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Photoemission Studies of The Layered Oxysulfide (CeO)CuS

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The layered oxysulfide (LaO)CuS is a wide gap *p*-type semiconductor, whose crystal structure belongs to tetragonal (LaO)AgS type structure (P4/nmm). Recently, we have succeeded in synthesis of (CeO)CuS with an iso-structure of (LaO)CuS. The new rare earth oxysulfide has strong blue luminescence band which is different from that of (LaO)CuS, and relatively low electrical resistivity. For this reason, this compound is expected to be a new optical material for LEDs. In this study, we have performed soft X-ray photoemission spectroscopy (PES) of (CeO)CuS to investigate the origin of blue luminescence.

The sample used in this study was a polycrystal synthesized by solid-state reaction. The starting materials were CeO_2, Ce_2S_3, CuO, and Cu powders. The appropriate amount of starting materials was mixed in Ar atmosphere and their mixture was pressed into bar shape and then, it was sintered at 900 degree C for 40 hours. Resonant PES and X-ray absorption spectroscopy of (CeO)CuS at the Ce 3d – 4f absorption edge were carried out on the BL25SU line at SPring 8. Clean surfaces were in situ obtained by scraping with a diamond file in the sample preparation chamber below 1 x 10^{-10} Torr.

The valence band of (CeO)CuS is expected to consist of Cu 3d –S 3p anti-bonding state, well localized Cu 3d non-bonding state, and Cu 3d – S 3p bonding state mainly, as well as those of (LaO)CuS. Figure 1 shows the valence-band PES spectra of (CeO)CuS at several excitation photon energies (*hv*). With increasing from *hv*=870 to 881 eV, a clear resonant peak is gradually enhanced just below the valence-band maximum (VBM) and at *hv* = 882.5 eV, the intensity becomes maximum. These results indicate that the Ce 4f orbital also constitutes the valence band of (CeO)CuS in addition to Cu 3d and S 3p orbitals. The new blue luminescence band may attribute to the Ce 4f orbital in the VBM.

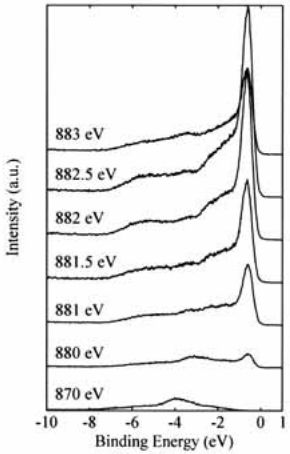

Figure 1. Valence-band PES spectra of (CeO)CuS in the Ce 3*d*-4*f* excitation region.

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Magnetization analysis of metallofullerenes by means of soft x-ray MCD

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Rare earth metallofullerenes are one of the promising candidates for ferromagnetic nano-particles applied for magnetic devices. A ferromagnetic metallofullerene particle is regarded as the smallest ferromagnetic nano-ball that helps realizing ultimate high-density storages. However, the desired ferromagnetic property has not been found in mono-metallofullerenes, yet.

Since rare earth mono-metallofullerenes have been characterized as the diamagnetic or paramagnetic, the di- or tri-metallofullerenes (multi-metallofullerenes) have attracted us as the research targets to discover a novel ferromagnetic metallofullerene. At present, the multi-metallofullerenes are found as the minor constituent of a synthesized material by means of a He arc-furnace. The chemical yield of metallofullerenes often becomes lower and lower with increasing the number of involved atoms in a cage. As the result, it takes more than few months in order to prepare the purified multi-metallofullerene sample for the conventional magnetization measurement.

Soft x-ray magnetic circular dichroism (SXMCD) is undeniably sensitive to a surface magnetization, therefore, is very suitable to investigate the magnetic properties of small quantities of metallofullerenes embedded on non-magnetic substrate. It is also great advantage of SXMCD technique that magnetization of involved rare earth ions is detected separately from a diamagnetic cage according to an element specificity of SXMCD.

In the present experiment, SXMCD measurements in rare earth metallofullerenes have been performed at BL25SU using an electromagnet type SXMCD spectrometer. Metallofullerene samples, $R_2@C_{84}$ (R=Er, Dy) were coated on the Cu sample plate, then, were baked at ~400K in the load-lock chamber under the pressure of 1×10^{-5} Pa. Sample was cooled down to 20K using a Liq.He continuous flow-type cryostat. SXMCD spectra and element specific magnetization curves were collected using the helicity switching technique of SXMCD measurements.

Some exciting results associating with the effective magnetic moment of rare earth ions are obtained, though no evidence of ferromagnetic properties has been found in measured metallofullerene samples. Nadaï *et al.* has reported on R magnetic moments of $R@C_{82}$ (R=Gd, Dy and Ho) by means of SXMCD measurements, where the magnetic moments are substantially reduced to almost the half of the expected values for free R ions [1]. In our results, some additional new properties of magnetic moment of rare earth metallofullerene were possibly elucidated, though analysis of MCD spectra has not been completed. Slight change in SXMCD spectral shapes depending on the number of atoms in a cage is found associating with the multiplet structure of the 4*f* final state. The relationship between the spectral shapes and a molecular motion of fullerene ball is worth to pay attention in the analysis.

[1] C.De. Nadaï *et al.*, Phys. Rev. B **69** (2004) 184421.