Infrared Spectroscopy under Extreme Conditions

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Abstract

We constructed a magneto-optical microspectroscopy apparatus in the infrared region using a synchrotron radiation, SPring-8. In the apparatus, an infrared microscope with the spatial resolution of $\sim$ 11 μm is combined with low temperatures ($\geq$ 3.5 K) and high magnetic fields ($\leq$ 14 T). The purpose is to investigate the electronic structure under extreme conditions of tiny materials such as organic conductors and of small region and the spatial distribution of electronic structures. After the installation of high pressure cells, optical measurements under multiple-extreme conditions is available. The specification of the apparatus and recent results of an organic superconductor are presented.

Key words: infrared spectroscopy; extremely conditions; synchrotron radiation; κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

Materials show their various properties under several extreme conditions. These properties are very different from that in ordinary conditions. Such extreme conditions can be produced by using superconducting magnets, low temperature cryostats, high pressure cells and so on. Because of the use of such special apparatuses, the space under extreme conditions is very narrow. Then fundamental experiments as transport and magnetic susceptibility have been only done. Recently, de Haas van Alphen and NMR experiments are done but the optical experiments in which individual electronic excitations can be observed have been done so much. Particularly, infrared (IR) spectroscopy that reflects electronic states near the Fermi level is unable to be performed under extreme conditions because of the low emittance of ordinary light sources such as a black body (BB) radiation. Then we constructed an apparatus for the IR spectroscopy under extreme conditions using high brilliant synchrotron radiation. In this paper, the present performance of the apparatus and recent obtained results are presented.

The apparatus was constructed at the IR beam line of SPring-8 that was dedicated in 2000. [1] The brilliance of the IR synchrotron radiation (IR-SR) from SPring-8 is 10^4 times higher than an ordinary BB light. [2] Actually, the focus size of the IR-SR is 10 times smaller and the light density is 10^2 times higher than BB light at the focal point in an infrared microscope. This means that the optical spectrum in a tiny region can be measured in the high contrast condition. We constructed the IR magneto-optical imaging apparatus in which an IR microscope is combined with a superconducting magnet depicted in Fig. 1. [3] The feature is to be able to measure reflectivity spectra and the magnetic circular dichroism [4] at 14 T at 3.5 K with the spatial resolution of 11 μm in diameter in the photon energy range of 0.1$\sim$2 eV. Using the apparatus, the spatial distribution of the reflectivity spectra as well as the electronic structure and the lattice vibration can be automatically measured and

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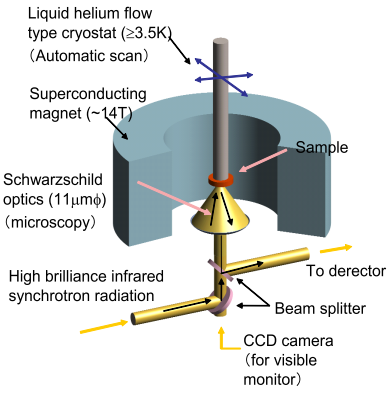

Fig. 1. Infrared magneto-optical imaging apparatus using a synchrotron radiation SPring-8 in which a superconducting magnet (maximum magnetic field is 14 T) and a sample cryostat (minimum temperature is 3.5 K) combined with an infrared microscope (focal size at the sample is 11 μm in diameter).

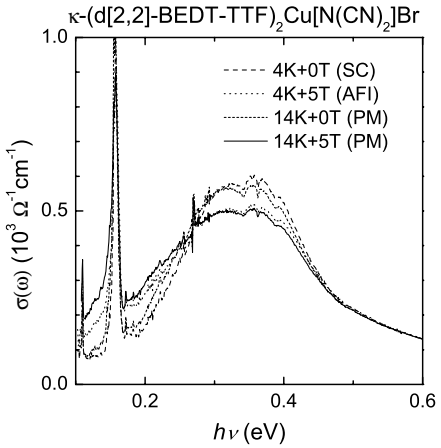

Fig. 2. Temperature and magnetic field dependences of the optical conductivity ($\sigma(\omega)$) spectrum of κ-(d[2,2]-BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. These spectra at 4 K and 0 T, at 4 K and 5 T and at 14 K and 0 T, 5 T belong to the superconductor (SC), the antiferromagnetic insulator (AFI) and the paramagnetic metal (PM) phases, respectively.

be imaged. The spatial resolution of 11 μm is much smaller than the high pressure space using a diamond anvil high pressure cell that has been installed in this apparatus recently. Then, if the spatial distribution of pressure is used, the pressure dependence of electronic structure can be investigated.

The measurement was performed to an organic superconductor, κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (T_C =11 K). The character changes from superconductor (SC) to antiferromagnetic insulator (AFI) across the quantum critical point of the Mott transition by the partially deuterated BEDT-TTF molecule. [5] Particularly, the cases of two sites substitution (d[2, 2]) and three sites substitution (d[3, 3]) in four is considered to be very near the quantum critical point. In d[2, 2], it was observed that the electric resistivity drastically jumps from SC to AFI by applying a magnetic field. [6] Since the sample size of such organic conductor is normally several 0.1 mm, it is difficult to measure the magneto-optical spectra at high magnetic fields. Then the apparatus reported here is suitable for the investigation of the change of the electronic structures from the SC to the AFI states under magnetic fields.

The magnetic field dependence of the optical conductivity ($\sigma(\omega)$) spectrum obtained from the Kramers-Kronig analysis of the reflectivity spectrum at 4 K is shown Fig. 2. In spite that the character of the material is SC at 4 K and 0 T and AFI at 4 K and 5 T, the $\sigma(\omega)$ spectrum does not change so much. This indicates that the electronic structure above 0.1 eV from the Fermi level does not change due to the field-induced SC - AFI transition. On the other hand, with increasing temperature, the state at 0 T first-ordered changes from a SC to a paramagnetic metal (PM) at 11 K with increasing temperature and it at high magnetic fields gradually changes from the AFI to the PM phases. The temperature dependence of the $\sigma(\omega)$ spectrum as well as the electronic structure is much larger than the field dependence as shown in Fig. 2. This indicates that the SC - PM transition at 0 T is a first order one and the AFI state at high field continuously connects to the PM state in the view of the electronic structure. The detail will appear in a separated paper.

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