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## Letter

# Splitting of the magnetic monopole pair-creation energy in spin ice

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## Abstract

The thermodynamics in spin-ice systems are governed by emergent magnetic monopole excitations and, until now, the creation of a pair of these topological defects was associated with one specific pair-creation energy. Here, we show that the electric dipole moments inherent to the magnetic monopoles lift the degeneracy of their creation process and lead to a splitting of the pair-creation energy. We consider this finding to extend the model of magnetic relaxation in spin-ice systems and show that an electric dipole interaction in the theoretically estimated order of magnitude leads to a splitting which can explain the controversially discussed discrepancies between the measured temperature dependence of the magnetic relaxation times and previous theory. By applying our extended model to experimental data of various spin-ice systems, we show its universal applicability and determine a dependence of the electric dipole interaction on the system parameters, which is in accordance with the theoretical model of electric dipole formation.


Keywords: spin ice, magnetic monopoles, magnetic relaxation, AC-susceptibility, electric dipoles

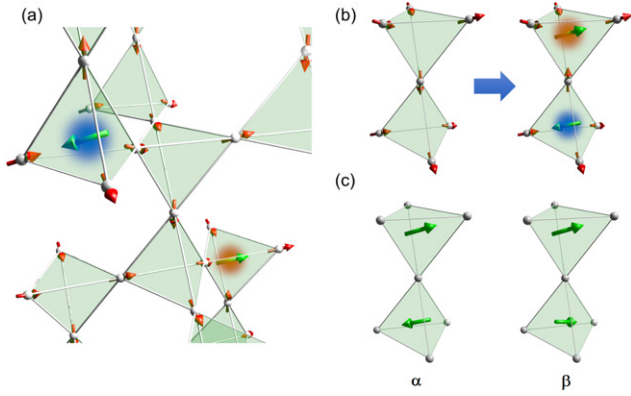
(Some figures may appear in colour only in the online journal)

Pyrochlore-oxides, with stoichiometry  $A_2B_2O_7$  are built up of two sublattices of corner-sharing tetrahedra of the A respectively B atoms. This unique structure, and the interplay of different types of interactions give rise to various exotic phenomena. The competing magnetic dipole and exchange interaction of Ising-spins in insulators, can lead to the so called all-in-all-out state [1] and the observation of an inverted magnetic hysteresis with a negative remanence [2], for a dominant exchange interaction. While a dominant dipole interaction

leads to the formation of the famous spin ice ground-state, with residual entropy [3] and emergent magnetic monopoles [4]. Additional terms of the exchange coupling or disorder can lead to even more exotic phenomena such as a quantum spin-ice state [5, 6]. In metallic systems, the complex magnetism can lead to a parity-breaking electronic nematic phase transition [7] while a related mechanism leads to the formation of electric dipole moments on the emergent magnetic monopoles in spin-ice systems [8, 9]. These electric dipole moments partially extend the analogy of the magnetic monopoles and their electric counterparts, but unlike spins, which cause magnetic-dipole moments connected to electrons, they are no conserved quantities. Therefore, they break the spatial symmetry of the magnetic-monopole pair-creation process without limiting the

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**Figure 1.** (a) Deconfined magnetic monopoles of 1-in-3-out/3-in-1-out excitations, indicated by the blue and red sphere. The red arrows represent the rare-earth Ising spins on the corner-sharing tetrahedra sublattice. The associated electric dipoles are indicated by green arrows. (b) Two neighboring tetrahedra in an exemplary ground state configuration (left) and with the shared spin flipped, resulting in two magnetic monopoles and the two corresponding electric dipoles (right). (c) The two different relative spatial orientations,  $\alpha$  and  $\beta$ , of the electric dipoles formed by the 18 possible spin configurations which lead to the formation of magnetic monopoles.

set of possible relative orientations. Although the theoretical estimates for the magnitude of these electric dipole moments suggested that they should have a measurable influence on the thermodynamics of spin-ice systems, the investigation of their effects is still at an early stage [9–13].

In spin-ice compounds, magnetic rare-earth ions are located on one sub-lattice (figure 1(a)) and a strong crystal electric field (CEF) generated by the surrounding oxygen ions lifts the degeneracy of the magnetic quantum number manifold [14], resulting in a doublet ground-state with a huge energy barrier to the next excited level. This forces the rare-earth magnetic moments to act like Ising spins which are confined to the local  $\langle 111 \rangle$  axis, pointing inward or outward of the tetrahedron center. An effective ferromagnetic next-neighbor interaction leads to a highly frustrated ground state, where two spins point in and two spins point out of each tetrahedron (2-in-2-out configuration). The magnetic excitations, three spins pointing in and one out or vice versa (3-in-1-out/1-in-3-out), of this spin ground state behave like magnetic monopoles [4]. For reasons of conservation of charges, these can only be created pairwise. This excitation process corresponds to a single spin flip in the degenerated 2-in-2-out ground state and was ascribed to a certain excitation energy. In the following, we briefly discuss the origin of the mentioned electric dipoles connected to the emergent magnetic monopoles, based on the work of Khomskii [8] and show how these partially lift the degeneracy of the monopole pair-creation process.

The Hubbard model,

$$H = -t \sum_{\langle ij, \sigma \rangle} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\downarrow} n_{i\uparrow}, \quad (1)$$

is used to describe the interplay between the matrix element of electron hopping  $t$  between neighboring sites  $\langle i, j \rangle$  and the Coulomb energy  $U$  between the electric charges. For  $U \gg t$ ,

the electrons are treated as localized, with antiferromagnetic nearest-neighbor exchange interaction  $J = 2t^2/U$ . But especially for frustrated systems it has been shown [15] that the electrons are not completely localized and spontaneous charge redistributions depending on the spin configuration can occur. In spin-ice systems, this leads to the formation of electric dipole moments pointing along a local  $\langle 111 \rangle$  direction towards the minority spin [8] on the less symmetric 3-in-1-out and 1-in-3-out configurations, which correspond to the magnetic monopole excitations. All other configurations, especially the 2-in-2-out ground-state configurations, show no net charge redistribution. Interestingly, the same result can be derived from applying the spin-current model to spin-ice systems [9].

The energy cost for a single isolated magnetic monopole  $\Delta$  is [4, 16],

$$\Delta = \frac{8}{3} \left[ 1 + \sqrt{\frac{2}{3}} \right] D - \frac{2J}{3}, \quad (2)$$

with the dipolar coupling constant,  $D$ , and the exchange interaction,  $J$ . We assume the electric Coulomb energy for creating the electric dipole moment to be already comprised in the value of the exchange interaction  $J = 2t^2/U$  which is a direct consequence of the Hubbard model.

When the shared spin of two adjacent 2-in-2-out tetrahedra is flipped, it creates a pair of oppositely charged magnetic monopoles and their associated electric dipole moments  $\vec{d}$  at a distance  $a_d$  (figure 1(b)). The energy cost for this process was presumed to be the energy cost for two isolated magnetic monopoles, plus their magnetic Coulomb energy at the distance  $a_d$ ,  $E_{mc}$  (which is negative for opposite charges) [4],

$$\Delta_p = 2\Delta + E_{mc}. \quad (3)$$

In addition, we are now taking account of the electric dipole interaction and the associated potential energy of the two dipoles  $\vec{d}_i$  and  $\vec{d}_j$  at a distance  $a_d$  [17],

$$E_{ed} = \frac{1}{4\pi\epsilon k_B} \frac{\vec{d}_i \cdot \vec{d}_j - 3(\vec{d}_i \cdot \vec{r}_{ij})(\vec{d}_j \cdot \vec{r}_{ij})}{a_d^3}, \quad (4)$$

where  $\vec{r}_{ij}$  is a unit vector pointing from the center of tetrahedron  $i$  to the center of tetrahedron  $j$  and the magnetic monopole pair-creation energy becomes:

$$\Delta_p = 2\Delta + E_{mc} + E_{ed}. \quad (5)$$

This equation now depends on the relative orientation between  $\vec{d}_i$  and  $\vec{d}_j$ . Considering the local Ising anisotropy and the structure of the pyrochlore lattice, the possible orientations between the electric dipole moments are countable and the electric dipole energy  $E_{ed}$  gets quantized. For two neighboring tetrahedra which fulfill the 2-in-2-out rule, there are 18 possible spin configurations. The flipping of the shared spin leads to the formation of an oppositely charged pair of magnetic monopoles and the incidental electric dipoles. As mentioned above, the electric dipole moments always point at the minority spin, along a local  $\langle 111 \rangle$  direction and these 18 initial spin configurations produce 2 relative spatial electric dipole orientations:  $\alpha$  which is 6-fold degenerated and  $\beta$  which is 12-fold degenerated (figure 1(c)) after the shared spin is flipped. Note that any

dipole orientation where the dipole points at the shared spin of two tetrahedra cannot be produced from the 2-in-2-out ground state. By applying formula (4), it turns out that the energies of the two relative electric dipole orientations have the same absolute value, while  $E_{ed}^\alpha$  is negative and  $E_{ed}^\beta$  positive:

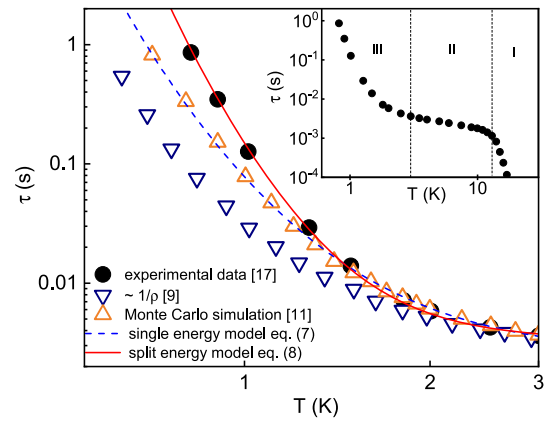
$$-E_{ed}^\alpha = E_{ed}^\beta. \quad (6)$$

Thus, there are two different monopole pair-creation energies, the smaller  $\Delta_p^\alpha$  which is six-fold degenerated and the larger  $\Delta_p^\beta$  which is 12-fold degenerated. In the following, we consider the implications of this finding for the magnetic relaxation in spin-ice systems.

Experimental data evidences, that the evolution of the spin-relaxation times in the low-temperature limit of spin-ice systems can be attributed to a thermally activated process. But until now, the associated energies could not be brought into agreement with any microscopic process and did not seem to show a universal energy scaling for different spin-ice compounds [16, 18–24]. In figure 2, we show the temperature-dependent spin-relaxation time  $\tau$  of  $\text{Dy}_2\text{Ti}_2\text{O}_7$  extracted from dynamic magnetic-susceptibility measurements [25]. Since they were already intensively studied by analytical [16] and numerical means [19], we want to use them, to briefly discuss the present understanding of the magnetic relaxation in spin-ice systems. The spin-relaxation time can be divided into three regions (see inset of figure 2) [19]. In the high-temperature regime I, the relaxation is attributed to thermal excitations of higher CEF levels [19, 26–29]. Due to the large CEF splitting such excitation becomes unlikely for lower temperatures and spin flips mainly occur via a tunneling between the two Ising states. The characteristic time constant of the tunneling sets the time scale for the dynamics and the relaxation time enters the so-called quasi-plateau region II [18, 19, 26, 27]. For even lower temperatures, the dynamics slow down and the spin-relaxation time again seems to acquire a thermally activated behavior in region III [18, 19, 30].

At low temperatures, where the excitation of higher CEF levels and double excitations (all-in and all-out) can be neglected, a spin flip either corresponds to a magnetic monopole pair creation/annihilation, or the hopping of a single monopole. Therefore, the temperature dependence of the spin-relaxation time is inextricably linked to either one or both of these processes. The temperature dependence of the relaxation times due to free diffusion of monopoles should behave like  $\tau \propto \rho^{-1}$ , where  $\rho$  is the temperature-dependent monopole density [16]. For the most simple approximation in the low-temperature limit,  $\rho \approx 2 \exp(-\Delta/T)$  and  $\rho^{-1}$ , therefore, resembles a thermally activated behavior with a barrier equal to the energy for a single isolated monopole. In the intermediate temperature regime II, this approach sufficiently captures the experimental data, if  $\rho$  is derived from Debye–Hückel theory [16]. But for lower temperatures it drastically underestimates the spin-relaxation times (figure 2).

Since the magnetic Coulomb interaction between the monopoles has a similar magnitude as the monopole energy itself, it drives them to annihilation, or binds them in pairs [19, 31–35]. Thus, the monopole mobility is heavily restricted and



**Figure 2.** Low temperature spin-relaxation times of  $\text{Dy}_2\text{Ti}_2\text{O}_7$  (black circles), extracted from dynamic magnetic-susceptibility measurements [25]. The blue downward triangles are a  $\tau \propto \rho^{-1}$  behavior taken from reference [16] representing relaxation due to monopole diffusion. The dashed blue line shows the results using equation (7), with a single energy  $\Delta_p = 5.8$  K as used in reference [19] representing the relaxation due to monopole pair creation without consideration of the electric dipole moments. The orange upward triangles are obtained from Monte Carlo simulations of the dipolar spin-ice model [19] (without consideration of the electric dipole moments). The red line shows the calculated results using equation (8) with the split monopole pair creation energy motivated in the main text. The used parameters are summarized in table 1. The inset shows the whole temperature range of the data provided in [25] divided in the three regions as described in the main text.

their free diffusion suppressed. Accordingly, we assume the monopole pair creation to become the predominant relaxation process in the low-temperature limit.

The monopole pair creation is a thermally activated process and described by an Arrhenius law on top of the quasi plateau, represented by  $\tau_0$ ,

$$\tau(T) = \tau_0 + A \exp(\Delta_p/T). \quad (7)$$

If we ignore the electric dipole interaction and take the value of  $\Delta_p = 5.8$  K from reference [19] while  $\tau_0$  and  $A$  remain free parameters, the analytic equation (7) perfectly reproduces the relaxation times obtained from Monte Carlo simulations (without considering the electric dipoles) [19]. This reinforces the assumption that the temperature dependence of the spin-relaxation times in the low-temperature limit is governed by monopole pair creation and annihilation, rather than free monopole diffusion.

However, the remaining discrepancies of the analytical and the numerical approach with the experimental data (figure 2) underpin the importance of a more accurate treatment of this process. Above, we have shown that the degeneracy of the magnetic monopole pair creation is lifted by the electric dipoles and that there are two different magnetic monopole pair-creation energies  $\Delta_p^\alpha$  and  $\Delta_p^\beta$ . Since the creation energy of a magnetic monopole pair is predetermined by the initial spin configuration at each lattice site, we propose to replace the temperature dependent part of equation (7) by an weighted arithmetic mean of two Arrhenius laws,

$$\tau = \tau_0 + A[1/3 \exp(\Delta_p^\alpha/T) + 2/3 \exp(\Delta_p^\beta/T)]. \quad (8)$$



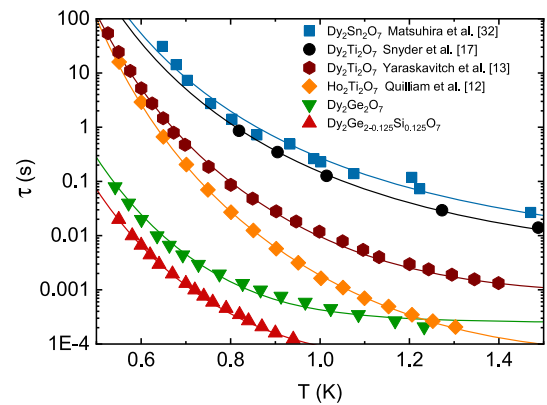
**Table 1.** The parameters used to describe the experimental data as well as other useful parameters of the investigated spin-ice systems. The asterisks mark the parameters used to describe the data which were corrected for demagnetization effects. Note that in [36, 40] an antiferromagnetic exchange interaction has a negative value by convention. Further  $E_{mc} = \mu_0 q_1 q_2 / 4\pi k_B a_d$ ,  $q_1, q_2 = \pm 2\mu/a_d$ ,  $a_d = \sqrt{3/2}a$  is the distance between the tetrahedron centers,  $J = 3J_{nn}$  and  $D = 3/5D_{nn}$ .

	Ho <sub>2</sub> Ti <sub>2</sub> O <sub>7</sub>	Dy <sub>2</sub> Sn <sub>2</sub> O <sub>7</sub>	Dy <sub>2</sub> Ti <sub>2</sub> O <sub>7</sub>	Dy <sub>2</sub> Ge <sub>2</sub> O <sub>7</sub>	Dy <sub>2</sub> Ge <sub>1.875</sub> Si <sub>0.125</sub> O <sub>7</sub>
$a$ (Å)	10.105 [37]	10.400 [36]	10.127 [37]	9.930 [40]	9.906 [40]
$a_d$ (Å)	4.376	4.503	4.385	4.300	4.289
$q$ ( $\mu_B \text{ \AA}^{-1}$ )	4.571	4.441	4.561	4.651	4.663
$D_{nn}$ (K)	2.278	2.089	2.263	2.400	2.417
$-J_{nn}/D_{nn}$	-0.27 [36]	-0.46 [36]	-0.49 [36]	-0.71 [40]	-0.74 [40]
$J$ (K)	1.845	2.883	3.326	5.113	5.367
$D$ (K)	1.366	1.254	1.357	1.440	1.451
$\Delta$ (K)	5.390	4.150	4.359	3.567	3.449
$E_{mc}$ (K)	-2.977	-2.730	-2.957	-3.136	-3.159
$E_{ed}$ (K)	2.897*	2.390	2.539/3.739*	3.171	3.302
$\tau_0$ (s)	$5.1 \times 10^{-5}$ *	$4.2 \times 10^{-3}$	$3.2 \times 10^{-3}/7.3 \times 10^{-4}$ *	$2.4 \times 10^{-4}$	$3.8 \times 10^{-5}$
$A$ (s)	$4.8 \times 10^{-8}$ *	$1.4 \times 10^{-4}$	$5.3 \times 10^{-5}/9.5 \times 10^{-7}$ *	$2.2 \times 10^{-7}$	$7.7 \times 10^{-8}$

Because this extended model gives similar good agreements with the experimental data for various triples of the parameters  $\Delta$ ,  $E_{mc}$  and  $E_{ed}$ , we have fixed  $\Delta$  and  $E_{mc}$  to values calculated with the system parameters obtained from references [36, 37] (see also table 1). Therefore, the absolute energy scale is set while the splitting of the two energy barriers is left as a free parameter. The parameters were chosen to fit the low-temperature limit, where our approach is able to reproduce the spin-relaxation times of Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> to a great extent and gives a significant improvement compared to previous approaches (solid line in figure 2).

The absolute value of the electric dipole interaction energy  $|E_{ed}|$  obtained from our fit is 2.54 K for Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>. With  $\epsilon \sim 64\epsilon_0$  [38], we obtain an electric dipole moment of  $\sim 5.7 \times 10^{-30}$  C m which is very close to the rough estimate of  $\sim 6 \times 10^{-30}$  C m given in reference [9].

To further test our model, we applied it to experimental data for various spin-ice systems. In figure 3 we show the relaxation times of Dy<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> [39], Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [21, 25], Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [20], Dy<sub>2</sub>Ge<sub>2</sub>O<sub>7</sub>, and Dy<sub>2</sub>Ge<sub>1.875</sub>Si<sub>0.125</sub>O<sub>7</sub>. To obtain the spin-relaxation time  $\tau$ , the temperature dependence of the complex dynamic susceptibility of Dy<sub>2</sub>Ge<sub>2</sub>O<sub>7</sub> and Dy<sub>2</sub>Ge<sub>1.875</sub>Si<sub>0.125</sub>O<sub>7</sub> was measured with a pair of compensated coils connected to a lock-in amplifier for various fixed frequencies  $f$  (figure 4). With the peak temperature,  $T_p$ , of the imaginary part of the dynamic magnetic susceptibility,  $\chi''$ ,  $\tau$  was obtained from the following relation:  $\tau(T_p) = (2\pi f)^{-1}$ . The same relation was used for Dy<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> in [39], while for Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [21, 25] and Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [20]  $\tau$  was obtained from the relation  $\tau(T) = (2\pi f_p)^{-1}$ , where  $f_p$  is the peak frequency of  $\chi''$ , by sweeping the frequency at a fixed temperature  $T$ . In [20] it was shown, that both methods can give slightly different results in the intermediate temperature range, but especially the values for  $\tau$  in the low-temperature limit are unaffected. Further it should be noted, that for Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [21] and Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [20] the experimental data was corrected for demagnetization effects. Unfortunately, a quantitative correct determination of the demagnetization factor for arbitrary given sample geometries

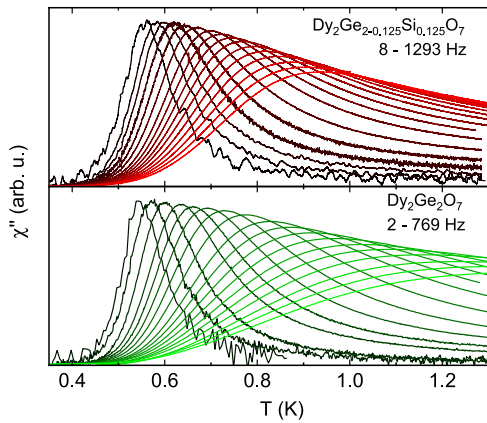


**Figure 3.** Temperature dependence of the relaxation times of Dy<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> (blue squares) [39], Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> (black circles) [25], Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> (brown hexagons) [21], Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> (orange tilted squares) [20], Dy<sub>2</sub>Ge<sub>2</sub>O<sub>7</sub> (green downward triangles), and Dy<sub>2</sub>Ge<sub>1.875</sub>Si<sub>0.125</sub>O<sub>7</sub> (red upward triangles). The solid lines illustrate the results obtained from equation (8). The used parameters can be found in table 1.

is not possible using analytical methods. Therefore, demagnetization effects were not taken into account for Dy<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> [39], Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> [25], Dy<sub>2</sub>Ge<sub>2</sub>O<sub>7</sub>, and Dy<sub>2</sub>Ge<sub>1.875</sub>Si<sub>0.125</sub>O<sub>7</sub>.

The correction of the experimental data in [20, 21] leads to a steeper slope of the relaxation times. In general, demagnetization corrections for the various spin ice systems investigated, are expected to have the same effect with similar strength on the obtained relaxation time, if no extreme sample geometries are used. The solid lines in figure 3 correspond to results of equation (8) with the parameters  $\Delta$  and  $E_{mc}$  fixed to values calculated with the system parameters obtained from references [36, 37, 40]. All the parameters used to describe the data, as well as other useful parameters of the investigated spin-ice systems are shown in table 1.

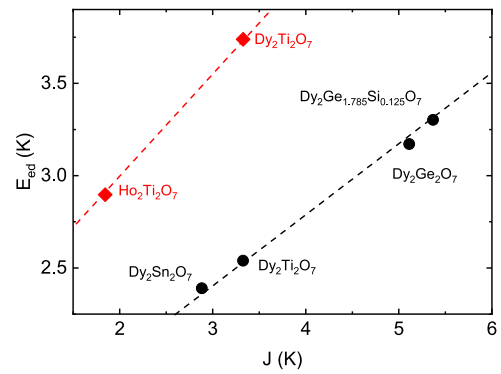
The most prominent difference in the family of spin-ice compounds is the huge variation of the absolute timescales, which are set by the spin-tunneling rates, represented by the parameters  $\tau_0$  and  $A$ , which change by orders of magnitude. But comparing only the extracted values of  $\tau_0$  and  $A$



**Figure 4.** Temperature dependence of the imaginary part of the complex dynamic susceptibility  $\chi''(T)$  of  $\text{Dy}_2\text{Ge}_{2-0.125}\text{Si}_{0.125}\text{O}_7$  (upper panel) and  $\text{Dy}_2\text{Ge}_2\text{O}_7$  (lower panel). From the frequency-dependent maxima, the temperature-dependent spin-relaxation times  $\tau$  can be extracted.

of the Dy compounds, a systematic dependence on the lattice parameter is found (table 1). By comparing the data for  $\text{Dy}_2\text{Ti}_2\text{O}_7$  from [21, 25] the influence of the correction for demagnetization effects becomes apparent. While both sets of data can be described with equation (8), the corrected data shows even larger discrepancies to previous models and reinforces the necessity of our new approach. The main difference is, that the electric dipole interaction energy  $E_{\text{ed}}$  needed to describe the data is 3.74 K instead of 2.54 K. This leads to an electric dipole moment of  $6.96 \times 10^{-30}$  C m which is still close to the mentioned estimate of  $\sim 6 \times 10^{-30}$  C m [9]. If we separately compare the electric dipole interaction energy needed to describe the corrected and uncorrected sets of data (figure 5), we see a systematic correlation with the exchange interaction  $J$ . By considering that  $J$  is a key parameter for the electric dipole formation, no matter which model is used for describing the underlying mechanism (the Hubbard model [8] or the spin-current model [9]), and that the size of the electric dipole moment is a monotonic function of  $J$  [9] this is exactly what would be expected. While the electric dipole interaction energy needed to describe the experimental data confirms the theoretical expectations, its magnitude implies that it should also have a non negligible influence on other thermodynamic quantities.

In [36, 40] the parameter  $J_{mn}$ , which we used to calculate  $\Delta$ , was obtained from comparing Monte Carlo simulations [41] to specific-heat data. In the underlying model, the electric dipole interaction was not considered and, therefore, the procedure used above is partially inconsistent. In contrast to the relaxation times, the specific heat is an equilibrium quantity, thus the splitting of the magnetic monopole pair-creation energy would not be directly observable since the energy of a single isolated monopole does not change. Nevertheless, similar as the magnetic Coulomb interaction [16], the electric dipole interaction should also lower the systems free energy. In consequence,  $\Delta$  obtained experimentally from specific heat would be larger and, hence,  $E_{\text{ed}}$  needed to explain the spin-relaxation time smaller. Of course, not only the specific heat



**Figure 5.** The electric dipole interaction  $E_{\text{ed}}$  needed to explain the relaxation times of the investigated spin-ice systems, plotted over the exchange interaction  $J$ . Values corrected (red diamonds) and not corrected for demagnetization (black circles) are shown. The dashed lines are a guide to the eye.

should be altered by the electric dipole interaction, but signatures of the split monopole pair-creation energy, in particular, should be present in other measurements.

A detailed discussion of all the existing experimental data of spin-ice systems with regard to our findings is out of the scope of this work, but we want to briefly mention some noticeable results. In reference [42], two different activation energies were identified by various heat-relaxation and thermal-transport measurements in  $\text{Dy}_2\text{Ti}_2\text{O}_7$ . Giblin *et al* [34] stated the necessity of two characteristic temperature scales to describe the relaxation after a magnetic-field pulse in the low-temperature spin-ice Coulomb phase of  $\text{Dy}_2\text{Ti}_2\text{O}_7$ . In the polarized neutron scattering data of  $\text{Ho}_2\text{Ti}_2\text{O}_7$  [32], a wave-vector-independent thermal contribution to the spin-flip scattering was described using two characteristic energies. Certainly, these findings were not attributed to the splitting of the magnetic monopole pair-creation energy due to electric dipoles at this time, but there are striking similarities to the expected implications from our findings. Another interesting point we want to mention, is the ordered monopole double-layer ground state predicted for a sufficient electric dipole interaction by Monte Carlo simulations [13]. As already mentioned, the values of  $J$  and  $E_{\text{ed}}$  shown in table 1 are probably slightly overestimated, and, therefore it is not clear where exactly the examined spin-ice compounds should be placed in the phase diagram of [13]. But it might be still possible, that some of them could evolve the mentioned bilayered monopole crystal structure at low temperatures. Although it is not clear on which timescales such a crystallization process would take place, it could be a possible explanation for the shortfall of the Pauling entropy in  $\text{Dy}_2\text{Ge}_2\text{O}_7$  and  $\text{Dy}_2\text{Ge}_{1.875}\text{Si}_{0.125}\text{O}_7$  [40] and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  [43], while it should be stated that the results in [43] are under debate [44]. Finally, we want to point out, that the proposed size of the electric dipole moments implies, that measurable effects could be achieved by applying technical feasible electrical fields in the order of  $10^7$  V  $\text{m}^{-1}$ . This opens new ways for future experiments and possible applications.

In conclusion, we have shown that the theoretically predicted electric dipole moments inherent to the magnetic monopole excitations, lift the degeneracy of their creation process and lead to a splitting of the pair-creation energy. We incorporated this finding in a semi-phenomenological model for the spin-relaxation times of spin-ice systems and showed that the splitting caused by electric dipole moments with magnitudes as theoretically estimated can explain the frequently mentioned discrepancies between the experimentally obtained temperature evolution and theoretical expectations. We applied our model to experimental data for various spin-ice systems and found that the electric dipole interaction energy necessary to explain the experimental data not only matches the theoretical estimate, but also shows a correlation to the system parameters which is in accordance with the microscopic mechanism of electric dipole formation and naturally explains the previously observed, seemingly nonuniversal energy scaling for different spin ice systems.

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