



DETECTION OF ORBITAL ORDER IN TRANSITION METAL OXIDES BY RESONANT X-RAY SCATTERING

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The transition metal oxides have fascinated chemists and physicists for many decades with their intriguing structural, magnetic and electronic properties. One of the remarkable phenomena displayed by a number of these compounds is the Jahn-Teller effect, in which an orbital degeneracy of the transition metal ion is lifted by a low-symmetry lattice distortion, which «chooses» one of the degenerate states. In a lattice of such ions one can encounter a cooperative

Jahn-Teller effect, in which the occupation of the degenerate orbitals displays long-range order, characterized by a well-defined wavevector. For example in LaMnO_3 , the Mn ions are partitioned in two sublattices, in which the $3d_{(3x^2-r^2)}$ or, respectively, the $3d_{(3y^2-r^2)}$ orbitals are occupied. It turns out that orbital order can be favoured not only by the consideration of lattice distortions, but also by other mechanisms, notably by magnetic interactions. This is because the relative orientation of occupied d orbitals in neighbouring ions determines the strength and sign of the indirect exchange (superexchange) interaction between their spins.

Recently, in view of the high current interest on transition metal oxides stimulated by the discovery of cuprate high-temperature superconductors and of colossal magneto-resistance in the manganates, orbital order phenomena have been paid much attention. One aspect which was discussed during the ESRF workshop on Magnetic Scattering is the possibility of a direct observation of orbital order by resonant x-ray scattering at the K edge of the transition metal. This could give a direct handle to the order parameter of this peculiar kind of ordered state, and a measure of its temperature dependence, and the relationship of its onset to structural or magnetic phase transitions, thus contributing to an understanding of its origin. In fact there is up to date no

satisfactory way to observe and measure the orbital order parameter, although in principle non-resonant diffraction should detect the corresponding change in the shape of the electron density. The corresponding signal is however expected to be weak (less than 0.1 electrons per unit cell).

In some recent theoretical work done at the ESRF by M. Fabrizio, M. Altarelli and M. Benfatto, the idea is to exploit the sensitivity of resonant scattering of polarized x-rays to the availability of empty intermediate states of the appropriate symmetry. In an orbitally-ordered system, they are available on one sublattice but not on the other. In this work the particularly intriguing case of V_2O_3 was considered; in this compound, the wavevector of the orbital order has never been experimentally measured and is not unambiguously determined by theory; recent indirect evidence, is in some cases, interpreted as strongly supporting the existence of orbital order, and in some others as putting the very existence of orbital order in question. The conclusion of the Authors, which has not yet been verified, is that orbital order should be directly observable by resonant scattering and therefore all controversy could be settled by experiments.

In experiments performed by Y. Murakami et al. on $\text{La}_{0.5}\text{Sr}_{1.5}\text{MnO}_4$ as well as on LaMnO_3 , a signal at the wavevector of the expected orbital order was detected, which was strongly resonant at the dipole edge corresponding to the Mn 1s to 4p transitions. In theoretical work done in Sendai by S. Ishihara and S. Maekawa, the sensitivity to orbital order was ascribed to a modulation of the energy of the empty 4p levels depending on the selective occupation of the 3d orbitals. These results of the Japanese groups were reproduced in Brookhaven by D. Gibbs and M. Blume. In our opinion, more work is called for, in order to verify whether the 4p level energy modulation is controlled by other factors or not, such as the displacement of the ligand oxygens.

In summary, the workshop provided a very timely opportunity to discuss the contribution of x-ray scattering to the investigation of orbital order, bringing together the groups involved in this problem at the ESRF, at NSLS and in Japan. ■

PROBE COHERENCE VOLUMES AND THE INTERPRETATION OF SCATTERING EXPERIMENTS

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One of the significant features of synchrotron radiation, especially that from 3rd generation insertion devices, is the small-energy bandpass of the emitted radiation. In the case of undulator beams monochromatized by Si crystals at the ESRF, the wavelength bandpass ($\Delta\lambda/\lambda$) attains $\sim 10^{-4}$. As a result the photon beam exhibits partial coherence with a longitudinal coherence length $\xi_L = \lambda/(\Delta\lambda/\lambda)$ of the order of $1 \mu\text{m}$ for 1 \AA x-rays. The possibility of coherence over such dimension is fundamental to the experiments involving «speckle» patterns at the ESRF, but it is also an important consideration when conventional scattering experiments are performed, especially when the quantity ξ_L becomes comparable to the dimensions of the material being studied, or with the extinction length; the latter being written as $1/\mu$ and corresponding to the length over which absorption reduces the intensity of the radiation by a factor $1/e$. Within a coherent volume the amplitudes rather than the intensities of the scattering process must be added. We emphasize here that these ideas are independent of the perfection of the crystal scattering the photon beam, i.e. they apply equally to both the kinematic and dynamic theories of diffraction, although the treatment in the two cases is, of course, different.

The standard procedure for correcting for absorption involves correcting the intensities diffracted from a volume of scattering, i.e. the interference between the amplitudes is first calculated to give the beam amplitude for a given reflection, and then the absorption correction is made on the final intensities. This is appropriate when the extinction length