Site-specific spectroscopic measurement of spin and charge in multiferroic superlattices

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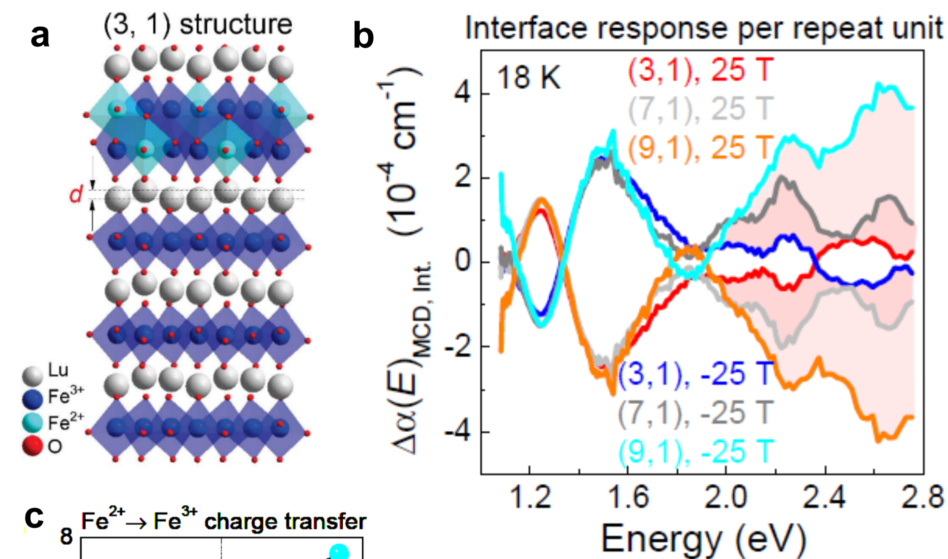
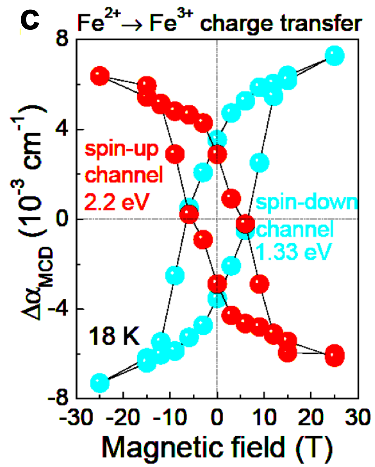
Electric fields can control the magnetism of multiferroic materials, making them technologically interesting. Finding a ferroelectric ferromagnet that is coupled at room temperature is the grand challenge of multiferroics and magnetoelectrics. Heteroepitaxy enlarges the materials design space to achieve this difficult goal. Superlattices of $(LuFeO_3)_m/(LuFe_2O_4)_n$ are room temperature multiferroics that sport ferroelectric ferrimagnetism. However, the microscopic nature of the interface effects and their connection to the robust magnetism in this system are under-explored and not understood.

The Figure summarizes the magnetic circular dichroic responses of all superlattices studied by this collaboration of MagLab users. Figure **(a)** shows the crystal structure of the (3,1) superlattice. **(b)** shows the interface response revealed by subtracting the spectra of bulk $LuFeO_3$ and $LuFe_2O_4$ in proper proportion. There is a dramatic intensity change above 2eV at the interfaces of the three superlattices. Analysis of the interface spectra at different energies reveals that increased Lu-layer distortion selectively enhances the spin-up charge transfer. *Because the dichroic signal is proportional to magnetization, this implies that enhanced magnetization in the $LuFe_2O_4$ layers boosts the Curie temperature in these multiferroic superlattices*. **(c)** shows how constant energy cuts of the dichroic spectra reveal optical hysteresis loops.





(a) Crystal structure of the (3,1) superlattice, showing one $LuFe_2O_4$ slab (with an iron oxide double layer with both Fe^{2+} and Fe^{3+} between a pair of Lu layers) for every three layers of $LuFeO_3$
(b) Interface magnetic circular dichroic spectra for (3,1), (7,1), and (9,1) superlattices for positive and negative 25T magnetic fields.
(c) Optical hysteresis for the spin-up and spin-down channel $Fe^{2+} \rightarrow Fe^{3+}$ charge transfer excitations.

Facility used: DC Field Facility: 25T split helix magnet

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