Resonant x-ray scattering at the Ru L-edges


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Resonant x-ray scattering provides an important tool for the investigation of magnetic and orbital order in transition metal oxides. This is particularly true for L-edge resonances, where the transitions directly probe the ordered d-states. For the 3d-transition metals, these energies lie in the soft x-ray region below 1 keV. In this energy range one deals with high x-ray absorption in air and the small accessible q-space region, which make measurements difficult. For the 4d-transition metals the respective energies lie in the more convenient x-ray region between 2 – 4 keV. Much of this energy range can be accessed at the hard x-ray beamline 4-ID-D at the Advanced Photon Source (APS) at Argonne National Laboratory (ANL). There, the vacuum chamber of the synchrotron has been modified in order to close the undulator gap and reach the required energies. At the same time the flexibility of the hard x-ray diffractometer setup, including polarization analysis, is preserved. Considerable effort had to be made to reduce the absorption in the beam path.

Different ruthenium compounds, including the single-layer compound Ca₂RuO₄ and the bi-layered system Ca₃Ru₂O₇, have been investigated at the ruthenium L_{II} and L_{III}-edges with photon energies of 2.968 keV and 2.837 keV, respectively, using the strong resonance enhancement of the signal due to magnetic and orbital order at the absorption edges. The resonance at the magnetically allowed (100) position of Ca₂RuO₄ is shown in Fig. 1 (left). A resonance enhancement of at least a factor of 500 is achieved at the L_{II}-edge [1].

The Ca₂RuO₄ compound shows a first-order metal-insulator transition at T_{MI}=356K. A tilt of the RuO₆-octahedra that varies slowly from low temperatures up to T_{MI} gives rise to superlattice reflections at the (100) position. The intensity of this structural reflection is almost constant over the whole temperature range below T_{MI}. The system orders antiferromagnetically at T_N=110K giving rise to superlattice reflections at (100) and (011). The antiferromagnetic order has been observed previously by neutron diffraction [2], and is also observed at the L-edge resonances for T<T_N (see upper right panel of Fig. 1).

Unexpectedly, the signal at the magnetically allowed (100) position does not vanish completely above T_N, but remains up to a temperature of T~260K (Fig. 1, right, lower panel). Exactly the same behaviour was observed also for the other magnetic reflection at (011). This temperature dependence of the magnetic intensities is clearly distinct from the one of the tilt-order reflection (110), which stays up to the MI transition [2]. There is only intensity in the \( \sigma−\pi' \)-channel and MuSR-data show no ordered magnetic moment above T_N. The additional phase transition is thus attributed to orbital order.

The bilayered compound Ca₃Ru₂O₇ shows antiferromagnetic order at T_N=56K followed by a metal-insulator transition at a lower temperature T_{MI}=48K. In the low temperature phase colossal magneto-resistance and quantum oscillations are observed [3]. These phenomena may originate in the strong coupling between spin and orbital order. A reorientation of the magnetic moment takes place around the MI transition temperature. This reorientation can be investigated by resonant x-ray scattering, which is sensitive to the direction of the magnetic moment with respect to the diffraction plane. By carrying out azimuthal dependences, i.e. a rotation of the crystal around the scattering vector, on different reflections, the moment directions can be determined by fitting the magnetic resonant scattering cross section to the data. Until now, the direct observation of orbital order was not possible at this compound also in our investigations, which indicates that ferro-orbital order might be present.
In summary, resonant x-ray diffraction at the L-edges offers a versatile tool for the investigation of orbital and magnetic order in small single crystal 4d-transition metal oxides.

Fig. 1: L_{II} and L_{III} resonances in Ca_{2}RuO_{4} at the (100) reciprocal space position (left). The inset shows the L_{III}-edge resonance together with the absorption coefficient $\mu$ as function of energy. On the right, the temperature dependence of the (100) intensity is shown both in linear (top) and logarithmic scale (bottom) with (open circle) and without (closed circle) polarization analysis. The green triangles show the integrated intensity of the second resonance peak 4eV above the edge.

References

