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Optical spectroscopic study of multiferroic $BiFeO_3$ and $LuFe_2O_4$[1]

XIAOSHAN XU, University of Tennessee, Knoxville

Iron-based multiferroics such as $BiFeO_3$ and $LuFe_2O_4$ exhibit the highest magnetic and ferroelectric ordering temperatures among known multiferroics. $LuFe_2O_4$ is a frustrated system with several phase transitions that result in electronically driven multiferroicity. To understand how this peculiar multiferroic mechanism correlates with magnetism, we studied electronic excitations by optical spectroscopy and other complementary techniques. We show that the charge order, which determines the dielectric properties, is due to the "order by fluctuation" mechanism, evidenced by the onset of charge fluctuation well below the charge ordering transition. We also find a low temperature monoclinic distortion driven by both temperature and magnetic field, indicating strong coupling between structure, magnetism and charge order. $BiFeO_3$ is the only known single phase multiferroics with room temperature magnetism and ferroelectricity. To investigate the spin-charge coupling, we measured the optical properties of $BiFeO_3$. We find that the absorption onset occurs due to on-site Fe^{3+} excitations at 1.41 and 1.90 eV. Temperature and magnetic-field-induced spectral changes reveal complex interactions between on-site crystal-field and magnetic excitations in the form of magnon sidebands. The sensitivity of the magnon sidebands allows us to map out the magnetic-field temperature phase diagram which demonstrates optical evidence for spin spiral quenching above 20 T and suggests a spin domain reorientation near 10 T. Work done in collaboration with T.V. Brinzari, R.C. Rai, M. Angst, R.P. Hermann, A.D. Christianson, J.-W. Kim, Z. Islam, B.C. Sales, D. Mandrus, S. Lee, Y.H. Chu, L. W. Martin, A. Kumar, R. Ramesh, S.W. Cheong, S. McGill, and J.L. Musfeldt.

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