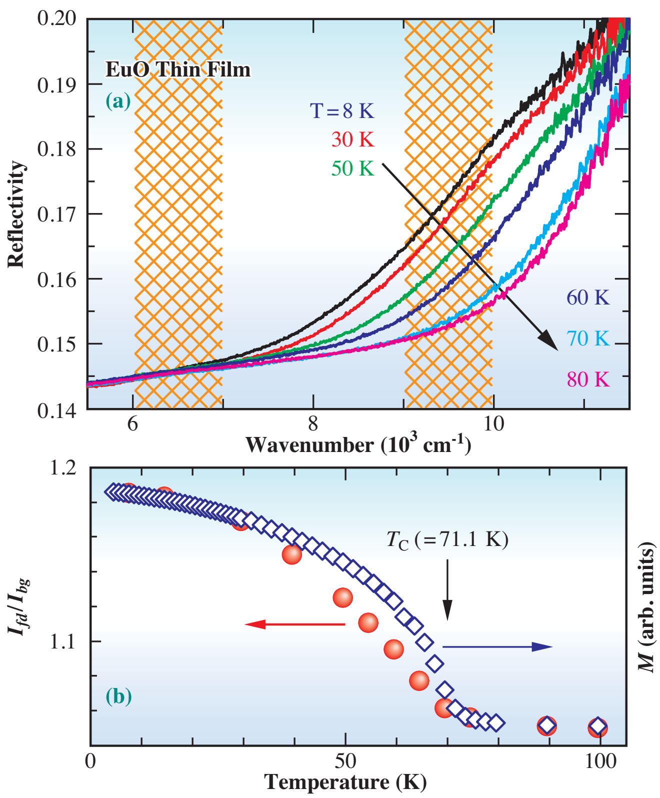

Fig. 1. **(a)** Temperature dependence of the reflectivity spectrum $R(\omega)$ of a EuO thin film in the wave number range of 4000 – 12000 cm^{-1}. **(b)** Temperature dependence of the intensity ratio between the absorption edge of the exciton of the Eu $4f \rightarrow 5d$ transition (I_{fd}) integrated at 9000 – 10000 cm^{-1} and the background intensity (I_{bg}) at 6000 – 7000 cm^{-1}. The temperature dependence of magnetization (open diamonds) is also plotted.

resolution better than 5 μm. The spatial images and the temperature dependence of the reflectivity spectrum were plotted using the intensity ratio of the absorption edge of the exciton of the Eu $4f \rightarrow 5d$ transition (I_{fd}) integrated over 9000 – 10000 cm^{-1} to the background intensity (I_{bg}) in the 6000 – 7000 cm^{-1} range, as shown in Fig. 1.

To investigate the phase separation in the magnetic-field-induced paramagnetic-to-ferromagnetic transition, we performed infrared imaging under magnetic fields at 80 K, which that is slightly higher than T_C, as shown in Fig. 2(a). The figure indicates that the paramagnetic state at $B = 0$ T becomes ferromagnetic as magnetic field increases. A similar magnetic-field-induced insulator-to-metal transition has been observed in manganites in spite of its different origin. The spatial distribution is plotted in Fig. 2(b). The figure shows that the distribution width normalized by that at $B = 0$ T exhibits an approximately twofold increase at 3 T and then decreases at 5 T. Such an anomalous temperature-dependent width can be explained in terms of the magnetic polaron scenario, in which a large magnetic polaron is generated by applying a magnetic field. In other words, the phase transition from a uniform paramagnetic state to a uniform ferromagnetic state via the inhomogeneous magnetic polaron state appears with increasing magnetic field at a temperature slightly higher than T_C.

We use these results as a basis for discussing the domain size of the magnetic polaron state in EuO. The spatial resolution of about 5 μm used in this work is larger than this domain size. However, changes in the spatial distribution width can be detected. In such cases, the domain size is about one-tenth of the spatial resolution of the used microscope, i.e., about several 100 nm. This is the first direct observation of the inhomogeneity due to magnetic polaron domains [5].

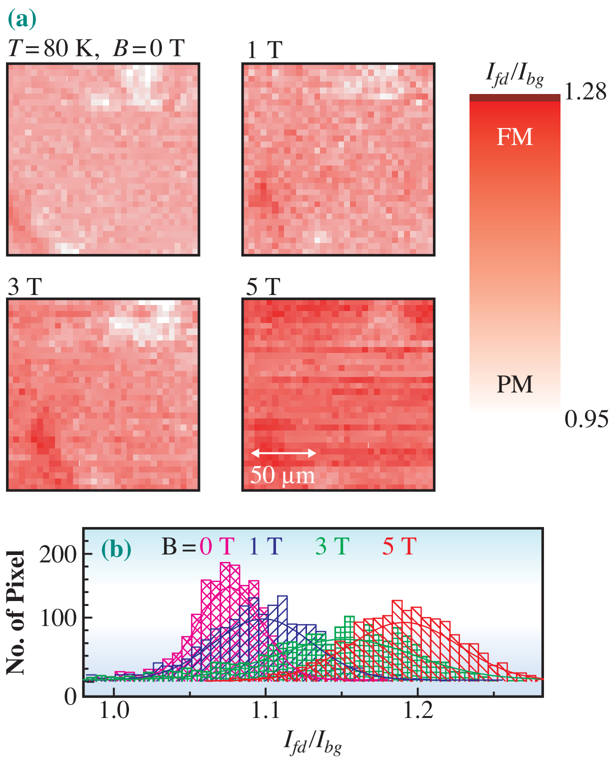

Fig. 2. **(a)** Magnetic field dependence of I_{fd}/I_{bg} of a EuO thin film at $T = 80$ K that is slightly higher temperature than T_C. **(b)** Statistical distributions and Gaussian fittings of I_{fd}/I_{bg} derived from **(a)**.

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References

[1] L. Zhang *et al.*: Science **298** (2002) 805.
[2] T. Nishi *et al.*: Phys. Rev. B **75** (2007) 014525.
[3] A. Mauger and C. Godart: Phys. Rep. **141** (1986) 51.
[4] S. Kimura *et al.*: Nucl. Instrum. Meth. A **467-468** (2001) 893-896.
[5] *S. Kimura, T. Ito, H. Miyazaki, T. Mizuno, T. Iizuka and T. Takahashi: Phys. Rev. B* ***78*** *(2008) 052409.*