What is the nature of the magnetic state in a photo-doped Mott insulator?

A central aim of condensed matter physics is to tune materials' properties in order to achieve new functionalities or to optimize existing functionality. Historically this has typically been accomplished using chemical substitution. Laser excitation now offers an exciting alternative method, with the advantage that the resulting states are tunable and reversible [1]. There are even reports of transient control of high temperature superconductivity in complex oxide materials [2]. So far, however, efforts to controllably access and refine the properties of new transient states with lasers have been held back by difficulties in probing the momentum and energy dependence of the electronic and magnetic correlations characterizing these states.

We recently made the first implementation of time-resolved resonant inelastic X-ray scattering (tr-RIXS) exploiting the ultrafast and highly brilliant X-ray pulses available at the SPring-8 Angstrom Compact free electron LAser (SACLA) and the Linear Coherent Light Source (LCLS) [3]. This technique was used to measure the nature of the magnetic state in the layered antiferromagnetic Mott insulator Sr_2IrO_4 after we "photo-doped" the material with a laser pulse designed to excite carriers across the insulating gap. We found that the non-equilibrium magnetic state, 2 ps after the excitation, exhibits strongly suppressed long-range magnetic order, but hosts photo- carriers that induces strong, non-thermal magnetic correlations. These two-dimensional (2D) antiferromagnetic correlations recover within a few picoseconds, whereas the three-dimensional (3D) long-range magnetic order restores on a fluence-dependent timescale of a few hundred picoseconds. These dramatically different timescales suggest that measuring the dimensionality of magnetic correlations will be vital for our efforts to understand ultra-fast magnetic processes and that tr-RIXS represents a very powerful new tool for this endeavor.

Figure 1 outlines our approach. Photo-excitation is achieved by visible 620 meV (2 μm) infrared photons incident on the sample. X-ray pulses with an energy of 11.215 keV then come in and are scattered from the Sr_2IrO_4 sample. This particular X-ray energy corresponds to a resonance in which an Ir 2*p* core electron is promoted into the Ir 5*d* valance band before an electron in a different valence state fills the core hole and an X-ray photon is re-emitted. Due to the properties of the core hole, this process is sensitive to the magnetic correlations [4] of the Ir spins. Elastic measurements of the scattered X-ray photons access the magnetic Bragg peak, which is primarily sensitive to the presence or absence of 3D antiferromagnetic order. We also implemented a spectrometer in order to energy-resolve the scattered photons and access the magnetic excitations that, due to the very weak interlayer magnetic coupling, provide information about the 2D magnetic correlations.

Our magnetic Bragg peak measurements at SACLA **BL3** are plotted in Figs. 2(a) and 2(b). Upon photo-excitation magnetic order is suppressed faster than 300 fs (the time resolution of our experiment). For fluences above 6 mJ/cm^2, 3D magnetic order is destroyed almost entirely. The magnetic recovery then occurs on two time-scales: partial recovery starts in a few ps; but complete recovery takes somewhere between 100 to several thousand ps [3].

We then used tr-RIXS to investigate the nature of the 2D correlations in the transient state, choosing a pump fluence of 6 mJ/cm^2 in order to destroy the 3D magnetic order. Figure 2(c) plots the RIXS spectra at a momentum transfer of Q = (π, 0), which is sensitive to nearest-neighbor correlations along the Ir-O-Ir bond direction. We find essentially the same spectra before and after photo-excitation proving for the first time that 2D magnetic correlations exist in the transient state, despite the suppression of 3D magnetic order. We then examined Q = (π, π) (Fig. 2(d) and 2(e)), which is sensitive to small 2D fluctuations of the antiferromagnetic order. These 2D magnetic correlations recover on a ps timescale. With this in mind, we reexamined the magnetic Bragg peak measurements and found that all the data could be fit

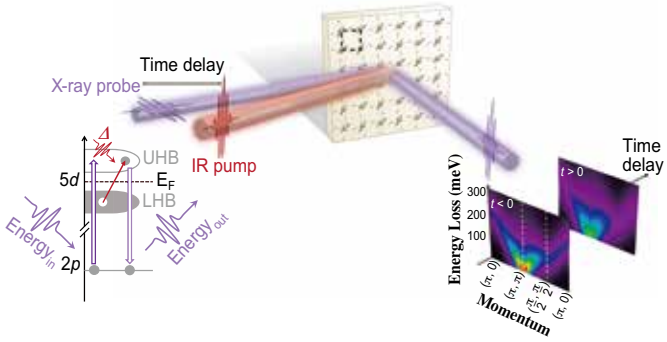

Fig. 1. An illustration of the experimental setup. Photo-excitation, depicted in red, drives electrons across the Mott gap of Sr_2IrO_4. X-rays, shown in purple, are sensitive to magnetism via the Ir *L*-edge resonant scattering cross-section. By measuring the scattered X-rays as a function of energy loss, momentum-transfer and time delay, we can infer 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.

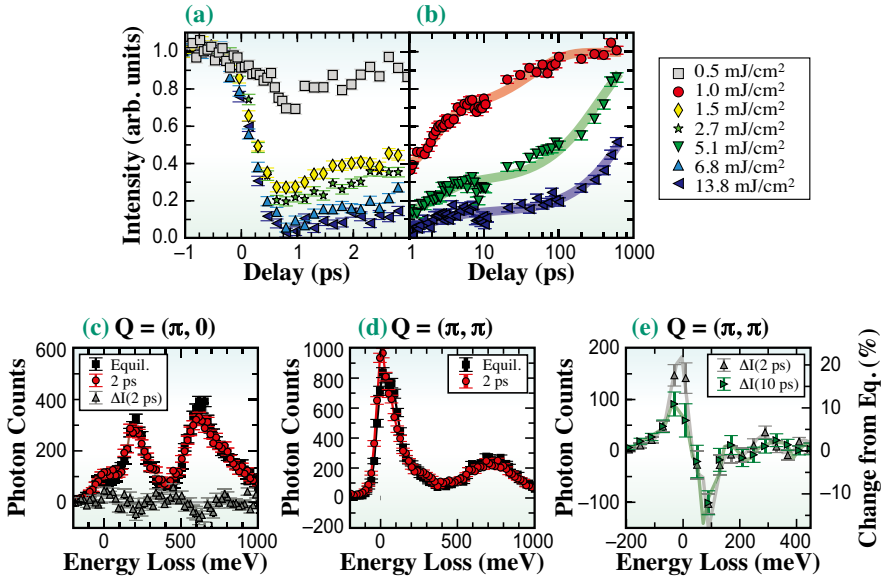

Fig. 2. **(a, b)** Measurements of the magnetic Bragg peak intensity, which is sensitive to the 3D antiferromagnetic order, as a function of time delay after photo-doping at different fluences. A fast suppression is followed by a two-stage recovery. **(c)-(e)** RIXS spectra at different Q points before and after photo-doping at 6 mJ/cm^2. (c) The spectrum at $(\pi, 0)$ indicating that 2D nearest neighbor magnetic interactions recover faster than 2 ps after photo-doping. Data at (π, π) are shown as total spectra (d) and difference spectra (e).

with a two-timescale recovery function (colored lines in Fig. 2(a) and 2(b)) with one fast (ps) timescale and one slow timescale from 100–1000 ps that we associate with 2D and 3D correlations respectively.

Figure 3 plots the dependence of the correlation time scales on the pump fluence. 2D correlations depend weakly on the fluence, changing from 1.5 ps at 0.5 mJ/cm^2 to 2 ps at 13.8 mJ/cm^2. Meanwhile 3D correlations are much slower with a timescale that varies roughly linearly with fluence up to 1,130 ps at 13.5 mJ/cm^2. After magnetic order is destroyed the 2D correlations recover over a few ps. As the 2D magnetic recovery is weakly dependent on the fluence, this process is likely controlled by the strength of intrinsic interactions (such as the magnetic exchange constant of 60 meV). The correlated Ir-O planes then develop inter-plane 3D correlations over hundreds of ps. Since this timescale is linear in fluence, we assume that this process is governed by the timescale over which spins can dump their excess energy into the lattice in order to evolve back towards the ground state.

A full understanding of magnetism in transient states requires studying both 2D and 3D correlation. Tr-RIXS is a uniquely powerful way to access these states on ultra-fast timescales and is therefore likely to play an important role in understanding and manipulating the properties of complex oxides with light.

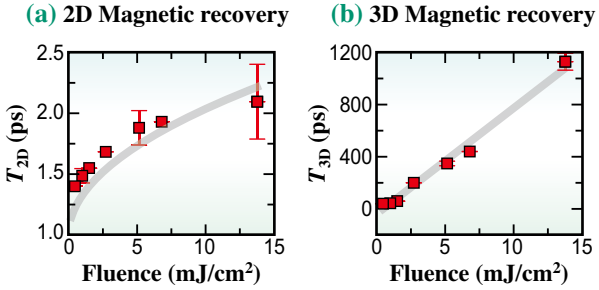

Fig. 3. **(a)** Fluence-dependent magnetic recovery timescales for the 2D correlations and **(b)** the 3D correlations.

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