Light–wave Control of Non–equilibrium Correlated States using Quantum Femtosecond Magnetism and Time–Periodic Modulation of Coherent Transport

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Lightwave quantum electronics utilizes the oscillating carrier wave of intense laser fields to control quantum materials properties. Using quantum kinetic equations of motion, we describe lightwave–driven nonlinear quantum transport of electronic spin and charge with simultaneous quantum fluctuations of non–collinear local spins. During cycles of field oscillations, spin–charge inter–atomic quantum excitations trigger non–adiabatic time evolution of an antiferromagnetic insulator state into a metallic non–equilibrium state with transient magnetization. Lightwave modulation of electronic hopping changes the energy landscape and establishes a non–thermal pathway to laser–induced transitions in correlated systems with strong local magnetic exchange interactions.

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Introduction.-- Emergent phenomena in quantum materials arise from competing and cooperative interactions between electronic, spin, and lattice degrees of freedom [1, 2]. Quasi–equilibrium (adiabatic) tuning of multi–component order parameters and microscopic interactions, e.g., by high pressure, magnetic or electric fields, has been used to control the complex phase diagram. However, static perturbations simultaneously affects other material properties that can act against the desired effects. Ultrashort laser pulses provide a different route for manipulating structural and electrical properties of quantum materials far from equilibrium. Ultrafast excitation nonlinear processes can give access to metastable and prethermalized non-equilibrium states in ways not possible through quasi–equilibrium processes [3–24]. Unlike for photoexcitation at optical frequencies, the advent of intense terahertz, midinfrared, and attosecond laser pulses with few cycles of oscillation and well–characterized electric–field temporal profiles has opened new opportunities for non–adiabatic quantum tuning [14, 16, 25, 33]. For example, the lightwave electric field can act as an oscillating force to accelerate electrons in controllable trajectories [15, 16, 25, 29]. Such electronic quantum transport during cycles of lightwave or lattice coherence oscillations can lead to the establishment of non–equilibrium or pre–thermalized states prior to relaxation [9, 13, 15, 16, 34, 35].

In this letter, we investigate the hypothesis that lightwave–driven quantum transport (coherent hopping) of electrons between competing sites with non–collinear spins can be used to coherently control non–equilibrium transitions and transient magnetization during cycles of oscillations [12, 26, 29, 36]. Our main focus is on the role of quantum spin non–thermal fluctuations driven by ultrafast modulation of inter–atomic coherent electronic hopping. We consider the strongly responsive spin background in a correlated magnetic system, where the (Born–Oppenheimer) adiabatic approximation of classical spins [38–40] breaks down during ultrashort timescales. By introducing quasi–particle Hubbard operators and applying a generalized–tight–binding mean field approximation [41], we treat quantum spin fluctuations during coherent electronic hopping between lattice sites in the strong coupling limit of infinite on–site magnetic interaction. Using quantum kinetic equations of motion for the Hubbard quasi–particle density matrix defined on a lattice, we describe the non–adiabatic time evolution of coupled spin–charge states driven by lightwaves.

The ability to experimentally control coherent electron transport on subcycle timescales sets the stage for attosecond magnetism [36], quantum femtosecond magnetism [12, 42–47], and lightwave quantum electronics [15, 16, 25, 28]. The non–thermal spin–charge–lattice pathway studied here can initiate a phase transition stabilized later by the lattice [8, 11, 17, 19, 24]. Here we show that non–thermal charge and spin populations and inter–site coherences can change drastically the shape of the total energy landscape as compared to equilibrium during multi–cycle electric field oscillations. Such “sudden” changes in the lattice forces prior to relaxation initiate coherent lattice displacements that can lead to a phase transition and establish new non–equilibrium states. We show that non–adiabatic quantum spin–charge dynamics evolves the AFM ground state into a non–equilibrium metallic state with transient magnetization, not possible in equilibrium.

Our results suggest a microscopic mechanism for quantum femtosecond/attosecond magnetism [12, 36, 37, 43]. In weakly correlated magnetic systems, it has been debated whether femtosecond magnetization dynamics arises from adiabatic processes associated with electron, spin and phonon populations, or from coherent processes associated with angular momenta interacting with photoexcited electrons [42, 43]. Here we study the limit of infinite on–site magnetic exchange interaction and demonstrate a lightwave driven transition from an AFM insulating ground state to a metallic non–equilibrium state with FM correlation. The predicted emergence of non–linear magnetization during cycles of light–wave oscillations is achieved by simultaneous control of electronic, magnetic, and lattice properties, which is essential for lightwave quantum magneto–electronics at the ultimate speed limit.

Quantum Kinetic Theory.— To model the strong coupling of
spin and charge excitations, we consider composite fermion quasi–particles created by Hubbard operators \[41\]. These Hubbard operators describe transitions between the multi–electron and multi–atom local configurations in the lattice unit cell, such as, e.g., the Zhang–Rice singlet Cu + O configuration in the cuprates or corresponding Mn + O configurations in manganese oxides \[39\]. We adopt a generalized tight-binding mean field approximation \[41\] and project out the high energy upper Hubbard band by assuming strong on–site interaction, e.g., Hund’s rule \[38, 48, 49\]. We consider a three–energy upper Hubbard band by assuming strong on–site inter–mean field approximation \[41\] and project out the high energy configurations have \( J=S+1/2 \) and \( J_z=M=-J \cdots J \). We also consider two orbital configurations \( \alpha \) and \( \beta \) per lattice site, which are split in energy by the local lattice displacement \( Q_i \), due to, e.g., the Jahn–Teller (JT) effect (Supplementary) \[38, 40\]. The spin local \( z \)-axis is defined by the local canting angle \( \theta_i \) that defines the equilibrium spin direction at site \( i \). \( M=S+1/2 \) and \( m=S \) then correspond to spins pointing along \( \theta_i \), as in the case of a classical spin background (Supplementary Sections S1–S2). For the antiferromagnetic (AFM) ground state here, \( \theta_i=0, \pi \).

The Hamiltonian is separated into on–site and inter–site terms, \( H(t) = H_{\text{local}} + H_{\text{hop}}(t) \). The local states discussed above are assumed to diagonalize the local Hamiltonian \( H_{\text{local}} \) in the absence of electronic hopping. Important for describing the quantum spin fluctuations of interest is that \( |\alpha M\rangle \) diagonalize the FM onsite interaction \( J_H S_i \cdot s_i \). In the
strong coupling limit of large $J_H$, we neglect population of the high energy upper Hubbard band, $J = S - 1/2$, which restricts the electronic motion as compared to the weak coupling limit. $H_{\text{local}}$ also includes the site–dependent local energy that depends on the lattice distortions $Q_i$ (e.g., Jahn–Teller (JT) effect) and Zeeman energies $E_{Bz}$ due to coupling of a weak external field $B_{ext}$, which breaks the symmetry and defines the global z–direction (Supplementary). We thus obtain an energy barrier between “bridge” ($Q_i = Q_B \neq 0$) and “corner” ($Q_i \approx 0$) sites leading to an insulator energy gap.

$H_{\text{hop}}(t)$ describes the coherent hopping of an itinerant spin–1/2 electron between lattice sites. The nearest neighbor hopping amplitude $V_{\alpha\beta}(i-j)$ is modulated from equilibrium by the oscillatory lightwaves (Supplementary Sections S3–S4). Coherent electronic hopping from site $j$ to site $i$ occurs via transitions $|\alpha M\rangle \rightarrow |j M-1/2\rangle$ and $|im\rangle \rightarrow |i + 1/2\rangle$, where spin is conserved. We derive and solve the real space density matrix equations of motion defined by Hubbard operators within a mean field approximation (Supplementary). The diagonal density matrix elements, $\rho_i^\alpha(M,t) = \langle |i \alpha M\rangle \langle i \alpha M| \rangle$ and $\rho_i^{\alpha M}(m,t) = \langle |i m\rangle \langle i m| \rangle$, describe the spin–resolved populations of the local configurations at each site $i$. Coherent electronic hopping is described by non–diagonal density matrix elements (quantum coherences) between all possible pairs of lattice site ($i,j$) configurations, with both light–driven and quasi–equilibrium contributions. Below we compare between adiabatic and non–adiabatic time evolution, driven by slow or fast time–dependent changes in the inter–site hopping amplitude $V(t)$.

Adiabatic Dynamics.— We first consider the adiabatic time evolution of a CE/AFM state with $V=0$, Fig. 1(c), driven by slowly varying hopping amplitude $V(t) = t_{\alpha\beta}(i-j) \frac{1}{\hbar}$. $T$ is sufficiently long so that the system reaches a stationary state, $\partial t \rho (t) = 0$, for $t > T$. The tight binding parameters $t_{\alpha\beta}$ used here were taken from Ref. [38], but may be obtained by fitting to ab–initio calculations for specific materials. The initial CE–AFM charge/orbital ordered (CO/COO) state, Fig. 1(c), consists of AFM–coupled zig–zag chains of FM–ordered spins with alternating full (bridge, total itinerant electron population $n=1$, energy $-E_{JT}$) and empty (corner, $n=0$, energy 0) sites (CO). These chains are located in identical x–y planes, which are AFM coupled along the z–direction. In the stationary state obtained after $t \geq T$, the populations of the bridge $M=2$ and corner $m=3/2$ local configurations have decreased, Figs 1(h) and (j) respectively, with a simultaneous increase of configurations with $m=3/2$ and $M=2$ respectively. This is expected for itinerant electron motion along a chain with parallel spins. However, we also see the development of new populations with $M=1$ and $m=1/2$, primarily at the corner sites, Fig. 1(h) and (j). These spin populations indicate local spin canting away from the z–axis and the pristine AFM magnetic order. Such quantum spin canting is seen for the full range of magnetic field and lattice displacements in Fig. S1. For $t > T$, all populations have reached stationary values within numerical accuracy (Fig. S2).

Non–adiabatic dynamics.— We now consider the non–adiabatic dynamics driven by time–periodic modulation of the electronic hopping amplitudes during electric field oscillations, Fig. 1(b). Fig. 2(a) shows the multi–cycle electric fields introduced here, with duration $t_p = 100$fs and frequency $\omega_p$ close to the inter–site energy barrier. These laser fields drive (i) inter–site coherences with dephasing times $T_2 \sim 20$fs ($T_2 < t_p$) that characterize the inter–site electronic coherent hopping, and (ii) non–thermal charge and spin coherent populations, Fig. 2(b). Fig. 2(c) shows the resulting local spin populations, Fig. 2(d), driven by the “Rabi energy” $dR = eaE$, where $a$ is the lattice constant, $e$ the electron charge, and $E$ the electric field amplitude. The initial condition for laser–driven dynamics is the stationary state after adiabatic turn–on of electronic hopping, Fig. 1. The light–induced quantum transport during cycles of electric field oscillations is described by the time evolution of the off–diagonal density matrix elements between all pairs of lattice sites ($i,j$). Fig. 2 shows the resulting local spin dynamics, $S_z(t)$, at the bridge, Fig. 2(b), and corner, Fig. 2(c), lattice sites. The difference in local spin changes between the two sites results in femtosecond magnetization for a small magnetic field $B_{ext}$ that breaks the symmetry. This magnetization is determined by the spin–resolved population dynamics, shown in Fig. 2(d). The charge imbalance between bridge and corner sites in the initial state gives way to a uniform charge distribution, which relaxes back to equilibrium.
Quantum spin canting is stronger on the corner sites. This is $m < 3/2$. Moreover, the population of $M < 2$ states, Fig. 1(e), comes from electronic hopping back to the bridge sites. At the bridge sites, the $M = 2$ majority population decreases from its ground state value, Fig. 1(b), with a simultaneous increase in the $m = 3/2$ local spin population, Fig. 1(c). This reflects the coherent hopping of an itinerant electron FM–coupled to the local spin from a bridge to a corner site during electric field cycles of oscillation. At the same time, new time–delayed local spin populations with $m < 3/2$ develop at the bridge sites, Fig. 1(e), which signifies non–instantaneous quantum canting of the local spin away from its equilibrium orientation $m = 3/2$. Moreover, the population of $M < 2$ states, Fig. 1(g), comes from electronic hopping back to the bridge sites. Quantum spin canting is stronger on the corner sites. This is seen in Figs 2(i–k) by the significant population of $m < 3/2$ and $M < 2$ configurations. This difference between corner and bridge sites leads to the development of FM correlation during lightwave cycles of oscillation, which arises from coherent hopping between sites with AFM spins simultaneously with quantum spin canting. Such lightwave quantum transport of spin and charge populates different spin states prior to relaxation ($T_1$) while simultaneously resulting in a more uniform charge distribution, $\Delta n \sim 0.5$.

So far, we have assumed that the lattice displacements are frozen during electronic hopping timescales. Fig. S1(d) shows that this approximation is justified for adiabatic turnover of $V(t)$, as the total energy landscape $E(Q_B)$ is not influenced significantly by the electronic charge transfer. In contrast, Fig. 3 shows that lightwave quantum transport of electron spin and charge significantly changes the energy landscape out of equilibrium. Fig. 3(a) shows the changes in total energy $E(Q_B, t)$ due to the light–driven spin–charge populations and coherences. Unlike for adiabatic hopping, the electronic quantum transport tuned by $d_R$ leads to significant changes in the overall shape of $E(Q_B, t)$ during cycles of oscillation (Fig. 3). Below electric field threshold, $d_R = 50$ meV in Fig. 3(a), the photoexcited system is insulating, since the total energy minimum remains at a finite $Q_B < Q_B^c$. The main effect with increasing $d_R$ is the softening of the phonon mode, as well as a non–parabolic dependence of $E(Q_B)$, which are evident for $d_R = 100$ meV after three cycles of oscillations (Fig. 3b). Such effects of lightwave charge and spin coherent populations induce anharmonic lattice nonlinear motion and forces. Above threshold, $d_R = 400$ meV, Fig. 3(c) shows that a new global minimum at $Q_B = 0$ develops during lightwave cycles. This change in the non–equilibrium total energy shape favors a metallic phase not possible in equilibrium. The dynamics of the phase transition, driven by coherent lattice displacements $Q_B(t)$ due to time–dependent forces $\frac{dE(Q_B, t)}{dQ_B}$, will be considered elsewhere.

In conclusion, inter–site excitations of itinerant electron spin and charge interacting strongly with an AFM spin background during cycles of oscillations of the modulated inter–site coherent hopping amplitude (i) Create a more homogeneous metallic–like nonthermal electronic population throughout the lattice, (ii) Drive transient magnetization from AFM–ordered spins via quantum spin fluctuations, and (iii) Destabilize the AFM/insulating phase with lattice displacements $Q_i \neq 0$ towards a transient metallic phase with $Q_i \sim 0$. Above electric field threshold, these non–equilibrium effects create non–thermally an initial condition for nonlinear lattice dynamics, Fig. 3 by drastically modifying the energy landscape in ways not possible close to equilibrium. The latter change in energy landscape makes the effects calculated for frozen lattice much more pronounced in a self–consistent calculation. We can thus envision ultrafast manipulation of an insulating phase with non–collinear spins by tunable laser pulse sequences, which can remove lattice distortions and coherently drive insulator–to–metal phase transitions simultaneously with transient magnetization via strong spin–charge quantum couplings.

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