Orbital ordering and the impurity effect in copper fluoride

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Introduction

It has been recognized that charge, spin, and orbital orderings play important roles in electronic and magnetic properties of strongly correlated electron systems. The long-range orderings have successfully been observed by resonant x-ray scattering (RXS) and neutron scattering and so on. Impurity effect on these ordered states has attracted a great deal of interest. However, few studies on the impurity effect are reported in orbitally ordered systems. We have investigated the effect on a typical orbital ordering system KCuF₃, K₂CuF₄ by substituting Zn (3d⁰) for Cu (3d⁹).

Experimental Results

Increasing Zn concentration we have found the difference between lattice constants a and c becomes small and to be zero around x = 0.6. This result indicates that a structural phase transition from tetragonal to cubic occurs near this concentration. It appears that Jahn-Teller distortion due to the orbital ordering in high Cu concentration releases with decreasing Cu concentration.

To confirm this we have applied the resonant x-ray scattering (RXS) technique to this system. The x-ray energy near the Cu K-edge (1s → 4p) energy was used for the RXS experiments. The RXS intensities rapidly decrease with decreasing Cu concentration and disappear around x = 0.6 at room temperature. This means the tetragonal and cubic phases correspond to an orbital ordered and disordered phases, respectively. We have made a concentration-temperature phase diagram from these diffraction data.

Figure 1 shows the RXS intensities at $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ of KCuₐ₀Znₐ₀F₃, together with the intensities at $Q = (2,2,2)$ which gives the instrumental resolution. This linewidth at $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ is much broader than that at $Q = (2,2,2)$, because the concentration $x = 0.60$ is near the critical concentration of the orbital ordering. The RXS intensities of this sample correspond to the scattering intensities from the percolating network of the orbital. This broad peak may reflect the scattering of the fractal structure in diluted orbital ordering system. In that case, a double-logarithmic plot of broad peak becomes to be linear. The scattering intensities for the fractal structure show where $Q$ is reciprocal vector, and $D_s$ is fractal dimension [1].

Figure 2 shows the double-logarithmic plot of Figure 1. We observed a linear part in a broad Q region. This means the dilute orbital structure has a self-similarity, i.e. a fractal structure. The obtained fractal dimension is about $D_s = 2.5$. This is the first experimental observation of the fractal structure in diluted orbital ordering system.

In conclusion, we have observed the RXS intensities at $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ of KCuₐ₀Znₐ₀F₃, and found the fractal structure in diluted orbital ordering system at the critical concentration.

References


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