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Optical properties of iron oxides¹

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Magnetoelectric coupling in materials like multiferroics, dilute magnetic semiconductors, and topological insulators has attracted a great deal of attention, although most work has been done in the static limit. Optical spectroscopy offers a way to investigate the dynamics of charge-spin coupling, an area where there has been much less effort. Using these techniques, we discovered that charge fluctuation in $LuFe_2O_4$, the prototypical charge ordered multiferroic, has an onset well below the charge ordering transition, supporting the "order by fluctuation" mechanism for the development of charge order superstructure. Bragg splitting and large magneto-optical contrast suggest a low temperature monoclinic distortion that can be driven by both temperature and magnetic field. At the same time, dramatic splitting of the LuO_2 layer phonon mode is attributed to charge-rich/poor proximity effects, and its temperature dependence reveals the antipolar nature of the W layer pattern. Using optical techniques, we also discovered that α -Fe₂O₃, a chemically-similar parent compound and one of the world's oldest and most iconic antiferromagnetic materials, appears more red in applied magnetic field than in zero field conditions. This effect is driven by a field-induced reorientation of magnetic order. The oscillator strength lost in the color band is partially transferred to the magnon side band, a process that also reveals a new exciton pattern induced by the modified exchange coupling. Analysis of the exciton pattern exposes C2/c monoclinic symmetry in the high field phase of hematite. Taken together, these findings advance our understanding of iron-based materials under extreme conditions.

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