Non-equilibrium itinerant-electron magnetism: a time-dependent mean-field theory

A. Secchi,1 A. I. Lichtenstein,2 and M. I. Katsnelson1

1Institute for Molecules and Materials, Radboud University Nijmegen, 6525 AJ Nijmegen, The Netherlands
2Institut für Theoretische Physik, Universität Hamburg, Jungiusstraße 9, D-20355 Hamburg, Germany

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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 which 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 non-equilibrium adiabatic regime, obtaining a closed expression for the transverse magnetic susceptibility. From this, we provide a rigorous definition of non-equilibrium (time-dependent) magnon frequencies and exchange parameters, expressed in terms of non-equilibrium single-electron Green functions and self-energies. In the particular case of equilibrium, we recover previously known results.

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The dynamical magnetic susceptibility of an electronic system is a key quantity in both theoretical and experimental studies of magnetism. 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) proper of the initial electronic systems when the magnetic moments undergo small rotations from their initial configuration. In the modern formulation, parameters are expressed in terms of single-electron Green functions (1EGFs) and self-energies. The original methods were recently extended to include unquenched electronic orbital degrees of freedom and relativistic interactions. 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, corrections are required to compute thermodynamic properties.

Experimental progresses allow to modify the magnetic properties of materials by applying time-dependent fields coupling with the electrons, thereby modulating the magnetic interactions in time. Particularly, sub-picosecond laser fields 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 non-equilibrium microscopic theory of magnetism. Computationally, strongly correlated systems are typically treated with Dynamical Mean-Field Theory or cluster perturbation theory and their non-equilibrium formulations. At the moment, the computation of full non-equilibrium two-electron Green functions (2EGFs), such as the dynamical magnetic susceptibility, is not feasible due to huge memory requirements (even the computation of non-equilibrium 1EGFs is, in general, very demanding). To avoid the computation of 2EGFs, the mapping to a dynamical classical spin model has been proposed, where the time-dependent magnetic parameters are expressed in terms of non-equilibrium 1EGFs and self-energies. Also in this case, the connection to the magnetic susceptibility is not obvious.

In this Article we derive the self-consistent equation for the non-equilibrium magnetic susceptibility and 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 Section I we introduce our notation and discuss the features of our non-equilibrium 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 Section II we introduce a generalized Bethe-Salpeter equation for the magnetic susceptibility, valid for arbitrary electronic models. The first two steps of approximations are taken in Section III where we apply the time-dependent Hartree-Fock approximation, and in Section IV where we restrict our theory to the (non-equilibrium) single-band Hubbard model. At this point, the problem can be solved in closed form in equilibrium, but not in the most general non-equilibrium case. The minimal non-equilibrium situation, which allows for a closed solution of the Bethe-Salpeter equa-
tion, corresponds to the adiabatic regime, which we introduce in Section [IV]. 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 Section [V] we characterize the non-equilibrium magnon frequencies by introducing non-equilibrium exchange parameters, and we recover well-known expressions valid in equilibrium as a particular case. In Section [VI] we show that our theory is consistent with the Goldstone theorem even out of equilibrium. Finally, in Section [VII] we summarize our results and mention possible future extensions. In the Appendices we include the most technical passages of the derivations, which can be useful to the reader in order to reproduce our main results, but are not essential to follow the discussion in the main text.

1. NOTATION

We formulate our non-equilibrium theory using the Kostantinov-Perel’ (KP) time contour \( \gamma = \gamma^+ \cup \gamma^- \cup \gamma^0_M \), where \( \gamma^\pm \) is the forward (backward) branch of the real-time (Keldysh) contour \( \gamma_K = \gamma^+ \cup \gamma^- \) and \( \gamma^0_M \) is the imaginary-time (Matsubara) branch\(^{36-39} \). If \( z \) denotes a contour time variable, we write \( z = t(\pm) \) if \( z \) lies on \( \gamma^\pm \), where \( t \in (t_0, \infty) \) denotes a physical time; \( t(+) < t(-) \) on the contour. We use letter \( M \) to denote the third component of spin of the electron fields; \( i, j \) to denote the sets of the other quantum numbers. \( S_t^a \) is the \( \alpha \) component of the vector of spin matrices for the \( i \)-th field, with dimensionality \( S_i \). The non-equilibrium Hamiltonian is

\[
\hat{H}(z) = \sum_{12} \psi_1^{\dagger} T_2(z) \psi_2^2 + \frac{1}{4} \sum_{1234} \psi_1^{\dagger} \psi_2^{\dagger} V_{3,4}^{1,2} \psi_3 \psi_4
\]

for \( z \in \gamma_K \), where \( 1 \equiv (i_1, M_j) \) is a complete set of electron field indices, the single-electron terms depends on the contour coordinate \( z \), and the interaction matrix element is antisymmetrized, \( V_{3,4}^{1,2} = V_{1,3}^{2,4} = -V_{1,4}^{2,3} = -V_{4,3}^{2,1} \). 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

\[
\hat{H}(z) = \hat{H}_{5M},
\]

independent of \( z \) for \( z \in \gamma_M \). 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 \( \hat{H}_{5M} \) 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 \( \hat{H}_{5M} \) 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 magnetic field. Also taking a low temperature, this 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 independently of the particular choice of the Hamiltonian on the Matsubara branch.

1EGFs and 2EGFs are denoted as

\[
G_{2z_2}^{1z_1} \equiv -i \left\langle T_\gamma \psi_1^{\dagger} \psi_2^{\dagger} \right\rangle,
\]

\[
G_{2z_2,4z_2}^{1z_1,3z_3} \equiv (-i)^2 \left\langle T_\gamma \psi_1^{\dagger} \psi_3^{\dagger} \psi_4 \psi_2^{\dagger} \right\rangle,
\]

where \( \left\langle \ldots \right\rangle \) denotes an expectation value computed using the contour evolution operators\(^{36-39} \). The contour 1EGFs are related to the lesser/greater Green functions via

\[
G_{2z_2}^{1z_1} = \Theta(z_1, z_2) \left( G^{>}_{2z_2} \right)^{1z_1}_{2z_2} + \Theta(z_2, z_1) \left( G^{<}_{2z_2} \right)^{1z_1}_{2z_2},
\]

\[
\left( G^{>}_{2z_2} \right)^{1z_1}_{2z_2} = -i \left\langle \psi_1^{\dagger} \psi_2^{\dagger} \right\rangle,
\]

\[
\left( G^{<}_{2z_2} \right)^{1z_1}_{2z_2} = i \left\langle \psi_2^{\dagger} \psi_1^{\dagger} \right\rangle.
\]

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

\[
id_2 G_{2z_2}^{1z_1} - \sum_d \left[ T_\gamma^{d} (z_2) G_{2z_2}^{2d} + \int d z_3 \Sigma^{1z_3}_{2z_2} G_{2z_2}^{2z_3} \right] = \delta^z_2 \delta(z, z_2),
\]

where the self-energy \( \Sigma \) is defined via

\[
\sum_d \int d z_3 \Sigma^{1z_3}_{2z_2} G_{2z_2}^{2z_3} \equiv \frac{1}{2} \sum_{345} V^{3,1}_{4,5} G^{4z_1,5z_3}_{2z_2,3(z_1+z_3)}.
\]

II. GENERALIZED BETHE-SALPETER EQUATION

The dynamical magnetic susceptibility tensor is

\[
\chi_{ij}^{\alpha\alpha'}(t, t') \equiv \frac{\delta \left\langle \hat{S}_i^{\alpha}(t) B \right\rangle}{\delta B_{j}^{\alpha'}(t')} \bigg|_{B=0} = -i \Theta(t-t') \left\langle \left[ \hat{S}_i^{\alpha}(t), \hat{S}_j^{\alpha'}(t') \right] \right\rangle,
\]

where \( \left\langle \ldots \right\rangle_B \) denotes an expectation value computed in the presence of the magnetic field \( B = \{ B_i(t) \} \) coupling with the spins, and \( \hat{S}_i^{\alpha}(t) \) is the \( \alpha \) component of the \( i \)-th spin operator of the system at time \( t \); \( \alpha \in \{ x, y, z \} \) or \( \alpha \in \{ +, - \} \). 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 \( \chi_q^{+}(\omega) \) are the magnon frequencies \( \omega_q \).
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

\[
\chi_{1,2,j}^{\alpha\alpha'}(z_1, z_2; t) \equiv \left( \chi_0^{\alpha\alpha'}(z_1, z_2; t') - \chi_1^{\alpha\alpha'}(z_1, z_2; t_{(z)}) \right),
\]

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).

\[
\chi_{1,2,j}^{\alpha\alpha'}(z_1, z_2; t') = \left( \chi_0^{\alpha\alpha'}(z_1, z_2; t') \right)_{2z_1,5w_5} + \sum_{\alpha''} \chi_{1,2,j}^{\alpha\alpha''}(z_1, z_2; w_5) \delta S^{\alpha''}\delta S^{\alpha'}(z_1, z_2; w_5)_{2z_1,5w_5},
\]

where we have introduced the quantities

\[
\left( \chi_0^{\alpha\alpha'} \right)_{2z_1,5w_5} = -i \left( S^{\alpha'} - S^\dagger \right)_{2z_1,5w_5},
\]

\[
\left( \Gamma^{\alpha\alpha''} \right)_{5w_5,6w_6} = \frac{i}{\delta S^{\alpha''}} \delta \left( S^{\alpha''} \right)_{5w_5,6w_6}.
\]

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

\[
\chi^{\alpha\alpha'}(t, t') = \sum_{s = \pm} \sum_{M} \chi_{M}^{\alpha\alpha'}(t_{(s)}),
\]

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) greatly simplifies its time-domain structure. In THF, the 2EGF is approximated as

\[
G_{B,C}^{A} \equiv G_B^{A} G_C^{A} - G_B^{A} G_C^{A},
\]

which yields the following expression for the self-energy:

\[
\Sigma_{2z_1}^{\alpha\alpha'} = -i\delta(z_1, z_2) \sum_{M} V_{4,2u}^{1,3} \left( G_C^{4u} \right)_{3t_4}.
\]

It should be noted that the 1EGFs appearing in Eqs. (14) and (15) are not the non-interacting Green functions which are used in the 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 functions are the solutions of an interacting problem, although simplified via the THF approximation. This amounts to the only approximation that the self-energy 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

\[
(\chi_0)_{ij}^{\alpha\alpha'}(t, t') = \sum_{M} (\chi_0)_{1M,1M;j}^{\alpha\alpha'}(t_{(i)}; t_{(j)}),
\]

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 which are analogous of those of the Stoner theory for the Hubbard model. In contrast, within RPA Eq. (16) would coincide with the bare magnetic susceptibility of a non-interacting system.

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

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

where

\[
\left( \Gamma^{\alpha\alpha''}_{\text{THF}} \right)_{5w_5,6w_6}^{4w_4,7w_7} = \sum_{M} \sum_{\alpha''} \sum_{\alpha'} \sum_{w_4} \sum_{t_{(i)}} \sum_{t_{(j)}} \sum_{w_5} \sum_{w_6} \sum_{w_7} S^{\alpha''} \delta S^{\alpha'} \delta S^{\alpha''}
\]

\[
\times \left( \chi_0^{\alpha\alpha'} \right)_{2z_1,5w_5} \left( \chi_0^{\alpha\alpha''} \right)_{2z_1,5w_5} \left( \chi_0^{\alpha\alpha'} \right)_{2z_1,5w_5} \left( \chi_0^{\alpha\alpha''} \right)_{2z_1,5w_5}.
\]

Inserting Eq. (18) into Eq. (19) yields the THF form of the GBSE,

\[
\chi_{1,2,j}^{\alpha\alpha'}(z_1, z_2; t') = \left( \chi_0^{\alpha\alpha'}(z_1, z_2; t') \right)_{2z_1,5w_5} + \sum_{\alpha''} \sum_{w_4} \int d\omega_4 \left( \chi_0^{\alpha\alpha''} \right)_{2z_1,5w_5} \left( \Gamma^{\alpha\alpha''}_{\text{THF}} \right)_{5w_5,6w_6}^{4w_4,7w_7} \times \chi_6^{\alpha\alpha''}(w_4 + \epsilon; t').
\]

If \( t_{(i)} \) is the physical time corresponding to the contour coordinate \( w_{4} \), then the quantity

\[
\chi_{6,7,j}^{\alpha\alpha'}(w_4 + \epsilon; t') = \chi_{6,7,j}^{\alpha\alpha'}(t_4; t').
\]
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
\begin{equation}
\chi^{\alpha\alpha'}_{ij}(t, t') = \sum_M \chi^{\alpha\alpha'}_{iM; ij}(t; t').
\end{equation}

Putting $z_1 = t(\alpha)$ and $z_2 = t(-\alpha)$, and
\begin{equation}
\chi^{\alpha\alpha'}_{12j}(t; t') \equiv \chi^{\alpha\alpha'}_{12}(t_{\alpha}(+), t_{\alpha}(-); t'),
\end{equation}
\begin{equation}
\chi^{\alpha\alpha'}_{01j}(t; t') \equiv \chi^{\alpha\alpha'}_{01}(t_{\alpha}(+), t_{\alpha}(-); t'),
\end{equation}
we then obtain, from Eq. (19), the THF GBSE in real-time coordinates:
\begin{equation}
\chi^{\alpha\alpha'}_{12j}(t; t')^{\text{THF}} = \chi^{\alpha\alpha'}_{01j}(t; t')^{\text{THF}} + \sum_{\alpha'\alpha''\alpha'''}\int_0^\infty dt'' \chi^{\alpha\alpha'''}_{07}(t, t''),
\end{equation}
where we have converted the contour integration to physical-time integration, and we have used the fact that $\chi^{\alpha\alpha'}_{7}(w_4, w_4 + \epsilon; t') = 0$ if $w_4 \in \gamma_M$, and we have introduced
\begin{equation}
\chi^{\alpha\alpha'''}_{07}(t, t') = \chi^{\alpha\alpha'''}_{07}(t),
\end{equation}
where
\begin{equation}
\chi^{\alpha\alpha'''}_{07}(t) = \chi^{\alpha\alpha'''}_{07}(t)\chi^{\alpha\alpha'''}_{07}(t).
\end{equation}

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 $S = 1/2$ at every site, and the interaction Hamiltonian becomes
\begin{equation}
\hat{V} = \frac{1}{4} \sum_{3456} V_{3456}^{\text{SOH}} \hat{n}_{3\uparrow} \hat{n}_{4\downarrow} \hat{\psi}_{5\uparrow} \hat{\psi}_{6\downarrow},
\end{equation}
which implies
\begin{equation}
V_{3456}^{\text{SOH}} = \delta_{i8} \delta_{i8} \delta_{46} \delta_{36} \delta_{M3} \delta_{M6} \left( \delta_{M3} - \delta_{M6} \right) U_{34}.
\end{equation}

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
\begin{equation}
\chi^{\alpha\alpha'}_{ij}(t, t') = \chi^{\alpha\alpha'}_{ij}(t, t')^{\text{SOH}} + \sum_k \int_0^\infty dt'' \chi^{\alpha\alpha'}_{ik}(t', t'') (-U_k) \chi^{\alpha\alpha'}_{kj}(t'', t),
\end{equation}
which is the non-equilibrium SOH version of the equation used in Ref. [2]. 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
\begin{equation}
\chi^{\alpha\alpha'}_{ij}(t, t')^{\text{SOH}} = i\theta(t - t') \left[ (G^\uparrow)^{ij\uparrow} (G^\downarrow)^{ij\downarrow} - (G^\downarrow)^{ij\downarrow} (G^\uparrow)^{ij\uparrow} \right],
\end{equation}
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 as
\begin{equation}
- i\partial_t \left( G^\uparrow \right)^{ij\uparrow} = \text{THF} \left( T \cdot G^\uparrow \right)^{ij\uparrow} + \left( G^\uparrow \right)^{ij\uparrow} \Sigma^{jM}(t'),
\end{equation}
\begin{equation}
- i\partial_t \left( G^\downarrow \right)^{ij\downarrow} = \text{THF} \left( T \cdot G^\downarrow \right)^{ij\downarrow} + \Sigma^{jM}(t') \left( G^\uparrow \right)^{ij\uparrow},
\end{equation}
where $\Sigma^{jM}(t') = \sum j \rho_{jM}(t')$ is the THF self-energy for the SOH model, with $\rho_{jM}(t') \equiv \langle \hat{\psi}_{j\uparrow}(t') \hat{\psi}_{j\downarrow}(t') \rangle$. We then obtain
\begin{equation}
\chi^{\alpha\alpha'}_{ij}(t, t')^{\text{THF}} = \delta(t - t') \left[ -i\partial_t - \Delta_j(t') \right],
\end{equation}
where $m_j(t') \equiv \rho_{j\uparrow}(t') - \rho_{j\downarrow}(t')$, and
\begin{equation}
\Delta_j(t') \equiv U_j m_j(t') \equiv -2\Sigma_j(t') \equiv \Sigma_{j\uparrow}(t') - \Sigma_{j\downarrow}(t'),
\end{equation}
is the time-dependent Stoner splitting, and
\begin{equation}
\Lambda_{ij}(t, t') = i\theta(t - t') \left[ (G^\uparrow)^{ij\uparrow} \left( T \cdot G^\uparrow \right)^{ij\downarrow} - (G^\downarrow)^{ij\downarrow} \left( T \cdot G^\downarrow \right)^{ij\uparrow} - (G^\uparrow)^{ij\uparrow} \left( G^\downarrow \right)^{ij\downarrow} + (G^\downarrow)^{ij\downarrow} \left( G^\uparrow \right)^{ij\uparrow} \right].
\end{equation}
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 NON-EQUILIBRIUM MAGNONS

We introduce the Wigner time coordinates, \( \tau \equiv t - t' \) and \( T \equiv (t + t')/2 \), which are respectively called the relative time and the total time. 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 put \( f(t, t') \equiv \tilde{f}(\tau, T) \) 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 \( \tau \) and the Fourier transform with respect to \( T \). We use the notation

\[
\tilde{f}(\omega, \Omega) \equiv \int_{-\infty}^{\infty} dT \, e^{i\Omega T} \int_{0}^{\infty} d\tau \, e^{i\omega \tau} \tilde{f}(\tau, T),
\]

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

\[
\tilde{\chi}_{ij}^{\perp}(\omega, \Omega) - (\tilde{\chi}_0)_{ij}^{\perp}(\omega, \Omega) \equiv -\int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} \sum_{k} (\tilde{\chi}_0)_{ik}^{\perp} \left( \omega + \Omega - \Omega' \right) U_k \tilde{\chi}_{kj}^{\perp} \left( \omega - \Omega', \Omega - \Omega' \right),
\]

where

\[
\lambda_{ij}(\omega) \equiv \omega - \Delta_{ij}(T) \equiv \frac{\delta_{ij} m_j(T) + \tilde{\Lambda}_{ij}(\omega; T)}{\omega - \lambda_{ij}(T)},
\]

while Eq. (33) simplifies into

\[
\sum_{k} \left[ \delta_{ik} + (\tilde{\chi}_0)_{ik}^{\perp}(\omega; T) U_k \right] \tilde{\chi}_{kj}^{\perp}(\omega; T) \equiv \tilde{\chi}_{ij}(\omega; T).
\]

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

\[
\tilde{\chi}_{ij}^{\perp}(\omega; T) \equiv \omega - \Delta_{ij}(T) \sum_{k} U_k F_{ik}^{-1}(\omega; T) \cdot \left[ \delta_{kj} m_j(T) + \tilde{\Lambda}_{kj}(\omega; T) \right],
\]

where we have introduced the matrix

\[
F_{ik}(\omega; T) \equiv \delta_{ik} \omega + U_i \tilde{\Lambda}_{ik}(\omega; T)
\]

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

\[
\sum_{i} F^{-1}_{li}(\omega; T) F_{ik}(\omega; T) = \delta_{lk}.
\]

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

\[
\Omega_{ij}(T) \equiv -U_i \tilde{\Lambda}_{ij}(0; T).
\]

The eigenvalues of \( \Omega \) can then be called non-equilibrium 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 which is obtained when the Hamiltonian is time-independent, \( \Omega_{ij} \) 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 non-equilibrium situations, valid in the adiabatic regime, consists in the fact that the magnon frequencies acquire a time dependence.

We note that the approximation which produces Eq. (12), namely replacing \( \tilde{\Lambda}_{ij}(\omega; T) \to \tilde{\Lambda}_{ij}(0; T) \), corresponds to linearizing the eigenvalue problem associated...
with Eq. (40). Corrections can be computed by keeping into account higher-order terms in the Taylor expansion of \( \tilde{\Lambda}_{ij}(\omega; T) \) in powers of \( \omega \); such analysis is beyond the scope of this work.

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

VI. NON-EQUILIBRIUM 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

\[
\Omega_{ij}(t, t') = \frac{T \mathrm{SH}}{2} \equiv -U_i \Lambda_{ij}(t, t'),
\]

and we express it in terms of non-equilibrium 1EGFs and self-energies. To this end, we use the non-equilibrium Dyson equations in the THF approximation, Eqs. (30), to eliminate the hopping matrix \( T \) from the expression of \( \Lambda \), Eq. (33). We obtain

\[
\Lambda_{ij}(t, t') \equiv \frac{i \theta(t - t')} {2 \Sigma_{ij}(t') - i \partial_{t'}} \left[ \left( G_{\uparrow}^<(t, t') \right)^{it} \left( G_{\uparrow}^>(t, t') \right)^{jt'} - \left( G_{\uparrow}^<(t, t') \right)^{jt'} \left( G_{\uparrow}^>(t, t') \right)^{it} \right].
\]

We split the exchange matrix into two parts,

\[
\Omega_{ij}(t, t') \equiv \frac{4} {m_i(t)} \left[ \tilde{\Omega}_{ij}(t, t') + X_{ij}(t, t') \right],
\]

where

\[
\tilde{\Omega}_{ij}(t, t') \equiv \frac{i \theta(t - t')} {2 \Sigma_{ij}(t') - i \partial_{t'}} \left[ \left( G_{\uparrow}^<(t, t') \right)^{it} \left( G_{\uparrow}^>(t, t') \right)^{jt'} - \left( G_{\uparrow}^<(t, t') \right)^{jt'} \left( G_{\uparrow}^>(t, t') \right)^{it} \right].
\]

B. One-time exchange parameters

If \( \tilde{\Omega}_{ij}(\omega; T) \) is almost independent of \( \omega \), we can determine a time-dependent pole of the non-equilibrium is the two-times exchange parameter (equivalent to the analogous quantity obtained in Ref. 33), and

\[
X_{ij}(t, t') \equiv \frac{1} {2} \Sigma_{ij}(t') \left[ \left( G_{\uparrow}^>(t, t') \right)^{it} \left( G_{\uparrow}^>(t, t') \right)^{jt'} - \left( G_{\uparrow}^<(t, t') \right)^{jt'} \left( G_{\uparrow}^<(t, t') \right)^{it} \right].
\]

We simplify the second term in the RHS of Eq. (48); after performing partial integration and using the relation \( (G^>)^i_{j,T} = -i \delta^i_j + (G^<)^i_{j,T} \), we obtain

\[
\tilde{\Omega}_{ij}(\omega; T) \equiv \frac{4} {m_i(T)} \left[ \tilde{J}_{ij}(\omega; T) + \tilde{X}_{ij}(\omega; T) \right].
\]

VI B. Switching again to the Wigner-coordinates representation and Laplace transforming with respect to relative time, we obtain

\[
\tilde{J}_{ij}(\omega; T) = -\frac{1} {2} \delta_{ij} \Sigma_{iS}(T) m_i(T)
\]

\[
+ \frac{1} {2} \Sigma_{iS}(T) \left[ \left( \frac{1} {2} \partial_T + i \omega \right) \right] \int_0^\infty \mathrm{d} \tau e^{i \omega \tau}
\]

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

The first term in the RHS of Eq. (48) involves the Laplace transform of the two-times exchange parameters,

\[
\tilde{J}_{ij}(\omega; T) \equiv i \Sigma_{iS}(T) \int_0^\infty \mathrm{d} \tau e^{i \omega \tau} \Sigma_{jS}(T - \tau/2) \left[ \left( G_{\uparrow}^>(t, T - \tau/2) \right)^{i,T+\tau/2} \left( G_{\uparrow}^>(t, T - \tau/2) \right)^{j,T+\tau/2} - \left( G_{\uparrow}^<(t, T - \tau/2) \right)^{i,T+\tau/2} \left( G_{\uparrow}^<(t, T - \tau/2) \right)^{j,T+\tau/2} \right].
\]
The equilibrium regime is a particular case of the adiabatic regime, such that 1EGFs depend only on the relative time \( \tau \) 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 \( \beta \rightarrow \infty \)) we can apply the analytical continuation from the real-time branches of the KP contour to the imaginary-time branch, and represent 1EGFs in the Matsubara formalism. In this case, we obtain [details are given in Appendix F]

\[
J_{ij}(T) = i \sum_{\tau} \lim_{\epsilon \to 0^+} \int_0^\infty d\tau e^{-\epsilon\tau} \sum_j \sum_{\epsilon>0} \left[ \left( G_{ij}^\downarrow \right)_{T+\tau/2} \left( G_{ji}^\uparrow \right)_{T-\tau/2} - \left( G_{ij}^\uparrow \right)_{T+\tau/2} \left( G_{ji}^\downarrow \right)_{T-\tau/2} \right].
\]

\[
X_{ij}(T) = -\frac{1}{2} \delta_{ij} \sum_j \sum_{\epsilon>0} \left( \left( G_{ij}^\downarrow \right)_{T+\tau/2} \left( G_{ji}^\uparrow \right)_{T-\tau/2} - \left( G_{ij}^\uparrow \right)_{T+\tau/2} \left( G_{ji}^\downarrow \right)_{T-\tau/2} \right).
\]

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 \( J_{ij}(T) \) given in Eq. (52) as the time-dependent exchange parameter due to its non-locality 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, and 8). The term \( X_{ij}(T) \) 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 Section VI C), of which this is the non-equilibrium generalization. The second line is a purely (non-local) non-equilibrium term with no analogue 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 Section 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 \( \tau \) 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 \( \beta \rightarrow \infty \)) we can apply the analytical continuation from the real-time branches of the KP contour to the imaginary-time branch, and represent 1EGFs in the Matsubara formalism. In this case, we obtain [details are given in Appendix F]

\[
\Omega_{ij}(T) = \lim_{\epsilon \to 0^+} \lim_{\omega \to 0} \frac{1}{4} \sum_j \left( \left( G_{ij}^\downarrow \right)_{T+\tau/2} \left( G_{ji}^\uparrow \right)_{T-\tau/2} - \left( G_{ij}^\uparrow \right)_{T+\tau/2} \left( G_{ji}^\downarrow \right)_{T-\tau/2} \right).
\]

where \( \omega \) and \( \epsilon > 0 \) are real, and

\[
J_{ij} = \frac{1}{2} \sum \sum \left( \left( G_{ij}^\downarrow \right)_{T+\tau/2} \left( G_{ji}^\uparrow \right)_{T-\tau/2} - \left( G_{ij}^\uparrow \right)_{T+\tau/2} \left( G_{ji}^\downarrow \right)_{T-\tau/2} \right),
\]

\[
X_{ij} = \frac{1}{4} \delta_{ij} \Delta_i m_i.
\]

This result agrees with the equilibrium formulas derived with different methods in Refs. 5, 6, and 35, specialized to the SOH model in the HF approximation. We see that Eq. (53) is the non-equilibrium 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 \( \mathbf{q} = \mathbf{0} \) (that is, \( \lim_{\mathbf{q} \to \mathbf{0}} \omega_q = 0 \)). We recover this result in our theory, even out of equilibrium, since it immediately follows from Eqs. (52) and (53) that

\[
\sum_j \Omega_{ij}(t, t') = 0 \Rightarrow \sum_j \Omega_{ij}(t, t') = 0,
\]

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 eigenvector corresponds indeed to the state with \( \mathbf{q} = \mathbf{0} \)). 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 [C].

The Goldstone theorem suggests a possible alternative definition for the exchange parameters contributing to the (one-time) exchange matrix. We can define \textit{stared} exchange parameters by combining Eq. (42) and the non-local part of Eq. (53) (second line). We get:

\begin{equation}
J_{ij}^\ast(T) = i \Sigma_{ij}(T) \lim_{\epsilon \to 0^+} \int_0^\infty d\tau e^{-\epsilon \tau} \left[ \Sigma_{ij}(T - \tau/2) - \frac{i}{4} \frac{\partial}{\partial \tau} \right] \left( G_i \g{}_{ij}^{T}(T - \tau/2) \right) \left( G_j \g{}_{ij}^{T}(T - \tau/2) \right) \left( G_i \g{}_{ij}^{T}(T + \tau/2) \right) \left( G_j \g{}_{ij}^{T}(T + \tau/2) \right).
\end{equation}

Combining this definition with Eq. (54), we can re-write Eq. (51) in terms of \( J^\ast \) only as

\begin{equation}
\Omega_{ij}(T) = \frac{-4}{m_i(T)} \left[ J_{ij}^\ast(T) - \delta_{ij} \sum_k J_{ik}^\ast(T) \right].
\end{equation}

VIII. SUMMARY

To summarize, we have presented a rigorous derivation of the transverse spin susceptibility in the non-equilibrium adiabatic regime for the SOH model within the THF approximation, leading to the definition of non-equilibrium 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. (49).

Concerning the possibility of developing a non-equilibrium theory beyond the THF approximation, we mention that using exact Greens functions but neglecting the vertices is not acceptable because it would break the Goldstone theorem. 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] and [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 Greens functions beyond Hartree-Fock was possible due to the assumed smallness of either the concentration of holes, or the inverse number of nearest neighbours, which allowed a linear approximation in one of those parameter. The generalization of this technique to the non-equilibrium regime is beyond the scope of the present work, where we have instead focused on obtaining the THF-approximated results without making any assumption on the electronic configuration.

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Appendix A: Derivation of the generalized Bethe-Salpeter equation

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

\begin{equation}
\sum_3 \int \gamma d\gamma \left( \overrightarrow{G}^{-1}_{B} \right)_{3z_{3}}^{1z_{1}} \left( G_{B} \right)_{2z_{2}}^{3z_{3}} = \delta_{2z_{1}}^{1z_{2}},
\end{equation}

\begin{equation}
\sum_3 \int \gamma d\gamma \left( G_{B} \right)_{2z_{2}}^{1z_{1}} \left( \overrightarrow{-G}^{-1}_{B} \right)_{3z_{3}}^{2z_{2}} = \delta_{2z_{1}}^{1z_{2}},
\end{equation}

where

\begin{equation}
\left( \overrightarrow{G}^{-1}_{B} \right)_{3z_{3}}^{1z_{1}} = \delta_{2z_{1}}^{1z_{2}} \delta(z_{1}, z_{3}) \overrightarrow{\partial}_{z_{3}} - (T_{B})_{3z_{3}}^{1z_{1}} - \left( \Sigma_{B} \right)_{3z_{3}}^{1z_{1}},
\end{equation}

\begin{equation}
\left( \overrightarrow{-G}^{-1}_{B} \right)_{2z_{2}}^{3z_{3}} = - \overrightarrow{\partial}_{z_{3}} \delta_{2z_{1}}^{1z_{2}} \delta(z_{3}, z_{2}) - (T_{B})_{2z_{2}}^{3z_{3}} - \left( \Sigma_{B} \right)_{2z_{2}}^{3z_{3}}.
\end{equation}

Here \( \Sigma_{B} \) 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,

\begin{equation}
(T_{B})_{3z_{3}}^{1z_{1}} = \delta(z_{1}, z_{3}) \left\{ \left( T(z_{1}) \right)^{1}_{3} + \left( \delta \right)_{3}^{1} B_{3i} z_{i} \right\} \left( S_{z_{1}} \right)_{M_{3}}^{M_{1}},
\end{equation}
where $|T(z_1)|^2$ 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

$$G_{jz}^{-1} \cdot G_B = 1 \Rightarrow \frac{\delta G_{jz}^{-1}}{\delta B_{jz}^{\alpha}} \cdot G_B + G_{jz}^{-1} \cdot \frac{\delta G_B}{\delta B_{jz}^{\alpha}} = 0 \Rightarrow G_B \cdot G_{jz}^{-1} \cdot \frac{\delta G_B}{\delta B_{jz}^{\alpha}} = -G_B \cdot \frac{\delta G_{jz}^{-1}}{\delta B_{jz}^{\alpha}} \cdot G_B. \quad (A4)$$

We can replace $G_B \cdot G_{jz}^{-1} \Rightarrow G_B \cdot G_B^{-1} \equiv 1$, since the two expressions differ only by boundary terms which vanish due to the Kubo-Martin-Schwinger relations on the KP contour. We then obtain the identity

$$\frac{\delta G_B}{\delta B_{jz}^{\alpha}} = G_B \cdot \frac{\delta \Sigma_B}{\delta B_{jz}^{\alpha}} \cdot G_B + G_B \cdot \frac{\delta T_B}{\delta B_{jz}^{\alpha}} \cdot G_B. \quad (A5)$$

We apply Eq. (A5) to Eq. (3), obtaining

$$\chi(\alpha, z_1, z_2; z_3) \equiv (\chi_0(\alpha, z_1, z_2; z_3) + \chi_{\Gamma}(\alpha, z_1, z_2; z_3), \quad (A6)$$

where

$$(\chi_0)_{\alpha, z_1, z_2; z_3} \equiv -i \left( S^\alpha \cdot G_{jz} \cdot S_j^\alpha \cdot G^{jz} \right)_{\alpha, z_1, z_2; z_3}^{12} \quad (A7)$$

$$\chi_{\Gamma}(\alpha, z_1, z_2; z_3) \equiv -i \left( S^\alpha \cdot G \cdot \frac{\delta \Sigma_B}{\delta B_{jz}^{\alpha}} \right)_{\alpha, z_1, z_2; z_3}^{12} \quad (A8)$$

$$= -i \int \gamma d(w_4, w_5) \sum_{4,5} \left( S^\alpha \cdot G_{4w_4} \right)_{\alpha, z_1, z_2; z_3}^{12} \frac{\delta \Sigma_B^{4w_4}}{\delta B_{jz}^{\alpha}} \bigg|_{B=0} \quad (A9)$$

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

$$\left( \sum_{\alpha''} S_k^{\alpha''} \cdot S_k^{\alpha''} \right)^M = \delta_{M, S_k} (S_k + 1), \quad (A10)$$

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

$$\delta (\Sigma_B)_{4w_4}^{5w_5} = \int \gamma d(w_6, w_7) \sum_{6,7} \frac{\delta \Sigma_B^{4w_4}}{\delta G_{6w_6}^{5w_5}} \delta (G_B)_{7w_7}^{6w_6} \quad (A11)$$

$$= \int \gamma d(w_6, w_7) \sum_{6,7} \frac{\delta (\Sigma_B)_{4w_4}}{\delta G_{6w_6}^{5w_5}} \frac{\delta (G_B)_{7w_7}^{6w_6}}{\delta G_{7w_7}^{6w_6}} \frac{1}{(S_{i4} + 1) S_{i6} (S_{i6} + 1)} \quad (A12)$$

$$\delta (\Sigma_B)_{4w_4}^{5w_5} = \int \gamma d(w_6, w_7) \sum_{6,7} \frac{\delta (\Sigma_B)_{4w_4}}{\delta G_{6w_6}^{5w_5}} \frac{\delta (G_B)_{7w_7}^{6w_6}}{\delta G_{7w_7}^{6w_6}} \frac{1}{(S_{i4} + 1) S_{i6} (S_{i6} + 1)} \quad (A13)$$

where we have introduced the quantities defined in Eqs. (1) and (2) of the main text. We note here that a more explicit form of Eq. (1) is

$$\Gamma_{\alpha'' \alpha'} (4w_4, 7w_7)_{5w_5, 6w_6} (2w_2, 4w_4)_{\alpha'' \alpha'} = \frac{1}{5w_5, 6w_6} \sum_{45} \chi_{\alpha'' \alpha'}^{2w_2} \chi_{\alpha'' \alpha'}^{12} \quad (A14)$$

Equation (A7) is related to Eq. (1) via the identity

$$(\chi_0)_{1,2, j}^{\alpha, \alpha'} (z_1, z_2; z_3) = \sum_{M} \left( \chi_0^{12} \right)_{2j,2z3}^{12} \quad (A15)$$

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

$$(\chi_0)_{1,2, j}^{\alpha, \alpha'} (z_1, z_2; t') \equiv (\chi_0)_{1,2, j}^{\alpha, \alpha'} (z_1, z_2; t_1^{+}) - (\chi_0)_{1,2, j}^{\alpha, \alpha'} (z_1, z_2; t_1^{-}). \quad (A16)$$

By inserting Eq. (A11) into Eq. (A9), one obtains the generalized Bethe-Salpeter equation, given by Eq. (2) of the main text.
Appendix B: Non-equilibrium dynamical spin susceptibility

In order to establish the relation between the supermatrix defined in Eq. (3) and the physical susceptibility defined in Eq. (1) of the main text, it is first convenient to define the quantity

$$\chi^\alpha_{ij} (z_1, z_2; z_3) \equiv \sum_M \chi^\alpha_{iM,iM,j} (z_1, z_2; z_3). \tag{B1}$$

We obtain the physical susceptibility from Eqs. (3) and (B1) as follows. From Eq. (B1) we get

$$\sum_j \sum_{\alpha'} \int d_z \chi^\alpha_{ij} (z_1, z_2; z_3) \delta B^\alpha_{jz} = -i \sum_j \sum_{\alpha'} \int d_z \delta \left( \frac{G^\alpha}{B^\alpha_{jz}} \right) \sum_{\gamma} \langle \chi_{ij}^{\alpha'} (t, t'; \gamma) \rangle \delta B^\alpha_{jz} = \delta \left[ -i \langle \chi_{ij}^{\alpha} (t, t') \rangle \right]. \tag{B2}$$

This quantity is equal to the variation of the local magnetic moment under a variation of the magnetic field, $\delta \langle S^z_i (t) \rangle$, 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

$$\delta \langle \hat{S}^z_i (t) \rangle = \sum_j \sum_{\alpha'} \int_0^\infty dt' \chi^\alpha_{ij} (t_{(+)}; t_{(-)}; t_{(+)}) \delta B^\alpha_{jz} = -\chi^\alpha_{ij} (t_{(+)}; t_{(-)}; t_{(+)}) \delta B^\alpha_{jz}, \tag{B3}$$

where we have put $\delta B^\alpha_{jz} = 0$ if $z \in \gamma_M$. Moreover, the variation of the magnetic field is physically meaningful only if $\delta B^\alpha_{jz} = 0$. This gives

$$\delta \langle \hat{S}^z_i (t) \rangle = \sum_j \sum_{\alpha'} \int_0^\infty dt' \chi^\alpha_{ij} (t, t') \delta B^\alpha_{jz} (t'), \tag{B4}$$

where the physical susceptibility is obtained as

$$\chi^\alpha_{ij} (t, t') = \chi^\alpha_{ij} (t_{(+)}; t_{(-)}; t_{(+)}) - \chi^\alpha_{ij} (t_{(+)}; t_{(-)}; t_{(-)}). \tag{B5}$$

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

Appendix C: Simplification of the Bethe-Salpeter equation in the case of the single-orbital Hubbard model

We here show 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 $[\hat{H}(t), \hat{S}^z] = 0$, so the total third component of the spin of the system is a good quantum number, we obtain

$$\chi^{\alpha''}_{iM,iM,j} (t; t') \equiv -i \theta (t - t') \sum_M \left( S^{\alpha''}_{ij} \right)^{M'}_M \delta^M \delta^j_{M'} \left[ \langle \hat{S}^\alpha_{ij} (t), \hat{S}^\gamma_{ij} (t') \rangle \right]$$

$$= -i \theta (t - t') \delta^j_D \left( S^{\alpha''}_{ij} \right)^{M''}_{M'} \left[ \langle \hat{S}^\alpha_{ij} (t), \hat{S}^\gamma_{ij} (t') \rangle \right]$$

$$= \delta^j_D \left( S^{\alpha''}_{ij} \right)^{M''}_{M'} \chi^{\alpha''}_{ij} (t, t'). \tag{C1}$$

Equation (24) simplifies as

$$(\chi_0)_{ij}^{\alpha''} (t, t') \equiv -i \theta (t - t') \delta^j_D \left( S^{\alpha''}_{ij} \right)^{M''}_{M'} \left[ \langle \hat{G}^\alpha_{ij} (t), \hat{G}^\gamma_{ij} (t') \rangle \right]$$

$$= \delta^j_D \left( S^{\alpha''}_{ij} \right)^{M''}_{M'} \chi^{\alpha''}_{ij} (t, t'). \tag{C2}$$

from which

$$(\chi_0)_{ij}^{\alpha''} (t, t') \equiv -i \theta (t - t') \left[ \langle \hat{G}^\alpha_{ij} (t), \hat{G}^\gamma (t') \rangle \right]$$

$$= \langle \hat{G}^\alpha_{ij} (t), \hat{G}^\gamma (t') \rangle. \tag{C3}$$

Equation (24) simplifies as

$$(\chi_0)_{ij}^{\alpha''} (t, t'') \equiv -i \theta (t - t'') \delta^j_D \left( S^{\alpha''}_{ij} \right)^{M''}_{M'} \left[ \langle \hat{G}^\alpha_{ij} (t), \hat{G}^\gamma (t') \rangle \right]$$

$$= \delta^j_D \left( S^{\alpha''}_{ij} \right)^{M''}_{M'} \chi^{\alpha''}_{ij} (t, t''). \tag{C4}$$

Using these expressions, from Eq. (24) one obtains Eq. (28) of the main text.

Appendix D: Derivation of the equations for the susceptibility in the frequency domain

We here show 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

$$\tilde{\chi}_{ij}^{\alpha''} (\tau, T) - (\tilde{\chi}_0)_{ij}^{\alpha''} (\tau, T) \equiv \sum_k \int_{-\infty}^\infty dt'' \left( \tilde{\chi}_0^{\alpha''}_{ik} (T + \frac{\tau}{2} - t'', T + \frac{\tau}{4} + t'' \right)$$

$$\times \left[ -(UK) \chi_{kj}^{\alpha''} (T'' - T + \frac{\tau}{2} - t'', T'' - \frac{\tau}{4} + t''), \right) \tag{D1}$$
where we have extended the lower boundary of integration over \( t'' \) to \(-\infty\). Applying the Laplace and Fourier transforms, we get
\[
\chi^{++}_{ij}(\omega, \Omega) - (\check{\chi}_0)_{ij}^{++}(\omega, \Omega) = \int_0^\infty dt e^{i\omega t} \int_0^\infty d\Omega e^{i\Omega t} \sum_k \int_0^\infty dt'' \times (\check{\chi}_0)_{ik}^{++} \left(T + \frac{t'}{2} - t'', \frac{T}{2} + \frac{t''}{4} + \frac{t'}{4}\right) \times (-U_k) \check{\chi}_{kj}^{++} \left(t'' - T + \frac{t''}{2} + \frac{T}{2} - \frac{\tau}{4}\right).
\]
We change variables according to \( t'' = \tau' - \frac{\tau}{2} + T \),
\[
\chi^{++}_{ij}(\omega, \Omega) - (\check{\chi}_0)_{ij}^{++}(\omega, \Omega) = -\int_0^\infty d\tau e^{i\omega \tau} \int_0^\infty d\Omega e^{i\Omega \tau} \int_0^\infty d\tau' \times \sum_k (\check{\chi}_0)_{ik}^{++} \left(\tau - \tau', T + \frac{\tau'}{2}\right) \times U_k \check{\chi}_{kj}^{++} \left(\tau', T - \frac{\tau'}{2}\right).
\]
Using the inverse Fourier transform on the second arguments of the two susceptibilities, we perform the integration over \( T \),
\[
\check{\chi}_{ij}^{++}(\omega, \Omega) - (\check{\chi}_0)_{ij}^{++}(\omega, \Omega) = -\int_0^\infty d\tau \int_0^\infty d\sigma \int_0^\infty d\Omega' \frac{e^{i(\omega + \frac{\Omega}{2})\tau}}{2\pi} e^{i(\omega - \frac{\Omega}{2})\tau'} \times \sum_k (\check{\chi}_0)_{ik}^{++} (\sigma; \Omega') U_k \check{\chi}_{kj}^{++} (\tau'; \Omega - \Omega') .
\]
For a fixed \( \tau' \), we substitute \( \sigma = \tau - \tau' \) and we obtain:
\[
\check{\chi}_{ij}^{++}(\omega, \Omega) - (\check{\chi}_0)_{ij}^{++}(\omega, \Omega) = -\int_0^\infty d\tau' \int_0^\infty d\sigma \int_0^\infty d\Omega' \frac{e^{i(\omega + \frac{\Omega}{2})\tau}}{2\pi} e^{i(\omega - \frac{\Omega}{2})\tau'} \times \sum_k (\check{\chi}_0)_{ik}^{++} (\sigma; \Omega') U_k \check{\chi}_{kj}^{++} (\tau'; \Omega - \Omega') .
\]
Finally, we notice that the integrand vanishes when \( \tau' < 0 \) because \( \check{\chi}(\tau' \ldots) \propto \theta(\tau') \), so we can restrict the integration over \( \tau' \) to the interval \((0, \infty)\). The integrand also vanishes when \( \sigma < 0 \) because \( \check{\chi}_0(\sigma \ldots) \propto \theta(\sigma) \), so we can also restrict the integration over \( \sigma \) to the interval \((0, \infty)\). We then recognize two Laplace transforms, and we obtain
\[
\check{\chi}_{ij}^{++}(\omega, \Omega) - (\check{\chi}_0)_{ij}^{++}(\omega, \Omega) = -\int_0^\infty d\Omega' \frac{\sum_k (\check{\chi}_0)_{ik}^{++} (\omega + \frac{\Omega - \Omega'}{2}, \Omega')}{2\pi} U_k \check{\chi}_{kj}^{++} \left(\omega + \frac{\Omega - \Omega'}{2}, \Omega'\right). 
\]
We now treat the equation for the bare susceptibility, Eq. (31). Introducing the Wigner coordinates, we obtain
\[
\check{\chi}_{ij}^{++}(\tau, T) = \left[\frac{\sigma}{2\Omega} - \frac{i}{2}T - \Delta_j (T - \frac{\tau}{2})\right]
\]
\[
\check{\chi}_{ij}^{++}(\tau, T) + \check{\chi}_{ij}^{++} (\tau; \Omega') \Delta_j (\Omega - \Omega').
\]
We partially integrate on the variable \( \tau \), and we note that the boundary terms vanish, respectively, because \( \Im(\omega) > 0 \) and \( (\check{\chi}_0)_{ij}^{++} (\tau; \Omega') \propto \theta(\tau) \). We then obtain Eq. (8) of the main text.

**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,
\[
f_{ij} = \frac{1}{N} \sum_q e^{i(\mathbf{R}_i - \mathbf{R}_j) \cdot \mathbf{q}} f_{\mathbf{q}} \Leftrightarrow f_{\mathbf{q}} = \frac{1}{N} \sum_{i,j} e^{-i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} f_{ij},
\]
\[
g_{ij} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{R}_i} g_{\mathbf{q}} \Leftrightarrow g_{\mathbf{q}} = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{q} \cdot \mathbf{R}_i} g_i.
\]
By applying \( \frac{1}{N} \sum_{i,j} e^{-i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} \) to both Eqs. (34) and (36), we obtain...
\[ \tilde{\chi}_q^{+-}(\omega, \Omega) - (\tilde{\chi}_0)_q^{+-}(\omega, \Omega) \equiv \text{THF SOH} \quad \Rightarrow \quad -U \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} (\tilde{\chi}_0)_q^{+-} (\omega + \frac{\Omega - \Omega'}{2}, \Omega') \tilde{\chi}_q^{+-} (\omega - \frac{\Omega'}{2}, \Omega - \Omega') , \]  

(E2)

\[
\left( \omega - \frac{\Omega}{2} \right) (\tilde{\chi}_0)_q^{+-}(\omega, \Omega) - \int_{-\infty}^{\infty} \frac{d\Omega'}{2\pi} (\tilde{\chi}_0)_q^{+-} (\omega + \frac{\Omega - \Omega'}{2}, \Omega') \tilde{\Delta}(\Omega - \Omega') \equiv \text{THF SOH} \quad \Rightarrow \quad 2\pi\delta(\Omega)\bar{m} + \tilde{\Lambda}_q(\omega, \Omega),
\]  

(E3)

where we have introduced the spatial averages

\[
\bar{g} \equiv \frac{1}{N} \sum_i g_i = \frac{g_{\alpha=0}}{\sqrt{N}},
\]

(E4)

and we have noticed that the average magnetic moment

\[
\frac{1}{N} \sum_i m_i(T) = \bar{m}
\]

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

\[
(\tilde{\chi}_0)_q^{+-}(\omega, \Omega) \equiv \frac{2\pi\delta(\Omega)\bar{m} + \tilde{\Lambda}_q(\omega, \Omega)}{\omega - \frac{\Omega}{2} - \tilde{\Delta}}.
\]

(E6)

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

\[
\tilde{\chi}_q^{+-}(\omega; T) \equiv \frac{(\tilde{\chi}_0)_q^{+-}(\omega; T)}{1 + U(\tilde{\chi}_0)_q^{+-}(\omega; T)} = \frac{\bar{m} + \tilde{\Lambda}_q(\omega; T)}{\omega - \left[-U\tilde{\Lambda}_q(\omega; T)\right]}.
\]

(E7)

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

\[
\omega_q(T) \equiv -U\tilde{\Lambda}_q(0; T).
\]

(E8)

**Appendix F: The equilibrium case**

In equilibrium we have the exact identity

\[ \tilde{A}(\omega, \Omega) = 2\pi\delta(\Omega)\Lambda(\omega) \]

(F1)

for the Fourier-Laplace transforms of the many-body functions of \( \tau \) 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. (30) and (33), and then by solving those equations directly, following exactly the same procedure which is discussed in the main text.

In particular, from Eq. (E2) we obtain the equilibrium exchange parameters as

\[
J_{ij} = i\Sigma_{ij}\Sigma_{ji} \lim_{\tau \to 0^+} \int_{-\infty}^{\infty} \frac{dt}{\beta} e^{-\beta t} \left[ (G^<)^{ij\uparrow} (G^>)^{\uparrow\downarrow} - (G^>)^{ij\downarrow} (G^<)^{\downarrow\uparrow} \right].
\]

(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

\[
J_{ij} = \Sigma_{ij}\Sigma_{ji} \lim_{\beta \to 0} \int_{-\beta}^{\beta} d\tau G^{ij\uparrow}(\tau) G^{\downarrow\uparrow}\uparrow(\tau),
\]

(F3)

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

\[
G(\tau) = \frac{1}{\beta} \sum_{\omega_n} e^{-i\omega_n\tau} G(i\omega_n),
\]

as well as using Eq. (E2) in the equilibrium case, we obtain Eqs. (E1) of the main text.

**Appendix G: A useful sum rule for non-equilibrium Green 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

\[
\sum_j \Lambda_{ij}(t, t') = 0.
\]

(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 Eqs. (30), in particular
\[
- i \partial_t \left( G^\geg \right)_{ij t'} = \sum_k \left( G^\geg \right)_{ik t} T^\gamma_{\gamma k} \left( G^\geg \right)_{ij t'} + \Sigma_j(t'),
\]
\[
i \partial_t \left( G^\geg \right)_{it t'} = \sum_k \left( T \cdot G^\geg \right)_{ik t'} + \Sigma_j(t') \left( G^\geg \right)_{it t'}.
\]
(G2)

We multiply the first equation by \( (G^\geg)_{it t'} \) and sum over \( j \); analogously, we multiply the second equation by \( (G^\geg)_{ij t'} \) and sum over \( i \). We obtain
\[
\sum_j \left( G^\geg \right)_{ij t'} \left( -i \partial_t \right) \left( G^\geg \right)_{it t'} = \sum_k \left[ \left( G^\geg \right)_{ik t} T^\gamma_{\gamma k} \left( G^\geg \right)_{it t'} \right]_i + \sum_k \left[ \left( G^\geg \right)_{ik t} \Sigma_j(t') \left( G^\geg \right)_{it t'} \right]_i.
\]
(G3)

We subtract Eq. (G3) from Eq. (G4), divide by 2, and we obtain
\[
\frac{1}{2} i \partial_t \left[ \left( G^\geg \right)_{it t'} \left( G^\geg \right)_{it t'}^\dagger \right]_i = \sum_k \left[ \left( G^\geg \right)_{ik t} \Sigma_j(t') \left( G^\geg \right)_{it t'} \right]_i.
\]
(G5)

The sum rule Eq. (G5) can be used to immediately check that Eqs. (44) and (45) indeed satisfy
\[
\sum_j \left[ J_{ij}(t, t') + X_{ij}(t, t') \right] = 0,
\]
which is in agreement with the Goldstone theorem.


