

Orbital Correlations in Complex Oxides

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Many new phenomena of high scientific and technological importance are strongly influenced by orbital degrees of freedom and their interplay with spin, charge and lattice degrees of freedom [1]. There are numerous examples where orbital correlations are believed to play a key role in determining the underlying physical properties, but direct experimental determination of their existence and influence is missing. Probably the two most prominent examples, which will be discussed in more detail here, are provided by the superconducting cuprates and colossal magnetoresistive manganites.

The superconducting transition in the cuprates is quite unusual and, despite more than a decade of intense efforts, still poorly understood. One of its most intriguing features is a pseudogap in the electronic excitations, resembling the superconducting gap in size and symmetry, that occurs in underdoped samples below a characteristic temperature T^* much higher than T_C , where superconductivity and phase coherence set in. Several theories propose that this pseudogap is due to the presence of some hidden order, which is not easily manifested in observable quantities such as for example the magnetization or other thermodynamic properties. These theories [2-4] are based on the existence of orbital currents around portions of the unit cell, which become ordered at T^* , producing antiferromagnetic moments that have not yet been observed, probably because they are exceedingly small. Such a state with ordered orbital currents however breaks time reversal symmetry. Using angle resolved photoemission with circularly polarized light [5], we do indeed observe a dichroism effect that provides evidence for both time reversal symmetry breaking in the pseudogap state and the strong influence of orbital degrees of freedom.

Similar to the cuprates, colossal magnetoresistance in manganites is believed to result from a strong competition between spin, charge, and orbital degrees of freedom. Detailed insight into the nanoscale phenomena arising from the frustration induced by this competition is obtained from neutron and synchrotron X-ray scattering studies. In the paramagnetic regime, a simultaneous development of conventional ferromagnetic correlations and strongly anisotropic Huang scattering due to the presence of quasistatic Jahn-Teller polarons is observed [6]. These polarons are orbitally polarized with a transition from a primarily out-of-plane to in-plane orbital occupation upon cooling [7]. In addition, there are short-range polaron correlations in the form of both longitudinal Jahn-Teller stripes competing with checkerboard-type charge fluctuations [8]. Below the ferromagnetic transition, which is close to being first order, and above a critical field line, the polarons are delocalized by the magnetic order, and the system becomes homogeneous.

While the above examples indirectly indicate the importance of orbital correlations, a direct experimental verification is still missing. The most direct tool to investigate orbital effects is probably provided by resonant X-ray techniques [9]. Resonant X-ray scattering for example has been very successfully applied in the hard X-ray regime for studying rare-earth systems, where a fifty-fold resonant enhancement at the Lanthanide L edge [10], and even larger effects of 5 to 7 orders of magnitude at the M-edges [11] have been observed. However, in transition metal ions, whose oxides provide the majority of

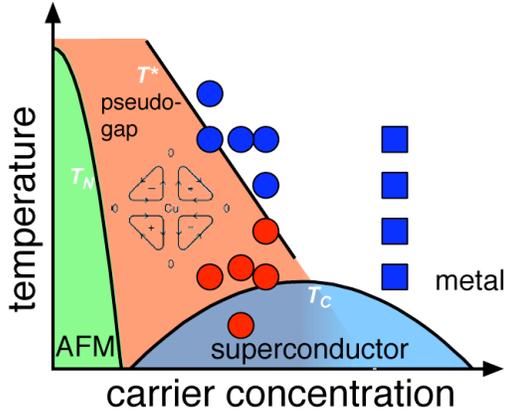


Fig. 1: The observation (red dots) of dichroism in angle resolved photoemission of the cuprate superconductors $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+d}$ provides evidence for time reversal symmetry breaking [5], possibly due to the presence of orbital currents as depicted for the model proposed by Varma [3].

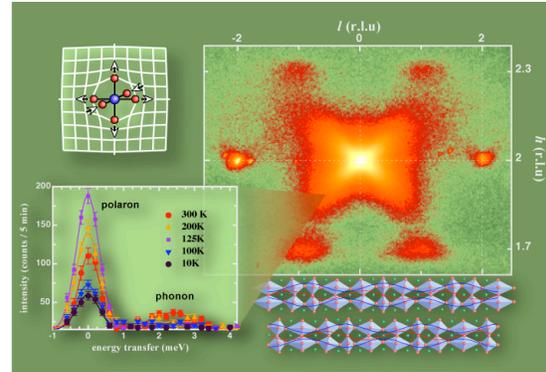


Fig. 2: The diffuse scattering observed from the bilayer manganese oxide $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ provides evidence for both Jahn Teller polarons as well as short range correlations in the form of orbital stripes [6-8].

systems of current scientific and technological importance, the hard X-ray resonant enhancement at the K-edges is much weaker (~ 5). Moreover, because these edges do not directly probe the 3d states, but rather 1s-4p excitations, the origin of these enhancements is controversial, and theoretical studies have shown that explanations other than orbital ordering are more likely. There has been far less attention for single crystal resonant scattering at the transition metal L and M edges, even though strong resonant enhancements in this soft X-ray regime have long been predicted [9]. This lack of interest is due to the perception that absorption by the sample at these X-ray energies would be too high. Recent measurements on single and double layer manganites have however shown that soft X-ray scattering does probe many unit cells and is not particularly surface sensitive [12,13]. Most importantly, these experiments showed that the observed resonant enhancement at the transition metal L edges in the presence of orbital/magnetic order is very large, indeed so large that it can exceed the nonresonant charge scattering. Resonant scattering and other soft X-ray resonant techniques therefore have a huge potential to provide invaluable information regarding orbital and magnetic correlations.

- [1] Y. Tokura and N. Nagaosa, *Science* 288, 462 (2000).
- [2] I. Affleck and J.B. Marston, *Phys. Rev. B* 37, 3774 (1998).
- [3] C.M. Varma, *Phys. Rev. B* 55, 14554 (1997).
- [4] S. Chakravarty *et al.*, *Phys. Rev. B* 64, 094503 (2001).
- [5] A. Kaminski *et al.*, *Nature* 416, 610 (2002).
- [6] L. Vasiliu-Doloc *et al.*, *Phys. Rev. Lett.* 83, 4393 (1999).
- [7] B.J. Campbell *et al.*, *Phys. Rev. B* 67, 020409 (2003).
- [8] B.J. Campbell *et al.*, *Phys. Rev. B* 65, 014427 (2001).
- [9] J.P. Hannon *et al.*, *Phys. Rev. Lett.* 61, 1245 (1988).
- [10] D. Gibbs *et al.*, *Phys. Rev. Lett.* 61, 1241 (1988).
- [11] E.D. Isaacs *et al.*, *Phys. Rev. Lett.* 62, 1671 (1989).
- [12] S.B. Wilkins *et al.*, *Phys. Rev. Lett.* 90, 187201 (2003).
- [13] S.B. Wilkins *et al.*, *Phys. Rev. Lett.* 91, 167205 (2003).