Antiphase oscillations in the time-resolved spin structure factor of a photoexcited Mott insulator on a square lattice

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Abstract.
We investigate momentum-dependent transient spin dynamics after photoirradiation in a two-dimensional Mott insulator described by a half-filled Hubbard model on a square lattice by using a numerically exact-diagonalization technique. We find a temporal oscillation in the static spin structure factor of a $\sqrt{18} \times \sqrt{18}$ periodic lattice. The oscillation exhibits an antiphase behavior between, for example, two orthogonal momentum directions parallel and perpendicular to the electric field of a pump pulse. The origin of the antiphase oscillation is attributed to a photoexcited wave function that has overlap with eigenstates producing the $B_{1g}$ bimagnon Raman intensity. Observing such antiphase oscillations will be a big challenge for time-resolved resonant elastic x-ray scattering experiments.

1. Introduction
The recent development of time-resolved resonant inelastic x-ray scattering (trRIXS) and time-resolved resonant elastic scattering (trREXS) tuning incident x rays for the $L$ edge of transition metals opens a new avenue for probing magnetic properties, from which one can investigate novel photoinduced nonequilibrium phenomena in the wide range of momentum and energy spaces [1, 2]. Motivated by the development, we theoretically investigate momentum-dependent spin dynamics that evolves after pumping within a femtosecond timescale in the antiferromagnetic Mott insulator on a square lattice. Using a numerically exact-diagonalization technique for a half-filled Hubbard model with a $\sqrt{18} \times \sqrt{18}$-site lattice, we find novel momentum-dependent transient spin dynamics. In particular, we demonstrate characteristic temporal oscillations for the static spin structure factor, showing an antiphase behavior for two orthogonal directions in the momentum space, which are parallel and perpendicular to the electric field of a pump pulse. Analyzing a time-dependent wave function after pumping, we find that their oscillation period in time is determined by bimagnon excitation in the Mott insulator [3]. This theoretical prediction will be confirmed for Mott insulating cuprates and iridates once trREXS is ready for a femtosecond timescale.

This paper is organized as follows. The half-filled Hubbard model, time-dependent wave function, and static spin structure factor are introduced in Sec. 2. Time-resolved static structure
factors after photoirradiation by a pump pulse is calculated in Sec. 3. Finally, a summary is given in Sec. 4.

2. Model and method
In order to describe Mott insulating state, we take a single-band Hubbard model on a square lattice at half filling given by

\[ \hat{H}_0 = -t_h \sum_{i,\delta} \hat{c}_{i\sigma}^\dagger \hat{c}_{i+\delta\sigma} + U \sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}, \]

where \( \hat{c}_{i\sigma}^\dagger \) is the creation operator of an electron with spin \( \sigma \) at site \( i \), number operator \( \hat{n}_{i\sigma} = \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \), \( i + \delta \) represents four nearest-neighbor sites around site \( i \) and \( t_h \), and \( U \) are the nearest-neighbor hopping and on-site Coulomb interaction, respectively.

We incorporate an external spatially homogeneous electric field applied along the \( x \) direction via the Peierls substitution in the hopping terms, \( \hat{c}_{i\sigma}^\dagger \hat{c}_{i+\delta\sigma} \rightarrow e^{-iA_x(t)\delta\sigma} \hat{c}_{i\sigma}^\dagger \hat{c}_{i+\delta\sigma} \), leading to the time-dependent Hamiltonian \( \hat{H}(t) \). Here, \( \delta_x \) is the \( x \) component of \( \delta \) and \( A_x(t) \) is the \( x \) component of vector potential given by

\[ A_x(t) = A_0 e^{-(t-t_0)^2/(2\Delta^2)} \cos[\omega_p(t-t_0)], \]

where a Gaussian-like envelope centered at \( t_0 \) has a temporal width \( \Delta \) and a central frequency \( \omega_p \). Hereafter we use \( t_h = 1 \) as the energy unit and \( 1/t_h \) as the time unit.

The time-dependent wave function \( |\psi(t)\rangle \) is described by the time-dependent Schrödinger equation. When a time step \( \Delta t \) between two wave functions is as small as \( \Delta t = 0.01 \) used in our calculations, the solution of the Schrödinger equation is given by

\[ |\psi(t + \Delta t)\rangle = e^{-i\hat{H}(t)\Delta t}|\psi(t)\rangle = \sum_n e^{-i\epsilon_n(t)\Delta t}|\phi_n(t)\rangle\langle\phi_n(t)|\psi(t)\rangle, \]

where \( \epsilon_n(t) \) and \( |\phi_n(t)\rangle \) are eigenvalue and eigenvector of \( \hat{H}(t) \). One of techniques for calculating (3) based on a exact-diagonalization scheme is the use of a Taylor expansion [4]:

\[ |\psi(t + \Delta t)\rangle = \sum_{n=0}^{\infty} \frac{(-i\hat{H}(t)\Delta t)^n}{n!}|\psi(t)\rangle \equiv \sum_{n=0}^{\infty} |\varphi_n(t)\rangle. \]

Each term is iteratively obtained by

\[ |\varphi_{n+1}(t)\rangle = -i\hat{H}(t)\Delta t/(n+1)|\varphi_n(t)\rangle \]

starting from \( |\varphi_0\rangle = |\psi(t)\rangle \). The summation can be truncated if the norm \( \langle\varphi_n(t)|\varphi_n(t)\rangle \) is smaller than a critical value such as 10^{-14}.

Since \( |\psi(t)\rangle \) can be expanded by the eigenstates \( |\varphi_n\rangle \) of \( \hat{H}_0 \), the weight of each eigenstate in a given \( |\psi(t)\rangle \) can be measured by a spectral representation such as

\[ W(\omega; t) = \sum_n |\langle\varphi_n|\psi(t)\rangle|^2 \delta(\omega - \epsilon_n + \epsilon_0), \]

where \( \epsilon_n \) \( (n = 0, 1, 2, \cdots) \) is the energenenergy of \( \hat{H}_0 \).

The time-resolved static spin structure factor is defined as

\[ S(q; t) = \langle\psi(t)|\hat{S}_z^q \hat{S}_z^q |\psi(t)\rangle, \]

where \( \hat{S}_z^i = \sum_{\sigma} e^{-i\mathbf{q} \cdot \mathbf{R}_i} \hat{S}_z^\sigma_i \) with \( \hat{S}_z^\sigma_i \) being the \( z \) component of spin operator at site \( i \) with position vector \( \mathbf{R}_i \).

We consider \( \hat{H}(t) \) for a \( \sqrt{18} \times \sqrt{18} \) lattice with periodic boundary conditions. We take \( U = 10 \), which is larger than the band width \( 8t_h \). For the pump pulse, we set \( A_0 = 0.5, t_d = 0.5, t_0 = 0, \) and \( \omega_p = 4, 10, \) and 20. Since the lower and upper bounds of optical absorption spectrum are around \( \omega = 6 \) and \( \omega = 17 \) for the \( \sqrt{18} \times \sqrt{18} \) Hubbard lattice [5], the energy absorbed into the system is small for \( \omega_p = 4 \) and 20 due to an nonresonance condition.

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3. Results

Figure 1 shows $S(q; t)$ for several values of $q$ together with the profile of a pump pulse. The values of $S(q; t)$ change under the pump pulse and show oscillating behaviors with $t$ after turning off the pump pulse. For $\omega_p = 4$ and 20 shown in Fig. 1(a) and 1(c), respectively, time-averaged values are almost the same as the values before pumping. This is due to an off-resonance condition of $\omega_p$. On the other hand, significant changes occur for $\omega_p = 10$: $S(q; t)$ for small $q$ such as $(\pi/3, \pi/3)$ enhances, while that for large $q$ such as $(2\pi/3, 2\pi/3)$ decreases. This is because resonant photoirradiation destroys short-ranged antiferromagnetic correlation, leading to the suppression of spin correlation for large $q$ but the enhancement for small $q$.

We find in Fig. 1 an interesting oscillating behavior of $S(q; t)$, where two momenta equivalent under the reflection with respect to the (1,1) axis [(2$\pi$/3, 0)/(0, 2$\pi$/3) and (\pi, $\pi$/3)/(\pi$/3$, $\pi$/3)] exhibit out-of-phase time dependence. To understand the origin of the antiphase oscillation, we focus on $S(q; t)$ at $q = (2\pi/3, 0)$ and (0, 2$\pi$/3). During pumping centered at $t = 0$ $S(q = (2\pi/3, 0); t)$ [S(q = (2$\pi$/3, 0); t)] decreases [increases] for $\omega_p = 20$ but vise versa for $\omega_p = 10$ and 4. This $\omega_p$ dependence is understood if one regards the pump pulse as an approximate periodic pulse and applies the Floquet theory for the Hubbard model as discussed in [3].

Following this initial change inducing the antiphase condition, an oscillation with the period $\sim 5$ emerges after turning off a pump pulse. At $\omega_p = 20$, this period produces a large peak at $\omega \sim 1$ in the power spectrum. Therefore, we need to clarify the origin of the energy $\omega \sim 1.25$ (not shown here). For this propose, it is reasonable to decompose the photoinduced wave function after pump pulse into the eigenstates of time-independent Hamiltonian $H_0$. Figure 2 shows $W(\omega; t)$ at $t = 2$, where the pump pulse is turned off. For simplicity, we neglect the $\omega = 0$ component and focus on finite energies. We find a weight at $\omega = 1.25$ for $\omega_p = 20$ as shown in Fig. 2(c). Analyzing eigenstates of a subspace with zero total momentum of $H_0$, we realize that the eigenstate at $\omega = 1.25$ has $B_{1g}$ symmetry. This means that the oscillation is a Rabi type between the ground state and the $B_{1g}$ eigenstate, leading the antiphase behavior [3]. We note that the $B_{1g}$ eigenstate is a Raman active state and gives the highest spectral weight in a bimagnon Raman process [6]. With decreasing $\omega_p$, higher-energy weights appear above the $B_{1g}$ state, and their weights become relatively large at $\omega_p = 4$ [see Fig. 2(a)]. Their symmetry is
predominantly $A_{1g}$ not contributing to the antiphase oscillations.

The oscillation period estimated from our calculation is $5/t_h$, which is roughly 30 fs. Recent trREXS has reported the time resolution of 150 fs [2]. Therefore, the resolution should be improved to detect the antiphase oscillation predicted by our calculation. We believe that recent rapid development of trREXS/trRIXS will enable this improvement in the near future.

4. Summary
We have investigated the momentum-dependent transient spin dynamics in the Mott insulator on a square lattice after photoirradiation. Using a exact diagonalization technique for a half-filled Hubbard model on the square lattice, we have found that after turning off a pump pulse, the static spin structure factor temporally oscillates. Examining the components of time-dependent wave function, we have confirmed that the oscillation comes from a Rabi-type one between the ground state and the Raman-active $B_{1g}$ state [3]. We have also found that an antiphase behavior in the oscillations between originally equivalent two momenta, for example, two orthogonal momentum directions parallel and perpendicular to the electric field of a pump pulse. We hope that trREXS will be able to detect these predicted oscillations in the near future.

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References
[1] Dean M P M et al. 2016 Nat. Mater. 15 601
[2] Mazzone D G et al. 2021 Proc. Natl. Acad. Sci. USA 118 e2103696118
[3] Tsutsui K, Shinjo K and Tohyama T 2021 Phys. Rev. Lett. 126 127404
[4] Shirakawa T, Miyakoshi S and Yunoki S 2020 Phys. Rev. B 101 174307
[5] Tohyama T, Inoue Y, Tsutsui K and Maekawa S 2005 Phys. Rev. B 72 045113
[6] Tohyama T, Onodera H, Tsutsui K and Maekawa S 2002 Phys. Rev. Lett. 89 257405, and references therein.