X-ray scattering from light-driven spin fluctuations in a doped Mott insulator

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Manipulating spin fluctuations with ultrafast laser pulses is a promising route to dynamically control collective phenomena in strongly correlated materials. However, understanding how photoexcited spin degrees of freedom evolve at a microscopic level requires a momentum- and energy-resolved characterization of their nonequilibrium dynamics. Here, we study the photoinduced dynamics of finite-momentum spin excitations in two-dimensional Mott insulators on a square lattice. By calculating the time-resolved resonant inelastic x-ray scattering cross-section, we show that an ultrafast pump above the Mott gap induces a prompt softening of the spin excitation energy, compatible with a transient renormalization of the exchange interaction. While spin fluctuations in a hole-doped system (paramagnons) are well described by Floquet theory, magnons at half filling are found to deviate from this picture. Furthermore, we show that the paramagnon softening is accompanied by an ultrafast suppression of \(d\)-wave pairing correlations, indicating a link between the transient spin excitation dynamics and superconducting pairing far from equilibrium.
Following the early demonstration of ultrafast demagnetization in ferromagnets, the optical manipulation of spin degrees of freedom emerged as an effective strategy for steering electronic properties in quantum materials. Over the years, light excitation protocols evolved from purely thermal effects to more sophisticated optical excitations involving Zeeman interaction, nonlinear phonon excitation, or the renormalization of local interactions. While most of these experimental efforts aim to manipulate magnetic orders, driving fluctuating spins with light could represent an effective dynamical control strategy for superconductivity in strongly correlated electron systems.

In the copper oxides, experimental and theoretical works suggest that spin fluctuations around the anti-ferromagnetic (AFM) wavevector \( q_{\text{AF}} = (\pi, \pi) \) could contribute to the pairing interaction and determine the superconducting critical temperature \( T_c \). Whether optically driving these spin fluctuations may explain the recent observation of light-induced superconductivity in certain copper oxides or be conducive to further optimized light-induced coherence is an open question.

Directly probing the effect of ultrafast light pulses on spin fluctuations at finite momentum \( q \) requires a tool beyond optical probes, which are only sensitive to long-wavelength (\( q \sim 0 \)) dynamics. Resonant inelastic X-ray scattering (RIXS) at transition-metal L-edge with cross-polarized incident and scattered X-rays (see Fig. 1a) is sensitive to magnetic excitations and able to map their dispersions thanks to the high photon momentum. Pioneering time-resolved RIXS (trRIXS) experiments in the hard X-ray regime provided first snapshots of pseudospin dynamics near \( q_{\text{AF}} \) in layered iridates and future soft X-ray experiments at the Cu and Ni L-edges will further expand the reach of this new ultrafast technique. Based on these developments, it becomes critical to theoretically, and possibly experimentally, investigate how dispersive spin fluctuations and superconducting pairing evolve in a prototypical correlated system driven far from equilibrium.

In this work, we present a trRIXS calculation of finite-momentum spin fluctuations in the paradigmatic single-band Hubbard model. We calculate the full trRIXS cross-section for a direct L-edge absorption for \( \pi-\sigma \) polarizations and for two different hole doping levels. We show that an ultrafast resonant pump induces a prompt softening of the short-ranged spin excitation spectrum, compatible with a light-induced renormalization of the exchange interaction [sketched in Fig. 1b], and a simultaneous suppression of \( d \)-wave pairing correlations. Our findings in a canonical Hubbard model demonstrate that light pulses can be used to tailor finite-momentum spin fluctuations and pave the way to novel strategies for the light control of unconventional superconductors.

**Results and discussion**

Microscopic model. With a particular focus on the high-\( T_c \) cuprates, we consider valence and conduction electrons described by the 2D single-band Hubbard model and for two different hole dopings (\( x = 0 \) and \( x = 0.167 \)). Since the X-ray scattering process explicitly involves an intermediate core level, the full Hamiltonian reads as

\[
\mathcal{H} = - \sum_{ij\sigma} t_{ij}^p (d^\dagger_{ij\sigma} d_{ij\sigma} + h.c.) + U \sum_i n_{i\uparrow \sigma} n_{i\downarrow \sigma} \\
+ \sum_{i\sigma} E_{\text{edge}} (1 - n_{i\sigma}) - U \sum_{i\sigma} n_{i\sigma} (1 - n_{i\sigma}) \\
+ \lambda \sum_{i\sigma\sigma'} p_{i\sigma\sigma'}^\dagger p_{i\sigma\sigma'}
\]

Here, \( d_{ij\sigma} (d^\dagger_{ij\sigma}) \) annihilates (creates) a \( 3d_{\sigma} \) electron at site \( i \) with spin \( \sigma \), while \( p_{i\sigma\sigma'} (p^\dagger_{i\sigma\sigma'}) \) annihilates (creates) a \( 2p_{\alpha} \) electron (\( \alpha = x, y, z \)). \( t_{ij}^p \) is the hopping integral between site \( i \) and \( j \), \( n_{i\sigma}^\dagger = \sum_{\sigma'} d^\dagger_{ij\sigma'} d_{ij\sigma'} \) and \( n_{i\sigma} = \sum_{\sigma'} d_{ij\sigma'} d^\dagger_{ij\sigma'} \) are the valence and core-level electron density operators, respectively. The nearest-neighbor hopping amplitude is set to \( t_h = 300 \) meV, the next-nearest neighbor hopping to \( t' = -0.3 t_h \), and the on-site repulsion to \( U = 8 t_h \). The core–hole potential \( U_c \) is instead fixed at \( 4 t_h \) and regarded identical for all \( 2p \) orbitals. \( E_{\text{edge}} = 938 \) eV represents instead the Cu L-edge absorption energy, i.e., the energy difference between the \( 3d \) and \( 2p \) orbitals without spin–orbit coupling. Finally, the spin–orbit coupling with the X-ray-induced core hole in the degenerate \( 2p \)-orbitals is accounted for by the last term in Eq. (1) with \( \lambda = 13 \) eV.

The trRIXS cross-section at a direct absorption edge can be written as (see Supplementary Note 1 for further details)

\[
\mathcal{I}(q_e, \omega_q, \omega_m, t) = \int \frac{d^{3}q}{(2\pi)^3} \int \frac{d^{3}t}{(2\pi)^3} g(t_2 - t_1) \chi_{\text{LSW}} (t_1) g^\dagger (t_2) \\
\times \sum_{\xi=x,y} \chi_{\text{LSW}} (t_2) \chi_{\text{LSW}} (t_1) \sum_{\xi=x,y} \chi_{\text{LSW}} (t_2) \chi_{\text{LSW}} (t_1) \\
\times l(t_1 = t), l(t_2 - t_1) dt_1 dt_2 dt d^3q_e 
\]

where \( q_e = q_c - q_s \) is the momentum transfer between the incident and scattered photons with energies \( \omega_m \) and \( \omega_q \), respectively, and \( l(t) = e^{-t/\tau_{\text{rel}}} \) is the core–hole lifetime decay, with \( \tau_{\text{rel}} = 0.5 t_h \) as its characteristic timescale. For a direct transition (e.g., Cu L-edge), the dipole operator is explicitly given by \( \mathcal{D}_{qe} = \sum_{\alpha\sigma} e^{-i\mathbf{q}_e \mathbf{r}_{\alpha}} (A_{\alpha\sigma} a^\dagger_{\alpha\sigma} p_{\alpha\sigma} + h.c.) \), where \( A_{\alpha\sigma} \) is the matrix element of the dipole transition associated with a \( \sigma \)-polarized radiation.
photon. As shown in Fig. 1a, we employed the $\pi-\sigma$ polarization configuration ($\epsilon_x$ parallel and $\epsilon_y$ perpendicular to the scattering plane) and fixed $\theta_{\text{in}} + \theta_{\text{out}} = 50^\circ$. This probe condition maximizes the spin-flip response in presence of the spin–orbit coupling term.\textsuperscript{26,31} Calculations for the $\pi-\pi$ polarization configuration are reported in Supplementary Note 2 for completeness. Due to the $O(N^2)$ complexity of the trRIXS calculation, we adopt the 12D Betts cluster as a compromise between complexity and finite size.

In our calculations, the vector potential of the optical pump field is

$$A^{\text{pump}}(t) = A_0 \ e^{-i \omega t / 2 \sigma_p} \ \cos(\Omega) \ \hat{e}_{\text{pol}},$$

with frequency $\Omega = 10 t_b \sim 3.0$ eV (above the Mott gap $\sim 4$–$5 t_b$) and duration $6 \sigma_t = 18 \ t_b^{-1} \sim 39.49$ fs, while the probe has a Gaussian envelope $g(t) = e^{-(t-t_0)^2/2 \sigma_p^2} / \sqrt{2\pi \sigma_p}$ with $\sigma_p = 1.5 \ t_b^{-1}$. $A_0$ is measured in natural units $ea_0 \ E_0 / \Omega$, where $A_0 = 0.1$ corresponds to a $E_0 = 79$ mV/Å peak electric field. The pump polarization $\hat{e}_{\text{pol}}$ is linear along the $x$-direction.

Here, we numerically calculate the trRIXS cross-section at each time step $t$ by scanning the full four-dimensional hyperplane defined by the time-integration variables in Eq. (2) at zero temperature. We first determine the equilibrium ground-state wavefunction with the parallel Arnoldi method\textsuperscript{37,38} and, then, calculate the time evolution of the system through the Krylov subspace technique\textsuperscript{39,40}.

**trRIXS and transient spin fluctuations.** In Figure 2, we show selected snapshots of the cross-polarized trRIXS spectrum at $\mathbf{q} = (\pi / 2, \pi / 2)$, where the magnon dispersion is maximized [see Supplementary Note 3 for a comparison with $\mathbf{q} = (0, \pi / 3)$]. At resonance ($\omega_{\text{in}} \sim 931$ eV for Cu $L_3$-edge in Fig. 2), the loss spectra below the Mott gap are dominated by a single intense peak at $\omega \sim 2 J = 0.3$ eV, where $J = 4t^2_f / U$ is the exchange interaction. The energy and shape of this peak reflect the distribution of spin excitations: for an undoped Hubbard model, these manifest as a coherent magnon due to the strong antiferromagnetic (AFM) order; in contrast, in the $\chi = 0.167$ case, the spin excitations jump into a short-ranged paramagnon\textsuperscript{41–45}. Negative time delays, when the system is close to equilibrium, our calculations reveal a splitting of the resonance along the $\omega_{\text{in}}$ axis at $x = 0.167$ doping. This reflects the presence of two different chemical environments, as the valence states can be either half-occupied or empty.

At the center of the pump pulse, the resonance undergoes (i) an overall suppression, and (ii) a redshift along the energy loss axis for both compositions. Since the center of the absorption peak does not exhibit a pump-induced shift along $\omega_{\text{in}}$, one can fix $\omega_{\text{in}} = 931.5$ eV and examine the time evolution of the magnon/paramagnon peak at $\mathbf{q} = (\pi / 2, \pi / 2)$ [see Fig. 3a, b]. The intensity drop and the peak shift closely follow the pump envelope and then persist long after the pump arrival due to the lack of dissipation. The photoexcited RIXS spectrum is visibly broadened at $x = 0$ and to a lesser extent at $x = 0.167$. Next, we analyze the dependence of the spin fluctuation peak as a function of the pump field strength $A_0$ up to a maximum value of 1. This physical regime is relevant to recent experiments on Sr$_2$IrO$_4$\textsuperscript{29} and Sr$_2$Ir$_x$O$_{2+y}$\textsuperscript{30}, which employed fields strengths $A_0$ up to 0.35 and 0.52, respectively. As $A_0$ increases, the peak position for both $x = 0$ and $\chi = 0.167$ shifts monotonically to lower energies [see Fig. 3c, d] by 80% and 50%, respectively. At the same time, the magnon/paramagnon peak broadens due to the generation of high-order fluctuations beyond simple spin-wave excitations and of photo-doped carriers which perturb the spin background. These observations imply that optical laser pulses with negligible momentum have tangible effects on the spin fluctuations at large momenta.

**Magnon/paramagnon energy renormalization.** While we report here an exact calculation of the full trRIXS cross-section, it is useful to discuss the microscopic physics at play in more intuitive terms. The magnon/paramagnon energy softening in our single-band Hubbard model can be attributed to photodoping, a dynamical Floquet-type renormalization of the spin-exchange interaction $J^{\text{FSLW}}$, or a combination of the two. Other experimentally relevant mechanisms, such as magnetophononics\textsuperscript{4,48,49}, would require extending the Hubbard Hamiltonian through the inclusion of additional degrees of freedom.

We first consider the photodoping contribution. Due to the light-induced holon-doublon excitations, the local spin moment $(n_{\uparrow}\downarrow)$ is diluted. In the simplest linear spin wave picture, this will induce a homogeneous softening of the spin structure factors across the entire Brillouin zone. To quantify this effect, we calculate the instantaneous local moment at $t = 0$, defined as $\gamma = \sqrt{(S^{\uparrow}t=0) / (S^{\uparrow}t=-\infty)}$, and plot the renormalized magnon/paramagnon peak energy $\omega_{\text{magnon}}^{\text{FSLW}}$ as dashed lines in Fig. 3e, f. Here, $\omega_{\text{magnon}}^{\text{FSLW}}$ is the numerically evaluated magnon energy at equilibrium. As visible from the comparison, the local photodoping effect, i.e., the dilution of the local spin moment, is too small to account for the observed energy softening.

The other possible mechanism to explain the peak shift hinges on a light-induced renormalization of the effective Hamiltonian. It has been demonstrated that for an ideal system driven by an infinitely long, nonresonant pump, the effective spin exchange energy $J$ is modified by a Floquet dressing of the intermediate doubly occupied states.\textsuperscript{47} In order to quantitatively understand our trRIXS spectrum, we apply Floquet linear spin-wave (FSLW) theory at different pump field amplitudes. In this framework, we assume the system at the center of the pump pulse to be in a steady-state, thus leading to a closed form for the spin excitations dispersion $\omega_{\text{FSLW}}^q$ (see “Methods” section.)

By including both the Floquet renormalization and photodoping effects, we calculate the transient magnon/paramagnon energy $\omega_{\text{magnon/paramagnon}}^{\text{FSLW}}$ and multiply it by a constant factor to match...
the equilibrium peak positions (see Fig. 3e, f). The scale factors are 1.15 for $x = 0$ and 1.02 for $x = 0.167$. At $x = 0$, the FLSW prediction tracks the magnon softening for weak pump fields but grossly deviates at higher pump strength. This is likely due to mobile photo-carriers which frustrate the AFM ground state, and reduces the spin excitation energy. While the former effect is otherwise inhibited. At equilibrium, this term partially cancels the energy cost of spin-flip events. At $x = 0.167$ hole-doping, however, FLSW theory closely aligns with the trRIXS calculation for all pump strengths. We tentatively attribute the nonequilibrium pairing correlations. We calculate the $d$-wave pairing correlation function $P_d = \langle \Delta_d \Delta_d \rangle$ at $x = 0.167$ for various pump strengths [see Fig. 4b], with the factor $\Delta_d$ defined as $\Delta_d = \frac{1}{\sqrt{N}} \sum_{k,k'} d_{k \downarrow}^* d_{-k \uparrow} \cos k_x - \cos k_y$. At the pump arrival, $P_d$ undergoes a rapid suppression, thus indicating a decrease of pairing in the $d$-wave channel. The drop in $P_d$ is found to be monotonic with the increase of the pump strength $A_0$. Intuitively, the value of $P_d$ is determined by both the quasiparticle density and the pairing interaction strength. In our calculations, the pump simultaneously increases quasiparticles via photodoping and reduces the spin excitation energy. While the former effect is favorable for superconductivity, it is the latter that dominates and reduces the nonequilibrium pairing correlations.

Conclusions

We studied the light-induced dynamics of finite-momentum spin excitations in the prototypical 2D Hubbard model through a full calculation of the $\pi - \sigma$ polarized trRIXS spectrum. By exciting a Mott insulator with an ultrashort pump pulse, we find that magnons and paramagnons undergo a dramatic light-induced softening, which cannot be exclusively explained by photodoping. Paramagnon excitations are quantitatively described by a Floquet renormalization of the effective exchange interactions, while magnons at high field strengths deviate from this picture. At optimal doping, we also observe a sizeable reduction of $d$-wave pairing correlations. While here we explore a paradigmatic optical excitation at the Mott gap, alternative mechanisms such as...
phonon Floquet\textsuperscript{57}, ligand manipulation\textsuperscript{2}, multi-band dynamical effects\textsuperscript{58,59} and coupling to cavities\textsuperscript{60,61} might lead to harder spin fluctuations and increased pairing\textsuperscript{47,62}. Future polarization-dependent trRIXS experiments and theoretical calculations will be key to investigate these photoinduced spin dynamics in a wide variety of quantum materials\textsuperscript{63,64}.

Methods

Floquet Linear-Spin-Wave Theory. To describe the evolution of magnetic excitations at finite momentum, we generalize the local Floquet renormalization of the spin-exchange interaction\textsuperscript{46,47} through FLSW theory. This approach is based on the assumption that the driven system can be instantaneously approximated by an effective Hamiltonian $\hat{H}_f$ and a steady state wavefunction $|\psi_f\rangle$. In the Floquet framework, the hopping $t_q$ in the effective Hamiltonian $\hat{H}_f$ is renormalized by a factor $\mathcal{J}(q)$, where $\mathcal{J}(q)$ is the Bessel function of the first kind and $A$ is the vector potential of an infinitely long driving field. Following second-order perturbation theory, the spin-exchange interaction between two coordinates is renormalized into\textsuperscript{57}

$$j_{ij}^\text{eff} = \frac{4t_q^2}{U} \sum_{m} \frac{\mathcal{J}(q)}{1 + mt_qU}.$$

Then, through the standard Holstein-Pikrovskii transformation and large-S expansion, one can obtain the linearized bosonic Hamiltonian

$$\hat{H}_\text{FLSW} = \sum_q \left( A_q - B_q \right) a_q^{\dagger} a_q = \sum_q \frac{\mathcal{J}(q)}{q} a_q^{\dagger} a_q,$$

where $a_q$ is the magnon annihilation operator and the coefficients $A_q$ and $B_q$ in the two-dimensional plane are\textsuperscript{65}

$$A_q = j_{xx} + j_{yy} - j_{xy} \left[ 1 - \cos(q_x + q_y) \right],$$

$$B_q = j_{xx} - j_{yy} \left[ 1 - \cos(q_x - q_y) \right],$$

$$(1, 1) \text{ cos } q_x + j_{xy} \text{ cos } q_y.$$}

Here, unlike the equilibrium counterpart, the coefficients are anisotropic due to the electric field polarization

$$j_{xx}^{\text{eff}} = \frac{4t_q^2}{U} \sum_{m} \frac{\mathcal{J}(q)}{1 + mt_qU},$$

$$j_{yy}^{\text{eff}} = \frac{4t_q^2}{U} \sum_{m} \frac{\mathcal{J}(q)}{1 + mt_qU}.$$}

This derivation can be extended to higher-order following the strategy of equilibrium LSW theory\textsuperscript{65}.

Data availability

The numerical data that support the findings of this study are available from the corresponding authors upon reasonable request.

Code availability

The relevant scripts of this study are available from the corresponding authors upon reasonable request.

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Author contributions

Y.W. and M.M. conceived the project. Y.W. and Y.C. performed the calculations. Y.W. and M.M. performed the data analysis. Y.W. and M.M. wrote the paper with help from Y.C., B.M. and T.P.D.

Competing interests

The authors declare no competing interests.

Additional information

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