Linear and nonlinear optical spectroscopy of multiferroics and related materials

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In this talk we report on the study of electronic structures in several groups of 3d transition-metal multiferroics and related materials using linear and nonlinear optical spectroscopy. The main experimental methods used are polarized optical absorption, variable angle spectroscopic ellipsometry, and second harmonic generation (SHG). Experiments were done in a wide spectral range from 0.5 to 5.4 eV, at temperatures from 2 to 300 K, and in magnetic fields up to 7 T.
The following materials and topics are discussed in the paper:
1. Ferroelectric-antiferromagnetic hexagonal manganites $RMnO_3$ (R=Sc, Y, In, Ho- Lu) where Mn^{3+} ($3d^4$) ions occupy unusually 5-fold coordinated positions. As a result the electronic structure of hexagonal manganites substantially differs from the structure of the orthorhombic perovskite-type manganites. We show that the optical absorption edge is determined by an abnormally strong (k~1) and narrow electric dipole transition with the center at approximately 1.6 eV with light polarization in the basal plane. We treat this transition as a charge-transfer from oxygen to manganese [1].
2. We studied optical dielectric functions of several iron oxides with different crystal structures, magnetic and electric properties. Among them are α-Fe_2O_3, $GaFeO_3$, $BiFeO_3$, $LiFe_5O_8$ [2]. These mutiferroic materials are among very few that have Curie and Néel temperatures above room temperature. In another iron compound $GdFe_3(BO_3)_4$ we studied optical absorption spectra, dielectric functions, and second harmonic generation [3]. For this compound, we prove for the first time possibility of SHG phase matching in magnetic materials.
3. We studied optical absorption and second harmonic generation spectra in noncentrosymmetric copper borate CuB_2O_4 where three types of magnetic-field-induced second harmonic (MFISH) generation are observed and provide selective access to nonequivalent copper sublattices [4]. Unusually sharp excitonic transitions were observed from the ground Cu^{2+} ion state (x^2-y^2) to excited states (xy), (xz,yz), and (z^2). These observations allowed us to calculate true values of the crystal-field parameters Dq, Ds, and Dt which we used for calculating

Jahn-Teller splitting in other cuprates like La_2CuO_4, Bi_2CuO_4, Nd_2CuO_4 et al.

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[3]. A. M. Kalashnikova, V. V. Pavlov, R. V. Pisarev, L. N. Bezmaternykh, M. Bayer, and Th. Rasing, JETP Letters **80**, 293 (2004).
[4]. R. V. Pisarev, I. Sänger, G. A. Petrakovskii, and M. Fiebig, Phys. Rev. Lett. **93**, 037204 (2004).