Time-dependent exchange creates the time-frustrated state of matter

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Magnetic systems governed by exchange interactions between magnetic moments harbor frustration that leads to ground state degeneracy and results in the new topological state often referred to as a frustrated state of matter (FSM). The frustration in the commonly discussed magnetic systems has a spatial origin. Here we demonstrate that an array of nanomagnets coupled by the real retarded exchange interactions develops a new state of matter, time frustrated matter (TFM). In a spin system with the time-dependent retarded exchange interaction, a single spin-flip influences other spins not instantly but after some delay. This implies that the sign of the exchange interaction changes, leading to either ferro- or antiferromagnetic interaction, depends on time. As a result, the system’s temporal evolution is essentially non-Markovian. The emerging competition between different magnetic orders leads to a new kind of time-core frustration. To establish this paradigmatic shift, we focus on the exemplary system, a granular multiferroic, where the exchange transferring medium has a pronounced frequency dispersion and hence develops the TFM.

The model
To reveal how the exchange retardation results in the TFM we focus first on an elemental building block of a granular-multiferroic, two adjacent magnetic granules interacting via a ferroelectric medium as schematically shown in Fig. 1. Figure 1a displaces two metallic granules carrying the opposite magnetic moments disposed over the ferroelectric substrate and Fig. 1b presents the same magnetic moments immersed into a ferroelectric medium. Since the dielectric constant of a ferroelectric environment typically has significant frequency dispersion, the retardation effects are inevitable. Indeed, in a simplest approximation taking into account the dielectric screening of the Coulomb interaction, one finds, following 10,11,24, that the dielectric constant appears in the effective exchange between two magnetic moments as

$$ J = \sum_{\mathbf{r}_1, \mathbf{r}_2} d \mathbf{r}_1 d \mathbf{r}_2 \sum_{k_1, k_2} \Psi^\dagger_k(\mathbf{r}_2) \Psi^\dagger_k(\mathbf{r}_1) \frac{\mathcal{e}^2}{\varepsilon |\mathbf{r}_1 - \mathbf{r}_2|^3} \Psi_k(\mathbf{r}_1) \Psi_k(\mathbf{r}_2), $$

where the sum is taken over the electron wave functions of each granule, and $\Psi(\mathbf{r};t)$ stand for the undisturbed electron wave functions. More precise calculation requires including the effect of the environment on the wave functions 26 and also accounting for the spatial dispersion effects. Yet, even in this first approximation, the frequency dispersion of the exchange integral $J(\omega)$ arises due to dispersion of $\varepsilon(\omega)$ the behavior of which is straightforwardly related to the dielectric permittivity tensor of the ferroelectric environment 10,11,12,14,24,25. Accordingly, we arrive at the model of a magnetic system with the effectively delayed exchange. In a temporal representation, this implies the delay in the interaction of magnetic moments: $\int J_{12}(t - t') \mathbf{m}_a(t') \mathbf{m}_b(t') dt'$, where $J_{12}(t - t')$ is the Fourier transform of $J(\omega)$. It is essential that $J_{12}(t - t')$ is purely retarded, that is $J_{12}(t - t') = 0$, if $t < t'$, so the causality is fulfilled.

In typical ferroelectrics (e.g., such as barium titanate (BTO) and lead zirconate titanate (PZT)), $\varepsilon(\omega)$ is large at low frequencies, $\varepsilon(\omega = 0) = \varepsilon_0 \gtrsim 1000$, and is of the order of unity for large frequencies, $\varepsilon(\omega = \infty) = \varepsilon_{\infty} \approx 1$. The frequency threshold is set by the phonon frequency which usually does not exceed 1 THz. Consequently, for small frequencies, we may treat $J(\omega)$ as vanishing, while at large frequencies, $J(\omega)$ tends to finite values. Accordingly, we put

$$ J_{12}(\omega = 0) = \int_0^\infty J_{12}(t) dt = 0, $$

implying that the function $J(t)$ is alternating in sign with time. Hence we arrive at the “time frustration” of the exchange interaction.
Let us consider now two adjacent magnetic moments $\mathbf{m}_1$, $\mathbf{m}_2$. The delay in the interaction implies a nonequilibrium regime at finite times, while due to the nonzero damping the magnetic moments assume stationary values, $\mathbf{m}_{\infty}^{\text{ext}}$, at $t \to \infty$. The final magnetic state is to be derived from the energy considerations using the effective exchange Hamiltonian $H = J_{\text{eff}}(\omega=0) \mathbf{m}_{\infty}^{\text{ext}} \cdot \mathbf{m}_{\infty}^{\text{ext}}$. The mutual orientation of magnetic moments is to be found by investigating the magnetization time evolution from the starting point to $t \to \infty$.

The magnetic granules are supposed to be semiclassical, hence granule’s magnetization should obey the non-local in time Landau-Lifshitz-Gilbert (LLG) equation. We consider an array of localized magnetic moments satisfying $\mathbf{m}(t)|=1$ condition. The equation of motion for $i$-th moment is

$$\dot{\mathbf{m}}_i(t) = -\gamma \mathbf{m}_i(t) \times \mathbf{h}_{\text{eff}}^{\text{ext}}(t) - \lambda \mathbf{m}_i(t) \times [\mathbf{m}_i(t) \times \mathbf{h}_{\text{eff}}^{\text{ext}}(t)],$$

where $\mathbf{h}_{\text{eff}}^{\text{ext}}(t)$ is an effective Weiss field at the $i$-th site defined as

$$\mathbf{h}_{\text{eff}}^{\text{ext}}(t) = \sum_{j=1}^N \int_{-\infty}^t J_{ij}(t-\tau) \mathbf{m}_j(\tau) d\tau + \mathbf{h}_{\text{int}}^{\text{ext}}(t),$$

where $\mathbf{h}_{\text{int}}^{\text{ext}}(t)$ is the weak external magnetic field, and the sum runs over the nearest neighbors to the site $i$ (we account for only the nearest-neighbor exchange interactions). Exchange integrals $J_{ij}(t)$ are time-dependent and preserve the causality. An instant interaction $J_{ij}(t'=t-\tau=0)J_{ij}$ corresponds to the standard LLG equation implying that the exchange energy assumes the usual form, $-1/2 \sum_{ij} J_{ij} \mathbf{m}_i \cdot \mathbf{m}_j$.

As usual, in the dimensionless equation (3) and (4) the magnetic moment and all the fields are normalized by the magnetic moment’s length ($\gamma M_s$) and field ($\gamma M_s H$), respectively. The time is measured in units of $M_s$ and $\gamma M_s H_0$, being the gyromagnetic ratio. This time scale has the order of $10^{12}$ s$^{-1}$ (corresponds to $1$ THz) with $\gamma_0 = 1.39 \times 10^8$ rad/s/A/m and $M_s \sim 10^6$ A/m (for FePt).

Stationary solutions to the LLG equation

Let us consider a stationary solution to Eq. (3) in which we hereafter set $\gamma = 1$ for simplicity. We assume that $\mathbf{m}(t)=\mathbf{m}$ is a set of stationary solutions to Eq. (3) in the absence of the external field, $\mathbf{h}_{\text{eff}}^{\text{ext}}(t) = 0$. For better visibility we further simplify the notations and write:

$$J_0 = \int_{-\infty}^t J(t-\tau) d\tau$$

$$\mathbf{h}_{\text{eff}}^{\text{ext}} = \sum_{\text{NN}} J_{ij} \mathbf{m}_i^{\text{ext}} 
$$

$$\mathbf{b}_i^{\text{ext}} = [\mathbf{m}_i \times \mathbf{h}_{\text{eff}}^{\text{ext}}],$$

where $\sum_{\text{NN}}$ stands for the sum over the nearest neighbors. In this transparent case, the LLG equation assumes the form

$$\dot{\mathbf{m}}_i(t) = -\mathbf{b}_i^{\text{ext}} - J[\mathbf{m}_i \times \mathbf{h}_{\text{eff}}^{\text{ext}}],$$

which requires that $\mathbf{b}_i^{\text{ext}} = 0$. Stationary solutions would realize for either

- Frustrated exchange with $J_0 = \int_{-\infty}^t J(t-\tau) d\tau = 0$ implying $\mathbf{h}_{\text{eff}}^{\text{ext}} = 0$, hence $\mathbf{b}_i^{\text{ext}} = 0$. Therefore, any magnetic configuration formally assumes a stationary solution. We address further the important question whether these solutions are stable with respect to small perturbations like noise or an external field, and show that only particular stationary configurations are stable.

- A non-frustrated exchange with $J_0 \neq 0$ implying $\mathbf{h}_{\text{eff}}^{\text{ext}} \neq 0$, which is satisfied for common FM, AFM structures and for collinear configurations for which $\sum_{\text{NN}} \mathbf{m}_i^{\text{ext}} = 0$, e.g., stripe structures in square lattice.

We see that for stationary solutions, frustrated and non-frustrated cases differ qualitatively. Hereafter we focus on a dynamically frustrated case.

Time-dependent magnetic moments

General equations

Let us derive the adjustments to the stationary solution discussed above arising due to time dependence of the magnetic moments. We consider the time-dependent part of the $i$-th magnetic moment $\mathbf{m}_i(t)$ being small and write magnetization as

$$\mathbf{m}_i(t) = \mathbf{m}_i^0 + \mathbf{m}_i(t).$$

In the corresponding linear approximation, the Landau-Lifshitz-Gilbert equation for $\mathbf{m}_i(t)$ in the frustrated case assumes the form, see Methods

$$\dot{\mathbf{m}}_i(t) = -[\mathbf{m}_i^0 \times \mathbf{h}_i(t)] - \lambda [\mathbf{m}_i^0 \times [\mathbf{m}_i^0 \times \mathbf{h}_i(t)]] .$$

To find the analytical solution to Eq. (10) we take its Fourier transform and obtain, see Methods,

$$i \omega \mathbf{m}_i^0(\omega) = -[\mathbf{m}_i^0 \times \mathbf{h}_i^0(\omega)] - \lambda [\mathbf{m}_i^0 \times [\mathbf{m}_i^0 \times \mathbf{h}_i^0(\omega)]] + \lambda \mathbf{h}_i^0(\omega).$$

Two-site cluster with the frustrated exchange

The above general reasoning holds for any arbitrary regular magnetic structure and, in particular, is not restricted to systems subject to nearest neighbor interactions constraint. To illustrate how the formation of the time-frustrated state occurs, we consider the simplest particular system, two interacting magnetic moments $\mathbf{m}_1$ and $\mathbf{m}_2$. To further simplify the problem, we analyze collinear stationary configurations, FM with $\mathbf{m}_1 = \mathbf{m}_2 \parallel z$ and AFM with $\mathbf{m}_1 = -\mathbf{m}_2 \parallel z$ (hereafter we use $\mathbf{m}_1^0 = \mathbf{m}_2^0 = 1$, $i = 1, 2$ and $\mathbf{m}_1^0 \times \mathbf{m}_2^0 = 0$).

Taking the simplest form of the exchange satisfying all the above-defined conditions

$$J(t) = G(\delta(t) - \omega t e^{-i\omega t})$$

where $\delta(t)$ is the Dirac delta function using its Fourier transform,

$$J(\omega) = G \frac{\omega}{\omega + i\omega_0},$$

see Fig. 2, which preserves the causality as the pole is in the lower half of the $\omega$-plane, and displays a reasonable asymptotic behavior: $J(\omega=0) = 0$, $J(\omega \to \infty) \to$ const. The characteristic value $\omega_0 \sim 1$ with the accepted time scale corresponds to $THz$ phonon frequency region of the common granular multiferroics, e.g. Pr$_{1-x}$SrMnO$_3$/LuMnO$_3$, NaNO$_2$/porous glass, SrTiO$_3$/rutile SrTiO$_3$/Teflon, La$_{1.25}$Ba$_{0.75}$MnO$_3$/LuMnO$_3$. To simplify further notations, we set it in that the magnetic moments and energy are properly normalized and are measured in dimensionless units. Thus, $G$, $\gamma$ and $\lambda$ also become dimensionless. Now we find the stability conditions ensuring the stationary solutions. For the FM case, $m_1^0 = m_2^0 = 1$, we obtain the stability condition as $G \lambda < \omega_0$. For the
AFM configuration, \( m_0 = -m_2 = +1 \), and the resulting stability condition is \( G \sqrt{1 + \lambda^2} < \omega_0 \).

Having established the ranges of stability within the linear approximation, let us turn to detailed investigating the time evolution of our system. The zero-frequency limit was discussed above. At high frequencies the ferroelectric degrees of freedom are frozen and the exchange is provided by the conventional electron clouds overlapping.

The time evolution appears radically different in the isotropic case and in the presence of the even weak uniaxial anisotropy. In the isotropic case, the asymptotic, \( t \to \infty \) state of magnetic moments, either in the FM or AFM case, is defined by the exchange potential parameters, mostly by its \( \delta \)-part. In the anisotropic case either the \( \delta \)-part or the exponential part of the time-depending exchange (12) dominates the system’s behavior. The results of the numerical calculations of the time evolution are displayed in Fig. 3. The panel Fig. 3a shows that the evolution of the isotropic system with the time-dependent exchange results in the FM state at \( t \to \infty \), as it is clearly seen from the evolution of magnetic moments projections. Remarkably, although perturbing the system by the half-sinusoidal pulse of the external magnetic field switches the FM state into the AFM one, see the panel Fig. 3b, this AFM state lives only for some finite time, and then the system returns back to the FM state.

This whole evolution picture is noise-resistant, it does not transform under the delta-correlated noise with the amplitude \( \langle \mathbf{h}_{\text{noise}} \rangle \) small relative to effective field \( \langle h_{\text{eff}(t)} \rangle \). The external perturbation in the form of the sequence of the alternating pulses successively converts FM to AFM and vice versa, see Supplementary Information (SI).

In the presence of the anisotropy, both states, the AFM and the FM, become stable. There exist two ways of switching the final destination of the system between these states. The first way is changing the parameters of the dynamically frustrated potential. The example of such a switch by changing the characteristic frequency \( \omega_0 \) is shown in Fig. 4a presenting the temporal evolution of quantity \( \mathbf{m}_1 \cdot \mathbf{m}_1 \), characterizing the state of the system. The second possibility of the switching is the perturbation in a form of the single half-sinusoidal external magnetic field pulse. In this case, in contrast to the isotropic one, the switched state is stable. Depending on the direction of the pulse, the final stable state is either the FM or the AFM state. Figure 4b shows an example of such a switch.

The revealed behaviours are of a general character and maintain for a general case of the system with the arbitrary number of the magnetic moments. The behaviour of the exemplary four-site cluster is presented in the Supplementary Information (SI).

**Discussion and conclusion**

We have studied spin system with retarded spin-spin interaction \( J_{ij} \). This implies the non-Markovian type of the time-dependent magnetic interaction and leads to nontrivial dynamics of the interacting magnetic moments. The time-frustrated case where \( J_{ij}(\omega = 0) = \int_0^\infty J_{ij}(t)dt = 0 \) is the most interesting regime because in this case the sign of \( J_{ij}(\omega = 0) \) does not naively predict the arising at \( t \to \infty \) magnetic configuration.

It is important to stress that the retardation causes the non-Hermiticity of the effective Hamiltonian of the interacting magnetic moments, therefore, the considered system is an effectively dissipative. Non-Hermitian quantum mechanics describing open dissipative systems is currently enjoying an intense explosive development \(^{35–38} \), and further aspects and implications of the non-Hermitian behavior of the system in hand will be a subject of the forthcoming publication.

The retarded spin-spin interaction is realized in the systems with the superexchange where magnetic moments interact indirectly through a medium with the pronounced frequency dispersion, granular multiferroics offering an appealing example. In multiferroics, magnetic granules interact through a ferroelectric medium. Its polarization comprises several contributions with the different characteristic times, \( P = P_{\text{el}} + P_{\text{mag}} + P_{\text{dipole}} + \ldots \). Here the first “elastic” contribution is the polarization of the outer electron shells, the second one is related to the ion shifts, and the third contribution is related to dipole moments of molecules; the second and the third terms typically are responsible for the ferroelectricity. It is important that all the contributions except the first one are relatively slow, with their relaxation times being larger or of order of the inverse phonon frequencies for which 1 THz is a natural scale \(^{39–41} \). At the same time, \( P_{\text{el}} \) relaxation time is electronic, having the optical frequencies, being thus by several orders of magnitude shorter (\( G \) in Eq. (12) is obviously define
Figure 4 | Control of the final stable state by the damping parameter or by the external pulse: the magnetic moments scalar product \( m_1 \cdot m_2 \) is depicted.

a. In the presence of weak anisotropy, the initial state transforms into different final stable states depending on damping parameter \( \omega_d \). If \( \omega_d \leq \omega^* \simeq 7.5 \) (relatively fast retardation), the final stable state is the FM, for \( \omega_d \geq \omega^* \geq 10 \) (relatively fast retardation), the final stable state is the AFM. The initial state is the slightly disturbed AFM (one magnetic moment infinitesimally tilted away from purely AFM arrangement). The asymptotic, \( t \gg 1 \), picture does not depend upon the initial state for \( \omega \neq (\omega^*, \omega^*) \). The Landau-Lifshitz-Gilbert equation parameters are \( \gamma = 1, \lambda = 1 \), anisotropy parameter \( \rho = 5 \), the retarded exchange amplitude is \( G = 10 \).

b. In the presence of weak anisotropy, the perturbation in the form of the magnetic pulse (pulse being the half sinuosoidal, \( A \sin(\alpha t) \)) allows to control the final stable state. For \( A \leq A^* - 15 \) (negative pulse) the final stable state is the FM, for \( A \geq A^* = 5 \) (positive pulse) the final stable state is the AFM. Here the initial state is the same as in the previous figure (the slightly disturbed AFM). Again, the asymptotic, \( t \gg 1 \), picture does not depend upon the initial state for \( A \neq (A^*, A^*) \). The Landau-Lifshitz-Gilbert equation parameters are \( \gamma = 1, \lambda = 1 \), anisotropy parameter \( \rho = 5 \), the retarded exchange amplitude is \( G = 10 \).

Methods

Derivation of a general equation  Taking magnetization as

\[
m_i(t) = m_i^0 + m_i^*(t),
\]

where \( m_i^0(t) \) is assumed small, one finds that the corresponding linear approximation of the Landau-Lifshitz-Gilbert equation for \( m_i^*(t) \) assumes the form

\[
m_i^*(t) = -b_i^*(t) - \lambda [m_i^0(t) \times b_i^0(t)] - \lambda [m_i^0 \times b_i^0(t)]
\]

(15)

\[
b_i^*(t) = [m_i^0(t) \times b_i^0(t)] + [m_i^0 \times b_i^0(t)]
\]

(16)

\[
h_i^*(t) = \sum_{NN} \int_{-\infty}^{\tau} J(t - \tau) m_N^0(t) d\tau + h^{\text{ext}}(t).
\]

(17)

In the frustrated case with \( \int_{-\infty}^{\tau} J(t - \tau) d\tau = 0 \), both \( b_i^0(t) = 0 \) and \( h_i^0(t) = 0 \) for large enough values of \( t \), and Eq. (15) reduces to Eq. (10) of the main text. Taking the Fourier transform of (10), one gets

\[
\int_0^{\infty} \omega m_i^0(\omega) = -[m_i^0 \times h_i^0(\omega)] - \lambda [m_i^0 \times [m_i^0 \times h_i^0(\omega)]],
\]

(18)

where

\[
h_i^0(\omega) = \sum_{NN} J(\omega) m_N^0(\omega) + h^{\text{ext}}(\omega).
\]

(19)

The double cross product in (18) is \( (m^0 \cdot m^0) = 1 \)

\[
[m_i^0 \times [m_i^0 \times h_i^0(\omega)] = m_i^0 \times (m_i^0 \cdot h_i^0(\omega) - h_i^0(\omega)),
\]

(20)

and the evolution equation for \( i \)-th for the Fourier transform of magnetic moment becomes Eq. (11) of the main text.

Two-site cluster with the frustrated exchange  For the two-site cluster the system of equations (11) for \( m_i^0(\omega), m_i^1(\omega) \) reads (we set \( h^{\alpha}(\omega) \parallel x \)):

\[
\int_0^{\infty} \omega m_i^0(\omega) = -[m_i^0 \times h^{\alpha}(\omega)] - J(\omega)[m_i^0 \times m_i^0(\omega)] + J h^{\alpha} + J(\omega) m_i^0(\omega)
\]

(21)

\[
\int_0^{\infty} \omega m_i^1(\omega) = -[m_i^1 \times h^{\alpha}(\omega)] - J(\omega)[m_i^1 \times m_i^0(\omega)] + J h^{\alpha} + J(\omega) m_i^1(\omega)
\]

(22)
\[
\Delta(\omega) = \begin{pmatrix}
  i\omega & 0 & -J(\omega) & -J(\omega)m_1^0 \\
 0 & i\omega & J(\omega)m_1^0 & -J(\omega) \\
 -J(\omega) & -J(\omega)m_1^0 & 0 & i\omega \\
 -J(\omega)m_1^0 & -J(\omega) & 0 & i\omega
\end{pmatrix}
\]

were \( m_0^2 = |m|^2 \).

Taking the exchange in the form Eq. (13) that not only preserves the causality, but also Re \( J(\omega) \) is even function of \( \omega \) like Re \( 1/\epsilon(\omega) \). Then the characteristic equation \( \Delta(\omega) = 0 \) has four roots (apart from four stationary roots \( \omega_0 = 0 \)):

\[
\omega_{1,2} = -i\omega_0 \pm iG \sqrt{\lambda + im_1^0} (\lambda + im_1^0)
\]

\[
\omega_{3,4} = -i\omega_0 \pm iG \sqrt{\lambda - im_1^0} (\lambda - im_1^0)
\]

For the FM case \( m_1^0 = m_2^0 = +1 \), we find

\[
\omega_{1,2} = -i\omega_0 \pm iG (\lambda + i)
\]

\[
\omega_{3,4} = -i\omega_0 \pm iG (\lambda - i)
\]

so the stability condition is \( G \lambda < \omega_0 \).

For the AFM configuration \( m_1^0 = -m_2^0 = +1 \), we have two double-degenerate roots

\[
\omega_{1,2} = -i\omega_0 \pm iG \sqrt{\lambda + i} \lambda^2
\]

and the resulting stability condition is \( G \lambda + \lambda^2 < \omega_0 \).

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Author contribution
N.M.C and A.V.M. conceived the work and performed calculations, V.E.V. carried out numerical simulations, V.M.V. took part in calculations and outlining the project, N.M.C, A.V.M., and V.M.V. lead the interpretation of the results and wrote the manuscript, all authors discussed the manuscript.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information is available in the online version of the paper.

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