Resonant X-ray scattering in $KCuF_3$

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Abstract

We study the resonant x-ray scattering (RXS) spectra around the Cu K edge in $KCuF_3$ on the basis of the band structure calculation combined with the LDA+U approximation. The lattice distortion is introduced as inputs of the calculation. We reproduce well the experimental spectra as a function of photon energy around the main–K–edge (MKE). The spectral structure around the MKE is independent of the magnetic order (MO) and the orbital order (OO) but strongly dependent on the magnitude of the Jahn–Teller distortion (JTD).

Key words: Resonant X-ray scattering ; Jahn–Teller distortion ; orbital order ; $KCuF_3$

1. Introduction

Recently, RXS spectra have been observed on the orbital and magnetic superlattice spots around the Cu K–edge in $KCuF_3$[1,2]. The intensities on the orbital superlattice spots have been directly connected to the OO. This interpretation does not seem compatible to the recent analyses [3–5] on the $LaMnO_3$, because $KCuF_3$ is known to be a typical material with a substantial magnitude of the JTD.

For the K edge in transition-metal compounds, RXS intensities arise from the modulation of $4p$ states in the intermediate states of the resonant process. The fact that $4p$ states are not the states of orbital ordering causes complications on the interpretation of RXS intensities. Two types of the mechanism for the modulation of the $4p$ states have been proposed. One relates the modulation to the $3d$ orbital polarization[6], while the other relates it to the lattice distortion[3–5].

Several *ab-initio* calculations[3–5] have revealed that $4p$ states of transition metal ion are so extending in space that they are considerably modified by neighboring electronic states through the JTD. It has been concluded that in $LaMnO_3$ the effect of the JTD on the RXS spectra is much larger than that of the OO.

The purpose of this paper is to elucidate the mechanism of the RXS spectra on the orbital superlattice spots through *ab-initio* calculations. We use the full-potential linear augmented plane wave (FLAPW) method, combined with the LDA+U in order to take account of the both effects of the JTD and the OO. We carry out calculations of the spectra in various magnetic phases and with changing the magnitude of the JTD.

2. Calculation

We assume the lattice constant determined by the experiment as inputs for the band calculation. The schematic crystal structure is shown in fig.1. For changing the magnitude of the JTD, we change $q = (l - s)/(l + s)$, fixing the lattice constants, the bond length m, and $l + s$ at the experimental values. The experimental value of q is 0.088. The local $3d$ Coulomb interaction is assumed as $U = 3$ eV and $J = 1$ eV.

Assuming the antiferromagnetic (AFM) ground state, which has been revealed by the experiment, we obtained the insulating ground state with the band

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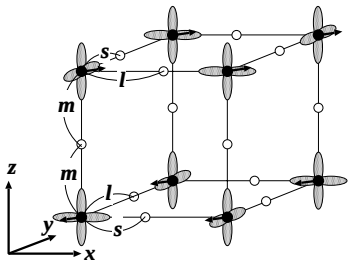

Fig. 1. Schematic crystal, magnetic, and orbital structure of $KCuF_3$. Solid and open circles represent the Cu and F ions, respectively. K ions are omitted. The arrows represent the direction of the spin moment. Each Cu site accommodates a hole in the $x^2 - z^2$–type orbital in the ionic limit.

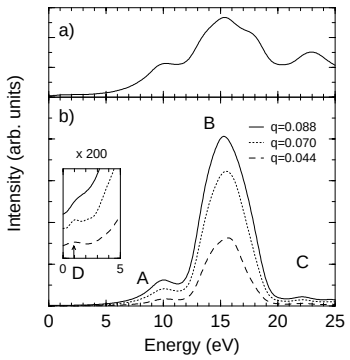

Fig. 2. (a) Calculated absorption spectra. (b) RXS spectra corresponding to the (331) reflection in Ref. [1,2]. They are calculated in the AFM ground state with the OO. Energy is set at the photon energy exciting an electron from 1s state to the states at the top of the valence band.

gap 0.8 eV and the Cu local spin moment $0.67\mu_B$. The calculated ground state has the OO compatible with the picture shown in fig.1. The degree of the OO little decreases, even though the magnitude of the JTD is reduced to $q = 0.044$. The ferromagnetic (FM) ground state also has the OO in the same degree as the AF ground state. The nonmagnetic (NM) ground states calculated with assuming $U = J = 0$ eV has no OO, even though the substantial magnitude of the JTD is assumed.

In the calculation of the x-ray spectra, we assume the core-hole lifetime broadening $\tau^{-1} = 1$ eV and neglect the effect of the core-hole potential to the $4p$ states. Fig.2 shows the calculated absorption and RXS spectra.

3. Results and Discussion

The calculated absorption spectra show good agreement with the experiment[1,2]. This indicates the reliability of the FLAPW calculation for describing the x-ray spectra.

The calculated RXS spectra around the MKE in the AFM ground state with the OO have two noticeable peaks denoted by A and B and a small structure denoted by C. These features are in agreement with the experiment[1,2]. In the FM ground state with the OO and the NM ground state without the OO, the calculated RXS spectra around the MKE are almost the same as the spectra in the AFM ground state. This indicates that the MO and the OO themselves have little influence on the intensity of the RXS spectra around the MKE. The MKE intensity decreases as the magnitude of the JTD is reduced, although the degree of the OO dose not decrease in the AFM ground states.

The inset in fig.2-b shows the magnified spectra around the pre–K–edge(PKE). The quite weak shoulder structure denoted by D is found on the foot of the MKE peaks. This structure becomes clearer as the magnitude of the JTD is reduced. This suggests that the PKE intensity may reflect the OO. The detailed discussion on the similar structure in the $LaMnO_3$ has been described in Ref.[7].

The result of the present paper suggests that the MKE intensity reflects mainly the lattice distortion, not the magnetic order nor the orbital order, while that PKE intensity may reflect the orbital order. In this respect, the large increase of the intensity around the MKE below the Neél temperature[1,2] indicates considerable increases of the magnitude of the JTD in the magnetic phase. More detailed studies on the PKE structure and also the studies on the magnetic scattering spectra are desirable.

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