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Satisfaction and Violation of the Fluctuation-Dissipation Relation in Spin Ice Materials

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We test the fluctuation-dissipation relation (FDR) in spin ice materials $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$ by measuring both the magnetic noise and the out-of-phase part of the susceptibility and comparing their ratio. We show that it is satisfied at temperatures well into the nonergodic region below 600 mK, indicating local equilibrium. In both materials, below 400 mK, low frequency violations develop, showing an excess of noise as in spin glasses, with a frequency threshold of 0.1 Hz. New relaxation pathways and aging properties are unveiled in this frequency range in the ac susceptibility. The FDR remains valid at higher frequencies down to 150 mK.

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At thermodynamic equilibrium, the fluctuation-dissipation theorem relates the spontaneous fluctuations in a system and the linear response function through the fluctuation-dissipation relation (FDR) [1]. When the system is not at equilibrium, deviations from the fluctuation-dissipation theorem are expected and were indeed reported in the context of glasses and liquid crystals [2–5]. A comprehensive theoretical description of the relation between fluctuations and dissipation in out-of-equilibrium systems nevertheless remains challenging [6].

In the specific context of magnetism, out-of-equilibrium properties have been mainly studied in spin glasses [7] and more recently in superspin glasses (assemblies of interacting nanoparticles) [8]. In these systems, simultaneous measurements of magnetic fluctuations and the response function have been performed to directly probe the FDR. They show that these systems can be described by an effective temperature different from the thermal bath which quantifies the deviations from the equilibrium case. In this description, both the fluctuations and response function are made of two contributions: quasiequilibrium or staggered, which respects time translation invariance, and aging, which represents the fact that the response and correlation functions decay slowly with time. This hypothesis, corresponding to a weak ergodicity breakdown, can be understood by considering that the system needs an infinite time to reach its equilibrium state, but that locally its behavior is governed by equilibrium dynamics [9].

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An interesting system where out-of-equilibrium properties are reported and which may break the fluctuation-dissipation relation is spin ice [10,11]. The spin ice state was originally observed in pyrochlore oxide compounds $R_2M_2O_7$, where the R and M ions lie on two interpenetrated corner sharing tetrahedron lattices, when $R =$ Ho, Dy and M is a nonmagnetic element. It is characterized by the "Pauling states," satisfying the ice rule with two spins pointing inward and two outward on each tetrahedron, stabilized experimentally below about 1 K. The Pauling states form a narrow quasidegenerate band, stabilized by geometrical frustration in the absence of structural disorder, retaining extensive entropy down to the lowest temperatures. Below 650 mK magnetization measurements show a clear bifurcation between the field-cooled and zero field-cooled curves [12,13], which indicates an ergodicity breaking and suggests that the system enters into an out-of-equilibrium state. This freezing results from the strong slowing down of the dynamics at these temperatures, due to the rarefaction of the excitations in the system, the so-called magnetic monopoles [14] (which correspond to a violation of the local ice rule, generating a "3-in, 1-out" or "3-out, 1-in" configuration in a tetrahedron). In addition, the system never reaches the predicted ordered ground state at very low temperature [15], even after months of waiting [16,17].

In this Letter, we address the nature of this low temperature state by probing the FDR in two spin ice systems $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$. In both compounds, we identify three characteristic regions: (i) the high temperature region, where the system is trivially in equilibrium, and the FDR is obeyed, as expected; (ii) the intermediate region, where ergodicity breaking is observed in the magnetization, but where we find that the FDR remains obeyed indicating that local equilibrium is preserved; (iii) the low temperature region, where the FDR is violated at low frequency, typically below 0.1 Hz, indicating that the system enters an out-of-equilibrium state. Furthermore, we observe a dissipation process that was not reported to date, as well as aging effects.

The FDR in a magnetic system relates the spectral noise density $S(f)$ to the dissipative part of the ac magnetic susceptibility $\chi''(f)$. In the case of a stationary system $S(f)$ corresponds to the Fourier transformed autocorrelation function of the magnetization $[\overline{M^2(f)}]$ and is characteristic of the spontaneous fluctuations. The relation is written as [18]

$$S(f) = D(f),$$

$$\text{where} \quad S(f) = \overline{M^2(f)} \quad \text{and} \quad D(f) = \frac{2k_B T}{\pi V}\frac{\chi''(f)}{f}, \qquad (1)$$

where T is the temperature, f is the frequency, k_B is the Boltzmann constant, and V is the sample volume. Satisfaction of the FDR indicates that the system is at least in local equilibrium, such that the exchange of energy to and from the heat bath is balanced. Its violation ensures an imbalance in this exchange which generates thermodynamic forces driving temporal evolution through configuration space and aging. Such violations are parametrized by the fluctuation-dissipation ratio $S(f)/D(f)$, characteristic of an effective temperature, equal to the ratio times the temperature of the heat bath [9]. In the aging regime of spin glasses, the effective temperature is observed to be greater than that of the heat bath [7].

To probe this relation, we have developed a superconducting quantum interference device magnetometer equipped with a dilution refrigerator, which allows us to measure the spectral noise density and the ac susceptibility of a zero field-cooled sample in the same series of measurements in order to compare both unambiguously (see Supplemental Material [19]). The experiments were carried out down to 150 and 163 mK, respectively, on two single crystals: a parallelepiped sample of $^{162}Dy_2Ti_2O_7$ of $3.7 \times 5.1 \times 14\ mm^3$ (with zero Dy nuclear moment) [16,20] and a nearly cylindrical $Ho_2Ti_2O_7$ sample of 4.8 mm diameter and 8.9 mm length [19].

In Figs. 1 and 2, the spectral noise density $S(f)$ is plotted together with $D(f)$ for $Dy_2Ti_2O_7$ and for $Ho_2Ti_2O_7$, respectively. In $Dy_2Ti_2O_7$, the measured noise is consistent with previous measurements [21–23]: above 0.6 K, $S(f)$ follows a power law dependence at high frequency and exhibits a plateau at low frequency; the separation between the two regimes defining the characteristic relaxation time. At lower temperature, the plateau regime is not reached in the measurements. In $Ho_2Ti_2O_7$, the shape of the noise is slightly different with the presence of two shoulders down to 1 K, due to the existence of two relaxation times in the

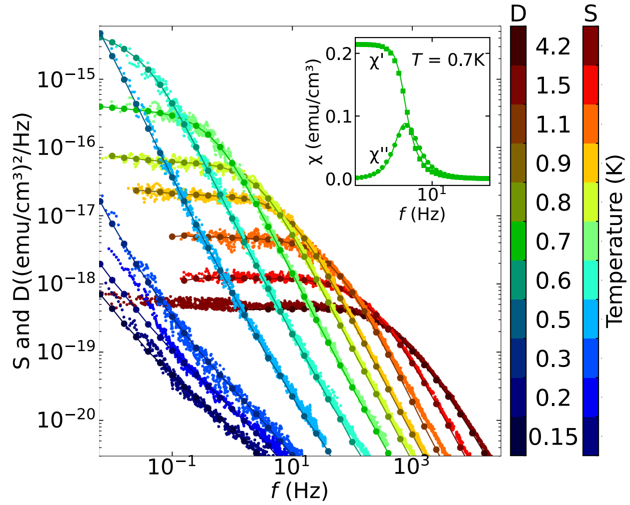

FIG. 1. FDR plot for $Dy_2Ti_2O_7$ on a logarithmic scale: $S(f)$ (small dots) and $D(f)$ (big dots) measured between 4.2 K and 150 mK. Lines are guides to the eye. Inset: ac susceptibility χ' and χ'' vs f measured at 700 mK. The solid lines show the fit to the Cole-Davidson equation (3) with $\tau = 0.429$ s, $\beta = 0.67$, $\chi_S = 0$ and $\chi_T = 0.213\ emu.cm^{-3}$.

system [24], which can be clearly seen in the ac susceptibility (see inset of Fig. 2). Despite these differences, in both systems, $S(f)$ and $D(f)$ overlap down to 300 and 400 mK, in $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$, respectively, thus well below the freezing temperature (650 mK) measured in magnetization measurements. This means that the FDR [Eq. (1)] is obeyed even in the nonergodic regime and suggests that, while the system fails to evolve toward its

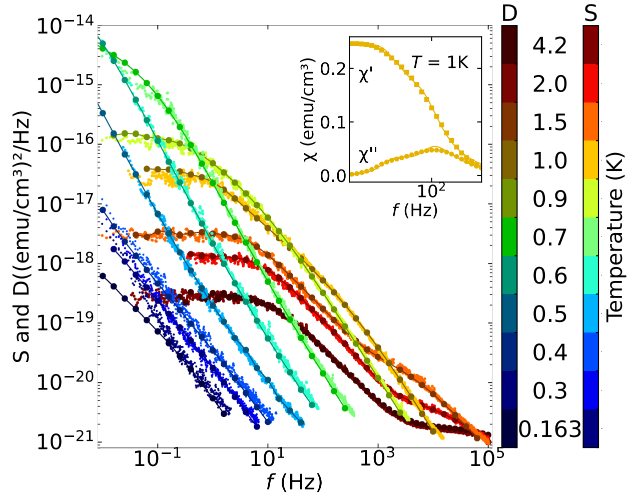

FIG. 2. FDR plot for $Ho_2Ti_2O_7$ on a logarithmic scale: $S(f)$ (small dots) and $D(f)$ (big dots) measured between 4.2 K and 163 mK. Lines are guides to the eye. Inset: ac susceptibility χ' and χ'' vs f measured at 1 K. The solid lines show the fit to a sum of two Cole-Davidson equations (3) with $\tau_1 = 0.098$ s, $\beta_1 = 0.31$, $\chi_{T1} = 0.116\ emu.cm^{-3}$, $\tau_2 = 0.002$ s, $\beta_2 = 0.41$, $\chi_{T2} = 0.13\ emu.cm^{-3}$ and $\chi_{S1} = \chi_{S2} = 0$.

equilibrium state when lowering the temperature or applying a magnetic field, the local dynamics of the spin ice state behave as if the system was at equilibrium. This is in agreement with simulations on the dumbbell model of spin ice parametrized for $Dy_2Ti_2O_7$. Using stochastic dynamics, it was found that the FDR is obeyed down to 400 mK, when the system is allowed to reach local equilibrium [11]. This contrasts, however, with the picture of spin ice behaving as a glass at low temperature [17,25]. This implies that, for all intents and purposes, down to these temperatures, the ac susceptibility and noise measurements are equivalent methods to probe the dynamics and correlation functions in the system. Nevertheless, the ease and precision of ac susceptibility measurements make it the better tool.

Below 300 mK for $Dy_2Ti_2O_7$ and 400 mK for $Ho_2Ti_2O_7$ deep in the nonergodic regime, an excess noise is observed in $S(f)$ compared to $D(f)$ at the lowest measured frequencies, typically below 0.1 Hz [see Figs. 3(a) and 3(b)]. The value of this excess noise is much larger than the noise measured at higher temperatures and in the empty sample holder experiment, which makes us confident that it is intrinsic to the physics of spin ice. The excess noise is associated with an abrupt increase of the $S(f)$ slope in the low frequency regime, while the dissipation part $D(f)$ obtained from the susceptibility increases smoothly. The difference between the fluctuation and the dissipation contributions indicate that the FDR is violated in this low temperature and low frequency regime for both $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$. In $Dy_2Ti_2O_7$, the fluctuation-

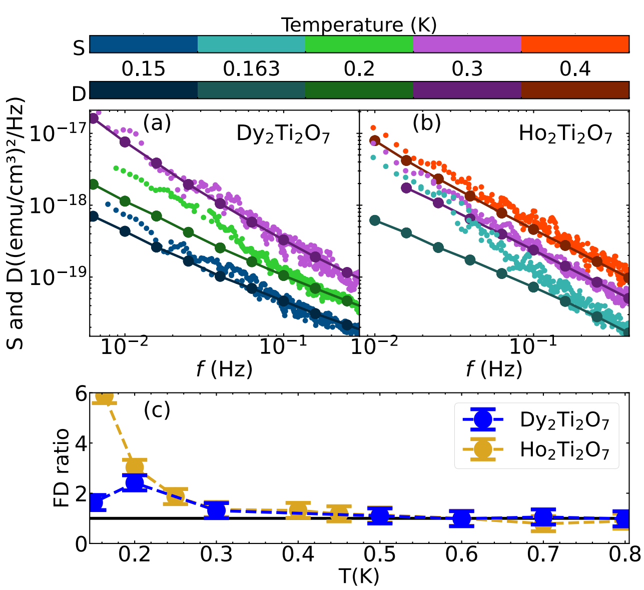

FIG. 3. FDR plot for (a) $Dy_2Ti_2O_7$ and (b) $Ho_2Ti_2O_7$ plotted on a logarithmic scale at low temperature and low frequency: $S(f)$ (small dots) and $D(f)$ (big dots). (c) Fluctuation-dissipation ratio $S(f)/D(f)$ calculated below 0.016 Hz for $Dy_2Ti_2O_7$ (in blue) and $Ho_2Ti_2O_7$ (in gold) from base temperature to 800 mK. The solid black line corresponds to a ratio equal to 1, which is expected when there is no violation of the FDR.

dissipation ratio reaches about 2 below 0.016 Hz and 200 mK. The amplitude of the violation is stronger in $Ho_2Ti_2O_7$ where the fluctuation-dissipation ratio reaches about 6 at 163 mK [see Fig. 3(c)]. This result means that, although the absolute scale is different, an equilibrium system would require a temperature of approximately one kelvin to achieve this ratio [9].

This scenario is characteristic of out-of-equilibrium systems where multiple timescales are present [26] and has been observed in spin-glasses [7]. In the high frequency regime, above the threshold of approximately 0.1 Hz, the FDR is satisfied, indicating local equilibrium over short time and length scales. The breakdown of the FDR with high effective temperature indicates the onset of nonergodicity over mesoscopic time and length scales with consequent temporal evolution and aging. In $Ho_2Ti_2O_7$, where the effect is stronger, a small temperature dependence is observed in the crossover frequency which moves from 0.05 to 0.16 Hz between 400 and 163 mK.

We have analyzed the detailed functional form of $S(f)$ and the ac susceptibility $\chi(f)$, extracting the characteristic relaxation time τ and exponents α and β from expressions

$$S(f) = \frac{S_0}{1 + (2\pi f \tau)^{\alpha}}, \tag{2}$$

$$\text{and} \quad \chi(f) = \chi_S + \frac{\chi_T - \chi_S}{(1 + i2\pi f \tau)^{\beta}} \tag{3}$$

[21,24]. S_0 is the noise at zero frequency, and χ_S and χ_T are the adiabatic and isothermal susceptibilities, respectively. As in previous experiments [22,24,27–29], we find (see Fig. 4) that the timescale τ diverges below 1 K but clearly above the temperature scale on which FDR violations develop. Hence, while the divergence of τ is compatible with the loss of global ergodicity and the zero field-cooled—field-cooled splitting for the static susceptibility, it does not appear to be relevant for the onset of FDR violations.

For $Dy_2Ti_2O_7$, three dynamical regimes can clearly be observed from the temperature dependence of the α parameter (or β for the susceptibility) [22]. Above 2 K and at least up to 4.2 K, when the system is not yet in the spin ice state, α is almost constant. It nevertheless remains below 2, the value for a random walk process, which would be expected for paramagnetic magnetic moments with a single relaxation time. When entering the spin ice regime, α increases, reaching about 1.6 and revealing correlated, constrained dynamics [30]. When entering the freezing regime below 700 mK, α decreases once again reaching 1.3 at 300 mK [see Fig. 4(a)]. These results further illustrate that, while FDR violations surely require complex correlated dynamics, the reverse is not true: α differs from two over a large range of temperatures where the FDR is satisfied indicating global or local equilibrium.

The analysis for $Ho_2Ti_2O_7$ turns out to be less straightforward due to the presence of two relaxation times [24].

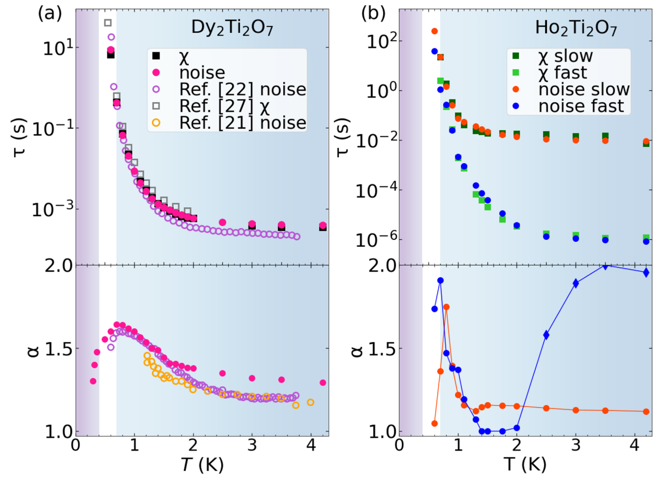

FIG. 4. (a) Relaxation time τ (top) and exponent α (bottom) in $Dy_2Ti_2O_7$ obtained from the fits of the noise data using Eq. (2), together with the τ obtained from the fits of $\chi(f)$ with Eq. (3) and the results from Refs. [21,22,27]. Below 600 mK, it is not possible to fit the noise and ac susceptibility due to the absence of shoulder in the noise and of maxima in χ''. The α parameter can nevertheless be obtained from the slope of the noise. (b) Relaxation times τ_{slow} and τ_{fast} (top) and exponents α_{fast} and α_{slow} (bottom) in $Ho_2Ti_2O_7$ obtained from noise measurements. Above 2 K, α_{fast} is obtained with a high uncertainty because the power law slope of the mode is at the limit of the frequency window (diamond symbols). The relaxation times obtained from ac susceptibility are also shown. The shaded backgrounds indicate the three temperature regimes: (i) equilibrium (blue), (ii) local equilibrium (white), and (iii) out of equilibrium (purple).

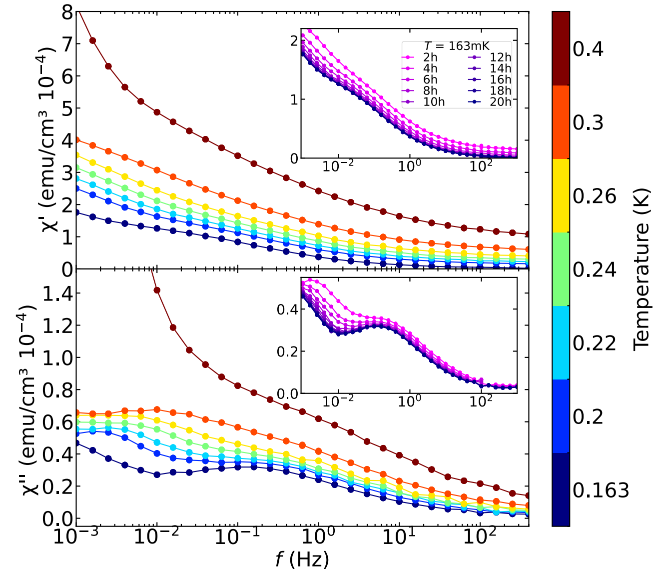

FIG. 5. ac susceptibility χ' (top) and χ'' (bottom) vs f for different temperatures below 300 mK in $Ho_2Ti_2O_7$. Insets: susceptibility measured at 163 mK after different waiting times.

Using the noise, as well as the in-phase and out-of-phase susceptibility, we could determine the relaxation times and α parameters [see Fig. 4(b)] by assuming a sum of two independent contributions following Eqs. (2) for $S(f)$ and (3) for $\chi(f)$. Compared to previous analysis with ac susceptibility only [24], the combination of the two sets of data allows us to deconvolute the contributions of the two relaxation times, especially at low frequency. We find that they have a similar temperature dependence, their relative intensity being almost constant as a function of temperature (see Supplemental Material [19]). These two timescales are thus probably related to the existence of two single spin flip tunneling times resulting from the local spin configurations as proposed theoretically [31,32]. Qualitatively, the relaxation times have a similar temperature dependence to those in $Dy_2Ti_2O_7$: they have a nearly constant value above 2.5 K, which corresponds to the intrinsic tunneling time and strongly increase at low temperature when entering the spin ice regime where slow monopole dynamics are present. The three regimes are also observed in the α parameters, especially the drop at low temperature below 700 mK.

In order to better understand the FDR violation, we have studied the ac susceptibility below 400 mK, where the signal is very small. Interestingly, our ac susceptibility data reveal a relaxation phenomenon that appears at very low temperature and is different from previously identified processes [24,27–29]. It manifests itself as two peaks in χ'' of very small amplitude, which we could observe thanks to a greatly improved sensitivity (see Fig. 5). The peaks appear in both $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$ samples, at slightly different frequencies [19]. They are not directly visible in the noise, but when converting $S(f)$ into $\chi''(f)$ using the FDR relation, the highest frequency peak, which sits in the frequency range where the relation is satisfied can just be resolved. However, the peak at the lowest frequency (below 0.1 Hz) is not visible, due to the presence of excess noise [19]. This low intensity signal shown in Fig. 5 has faster characteristic times (typically a few seconds compared to a few hundreds of seconds for the processes described above) with little temperature dependence compared to the majority relaxation processes. It is thus clearly associated with a new relaxation mechanism, which appears masked at higher temperature, becoming visible only when the other relaxation channels are too slow. In addition to these relaxation processes, ac susceptibility exhibits aging precisely in the regime where the FDR breakdown occurs (see inset of Fig. 5) in close analogy with spin glasses in the same regime [7].

The FDR violations and aging could be due to the presence of trapped noncontractible monopole pairs [11] which can drive the system out of equilibrium [33–36], although this seems unlikely as such pairs are only short-lived at 300 mK [11,33] and require rapid thermal quenches to be stabilized at lower temperatures. However, it is possible that the spin ice materials begin to be sensitive to the degeneracy lifting of the Pauling states induced by the dipole interactions [15,37] in this temperature range. This finite energy bandwidth, which is a correction to the monopole picture of spin ice could lead to a rough free energy surface that drives FDR violations, as in spin glasses [38]. The emerging short timescale could then correspond to monopoles being confined in short loops, with the long timescale a deconfinement time. This separation of scales would be explicitly due to finite bandwidth of energy barriers between Pauling states [22] which is absent in the idealized monopole picture [11]. These ideas could be tested but the investigation of aging effects in the experimentally observed temperature range would require challenging simulations using the full dipolar spin ice Hamiltonian [17,22].

An important remark concerns the role of demagnetizing effects. Because of the ferromagnetic correlations in spin ice, corrections to the susceptibility due to demagnetizing effects can be very important [28,39]. When the real part of the ac susceptibility is sizable, such corrections mix the in-phase and out-of-phase parts of the susceptibility, resulting in a larger intrinsic χ_i'' that is shifted to higher frequencies compared to measured data. However, in our experimental data of Figs. 1 and 2, the FDR is obeyed when comparing $S(f)$ with $D(f)$ which has not been corrected for demagnetizing effects. This seems counterintuitive as noise fluctuations are measured in a zero field environment [19], but presumably fluctuations do create their own internal fields that must also be influenced by the sample shape. Corrections should therefore also be performed on the noise measurements to obtain the "intrinsic" noise response. We are not aware of the correct procedure for this, although we stress that, while corrections change the scale of the response, they do not remove information and the important thing is the measured magnetic noise should scale with the measured susceptibility as first pointed out in Ref. [40]. To estimate the effects of demagnetization corrections on the noise, one can compare $D_i(f)$, obtained from the corrected $\chi_i''(f)$ susceptibility with the measured noise [19]. We note that at low temperature, typically below 500 mK, these corrections become almost negligible due to the very small value of χ' and are thus not relevant in the range where FDR violations are observed. These results also point to the difficulty of making quantitative comparisons between noise from experiments and from models in which the intrinsic response is simulated using periodic boundaries and the Ewald summation [22,30].

In summary, by measuring the FDR for $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$ deep into the highly correlated spin ice regime, we show that the FDR is satisfied in a large frequency range in both systems, including in the nonergodic region below 650 mK. We have confirmed the existence of two distinct relaxation times in $Ho_2Ti_2O_7$, which could be associated with the two tunneling relaxation times proposed theoretically [32]. Below 300 and 400 mK in $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$, respectively, a clear violation to the FDR develops, with an excess noise appearing below 0.1 Hz in both compounds, leading to a fluctuation-dissipation ratio larger than 1 at 0.01 Hz. Such a deviation can only be explained by the presence of several timescales in the system. This violation of the FDR may be the signature of the sensitivity of the system to the Pauli states bandwidth. It is observed in a regime where the ac susceptibility possesses some low intensity residual relaxation processes and presents aging properties. To further characterize these out-of-equilibrium processes and tune the density of monopoles in the low temperature state, an important step forward would be to probe the FDR after controlled quench protocols, which is especially challenging for this kind of experiment. Our study also opens the way to new theoretical developments to understand the nature of the novel relaxation processes in this regime.

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