Theory of photoinduced ultrafast switching to a spin-orbital ordered hidden phase

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Photo-induced hidden phases are often observed in materials with intertwined orders. Understanding the formation of these non-thermal phases is challenging and requires a resolution of the cooperative interplay between different orders on the ultra-short timescale. In this work, we demonstrate that non-equilibrium photo-excitations can induce a state with spin-orbital orders entirely different from the equilibrium state in the three-quarter-filled two-band Hubbard model. We identify a general mechanism governing the transition to the hidden state, which relies on a non-thermal partial melting of the intertwined orders mediated by photoinduced charge excitations in the presence of strong spin-orbital exchange interactions. Our study theoretically confirms the crucial role played by orbital degrees of freedom in the light-induced dynamics of strongly correlated materials and it shows that the switching to hidden states can be controlled already on the fs timescale of the electron dynamics.
Photo-induced phase transitions open the intriguing perspective of controlling complex materials on ultra-short timescales, with promising applications in information storage and processing. An intense laser pulse can impulsively create charge excitations, and induce electronic processes which cannot be described in terms of a quasi-equilibrium scenario. This gives rise to rich physics in particular in Mott insulators, where the extensive studies of the Hubbard model have provided insights. This gives rise to rich physics in particular in Mott insulators and explores the landscape of different orders in Mott insulators and explore the landscape of hidden states.

In this work, we investigate the non-equilibrium evolution of the intertwined spin-orbital-ordering and a resulting hidden phase in transition-metal compounds with a partially filled d-shell. In the representative case of one electron (or hole) in two $e_g$ orbitals, such as $d^1$ or $d^9$ configurations, spin and orbital exchange interactions can emerge due to the superexchange mechanism, and result in a spatially ordered pattern for both the spin orientations and orbital occupations. The orbital-ordering drives the lattice to form Jahn–Teller-like distortions. This scenario offers the intriguing opportunity to simultaneously switch spin and orbital orders through non-equilibrium protocols on the ultra-fast timescale.

**Results**

Spin and orbital order in the two-band Hubbard model. We consider a system with a partially filled 3d band in a cubic crystal, such that the $d$-shell is split into two $e_g$ and three $t_{2g}$ orbitals. Typical representatives are the perovskites, with a cubic arrangement of transition-metal ions in an octahedral environment of ligand atoms. We assume the $t_{2g}$ orbitals are inactive (filled or empty), so that the system can be described by a two-band Hubbard model with two $e_g$ orbitals $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ at each site. The local interaction is given by

$$H_U = U \sum_{\gamma} n_{\gamma \uparrow} n_{\gamma \downarrow} + \sum_{\gamma \sigma, \delta \Phi} \left( U - J_H \delta_{\alpha \beta} \right) n_{\gamma \sigma} n_{\delta \sigma} + J_H \sum_{\gamma \Phi} \left( \epsilon_{\gamma \sigma} \epsilon_{\gamma \sigma}^* - \frac{1}{2} J H \delta_{\alpha \beta} \right),$$

where $\gamma$ labels sites and $\alpha = d_{x^2-y^2}, d_{3z^2-r^2}$ is the orbital index. $J_H$ is the Hund’s coupling and $U' = U - 2J_H$. The hopping Hamiltonian is given by

$$H_0 = -t_0 \sum_{\langle \gamma \Phi \sigma \rangle} \epsilon_{\gamma \sigma} \epsilon_{\gamma \sigma}^*$$

where the structure of the $2 \times 2$ hopping matrices $\tilde{T}^{\sigma}$ along the bonds $(ij)||\alpha = x, y, z$ is imposed by the cubic symmetry, and electric fields can be included via a Peierls phase $\phi_j$ (see methods). The hopping amplitude $t_0 = 1$ sets the energy scale. We use non-equilibrium dynamical mean-field theory (DMFT) to solve this problem (see methods).

We consider the case of three-quarter-filling, and choose $U/t_0 = 7$ and $J_H/t_0 = 0.1$ to roughly match the realistic parameter regime of KCuF$_3$, with an insulating gap of $E_g \approx 3$ eV. The time unit $\hbar/t_0$ and the initial temperature $0.01$ then correspond to about 1 fs and 100 K, respectively. At this temperature, the DMFT calculation predicts A-type antiferromagnetic spin-ordering (A-AFM) and antiferro-orbital-ordering (AFO), consistent with both ab initio and mean-field results.

The local spins align ferromagnetically in the $xy$-plane and antiferromagnetically along the $z$-axis, and the hole approximately occupies the orbitals $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ in an alternating pattern (Fig. 1). To represent the orbital order, it is convenient to combine the two orbitals into a spinor $\psi = (c_{x^2-y^2}, c_{3z^2-r^2})$, and define the pseudospin components of the hole $Z_{\alpha} = \frac{1}{2} \psi \sigma_\alpha \psi$, with the Pauli matrices $\sigma_1, 2, 3$. A transformation of the basis orbitals to $\left( d_{x^2-y^2}, d_{3z^2-r^2} \right)$ and $\left( d_{z^2-r^2}, d_{3z^2-r^2} \right)$ corresponds to successive $\theta = 120^\circ$ rotations around the $Z_{\alpha}$-axis, using the rotation matrix $R(\theta) = e^{i\theta} 3/2$, and the resulting pseudospin components in this new basis will be denoted by $X_{\alpha}$ and $Y_{\alpha}$, respectively (Fig. 1).
Concentrating on the site A in Fig. 1 and defining $d_{z^2-r^2}$ and $d_{y^2-z^2}$ as orbital 1 and 2, respectively, the A site has two electrons in orbital 2 and one spin-down electron in orbital 1. The orbital-order parameter can be defined as the occupation difference between the two orbitals, which is the component $X_3 = \frac{1}{2} (n_2 - n_1)$ of the orbital pseudospin. The orbital pseudospin vector $(X_3 \parallel)$ on the B sites is a reflection of $(X_3 \perp)$ w.r.t. the $Z_3$-direction, i.e., the order parameter approximately alternates between the $X_3$ and $Y_3$ directions on neighboring lattice sites, and the ordered phase is invariant under the transformation $\psi_{i\sigma} \rightarrow \psi_{i+\hat{Z},\sigma}$ and $\psi_{i\sigma} \rightarrow \psi_{i+\hat{Z},-\sigma}$. This symmetry is preserved out of equilibrium, so that the general spin-orbital order is given by a three-dimensional manifold parametrized by the magnitudes of $S_z$ and $X_3$ and the direction of the pseudospin, $\tan \theta = X_3/X_1$.

Photo-induced reduction of spin and orbital order. We consider three non-equilibrium protocols: excitation of the $e_g$ system with an electric field pulse with polarization parallel or perpendicular to the ferromagnetic planes, and a photo-doping, i.e., a sudden excitation of electrons into (out) of the $e_g$ manifold by a resonant laser excitation from (to) other bands (see methods). We first study the relaxation after an electric pulse polarized in the diagonal of the $xy$-plane, taken to be a single cycle pulse of period $T \approx 2$. The pulse creates non-equilibrium charge excitations and causes a reduction or melting of the spin and orbital orders, as shown in Fig. 2. Furthermore, while the equilibrium orbital pseudospin $X$ corresponds to a real superposition of orbital states in the $X_1 - X_3$ plane, transiently a small non-zero $X_2$ component emerges, indicating precession dynamics induced by the excitation.\(^{31}\) The long-time relaxation of order parameters can be examined by fitting the time evolution of both $S_z$ and $X_3$ by exponential functions, $S_z(t) = S_z(\infty) + C_\text{S} e^{-t/\tau_3}$ and $X_3(t) = X_3(\infty) + C_\text{X} e^{-t/\tau_X}$, to extract the decay rates $\tau^{-1}_X$ (Fig. 2c) and the extrapolated order parameters at $t = \infty$ (Fig. 2b).

Hidden state. In the long-time limit, the presence of charge excitations leads to a quasi-steady photo-excited state, which does not thermalize on the 100 fs timescale of the simulation due to the Mott gap. We now compare the multi-dimensional order parameter (given by $S_z(\infty)$, $X_3(\infty)$, and the angle $\theta = X_3(\infty)/S_z(\infty)$) in the photo-excited state at different excitation densities, and in various equilibrium states. We first look at the relative magnitude of $S_z$ to $X_3$ by plotting $X_3$ against $S_z$ in different states (Fig. 3a). In thermal equilibrium, as the temperature increases, the $S_z - X_3$ curve first drops to $S_z = 0$ and then proceeds to the high-temperature state $X_3 = S_z = 0$, reflecting the lower critical temperature of the A-AFM order compared to the AFO order. Chemically doped systems follow similar paths, as shown by the red and blue dot-dashed lines for doping $\delta n = \pm 0.01$. The photo-excited states, in contrast, follow a smooth curve in the $S_z - X_3$ plane with $S_z > X_3$ when $n_{\text{ex}}$ is increasing, for all three non-equilibrium protocols. Photo-doping electrons leads to the weakest AFM order, indicating that 4-particle excitations most efficiently destroy the spin order. The photo-excited states at a given density $n_{\text{ex}} = 0.01$ (see square symbols) exhibit different order parameters than the equilibrium system with the same hole or electron doping $\delta n = \pm 0.01$, independent of temperature. Hence the states reached by ultra-fast laser excitation are not accessible under equilibrium conditions. Furthermore, the direction of $X(\infty)$ in the pseudospin space shows that the polarization of orbital order is different in the equilibrium and non-equilibrium states (Fig. 3b). In equilibrium, the angle between $X_A$ and $X_B$ of the two sublattices (~120°) increases with temperature and stays at 180° after the AFM order has melted. On the contrary, photo-excitation results in a decreasing angle, which evolves towards 0° in the strong excitation limit, corresponding to a ferro-axial (FO) ordering with a small magnitude $|X|$.

We now explain the mechanism which drives the system into the hidden state. It follows from the non-thermal nature of quenching the orders by photo-induced carriers, and the spin-orbital exchange interactions, which act differently in this unconventional quenched state compared to the equilibrium state.

Femtosecond quench of spin and orbital order. While thermal melting of spin and orbital order is due to the population of (orbital) spin-waves, the partial quench of the two order parameters after photo-excitation follows an entirely different mechanism: It occurs on the femtosecond timescale, as the motion of charge excitations leaves a string of defects in the
ordered background, similar to the case of the single-band AFM\textsuperscript{19,36,37}. Due to the in-plane ferromagnetism in the \textit{xy}-plane, only the motion of charge excitations along the \textit{z} direction affects the spin order (process 1 in Fig. 4b), while orbital order can be affected by hopping processes in all three directions (process 2 in Fig. 4b) and thus should decrease faster.

To confirm this mechanism, we first note that it corresponds to a transfer of kinetic energy from the charge carriers to the ordered background. This can be seen directly by looking at the electronic distribution functions, which show the relaxation to a low-temperature cold distribution within the electronic hopping (Fig. 4a). Furthermore, at early times, the anisotropy is also reflected in the polarization dependence of the excitation (Fig. 4c). During the pulse, the suppression of the magnetic order is lower for \( E \parallel x,y \), consistent with the fact that this creates mostly in-plane spin-triplet excitations (process 4 in Fig. 4b), while the perpendicular polarization \( E \parallel z \) directly creates spin-singlet doublons (process 3 in Fig. 4b), affecting A-AFM and AFO in the same manner. After several hopping times, the decay rates of the A-AFM and AFO order for both polarizations differ by roughly a factor of two to three, consistent with an independent melting of the two orders due to the hoppings along the different directions. Note that a larger Hund’s coupling \( J_{H} \) might further suppress out-of-plane hopping by energetically penalizing the conversion of spin-triplet into spin-singlet doublons.

**Orbital-order polarization.** After the non-thermal reduction of \( X_{3} \) and \( S_{c} \), the unconventional polarization of the orbital order qualitatively follows from the intertwined dynamics of the two orders due to the spin-orbital exchange interactions. In the Mott phase at \( U > U_{0} \), the latter are described by the Kugel–Khomskii model\textsuperscript{29},

\[
H = \sum_{\langle ij \rangle} \xi_{x}X_{i}X_{j} + \sum_{\langle ij \rangle} \xi_{y}Y_{i}Y_{j} + \sum_{\langle ij \rangle} \xi_{z}Z_{i}Z_{j} + \sum_{i} (\eta_{x}X_{i} + \eta_{y}Y_{i} + \eta_{z}Z_{i}).
\]

Here \( \xi_{\alpha}, \eta_{\alpha} \) are orbital exchange parameters that depend on the spin configuration on the bond \( \langle ij \rangle \parallel \alpha \) with \( \alpha = x, y, z \). In particular,

\[
\eta_{\alpha} = J_{T} \frac{1}{4} \mathbf{S} \cdot \mathbf{S} \text{,}
\]

with a positive exchange interaction \( J_{T} \) obtained through the Schrieffer–Wolff transformation\textsuperscript{38}. The parameter \( \eta_{\alpha} \) is maximized for spin-singlet and minimized for spin-triplet, thus \( \eta_{z} > \eta_{x} = \eta_{y} \) under A-AF spin order.

As \( S_{c} \) vanishes for increasing temperatures, the compass parameters \( \xi_{\alpha} \) and \( \eta_{\alpha} \) become isotropic for \( \alpha = x, y, z \), and the 180° orbital-ordering minimizes the mean-field energy\textsuperscript{24,39}. In the photo-excited states, however, the compass parameters remain anisotropic with a finite \( S_{c} \), while the strong reduction in \( |X| \) renders the linear terms on the second line of Eq. (3) dominant. Therefore, the pseudospins align along the negative \( Z_{3} \)-direction (orbital \( d_{3z2−r}^\alpha \)) due to the dominant \( \eta_{z} \).

**Discussion**

In summary, our finding suggests a pathway to reach hidden states in correlated electron systems with intertwined spin and orbital order on the ultimately short timescale of the electronic hopping. The non-thermal quench transfers energy from photo-induced charge excitations into the A-AFM and AFO ordered backgrounds at different rates. Due to the exchange-coupling between the order parameters, this drives the system to a state that features spin-orbital orders unaccessible in an equilibrium state. In particular, starting with a near-120° AFO ordering, the photo-excited system approaches a ferro-orbital-ordering in the strong excitation limit, which is qualitatively different from the behavior of the equilibrium system.

In particular, the Jahn–Teller effect can be non-negligible in realistic perovskites\textsuperscript{80,81}, but the electronic mechanism would still be a key driving force, among other effects, of the full electron-lattice dynamics. One possible scenario is suggested by recent experiments\textsuperscript{7,14}, where the subsequent lattice dynamics is driven by the fast change in electronic degrees of freedom. This should be the case when the electron-lattice coupling is weak enough and only affects the dynamics on longer timescales than the electronic processes. In the strong coupling limit, on the other hand, the electrons can be dressed with lattice distortions to form

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**Fig. 3** The extrapolated spin and orbital order in the long-time limit. **a** The three different non-equilibrium protocols (electric field pulse, photo-doping in/out electrons) all lead to stronger spin-ordering than orbital-ordering, which is qualitatively different from the behavior of the equilibrium system with increasing temperature (yellow double-dotted line) at integer filling and for the chemically doped systems with \( d_{n} = 0.01 \) (red/blue double-dotted lines). **b** Normalized orbital-order components \( X_{1} \) and \( X_{2} \) in the long-time limit shown on the pseudospin compass. Rising equilibrium temperature and photo-excitation cause the orbital pseudospin to rotate in opposite directions.
polarons. With renormalized parameters, the electronic mechanism of weaker and slower melting of AFO than A-AFM could still be established in the polaron dynamics. Thus, in both cases, the opposite rotation of the orbital pseudospin in the equilibrium and non-equilibrium regimes might reveal itself by inducing opposite Jahn–Teller-like distortions through electron-phonon coupling, which can possibly be detected by time-resolved X-ray diffraction techniques. The competition between Jahn–Teller effect and the electronic mechanism in the intermediate coupling regime can be more complicated and requires further studies.

The existence of new types of order upon photo-excitation is in sharp contrast to the one-band Hubbard model, where the excitation density and effective temperature exclusively determine the spectral properties and exchange interactions in the photo-excited state. In addition to the multi-component order-parameter, the \( \epsilon_f \)-orbital degeneracy allows for multiple types of charge excitations (in particular, there are three different doubly occupied sites). Thus, even locally there are electronically excited states described by a continuum of parameters, potentially giving rise to many near-degenerate phases. In fact, in the present case, along with the different orders, also the probability distribution of the electronic processes occurring in the photo-excited state. In addition to the multi-component order-parameter, the orbital orders on the two sublattices can be related by a \( \epsilon_x \)-field. This self-consistency represents a compass model and has been intensively studied in the literature. Motivated by the mean-field theory, the lattice system is approximated by an effective impurity model coupled to a non-interacting bath. The impurity action approximating the lattice system takes the form \( \mathcal{S} = \mathcal{S}_{\text{int}} - i \sum_{\sigma} f \bar{\psi} \phi(t) \bar{\Delta}(t) \psi(t) \phi(t) \). The two-band Hubbard model involves orbital-mixing terms, but conserves the total spin \( S \) component. Therefore, the hybridization function of the bath \( \Delta_{\sigma}(t, \tau) \), as well as the Green’s functions, are diagonal in the spin indices. We choose the non-crossing approximation (NCA) as the impurity solver, which yields reliable results when the two-band Hubbard model at large \( U \) is considered.

### Methods

#### The two-band Hubbard model

The hopping matrices \( T^\sigma \) are imposed by the cubic symmetry, or in particular, the permutation symmetry of \( x, y, z \)-bonds. \( T^z \), the hopping along the \( z \)-bond, is determined by the only non-vanishing matrix element between \( d_{x^2-y^2} \) orbitals. All other matrices can be determined through rotations:

\[
\begin{align*}
T^x &= \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}, \\
T^y &= \begin{pmatrix} 3 & -\sqrt{3} \\ -\sqrt{3} & 1 \end{pmatrix}, \\
T^z &= \begin{pmatrix} 3 & \sqrt{3} \\ \sqrt{3} & 1 \end{pmatrix}.
\end{align*}
\]

In the one-hole- (three-electrons) or one-electron-filled case, using a Schrieffer–Wolff transformation, an effective Hamiltonian (Kugel–Khomskii model) in terms of spin and orbital pseudospin can be obtained, which preserves the threefold rotational symmetry in the pseudospin space. This model is an example of a compass model and has been intensively studied in the literature.

#### Dynamical mean-field theory

In a non-equilibrium dynamical mean-field theory, the lattice system is approximated by an effective impurity model coupled to a non-interacting bath. The impurity action approximating the lattice system takes the form \( \mathcal{S} = \mathcal{S}_{\text{int}} - i \sum_{\sigma} f \bar{\psi} \phi(t) \bar{\Delta}(t) \psi(t) \phi(t) \). The two-band Hubbard model involves orbital-mixing terms, but conserves the total spin \( S \) component. Therefore, the hybridization function of the bath \( \Delta_{\sigma}(t, \tau) \), as well as the Green’s functions, are diagonal in the spin indices. We choose the non-crossing approximation (NCA) as the impurity solver, which yields reliable results when the two-band Hubbard model at large \( U \) is considered.

The lattice consists of two sublattices \( A \) and \( B \) with different orbital occupations. The self-consistency condition for the hybridization function reads \( \Delta_{\sigma}(t, \tau) = k \sum_{\epsilon} \epsilon^2 \Delta_{\epsilon}(t, \tau) \phi_\epsilon(t) \bar{\Delta}(t) \psi_\epsilon(t) \). The two-band Hubbard model involves orbital-mixing terms, but conserves the total spin \( S \) component. Therefore, the hybridization function of the bath \( \Delta_{\sigma}(t, \tau) \), as well as the Green’s functions, are diagonal in the spin indices. We choose the non-crossing approximation (NCA) as the impurity solver, which yields reliable results when the two-band Hubbard model at large \( U \) is considered.

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Motivated by the mean-field solutions, we consider an intertwined spin and orbital-ordered phase, where \( A \)-type antiferromagnetism and antiferro-orbital order are assumed. The orbital orders on the two sublattices can be related by a unitary rotation \( \mathcal{R} = \hat{R} \), in the pseudospin space which flips the pseudospin w.r.t. the \( Z \)-direction, i.e., \( \tilde{\mathcal{G}}_{\sigma} = \mathcal{R} \tilde{\mathcal{G}} \mathcal{R} \), where the spin \( \sigma \) is determined by the bond, with \( \sigma = \sigma \) for \( a = x, y \) and \( \sigma = -\sigma \) for \( a = z \).

#### Characterization of the photo-excited state

The laser excitation is included in the model by the Peierls phase, as indicated in Eq. (2). Photo-doping is realized by connecting the lattice to an empty or filled Fermion bath for a short time \( t \leq 2 \). The photo-doped system can be characterized by the excitation density \( n_x, n_{\sigma} \) as defined as the sum of excited doublon and 4-particle excitations. In the electronic-pulse case, this quantity can be calculated as the pulse-induced growth in the
probabilities of local 4-particle and 2-particle states. In the photo-doping case, it can be measured by the change in the total particle number after the photo-doping (coupling to the fermion bath).

In Fig. 4, the time-dependent density of states and occupied density of states are computed through Fourier transforms over the relative time variables $A(\omega,t) = -\frac{\text{Im} \{ \text{Tr}_{\xi_d} \text{exp}[G(t/t_\delta/2 - t/2)]/\pi \} \text{and} \} \text{Tr}_{\xi_d} \text{exp}[G(t/t_\delta + t - t/2)]$, which are traced over orbitals and spins.

Data availability
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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M.E. and J.L. conceived the project. J.L. has run the DMFT simulations. M.E., H.U.R.S., B.E., J.I.S. and M.E. performed the calculations. All authors contributed to the discussion and the writing of the manuscript.

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