Nonequilibrium itinerant-electron magnetism: A time-dependent mean-field theory

A. Secchi, 1,* A. I. Lichtenstein, 2 and M. I. Katsnelson 1

¹Institute for Molecules and Materials, Radboud University Nijmegen, 6525 AJ Nijmegen, The Netherlands ²Institut für Theoretische Physik, Universitat Hamburg, Jungiusstraße 9, D-20355 Hamburg, Germany (Received 12 February 2016; revised manuscript received 26 June 2016; published 31 August 2016)

We study the dynamical magnetic susceptibility of a strongly correlated electronic system in the presence of a time-dependent hopping field, deriving a generalized Bethe-Salpeter equation that is valid also out of equilibrium. Focusing on the single-orbital Hubbard model within the time-dependent Hartree-Fock approximation, we solve the equation in the nonequilibrium adiabatic regime, obtaining a closed expression for the transverse magnetic susceptibility. From this, we provide a rigorous definition of nonequilibrium (time-dependent) magnon frequencies and exchange parameters, expressed in terms of nonequilibrium single-electron Green's functions and self-energies. In the particular case of equilibrium, we recover previously known results.

DOI: 10.1103/PhysRevB.94.085153

The dynamical magnetic susceptibility of an electronic system is a key quantity in both theoretical and experimental studies of magnetism [1,2]. In addition to its physical meaning as the first-order response function of the local magnetic moments to the application of a (space- and time-dependent) magnetic field, its relevance is due to the fact that its frequency spectrum contains all the magnetic excitations of the system. In particular, the spectrum of the transverse component of the magnetic susceptibility tensor contains the magnon frequencies.

To enable theoretical analysis, it is desirable to compute the magnon spectrum directly from a closed formula, rather than doing a numerical search of the poles of the transverse susceptibility. For strongly correlated systems in equilibrium, methods were developed to map electronic Hamiltonians onto effective classical spin models, from which one extracts the magnetic parameters (e.g., exchange) that are appropriate for the initial electronic systems when the magnetic moments undergo small rotations from their initial configuration [3,4]; in the modern formulation, parameters are expressed in terms of single-electron Green's functions (1EGFs) and selfenergies [5,6]. The original methods were recently extended to include unquenched electronic orbital degrees of freedom and relativistic interactions [7–11]. However, a direct connection between the magnetic parameters so determined and the poles of the transverse susceptibility is not obvious; within the framework of spin-density functional theory, it has been shown that the original formulas yield accurate low-wavelength magnon frequencies for ferromagnetic systems within the local spin-density approximation [12]; corrections are required to compute thermodynamic properties [13].

Experimental progress has allowed us to modify the magnetic properties of materials by applying time-dependent fields coupling with the electrons, thereby modulating the magnetic interactions in time. In particular, subpicosecond laser fields [14–22] promise to provide the fastest possible modifications of magnetic states and, in the future, the fastest memory devices. Understanding how the magnetic properties are modulated in time requires a nonequilibrium microscopic theory of magnetism. Computationally, strongly correlated systems are

typically treated with dynamical mean-field theory [23–26] or cluster perturbation theory [27–29] and their nonequilibrium formulations [30–33]. At the moment, the computation of full nonequilibrium two-electron Green's functions (2EGFs), such as the dynamical magnetic susceptibility, is not feasible due to huge memory requirements (even the computation of nonequilibrium 1EGFs is, in general, very demanding [34]). To avoid the computation of 2EGFs, mapping to a dynamical classical spin model has been proposed [35], where the time-dependent magnetic parameters are expressed in terms of nonequilibrium 1EGFs and self-energies. Also in this case, the connection to magnetic susceptibility is not obvious.

In this article, we derive a self-consistent equation for nonequilibrium magnetic susceptibility, and we solve it for the Hubbard model within the time-dependent Hartree-Fock approximation in the adiabatic regime. We show that the effect of an external time-dependent field acting on the electrons (such as that of a laser or a phonon distribution) can be described by endowing the transverse magnetic susceptibility with time-dependent poles, i.e., time-dependent magnon frequencies.

The remainder of this article is organized as follows. In Sec. I, we introduce our notation and discuss the features of our nonequilibrium theory. We then present the problem in its most general formulation, before successively applying several approximations to reduce it to a solvable one. Therefore, in Sec. II we introduce a generalized Bethe-Salpeter equation for magnetic susceptibility that is valid for arbitrary electronic models. The first two steps of the approximations are taken in Sec. III, where we apply the time-dependent Hartree-Fock approximation, and in Sec. IV, where we restrict our theory to the (nonequilibrium) single-band Hubbard model. At this point, the problem can be solved in closed form in equilibrium, but not in the most general nonequilibrium case. The minimal nonequilibrium situation, which allows for a closed solution of the Bethe-Salpeter equation, corresponds to the adiabatic regime, which we introduce in Sec. V. In this regime, the system sustains time-dependent magnon excitations, meaning that the magnon frequencies are modulated in time by the action of the external field, but the magnon concept is still valid. In Sec. VI, we characterize the nonequilibrium magnon frequencies by introducing nonequilibrium exchange parameters, and we recover well-known expressions that are

^{*}Present address: Istituto Italiano di Tecnologia, 16163 Genova, Italy, andrea.secchi@iit.it, andrea.secchi@gmail.com

valid in equilibrium as a particular case. In Sec. VII, we show that our theory is consistent with the Goldstone theorem even out of equilibrium. Finally, in Sec. VIII we summarize our results and mention possible future extensions. In the Appendixes, we include the most technical passages of the derivations, which can be useful to the reader in order to reproduce our main results, but they are not essential for following the discussion in the main text.

I. NOTATION

We formulate our nonequilibrium theory using the Kostantinov-Perel' (KP) time contour $\gamma = \gamma_+ \cup \gamma_- \cup \gamma_{\mathbb{M}}$, where γ_\pm is the forward (backward) branch of the real-time (Keldysh) contour $\gamma_{\mathbb{K}} = \gamma_+ \cup \gamma_-$, and $\gamma_{\mathbb{M}}$ is the imaginary-time (Matsubara) branch [36–39]. If z denotes a contour time variable, we write $z = t_{(\pm)}$ if z lies on γ_\pm , where $t \in [t_0, \infty)$ denotes a physical time; $t_{(+)} < t_{(-)}$ on the contour. We use M to denote the third component of spin of the electron fields, and i, j, k to denote the sets of the other quantum numbers. S_i^α is the α component of the vector of spin matrices for the ith field, with dimensionality S_i . The nonequilibrium Hamiltonian is

$$\hat{H}(z) = \sum_{12} \hat{\psi}_1^{\dagger} T_2^{1}(z) \,\hat{\psi}^2 + \frac{1}{4} \sum_{1234} \hat{\psi}_1^{\dagger} \hat{\psi}_2^{\dagger} V_{3,4}^{2,1} \hat{\psi}^3 \hat{\psi}^4 \quad (1)$$

for $z \in \gamma_{\mathbb{K}}$, where $1 \equiv (i_1, M_1)$ is a complete set of electron field indices, the single-electron terms depend on the contour coordinate z, and the interaction matrix element is antisymmetrized, $V_{3,4}^{2,1} = V_{4,3}^{1,2} = -V_{3,4}^{1,2} = -V_{4,3}^{2,1}$. The single-electron Hamiltonian includes the time-dependent terms generated by the coupling of the electrons with an external time-dependent field. On the Matsubara branch, the Hamiltonian may have a different form [38], which we denote, in general, as

$$\hat{H}(z) = \hat{H}_{\mathbb{M}},\tag{2}$$

independent of z for $z \in \gamma_{\mathbb{M}}$. The Hamiltonian on the Matsubara branch should be considered as a tool to prepare the system in some known state at the initial time t_0 ; it might coincide (up to conserved quantities) with the physical Hamiltonian at the initial time t_0 , in which case the system is prepared in a thermal superposition. Alternatively, one can choose $\hat{H}_{\mathbb{M}}$ as an effective projector over a state or a set of states of interest. For example, to prepare the system in a fully spin-polarized state, one can include in $\hat{H}_{\mathbb{M}}$ a Zeeman term coupling the spins with an auxiliary uniform magnetic field, despite the fact that the Hamiltonian of the system of interest (on the real-time branches) might not include such a magnetic field. In this situation, taking a low temperature effectively restricts the system to a broken-symmetry configuration, which would not be captured in the absence of the auxiliary magnetic field. The results that we present in this work hold regardless of the particular choice of the Hamiltonian on the Matsubara branch.

1EGFs and 2EGFs are denoted as

$$G_{2z_{2}}^{1z_{1}} \equiv -i \langle \mathcal{T}_{\gamma} \hat{\psi}^{1z_{1}} \hat{\psi}_{2z_{2}}^{\dagger} \rangle,$$

$$G_{2z_{2},4z_{4}}^{1z_{1},3z_{3}} \equiv (-i)^{2} \langle \mathcal{T}_{\gamma} \hat{\psi}^{1z_{1}} \hat{\psi}^{3z_{3}} \hat{\psi}_{4z_{4}}^{\dagger} \hat{\psi}_{2z_{2}}^{\dagger} \rangle,$$
(3)

where $\langle \cdots \rangle$ denotes an expectation value computed using the contour evolution operators [36–38]. The contour 1EGFs are

related to the lesser/greater Green's functions via

$$G_{2z_{2}}^{1z_{1}} \equiv \Theta(z_{1}, z_{2})(G^{>})_{2t_{2}}^{1t_{1}} + \Theta(z_{2}, z_{1})(G^{<})_{2t_{2}}^{1t_{1}},$$

$$(G^{>})_{2t_{2}}^{1t_{1}} = -i\langle \hat{\psi}^{1t_{1}} \hat{\psi}^{\dagger}_{2t_{2}} \rangle, \quad (G^{<})_{2t_{1}}^{1t_{1}} = i\langle \hat{\psi}^{\dagger}_{2t_{2}} \hat{\psi}^{1t_{1}} \rangle,$$

$$(4)$$

where $\Theta(z_1, z_2)$ is the step function on the KP contour. Finally, the Dyson equation reads

$$i\partial_z G_{2z_2}^{1z} - \sum_3 \left[T_3^1(z) G_{2z_2}^{3z} + \int_{\gamma} dz_3 \Sigma_{3z_3}^{1z} G_{2z_2}^{3z_3} \right] = \delta_2^1 \delta(z, z_2), \tag{5}$$

where the self-energy Σ is defined via

$$\sum_{5} \int_{\gamma} dz_5 \Sigma_{5z_5}^{1z_1} G_{2z_2}^{5z_5} \equiv \frac{i}{2} \sum_{345} V_{4,5}^{3,1} G_{2z_2,3(z_1+\epsilon)}^{4z_1,5z_1}.$$
 (6)

II. GENERALIZED BETHE-SALPETER EQUATION

The dynamical magnetic susceptibility tensor is

$$\chi_{ij}^{\alpha\alpha'}(t,t') \equiv \frac{\delta \langle \hat{S}_{i}^{\alpha}(t) \rangle_{B}}{\delta B_{j}^{\alpha'}(t')} \bigg|_{B=0}$$

$$= -i\Theta(t-t') \langle [\hat{S}_{i}^{\alpha}(t), \hat{S}_{j}^{\alpha'}(t')] \rangle, \tag{7}$$

where $\langle \cdots \rangle_B$ denotes an expectation value computed in the presence of the magnetic field $\mathbf{B} \equiv \{\mathbf{B}_i(t)\}$ coupling with the spins, and $\hat{S}_i^{\alpha}(t)$ is the α component of the ith spin operator of the system at time t; $\alpha \in \{x,y,z\}$ or $\alpha \in \{+,-,z\}$. The second line of Eq. (7) is the Kubo formula, which connects $\chi_{ij}^{\alpha\alpha'}(t,t')$ to relevant many-body quantities. For example, for a ferromagnetic lattice in equilibrium, the low-energy poles of the Laplace transform of the transverse magnetic susceptibility $\chi_a^{\mu-}(\omega)$ are the magnon frequencies ω_a .

 $\chi_q^{+-}(\omega)$ are the magnon frequencies ω_q . We now generalize the Bethe-Salpeter equation (BSE) for the magnetic susceptibility to the case of the most arbitrary electronic system out of equilibrium. It is convenient to define the matrices

$$\chi_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};z_{3}) \equiv -i \frac{\delta(S^{\alpha}G_{B})_{2z_{2}}^{1z_{1}}}{\delta B_{jz_{3}}^{\alpha'}} \bigg|_{B=0},
\chi_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t') \equiv \chi_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t'_{(+)}) - \chi_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t'_{(-)}),$$
(8)

where the magnetic field is allowed to take different values for the two Keldysh coordinates corresponding to the same physical time. The susceptibility matrix defined in Eq. (8) satisfies the following generalized Bethe-Salpeter equation (GBSE) on the KP contour [for the full derivation, see Appendix A]:

$$\chi_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t') = (\chi_{0})_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t')$$

$$+ \sum_{\alpha''\alpha'''} \sum_{4567} \int_{\gamma} d(w_{4},w_{5},w_{6},w_{7}) (\chi_{0}^{\alpha\alpha''})_{2z_{2},4w_{4}}^{1z_{1},5w_{5}}$$

$$\times (\Gamma^{\alpha''\alpha'''})_{5w_{6},6w_{7}}^{4w_{4},7w_{7}} \chi_{6,7;i}^{\alpha'''\alpha'}(w_{6},w_{7};t'), \qquad (9)$$

where we have introduced the quantities

$$\left(\chi_0^{\alpha\alpha''}\right)_{2z_2,4w_4}^{1z_1,5w_5} \equiv -i(S^{\alpha}GS^{\alpha''})_{4w_4}^{1z_1}G_{2z_2}^{5w_5},\tag{10}$$

$$(\Gamma^{\alpha''\alpha'''})_{5w_5,6w_6}^{4w_4,7w_7} \equiv \frac{i}{S_{i_*}(S_{i_*}+1)} \frac{\delta(S^{\alpha''}\Sigma)_{5w_5}^{4w_4}}{\delta(S^{\alpha'''}G)_{5w_5}^{6w_6}},\tag{11}$$

$$(\chi_0)_{1,2;j}^{\alpha\alpha'}(z_1,z_2;t') = \sum_{s=\pm} s \sum_{M} \left(\chi_0^{\alpha\alpha'}\right)_{2z_2, jMt'_{(s)}}^{1z_1, jMt'_{(s)}}.$$
 (12)

The physical susceptibility given by Eq. (7) can be obtained from Eq. (8) via the relation

$$\chi_{ij}^{\alpha\alpha'}(t,t') = \sum_{M} \chi_{iM,iM;j}^{\alpha\alpha'}(t_{(+)},t_{(-)};t'), \tag{13}$$

as detailed in Appendix B.

III. TIME-DEPENDENT HARTREE-FOCK APPROXIMATION

Equation (9) is exact, but its matrix structure is very complicated. The time-dependent Hartree-Fock approximation (THF) [38] greatly simplifies its time-domain structure. In THF, the 2EGF is approximated as

$$G_{B,D}^{A,C} \stackrel{\text{THF}}{=} G_B^A G_D^C - G_D^A G_B^C, \tag{14}$$

which yields the following expression for the self-energy:

$$\Sigma_{2z_2}^{1z_1} \stackrel{\text{THF}}{=} -i\delta(z_1, z_2) \sum_{2A} V_{2,4}^{1,3} (G^{<})_{3t_1}^{4t_1}.$$
 (15)

It should be noted that the 1EGFs appearing in Eqs. (14) and (15) are *not* the noninteracting Green's functions that are used in conventional many-body perturbation theory for weakly correlated systems, where the electron-electron interaction is the small parameter. In that case, Eqs. (14) and (15) would reduce to the RPA scheme. In our case, instead, single-particle Green's functions are the solutions of an *interacting* problem, although simplified via the THF approximation, which keeps only the part of the self-energy that is local in time. Although the equations are formally similar, this difference between THF and RPA is crucial to properly describe the magnon excitations for strongly correlated systems.

With this distinction in mind, we now introduce

$$(\chi_0)_{ij}^{\alpha\alpha'}(t,t') = \sum_{M} (\chi_0)_{iM,iM;j}^{\alpha\alpha'}(t_{(+)},t_{(-)};t'), \qquad (16)$$

which is a physical quantity defined in terms of 1EGFs, whose meaning depends on the approximation scheme. In our case, it can be called *Stoner susceptibility*, since its spectrum contains only electron-hole excitations that are analogous to those of the Stoner theory for the Hubbard model. In contrast, within the RPA, Eq. (16) would coincide with the bare magnetic susceptibility of a noninteracting system.

Applying Eq. (15) to Eq. (11), we obtain

$$(\Gamma^{\alpha''\alpha'''})_{5w_5,6w_6}^{4w_4,7w_7} \stackrel{\text{THF}}{=} \delta(w_4, w_5)\delta(w_4, w_6)\delta(w_7, w_4 + \epsilon) \left(\Gamma^{\alpha''\alpha'''}_{\text{THF}}\right)_{5,6}^{4,7},$$
(17)

where

$$\left(\Gamma_{\text{THF}}^{\alpha''\alpha'''}\right)_{5,6}^{4,7} \equiv \sum_{MM'} \frac{\left(S_{i_4}^{\alpha''}\right)_{M'}^{M_4} V_{5,(i_6M)}^{(i_4M'),7} \left(S_{i_6}^{\alpha'''}\right)_{M_6}^{M}}{S_{i_4} (S_{i_4} + 1) S_{i_6} (S_{i_6} + 1)}.$$
 (18)

Inserting Eq. (18) into Eq. (9) yields the THF form of the GBSE.

$$\chi_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t') \stackrel{\text{THF}}{=} (\chi_{0})_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};t')$$

$$+ \sum_{\alpha''\alpha'''} \sum_{4567} \int_{\gamma} dw_{4} (\chi_{0}^{\alpha\alpha''})_{2z_{2},4w_{4}}^{1z_{1},5w_{4}} (\Gamma_{\text{THF}}^{\alpha''\alpha''})_{5,6}^{4,7}$$

$$\times \chi_{6.7:j}^{\alpha'''\alpha'}(w_{4},w_{4}+\epsilon;t').$$
(19)

If t_4 is the physical time corresponding to the contour coordinate w_4 , then the quantity

$$\chi_{6,7;i}^{\alpha'''\alpha'}(w_4, w_4 + \epsilon; t') \equiv \chi_{6,7;i}^{\alpha'''\alpha'}(t_4; t') \tag{20}$$

depends only on t_4 , independently of the contour branch on which w_4 lies. This can be seen by applying Eq. (8) with $z_1 = w_4$ and $z_2 = w_4 + \epsilon$. In terms of Eq. (20), we have

$$\chi_{ij}^{\alpha\alpha'}(t,t') \equiv \sum_{M} \chi_{iM,iM;j}^{\alpha\alpha'}(t;t'). \tag{21}$$

Setting $z_1 = t_{(+)}$ and $z_2 = t_{(-)}$, and

$$\chi_{1,2;j}^{\alpha\alpha'}(t;t') \equiv \chi_{1,2;j}^{\alpha\alpha'}(t_{(+)},t_{(-)};t'), (\chi_0)_{1,2;j}^{\alpha\alpha'}(t;t') \equiv (\chi_0)_{1,2;j}^{\alpha\alpha'}(t_{(+)},t_{(-)};t'),$$
(22)

we then obtain, from Eq. (19), the THF GBSE in real-time coordinates:

$$\chi_{1,2;j}^{\alpha\alpha'}(t;t') \stackrel{\text{THF}}{=} (\chi_0)_{1,2;j}^{\alpha\alpha'}(t;t') + \sum_{\alpha''\alpha'''} \sum_{4567} \int_{t_0}^{\infty} dt'' \chi_0^{\alpha\alpha''}(t,t'')_{2,4}^{1,5} \times (\Gamma_{\text{THF}}^{\alpha''\alpha''})_{5,6}^{4,7} \chi_0^{\alpha'''\alpha'}(t'';t'), \tag{23}$$

where we have converted the contour integration to physicaltime integration, we have used the fact that $\chi_{6,7;j}^{\alpha'''\alpha'}(w_4, w_4 + \epsilon; t') = 0$ if $w_4 \in \gamma_M$, and we have introduced

$$\begin{split} \chi_{0}^{\alpha\alpha''}(t,t'')_{2,4}^{1,5} \equiv & \left(\chi_{0}^{\alpha\alpha''}\right)_{2t_{(-)},4t_{(+)}^{(\prime)}}^{1t_{(+)},5t_{(+)}^{\prime\prime}} - \left(\chi_{0}^{\alpha\alpha''}\right)_{2t_{(-)},4t_{(-)}^{\prime\prime}}^{1t_{(+)},5t_{(-)}^{\prime\prime}} \\ = & -i\theta(t-t'') \left[\left(S^{\alpha} \left(G^{>}\right)_{t''}^{t} S^{\alpha''}\right)_{4}^{1} \left(G^{<}\right)_{2t'}^{5t''} \\ & - \left(S^{\alpha} \left(G^{<}\right)_{t''}^{t} S^{\alpha''}\right)_{4}^{1} \left(G^{>}\right)_{2t'}^{5t''} \right], \end{split}$$
(24)

and therefore

$$(\chi_0)_{1,2;j}^{\alpha\alpha'}(t;t') = \sum_{M} \chi_0^{\alpha\alpha'}(t,t')_{2,jM}^{1,jM}.$$
 (25)

IV. SINGLE-ORBITAL HUBBARD MODEL

To achieve a further simplification, we restrict our theory to the single-orbital Hubbard model (SOH). In this case, the spin space has dimensionality $\mathcal{S}=1/2$ at every site, and the interaction Hamiltonian becomes

$$\hat{V} = \frac{1}{4} \sum_{3456} V_{5,6}^{4,3} \hat{\psi}_3^{\dagger} \hat{\psi}_4^{\dagger} \hat{\psi}^5 \hat{\psi}^6 \stackrel{\text{SOH}}{=} \sum_i U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}, \qquad (26)$$

which implies

$$V_{5,6}^{4,3} \stackrel{\text{SOH}}{=} \delta_{i_5}^{i_4} \delta_{i_5}^{i_3} \delta_{i_6}^{i_3} \delta_{\overline{M}_3}^{M_4} \delta_{M_6}^{\overline{M}_5} (\delta_{M_5}^{\overline{M}_3} - \delta_{M_5}^{M_3}) U_{i_3}. \tag{27}$$

The SOH Hamiltonian is spin-independent: $[\hat{H}(t), \hat{S}^z] = 0$, so the total third component of the spin of the system is a good quantum number. The transverse component of

Eq. (23), corresponding to $(\alpha, \alpha') = (+, -)$, then simplifies as

$$\chi_{ij}^{+-}(t,t') \stackrel{\text{THF SOH}}{=} (\chi_0)_{ij}^{+-}(t,t') + \sum_k \int_{t_0}^{\infty} dt''$$

$$\times (\chi_0)_{ik}^{+-}(t,t'')(-U_k)\chi_{ki}^{+-}(t'',t'), \quad (28)$$

which is the nonequilibrium SOH version of the equation used in Ref. [12]. Compared to the general case, this form of the BSE has the simplest possible structure in both time and spin domains. Details about the derivation of Eq. (28) are given in Appendix C.

The Stoner transverse susceptibility is

$$(\chi_0)_{ij}^{+-}(t,t') \stackrel{\text{SOH}}{=} i\theta(t-t') \left[(G^<)_{j\downarrow t'}^{i\downarrow t} (G^>)_{i\uparrow t}^{j\uparrow t'} - (G^>)_{i\downarrow t'}^{i\downarrow t} (G^<)_{i\uparrow t}^{j\uparrow t'} \right]. \tag{29}$$

We now derive an effective equation for this quantity by applying the operator $-i\partial_{t'}$ to Eq. (29) and using the Dyson equations in the THF approximation, which read

$$-i \partial_{t'} (G^{\lessgtr})_{jMt'}^{iMt} \stackrel{\text{THF}}{=} (G^{\lessgtr}T)_{jMt'}^{iMt} + (G^{\lessgtr})_{jMt'}^{iMt} \Sigma_{jM}(t'),$$

$$i \partial_{t'} (G^{\lessgtr})_{iMt}^{jMt'} \stackrel{\text{THF}}{=} (TG^{\lessgtr})_{iMt}^{jMt'} + \Sigma_{jM}(t') (G^{\lessgtr})_{iMt}^{jMt'},$$

$$(30)$$

where $\Sigma_{jM}(t') \equiv U_j \rho_{j\overline{M}}(t')$ is the THF self-energy for the SOH model, with $\rho_{j\overline{M}}(t') \equiv \langle \hat{\psi}^{\dagger}_{j\overline{M}}(t') \hat{\psi}^{j\overline{M}}(t') \rangle$. We then obtain

$$(\chi_0)_{ij}^{+-}(t,t')[-i\overleftarrow{\partial_{t'}} - \Delta_j(t')]$$

$$\stackrel{\text{SOH THF}}{=} \delta(t-t')\delta_{ij}m_i(t') + \Lambda_{ij}(t,t'), \tag{31}$$

where $m_i(t') \equiv \rho_{i\uparrow}(t') - \rho_{i\downarrow}(t')$,

$$\Delta_{i}(t') \equiv U_{i} m_{i}(t') \equiv -2\Sigma_{iS}(t') \equiv \Sigma_{i\downarrow}(t') - \Sigma_{i\uparrow}(t') \quad (32)$$

is the time-dependent Stoner splitting, and

$$\Lambda_{ij}(t,t') = i\theta(t-t') \Big[(G^{>})_{j\downarrow t'}^{i\downarrow t} (TG^{<})_{i\uparrow t}^{j\uparrow t'} - (G^{<})_{j\downarrow t'}^{i\downarrow t} (TG^{>})_{i\uparrow t}^{j\uparrow t'} \\
- \left(G^{>}T \right)_{i\downarrow t'}^{i\downarrow t} (G^{<})_{i\uparrow t}^{j\uparrow t'} + (G^{<}T)_{i\downarrow t'}^{i\downarrow t} (G^{>})_{i\uparrow t}^{j\uparrow t'} \Big].$$
(33)

We now determine the transverse magnetic susceptibility from the BSE, Eq. (28), and the approximate equation for the bare susceptibility, Eq. (31).

V. ADIABATIC APPROXIMATION AND NONEOUILIBRIUM MAGNONS

We introduce the Wigner time coordinates, $\tau \equiv t - t'$ and $T \equiv (t + t')/2$, which are called the relative time and the total time, respectively. In this section, we send the initial time $t_0 \to -\infty$, so that the domain of T is $(-\infty,\infty)$, and we can define the Fourier transforms with respect to T on the whole real axis. We set $f(t,t') \equiv \widetilde{f}(\tau,T)$ to distinguish the representations of a function in terms of the individual fermionic time arguments versus the Wigner coordinates. We apply to both Eqs. (28) and (31) the Laplace transform with respect to T and the Fourier transform with respect to T. We use the notation

$$\widetilde{f}(\omega,\Omega) \equiv \int_{-\infty}^{\infty} dT \, e^{i\Omega T} \int_{0}^{\infty} d\tau \, e^{i\omega \tau} \, \widetilde{f}(\tau,T), \qquad (34)$$

where $\text{Im}(\omega) > 0$. We obtain the following representations of Eqs. (28) and (31) in the frequency domain [the full derivation can be found in Appendix D]:

$$\widetilde{\chi}_{ij}^{+-}(\omega,\Omega) - (\widetilde{\chi}_0)_{ij}^{+-}(\omega,\Omega) \stackrel{\text{THF SOH}}{=} - \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} \sum_{k} (\widetilde{\chi}_0)_{ik}^{+-} \left(\omega + \frac{\Omega - \Omega'}{2}, \Omega'\right) U_k \widetilde{\chi}_{kj}^{+-} \left(\omega - \frac{\Omega'}{2}, \Omega - \Omega'\right), \tag{35}$$

$$\left(\omega - \frac{\Omega}{2}\right)(\widetilde{\chi}_0)_{ij}^{+-}(\omega, \Omega) - \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi}(\widetilde{\chi}_0)_{ij}^{+-}\left(\omega + \frac{\Omega - \Omega'}{2}, \Omega'\right) \Delta_j(\Omega - \Omega') \stackrel{\text{THF SOH}}{=} \delta_j^i m_j(\Omega) + \widetilde{\Lambda}_{ij}(\omega, \Omega). \tag{36}$$

In the nonequilibrium adiabatic (AD) regime, we assume that the susceptibilities are nonzero only when the frequencies related to the Fourier transforms with respect to the total time T are much smaller than the frequencies related to the Laplace transforms with respect to the relative time τ . In this case, Eq. (36) simplifies to

$$(\widetilde{\chi}_0)_{ij}^{+-}(\omega;T) \stackrel{\text{THF SOH AD}}{=} \stackrel{\delta_{ij}m_j(T) + \widetilde{\Lambda}_{ij}(\omega;T)}{\omega - \Delta_j(T)}, \quad (37)$$

while Eq. (35) simplifies to

$$\sum_{k} [\delta_{ik} + (\widetilde{\chi}_0)_{ik}^{+-}(\omega; T) U_k] \widetilde{\chi}_{kj}^{+-}(\omega; T)$$

$$\stackrel{\text{THF SOH AD}}{=} = (\widetilde{\chi}_0)_{ij}^{+-}(\omega; T). \tag{38}$$

We substitute Eq. (37) into Eq. (38), and, after some algebra, we get

$$\widetilde{\chi}_{ij}^{+-}(\omega;T) \stackrel{\text{THF SOH AD}}{=} \stackrel{\text{DD}}{=} \frac{\omega - \Delta_{i}(T)}{\omega - \Delta_{j}(T)} \sum_{k} \frac{U_{k}}{U_{i}} \overrightarrow{F}_{ik}^{-1}(\omega;T) \times [\delta_{ki} m_{i}(T) + \widetilde{\Lambda}_{ki}(\omega;T)], \quad (39)$$

where we have introduced the matrix

$$F_{ik}(\omega;T) \equiv \delta_{ik}\omega + U_i \widetilde{\Lambda}_{ik}(\omega;T) \tag{40}$$

and its left inverse $\overrightarrow{F}^{-1}(\omega;T)$, defined via

$$\sum_{i} \overrightarrow{F}_{li}^{-1}(\omega; T) F_{ik}(\omega; T) = \delta_{lk}. \tag{41}$$

The susceptibility has a pole when the matrix (40) has a null eigenvalue. If we assume that $\widetilde{\Lambda}_{ik}(\omega;T)$ is almost independent of ω at frequencies much smaller than the Stoner excitations, then the poles are obtained when ω is an eigenvalue of the

time-dependent matrix,

$$\Omega_{ij}(T) \equiv -U_i \widetilde{\Lambda}_{ij}(0;T). \tag{42}$$

The eigenvalues of (42) can then be called *nonequilibrium* magnon frequencies, and they are time-dependent due to the action of the external field. It should be noted that the system given by the union of the magnetic medium and the external field might in general have a lower spatial symmetry than the lattice of the magnetic medium in the absence of the field (the field typically has some privileged directions, such as the polarization and direction of propagation for an electromagnetic wave). If such symmetry lowering is absent or negligible, one can exploit the symmetry of the magnetic lattice to diagonalize $\Omega_{ij}(T)$ (see Appendix E).

In equilibrium, which is formally a particular case of this treatment that is obtained when the Hamiltonian is time-independent, Ω_{ij} is independent of T and its eigenvalues are the conventional magnon frequencies. Therefore, we have formally demonstrated that the minimal correction to the transverse magnetic susceptibility in nonequilibrium situations, valid in the adiabatic regime, consists in the fact that the magnon frequencies acquire a time dependence.

We note that the approximation that produces Eq. (42), namely replacing $\widetilde{\Lambda}_{ij}(\omega;T) \to \widetilde{\Lambda}_{ij}(0;T)$, corresponds to linearizing the eigenvalue problem associated with Eq. (40). Corrections can be computed by taking into account higher-order terms in the Taylor expansion of $\widetilde{\Lambda}_{ij}(\omega;T)$ in powers of ω ; such analysis is beyond the scope of this work.

We now characterize the nonequilibrium magnon frequencies and establish the correspondence to the previous literature by introducing two different forms of nonequilibrium exchange parameters.

VI. NONEQUILIBRIUM EXCHANGE PARAMETERS

A. Two-times exchange parameters

We first switch back from the frequency-domain representation to the time-domain representation. We define the two-times exchange matrix

$$\Omega_{ij}(t,t') \stackrel{\text{THF SOH}}{=} -U_i \Lambda_{ij}(t,t'), \tag{43}$$

and we express it in terms of nonequilibrium 1EGFs and self-energies. Toward that end, we use the nonequilibrium Dyson equations in the THF approximation, Eq. (30), to eliminate the hopping matrix T from the expression of Λ , Eq. (33). We obtain

$$\Lambda_{ij}(t,t') \stackrel{\text{THF}}{=} i\theta(t-t') [2\Sigma_{jS}(t') - i \overrightarrow{\partial}_{t'}]
\times \left[(G_{\downarrow}^{<})_{jt'}^{it} (G_{\uparrow}^{>})_{it}^{jt'} - (G_{\downarrow}^{>})_{jt'}^{it} (G_{\uparrow}^{<})_{it}^{jt'} \right].$$
(44)

We split the exchange matrix into two parts,

$$\Omega_{ij}(t,t') \stackrel{\text{THF SOH}}{=} \frac{4}{m_i(\frac{t+t'}{2})} [J_{ij}(t,t') + X_{ij}(t,t')],$$
(45)

where

$$J_{ij}(t,t') \equiv i\theta(t-t') \, \Sigma_{iS} \left(\frac{t+t'}{2}\right) \Sigma_{jS}(t')$$

$$\times \left[(G_{\downarrow}^{<})_{jt'}^{it} (G_{\uparrow}^{>})_{it}^{jt'} - (G_{\downarrow}^{>})_{jt'}^{it} (G_{\uparrow}^{<})_{it}^{jt'} \right]$$
(46)

is the two-times exchange parameter (equivalent to the analogous quantity obtained in Ref. [35]), and

$$X_{ij}(t,t') \equiv \theta(t-t') \frac{1}{2} \Sigma_{iS} \left(\frac{t+t'}{2}\right)$$

$$\times \overrightarrow{\partial}_{t'} \left[(G_{\downarrow}^{<})_{jt'}^{it} (G_{\uparrow}^{>})_{it}^{jt'} - (G_{\downarrow}^{>})_{jt'}^{it} (G_{\uparrow}^{<})_{it}^{jt'} \right]$$
(47)

is a quantity whose meaning will be clarified in Sec. VIB. Switching again to the Wigner-coordinates representation and Laplace transforming with respect to relative time, we obtain

$$\widetilde{\Omega}_{ij}(\omega;T) \stackrel{\text{THF SOH}}{=} \frac{4}{m_i(T)} [\widetilde{J}_{ij}(\omega;T) + \widetilde{X}_{ij}(\omega;T)].$$
 (48)

We simplify the second term on the right-hand side of Eq. (48); after performing partial integration and using the relation $(G^{>})_{i,T}^{i,T} = -i\delta_i^i + (G^{<})_{i,T}^{i,T}$, we obtain

$$\widetilde{X}_{ij}(\omega;T) = -\frac{1}{2}\delta_{ij}\Sigma_{iS}(T)m_{i}(T)$$

$$+\frac{1}{2}\Sigma_{iS}(T)\left(\frac{1}{2}\overrightarrow{\partial}_{T} + i\omega\right)\int_{0}^{\infty}d\tau e^{i\omega\tau}$$

$$\times \left[(G_{\downarrow}^{<})_{j,T-\tau/2}^{i,T+\tau/2}(G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2}$$

$$-(G_{\downarrow}^{>})_{i,T-\tau/2}^{i,T+\tau/2}(G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2}\right]. \tag{49}$$

The first term on the right-hand side of Eq. (48) involves the Laplace transform of the two-times exchange parameters,

$$\widetilde{J}_{ij}(\omega;T) \equiv i \Sigma_{iS}(T) \int_{0}^{\infty} d\tau \, e^{i\omega\tau} \Sigma_{jS}(T - \tau/2)
\times \left[(G_{\downarrow}^{<})_{j,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{>})_{i,T+\tau/2}^{j,T-\tau/2} \right.
\left. - (G_{\perp}^{>})_{i,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2} \right].$$
(50)

B. One-time exchange parameters

If $\widetilde{\Omega}_{ij}(\omega;T)$ is almost independent of ω , we can determine a *time-dependent* pole of the nonequilibrium transverse susceptibility, which is a generalization of the magnon frequency to the nonequilibrium adiabatic regime, as in Eq. (42). More explicitly, from Eq. (48) we write

$$\Omega_{ij}(T) \equiv \lim_{\epsilon \to 0^{+}} \lim_{\omega \to 0} \widetilde{\Omega}_{ij}(\omega + i\epsilon; T)$$

$$\equiv \frac{4}{m_{i}(T)} [J_{ij}(T) + X_{ij}(T)], \tag{51}$$

where ω and $\epsilon > 0$ are real, and

$$J_{ij}(T) = i \Sigma_{iS}(T) \lim_{\epsilon \to 0^{+}} \int_{0}^{\infty} d\tau \, e^{-\epsilon \tau} \, \Sigma_{jS}(T - \tau/2)$$

$$\times \left[(G_{\downarrow}^{<})_{j,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2} - (G_{\downarrow}^{<})_{j,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2} \right], \qquad (52)$$

$$X_{ij}(T) = -\frac{1}{2} \delta_{ij} \Sigma_{iS}(T) m_{i}(T)$$

$$+ \frac{1}{4} \Sigma_{iS}(T) \lim_{\epsilon \to 0^{+}} \overrightarrow{\partial}_{T} \int_{0}^{\infty} d\tau \, e^{-\epsilon \tau}$$

$$\times \left[(G_{\downarrow}^{<})_{j,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2} - (G_{\downarrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2} \right]. \qquad (53)$$

As seen in Eq. (51), both terms $J_{ij}(T)$ and $X_{ij}(T)$ contribute on the same footing to the time-dependent magnon dispersion. We identify $J_{ii}(T)$ given in Eq. (52) as the time-dependent exchange parameter due to its nonlocality in space and its general structure that can be schematically denoted as $\Sigma G \Sigma G$, which is analogous to the structure found for the equilibrium exchange parameters in equilibrium theories (see, e.g., Refs. [5,6,8]). The term $X_{ii}(T)$ defined in Eq. (53) is given by two contributions. The first line is local in space; an analogous term appears in the expression of the dynamical transverse susceptibility in equilibrium (see Sec. VIC), of which this is the nonequilibrium generalization. The second line is a purely (nonlocal) nonequilibrium term with no analog in equilibrium. In fact, the Green's functions would not depend on T in that case, so the derivative would vanish. Out of equilibrium, instead, the T dependence is not trivial, due to the time-dependent hopping. This term is explicitly related to the dynamical variation of the sites' electronic population. The presence of the term $X_{ij}(T)$ in the expression of the susceptibility has an important role in showing that the magnon dispersion satisfies the Goldstone theorem, even out of equilibrium (see Sec. VII).

C. Equilibrium exchange parameters

The equilibrium regime is a particular case of the adiabatic regime, such that 1EGFs depend only on the relative time τ and not on the total time T, while THF self-energies are time-independent. The equilibrium exchange parameters are obtained from Eqs. (52) and (53) by removing the dependence on T. If the state of the system is given by a thermal distribution, in the limit of zero temperature (or inverse temperature $\beta \to \infty$) we can apply the analytical continuation from the real-time branches of the KP contour to the imaginary-time branch, and we represent 1EGFs in the Matsubara formalism. In this case, we obtain (details are given in Appendix F)

$$J_{ij} = \frac{1}{2} \lim_{\beta \to \infty} \frac{1}{\beta} \sum_{\omega_n} \Delta_i G_{ij}^{\downarrow}(i\omega_n) \Delta_j G_{ji}^{\uparrow}(i\omega_n),$$

$$X_{ij} = \frac{1}{4} \delta_{ij} \Delta_i m_i.$$
(54)

This result agrees with the equilibrium formulas derived with different methods in Refs. [5–8,35], specialized to the SOH model in the HF approximation. We see that Eq. (53) is the nonequilibrium generalization of the last term of Eq. (31) in Ref. [12].

VII. GOLDSTONE THEOREM

The SOH model is not relativistic, therefore rotating all the electronic spins of the same angle with respect to a given axis costs no energy. Since this is a continuous symmetry, the Goldstone theorem predicts that the exchange matrix has a null eigenvalue, which in a lattice corresponds to the eigenstate with q = 0 (that is, $\lim_{q\to 0} \omega_q = 0$). We recover this result in our theory, even out of equilibrium, since it immediately follows

from Eqs. (33) and (43) that

$$\sum_{i} \Lambda_{ij}(t, t') = 0 \Rightarrow \sum_{i} \Omega_{ij}(t, t') = 0, \tag{55}$$

hence the vector $(1,1,1,\ldots,1)$ is an eigenvector of the exchange matrix $\Omega(t,t')$, with eigenvalue $\omega=0$ (if the system is a lattice, such an eigenvector corresponds indeed to the state with q=0). Obviously, this property holds also in equilibrium, as a particular case. An alternative way to check that our theory is consistent with the Goldstone theorem is shown in Appendix G.

The Goldstone theorem suggests a possible alternative definition for the exchange parameters contributing to the (one-time) exchange matrix. We can define *starred* exchange parameters by combining Eq. (52) and the nonlocal part of Eq. (53) (second line). We get

$$J_{ij}^{\star}(T) = i \sum_{i,S} (T) \lim_{\epsilon \to 0^{+}} \int_{0}^{\infty} d\tau \ e^{-\epsilon \tau} \left[\sum_{j,S} (T - \tau/2) - \frac{i}{4} \stackrel{\rightarrow}{\theta}_{T} \right]$$

$$\times \left[(G_{\downarrow}^{<})_{j,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{>})_{i,T+\tau/2}^{j,T-\tau/2} - (G_{\downarrow}^{>})_{i,T-\tau/2}^{i,T+\tau/2} (G_{\uparrow}^{<})_{i,T+\tau/2}^{j,T-\tau/2} \right].$$

$$(56)$$

Combining this definition with Eq. (55), we can rewrite Eq. (51) in terms of J^* only as

$$\Omega_{ij}(T) \equiv \frac{4}{m_i(T)} \left[J_{ij}^{\star}(T) - \delta_{ij} \sum_{k} J_{ik}^{\star}(T) \right]. \tag{57}$$

VIII. SUMMARY

To summarize, we have presented a rigorous derivation of the transverse spin susceptibility in the nonequilibrium adiabatic regime for the SOH model within the THF approximation, leading to the definition of nonequilibrium magnon frequencies and exchange parameters. Our results should be relevant to interpret the physics associated with ultrafast laser experiments, and possibly to unravel the effect of phonons on the magnetic properties of materials, provided that the frequencies of the oscillating fields are much smaller than the Stoner excitations. Further work can be envisaged to remove the THF approximation and extend to more general electronic systems, including relativistic interactions. The starting point for these possible developments is given by the GBSE, Eq. (9).

With regard to the possibility of developing a nonequilibrium theory beyond the THF approximation, we mention that using exact Green's functions but neglecting the vertices is not acceptable because it would break the Goldstone theorem [40]. The possibility of obtaining a problem that can be solved in closed form without employing the THF approximation must rely on the assumption of some small parameter (and therefore a necessary loss of generality with respect to the unspecified electronic configuration that we have considered here). In equilibrium, a technique involving exact Green's functions of the Hubbard X-operators was presented in Refs. [41,42], applied to study a fully spin-polarized electronic system with a small concentration of holes, with emphasis on the two-magnon scattering processes. The inclusion of full Green's functions beyond the Hartree-Fock approximation was possible due to the assumed smallness of either the concentration of holes or the inverse number of nearest neighbors, which allowed a linear approximation in one of those parameters. The generalization of this technique to the nonequilibrium regime is beyond the scope of the present work, where we have focused instead on obtaining the THFapproximated results without making any assumption about the electronic configuration.

ACKNOWLEDGMENTS

A.S. and M.I.K. acknowledge funding from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO) via a Spinoza Prize; A.I.L. acknowledges funding from the SFB-925 grant of Deutsche Forschungsgemeinschaft (DFG). We thank Mikhail Titov and Alessandro Principi for useful discussions.

APPENDIX A: DERIVATION OF THE GENERALIZED BETHE-SALPETER EQUATION

We derive the generalized Bethe-Salpeter equation using the properties of the nonequilibrium Green's functions. The Dyson equations on the KP contour are written as

$$\sum_{3} \int_{\gamma} dz_{3} (\overrightarrow{G^{-1}}_{B})_{3z_{3}}^{1z_{1}} (G_{B})_{2z_{2}}^{3z_{3}} = \delta_{2}^{1} \delta(z_{1}, z_{2}),$$

$$\sum_{3} \int_{\gamma} dz_{3} (G_{B})_{3z_{3}}^{1z_{1}} (\overrightarrow{G^{-1}}_{B})_{2z_{2}}^{3z_{3}} = \delta_{2}^{1} \delta(z_{1}, z_{2}),$$
(A1)

where

$$(\overrightarrow{G^{-1}}_{B})_{3z_{3}}^{1z_{1}} = \delta_{3}^{1}\delta(z_{1}, z_{3})i\overrightarrow{\partial}_{z_{3}} - (T_{B})_{3z_{3}}^{1z_{1}} - (\Sigma_{B})_{3z_{3}}^{1z_{1}}, (\overrightarrow{G^{-1}}_{B})_{2z_{2}}^{3z_{3}} = -i\overleftarrow{\partial}_{z_{3}}\delta_{2}^{3}\delta(z_{3}, z_{2}) - (T_{B})_{2z_{2}}^{3z_{3}} - (\Sigma_{B})_{2z_{2}}^{3z_{3}}.$$
(A2)

Here Σ_B and T_B denote, respectively, the self-energy and single-particle Hamiltonian matrix in the presence of a magnetic field B depending on the KP coordinate. In particular,

$$(T_{\boldsymbol{B}})_{3z_{3}}^{1z_{1}} \equiv \delta(z_{1}, z_{3}) \left\{ [T(z_{1})]_{3}^{1} + \delta_{i_{3}}^{i_{1}} \boldsymbol{B}_{i_{1}z_{1}} \cdot (\boldsymbol{S}_{i_{1}})_{M_{3}}^{M_{1}} \right\}, \quad (A3)$$

where $[T(z_1)]_3^1$ is the hopping term that does not depend on B, but is time-dependent as well, since it includes all the external fields acting on the electrons. Using a condensed notation, where the sums over all matrix indices and integrations over intermediate times are implied, we can write

$$\overrightarrow{G^{-1}}_{B} \cdot G_{B} = 1 \Rightarrow \frac{\delta \overrightarrow{G^{-1}}_{B}}{\delta B_{jz_{3}}^{\alpha'}} \cdot G_{B} + \overrightarrow{G^{-1}}_{B} \cdot \frac{\delta G_{B}}{\delta B_{jz_{3}}^{\alpha'}} = \mathbf{0}$$

$$\Rightarrow G_{B} \cdot \overrightarrow{G^{-1}}_{B} \cdot \frac{\delta G_{B}}{\delta B_{jz_{3}}^{\alpha'}} = -G_{B} \cdot \frac{\delta \overrightarrow{G^{-1}}_{B}}{\delta B_{jz_{3}}^{\alpha'}} \cdot G_{B}.$$
(A4)

We can replace $G_B \cdot \overrightarrow{G^{-1}}_B \to G_B \cdot \overleftarrow{G^{-1}}_B \equiv 1$, since the two expressions differ only by boundary terms that vanish due to the Kubo-Martin-Schwinger relations [43] on the KP contour. We then obtain the identity

$$\frac{\delta G_{B}}{\delta B_{jz_{3}}^{\alpha'}} = G_{B} \cdot \frac{\delta \Sigma_{B}}{\delta B_{jz_{3}}^{\alpha'}} \cdot G_{B} + G_{B} \cdot \frac{\delta T_{B}}{\delta B_{jz_{3}}^{\alpha'}} \cdot G_{B}. \tag{A5}$$

We apply Eq. (A5) to Eq. (8), obtaining

$$\chi_{1,2;j}^{\alpha\alpha'}(z_1, z_2; z_3) \equiv (\chi_0)_{1,2;j}^{\alpha\alpha'}(z_1, z_2; z_3) + (\chi_{\Gamma})_{1,2;j}^{\alpha\alpha'}(z_1, z_2; z_3),$$
(A6)

where

$$(\chi_{0})_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};z_{3}) \equiv -i\left(S^{\alpha}G_{jz_{3}}S_{j}^{\alpha'}G^{jz_{3}}\right)_{2z_{2}}^{1z_{1}}$$

$$= -i\sum_{M}(S^{\alpha}GS^{\alpha'})_{jMz_{3}}^{1z_{1}}G_{2z_{2}}^{jMz_{3}}, \quad (A7)$$

$$(\chi_{\Gamma})_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};z_{3}) \equiv -i\left(S^{\alpha}G\frac{\delta\Sigma_{B}}{\delta B_{jz_{3}}^{\alpha'}}\Big|_{B=0}G\right)_{2z_{2}}^{1z_{1}}$$

$$= -i\int_{\gamma}d(w_{4},w_{5})\sum_{4,5}\left(S^{\alpha}G_{4w_{4}}\right)^{1z_{1}}G_{2z_{2}}^{5w_{5}}$$

$$\times \frac{\delta(\Sigma_{B})_{5w_{5}}^{4w_{4}}}{\delta B_{jz_{3}}^{\alpha'}}\Big|_{B=0}. \quad (A8)$$

We now perform some manipulations on Eq. (A8). If the dimensionality of the spin associated with quantum numbers k is S_k , then a fundamental property of the spin matrices is that

$$\left(\sum_{\alpha''} S_k^{\alpha''} S_k^{\alpha''}\right)_{M'}^M = \delta_{M'}^M \mathcal{S}_k(\mathcal{S}_k + 1), \tag{A9}$$

with $\alpha'' \in \{x, y, z\}$. Using this relation, we obtain

$$\frac{\delta(\Sigma_{B})_{5w_{5}}^{4w_{4}}}{\delta B_{jz_{3}}^{\omega'}} = \int_{\gamma} d(w_{6}, w_{7}) \sum_{6,7} \frac{\delta \Sigma_{5w_{5}}^{4w_{4}}}{\delta G_{7w_{7}}^{6w_{5}}} \frac{\delta(G_{B})_{7w_{7}}^{6w_{6}}}{\delta B_{jz_{3}}^{\omega'}} \\
= \int_{\gamma} d(w_{6}, w_{7}) \sum_{6,7} \frac{1}{S_{i_{4}}(S_{i_{4}} + 1)S_{i_{6}}(S_{i_{6}} + 1)} \\
\times \sum_{\alpha''} \frac{\delta \left(S_{i_{4}}^{\alpha''} S_{i_{4}}^{\alpha''} \Sigma\right)_{5w_{5}}^{4w_{4}}}{\delta G_{7w_{7}}^{6w_{6}}} \sum_{\alpha'''} \frac{\delta \left(S_{i_{6}}^{\alpha'''} S_{i_{6}}^{\alpha'''} G_{B}\right)_{7w_{7}}^{6w_{6}}}{\delta B_{jz_{3}}^{\alpha''}} \\
= \int_{\gamma} d(w_{6}, w_{7}) \sum_{6,7} \frac{1}{S_{i_{4}}(S_{i_{4}} + 1)S_{i_{6}}(S_{i_{6}} + 1)} \\
\times \sum_{\alpha'''} \sum_{M'} \left(S_{i_{4}}^{\alpha''}\right)_{M_{4}}^{M'} \frac{\delta(S^{\alpha''} \Sigma)_{5w_{5}}^{4w_{4}}}{\delta G_{7w_{7}}^{6w_{6}}} \\
\times \sum_{\alpha''''} \sum_{M} \left(S_{i_{6}}^{\alpha'''}\right)_{M_{6}}^{M} \frac{\delta \left(S^{\alpha'''} G_{B}\right)_{7w_{7}}^{6w_{6}}}{\delta B_{jz_{3}}^{\alpha''}}. \quad (A10)$$

Inserting this into Eq. (A8) yields

$$(\chi_{\Gamma})_{1,2;j}^{\alpha\alpha'}(z_{1},z_{2};z_{3})$$

$$\equiv \sum_{\alpha''\alpha'''} \int_{\gamma} d(w_{4},w_{5},w_{6},w_{7}) \sum_{4567} (\chi_{0}^{\alpha\alpha''})_{2z_{2},4w_{4}}^{1z_{1},5w_{5}}$$

$$\times (\Gamma^{\alpha''\alpha'''})_{5w_{5}}^{4w_{4},7w_{7}} \chi_{6,7;}^{\alpha'''\alpha'}(w_{6},w_{7};z_{3}), \tag{A11}$$

where we have introduced the quantities defined in Eqs. (10) and (11). We note here that a more explicit form of

Eq. (11) is

$$(\Gamma^{\alpha''\alpha'''})_{5w_5,6w_6}^{4w_4,7w_7} = \frac{i}{S_{i_4}(S_{i_4} + 1)S_{i_6}(S_{i_6} + 1)} \times \sum_{MM'} \left(S_{i_4}^{\alpha''}\right)_{M'}^{M_4} \frac{\delta \Sigma_{5w_5}^{i_4M'w_4}}{\delta G_{7w_7}^{i_6Mw_6}} \left(S_{i_6}^{\alpha'''}\right)_{M_6}^{M}. \quad (A12)$$

Equation (A7) is related to Eq. (10) via the identity

$$(\chi_0)_{1,2;j}^{\alpha\alpha'}(z_1,z_2;z_3) = \sum_{M} (\chi_0^{\alpha\alpha'})_{2z_2,jMz_3}^{1z_1,jMz_3},$$
(A13)

and the quantity defined in Eq. (12) is related to Eq. (A7) via

$$(\chi_0)_{1,2;i}^{\alpha\alpha'}(z_1,z_2;t')$$

$$\equiv (\chi_0)_{1,2;j}^{\alpha\alpha'}(z_1,z_2;t'_{(+)}) - (\chi_0)_{1,2;j}^{\alpha\alpha'}(z_1,z_2;t'_{(-)}). \tag{A14}$$

By inserting Eq. (A11) into Eq. (A6), one obtains the generalized Bethe-Salpeter equation, given by Eq. (9) of the main text.

APPENDIX B: NONEQUILIBRIUM DYNAMICAL SPIN SUSCEPTIBILITY

To establish the relation between the supermatrix defined in Eq. (8) and the physical susceptibility defined in Eq. (7), it is first convenient to define the quantity

$$\chi_{ij}^{\alpha\alpha'}(z_1, z_2; z_3) \equiv \sum_{M} \chi_{iM, iM; j}^{\alpha\alpha'}(z_1, z_2; z_3).$$
(B1)

We obtain the physical susceptibility from Eqs. (8) and (B1) as follows. From Eq. (B1) we obtain

$$\sum_{j} \sum_{\alpha'} \int_{\gamma} dz_3 \chi_{ij}^{\alpha\alpha'}(z_1, z_2; z_3) \delta B_{jz_3}^{\alpha'}$$

$$= -i \sum_{j} \sum_{\alpha'} \int_{\gamma} dz_3 \operatorname{Sp} \left\{ S_i^{\alpha} \frac{\delta(G_B)_{iz_2}^{iz_1}}{\delta B_{jz_3}^{\alpha'}} \Big|_{B=0} \right\} \delta B_{jz_3}^{\alpha'}$$

$$= \delta \left[-i \operatorname{Sp} \left(S_i^{\alpha} G_{jz_3}^{iz_1} \right) \right]. \tag{B2}$$

This quantity is equal to the variation of the local magnetic moment under a variation of the magnetic field, $\delta \langle \hat{S}_i^{\alpha}(t) \rangle$, if we take $z_1 = t_{(+)}$ and $z_2 = t_{(-)}$. Moving from the KP coordinates to physical times $(z_3 \to t'_{(+)})$ if $z_3 \in \gamma_+$ and $z_3 \to t'_{(-)}$ if $z_3 \in \gamma_-$) gives

$$\delta \langle \hat{S}_{i}^{\alpha}(t) \rangle = \sum_{j} \sum_{\alpha'} \int_{t_{0}}^{\infty} dt' \left[\chi_{ij}^{\alpha \alpha'}(t_{(+)}, t_{(-)}; t'_{(+)}) \, \delta B_{jt'_{(+)}}^{\alpha'} - \chi_{ij}^{\alpha \alpha'}(t_{(+)}, t_{(-)}; t'_{(-)}) \delta B_{jt'_{(-)}}^{\alpha'} \right], \tag{B3}$$

where we have set $\delta B_{jz}^{\alpha'}=0$ if $z\in\gamma_{\mathbb{M}}$. Moreover, the variation of the magnetic field is physically meaningful only if $\delta B_{jt'_{(+)}}^{\alpha'}=\delta B_{jt'_{(-)}}^{\alpha'}\equiv\delta B_{j}^{\alpha'}(t')$. This gives

$$\delta \langle \hat{S}_{i}^{\alpha}(t) \rangle = \sum_{i} \sum_{\alpha'} \int_{t_{0}}^{\infty} dt' \chi_{ij}^{\alpha\alpha'}(t, t') \delta B_{j}^{\alpha'}(t'), \tag{B4}$$

where the physical susceptibility is obtained as

$$\chi_{ij}^{\alpha\alpha'}(t,t') \equiv \chi_{ij}^{\alpha\alpha'}(t_{(+)},t_{(-)};t'_{(+)}) - \chi_{ij}^{\alpha\alpha'}(t_{(+)},t_{(-)};t'_{(-)}).$$
 (B5)

Using Eq. (B1), we immediately obtain that the relation between Eqs. (7) and (8) is given by Eq. (13).

APPENDIX C: SIMPLIFICATION OF THE BETHE-SALPETER EQUATION IN THE CASE OF THE SINGLE-ORBITAL HUBBARD MODEL

We show here the details of the simplification of the THF Bethe-Salpeter equation for transverse susceptibility in the single-orbital Hubbard model (SOH). Using the fact that $[\hat{H}(t), \hat{S}^z] = 0$, thus the total third component of the spin of the system is a good quantum number, we obtain

$$\chi_{iM'',iM';j}^{\alpha'''-}(t;t')$$

$$\stackrel{\text{SOH}}{=} -i\theta(t-t') \sum_{M} (S^{\alpha'''})_{M}^{M''} \delta_{\downarrow}^{M} \delta_{M'}^{\uparrow} \langle \left[\hat{\rho}_{i\uparrow}^{i\downarrow}(t), \hat{S}_{j}^{-}(t') \right] \rangle$$

$$= -i\theta(t-t') \delta_{M'}^{\uparrow} (S^{\alpha'''})_{\downarrow}^{M''} \langle \left[\hat{S}_{i}^{+}(t), \hat{S}_{j}^{-}(t') \right] \rangle$$

$$= \delta_{M'}^{\uparrow} (S^{\alpha'''})_{\downarrow}^{M''} \chi_{ii}^{+-}(t,t'). \tag{C1}$$

Equation (25) simplifies as

$$(\chi_0)_{1,2;j}^{+-}(t,t') \stackrel{\text{SOH}}{=} -i\theta(t-t')\delta_{\uparrow}^{M_1} [(G^{>})_{j\downarrow t'}^{i_1\downarrow t}(G^{<})_{2t}^{j\uparrow t'} - (G^{<})_{j\downarrow t'}^{i_1\downarrow t}(G^{>})_{2t}^{j\uparrow t'}], \tag{C2}$$

from which

$$(\chi_0)_{ij}^{+-}(t,t') \stackrel{\text{SOH}}{=} -i\theta(t-t') \left[(G^>)_{j\downarrow t'}^{i\downarrow t} (G^<)_{i\uparrow t}^{j\uparrow t'} - (G^<)_{i\downarrow t}^{i\downarrow t} (G^>)_{i\uparrow t}^{j\uparrow t'} \right]. \tag{C3}$$

Equation (24) simplifies as

$$\begin{split} \chi_0^{+\alpha''}(t,t'')_{iM,4}^{iM,5} &\overset{\text{SOH}}{=} -i\theta(t-t'')\delta_{\uparrow}^M \delta_{\uparrow}^{M_5}(S^{\alpha''})_{M_4}^{\downarrow} \\ & \times \left[(G^>)_{i_4\downarrow t''}^{i\downarrow t}(G^<)_{i\uparrow t}^{i\varsigma t''} \right. \\ & \left. - (G^<)_{i_4\downarrow t''}^{i\downarrow t}(G^>)_{i\uparrow t}^{i\varsigma t''} \right] \\ & \equiv \delta_{\uparrow}^M \delta_{\uparrow}^{M_5}(S^{\alpha''})_{M_4}^{\downarrow}(\chi_0)_{i,i_4i_5}^{+-}(t,t''). \end{split} \label{eq:continuous} \tag{C4}$$

Using these expressions, from Eq. (23) one obtains Eq. (28).

APPENDIX D: DERIVATION OF THE EQUATIONS FOR THE SUSCEPTIBILITY IN THE FREQUENCY DOMAIN

We show here the detailed derivation of the frequency-domain representations of the Bethe-Salpeter equation, Eq. (28), and the equation for the bare susceptibility, Eq. (31).

We start from the Bethe-Salpeter equation. Equation (28) becomes

$$\widetilde{\chi}_{ij}^{+-}(\tau,T) - (\widetilde{\chi}_0)_{ij}^{+-}(\tau,T)
\stackrel{\text{THF SOH}}{=} \sum_{k} \int_{-\infty}^{\infty} dt''(\widetilde{\chi}_0)_{ik}^{+-} \left(T + \frac{\tau}{2} - t'', \frac{T}{2} + \frac{\tau}{4} + \frac{t''}{2}\right)
\times (-U_k)\widetilde{\chi}_{kj}^{+-} \left(t'' - T + \frac{\tau}{2}, \frac{t''}{2} + \frac{T}{2} - \frac{\tau}{4}\right), \tag{D1}$$

where we have extended the lower boundary of integration over t'' to $-\infty$. Applying the Laplace and Fourier transforms,

we get

$$\widetilde{\chi}_{ij}^{+-}(\omega,\Omega) - (\widetilde{\chi}_0)_{ij}^{+-}(\omega,\Omega)$$

$$\stackrel{\text{THF SOH}}{=} \int_0^\infty d\tau \, e^{i\omega\tau} \int_{-\infty}^\infty dT \, e^{i\Omega T} \sum_k \int_{-\infty}^\infty dt''$$

$$\times (\widetilde{\chi}_0)_{ik}^{+-} \left(T + \frac{\tau}{2} - t'', \frac{T}{2} + \frac{\tau}{4} + \frac{t''}{2}\right)$$

$$\times (-U_k)\widetilde{\chi}_{kj}^{+-} \left(t'' - T + \frac{\tau}{2}, \frac{t''}{2} + \frac{T}{2} - \frac{\tau}{4}\right). \quad (D2)$$

We change variables according to $t'' = \tau' - \frac{\tau}{2} + T$,

$$\widetilde{\chi}_{ij}^{+-}(\omega,\Omega) - (\widetilde{\chi}_0)_{ij}^{+-}(\omega,\Omega)$$

$$\stackrel{\text{THF SOH}}{=} - \int_0^\infty d\tau \ e^{i\omega\tau} \int_{-\infty}^\infty dT \ e^{i\Omega T} \int_{-\infty}^\infty d\tau'$$

$$\times \sum_k (\widetilde{\chi}_0)_{ik}^{+-} \left(\tau - \tau', T + \frac{\tau'}{2}\right)$$

$$\times U_k \, \widetilde{\chi}_{kj}^{+-} \left(\tau', T - \frac{\tau - \tau'}{2}\right). \tag{D3}$$

Using the inverse Fourier transform on the second arguments of the two susceptibilities, we perform the integration over T,

$$\begin{split} \widetilde{\chi}_{ij}^{+-}(\omega,\Omega) &- (\widetilde{\chi}_0)_{ij}^{+-}(\omega,\Omega) \\ \stackrel{\text{THF SOH}}{=} &- \int_0^\infty d\tau \int_{-\infty}^\infty d\tau' \int_{-\infty}^\infty \frac{d\Omega'}{2\pi} e^{i(\omega + \frac{\Omega - \Omega'}{2})\tau} e^{-i\frac{\Omega}{2}\tau'} \\ &\times \sum_k (\widetilde{\chi}_0)_{ik}^{+-}(\tau - \tau';\Omega') U_k \widetilde{\chi}_{kj}^{+-}(\tau';\Omega - \Omega'). \end{split} \tag{D4}$$

For a fixed τ' , we substitute $\sigma = \tau - \tau'$ and we obtain

$$\widetilde{\chi}_{ij}^{+-}(\omega,\Omega) - (\widetilde{\chi}_{0})_{ij}^{+-}(\omega,\Omega)
\stackrel{\text{THFSOH}}{=} - \int_{-\infty}^{\infty} d\tau' \int_{-\tau'}^{\infty} d\sigma \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} e^{i(\omega + \frac{\Omega - \Omega'}{2})\sigma} e^{i(\omega - \frac{\Omega'}{2})\tau'}
\times \sum_{i} (\widetilde{\chi}_{0})_{ik}^{+-}(\sigma;\Omega') U_{k} \widetilde{\chi}_{kj}^{+-}(\tau';\Omega - \Omega').$$
(D5)

Finally, we notice that the integrand vanishes when $\tau' < 0$ because $\widetilde{\chi}(\tau';\ldots) \propto \theta(\tau')$, so we can restrict the integration over τ' to the interval $(0,\infty)$. The integrand also vanishes when $\sigma < 0$ because $\widetilde{\chi}_0(\sigma;\ldots) \propto \theta(\sigma)$, so we can also restrict the integration over σ to the interval $(0,\infty)$. We then recognize two Laplace transforms, and we obtain

$$\widetilde{\chi}_{ij}^{+-}(\omega,\Omega) - (\widetilde{\chi}_0)_{ij}^{+-}(\omega,\Omega)
\stackrel{\text{THFSOH}}{=} - \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} \sum_{k} (\widetilde{\chi}_0)_{ik}^{+-} \left(\omega + \frac{\Omega - \Omega'}{2}, \Omega'\right)
\times U_k \, \widetilde{\chi}_{kj}^{+-} \left(\omega - \frac{\Omega'}{2}, \Omega - \Omega'\right).$$
(D6)

We now treat the equation for the bare susceptibility, Eq. (31). Introducing the Wigner coordinates, we obtain

$$(\widetilde{\chi}_{0})_{ij}^{+-}(\tau,T) \left[i \stackrel{\leftarrow}{\partial_{\tau}} - \frac{i}{2} \stackrel{\leftarrow}{\partial_{T}} - \Delta_{j} \left(T - \frac{\tau}{2} \right) \right]$$

$$\stackrel{\text{SOH THF}}{=} \delta(\tau) \delta_{ij} m_{j}(T) + \widetilde{\Lambda}_{ij}(\tau,T). \tag{D7}$$

We multiply both sides of the previous equation by $e^{i\omega\tau}e^{i\Omega T}$ and we integrate over τ and T, in both cases on the full real axis $(-\infty,\infty)$. We obtain

$$\begin{split} \delta_{ij} m_{j}(\Omega) + \widetilde{\Lambda}_{ij}(\omega, \Omega) \\ &\stackrel{\text{SOH THF}}{=} \int_{-\infty}^{\infty} d\tau \int_{-\infty}^{\infty} dT \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} e^{-i\Omega'T} \\ &\times (\widetilde{\chi}_{0})_{ij}^{+-}(\tau; \Omega') \bigg(i \overleftarrow{\partial_{\tau}} - \frac{\Omega'}{2} \bigg) e^{i\omega\tau} e^{i\Omega T} \\ &- \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} (\widetilde{\chi}_{0})_{ij}^{+-} \bigg(\omega + \frac{\Omega - \Omega'}{2}, \Omega' \bigg) \Delta_{j}(\Omega - \Omega'). \end{split} \tag{D8}$$

We partially integrate on the variable τ , and we note that the boundary terms vanish, respectively, because $\text{Im}(\omega) > 0$ and $(\widetilde{\chi}_0)_{ij}^{+-}(\tau;\Omega') \propto \theta(\tau)$. We then obtain Eq. (36).

APPENDIX E: SIMPLIFICATIONS FOR SPATIALLY PERIODIC SYSTEMS

If the system is spatially periodic (and stays so under the application of the time-dependent external field), it is convenient to write and solve the equations for the susceptibility in wave-vector space. We define the spatial Fourier transforms according to the usual conventions,

$$f_{ij} = \frac{1}{N} \sum_{\mathbf{q}} e^{i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} f_{\mathbf{q}} \Leftrightarrow f_{\mathbf{q}} = \frac{1}{N} \sum_{i,j} e^{-i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} f_{ij},$$

$$g_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{R}_i} g_{\mathbf{q}} \Leftrightarrow g_{\mathbf{q}} = \frac{1}{\sqrt{N}} \sum_{i} e^{-i\mathbf{q} \cdot \mathbf{R}_i} g_{i}.$$
(E1)

By applying $\frac{1}{N} \sum_{i,j} e^{-i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)}$ to both Eqs. (35) and (36), we obtain

$$\widetilde{\chi}_{q}^{+-}(\omega,\Omega) - (\widetilde{\chi}_{0})_{q}^{+-}(\omega,\Omega)
\stackrel{\text{THF SOH}}{=} - \overline{U} \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} (\widetilde{\chi}_{0})_{q}^{+-} \left(\omega + \frac{\Omega - \Omega'}{2}, \Omega'\right)
\times \widetilde{\chi}_{q}^{+-} \left(\omega - \frac{\Omega'}{2}, \Omega - \Omega'\right), \qquad (E2)
\left(\omega - \frac{\Omega}{2}\right) (\widetilde{\chi}_{0})_{q}^{+-}(\omega,\Omega) - \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} (\widetilde{\chi}_{0})_{q}^{+-}
\times \left(\omega + \frac{\Omega - \Omega'}{2}, \Omega'\right) \overline{\Delta} (\Omega - \Omega')
\stackrel{\text{THF SOH}}{=} 2\pi \delta(\Omega) \overline{m} + \widetilde{\Lambda}_{q}(\omega,\Omega), \qquad (E3)$$

where we have introduced the spatial averages

$$\overline{g} \equiv \frac{1}{N} \sum_{i} g_i = \frac{g_{q=0}}{\sqrt{N}},\tag{E4}$$

and we have noticed that the average magnetic moment

$$\frac{1}{N} \sum_{i} m_i(T) \equiv \overline{m} \tag{E5}$$

is independent of time. If $U_i \to \underline{U}$ is spatially uniform, as we shall assume, then also $U\overline{m} = \overline{\Delta}$ is time-independent, then $\overline{\Delta}(\Omega - \Omega') \to 2\pi\delta(\Omega - \Omega')\overline{\Delta}$, and Eq. (E3) can be solved without further approximations:

$$(\widetilde{\chi}_0)_q^{+-}(\omega,\Omega) \stackrel{\text{THF SOH}}{=} \frac{2\pi\delta(\Omega)\overline{m} + \widetilde{\Lambda}_q(\omega,\Omega)}{\omega - \frac{\Omega}{2} - \overline{\Delta}}.$$
 (E6)

By inserting this result into Eq. (E2), applying the adiabatic approximation, and switching to the representation in terms of $(\omega; T)$, we obtain

$$\widetilde{\chi}_{q}^{+-}(\omega;T) \stackrel{\text{THF SOH AD}}{=} \stackrel{\text{ED}}{=} \frac{(\widetilde{\chi}_{0})_{q}^{+-}(\omega;T)}{1 + U(\widetilde{\chi}_{0})_{q}^{+-}(\omega;T)}$$

$$= \frac{\overline{m} + \widetilde{\Lambda}_{q}(\omega;T)}{\omega - [-U\widetilde{\Lambda}_{q}(\omega;T)]}. \quad (E7)$$

If $\widetilde{\Lambda}_q(\omega;T)$ is almost independent of ω at frequencies that are small with respect to the Stoner excitations, we can define the *time-dependent magnon frequency* as

$$\omega_{\mathbf{q}}(T) \equiv -U\widetilde{\Lambda}_{\mathbf{q}}(0;T).$$
 (E8)

APPENDIX F: THE EQUILIBRIUM CASE

In equilibrium, we have the exact identity

$$\widetilde{A}(\omega,\Omega) = 2\pi \delta(\Omega)A(\omega)$$
 (F1)

for the Fourier-Laplace transforms of the many-body functions of τ and T involved in our derivation, since the latter do not depend on the total time T. This is a particular case of the adiabatic regime discussed in the main text, so all the equilibrium results can be immediately recovered from those valid in the adiabatic regime by just removing the dependence of the exchange matrix (and, therefore, of the magnon frequencies) on the total time T. This can also be checked by using Eq. (F1) to simplify Eqs. (36) and (35), and then by solving those equations directly, following exactly the same procedure that is discussed in the main text

In particular, from Eq. (52) we obtain the equilibrium exchange parameters as

$$J_{ij} = i \sum_{iS} \sum_{jS} \lim_{\epsilon \to 0^{+}} \int_{0}^{\infty} dt e^{-\epsilon t} \left[(G^{<})_{j\downarrow 0}^{i\downarrow t} (G^{>})_{i\uparrow t}^{j\uparrow 0} \right] - (G^{>})_{i\downarrow 0}^{i\downarrow t} (G^{<})_{i\uparrow t}^{j\uparrow 0} \right].$$
 (F2)

In the Matsubara representation, it is assumed that the statistical preparation of the initial state follows a thermal distribution. At zero temperature (or $\beta \to \infty$), the above expression is equivalent to

$$J_{ij} = \sum_{iS} \sum_{jS} \lim_{\beta \to \infty} \int_{-\beta}^{\beta} d\tau \ G_{j\downarrow}^{i\downarrow}(\tau) \ G_{i\uparrow}^{j\uparrow}(-\tau), \qquad (F3)$$

where $G(\tau)$ denotes a Matsubara Green's function in the imaginary-time (here denoted as τ) representation. Switching to the representation in terms of Matsubara frequencies ω_n ,

$$G(\tau) = \frac{1}{\beta} \sum_{\omega} e^{-i\omega_n \tau} G(i\omega_n), \tag{F4}$$

as well as using Eq. (32) in the equilibrium case, we obtain Eq. (54).

APPENDIX G: A USEFUL SUM RULE FOR NONEQUILIBRIUM GREEN'S FUNCTIONS

As mentioned in the main text, the fact that our theory is consistent with the Goldstone theorem, even out of equilibrium, can be immediately seen from the fact that

$$\sum_{i} \Lambda_{ij}(t, t') = 0. \tag{G1}$$

The Goldstone theorem can also be checked in an alternative way by using a sum rule that we derive here, valid in and out of equilibrium within the THF approximation. The Dyson equations are given by Eq. (30), in particular

$$-i\partial_{t'}(G^{\lessgtr})_{j\downarrow t'}^{i\downarrow t} \stackrel{\text{THF}}{=} (G^{\lessgtr}T)_{j\downarrow t'}^{i\downarrow t} + (G^{\lessgtr})_{j\downarrow t'}^{i\downarrow t} \Sigma_{j\downarrow}(t'),$$

$$i\partial_{t'}(G^{\gtrless})_{i\uparrow t}^{j\uparrow t'} \stackrel{\text{THF}}{=} (TG^{\gtrless})_{i\uparrow t}^{j\uparrow t'} + \Sigma_{j\uparrow}(t')(G^{\gtrless})_{i\uparrow t}^{j\uparrow t'}.$$
(G2)

We multiply the first equation by $(G^{\gtrless})_{i\uparrow t}^{j\uparrow t'}$ and sum over j; analogously, we multiply the second equation by $(G^{\lessgtr})_{j\downarrow t'}^{i\downarrow t}$ and sum over j. We obtain

$$\sum_{j} (G^{\lessgtr})_{j\downarrow t'}^{i\downarrow t} (-i \overleftarrow{\partial}_{t'}) (G^{\gtrless})_{i\uparrow t'}^{j\uparrow t'}$$

$$\stackrel{\text{THF}}{=} \left[(G^{\lessgtr})_{\downarrow t'}^{\downarrow t} T(t') (G^{\gtrless})_{\uparrow t}^{\uparrow t'} \right]_{i}^{i}$$

$$+ \left[(G^{\lessgtr})_{\downarrow t'}^{\downarrow t} \Sigma_{\downarrow} (t') (G^{\gtrless})_{\uparrow t'}^{\uparrow t'} \right]_{i}^{i}, \qquad (G3)$$

$$\sum_{j} (G^{\lessgtr})_{j\downarrow t'}^{i\downarrow t} (i \overrightarrow{\partial}_{t'}) (G^{\gtrless})_{i\uparrow t'}^{\uparrow t'}$$

$$\stackrel{\text{THF}}{=} \left[(G^{\lessgtr})_{\downarrow t'}^{\downarrow t} T(t') (G^{\gtrless})_{\uparrow t}^{\uparrow t'} \right]_{i}^{i}$$

$$+ \left[(G^{\lessgtr})_{\downarrow t'}^{\downarrow t} \Sigma_{\uparrow} (t') (G^{\gtrless})_{\uparrow t'}^{\uparrow t'} \right]_{i}^{i}. \qquad (G4)$$

We subtract Eq. (G3) from Eq. (G4), divide by 2, and we obtain

$$\frac{1}{2}i\overrightarrow{\partial}_{t'}\left[(G^{\lessgtr})^{\downarrow t}_{\downarrow t'}(G^{\gtrless})^{\uparrow t'}_{\uparrow t}\right]^{i}_{i}$$

$$\stackrel{\text{THF}}{=}\left[(G^{\lessgtr})^{\downarrow t}_{\downarrow t'}\Sigma_{S}(t')(G^{\gtrless})^{\uparrow t'}_{\uparrow t}\right]^{i}_{i}.$$
(G5)

The sum rule Eq. (G5) can be used to immediately check that Eqs. (46) and (47) indeed satisfy

$$\sum_{j} [J_{ij}(t,t') + X_{ij}(t,t')] = 0,$$
 (G6)

which is in agreement with the Goldstone theorem.

- [1] K. Yosida, *Theory of Magnetism* (Springer-Verlag, Berlin, 1996).
- [2] R. M. White, Quantum Theory of Magnetism, 3rd ed. (Springer-Verlag, Berlin, 2007).
- [3] A. I. Liechtenstein, M. I. Katsnelson, and V. A. Gubanov, J. Phys. F 14, L125 (1984).
- [4] A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, and V. A. Gubanov, J. Magn. Magn. Mater. 67, 65 (1987).
- [5] M. I. Katsnelson and A. I. Lichtenstein, Phys. Rev. B 61, 8906 (2000).
- [6] M. I. Katsnelson and A. I. Lichtenstein, Eur. Phys. J. B 30, 9 (2002).
- [7] A. Secchi, A. I. Lichtenstein, and M. I. Katsnelson, Ann. Phys. (N.Y.) 360, 61 (2015).
- [8] A. Secchi, A. I. Lichtenstein, and M. I. Katsnelson, J. Magn. Magn. Mater. 400, 112 (2016).
- [9] M. I. Katsnelson, Y. O. Kvashnin, V. V. Mazurenko, and A. I. Lichtenstein, Phys. Rev. B 82, 100403(R) (2010).
- [10] L. Udvardi, L. Szunyogh, K. Palotás, and P. Weinberger, Phys. Rev. B 68, 104436 (2003).
- [11] A. Szilva, M. Costa, A. Bergman, L. Szunyogh, L. Nordström, and O. Eriksson, Phys. Rev. Lett. 111, 127204 (2013).
- [12] M. I. Katsnelson and A. I. Lichtenstein, J. Phys.: Condens. Matter 16, 7439 (2004).
- [13] P. Bruno, Phys. Rev. Lett. 90, 087205 (2003).
- [14] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. 76, 4250 (1996).
- [15] J. Hohlfeld, E. Matthias, R. Knorren, and K. H. Bennemann, Phys. Rev. Lett. 78, 4861 (1997).
- [16] A. Scholl, L. Baumgarten, R. Jacquemin, and W. Eberhardt, Phys. Rev. Lett. 79, 5146 (1997).
- [17] J. Güdde, U. Conrad, V. Jähnke, J. Hohlfeld, and E. Matthias, Phys. Rev. B **59**, R6608(R) (1999).
- [18] G. P. Zhang and W. Hübner, Phys. Rev. Lett. 85, 3025 (2000).
- [19] A. V. Kimel, A. Kirilyuk, P. A. Usachev, R. V. Pisarev, A. M. Balbashov, and Th. Rasing, Nature 435, 655 (2005).
- [20] M. G. Münzenberg, Nat. Mater. 9, 184 (2010).
- [21] S. L. Johnson et al., Phys. Rev. Lett. 108, 037203 (2012).

- [22] A. Kirilyuk, A. V. Kimel, and Th. Rasing, Rev. Mod. Phys. **82**, 2731 (2010).
- [23] W. Metzner and D. Vollhardt, Phys. Rev. Lett. 62, 324 (1989).
- [24] A. Georges and Gabriel Kotliar, Phys. Rev. B 45, 6479 (1992).
- [25] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. 68, 13 (1996).
- [26] G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).
- [27] D. Sénéchal, D. Perez, and M. Pioro-Ladrière, Phys. Rev. Lett. 84, 522 (2000).
- [28] D. Sénéchal, D. Perez, and D. Plouffe, Phys. Rev. B 66, 075129 (2002).
- [29] C. Gros and R. Valentí, Phys. Rev. B 48, 418 (1993).
- [30] J. K. Freericks, V. M. Turkowski, and V. Zlatić, Phys. Rev. Lett. 97, 266408 (2006).
- [31] H. Aoki, N. Tsuji, M. Eckstein, M. Kollar, T. Oka, and Ph. Werner, Rev. Mod. Phys. 86, 779 (2014).
- [32] M. Eckstein and P. Werner, Phys. Rev. B 82, 115115 (2010).
- [33] M. Balzer and M. Potthoff, Phys. Rev. B 83, 195132 (2011).
- [34] K. Balzer and M. Eckstein, Phys. Rev. B 89, 035148 (2014).
- [35] A. Secchi, S. Brener, A. I. Lichtenstein, and M. I. Katsnelson, Ann. Phys. (N.Y.) 333, 221 (2013).
- [36] O. V. Konstantinov and V. I. Perel', Sov. Phys. JETP 12, 142 (1961).
- [37] L. P. Kadanoff and G. Baym, Quantum Statistical Mechanics (Benjamin, New York, 1962).
- [38] G. Stefanucci and R. van Leeuwen, *Nonequilibrium Many-Body Theory of Quantum Systems* (Cambridge University Press, New York, 2013).
- [39] A. Kamenev, Field Theory of Non-Equilibrium Systems (Cambridge University Press, New York, 2011).
- [40] J. A. Hertz and D. M. Edwards, J. Phys. F 3, 2174 (1973).
- [41] V. Yu. Irkhin and M. I. Katsnelson, J. Phys. C 18, 4173 (1985).
- [42] V. Yu. Irkhin and M. I. Katsnelson, Zh. Eksp. Teor. Fiz. 88, 522 (1985) [Sov. Phys. JETP 61, 306 (1985)].
- [43] P. C. Martin and J. Schwinger, Phys. Rev. 115, 1342 (1959); R. Kubo, J. Phys. Soc. Jpn. 12, 570 (1957).