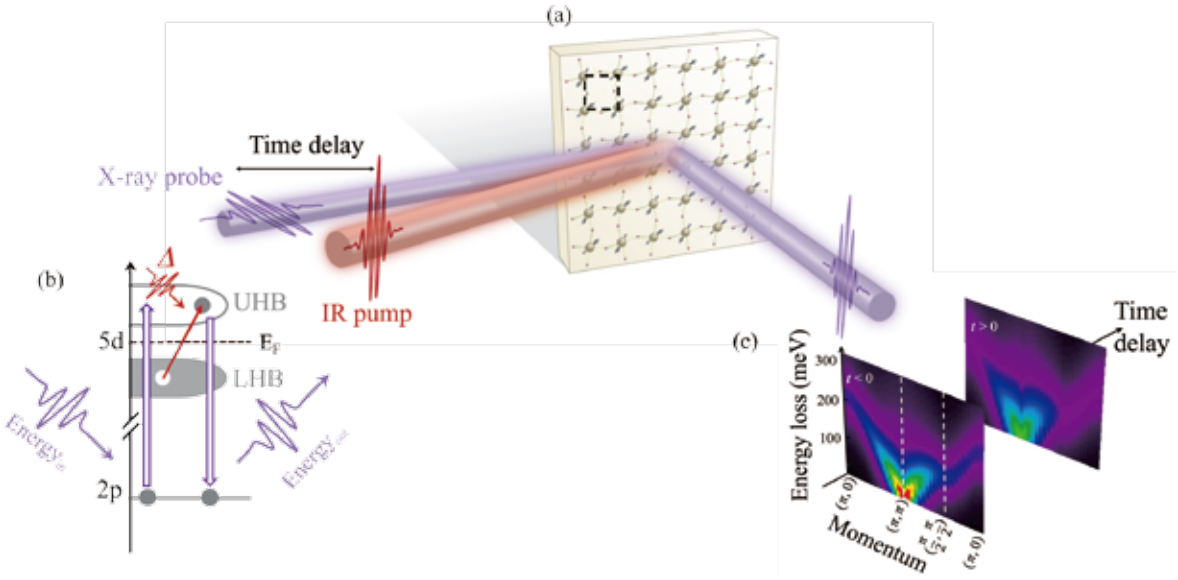
Ultra-fast X-ray Lasers Illuminate Elusive Atomic Spins

A quick flash of light can make ordinary materials extraordinary, potentially inducing qualities such as the perfect efficiency of superconductivity even at room temperature. These subatomic transformations, however, are infamously fleeting – they vanish in just trillionths of a second. Subsequently, how to measure and characterize the manifested properties in such a short time frame becomes extremely challenging. Very few techniques or approaches are available for related studies.

Now, an international team, including scientists from China, the U.S., Germany, Japan, Spain, and the UK, has used synchronized infrared and X-ray laser pulses to simultaneously manipulate and reveal the ultra-fast magnetic properties of this promising quantum landscape. The rapid, light-driven switching between magnetic states, explored by them with unprecedented precision, could one day revolutionize the reading and writing of data in computers and other digital devices.

The results of their study were published on May 9, 2016 in the journal *Nature Materials*, entitled as "Ultrafast energy- and momentum-resolved dynamics of magnetic correlations in the photo-doped Mott insulator Sr_2IrO_4"

Nowadays, large X-ray free electron laser (FEL)



Experimental configuration. a, The scattering set-up. The vertically polarized pump pulse (shown in red) is incident on the *ab*-face of Sr_2IrO_4. X-ray pulses from a free-electron laser (shown in purple) probe the resulting transient state. X-rays that are scattered close to 90° are either directly measured, to access the magnetic Bragg peak that probes the presence or absence of 3D magnetic order, or energy analysed to access the inelastic spectrum, which is particularly sensitive to the 2D magnetic correlations. The basic in-plane structural unit of Sr_2IrO_4 is outlined with a dotted black line. **b,** An illustration of the pump and probe processes. The 620 meV (2 μm) pump beam (in red) photo-dopes the sample, exciting an electron across the Fermi energy, EF , from the lower Hubbard band (LHB) to the upper Hubbard band (UHB). Horizontally polarized 11.215 keV X-ray pulses from a free-electron laser (shown in purple) probe the resulting transient state. The incident X-ray pulses excite an Ir 2*p* core electron into the 5d valence band, to couple to the spin degree of freedom. The resulting emitted photon encodes the magnetic and orbital configuration of the transient state[1]. **c,** Illustration of the detection of X-rays as a function of energy loss, momentum transfer and time delay, encoding the time-dependent magnetic correlations in the transient state. The RIXS planes plot simple spin-wave calculations based on an increased thermal population of magnons after the pulse. (Source: *Nature Materials*, 2016, DOI: 10.1038/NMAT4641)

[1] Ament, L. J. P., van Veenendaal, M., Devereaux, T. P., Hill, J. P. & van den Brink, J. Resonant inelastic X-ray scattering studies of elementary excitations. *Rev. Mod. Phys.* 83, 705–767 (2011).

facilities, such as LCLS and SACLA, can supply high quality X-ray pulses with remarkable short time length of femtoseconds. The challenge is how to use such powerful tool to detect the dynamic response of the spins. That is, a specialized X-ray detection system or "camera" is needed. The scientists developed a highly specialized resonant inelastic X-ray scattering (RIXS) spectrometer for FEL, which used millimeter-sized silicon crystals to measure the exact energy of the rebounding X-rays, allowing reduction of the transient electronic and magnetic qualities.

In this work, an infrared laser was used to manipulate the spin, and time-resolved RIXS (tr-RIXS) was used to catch the spin in motion. The data revealed a clear difference in the propagation and timescale of the magnetic phenomena, with the inter-layer correlations taking hundreds of times longer to recover than those within each layer. Further, and more importantly, the magnetic dynamics showed significant difference between the equilibrium state and the transient state, where the low energy magnon spectral weight was largely enhanced. This indicates that the low energy spin dynamics is strongly coupled to the carriers, which come from photo-doping in this case.

These findings demonstrate the strength and precision of tr-RIXS, which opens the door to revealing femtosecond magnetic dynamics in as yet unseen detail. This made one more step closer to perfecting a recipe for manipulating materials on ultra-fast time scales.

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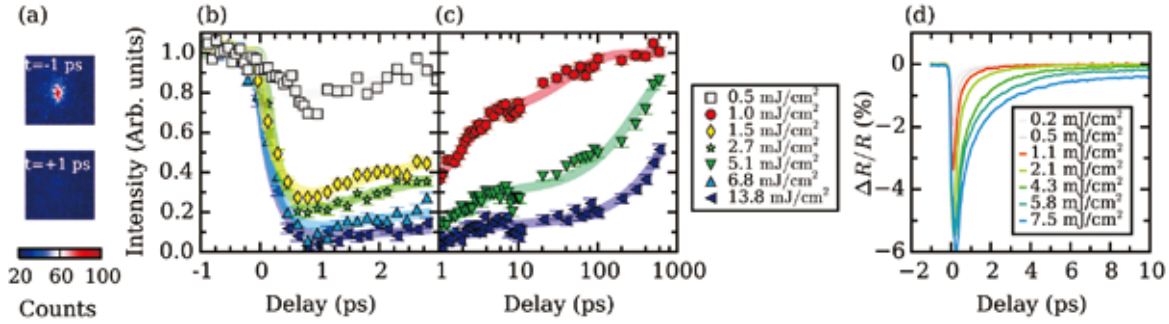
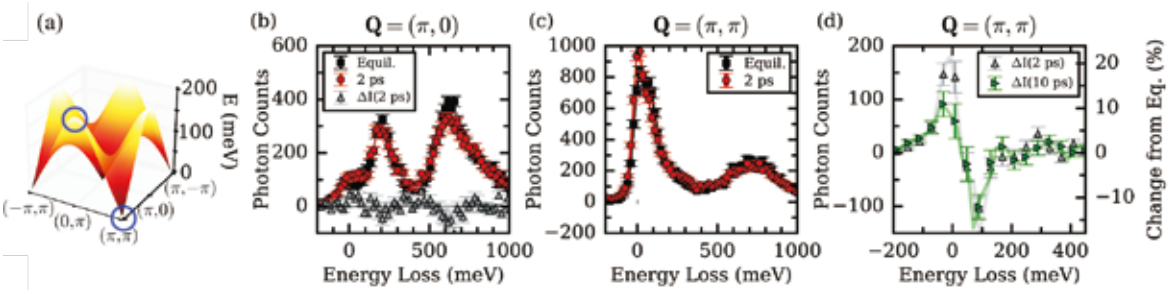


Destruction and recovery of charge and 3D magnetic order in Sr_2IrO_4 . a, Intensity of the (−3, −2, 28) magnetic Bragg peak 1 ps before (top panel) and 1 ps after (bottom panel) excitation at 6.8 mJ cm^{-2} . **b, c,** Intensity of the magnetic Bragg peak as a function of probe delay focusing on the short (b) and long (c) timescales. The lines show the result of fitting the model, which incorporates one decay timescale and two recovery timescales. **d,** Relative change in the 800 nm optical reflectivity ($\Delta R/R$) of Sr_2IrO_4 after excitation with a 620 meV pump at different fluences. All data are taken at 110 K. Error bars represent the statistical uncertainty in the intensities assuming Poisson counting statistics. (Source: *Nature Materials*, 2016, DOI: 10.1038/NMAT4641)



2D magnetic correlations before and after photo-excitation. a, Equilibrium state magnetic dispersions of Sr_2IrO_4 based on a spin-wave fit to measurements in previous research by Kim, J. *et al.*[2]. The **Q**-vectors studied are outlined in blue. **b,c,** Time-resolved RIXS (tr-RIXS) spectra showing magnetic excitations (0–200 meV) and orbital excitations (600 meV) in the equilibrium state 50 ps before photo-excitation (labelled Equil.) and 2 ps after photo-excitation at 6 mJ cm^{-2} . **b,** Spectra show that high-energy nearest-neighbour 2D magnetic correlations, as probed at **Q** = (π, 0), have completely recovered 2 ps after the pump. **c,** Spectra show that magnetic fluctuations at **Q** = (π, π) with relatively low energy arise from a small disturbance of the Néel order. **d,** Intensity difference spectra, ΔI, between the equilibrium state and the 2 ps transient state (from **c**) and between the equilibrium state and 10 ps. This shows a depletion of approximately 20% of the magnetic spectral weight around 100 meV and additional spectral intensity appearing at very low energy. Poisson counting statistics were used to determine the error bars in **b–d.** (Source: *Nature Materials*, 2016, DOI: 10.1038/NMAT4641)

[2] Kim, J. *et al.* Magnetic excitation spectra of Sr_2IrO_4 probed by resonant inelastic X-ray scattering: establishing links to cuprate superconductors. *Phys. Rev. Lett.* 108, 177003 (2012).