Correlated noise in Brownian motion allows for super resolution

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Diffusion broadening of spectral lines is the main limitation to frequency resolution in non-polarized liquid state nano-NMR. This problem arises from the limited amount of information that can be extracted from the signal before losing coherence. For liquid state NMR as with most generic sensing experiments, the signal is thought to decay exponentially, severely limiting resolution. However, there is theoretical evidence that predicts a power law decay of the signal’s correlations due to diffusion noise in the non-polarized nano-NMR scenario. In this work we show that in the NV based nano-NMR setup such diffusion noise results in high spectral resolution.

Spectral analysis is of utmost importance in a wide variety of fields from material science, to biology and medicine. Among the most widespread techniques to obtain structural information in the form of a spectrum is Nuclear Magnetic Resonance (NMR), which is nonetheless hindered by low sensitivity. One promising approach to improve the capacities of NMR is to reduce the sample to the nano-scale. This technique, however, is still limited by the finite resolution of spectral features. A possible solution is to use polarized samples as in conventional NMR, but this approach requires either large samples or a substantial increase in experimental complexity. In this work we challenge the claim that working with nano-sized samples limits resolution, and provide analytical and numerical evidence supporting the viability of the non-polarized setup as an alternative route to nano-NMR.

NV centers have been used extensively in the past as quantum sensors for the implementation of the nano-NMR scheme1–3. In particular, the use of quantum heterodyne (Qdyne) measurement techniques (know as well as synchronized measurements), together with a suitable data-analysis algorithm has demonstrated that resolving two close frequencies requires no more than accumulating a sufficient number of measurements11,12. These techniques, however, are computationally heavy since they need to solve a global maximization problem in a large dimensional space that grows linearly with measurement time.

Measuring a spectrum that contains two (or more) similar frequencies that are closer than the characteristic wavelength used to image them. Here, the width of the line-shape plays the role of the wavelength. This resolution problem for two close frequencies can best be understood by looking at the change in the spectrum (Fig. 1). The intuition behind the limited resolution can be understood in terms of the Rayleigh criterion from optics, where two images are resolvable only up to the finite frequency difference has a very small effect on the spectrum (Fig. 1a), whereas for a sharp-peak function the change is more pronounced (Fig. 1b). This suggests that for a sharp-peaked spectrum, spectral-resolution could be improved.

Spectral resolution in NV based liquid-state nano-NMR is limited mainly by the diffusion of nuclei in the sample13–16. When measuring a noisy signal oscillating at frequency δ, the amount of information that can be extracted from the signal is limited by the noise coherence time. For diffusion noise in liquid state nano-NMR, C(t) is generally considered to be an exponentially decaying function leading to Lorentzian spectral line-shapes, impeding high spectral resolution. In this manuscript, we challenge this framework by building on the work of Cohen et al.17, which reported that a significant deviation from the Lorentzian line-shape paradigm occurs when measuring a magnetic field of a non-polarized nano-sized liquid sample with a shallow NV. We show that diffusion does not limit resolution and that the analysis is computationally amenable and can be done with simple algorithms such as Fourier spectrum analysis.

The effect in17 can be understood as follows. The effective sensitivity of an NV located at depth d beneath a sample extends to a semi-sphere of radius d above the surface that contains N ∝ d3 non-polarized nuclei. The rms of the magnetic field sensed by the NV is thus $B_{\text{rms}} \propto \sqrt{N}/d$, where the sample is due to the dipole-dipole interaction.

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The peak of the power spectrum is thus $S(\delta = 0) \propto B_{\text{rms}}^2 T_\phi \propto 1/d$, with $T_\phi \propto d^1$ the characteristic time that it takes the nuclei to diffuse out of the semi-sphere (i.e., the inverse of the signal bandwidth). When, for example, applying dynamical decoupling (DD) sequence with detuning $\delta$ from the nuclei Larmor frequency, a new length scale is introduced, i.e., $\ell = \sqrt{D/d}$; this length scale can be understood as a cut-off for the interaction between NV and distant nuclei; fields coming from these nuclei are slow changing and thus attributed to low frequency. Using the same reasoning as before, the power spectrum around the peak is $S(\delta) \propto 1/d - \alpha/\ell = 1/d - \alpha/\sqrt{D/D}$, where $\alpha$ is a positive number. Therefore the power spectrum in NV based nano-NMR of liquid samples is a sharp-peaked function. A similar effect has also been observed in diffusing atom systems\textsuperscript{19}. Conversely, in the time domain, where the resolution problem is manifested by our ability to see a beat between close frequencies, the beating between close frequencies can be observed, allowing higher resolution.

Results

Fl analysis. We now analyze the effect of long-lived correlations on frequency estimation and resolution. The resolution problem is characterized by an estimation error for the frequencies that diverges when the frequency difference is much smaller than the characteristic noise frequency, $T_\phi^{-1}$, as demonstrated by a vanishing amount of information extracted from the signal\textsuperscript{19}. For a noise that is a stationary Gaussian process, with a covariance function of the form $\text{Cov}(t) \propto C(t) \sum_{j=1}^{N} \cos(\delta_j t)$, the resolution problem occurs for $|\delta_i - \delta_j|/T_\phi < 1$. We restrict the derivation to the estimation of a small single frequency, which is a good model for the resolution problem since the average frequency is generally easier to estimate. We analyze the three possible measurement scenarios, i.e. correlation spectroscopy\textsuperscript{4,20}, Qdyne/synchronized measurement protocol\textsuperscript{1,8,9}, and power spectrum probing\textsuperscript{21}. For the full details of this derivation and schematics of each protocol we refer the reader to the Supplementary Information.

Correlation spectroscopy. The fluorescence response of the NV can be modeled by a Poisson distribution with a rate parameter that depends on the NV state ($m = 0, 1$). In the correlation spectroscopy scenario, the average number of photons detected is given by\textsuperscript{4,20}

$$p = \eta + \frac{c}{2} \langle \sin(\phi) \sin(\phi_{i+1}) \rangle,$$

where $\eta, c$ are the average detection rate and contrast, and $\phi, (\phi_{i+1})$ is the phase accumulated by the NV during the first (second) interrogation time ($\tau$). These phases are calculated by integrating over the magnetic field. We model the magnetic field as stationary Gaussian processes oscillating at frequency $\delta$, with a characteristic correlation time $T_\phi$ and a mean field strength of $B_{\text{rms}}$. Averaging over realizations of the magnetic field yields...
\[ p = \eta + \frac{c}{2} e^{-\phi_{\text{rms}}^2 \sinh(\phi_{\text{rms}}^2 \cos(\delta t) C(t/T_\phi))}, \]

where \( C(\cdot) \) is the correlation function (envelope) of the phases. The rms of the accumulated phase and its correlation function can be approximated by \( \phi_{\text{rms}} \approx \sqrt{B_\text{rms} \tau} \) and \( C(t/T_\phi) \approx \text{corr}(B_\phi, B_\phi^{+}) \) for a short interrogation time \( \tau \ll T_\phi \), where \( \gamma \) is the gyromagnetic ratio of the NV. For a weak signal (i.e., \( \phi_{\text{rms}}^2 \ll 1 \)) Eq. (2) can be approximated by

\[ p \approx \eta + \frac{c}{2} \phi_{\text{rms}}^2 \cos(\delta t) C(t/T_\phi). \]

The FI of \( \delta \) from a single measurement (a single choice of \( t \)) is given by

\[ j_{\delta,\beta} \approx \frac{c^2}{4\eta + c^2 \phi_{\text{rms}}^4 t^2 \sin^2(\delta t) C^2(t/T_\phi)}, \]

in the weak signal regime. Eq. (4) shows that the sine term is the reason for the limited resolution. The maximum amount of information from a single measurement (for small \( \delta \)) depends on the correlation function. An exponential decay imposes an optimal measurement time that scales as \( t_{\text{opt}} \propto T_\phi \); i.e., the longest time possible before the correlation is exponentially small. Thus the information scales as \( j_{\delta,\beta} \propto \delta^2 T_\phi^4 \), and vanishes for \( \delta \to 0 \). By contrast, for a slow polynomial decay (i.e., \( C(z) \propto z^{-n} \) for large \( z \) and \( 0.5 < n < 1.5 \), with \( z \) henceforth being \( z = t/T_\phi \) the optimal measurement time scales as \( t_{\text{opt}} \propto \delta^{-1} \); i.e., the correlations are significant enough such that the sine term poses no problems. Thus the information scales as \( j_{\delta,\beta} \propto \delta^{2n-2} T_\phi^{2n} \), with a weaker dependence on frequency. With respect to the measurement time, the information rate is \( j_{\delta,\beta}/T_{\text{tot}} \propto \delta^{2n-1} T_\phi^2 \); consequently, for correlations with \( n < 1.5 \) there is a slight improvement in resolution, and for \( n = 1.5 \), as in \( 17 \) (Eq. (13)), there is no improvement over exponential correlations. For this reason it may be desirable to consider different measurement protocols.

**Qdyme/synchronized measurements.** Further improvement can be made considering a synchronized measurement protocol\(^{14,18}\). In this scenario, the fluorescence response of the NV has a detection rate of

\[ q_t = \eta + \frac{c}{2} \sin(\phi_t). \]

Thus the average probability for measuring the pair \((y_t, y_{t+\Delta})\) of number of photons is

\[ \langle q_t q_{t+\Delta} \rangle = \eta^2 + \frac{c^2}{4} e^{-\phi_{\text{rms}}^2 \sinh(\phi_{\text{rms}}^2 \cos(\delta t) C(t/T_\phi))}. \]

Estimating the signal using the covariance between the number of photons detected at different times, the information about \( \delta \) (from two measurements with a time difference \( t \)) is given by

\[ j_{\delta,\beta} = \frac{c^4}{(4\eta + c^2)^2} \phi_{\text{rms}}^4 t^2 \sin^2(\delta t) C^2(t/T_\phi) + \tilde{C} (\phi_{\text{rms}}^4). \]

This FI is obtained for a weak signal by (least-squares) fitting of the correlation function. With each additional measurement (performed at time \( t + \tilde{t} \)) we effectively obtain \( t/\tilde{t} \) additional “measurements” by correlating with all previous measurements. For small rms we can safely assume that the noise in the “measurements” is uncorrelated. For data taken at times \( t_m = mt \), the total FI is given by

\[ j_{\delta,\beta} \approx \frac{c^4}{(4\eta + c^2)^2} \phi_{\text{rms}}^4 T_{\text{tot}}^3 \tilde{t}^2 \mathcal{F}, \]

where we assumed \( \delta \tilde{t} \) and \( \tilde{t}/T_\phi \) to be small. The behavior of the integral in Eq. (9) for small \( \delta \) depends on the correlation function. For an exponential decay, \( \mathcal{F} \propto \delta^2 T_\phi T_{\text{tot}} \) in the regime of \( \delta T_\phi \ll 1 \ll \delta T_{\text{tot}} \), whereas for polynomial decay

\[ \mathcal{F} \propto \begin{cases} \frac{1}{\delta T_{\text{tot}}} & , n < 1.5 \\ \frac{\delta T_{\text{tot}}}{\delta T_\phi}^{1-2n} & , 1.5 < n < 2.5 \\ \frac{\delta^2 T_\phi}{\delta T_{\text{tot}}} & , n > 2.5 \end{cases} \]

in other words, there is a minute correction for small \( \delta \) when the polynomial decay is slower than 2.5. For decay rates slower than 1.5 the information is independent of \( \delta \), and the information rate increases with time (\( \propto \delta T_{\text{tot}}^{2-2n} \)) (see Fig. 2). In the limiting case of \( n = 1.5 \), \( \mathcal{F} \propto \log(\delta T_{\text{tot}})T_{\text{tot}}/T_\phi \) and the correction grows logarithmically when \( t_{\text{tot}} \) is large.
The fluorescence response of the NV is given by \( \eta \propto T^{-\frac{1}{2}} \exp(\delta T) \). Different polynomial scalings are presented in different colors. The case of exponential correlation is presented as a dashed line. The information per unit of time saturates for \( \delta T_{\text{tot}} \gtrsim 1 \), for correlations with slow polynomial decay ( \( n < 1.5 \)). For faster decays ( \( 1.5 < n < 2.5 \)) the characteristic time changes continuously towards \( \delta T_{\phi} \gtrsim 1 \) (see top horizontal axis). For the limiting case of \( n = 1.5 \) the information rate changes its behavior for \( \delta T_{\phi} \gtrsim 1 \), but only saturates for \( \delta T_{\phi} \gtrsim 1 \), which is attributed to the small logarithmic correction \( \log(\delta T_{\phi}) \).

Figure 2. Scaling of the FI rate about \( \delta \) as a function of \( \delta \) (Eqs. 8, 9); for this plot we set \( T_{\text{tot}} = 10^4 T_{\phi} \). Different polynomial scalings are presented in different colors. The restriction on the interrogation time poses an extra limit on the field strength being obtained at post-processing rather than on the NV. Nevertheless, this factor is compensated for by the fact that more statistics are gathered in Qdyne; i.e., roughly a factor of \( 2 \) in Eq. (7) suffers from an extra constant factor on the information, as most of the information comes from long-time correlations. Note that for correlation spectroscopy the shortest correlation time is limited by the DD sequence (which must be shorter than the coherence time of the signal), whereas for Qdyne is limited also by the readout/initialization time ( \( \bar{T} - \tau \approx 2.1 \mu s \), see for example); for exponential correlations this limits the Qdyne technique to samples with coherence time longer than the readout time. But for a slow polynomial decay this induces only a small constant factor on the information, as most of the information comes from long-time correlations.

**Power spectrum measurements.** In the power spectrum measurement scenario, the interrogation time, \( \tau \), must be increased beyond the correlation time of the noise, which in most cases is impossible since the coherence time of the NV ( \( T_2^N \)) is too short. The fluorescence response of the NV is given by

\[
\langle y(\omega) \rangle = \eta - \frac{c}{2} \exp \left( -\frac{1}{2} \omega^2 B_{\text{rms}}^2 T_{\phi} \tau \mathcal{S}_\tau(\omega) \right),
\]

where \( \mathcal{S}_\tau(\omega) \) is the unit-less (normalized by \( T_{\phi} \tau \)) power spectrum (convoluted with the filter function defined by the DD protocol). The restriction on the interrogation time poses an extra limit on the field strength being probed \( \gamma^2 B_{\text{rms}}^2 T_{\phi} \tau \lesssim 1 \) (i.e., a large rms value will saturate the signal exponentially fast). In addition, the inverse interrogation time sets the resolution for this measurement protocol; i.e., in order to resolve a frequency difference \( \delta \) we must set \( \tau > \delta^{-1} \).

When these requirements are met, the shape of the spectrum will dictate the information scaling; correlations that decay with a power law \( \sim n \) correspond to a spectrum that scales with a power law \( n - 1 \) around the peak. For a smooth spectrum ( \( n > 2 \)) the information scales as the derivative of the spectrum (squared), \( j_{\delta,\delta} \propto T_2^N(\delta T_{\phi})^{-n} \) at \( \omega = 0 \). For a sharp spectrum (derivative is discontinuous at the peaks, \( 1 < n < 2 \)) the optimal measurement is performed at \( \omega - \delta \approx \tau^{-1} \) (as close as possible to the peak), before the shape of the filter function starts to dominate) and the information scales as \( j_{\delta,\delta} \propto (\tau/T_{\phi})^{-4} \). For the former case, resolution limit is set by \( T_{\phi}^{-1} \), albeit with a reduced “penalty”, and by \( \tau^{-1} \) for the latter.

**Nano-NMR signal analysis.** We now demonstrate resolution and verify the theoretical analysis by simulating and analyzing both single and multi-frequency signals. The procedure is as follows; First, we generate accumulated phases \( \phi_i \) (Eq. 5) by either using molecular dynamic (MD) simulations for a more accurate description of an experimental situation (see “Methods”), or we sample a multivariate Gaussian distribution which simplifies the theoretical analysis. These phases are then used to simulate measurement vectors in a Qdyne protocol. Parameter estimation is then performed by least squares fitting the signal correlation function to the theoretical model.
showing that the MSE is commensurate with the multi-frequency analysis. In yellow, we generate a single-frequency signal. A signal with one frequency is estimated for exponentially decaying correlations the same signal produces a histogram in which no single frequency can be pinpointed. Furthermore, we include the histogram corresponding to a signal with a non-polarized sample. This is compared to a signal generated with exponential correlations, which does not allow for resolution of the frequencies.

Resolution. Figure 3 illustrates resolution beyond the Rayleigh Limit. We generate the signals of the magnetic fluid, whose correlations behave as \( C(z \gg 1) \propto z^{-3/2} \) to zero. For more information about the numerical procedure see “Methods”. In Fig. 3a we depict the estimation of a single frequency for 600 measurement vectors, each composed of 2^{12} measurements. In fitting the correlation function Eq. (12), a fitting is only accepted if \( r^2 > 0.95 \). In both cases, the frequencies were not resolved for the same parameters but rather with exponential correlations. Estimating close frequencies is a global optimization problem whose complexity increases exponentially in parallel with the size of the search space in which the frequencies live. In fitting the correlation function Eq. (12), a fitting is only accepted if \( r^2 > 0.95 \). In both cases, the frequencies were not resolved for the same parameters but rather with exponential correlations.

Scaling analysis. We now proceed to the numerical analysis of the theoretical model presented in the previous section, in the case of one and two frequency signals. We show that for the anticipated signal in the nano-NMR scenario, the characteristic time for resolution is the total measurement time. In this case, we simulate synchronized measurements by generating signals with an analytical correlation function \( C(t/T_\phi) \) where the noise comes from sampling a multivariate Gaussian distribution mimicking the scenario of small \( \phi_{\text{rms}} \). We focus here on the case of \( n = 1.5 \) in Eq. (10) corresponding to the correlation function in Eq. (13) \( C(z \gg 1) \propto z^{-3/2} \) which corresponds to Eq. (6) for weak signals. \( C(z) \) is considered either as polynomial correlations \( \propto z^{-3/2} \) corresponding to Eq. (13) from \( 17 \) (henceforth \( C(z \gg 1) \propto z^{-3/2} \)), or an exponential correlation \( \exp(-z) \) for comparison purposes. The \( \phi_i \) in Eq. (12) is a dummy parameter added for numerical reasons, and which tends to zero. For more information about the numerical procedure see “Methods”. We focus here on the case of \( n = 1.5 \) in Eq. (10) corresponding to the correlation function in Eq. (13) \( C(z \gg 1) \propto z^{-3/2} \)
from\textsuperscript{17}. A point in Fig. 4 corresponds to the MSE of a histogram composed of \( N = 2^8 \) measurement vectors each, with \( 2^{14} \) measurements.

Figure 4a displays the behavior of the MSE of the estimator as a function of \( \phi_{\text{rms}} \). For fixed \( \delta T_\phi = 0.5[2\pi] \), below the Rayleigh Limit such that the signal with an exponential correlation could not be resolved, we simulate signals with varying \( \phi_{\text{rms}} \). According to Eq. (10), for a weak signal the MSE (i.e. \( 1/\delta T_\phi \)) diverges as \( \phi_{\text{rms}}^{-2} \) as we observe in Fig. 4b, thus setting the optimal region for nano-NMR around \( \phi_{\text{rms}} = 1 \). For strong signals, the information rate is exponentially suppressed. The scaling in the case of one frequency is not fundamentally different from that of two frequencies.

In Fig. 4b we set \( \phi_{\text{rms}} = 0.6 \) and study the behavior with \( \delta T_\phi \). Here we can observe the difference caused by extended correlations in the information rate and thus the resolution capacity. While for exponential correlations the MSE diverges quadratically with \( \delta \), and rapidly saturates the histogram, for polynomial decays the divergence is slower. In the case of \( C(z \gg 1) \propto z^{-3/2} \) the divergence is logarithmic in \( \delta \) (see Eq. (10)), as we see in Fig. 4b, i.e., it can easily be compensated for by increasing the measurement time. Note in addition that since \( \phi_{\text{rms}} (B_{\text{rms}}) \sim 1/d^{3/2} \) and \( T_\phi \sim d^{21/2} \), for \( C(z \gg 1) \propto z^{-3/2} \) according to Eq. (10) the MSE is independent of the depth of the NV, as occurs with polarized nano-NMR.

**Discussion**

We showed that spectral resolution in non-polarized liquid state nano-NMR is not necessarily limited by the broadening of spectral lines due to diffusion. While for exponential correlations the resolution is limited by the inverse characteristic coherence time of the signal, we demonstrate that for (slow) polynomial correlations, as predicted by\textsuperscript{9}, resolution is not limited.

We analyzed the scenario in which the sensor is a shallow NV center. In this case, the correlations decay as \( C(z \gg 1) \propto z^{-3/2} \) at long times, producing sharp spectral features. Moreover, increasing the number of frequencies analyzed does not hinder resolution.

Comparing the three measurement protocols we observe that for exponential correlations, the resolution problem always appears for \( \delta T_\phi < 1 \), but the sensitivity of Qdyne is different by a factor of about \( (c^2/\eta)(T_\phi/\tau)^2 \). For a low viscosity, water-like fluid this could still prove beneficial, despite the low contrast in state of the art systems \( (c^2/\eta \approx 0.016) \). For power-law decay (with power of 3/2), while the sensitivity remains the same as the exponential case, the resolution capabilities of the power spectrum measurement and Qdyne protocols are extended. For power spectrum measurements, the protocol is limited by the time of a single measurement \( (\tau) \).
which is only restricted by the coherence time $T_2$ of the NV sensor. The Qdyne protocol is virtually not limited by diffusion as the only limitation is the total measurement time.

The power law analysis presented here is so far based on theoretical grounds. Nonetheless, experimental evidence for a deviation from the exponential correlations paradigm already exist. In fact, Staudacher et al. found in\textsuperscript{20} a correlation function for a non-polarized liquid state nano-NMR experiment which exhibits a long-lived tail. Such behaviour was attributed to a surface effect which creates a thin layer of static, rotating molecules close to the surface of the diamond, finding a reasonably good agreement between the model and the experimental results. It is clear that the assumption of macroscopic Brownian motion with a Lorentzian profile and exponential correlations is too crude an approach to the non-polarized nano-NMR setting. As such, the diffusion induced long-lived correlations described in\textsuperscript{17}, which we have demonstrated lead to enhanced resolution, are but a lower limit on the achievable resolution scaling of the non-polarized nano-NMR setup. Different physical effects such as those described in\textsuperscript{20} demonstrate that even longer-lived correlations can be expected to exist. As our analysis demonstrates, harnessing these power-law correlations leads to an increase of the information gathered (see Fig. 2), resulting in even better scaling for resolution of frequencies in a nano-NMR spectra.

Methods
Noise model for diffusing particles. Each nucleus composing the sample substance interacts with the NV center via dipolar coupling; in the nano-NMR setting, nucleus dynamics manifests through the dephasing rate of the NV center. Calculating this dephasing rate involves solving the drift-diffusion dynamics equation. For an NV situated at a depth $d$ from the diamond surface and assuming that the liquid fills a semi-infinite volume above the diamond surface, the correlation function for the nucleus distribution is\textsuperscript{17}
\[ C(z) = \frac{4}{\sqrt{\pi}} \left( z^2 - \frac{3}{2}z^2 \frac{1}{z^2} + \frac{\sqrt{3}}{4} + 3\sqrt{2} - \frac{3}{2}\sqrt{\pi} z \frac{1}{z} + \sqrt{\frac{\pi}{z}} \text{erfc} \left( z^{-\frac{1}{2}} \right) \exp \left( -\frac{z}{2} \right) + \frac{7}{4} \sqrt{2} z + \frac{3}{2} z^2 \right) \],

with \( z = d^{-2} Dt \) where \( D \) is the diffusion coefficient for the fluid.

To accurately simulate the NV response signal to the magnetic field generated by a distribution of diffusing molecules used to demonstrate resolution in an experimental-like scenario, we perform molecular dynamics simulations.

For the molecular dynamics we consider \( N \approx 46k \) dipolar particles within a simulation box of size \( L_{x,y,z} \approx (50,50,24) \), with a NV located at depths in the range of \((0.3,5)\). The particles within the box are simulated as a Lennard–Jones fluid with normalized parameters \( \varepsilon = \sigma = 1 \) and are initialized into a thermal state at temperature \( T = 1 \). During the simulation, the magnetic field induced by the particles at the NV position is measured along the \( z \) direction for several NV depths.

Analysis of the generated magnetic fields at different NV depths shows that the data have no trend and that the standard deviation remains scale invariant. This means we can compare different depths if appropriately scaled. This is done by calculating the correlations and partial correlations of the different time series. An example of a time series can be found in Fig. 5a.

In Fig. 5b we analyze the temporal correlation in the magnetic field as a function of NV depth. The correlation, which is akin to the autocorrelation after correcting by the mean, tells us how a point in the time series is related to itself after \( \kappa \) time-steps. We observe that it is highly dependent on the depth of the NV, as expected from the relation \( T_\phi \propto d^2 \) (the diffusion coefficient \( D \) is the same for all depths). Since resolution depends on \( \delta T_\phi \) Fig. 5b gives us information about which depth is more convenient, depending on the characteristics of the signal that we want to analyze.

Figure 5c depicts the correlation corrected by depth. Note the deviation from exponential decay at long times, as described by Eq. (13), which is responsible for long-lived correlations. Moreover, this deviation is independent of the depth of the NV, which means that the same description is valid for the magnetic field at any NV depth.

When using MD vectors to simulate the accumulated phases \( \phi_t \), we avoid correlations among different MD vectors by calculating each noise realization by randomly sampling two different instances of magnetic fields in the corresponding NV depth.

**Numerical calculations.** Parameter estimation is done by numerically fitting each measurement vector to the theoretical model

\[ \sum_i (\phi_{rms}^{(i)})^2 \cos(\delta t + \phi)C(t/T_\phi). \]

The fitting is done by a non-linear least squares algorithm with finite-difference estimation of gradient. Each fitting is initialized with random values taken from uniform distributions around the mean signal values for each parameter in Eq. (12). The width of the distributions coincides as well with the allowed search regions in the fitting process. These are, respectively, \( \phi_{rms} \in [\phi_{rms}/2, 3\phi_{rms}/2] \), \( \delta \in [\delta_{avg}/2, 3\delta_{avg}/2] \), \( \psi \in [0, 2\pi] \) and \( T_\phi \in [T_\phi/100, 100 T_\phi] \). Average values are estimated from the signal for the \( \phi_{rms} \) or from the signal FT for \( \delta \).

The \( \phi_t \) in Eq. (14) is non-physical and is included for reasons of numerical stability. In all of the fittings it tends to either 0 or 2\( \pi \).

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Author contributions
A.Re. conceived the idea. A.Ro. and S.O.C. performed the analytical and numerical analysis. R.N. provided the molecular dynamics simulations. A.Ro. and S.O.C. wrote the manuscript, which was revised, commented, and appropriately updated by all of the authors.

Competing interests
The authors declare no competing interests.

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