

Resonant X-ray Diffraction Studies of LaTiO₃ and LaVO₃ Perovskites

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Introduction: Lately, transition-metal oxides have attracted a lot of interest due to their fascinating properties, the most conspicuous being Hi-Tc superconductivity and Colossal Magneto Resistance. The study of these systems is not only important from possible applications, but also for fundamental understanding of strongly correlated electron systems. The strong interaction and ordering of orbital, spin and charge degrees of freedom underlies the unusual behavior and rich phase diagrams of these systems.

Anomalous x-ray diffraction with photon energies near the K absorption edge of the transition metal ion has recently been established as a very sensitive and direct probe of orbital and charge ordering in Mn perovskites¹. The scattered intensity of superlattice reflections due to orbital and charge ordering is greatly enhanced at the absorption edge. The dependence of the scattered superlattice intensity with the incident photon polarization (accessible via the use of a polarization analyzer crystal) and its azimuthal dependence provide detailed information on the symmetry and wavevectors of the ordered phases.

We have therefore employed resonant techniques in the ongoing study of other transition-metal oxides perovskite systems. The samples were single crystals grown by floating zone technique at the University of Tokyo, cut and polished along a direction of interest in each case.

Of these other systems, the antiferromagnetic (spin- $\frac{1}{2}$) insulator LaTiO₃ is particularly interesting. Neutron scattering studies² have revealed a nearly isotropic spin wave spectrum. This is surprising, in view of the absence of a measurable Jahn-Teller distortion of the TiO₆ octahedra that could quench the orbital angular momentum. We have therefore carried out an extensive search for superlattice reflections at several reciprocal space points such as ($\frac{1}{2}, \frac{1}{2}, 0$), the ordering wavevector expected for t_{2g} orbitals with G-type spin structure, near the Ti K-edge (4.966 keV). No evidence of orbital order (as seen in the manganites) was found, suggesting strong quantum fluctuations of the orbital degrees of freedom. This observation is in agreement with a recent theoretical development³ that establishes LaTiO₃ as a novel and unique realization of an orbital liquid state in a magnetic insulator.

We are also studying the analogous vanadate (spin-1) system LaVO₃. It undergoes a crystallographic (tetragonal to orthorhombic) and magnetic (antiferromagnetic ordering) transition at about 140K⁴. In a preliminary study we have focused on the superlattice reflections at (0,3,0) and (0,1,0) (*Pbnm* orthorhombic notation). A resonant behavior is observed around the V edge for both peaks. The observed azimuthal dependence is of the sine-squared type, and polarization analysis indicates that the scattered intensity is predominantly in the $\sigma \rightarrow \pi$ channel. A pre-edge feature, similar to that of the manganites, is also observed in the resonant energy lineshape. This feature may be due to a quadrupole transition, in which case it provides a direct probe of the 3d states. The scattered intensity is present from low temperature to room temperature and appears to be unaffected by the structural-magnetic transition. A more detailed study of is needed to establish the existence and nature of the ordered phase.

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References: ¹Y. Murakami *et al.*, Phys. Rev. Lett. **81**, 582 (1998), also see M. v. Zimmermann *et al.*, Phys. Rev. Lett. **83**, 4872 (1999). ²B. Keimer *et al.*, Phys. Rev. Lett. **85**, 3496 (2000). ³G. Khaliullin *et al.*, Phys. Rev. Lett. **85**, 3450 (2000). ⁴A. Mahajan *et al.*, Phys. Rev. B **46**, 10966 (1992).