Ultrafast quenching of the exchange interaction in a Mott insulator

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We investigate how fast, and how effective, photo-carrier excitation can modify the exchange interaction \( J_{\text{ex}} \) in the prototype Mott-Hubbard insulator. We demonstrate an ultrafast quenching of \( J_{\text{ex}} \) both by evaluating exchange integrals from a time-dependent response formalism, and by explicitly simulating laser-induced spin precession in an antiferromagnet that is canted by an external magnetic field. In both cases, the electron dynamics is obtained from nonequilibrium dynamical mean-field theory. We find that the modified \( J_{\text{ex}} \) emerges already within a few electron hopping times after the pulse, with a reduction that is comparable to the effect of chemical doping.

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Magnetic long-range order and the dynamics of spins in magnetic materials is governed by the exchange interaction \( J_{\text{ex}} \), the strongest force of magnetism. Because \( J_{\text{ex}} \) emerges from the Pauli principle and the electrostatic Coulomb repulsion between electrons, it is sensitive to purely nonmagnetic perturbations. This fact implies intriguing and largely unexplored possibilities for the ultrafast control of magnetism by femtosecond laser-pulses, which is currently a very active research area [1]. In principle, laser-excitation can effect \( J_{\text{ex}} \) by modulating the electronic structure (electron hopping, Coulomb repulsion) and by creating a nonequilibrium distribution of photo-excited carriers (photo-doping). A modification of \( J_{\text{ex}} \) has been discussed within the context of experiments on manganites [2–4], magnetic semi-conductors [5], and, using static field gradients, ultra-cold atoms in optical lattices [6, 7]. While it might play a role as well in metallic magnetic materials is governed by the exchange interaction, it is sensitive to purely nonmagnetic perturbations. Recently, Secchi et al. defined the nonequilibrium exchange interaction via an effective action that governs the spin dynamics out of equilibrium, leading to an expression in terms of nonequilibrium electronic Green’s functions [16]. Here we apply this framework to the paradigm single band Mott-Hubbard insulator at half-filling, for which the concept of exchange interaction in equilibrium is very well understood. To directly assess the nonequilibrium electron dynamics and evaluate the nonequilibrium Green’s functions, we employ nonequilibrium Dynamical Mean Field Theory (DMFT). Previous investigations of the antiferromagnetic phase in the Hubbard model have demonstrated ultrafast melting of long-range order after an interaction quench [17, 18]. Here, we will focus on the excitation with an electric field pulse and weaker excitation strength, to assess the control of the exchange interaction within the ordered phase, and to determine how fast a rigid spin dynamics emerges after the excitation.

Model — In this work we study the antiferromagnetic phase of repulsive Hubbard model at half-filling,

\[
H = -t_0 \sum_{\langle ij \rangle \sigma} c_{i \sigma}^\dagger c_{j \sigma} + U \sum_i n_{i \uparrow} n_{i \downarrow} + B_x \sum_j S_{j x}.
\]  

Here \( c_{i \sigma}^\dagger \) creates an electron at site \( i \) with spin \( \sigma = \uparrow, \downarrow \) along a given spin quantization axis (the \( z \)-axis). The first two terms describe nearest-neighbor hopping \( t_0 \) and repulsive on-site interaction \( U \). The third term introduces coupling of the spin \( S_{j \sigma} = \frac{1}{2} \sum_{\sigma' \sigma''} c_{j \sigma'}^\dagger \bar{\sigma}_{j \sigma''} c_{j \sigma''} \) to a homogeneous magnetic field \( B_x \) along the \( x \)-axis (\( x = x, y, z; \bar{\sigma}_\alpha \) denote the Pauli matrices). The latter allows us to probe transverse dynamics of the antiferromagnetic order parameter in the \( y-z \) plane; the \( x \)-component of the total spin \( \langle S_x \rangle \) is conserved.

To solve the electron dynamics in the Hubbard model we use nonequilibrium DMFT [19, 20]. Within DMFT [21], which becomes exact in the limit of infinite dimensions [22], local correlation functions are obtained from an effective impurity model in which one site of the lattice is coupled to a non-interacting bath. In the presence of a transverse magnetic field \( B_x \), one must include spin-flip terms in the effective impurity action, which thus takes the form \( \bar{S} = S_{\text{loc}} - \frac{i}{\hbar} \int dt \sum_{\sigma \sigma'} c_{\sigma}^\dagger (t) \Delta_{\sigma \sigma'} (t,t') c_{\sigma'} (t') \). Here \( \Delta_{\sigma \sigma'} (t,t') \) is the hybridization function of the bath, that is determined self-consistently. The impurity model is solved within the perturbative hybridization expansion (non-crossing approximation, NCA), which is reliable in the Mott insulating phase [23]. The incorporation of spin-flip terms \( \Delta_{\uparrow \downarrow} \) is a straightforward extension to the nonequilibrium DMFT implementation and the NCA, which have been explained in Refs. [20] and [23]. For
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Secchi and coworkers \[16\]. The essential idea is to define the dynamics of the full electronic model. A formal derivation of the spin interaction in such a model has been given by Secchi and coworkers \[16\]. The essential idea is to define the effective spin interaction in terms of time-dependent rotations of the spin quantization axes \( \xi_i(t) \), as described by Holstein-Primakov bosons \( \xi_i(t) \). Starting from the electronic partition function as a path integral over fermionic fields \( \phi \), one introduces rotated fermion fields \( \psi \) and then expands the action to second order in \( \xi \). The rotated fermionic fields are integrated out, which leads to spin action with an interaction term of the form \( \mathbb{S}_{\mathrm{spin}}[\xi^i, \xi^j] = \sum_{ij} \int dt \int dt' \xi^i(t) A_{ij}(t, t') \xi^j(t') \). The coupling \( A_{ij}(t, t') \) between spin-rotations at different times and different sites \( i \neq j \) is expressed in terms of the spin-(\( \sigma \))-dependent single-particle Green’s functions \( G_{\sigma}^{\xi}(t, t') \) and the self energies \( \Sigma_{\sigma}^{\xi}(t, t') \).

\[
A_{ij}(t, t') = R_{ij}^x(t, t') R_{ij}^x(t', t) + S_{ij}^x(t, t') S_{ij}^x(t', t) - T_{ij}^x(t, t') G_{\uparrow}^{\xi}(t, t') - G_{\downarrow}^{\xi}(t, t') T_{ij}^x(t, t'),
\]

where \( T_{ij}^x(t, t') = \Sigma_{\sigma}^{\xi}(t, t')-\mathbb{E}\cdot \mathbf{g}_{\sigma}^{\xi}(t, t') \), \( R_{ij}^x(t, t') = \mathbb{E} \cdot \mathbf{g}_{\sigma}^{\xi}(t', t) \), and \( S_{ij}^x(t, t') = (\mathbb{E} \cdot \mathbf{g}_{\sigma}^{\xi}(t, t')) \). These formulas are a direct generalization of the equilibrium formalism \[24, 25\], which is based on variations of the total (free) energy \( \delta E = J_{ex} \theta^2 \) for static spin rotations by a small angle \( \theta \). We emphasize that Eq. (2) is valid for arbitrary fast and/or strong fields, apart from neglecting of vertex corrections \[16\]. In addition, the expressions assume rotations from a collinear state. Reduction of the action with a retarded (two-time) exchange coupling to a spin Hamiltonian with an instantaneous (possibly time-dependent) interaction is possible when the rotations of the quantization axes are much slower than the electron dynamics, and in particular slower than time-dependent fluctuations of the local magnetic moments themselves. Then, we can average over the fast electron dynamics,

\[
J_{ij}(t) = \text{Im} \int_0^\infty ds A_{ij}^{\text{ret}}(t, t - s).
\]

Still, \( J_{ij}(t) \) contains not only the exchange interactions, but also the time-averaged reduction of the local spin by fluctuations. The “bare” exchange interactions between spin vectors \( \langle S_i \rangle \) is finally given by

\[
J_{ij}^{\text{bare}}(t) = \frac{1}{4} \frac{J_{ij}(t)}{\langle S_i \rangle \langle S_j \rangle}.
\]

In the regime where a rigid macrospin model is valid, \( J_{ij}^{\text{bare}} \) should determine the spin dynamics by a Landau-Lifshitz equation. For example, for a canting antiferromagnet on a bipartite lattice in a transverse magnetic field \( B_z \), we can write \( \langle S_i \rangle = -\langle S_i \rangle \times B_{\text{eff}} \), where \( B_{\text{eff}} = -2J_{ex}\langle S_2 \rangle + B_z e_x \). Here \( \langle S_{1,2} \rangle \) correspond to the spin on the two sublattices, and the effective exchange interaction is \( J_{ex} = \sum_{ij} J_{ij}^{\text{bare}} \). Using Neel symmetry \( \langle S_{1z} \rangle = -\langle S_{2z} \rangle, \langle S_{1x} \rangle = +\langle S_{2x} \rangle \) we can thus infer the exchange interaction in the canted geometry from the spin dynamics,

\[
J_{ex}^{\text{canted}} = \frac{B_z}{4\langle S_1 \rangle} + \frac{1}{4\langle S_1 \rangle} \langle S_{1x} \rangle \langle S_{1z} \rangle.
\]

The validity of the instantaneous approximation is a fundamental question that is not resolved in general, and which will be partially addressed below by comparison of the two expressions (4) and (5).

**Results** — We first solve the DMFT equations on the Bethe lattice with a semi-elliptic density of states \( D(\epsilon) = \sqrt{4 - \epsilon^2}/2\pi \). This setup implies a closed-form self-consistency condition and thus allows us to compute the electronic dynamics to long times, as needed for an accurate evaluation of the integral in Eq. (3). In fact, to evaluate the exchange formulas (2) one typically needs a twice smaller step size in time than to evaluate the Green’s functions themselves, because we obtain the self-energy by computing numerical derivates of the Green’s functions (see supplementary material).

Before exploring nonequilibrium, it is illustrative to evaluate the exchange interaction (4) in the familiar equilibrium case. For the Mott-insulator at half-filling, the static exchange interaction at zero temperature can be obtained from a perturbation expansion in the hopping, which yields \( |J_{ex}^{\text{bare}}| = 2t^2/U \). In Figure 1 we compare the analytical value \( |J_{ex}^{\text{bare}}| \) (dashed lines) and the bare exchange interaction \( |J_{ex}^{\text{bare}}| = |J_{ex}^{\text{canted}}| \) computed from the collinear DMFT solution using Eq. (4) (red circles) as function of temperature for three values of \( U \). In addition, we solve the DMFT equations for the antiferromagnetic Mott-insulator in a weak transverse field of strength \( B_z \), and obtain an estimate \( |J_{ex}^{\text{canted}}| = B_z/4\langle S_x \rangle \) by comparing the canting \( \langle S_x \rangle \) of spins to the prediction from a rigid macrospin
model Eq. 5 in the static limit (blue solid discs). We choose $B_t = 0.64 t_0^2 / U$, such that the deviation from the collinear geometry at low temperature is about 10 degrees for all $U$. At large $U$ we find excellent agreement between $J_{\text{ex}}^0$, $J_{\text{ex}}$ and $J_{\text{ff}}$, where deviations between $J_{\text{ex}}$ and $J_{\text{ff}}$ are in the order of $(t / U)^2$. For smaller $U$, the deviation of $J_{\text{ex}}^0$ from $J_{\text{ex}}$ becomes more pronounced, up to 25% at $U = 4$. The differences between the two expressions 4 and 5 may have several possible origins: (i) At small values of $U$ the rigid macrospin model is no longer valid, because retardation effects in $A(t, t')$ become relevant, (ii), vertex corrections to Eq. 2 become important, or (iii), $J_{\text{ex}}^0$ is a nearest neighbor interaction while Eq. 5 also takes into account next-nearest neighbor terms. Below we will study nonequilibrium exchange at large values of $U$. Nevertheless, for moderate $U$, where retardation effects to the exchange become important, we can still use Eq. 5 as a heuristic measure for $J_{\text{ex}}$, in the sense that it is the best estimate of an instantaneous exchange interaction which is in accordance with an observed spin dynamics.

Next, we generate nonequilibrium electron dynamics by suddenly changing $U$. It has been demonstrated recently that after an interaction quench the order parameter $m$ quickly relaxes to a quasi-stationary but non-thermal stationary [17] that is protected from further decay by the slow recombination rate of doublons and holes [24–29]. This transient state thus resembles properties of a photo-doped system in which charge carriers are created by a short laser pulse. We will refer to the quench-induced change of the doublon and hole densities $d$ and $h$ with respect to their equilibrium values $d_0$ and $h_0$ as photo-doping $\Delta U = d + h - d_0 - h_0 = 2(d_0 - h_0)$. The inset of Figure 2 shows the evolution of the time-dependent nonequilibrium exchange interaction (solid line) and order parameter (dashed line) for a quench $U = 4 \rightarrow 8$. (A Gaussian window $\exp(-t^2/\omega^2)$ of length $\omega = 10 t_0 / \pi$ was used in Eq. 5 to ensure a smooth cut-off of the upper integration limit.) We find that $J_{\text{ex}}^0$, like $m$, becomes stationary already on an electronic timescale, which in this case is a few tens of the inverse hopping.

To study how effective the exchange interaction is modified, we evaluate it in the quasi-stationary state after different excitation strengths $\Delta U = U_t - U_s = 0 . . . 4$, with final $U_t = 8$. The result is shown by red open circles in Figure 2 as a function of “photo-doping” $\Delta U$, demonstrating a reduction of the exchange interaction to a value significantly below the equilibrium difference $J_{\text{ex}}^0(U_t) - J_{\text{ex}}^0(U_s)$. The results are independent of a Gaussian cut-off in Eq. 5 for $w = 60 t_0 / \pi$. Only for the largest $\Delta U$ we find a slight dependence on $w$ which indicates that $J_{\text{ex}}^0$ is not yet fully stationary. Further, blue lines in Fig. 2 show the equilibrium exchange interaction at chemical doping for different temperatures. These results confirm the conclusions obtained from analyzing the electronic spectrum [17], that properties of the photo-doped state with added doublons and holes resemble those of the chemically doped state with the same total number of carriers: Adding doublons and holes causes an ultrafast quenching of the exchange interaction by an amount comparable to that of chemical doping. The weakening of the antiferromagnetic exchange can qualitatively understood in the limit of large $U$, where in equilibrium a weak deviation from half-filling favors a ferromagnetic state [50].

**Photo-excitation** To further demonstrate the possibility of changing the exchange interaction in a setup that is closer the laser excitation of condensed matter systems, we study the Hubbard model driven by an external electric field. This is implemented for the infinite-dimensional hyper-cubic lattice with density of states $I(e) = \exp(-e^2) / \sqrt{\pi}$, with the electric field pointing along the body diagonal [20, 31]. Photo-excited carriers are created by a single-cycle pulse $E(t) = E_0 \sin(\omega t) \exp(-\alpha(t-t_c)^2)$, $t_c = \pi / \omega, \alpha = 4.6 / t_c^2$ with a Gaussian envelope and a centre frequency of $\omega = U$. To measure directly the transverse spin dynamics associated with the exchange interaction, we study the system in a canted geometry created by a homogeneous magnetic field $B_z$. Before laser excitation, the system is prepared in equilibrium with a canting angle determined by the balance of the external field and the exchange interaction. When the exchange interaction is changed, this balance will be broken and a spin resonance will be excited. Such spin resonances can in principle be detected experimentally using magneto-optical techniques [1]. In our simulations, we extract the nonequilibrium exchange interaction by comparing the spin dynamics obtained within DMFT to the rigid macrospin model, cf. Eq. 5. The results of this approach are shown in Figure 3 computed at $U = 8$ and $B_z = 0.01$. The top panel shows that the sublattice magnetization is initially in the $x - z$ plane. Light to dark colors indicate excitation strengths ranging from $|E_0|/t_0 = 1 . . . 5.5$. The bottom panel shows $\Delta J_{\text{ex}}^0 \sim \langle S_{1y} \rangle / \langle S_{1z} \rangle$, cf. Eq. 5, where $\langle S_{1y} \rangle$ is computed from the time-trace of $\langle S_{1y}(t) \rangle$. We observe three different time scales in our simulations: (i) fast $1/U$ oscillations on the timescale of the laser excitation, as
is the numerical accuracy. The values indicated as dots in the bottom panel of Fig. 3. Similar as for the excitation density, as the critical excitation for melting the antiferromagnetic order is approached. Furthermore, we find the set of rigid rotation of the spin sublattices at quasi-stationary stabilization of the local magnetic moments. (ii) Relaxation of the exchange interaction in the chemically doped system (blue lines).

FIG. 3: (Color online) Induced spin dynamics (top) and modification of the exchange interaction (bottom) caused by excitation with an electric field (hypercubic lattice, \( U = 8 \)).

FIG. 4: (Color online) Comparison of the nonequilibrium exchange interaction (red open circles) computed from the induced precession (Fig. 3), with the equilibrium exchange interaction in the chemically doped system (blue lines).

most clearly seen in the bottom panel. This characterizes the stabilization of the local magnetic moments. (ii) Relaxation of the order parameter and the exchange interaction. (iii) The onset of rigid rotation of the spin sublattices at quasi-stationary values \( |\langle S_1 \rangle| \) and \( J_{ex} \). We estimate the time \( t_e \) that it takes for \( J_{ex}^f \) to become stationary from \( J_{ex}^f(t_e) - J_{ex}(t_{max}) < \varepsilon \), where \( \varepsilon \) is the numerical accuracy. The values \( t_e \) thus obtained are indicated as dots in the bottom panel of Fig. 3. Similar as for the sudden change of \( U \), a quasi-stationary state is formed after a few tens of the hopping. This relaxation time increases with the excitation density, as the critical excitation for melting the antiferromagnetic order is approached. Furthermore, we find that direct photo-excitation has a similar effect as the interaction quench, i.e. the efficiency of the modification of the exchange interaction is determined by the number of photo-excited carriers. This is demonstrated in Fig. 4 by plotting the extracted exchange interaction in the quasi-stationary state as a function of the photo doping, together with equilibrium calculations in the canted geometry with chemical doping. In the hyper cubic lattice, we observe that photo excitation modifies \( J_{ex}^f \) slightly stronger than chemical doping. In addition, there is a more pronounced temperature dependence of \( J_{ex}^f \) in equilibrium. Both effects might be related to a slightly different dynamics of low energy (photo) doped carriers in the Bethe lattice and the hypercubic lattice, where the latter does not have a sharp band edge in the density of states.

In summary, we report that the photo-excitation causes an ultrafast quenching of the exchange interaction. An effectively static exchange interaction can be defined already on the ultrafast time-scale in the order of a few tens inverse hopping times, which is similar to the relaxation time of the order parameter. The reduction of the exchange interaction is comparable to that of a chemically doped state when measured in terms of the total number of excited carriers. These results demonstrate intriguing possibilities to control magnetic order without magnetic fields and can be of direct relevance to experiments on prototype Mott-insulators such as \( V_2O_3 \). Similar, or even more efficient ways to control \( J_{ex} \) under nonequilibrium conditions might be found by extending our work to more complex multi-band systems with different exchange mechanisms.

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Supplementary material

Implementation of nonequilibrium DMFT with a transverse magnetic field

In the supplementary material we describe in detail how we implement the nonequilibrium DMFT for the antiferromagnetic phase of the Hubbard model in a transverse magnetic field,

\[
H = - \sum_{(ij)\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} + B_s \sum_j S_{jx} - \mu \sum_{j\sigma} n_{j\sigma}.
\]

Apart from the incorporation of spin-flip terms in the DMFT impurity action and the self-consistency relations, the resulting equations are a straightforward extension of the equations for the paramagnetic phase and the collinear antiferromagnet, which have been described previously [20, 23].
The impurity model

To describe a magnetically ordered system, we introduce Keldysh Green’s functions which are $2 \times 2$ matrices in spin-space,

$$
\hat{G}_{J}(t, t') = -i\langle TC\hat{\psi}_J(t)\hat{\psi}^\dagger_J(t)S \rangle 
$$

(7)

$$
\approx (\langle -i\langle TC c_{J\uparrow}(t) c_{J\uparrow}(t)S \rangle - i\langle TC c_{J\downarrow}(t) c_{J\downarrow}(t)S \rangle ) \quad (8)
$$

Here $\hat{\psi}_J$ is the spinor

$$
\hat{\psi}_J = \left( \begin{array}{c} c_{J\uparrow} \\ c_{J\downarrow} \end{array} \right),
$$

(9)

$C$ is the $L$-shaped Keldysh contour that extends from 0 to some maximum time $t_{\text{max}}$ along the real axis, back to 0, and to $-i\beta$ along the imaginary time axis, and

$$
\langle TC \cdots S \rangle \equiv \text{tr} [TC e^{S} \cdots] / \text{tr} [TC e^{S}]
$$

(10)

denotes the contour-ordered expectation value for an action $S$; the action for the lattice model (6) is given by $S = -i \int_0^\beta DT \hat{H}(t)$. We follow Ref. [20] for the notation for Keldysh Green’s and their convolution and time-derivatives along $C$.

The antiferromagnetic DMFT solution is obtained on a bipartite lattice at and close to half-filling. The local Green’s function $\hat{G}_\alpha$ for a site on sub-lattice $\alpha = A, B$ of the bipartite lattice is obtained from an impurity model with action

$$
\hat{S}_\alpha = -i \int_C dt H_{bc,\alpha}(t) - i \int_C dt d\tau \hat{\psi}^\dagger(t)\Delta_\alpha(t, \tau)\hat{\psi}(\tau),
$$

(11)

where $H_{bc,\alpha}(t)$ is the local part of Hamiltonian (6), and $\Delta_\alpha(t, \tau)$ is the hybridization matrix, which is later determined self-consistently.

In order to compute the impurity Green’s function

$$
\hat{G}_\alpha(t, t') = -i\langle TC\hat{\psi}_\alpha(t)\hat{\psi}^\dagger_\alpha(t)\rangle_{\hat{S}_\alpha},
$$

(12)

we use the lowest strong-coupling impurity solver [23] (non-crossing approximation, NCA), which is a self-consistent expansion in the hybridization function $\Delta_\alpha(t, \tau)$. The hybridization expansion can be formulated in terms of pseudo-particle propagators $G_{nm}$, whose flavor indices $n, m$ correspond to the many-body states of the impurity. These propagators satisfy a Dyson equation, with a self-energy $\Xi$ that is given by a diagrammatic expansion in the hybridization function. In the present case, a basis of the local Hilbert space at the impurity model is given by the four states $|0\rangle, |\sigma\rangle \equiv c_{\uparrow \downarrow}$ (for $\sigma = \pm$), and $|2\rangle \equiv c_{\uparrow \downarrow}^\dagger c_{\uparrow \downarrow}$. Because the $H_{bc}$ and $\Delta_\alpha(t, \tau)$ allow spin-flip terms, pseudo-particle propagators $G_{nm}$ are only diagonal in particle number of $|n\rangle$ and $|m\rangle$, but not in spin. Hence, we introduce three propagators, $G^{(0)} = \langle 0|0\rangle$, $G^{(1)} = \langle 0|1\rangle$, and $G^{(2)} = \langle 2|2\rangle$, and $G^{(1)}_{\sigma\sigma'} = G^{(1)}_{|\sigma\rangle,|\sigma'|\rangle}$, and corresponding self-energies $\Xi^{(0)}(t, t')$, $n = 0, 1, 2$. From the diagrammatic rules for a general multi-orbital case as stated in Ref. [23], we obtain

$$
\Xi^{(0)}(t, t') = -i \sum_{\sigma\sigma'} G^{(1)}_{\sigma\sigma'}(t, t') \Delta_{\sigma\sigma'}(t', t)
$$

(13)

$$
\Xi^{(1)}_{\sigma\sigma'}(t, t') = i G^{(0)}(t, t') \Delta_{\sigma\sigma'}(t', t) - i \bar{\sigma} \bar{\sigma'} G^{(2)}(t, t') \Delta_{\sigma\sigma'}(t', t).
$$

(14)

Finally, the local Green’s function is given by evaluation of the “bubble diagram” [23]

$$
G_{\sigma\sigma'}(t, t') = i G^{(0)}(t, t') G^{(1)}_{\sigma\sigma'}(t, t') - i \sigma \sigma' G^{(2)}(t, t') G^{(2)}(t, t').
$$

(16)

DMFT self-consistency for the Bethe lattice

For a Bethe lattice with nearest neighbor hopping $t_{ij} \equiv t_0/\sqrt{Z}$ in the limit $Z \to \infty$, which has a semi-elliptic density of states $D(\epsilon) = \sqrt{4 - \epsilon^2/2\pi}$ for $t_0 = 1$, the hybridization function is determined by the closed form self-consistency equation [21]

$$
\hat{\Delta}_\alpha(t, \tau) = \tilde{\gamma}_0 \hat{G}_\alpha(t, \tau),
$$

(17)

where $\tilde{\gamma}_0 = B(A)$ for $\alpha = A(B)$. The NCA equations together with Eq. (17) provide a closed set of equations that is numerically propagated in time as described in Ref. [20].

DMFT self-consistency for the hypercubic lattice

To solve the DMFT equation on a cubic lattice with $A/B$ sub-lattice symmetry breaking, we let $L'$ denote the magnetic superlattice of points $R$, and $L$ is the full lattice with atoms at coordinates $r_{ja} = R_{ja} + \delta_a$, $a = A, B$. For example, we can choose $L'$ as the A-sublattice of the bipartite cubic lattice, such that $\delta_A = 0, \delta_B = (1, 0, ...)$. We then introduce the Fourier transform with respect to the coordinate $R_{ja}$,

$$
\tilde{c}_{j\sigma\tau} = \frac{1}{\sqrt{L'}} \sum_{\vec{R} \in L'} \tilde{c}_{\vec{R} + j\sigma\tau} e^{-iK(R_{ja} + \delta_{a\sigma\tau})},
$$

(18)

$$
\tilde{c}_{j\sigma\tau} = \frac{1}{\sqrt{L'}} \sum_{\vec{R} \in B'} \tilde{c}_{\vec{R} + j\sigma\tau} e^{-iK(R_{ja} + \delta_{a\sigma\tau})},
$$

(19)

where $L'$ is the number points in $L'$, and $B'$ is the first Brillouin zone of the magnetic superlattice. To describe the broken symmetry phase, we introduce super-spinors

$$
\tilde{\Psi}_K = \left( \begin{array}{c} \tilde{c}_{K, A, \uparrow} \\ \tilde{c}_{K, A, \downarrow} \\ \tilde{c}_{K, B, \uparrow} \\ \tilde{c}_{K, B, \downarrow} \end{array} \right) \equiv (\tilde{\psi}_{K, A} \begin{array}{c} \tilde{\psi}_{K, A} \\ \tilde{\psi}_{K, B} \end{array}),
$$

(20)
and corresponding Green’s functions
\[
\hat{G}_K(t) = -i \langle \tau_C \hat{\Psi}_K(t) \hat{\Psi}_K(t') \rangle \equiv \begin{pmatrix} \hat{G}_{K,AA}(t, t') & \hat{G}_{K,AB}(t, t') \\ \hat{G}_{K,BA}(t, t') & \hat{G}_{K,BB}(t, t') \end{pmatrix},
\]
where the second expression is a block-matrix with entries \(\hat{G}_{K,\alpha\beta}(t, t') = -i \langle \tau_C \hat{\Psi}_{\alpha}(t) \hat{\Psi}_{\beta}^\dagger(t') \rangle\). With this, the quadratic part of the Hamiltonian \(\hat{V}\) can be rewritten as
\[
\sum_{K \in B'} \hat{V}_K \hat{\Psi}_K \hat{\Psi}_K,
\]
with
\[
\hat{H}_K = \begin{pmatrix} \hat{H}_{loc,A} & \hat{\epsilon}_K \\ \hat{\epsilon}_K & \hat{H}_{loc,B} \end{pmatrix},
\]
where \(\hat{H}_{loc,A} = \hat{H}_{loc,B} = \hat{\epsilon}_K B\), and \(\hat{\epsilon}_K = \epsilon_K \hat{1}\), with the 2 \times 2 unit matrix \(\hat{1}\). The electronic dispersion \(\epsilon_K\) may be time-dependent due to inclusion of a external electric field via the Peierls substitution (see below). The Dyson equation has a 4 \times 4-structure,
\[
\hat{G}_K^{-1}(t, t') = \delta_\Omega(t, t')[i\partial_t + \mu] \hat{1} - \hat{H}_K - \hat{\Sigma}(t, t'),
\]
with the spatially local self energy
\[
\hat{\Sigma}(t, t') = \begin{pmatrix} \hat{\Sigma}_A(t, t') & 0 \\ 0 & \hat{\Sigma}_B(t, t') \end{pmatrix},
\]
\[
\hat{\Sigma}_\alpha(t, t') = \begin{pmatrix} \Sigma_{\alpha,\uparrow\uparrow}(t, t') & \Sigma_{\alpha,\uparrow\downarrow}(t, t') \\ \Sigma_{\alpha,\downarrow\uparrow}(t, t') & \Sigma_{\alpha,\downarrow\downarrow}(t, t') \end{pmatrix}.
\]
Numerically, it is convenient to solve the DMFT equations without explicitly solving for the self energy. By introducing \(\hat{Z}_\alpha = [i\partial_t + \mu - \hat{H}_{loc,\alpha} - \hat{\Sigma}_\alpha]^{-1}\), the impurity Dyson equation reads
\[
\hat{G}_\alpha = \hat{Z}_\alpha + \hat{Z}_\alpha \hat{\Delta}_\alpha \hat{G}_\alpha,
\]
and the lattice Dyson equation is given by
\[
\hat{G}_K = \begin{pmatrix} \hat{Z}_A & 0 \\ 0 & \hat{Z}_B \end{pmatrix} + \begin{pmatrix} \hat{Z}_A & 0 \\ 0 & \hat{Z}_B \end{pmatrix} \hat{\epsilon}_K \begin{pmatrix} 0 \\ \hat{\epsilon}_K \end{pmatrix} \hat{G}_K.
\]
The lattice Dyson equation can be written explicitly for its four 2 \times 2 components,
\[
\hat{G}_{K,AA} = \hat{Z}_A + \hat{\Delta}_A \hat{G}_{K,AA},
\]
\[
\hat{G}_{K,AB} = \hat{Z}_A + \hat{\Delta}_A \hat{G}_{K,AB},
\]
\[
\hat{G}_{K,BA} = \hat{Z}_B + \hat{\Delta}_B \hat{G}_{K,BA},
\]
\[
\hat{G}_{K,BB} = \hat{Z}_B + \hat{\Delta}_B \hat{G}_{K,BB}.
\]
(where we have reinserted the expressions for \(\hat{G}_{K,AB}\) and \(\hat{G}_{K,BA}\) into the equations for \(\hat{G}_{K,AA}\) and \(\hat{G}_{K,BB}\).) By summing these equations over \(K\) and comparing with the impurity Dyson equation, we then obtain an explicit equation for the hybridization function (for \(\alpha = A, B\)). For this it is convenient to introduce the moments
\[
\hat{G}_\alpha = \frac{1}{L} \sum_{K \in B'} \hat{G}_{K,\alpha\alpha},
\]
\[
\hat{G}_\alpha^{(1)} = \frac{1}{L} \sum_{K \in B'} \hat{\epsilon}_K * \hat{Z}_\alpha * \hat{G}_{K,\alpha\alpha},
\]
\[
\hat{G}_\alpha^{(2)} = \frac{1}{L} \sum_{K \in B'} \hat{\epsilon}_K * \hat{Z}_\alpha * \hat{\Delta}_\alpha * \hat{G}_{K,\alpha\alpha} * \hat{\epsilon}_K * \hat{Z}_\alpha * \hat{\epsilon}_K
\]
\[
\hat{\Delta}_\alpha + \hat{\Delta}_\alpha * \hat{\Delta}_\alpha = \hat{G}_\alpha^{(2)},
\]
from which the hybridization can be determined, thus closing the DMFT self-consistency.

Throughout this work we consider magnetic fields along \(x\), perpendicular to the antiferromagnetic order parameter. In this case, the system is invariant under a translation by one lattice constant and spin rotation by \(\pi\) around the axis of the \(B\)-field. This symmetry can be used to relate local quantities at the \(A\) and \(B\) sites, i.e.,
\[
\hat{\Sigma}_B = \hat{\sigma}_x \hat{\Sigma}_A \hat{\sigma}_x,
\]
and analogous for the functions \(\hat{Z}_B\), \(\hat{G}_B\), and \(\hat{\Delta}_B\). Explicitly,
\[
\begin{pmatrix} \Sigma_{B,\uparrow\uparrow}(t, t') & \Sigma_{B,\uparrow\downarrow}(t, t') \\ \Sigma_{B,\downarrow\uparrow}(t, t') & \Sigma_{B,\downarrow\downarrow}(t, t') \end{pmatrix} = \begin{pmatrix} \Sigma_{A,\uparrow\uparrow}(t, t') & \Sigma_{A,\uparrow\downarrow}(t, t') \\ \Sigma_{A,\downarrow\uparrow}(t, t') & \Sigma_{A,\downarrow\downarrow}(t, t') \end{pmatrix}.
\]
This symmetry leads to a considerable reduction of the numerical complexity, because one can make the 4 \times 4 Dyson equation \(2 \times 2\) block-diagonal with the basis change
\[
\hat{V} = \frac{1}{\sqrt{2}} \begin{pmatrix} \hat{1} & \hat{\sigma}_x \\ \hat{\sigma}_x & -\hat{1} \end{pmatrix}.
\]
The symmetry \(\hat{V}\) implies
\[
\hat{V} \begin{pmatrix} \hat{Z}_A & 0 \\ 0 & \hat{Z}_B \end{pmatrix} \hat{V}^\dagger = \begin{pmatrix} \hat{Z}_A & 0 \\ 0 & \hat{Z}_B \end{pmatrix},
\]
and we have
\[
\hat{V} \begin{pmatrix} \hat{\epsilon}_K & 0 \\ 0 & \hat{\epsilon}_K \end{pmatrix} \hat{V}^\dagger = \begin{pmatrix} \hat{\epsilon}_K \hat{\sigma}_x & 0 \\ 0 & -\hat{\epsilon}_K \hat{\sigma}_x \end{pmatrix}.
\]
Thus the Dyson equation for the transformed 4 \times 4 Green’s functions
\[
\hat{G}_K(t, t') = \hat{V} \hat{G}_K(t, t') \hat{V}^\dagger.
\]
is block-diagonal: When we introduce the notation
\[ \tilde{G}_K = \begin{pmatrix} \tilde{G}_K^+ & 0 \\ 0 & \tilde{G}_K^- \end{pmatrix}, \]
the two blocks are obtained by solving two Dyson equations,
\[ \tilde{Z}_A + \tilde{Z}_A \ast \epsilon_K \tilde{\sigma}_z \ast \tilde{G}_K^+ = \tilde{G}_K^+, \]
\[ \tilde{Z}_A - \tilde{Z}_A \ast \epsilon_K \tilde{\sigma}_z \ast \tilde{G}_K^- = \tilde{G}_K^-, \]
where we have again used the symmetry \( \tilde{Z}_B = \tilde{\sigma}_z \tilde{Z}_A \tilde{\sigma}_z \) in the second equation. The back-transformation to the original basis, \( \tilde{G}_K = \tilde{V}^i \tilde{G}_K \tilde{V}, \) gives
\[ \tilde{G}_K = \frac{1}{2} \left( \tilde{G}_K^+ + \tilde{G}_K^- \right) \left( \tilde{\sigma}_x (\tilde{G}_K^+ - \tilde{G}_K^-) \tilde{\sigma}_z \right). \]  
(48)
The \( K \)-summed quantities \( (33), (34), \) and \( (36) \) are thus obtained as
\[ \tilde{G}_A = \frac{1}{2L^2} \sum_{K \in B^2} \tilde{G}_K^+ + \tilde{G}_K^-, \]
\[ \tilde{G}_A^{(1)} = \frac{1}{2L^2} \sum_{K \in B^2} \epsilon_K \tilde{\sigma}_x \ast (\tilde{G}_K^+ - \tilde{G}_K^-), \]
\[ \tilde{G}_A^{(2)} = \frac{1}{2L^2} \sum_{K \in B^2} \epsilon_K \tilde{\sigma}_x \ast (\tilde{G}_K^+ + \tilde{G}_K^-) \ast \tilde{\sigma}_z \epsilon_K, \]
Here we have used Eqs. \( (31) \) and \( (32) \) in Eqs. \( (33), (34), \) and \( (36), \) and then replaced \( \tilde{G}_{K_{x,y}} \) by the explicit expressions obtained from Eq. \( (48). \) Because all convolutions involve the time-local functions \( \epsilon_K, \) they are evaluated without numerical cost.

The final set of DMFT equations, to be solved successively timestep after timestep, is thus given by: (i) Solve one impurity model (the one on the \( A \)-lattice, i.e., compute \( \tilde{G}_A \) [Eq. \( (12) \)] from the action \( (11) \) with hybridization \( \lambda_\Delta \). (ii) Solve Eq. \( (27) \) for \( \tilde{Z}_A \). (iii) Solve two equations \( (46) \) and \( (47) \) for \( \tilde{G}_K^+ \) and \( \tilde{G}_K^- \). (iv) Evaluate the sums Eq. \( (49) \) to \( (51) \). (v) Compute the new hybridization function \( \lambda_\Delta \) from Eq. \( (38) \).

Finally, the summation over \( K \) is reduced to an integral over the density of states, as described in Ref. \( [31]. \) We consider a cubic lattice with pure nearest neighbor hopping, and an electric field \( E(t) = E(t)(1, 1, 1, \ldots) \) which is pointing along the body-diagonal of the unit cell. Then
\[ \epsilon_K = \frac{-2 \pi}{\sqrt{2d}} \sum_{\alpha=1}^{d} \cos(k_\alpha - A(t)), \]
\[ = \cos(A(t)) \epsilon_K^0 + \sin(A(t)) \epsilon_K^0, \]
(52)
with the density of states for the reduced zone
\[ D'(\epsilon, \tilde{\epsilon}) = \frac{1}{L'} \sum_{K \in B'} \delta(\epsilon - \epsilon_K^0) \delta(\tilde{\epsilon} - \tilde{\epsilon}_K^0). \]  
(54)
Because all points in the full BZ \( B \) can be obtained by \( \{ K, K + Q \} \) with \( K \in B' \) and \( Q = (\pi, \pi, \ldots) \), and because \( \epsilon_K^0 + \epsilon_Q^0 = -\epsilon_K^0 \), we can choose the reduced BZ \( B' \) as all \( K \) with \( \epsilon_K^0 < 0 \). Hence we have
\[ D'(\epsilon, \tilde{\epsilon}) = 2\theta(-\epsilon)D(\epsilon, \tilde{\epsilon}), \]
(55)
where \( D(\epsilon, \tilde{\epsilon}) \) is the density of states for the full BZ. We will work in the limit of infinite dimensions, with \( D(\epsilon, \tilde{\epsilon}) = e^{-\epsilon^2 \pi^2} \).

We close with the remark that the DMFT equations conserve the total spin along the direction of \( B \). The magnetic field \( B \) thus determines only the time-independent expectation value of the initial field, while any time-dependence of a homogeneous magnetic field implies a trivial time-dependent rotation of the Green’s functions in spin space.

Evaluation the exchange formulas

In this section we describe how we evaluate numerically the exchange interactions [Eq. \( (2) \) of the main text] within DMFT. In contrast to the evaluation of the DMFT self-consistency described above, this requires an explicit knowledge of the self energy. Below we discuss how the self energy \( \Sigma(t, t') \) is evaluated using numerical derivatives. Once \( \Sigma(t, t') \) is computed, the exchange interactions are evaluated by making the appropriate products and convolutions.

Within NCA, the self-consistent solution of the impurity model gives us direct access to the local Green function \( G_i(t, t') \) and the hybridization function \( \Delta_i(t, t') \) (where possible we omit the spin index \( \sigma \)). \( \Sigma_i(t, t') \) is related to \( G_i(t, t') \) and \( \Delta_i(t, t') \) by the impurity Dyson equation:
\[ i\partial_t G_i - [\Delta_i G_i] = 1 + [\Sigma_i G_i], \]
(56)
where \( \Delta \) indicates the delta function \( \delta_\Delta(t, t') \) on the Keldysh contour. To be able to handle the equal-time discontinuities of the Green’s functions and self energies analytically, we write the self-energy as \( \Sigma_i = \Sigma^{i}_{1} + \Sigma^{i}_{2} \). Here \( \Sigma(t, t') = \delta_\Delta(t, t') \Sigma^{i}_{2}(t) \) is the Hartree component of the self-energy and \( \Sigma^{i}_{1}(t, t') \) is the part of the self-energy which is finite at \( t = t' \).

The Hartree component is computed by invoking a first numerical derivative, indicated by \( \delta_\Sigma \), which is evaluated only for \( t = t' \). Denoting \( F_i = \Sigma^{i}_{1} G_i \) we write the Dyson equation as:
\[ F_i = i\partial_t G_i - 1 - \Delta_i * G_i \]
\[ = (i\delta_\Sigma G_i + 1) - 1 - \Delta_i * G_i = i\delta_\Sigma G_i - \Delta_i * G_i, \]
\( \Sigma^{i}_{1}(t) \) now follows directly from the equal time contribution \( F_i(t, t) \). On the real-time axis we have
\[ F_{i}^{1+}(t, t) - F_{i}^{1-}(t, t) = -i \Sigma^{i}_{1}(t), \]
(58)
where we used that $G^\tau(t,t) - G^\tau(t,t) = -i$. On the Matsubara axis we have equivalently $F^\tau_i(0) + F^\tau_i(\beta) = -\Sigma_i^H(0)$.

To compute the component $\Sigma_i'(t,t')$ we invoke a second derivative and write the local $T$ matrix as:

$$T_i = i\partial_t F_i^\dagger - \Delta_i * F_i^\dagger = [i\partial_t G_i - \Delta_i * G_i] * \Sigma_i$$

$$= [i\partial_\beta G_i + \Delta_i * G_i] * \Sigma_i$$

$$= i\partial_\beta G_i * \Sigma_i + \partial_\beta G_i * \Sigma_i$$

(59)

In addition, we have

$$T_i = [i\partial_t G_i - \Delta_i * G_i] * \Sigma_i = \Sigma_i + \partial_\beta G_i * \Sigma_i$$

$$= \Sigma_i + \partial_\beta G_i * \Sigma_i$$

(60)

By subtracting Eq. (59) and Eq. (60) we obtain:

$$\Sigma_i + \Sigma_i = \partial_\beta G_i * \Sigma_i$$

(61)

Finally, the self-energy $\Sigma_i'$ is evaluated by inverting Eq. (61). This integral equation corresponds to a Volterra equation of the second kind, which is numerically well conditioned.