

Low-temperature spin dynamics and absence of magnetic order in layered α -RuI₃

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(Received 6 June 2024; revised 9 March 2026; accepted 11 June 2026; published 10 July 2026)

The honeycomb-lattice system α -RuI₃ is isostructural to the widely studied α -RuCl₃ compound which was identified as a potential Kitaev system but exhibits, instead of spin liquid behavior, a magnetically ordered zigzag ground state which sets in below 14 K. Here we show experimentally that, in contrast, the spins in α -RuI₃ remain dynamic down to at least 50 mK. We study the spin dynamics using muon-spin relaxation methods and determine the presence of low-frequency fluctuations which are characteristic of a two-dimensional system.

DOI: [10.1103/kxbj-gp8z](https://doi.org/10.1103/kxbj-gp8z)

I. INTRODUCTION

Materials with magnetic ions decorating a layered honeycomb lattice are of great current interest due to the geometric frustration that is present when the magnetic ions are antiferromagnetically coupled [1]. In this context, α -RuCl₃ has been widely studied as a potential quantum spin liquid (QSL) material. The Ru³⁺ ions in this compound form a layered honeycomb structure [2] and this highlighted α -RuCl₃ as a potential realization of the exactly solvable Kitaev model [3]. The octahedral crystal field and spin-orbit coupling acting on the 4d⁵ electrons produce a local effective $J = \frac{1}{2}$ ground state and lead to anisotropic interactions that are potentially described by the Kitaev model [4,5].

However, α -RuCl₃ orders magnetically at around 7 K to 14 K in zero field, the temperature probably depending on stacking faults [6–11]. It only enters a possible spin liquid state under an applied magnetic field at 10 T [12,13]. Meanwhile, α -RuBr₃ was found also to possess a zigzag antiferromagnetic ground state with a higher transition temperature at 34 K [14,15]. The magnetic ordering in both compounds has been attributed to non-Kitaev interactions in the Hamiltonian and both compounds are now thought to realize the more general J - K - Γ model [16], the Hamiltonian for which is given by

$$\hat{H} = \sum_{\langle i,j \rangle} J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha), \quad (1)$$

where J is the Heisenberg exchange, K is the Kitaev exchange, and Γ is the off-diagonal interaction. On each bond, one spin direction is chosen as γ , with α and β denoting the remaining directions. Although both the chloride and bromide salts exhibit magnetic order, the magnetic order in α -RuBr₃ appears more robust. The larger ionic radius of the Br[−] anion both expands the lattice and increases the covalency of the Ru-halide bond due to the upfloating of Br 4p bands compared with Cl 3p bands [14], both effects of which tend to enhance the Kitaev interaction.

The isostructural iodide α -RuI₃ has only been recently synthesized and has shown promising potential to become another candidate QSL [17]. Magnetic susceptibility measurements of polycrystalline samples found α -RuI₃ to be weakly paramagnetic and no magnetic ordering was observed down to 1.8 K. The magnetization data show a linear dependence on the external field at 200 K, but at low temperature (2 K) there is an S-shaped feature which cannot be completely explained by a Brillouin function [17]. Heat capacity (C_p) measurements also found no indication of a phase transition between 0.35 and 10 K, but did identify a large T -linear term (29.3 mJ mol^{−1} K^{−2}, which is larger than other layered insulating metal trihalides such as CrI₃, 1.17 mJ mol^{−1} K^{−2}, and in contrast with α -RuCl₃ in which the γT term vanishes) [17].

Resistivity measurements reveal metallic behavior [17,18], consistent with density functional theory (DFT) calculations that identify metallic states near the Fermi surface [17–19] and in contrast with the insulating nature of α -RuCl₃. However, it has been suggested that the metallic nature of the sample may be due to a grain boundary effect and might not be intrinsic [20], but recent photoemission data [21], resonant inelastic x-ray scattering (RIXS) data [22], and Raman studies [23] support the existence of a metallic state in α -RuI₃. In this Letter, we report muon spin relaxation (μ SR) data obtained on α -RuI₃ and thereby demonstrate that the Ru³⁺ ions in this compound do not form a magnetically ordered ground state down to 50 mK, demonstrating that this compound may realize a spin liquid ground state.

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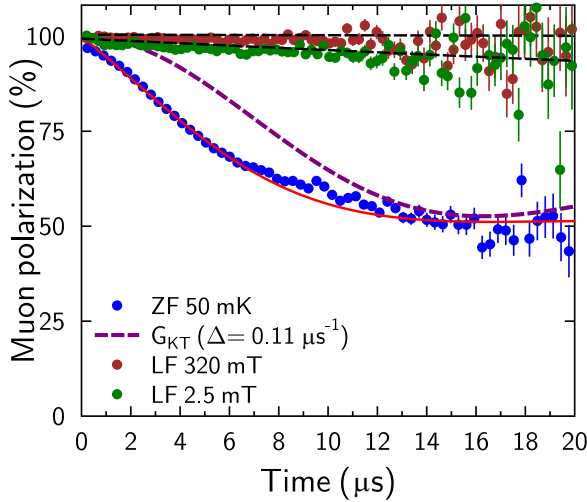


FIG. 1. Blue: ZF- μ SR polarization measured at 50 mK with error bars and a fitted line (red) by Eq. (2). Purple: a Kubo-Toyabe relaxation with $\Delta \approx 0.11 \mu\text{s}^{-1}$ as a comparison to a pure relaxation by nuclear moments. Green and Brown: LF- μ SR polarization with 2.5 mT and 320 mT applied at 100 mK, with black dashed lines indicating the fit to an exponential decay function $e^{-\lambda t}$.

II. MUON SPIN SPECTROSCOPY (μ SR)

A. Zero field μ SR

The polycrystalline sample used in our experiment was prepared using a self-flux method (as described in [17]) and has been characterized using x-ray diffraction. In a μ SR experiment [24] spin-polarized muons are implanted in a sample and interact with the local magnetic field. They subsequently decay, after an average time of $2.2 \mu\text{s}$, and the angular distribution of the detected positrons, each of which is preferentially emitted in the direction of the muon spin at the time of decay, allows us to deduce the polarization of the muon-spin ensemble. We performed our zero-field (ZF) μ SR spectroscopy using the HiFi instrument at the ISIS Neutron and Muon Source with the sample loaded into a packet made of $25 \mu\text{m}$ silver foil, placed on a silver plate, and mounted in a dilution refrigerator [25]. The results, plotted in Fig. 1, show a monotonic relaxation of the muon asymmetry at all temperatures, with no evidence of any precession signal that would indicate magnetic ordering. The asymmetry instead exhibits an initial Gaussian-like decay but becomes exponential at late times. This behavior can be crudely fitted by

$$P(t) = P_r G_{KT}(t, \Delta) e^{-\lambda t} + P_{bg}, \quad (2)$$

where P_r and P_{bg} are the amplitudes relating to the relaxing component (due to muons implanted in the sample) and background component (due to muons implanted in the silver packet and plate). This can be interpreted by assigning the function $G_{KT}(t, \Delta)$, a Kubo-Toyabe relaxation function, to a distribution of static, but randomly oriented, nuclear moments and assuming that the muon is also coupled to dynamical fluctuations in the field at the muon site which arise from fluctuations in the electronic Ru^{3+} moments, giving rise to the weak exponential decay term $e^{-\lambda t}$. To illustrate the effect

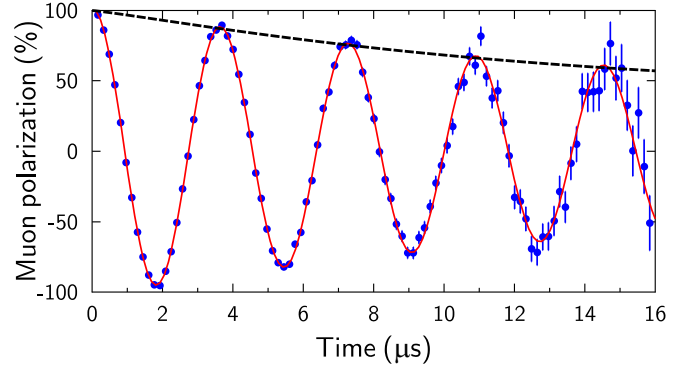


FIG. 2. Weak transverse field muon asymmetry (blue) at 61 mK with a 2.0 mT transverse field and the fitted line (red) using Eq. (3). The dashed line (black) shows the relaxation envelope.

of this exponential, the pure Kubo-Toyabe function with field distribution width $\Delta = 0.11 \mu\text{s}^{-1}$ and no extra decay term is plotted as a dashed line in Fig. 1 (this value of Δ has been chosen to be consistent with the expected nuclear broadening at the calculated muon site, as will be discussed later). We found no possible fit to the data using just $G_{KT}(t, \Delta)$, which indicates that there is an additional relaxation process occurring in addition to the effect of static magnetic fields from nuclear moments. These ZF- μ SR results therefore provide strong evidence that the sample does not undergo magnetic ordering down to 50 mK, in strong contrast to the spin precession signals that are obtained in ZF- μ SR experiments on $\alpha\text{-RuCl}_3$ [11] and $\alpha\text{-RuBr}_3$ [26] below their antiferromagnetic ordering transitions.

Further confirmation of this comes from weak transverse field measurements. Data taken with a $B = 2.0 \text{ mT}$ transverse field at 61 mK exhibit a precession signal at the Larmor frequency with weak damping, as shown in Fig. 2. By fitting the TF- μ SR spectrum to a cosine function multiplied by a relaxation envelope, given by

$$P(t) = (P_r e^{-\lambda t} + P_{bg}) \cos(2\pi \nu t + \phi), \quad (3)$$

and using the same relaxing asymmetry P_r as in the ZF measurement [see Eq. (2)], the precession frequency $\nu = \gamma_\mu B / (2\pi)$ is consistent with the applied field (where the muon gyromagnetic ratio γ_μ is given by $\gamma_\mu = 2\pi \times 135.5 \text{ MHz T}^{-1}$ and λ is found to be $0.074(2) \mu\text{s}^{-1}$). We also fitted the spectrum with a stretched exponential decay $e^{(-\lambda t)^\beta}$ and obtained $\beta = 1.17(4)$, which is consistent with a Lorentzian field distribution superimposed on top of the applied field, instead of a Gaussian one ($\beta = 2$). This reflects persistent spin dynamics and a fluctuating local field at the muon site.

Using DFT (in particular, the DFT + μ method [27–29]), it is possible to identify the muon site. Our calculations reveal a muon site in $\alpha\text{-RuI}_3$ at $[0.544, 0.180, 0.286]$, within a RuI_3 layer, and close to the center of a Ru hexagon, as shown in Figs. 3(a) and 3(b). The site is in close proximity to two I^- ions at distances of $1.91 \text{ \AA}$ and $2.03 \text{ \AA}$ from the muon and with an $\text{I}-\mu-\text{I}$ angle of 156.84° . The compound consists of stacks of RuI_3 hexagonal layers [see Fig. 3(c)] and this site implies that

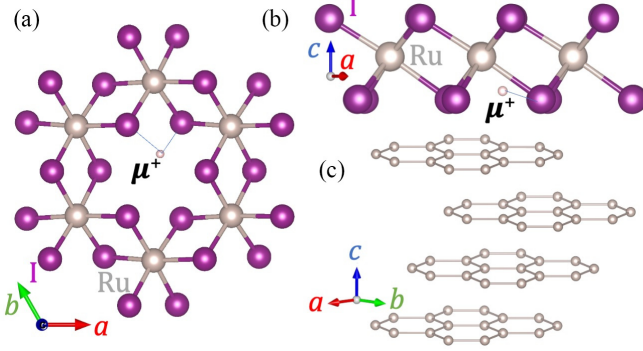


FIG. 3. Muon site for α -RuI₃, viewed (a) along the c axis, showing the structure of the layer, and (b) viewed along the b axis, showing the layer edge-on. (c) The honeycomb Ru³⁺ layers stacked along the c axis.

the muon will be particularly sensitive to the moments within a single layer.

B. Longitudinal field μ SR

To probe the nature of fluctuations in the local magnetic field at the muon site, we studied the effect of an applied longitudinal field (LF) at five temperatures from 0.1 K up to 40 K. The purpose of applying a longitudinal field is to quench the relaxation due to any static magnetic field from nearby nuclear moments and study the dynamical relaxation. In materials without electronic magnetism, a small applied field of 2.5 mT is sufficient to decouple the muon from the field produced by surrounding nuclear moments. Our results show that there is a significant recovery of muon polarization at 2.5 mT, which confirms that the muon experiences significant relaxation from nuclear moments. However, a small fraction of the relaxation in the form of $e^{-\lambda t}$ remains, as shown in Fig. 1, which can be traced back to the effect of the fluctuating Ru³⁺ electronic moments. The dynamical relaxation weakens with increasing longitudinal field. This is demonstrated by Fig. 4, which plots the relaxation rate λ at 0.1 K as a function

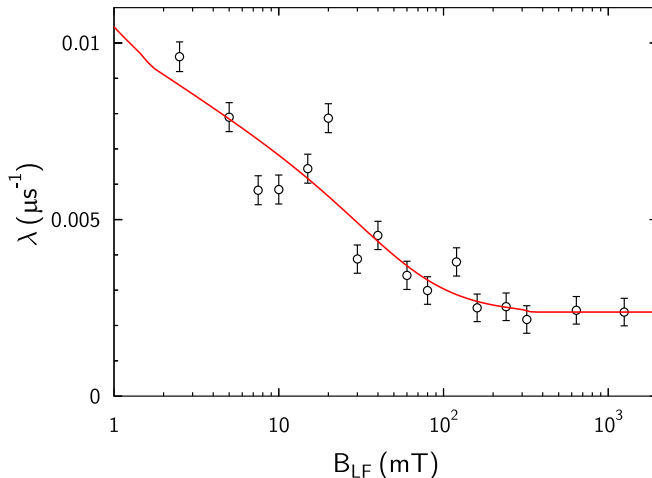


FIG. 4. Dynamical relaxation rate λ as a function of applied longitudinal field B_{LF} applied on α -RuI₃ at 0.1 K, fitted to Eq. (4).

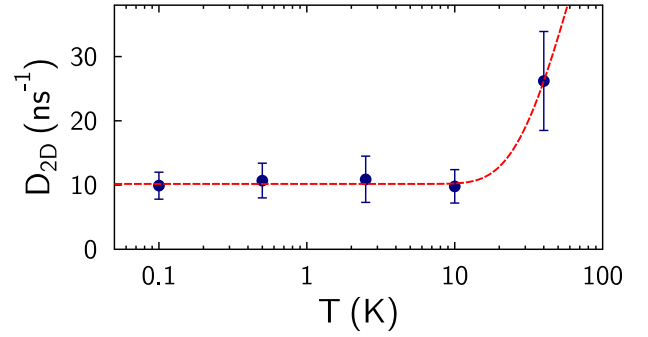


FIG. 5. Fast diffusion rate $D_{2\text{D}}$ of the 2D spin diffusion model extracted by fits in Fig. 4 at five temperatures from 0.1 K to 40 K.

of applied longitudinal field for fields between 2.5 mT and 1.25 T. The plot could be interpreted as a combination of a field-dependent term and a field-independent contribution such that

$$\lambda(B_{\text{LF}}) = \lambda_{2\text{D}}(B_{\text{LF}}) + \lambda_0, \quad (4)$$

where $\lambda_{2\text{D}}(B_{\text{LF}})$ refers to the diffusive spin excitation. Figure 4 also shows that a field of around 0.5 T is required to fully suppress the field-dependent spin diffusion in α -RuI₃, in which case the remaining relaxation is then due to the effect of the localized spin excitation λ_0 [30]. This localized term may originate from the small quantity of orphan spins (identified in the magnetic susceptibility measurements; see Appendix A) or from some contribution from the intrinsic spins, possibly due to dipolar coupling with a mobile electronic excitation that fluctuates much faster than the muon Larmor frequency [30].

To study the spin correlations in α -RuI₃, we fitted the relaxation rate $\lambda(B)$ at five temperatures against both the zero-dimensional (0D) Redfield model, where spin fluctuations are local and are not correlated to any of the neighbours, and a 2D spin diffusion model. For the data in Fig. 4, the 2D spin diffusion model gave a reduced χ^2 value of 5.0, while the 0D Redfield model gave 6.3. Both models have three free parameters and, while the scatter in the data (originating from the very low λ values) is high and leads to large reduced χ^2 values, the 2D model, which accounts for the field-dependent spin excitation term [30,31], was consistently found to fit the LF data set better than the zero-dimensional model. This is because the latter results in a relaxation rate plateau at low fields, but experimentally the low-field relaxation rate is found to increase with decreasing field, as shown in Fig. 4.

The spin diffusion rate $D_{2\text{D}}$, plotted as a function of temperature T in Fig. 5, stays around 10 ns⁻¹ between 0.1 K and 10 K, but shoots up to 26 ns⁻¹ at 40 K (the line in Fig. 5 is a guide to the eye, but assumes that there is a temperature independent and an activated temperature dependent process contributing in parallel). The rising diffusion rate corresponds to a falling entanglement length and is consistent with the scenario where in the high- T classical regime the Ru³⁺ moments are correlated to only their nearest neighbors, whereas at low T (in the quantum regime) the QSL state has a longer entanglement range between spin pairs [31].

We note that the reported Curie-Weiss temperature (15.4 K [20]) lies within the crossover regime between 10 K and 40 K.

The LF- μ SR analysis therefore demonstrates that α -RuI₃ shows the characteristic field dependence of a 2D spin diffusion model, so that there is a fast diffusion within the *ab* plane (deduced by the crystal symmetry) and slow diffusion along *c*. This interpretation supports the idea that there are antiferromagnetic spin correlations among the weak Ru³⁺ moments beyond the nearest neighbors and also that there is a crossover from the quantum to classical regime between 10 K and 40 K, based on the fitted spin diffusion rate D_{2D} .

III. DISCUSSION

We have estimated the relaxation due to the magnetic field resulting from randomly oriented nuclear moments at the muon site identified by the DFT + μ method, as well as a couple of other less likely, but possible, muon sites (which have higher energy). These estimates imply a Kubo-Toyabe relaxation with a value of Δ [see Eq. (2)] between 0.03 μ s⁻¹ and 0.08 μ s⁻¹, since the shortest μ -I bond ranges from 1.67 Å to 1.93 Å and the effect is mostly dominated by the distance to the nearest I⁻ ion(s) as the Van Vleck sum goes as $\sigma^2 \propto \sum_i \frac{\mu_{iI}}{r_{iI}^3}$ (and is dominated by the spin $I = \frac{5}{2}$ iodine nuclei, which each have a moment 7.5 times larger than that of ruthenium nuclei on average, considering all relevant isotopes with their natural abundances). Fitting the 50 mK ZF- μ SR data to a Kubo-Toyabe function together with an exponential relaxation to account for fluctuating moments gives a fitted value of $\Delta = 0.11 \mu$ s⁻¹, confirming the muon's proximity to the nearest I⁻ ion. We have also estimated the magnitude of the static field that would result from antiferromagnetic order, as is observed in the case of α -RuCl₃ and α -RuBr₃. We find that all conceivable muon sites should have a dipolar field of at least 1.5 mT (or muon precession frequency of 0.2 MHz) if Ru³⁺ moments are ordered antiferromagnetically with a moment size of 0.1 μ_B each. This effect would correspond to an oscillation period of less than 5 μ s and this effect is certainly not observed in our ZF- μ SR data (see Fig. 1). In addition, should there be a magnetic order the local static field would have damped the 2 mT weak transverse field- μ SR measurements very effectively (see Fig. 2), which they do not. Therefore, we can conclusively rule out static order for α -RuI₃ down to 50 mK, in stark contrast to what has been observed with μ SR for both α -RuCl₃ [11] and α -RuBr₃ [26].

Due to the more delocalized 4*d* orbitals in RuI₃ than in RuCl₃ [20], *d*-*p* hybridization is understood to be stronger in RuI₃ [32]. It was reported that while the zigzag ordered state was calculated to be the energetically favorable state in RuCl₃ and in RuBr₃, all magnetic states considered for RuI₃ were very similar in energy, which has motivated the search for a QSL state in this compound [20]. DFT calculations performed on RuI₃ have predicted the magnitude of the Ru³⁺ moments to be very small, of the order of 0.1 μ_B [20,33]. RIXS and Raman spectra imply that the low-temperature magnetism can be de-

scribed using $J = \frac{1}{2}$ pseudospins, even though the compound is metallic [22,23]. This is all consistent with susceptibility measurements [17], torque measurements [34], and our μ SR results presented here. An alternative scenario would be to describe α -RuI₃ as a correlated metal exhibiting some kind of paramagnon-like fluctuations. However, there is no available theoretical prediction from such a picture, nor is it clear that the fluctuations would be of the correct frequency to account for the observed behavior.

IV. CONCLUSION

Therefore, through zero-field and weak transverse-field μ SR measurements, we are able to demonstrate the absence of magnetic order in α -RuI₃ down to 50 mK and that its local magnetism can be explained in terms of the effect of both the static nuclear moments and the fluctuating electronic moments. Because these fluctuating moments are small in magnitude, their presence is only directly observable at late times, from $t = 6 \mu$ s onwards, when the nuclear moments-led relaxation fades out. Despite the small size of the Ru³⁺ moments, their fluctuating dynamics are revealed by using an applied longitudinal field that suppresses the relaxation from the nuclear moments. Our analysis shows that the nature of the spin correlations fits well to a 2D spin diffusion model which has a crossover between a quantum and a classical regime between 10 K and 40 K. The results therefore demonstrate the strong likelihood that α -RuI₃ hosts a quantum spin liquid state.

ACKNOWLEDGMENTS

The μ SR experiments were carried out at the ISIS neutron and muon source, UK, and were supported by beam-time Allocation No. RB2310371. We acknowledge the support by the ISIS Neutron and Muon Source, the scholarship funding by the Croucher Foundation, and the UK Research and Innovation (UKRI) under the UK Government's Horizon Europe funding guarantee Grant No. EP/X025861/1.

APPENDIX A: MAGNETIC SUSCEPTIBILITY

Magnetic susceptibility measurements were made using a Quantum Design MPMS3 system at the Clarendon Laboratory, University of Oxford, UK, between 1.8 K and 300 K with a DC field of 500 G. The results show a broadly temperature-independent response above 50 K, consistent with Pauli paramagnetism, while below 50 K there is an upturn in χ , which can be explained by a small fraction of uncorrelated orphan Ru³⁺ spins. The data are fitted against a model $\chi = \frac{C_{\text{orphan}}}{T} + \chi_{\text{Pauli}}$ corresponding to these two effects and yield $C_{\text{orphan}} = 0.0072 \text{ emu K mol}^{-1} \text{Oe}^{-1}$ and $\chi_{\text{Pauli}} = 6.9 \times 10^{-4} \text{ emu mol}^{-1} \text{Oe}^{-1}$. Assuming an orphan Ru³⁺ adopts the low-spin moment of 1.73 μ_B , we estimate the fraction of orphan spins could be estimated to be around 2% (see Fig. 6).

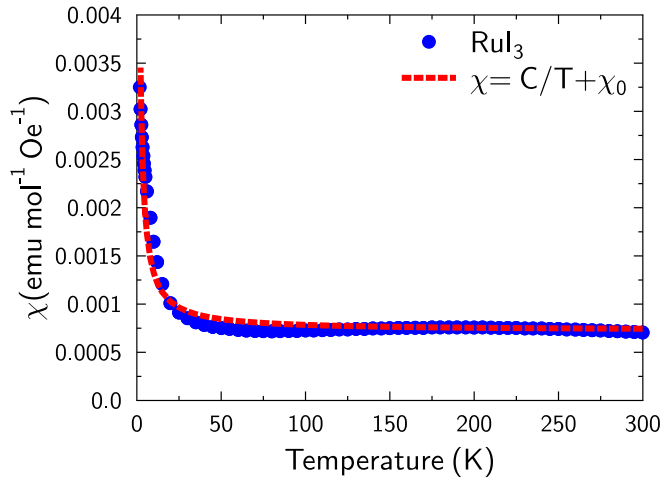


FIG. 6. Magnetic susceptibility of α -RuI₃ showing a flat feature at high temperature and an upturn at low temperature which are represented by Pauli paramagnetism and a small fraction of uncorrelated orphan Ru³⁺ spins.

APPENDIX B: SPIN DIFFUSION MODEL FOR LF- μ SR DATA FITTING

The relaxation rate extracted from LF- μ SR is plotted against the applied field which is then fitted with a combination of a two-dimensional spin diffusion model and a constant background such that

$$\lambda_{\text{LF}}(B_{\text{LF}}) = \lambda_{2\text{D}}(B_{\text{LF}}) + \lambda_0, \quad (\text{B1})$$

where $\lambda_{2\text{D}} = \frac{A^2}{4} J_{2\text{D}}(D_{2\text{D}}, \omega_e)$, with A being the hyperfine coupling constant, $\omega_e = \gamma_e B_{\text{LF}}$, and $J_{2\text{D}}$ is the spectral density of the 2D random walk of the mobile spin excitations (spinons) with diffusion rate $D_{2\text{D}}$ [35]. The spectral density $J_{2\text{D}}$ is given by the Fourier transform of the autocorrelation function for a 2D random walk [30].

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