Non-equilibrium spin-spin interactions in strongly correlated systems

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Abstract We develop a theory for magnetism of strongly correlated systems driven out of equilibrium by an external time-dependent electric field. We provide expressions for computing the effective interaction parameters between electronic spins, including a new interaction that we name *twist exchange*. Our theory is suitable for laser-induced ultrafast magnetization dynamics.

Introduction and Method

The theoretical study of magnetization in realistic systems is a challenging problem. In equilibrium, magnetic interactions in magnetic metals and semiconductors are non-Heisenberg [1-4], i.e., the lengths of magnetic moments and values of exchange parameters depend on the magnetic configuration for which they are computed, which calls for an accurate *ab initio* formulation. The expressions for the exchange-interaction parameters were given years ago [5, 6], and they can be used within a classical Heisenberg model to simulate spin dynamics. However, this treatment is not expected to be appropriate for ultrafast dynamics [7], since the time scale of the excitations is faster than the typical scale of exchange interactions (10÷100 fs). Exchange parameters are thus time-dependent. Our purpose is to derive their expressions in a general non-equilibrium framework, including external time-dependent fields [8]. The resulting formulas are valid for any time scale, thus they are applicable to ultrafast spin dynamics.

We consider a system described by the time-dependent multi-band Hubbard Hamiltonian,

$$\hat{H}(t) \equiv \sum_{a}\sum_{b}\sum_{\sigma=\uparrow,\downarrow}\hat{\phi}^{+}_{a\sigma}T_{ab}(t)\hat{\phi}_{b\sigma} + \hat{H}_V , \qquad 1$$

where italic letters a, b denote sets of site and orbital indexes; $\hat{H}_V$ accounts for on-site interactions between the electrons. The matrix element $T_{ab}(t)$ accounts for in-

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ter-site hopping and orbital hybridization due to both the equilibrium structure and an external time-dependent electric field, switched on at time t_0. Here we do not consider relativistic effects nor external magnetic fields, so $T_{ab}(t)$ is spin-independent.

We write the partition function for the electronic system in the Kadanoff-Baym formalism and we derive an effective action that describes the system in equilibrium for $t \le t_0$, as well as out of equilibrium for $t > t_0$. We assume spontaneous symmetry breaking (SSB) along the direction z, specified by the unit vector $\vec{u}_z$, and we study the spin excitations on the top of the collinear equilibrium ground state. Thus, we apply time- and site- dependent Holstein-Primakoff spin rotations to map the old fermionic fields $\phi_{a\sigma}$ into new fields $\psi_{a\sigma}$ having their spins aligned with t-dependent vectors $\vec{e}_a(t)$, which are the unit vectors of the (classical) magnetic moments. The deviations of the $\vec{e}_a(t)$'s from $\vec{u}_z$ are described by auxiliary bosonic fields; in the low-energy sector we assume such deviations to be small, obtaining an action quadratic in the bosons. Integrating out the fermionic fields $\psi_{a\sigma}$ gives an effective bosonic action, and we finally map the bosons to the $\vec{e}_a(t)$'s to obtain the spin-spin interactions.

Results

Our results are presented in Ref.[8]. Here we summarize some of the main points.

We find that there are two forms of quadratic interactions between magnetic moments. The first form, $\propto \vec{e}_a \cdot \vec{e}_b$, is exchange. The second form, $\propto \left(\vec{e}_a \times \vec{e}_b\right) \cdot \vec{u}_z$, looks similar to an effective Dzyaloshinskii-Moriya interaction (DMI), but this interpretation is not correct since relativistic effects are absent in our model. We note, instead, that an *effective three-body* interaction of the form $\left(\vec{e}_a \times \vec{e}_b\right) \cdot \vec{e}_c$ generates precisely this term when up to second-order deviations of the magnetic moments from $\vec{u}_z$ are considered. We have proposed the name *twist exchange* for this new interaction [9]. Our method provides both the equilibrium and the non-equilibrium formulas for the coefficients describing exchange and twist-exchange, which are obtained after the fermionic field integration as convolutions of electronic Green functions and self-energies. These can be computed by the means of non-equilibrium dynamical mean-field theory. The formulas for exchange parameters reproduce, in the particular case of the single-band Hubbard model in equilibrium, the results of Ref.[6].

We find that the non-equilibrium spin-spin interactions are not, in general, local in time, i.e., the system has *memory*: the parameters depend on two times. We here present the formula for non-equilibrium exchange parameters in the Hartree-Fock approximation (Ref.[8] contains the more general formulas, also for twist exchange):

$$
\begin{aligned}
J_{ab}(t,t') = {} & \frac{1}{4}\delta(t-t')\,\mathrm{Re}\left\{ \int_{-\infty}^{+\infty} \frac{\mathrm{d}\omega}{2\pi} f(\omega,t) \left[\overline{\Sigma}_{ab}^{\downarrow}(t)\, A_{ba}^{\uparrow}(\omega,t) + A_{ab}^{\downarrow}(\omega,t) \overline{\Sigma}_{ba}^{\uparrow}(t) \right] \right\} \\
& + \frac{\mathrm{sign}(t_r)}{8}\,\mathrm{Im}\left\{ \int_{-\infty}^{+\infty} \frac{\mathrm{d}\omega}{2\pi} \int_{-\infty}^{+\infty} \frac{\mathrm{d}\omega'}{2\pi} e^{-i(\omega-\omega')t_r} \left[f(\omega,t_c) - f(\omega',t_c) \right] \right. \\
& \cdot \left\{ \left[\overline{\Sigma}^{\downarrow}(t) \cdot A^{\downarrow}(\omega,t_c) \right]_{ab} \left[\overline{\Sigma}^{\uparrow}(t') \cdot A^{\uparrow}(\omega',t_c) \right]_{ba} - A_{ab}^{\downarrow}(\omega,t_c) \left[\overline{\Sigma}^{\uparrow}(t') \cdot A^{\uparrow}(\omega',t_c) \cdot \overline{\Sigma}^{\uparrow}(t) \right]_{ba} \right. \\
& \left. \left. + \left[A^{\downarrow}(\omega,t_c) \cdot \overline{\Sigma}^{\downarrow}(t') \right]_{ab} \left[A^{\uparrow}(\omega',t_c) \cdot \overline{\Sigma}^{\uparrow}(t) \right]_{ba} - \left[\overline{\Sigma}^{\downarrow}(t) \cdot A^{\downarrow}(\omega,t_c) \cdot \overline{\Sigma}^{\downarrow}(t') \right]_{ab} A_{ba}^{\uparrow}(\omega',t_c) \right\} \right\}.
\end{aligned}
\tag{2}
$$

Here $t_r = t - t'$, $t_c = (t+t')/2$, and $\overline{\Sigma}^{\sigma}(t)$ is the Hartree-Fock self-energy (because of SSB, $\overline{\Sigma}^{\downarrow} \neq \overline{\Sigma}^{\uparrow}$), while $f(\omega,t)$ and $A^{\sigma}(\omega,t)$ are, respectively, the time-dependent occupation number and spectral function, related to the non-equilibrium Green functions via the relation $G_{ab}^{<}(\omega,t) \equiv i f(\omega,t) A_{ab}(\omega,t)$. As an application, in Ref.[8] we also derive the time-dependent spin stiffness tensor for a ferromagnet, which may be relevant for the interpretation of experiments where spin dynamics is induced by a laser beam of macroscopic size [7].

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