Observation of magnetic structural universality using transverse NMR relaxation

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Transverse NMR relaxation from spins diffusing through a random magnetic medium is sensitive to its structure on a mesoscopic scale. In particular, this results in the time-dependent relaxation rate. We show analytically and numerically that this rate approaches the long-time limit in a power-law fashion, with the exponent reflecting the disorder class of mesoscopic magnetic structure. The spectral line shape acquires a corresponding non-analytic power law singularity at zero frequency. We experimentally detect a change in the dynamical exponent as a result of the transition into a maximally random jammed state characterized by hyperuniform correlations.

In this work, we consider the loss of Larmor precession coherence in media with a static magnetic structure on a much larger, mesoscopic scale9–11 relevant for NMR experiments in porous rocks7,8 and in biological tissues.12–21 Here, the individual precession phases \( \varphi(t) = \int_0^t \Omega(r) \, dr \) decohere due to the path-dependent Larmor frequency offset \( \Omega(r) \) on their Brownian trajectories \( r \) induced by heterogeneous medium’s magnetic susceptibility. The macroscopic \( t \to \infty \) rate \( R^\infty \sim \langle \Omega^2(r) \rangle t_c \) decreases for a faster diffusion constant \( D \), as the time \( t_c \sim l_0^2/D \) to travel across the disorder correlation length \( l_c \) shortens, exemplifying the diffusion narrowing.14,16,17,26 Importantly, the typical mesoscopic correlation times \( t_c \) can be of the order of the NMR measurement time scale, which makes it possible to explore the transient signal evolution \( s(t) \) before the long-time monoexponential limit is reached, via studying the corresponding time-dependent relaxation rate \( R(t) = -d \ln s(t)/dt \).

The mesoscopic contribution to the transverse relaxation, in principle, depends on myriads of parameters characterizing the spatial organization of susceptibility-induced \( \Omega(r) \). It is generally non-universal, and sensitive to the shape of magnetic inclusions (e.g. cells).18,27 Our main result is the universal feature of the mesoscopic relaxation which manifests itself in the power-law tail in the approach of \( R(t) \) to \( R^\infty \),

\[
dR_2(t) \sim t^{-\nu} \quad \text{for} \quad t \gg t_c \tag{1}
\]

such that \( R^\infty - R(t) \sim t^{-\nu+1} \) for \( \nu > 1 \), and \( R(t) \sim \ln t \) for \( \nu = 1 \); an upper temporal limit on this behavior is discussed below. We relate the dynamical relaxation exponent

\[
\nu = \frac{p + d}{2} \tag{2}
\]

to the large-scale statistics of the structural organization: The relevant signature of the \( d \)-dimensional medium, represented by magnetic susceptibility \( \chi(r) \) varying on the mesoscopic scale, is embodied by the (magnetic) structural exponent \( p \), which we define via the \( k \to 0 \) scaling of the power spectrum

\[
\Gamma_2(k) = \int d^d r \, e^{-ikr} \langle \chi(r_0) \chi(r) \rangle_{r_0} \sim k^{p}, \quad k \to 0 . \tag{3}
\]
**FIG. 1. Illustration of the universality relations (1) – (3) for the disorder classes in $d = 3$ dimensions.** Columns show the results for five fully permeable synthetic media with qualitatively distinct structural fluctuations (see Methods). First row shows a quarter of the Monte Carlo simulation box in each dimension; second row shows the angular-averaged power spectra $\Gamma_2(\mathbf{k})$, Eq. (4); third and fourth rows show the self-energy part $\Sigma(\omega)$ (see text) and $dR_2(t)/dt$, numerically obtained from the simulations. The packing of long ellipsoids has large fluctuations at small $k$. This is a finite-size effect that is alleviated by the ensemble averaging (thicker yellow line) over 10 disorder realizations (thin gray lines). $\Sigma(\omega)$ and $dR_2(t)/dt$ obtained from MC simulations (colors) are compared with the leading-order calculation (black lines) by integrating the numerically found power spectra according to Eqs. (9) and (10), respectively, while neglecting $R_\infty^2$ on the right-hand side. For $dR_2(t)/dt$ (fourth row) this agrees well with the asymptotic limit (1) with the exponent (2) (dashed lines). Parameters $\rho$, $\delta\Omega$, $\alpha$, and $t_c$ are defined in Methods.

The exponent $p$ takes a few discrete values, characterizing distinct universality classes of structural disorder.

The key relation (2) is illustrated in Fig. 1 for five statistically isotropic disorder classes in $d = 3$ dimensions using Monte Carlo (MC) simulations, where we identify the exponent $p$ in the angular-averaged power spectra

$$\Gamma_2(k) = \frac{\langle \Omega - k\Omega_k \rangle_k}{V} = c_d(4\pi\gamma B_0)^2 \cdot \Gamma_\chi^2(k) \sim k^p$$  (4)

of the susceptibility-induced Larmor frequency offset

$$\Omega_k = 4\pi\gamma B_0 \chi k Y_k$$  (Fig. 2), where $Y_k = 1/3 - k_z^2/k^2$ is the elementary dipole field, $\gamma B_0$ is the average Larmor frequency, and $V$ is the sample volume. The scaling of the power spectra of the structure and of the induced frequency with $k = |\mathbf{k}|$ is similar due to the $Y(r) \sim 1/\rho^d$ dependence of the dipole field, with $c_3 = \langle |Y_k|^2 \rangle_k = 4/45$, meaning that the transverse relaxation effectively samples the structure of the medium (i.e. $\chi(r)$) directly, even though it senses the induced $\Omega(r)$.

The universality (2) in the diffusion-narrowing regime can be used as a probe of the global structural organization of magnetically heterogeneous media, and for the mesoscopic model selection. In Fig. 1, four kinds of identical sphere arrangements, and one with randomly placed long (prolate) ellipsoids, represent five distinct universality classes. In particular, Order, represented by a cubic lattice of spheres, shows no long-range fluctuations and $\Gamma_2(k) \equiv 0$ for small $k$, which
can be associated with an exponent \( p = \infty \), yielding an exponentially fast decay of \( dR_2/dt \) (faster than any inverse power law). For \textit{hyperuniform disorder,} \( p > 0 \), these fluctuations are not completely absent but are suppressed. Examples are a \textit{shuffled lattice,} \( \Omega \) where the lattice objects are randomly displaced from their original positions, showing a quadratic behavior \( \Gamma_2[k \to 0] \sim k^2 \), with \( p = 2 \) yielding \( \nu = 5/2 \); and a \textit{maximally random jammed (MRJ) packing,} \( \Omega \) where \( \Gamma_2[k \to 0] \sim k \), with a nontrivial exponent \( p = 1 \), which manifests itself in \( dR_2/dt \sim 1/t^2 \). \textit{Short-range disorder} (the most widespread disorder class, characterized by a finite correlation length, e.g. as in the Poissonian objects' placement) is represented here by the random packing of non-overlapping spheres. Its power spectrum is characterized by a finite plateau at small \( k \), \( \Gamma_2(k)_{k \to 0} = \text{const} \), such that \( p = 0 \) and \( \nu = d/2 \). Finally, \textit{strong disorder} is characterized by the diverging structural fluctuations resulting in the exponent \( p < 0 \). An example are randomly placed “rods”, such as vesicles or capillaries in the brain (here represented by highly prolate ellipsoids whose long axis exceeds the range of diffusion lengths), yielding \( \Gamma_2[k \to 0] \sim k^{-1} \), \( p = -1 \), such that \( \nu = 1 \) and the relaxation rate \( R_2(t) \sim \ln t \) diverges.

In what follows, we will provide a qualitative coarse-graining argument for the universal relation (2), Fig. 2, followed by the self-consistent approximation (10) for the signal, Fig. 3, and then demonstrate experimentally how the change in the disorder universality class due to the jamming transition, \( p = 0 \to p = 1 \), can be detected via the dynamics (1) of the measured bulk transverse relaxation, Fig. 4.

An intuition behind the relation of \( dR_2/dt \) to the spatial fluctuations stems from realizing that the \( \Omega(t) \) in the Larmor frequency, \( \Omega(r) \rightarrow \Omega_t(r) \), effectively “seen” by the spins, Fig. 2. If we were to begin the evolution of magnetization at such \( t \) foregoing shorter times, then the coarse-grained \( \Omega_t(r) \) would have the effective correlation length \( L(t) > \infty \), and the variance \( \langle \Omega^2_t(r) \rangle \sim \langle \Omega^2_t(r) \rangle_{L(t)} \sim \langle \Omega^2 \rangle \sim \langle \Omega^2_e \rangle \), for the short-range disorder due to Poissonian statistics. We now apply the conventional diffusion-narrowing argument \( dR_2/dt \sim \langle \Omega^2 \rangle \) to our effective \( \Omega_t(r) \), by identifying \( t_c \to t \) and \( \langle \Omega^2 \rangle \to \langle \Omega^2_e \rangle \):

\[
\frac{dR_2(t)}{dt} \approx \langle \Omega^2_e(r) \rangle \sim t^{-\nu},
\]

with \( \nu = d/2 \) for the Poissonian case of \( p = 0 \).

One can view Eq. (5) as a real-space renormalization group equation on the effective macroscopic parameter \( R_2 \) over the increasing diffusion length scale \( L(t) \). We immediately see that the rate \( R_2(t) \) \textit{always increases} with \( t \), such as both length scale contributes a strictly positive frequency variance to the relaxation; however, its growth slows down due to the self-averaging, as the instantaneous distribution \( \Omega_t(r) \) becomes narrower with \( t \), Fig. 2. Moreover, the self-averaging will be faster when the fluctuations \( \langle \Omega^2_t(r) \rangle \) decrease faster than the inverse “diffusion volume” \( L^{-d}(t) \) (which happens for hyperuniform media, \( p > 0 \)), and slower for strong disorder with diverging fluctuations, \( p < 0 \), in agreement with Fig. 1.

This “RG flow” must eventually stop for \( t > t_e \) such that \( t_e R_2(t_e) \sim 1 \), when \( R_2(t) \) becomes so large that the mesoscopic signal is suppressed exponentially before spins can sample fluctuations of \( \Omega(r) \) at the scales exceeding \( L(t_e) \). After \( t > t_e \), the power law (1) gets cut-off, Fig. 3, and by then the mesoscopic signal \( s(t) \leq 1 \). Hence, the scaling (1) is detectable for \( t_e < t \leq t_e \), provided that the relaxation is sufficiently weak, i.e. \( R_2(t_e) t_e \ll 1 \), which is equivalent to a small “single-step” phase variance \( \langle \delta^2 \rangle \sim \langle \Omega^2 \rangle t_e^2 < 1 \).

The above intuition is supported by finding the disorder-averaged Green’s function \( G_{t,x-r_0} = \langle g_{t,x-r_0} \rangle_t \) of the mesoscopic Bloch-Torrey equation for the transverse nuclear magnetization \( 14,21,30 \)

\[
[\partial_t - \partial_r D(r) \partial_r + i \Omega(r)] G_{t,x-r_0} = \delta(t) \delta(r-r_0),
\]

where \( D(r) \) is the local diffusion coefficient. The mesoscopic contribution \( s(t) \) to the NMR signal is helpful to think of in terms of “spin packets”, the groups of spins emanating from the same point \( r_0 \). The magnetization of a spin packet is \( \int dr \, g_{t,x-r_0} \); the \( r_0 \)- and \( t \)-dependence of this quantity embodies the coarse-graining discussed above. Acquisition from a macroscopic sample entails ensemble-averaging of the spin packet magnetization, \( s(t) = \frac{1}{\Omega} \int dr \, g_{t,x-r_0} \equiv \int dr \, G_{t,x} \). In other words, the signal \( s(t) \equiv G_{t,q} = 0 \) is the Fourier transform of \( G_{t,q} \) for the wavenumber \( q = 0 \). Here and in what follows, we factor out the molecular relaxation; the experimentally observable signal \( S(t) = e^{-\mu q^{-2}} s(t) \).

We represent the disorder-averaged propagator of Eq. (6) \( G_{t,q} \)

\[
G_{\omega,q} = \frac{1}{-i\omega + D_{\infty} q^2 - \Sigma_{\omega,q}}
\]

in terms of the self-energy part \( 21,31,32 \) \( \Sigma_{\omega,q} \) that collects all one-particle irreducible Feynman diagrams accounting for \( \Omega(r) \) and the deviation \( D(r) - D_{\infty} \) from the macroscopic diffusion constant \( D_{\infty} \). The expansion of \( \Sigma_{\omega,q} \) in the powers of \( q \) reflects the measurable mesoscopic effects in the bulk relaxation \( 21 \) (due to even-order correlators \( \langle \Omega(r_1) \Omega(r_2) \rangle \)), frequency shift (similar correlators of odd orders), diffusion \( 21 \) (due to \( \langle D(r_1)D(r_2) \rangle \)), and apparent diffusion \( 12 \) (due to the cross-terms \( \langle \Omega(r_1)D(r_2) \rangle \)). In particular, the mesoscopic spectral lineshape \( s(\omega) = \left[ -i\omega - \Sigma(\omega) \right]^{-1}, \) where \( \Sigma(\omega) \equiv \Sigma_{\omega,q} = 0 \).

We consider the self-energy part in the self-consistent Born approximation, \( 21 \) equivalent to summing up the “rainbow” di-
The dynamical exponent (2) changes from $\nu = 3/2$ to $\nu = 2$ in aqueous suspension of polystyrene microbeads, according to the change of the disorder universality class between the dilute suspension with short-range disorder, $p = 0$ (panel a), and an MRJ sediment with the predicted exponent $p = 1$ (panel b). Shown are representative data sets for water doped with paramagnetic agent (HOOCl₃ · 6H₂O) with concentrations 1.5, 2.0 and 2.5 mmol/L to create a magnetic susceptibility contrast with the microbeads (see Methods for experimental details).
substitution $R_\infty^2 \rightarrow R_{mol}^2 + R_\infty^2$.

The present discussion shows that the universality, our main result expressed by Eqs. (1) and (2), is mapped onto the behavior of the structural power spectrum for small $k$, which is independent of individual properties of the magnetized objects. This is illustrated in supplementary Fig. S1 that shows results of MC simulations for the same media as in Fig. 1, but with the magnetized objects made impermeable for diffusing spins.

As an application of the developed formalism, in Fig. 4 we experimentally demonstrate the change of the disorder universality class $p = 0 \rightarrow 1$ after reaching the maximally random jammed state for mono-dispersed spheres, where the nontrivial exponent $p = 1$ was predicted numerically.\textsuperscript{25} NMR relaxation in two micro bead packings (suspension and densely-packed sediment) exhibit distinct exponents for the time derivative of the relaxation rate, which makes it possible to distinguish the two packings using a macroscopic NMR measurement in contrast to the microscopic character of up-to-date observations.\textsuperscript{33-35} This remarkable sensitivity of the macroscopic measurement to the nontrivial mesoscopic structure is enabled by the time-dependent coarse-graining window, Fig. 2, that effectively samples the mesoscopic medium’s power spectrum (3).

To conclude, we have shown analytically and numerically, that the mesoscopic component of the transverse relaxation rate displays a universal scaling behavior that is sensitive to the statistics of large-scale organization of tissue magnetic susceptibility. This allowed us to provide the first macroscopic experimental observation of the MRJ transition in spherical micro bead packings using NMR relaxation. Our results provide a framework for noninvasive investigation of the structure of complex materials and in biomedical magnetic resonance imaging, where both native and added susceptibility contrast is ubiquitous.

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METHODS

Synthetic media. In Fig. 1, we consider five representative media: four types of differently arranged identical spheres with radius \( \rho \), and one with randomly placed long (prolate) ellipsoids (here \( \rho \) is the radius of the short axes, the long semi-axis is \( 40 \rho \)), which were generated as follows. A simple cubic lattice of spheres with a volume fraction of \( \zeta = 34\% \) represents perfect order. To create the shuffled lattice, the spheres of the regular lattice were randomly displaced from their original positions by nine discrete values within the distance \( \pm 0.2335 \rho \) in each direction rejecting steps that caused overlap with a neighbor. The MRJ packing was generated using an event-driven molecular dynamics simulation using the code downloadable from the authors’ website. The resulted medium had the volume fraction of spheres \( \zeta = 65\% \). For the short-range disorder, the spheres were randomly added rejecting steps leading to the overlap with already existing spheres (\( \zeta = 34\% \)). The same algorithm was used for the random arrangement of long ellipsoids whereas the non-overlap condition was released resulting the ellipsoids with the summed volume of 15% of the simulation volume forming a structure with the overall volume fraction \( \zeta = 14\% \). To alleviate finite-size effects in the diverging power spectrum of the long ellipsoids at small \( k \) (Fig. 1), we further averaged the MC runs over ten different disorder realizations. All media were sampled on a 1024\(^3\) cubic grid for numerical computation of \( \Omega(r) \) and successive MC simulations.

The Larmor frequency shift, \( \Omega(r) \), was calculated as the convolution with the elementary dipole field. To characterize the scale (the dephasing strength) of field variations, we use the dephasing introduced by a single object: \( \Omega l^2 = \frac{4}{3} \pi \chi \gamma B_0 \) for spheres and \( \Omega l^2 = 2 \pi \chi \gamma B_0 \) for the long ellipsoids, where \( \chi \) is the susceptibility difference with the background, and \( \gamma B_0 \) the Larmor frequency in the external field. The disorder correlation length \( l_c \) was defined starting with \( k_c = \pi / \rho \), which is close to the pronounced peak of \( \Gamma_2(k) \) for the considered sphere packings, corresponding \( l_c = 1 / k_c \) and the correlation time \( t_c = 1 / (D k_c^2) \). The dimensionless parameter \( \alpha = \delta t_c \Omega t_c \) instantiates the typical spin phase, \( \langle \varphi^2 \rangle \), acquired when moving over the disorder correlation length; the diffusion-narrowing takes place when \( \alpha < 1 \).

Numerical calculations to obtain \( \Sigma(\omega) \) and \( d R_2(t)/dt \) to the leading order were performed by integrating the Larmor frequency power spectrum \( \Gamma_2(k) = \Omega \chi \Omega k^2 / V \), cf. Eq. (4), for the five synthetic media according to Eqs. (9) and (10), respectively, while neglecting \( R_2^\infty \) on the right-hand side.

Monte Carlo simulations of the mesoscopic relaxation for freely diffusing spins were performed with \( N_s = 10^8 \) spins randomly hopping on the sample grid of the above described media imitating the dephasing strength \( \alpha = 0.2 \) (spheres) and \( \alpha = 0.05 \) (long ellipsoids). The increment of the random walker’s spin phase was calculated using the mean of \( \Omega(x) \) before and after each hop. The mesoscopic NMR signal at each time moment \( t \) was calculated as the mean of all accumulated phase factors \( s(t) = \exp(-i \Omega t) \). The second derivative of \( \ln s(t) \) was calculated using third-order polynomial fitting, based on Savitzky-Golay filtering, \( \gamma(t) \) with a linearly increasing filter width of 0.6\( t \).

Microbead samples. Polystyrene microbeads (DYN005 TSL; Microbeads AS, Skedsmokorset, Norway) were suspended in an aqueous solution of sodium chloride doped with Holmium(III) chloride hexahydrate (HoCl\(_3\cdot6\)H\(_2\)O) in various concentrations to adjust the solution density and magnetic susceptibility, respectively. Suspensions with 30% volume fraction of microbeads were prepared using a particle-density matched sodium chloride solution (\( c_{NaCl} = 1.28 \) mol/L for \( T = 309 K \)) to avoid sedimentation. MRJ samples were prepared by particle sedimentation in \( c_{NaCl} = 0.33 \) mol/L solution and careful removal of particle-free fluid from the top. All samples were prepared in standard 5-mm NMR tubes.

NMR measurements were performed on a DPX 200 MHz spectrometer (Bruker, Ettlingen, Germany) using a standard zg30 sequence (flip angle = 30\(^\circ\), acquisition time = 4\( s \), 16 averages, relaxation delay = 3\( s \), no spinning) at \( T = 309 K \). The shim fields were adjusted on a pure D\(_2\)O sample and then kept for all samples within a measurement series. To obtain \( d R_2 / dt \), the measured FID signals were processed with the same fitting algorithm as applied for MC simulations using a filter width of 0.7\( t \).

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SUPPLEMENTAL INFORMATION

Monte Carlo simulations for hindered diffusion were performed within the same media and with the same parameters as in Fig. 1. Impermeable spheres and ellipsoids were simulated by discarding Monte Carlo steps that lead inside the objects in which case the random walkers did not move during the given time step. The results shown in Fig. S1 support the universality of the dynamical exponent, while the coefficients in front of $t^{-\nu}$ and $|\omega|^{\nu-1}$ are non-universal. Note that the renormalization of the diffusion constant with its long-time asymptote (Fig. S2) is not sufficient to reproduce the non-universal coefficients (data not shown), which are strongly modified for the media with higher volume fraction $\zeta$.

Order: 
- Regular lattice
- Hyperuniform disorder: 
  - Shuffled lattice MRJ packing
- Short-range disorder: 
  - Random packing
- Strong disorder: 
  - Long ellipsoids

FIG. S1. Results for hindered diffusion in the five three-dimensional synthetic media from Fig. 1 representing different disorder classes. The second row shows the derivative of the time-dependent relaxation rate, $dR_2/dt$. MC results for hindered diffusion (colored curves) in general differ from the leading-order calculation (black curves, the same as in Fig. 1), which assumes permeable objects, but follow the same characteristic exponents (dashed lines). The same observation holds for the equivalent depiction in frequency domain (first row). While the self-energy curves also differ quantitatively from the leading-order calculation, the qualitative behavior embodied by the power-law exponent at $\omega = 0$ is similar. For strong disorder the difference between hindered and free diffusion is actually negligible, which follows from the low volume fraction of $\zeta = 14\%$ for the long ellipsoids.

FIG. S2. Instantaneous diffusion coefficient \[^{31}\] $D(t) = \frac{1}{t} \frac{d}{dt} \langle x^2(t) \rangle$ for the hindered diffusion within the investigated media, normalized to the free diffusion coefficient $D_0$. The temporal derivative of the mean square displacement $\langle x^2(t) \rangle$ of the MC random walkers was computed using the Savitzky-Golay filter \[^{37}\] based on the second-order polynomial, with a width of about $t_c$. 

\[^{31}\] $D(t) = \frac{1}{t} \frac{d}{dt} \langle x^2(t) \rangle$ 

\[^{37}\] Savitzky-Golay filter