Field evolution of the magnetic structures in $\text{Er}_2\text{Ti}_2\text{O}_7$ through the critical point.

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(Dated: August 19, 2010)

We have measured neutron diffraction patterns in a single crystal sample of the pyrochlore compound $\text{Er}_2\text{Ti}_2\text{O}_7$ in the antiferromagnetic phase ($T=0.3$ K), as a function of the magnetic field, up to 6 T, applied along the [110] direction. We determine all the characteristics of the magnetic structure throughout the quantum critical point at $H_c=2$ T. As a main result, all Er moments align along the field at $H_c$ and their values reach a minimum. Using a four-sublattice self-consistent calculation, we show that the evolution of the magnetic structure and the value of the critical field are rather well reproduced using the same anisotropic exchange tensor as that accounting for the local paramagnetic susceptibility. In contrast, an isotropic exchange tensor does not match the moment variations through the critical point. The model also accounts semi-quantitatively for other experimental data previously measured, such as the field dependence of the heat capacity, energy of the dispersionless inelastic modes and transition temperature.

I. INTRODUCTION

Geometrical magnetic frustration allows one to obtain materials with tunable properties. It yields a large landscape of possible magnetic ground states, due to the inability of the system to choose a unique spin configuration which minimizes the energy. Rare earth pyrochlores $\text{R}_2\text{Ti}_2\text{O}_7$, where the R magnetic moments reside on the sumits of corner sharing tetrahedra, are model systems to study such effects. Here, geometrical frustration does not arise from the competition of magnetic interactions, but rather emerges in the context of a highly symmetrical structure, from the subtle interplay of three main energy terms: the single ion crystal field anisotropy, the exchange interaction and the magnetic dipolar coupling. In $\text{R}_2\text{Ti}_2\text{O}_7$, the trigonal symmetry of the crystal field at the R site comes from the oxygen environment. At low temperature, this yields two generic behaviors, Ising-like ($\text{Ho}$, $\text{Dy}$, $\text{Tb}$) or XY-like ($\text{Er}$, $\text{Yb}$), depending on whether the <111> axes are easy or hard anisotropy axes for the magnetic moments. The final selection of a magnetic state within the ground state manifold is determined by the nature, lengthscale and sign of the magnetic interaction, by perturbation energy terms, or by "order by disorder" processes.

The spin ice compounds $\text{Ho}_2\text{Ti}_2\text{O}_7$ and $\text{Dy}_2\text{Ti}_2\text{O}_7$, combining a strong Ising anisotropy with an effective ferromagnetic exchange, show exotic short range orders with macroscopic entropy and peculiar excitations, where the R dipolar moments fractionalize into magnetic monopoles. Here we focus on $\text{Er}_2\text{Ti}_2\text{O}_7$, a compound with "reversed" behavior, namely with planar XY anisotropy and antiferromagnetic (AF) interactions, and showing magnetic order below 1.2 K.

$\text{Er}_2\text{Ti}_2\text{O}_7$ was proposed to realize a model type XY antiferromagnet, for which theory predicts a fluctuation induced symmetry breaking, leading to magnetic long range ordering. Indeed, for the crystal field ground Kramers doublet of the $\text{Er}^{3+}$ ion, the threefold symmetry [111] axis is a hard magnetic axis, the easy plane being the local (111) plane. Below $T_K=1.2$ K, the antiferromagnetic structure has a $k=0$ propagation vector and it is defined by the basis vectors $\psi_2$, which transform according to the irreducible representation $\Gamma_5$, following the Kovalev notations used in Refs.6 and 7. It consists of six equally populated domains, as shown by spherical neutron polarimetry. The selection of this particular state among the possible basis states of the $k=0$ manifold is still subject to discussion. Surprisingly, it is the only non-coplanar structure among all others, whereas order by disorder processes usually select coplanar or collinear orders. It also differs from the the so-called Palmer-Chalker state, predicted to be the ground state in the presence of AF isotropic exchange and dipolar interactions. An energetic selection of the ordered states was recently proposed, arising from sixth order terms in the crystal field Hamiltonian, together with possible anisotropic exchange, dipolar and Dzyaloshinskii interactions. An anisotropic molecular field tensor with antiferromagnetic components, reinforcing the crystal field planar anisotropy, has indeed been shown to be present in $\text{Er}_2\text{Ti}_2\text{O}_7$ using polarized neutron diffraction, by measuring the local susceptibility in the paramagnetic phase.

The magnetic structure near $T=0$ has been shown to coexist with spin fluctuations by $\mu$SR spectroscopy: a non-vanishing spin dynamics washes out the precession signal usually observed by muons in ordered magnets. This is confirmed by the presence of soft collective excitations probed by inelastic neutron scattering. In this latter work, it is shown that application of a magnetic order, by perturbation energy terms, or by "order by disorder" processes.
field (along [110]) decreases the Néel temperature, resulting in a zero temperature phase transition at a critical field \( H_c \approx 1.8 \) T. The magnetic ordered state is suggested to transform into some kind of spin liquid state above the critical field, through a second order quantum phase transition driven by spin fluctuations. The field evolution of the magnetic structure was qualitatively understood as a smooth deformation from the zero field \( \psi_2 \) configuration to a field configuration above \( H_c \) where the moments are aligned with or close to the field direction.

In these studies, however, the spin configurations could not be characterised in detail, considering the limited information provided by the cold neutron measurements.

In this work, we present detailed in-field neutron diffraction experiments in the AF phase of \( \text{Er}_2\text{Ti}_2\text{O}_7 \) using hot neutrons. This allows us to follow the evolution of the ground state induced by a magnetic field applied along [110], through the quantum critical point and up to a field of 6 T, and to clarify both questions of the domain structure and of the microscopic local spin structure within a tetrahedron. We show that the Er moment magnitude presents a minimum around \( H_c \), which can be considered as the spin-flip field for the [110] direction. Up to 6 T, the Er moments do not recover a fully collinear structure because [110] is not a principal direction for the local g-tensor of half the Er sites. We apply a self-consistent calculation, in the molecular field approximation, to try and explain this evolution quantitatively; our model takes into account the crystal field interaction previously studied in Refs. and together with an anisotropic molecular field tensor, arising from exchange couplings with the 6 nearest neighbours of a given ion. We show that we can account for the field evolution of the magnetic structure in the AF phase (except in a limited field range below \( H_c \)) by the same molecular field tensor which accounts for the thermal variation of the local susceptibility in the paramagnetic phase. We apply our model to compute the lowest excitation energies at 0.05 K, the heat capacity in the paramagnetic phase, and the \((H, T_N)\) phase diagram; a reasonable agreement is obtained with the experimental data of Refs. and .

II. EXPERIMENTAL

A single crystal of \( \text{Er}_2\text{Ti}_2\text{O}_7 \) was grown by the floating zone technique, using a mirror furnace. It was placed at the bottom of a dilution inset, inside a superconducting coil. The vertical axis of the magnetic field was aligned with the [110] axis, with a slight misorientation discussed below. Neutron diffraction measurements were performed at the ORPhee reactor of the Laboratoire Léon Brillouin, on the Super-6T2 spectrometer in the unpolarized neutron version, with an incident neutron wavelength \( \lambda_n = 0.9 \) Å. The nuclear structure was characterized by zero field neutron diffraction at two temperatures above \( T_N \), 100 K and 5 K, allowing the lattice con-

![FIG. 1. Integrated intensities versus the field \( H \) applied along [110] of some typical Bragg peaks measured at 0.3 K in \( \text{Er}_2\text{Ti}_2\text{O}_7 \).](image)

constant, positional parameters, occupancy factors, isotropic temperature factors and extinction parameters to be refined, within the space group \( F\bar{3}m \). To determine the magnetic structures, about 300 Bragg peaks were collected at 0.3 K, in zero field and for 10 field values in the range 0 - 6 T.

III. EVOLUTION OF THE MAGNETIC STRUCTURE

Figure I shows the field dependence of the integrated intensities of some particular Bragg peaks. The \((h, -h, l)\) peaks, situated in the horizontal plane, have a field dependence similar to that previously measured. The lifting counter geometry of the diffractometer allowed us to measure the equivalent out-of-plane \((h, h, l)\) reflections plotted in the figure, which show different field dependencies. These allow several regions of interest to be distinguished. The low field region \( H < 0.1 \) T shows opposite variations of the \((220)\) and \((022)/(202)\) Bragg peaks, which are clearly assigned to the reorientation of the magnetic domains by the applied field. At higher fields, a single domain is stabilized, and the variations of the Bragg peaks arise from moment reorientations inside a tetrahedron. The \((002)\) and \((111)\) Bragg peaks show an extremum at 1.5 T, somehow below the critical field. The evolution of the magnetic peaks with the field is well accounted for by the magnetic refinements described below.

Magnetic refinements were performed with the program Fullprof. In zero field, the magnetic structure is a \( k=0 \) structure, which means that the 4 tetrahedra in the cubic unit cell have the same moment orientations. These orientations are those corresponding to the \( \psi_2 \) state, with moments in the easy planes along \( <211> \) axes. The magnetic moment at 0.3 K was refined as \( m = 3.25(20) \mu_B \) per Er ion. The domain populations were refined, yielding...
three equally populated magnetic domains (together with the opposite domains giving the same contribution), as also found by polarized neutron measurements.

Above 0.1 T, good refinements (with typical agreement factors $R_F=5\%$) were obtained by considering a single magnetic domain. The evolution of the magnetic structure with the field was determined by refining the integrated magnetic intensities, with moment values and angles as parameters. For the [110] field direction, the Er sites split into two sets, or chains: the $\alpha$ sites, with a ternary axis at an angle $\theta = \arcsin(1/\sqrt{3}) \simeq 35.3^\circ$ from $H$, and the $\beta$ sites with their ternary axis perpendicular to the field. In the $\psi_2$ state, the $\alpha$ sites split further into $\alpha_1$ and $\alpha_2$, with moments which are not symmetrical with respect to the field direction. We also split here the $\beta$ moments into $\beta_3$ and $\beta_4$ (see Fig. 2). In our experimental setup, there is a slight misorientation of the applied field with respect to the [110] direction: the polar angle $\theta$ of the field is $93.8^\circ$ (instead of $90^\circ$) and its azimuthal angle $\varphi$ is $45.8^\circ$ (instead of $45^\circ$). In the calculation described in the following section, we will neglect the small azimuthal misorientation and assume $\varphi = 45^\circ$, but we take into account the exact $\theta$ value. In the refinements, in order to reduce the number of fitting parameters, we assumed equal magnitudes for the two $\beta$ moments, which was a posteriori justified by the calculation. The preferred domain orientation (above a field of about 0.1 T) is such that the $\alpha$ moments lie in the upper hemisphere.

The moment orientations in a given tetrahedron are sketched for selected fields in Fig. 3. All the moments are seen to rotate towards the field direction (see c or d in Fig. 3) up to a field of about 2 T, which can be identified with the quantum critical field $H_c$ of Ref. 13. For this field value, all the moments are practically aligned along the field, so that $H_c$ can be considered as the spin-flip field of the $\mathrm{Er}_2\mathrm{Ti}_2\mathrm{O}_7$ magnetic structure for the [110] field direction. On further increase of $H$ beyond 2 T, the $\beta$ moments remain along the field whereas the $\alpha$ moments tend towards an asymptotic orientation, symmetric with respect to $H$.

The data points in Fig. 4 represent the evolution of the moment values and orientations at the 4 Er sites as the field increases. Pronounced anomalies are seen in the quantum critical region. As a major effect, all moment values show a minimum at $H_c=2$ T (Fig. 4a). This minimum is much more pronounced for the $\alpha$ moments, which get the closest to their hard local ternary axis for this field value. The $\alpha_1$ moment decreases down to $1 \mu_B$ whereas the $\alpha_2$ moment reaches $2 \mu_B$. As to the $\beta$ moments, which remain in their easy plane while rotating, they show a much shallower minimum of about $3 \mu_B$.

The evolution of the angles of the moments with the field is shown in Figs. 4c and 4d. At 2 T, all the angles are close to zero. The “flip” of the $\alpha_1$ moment is reflected by the change of sign of its angle with $H$, when it enters the lower hemisphere (see Fig. 4c above 2 T). By contrast, the $\alpha_2$ moment approaches the field direction at 2 T, then tilts away, always remaining in the upper hemisphere. Above 2 T, the two $\alpha$ moments tend towards an orientation at 20-25° on either side of the field. The $\beta$ moments show a much smoother field variation: they progressively reorient towards the field and remain aligned along $H$ from 2 T upwards. The angles of the moments with their local ternary axis (referred to as OZ in Fig. 4b) confirm that the $\beta$ moments always remain in their easy plane, while the $\alpha$ moments approach their hard axis in the critical region.

Our model calculation, to be described in the next sec-
FIG. 4. [Color on line] Variation with the field, applied along [110], of the moment magnitude (a) and orientation with respect to the local ternary axis (b) and to the field direction (c and d), for the 4 Er sites in Er$_2$Ti$_2$O$_7$ at 0.3 K. Our convention for the angle with the field direction is that it is negative if the moment lies in the lower hemisphere. Solid lines in (a)-(c) are self-consistent calculations with $\lambda_1 = -0.51$ T$/\mu_B$ and $\lambda_1 = -0.06$ T$/\mu_B$; Solid lines in (d) are calculations with $\lambda_X = -0.54$ T$/\mu_B$, $\lambda_Y = -0.48$ T$/\mu_B$ and $\lambda_\parallel = -0.06$ T$/\mu_B$; dashed lines are interpolated ones.

tion, accounts for most features of this evolution.

IV. MODEL CALCULATION

We implemented a model intended to reproduce the experimental data concerning both the thermal variation of the local magnetic susceptibility in the paramagnetic phase, measured in Ref.11 and the evolution with field of the magnetic structure at 0.3 K, measured in the present work. This model performs mean field self-consistent calculations and uses as ingredients the crystal field parameters of Er$^{3+}$ in Er$_2$Ti$_2$O$_7$ as in Ref.11 and anisotropic two-ion exchange of the type:

$$\mathcal{H}_{ex} = -J_\parallel S_{1\parallel}S_{2\parallel} - J_\perp(S_{1X}S_{2X} + S_{1Y}S_{2Y}),$$

where the sum runs over the 6 nearest neighbors. The relationship between the components of the $\lambda$ and $\tilde{J}$ tensors is:

$$\lambda_i = 6J_i \left( \frac{gJ_i - 1}{gJ_i} \right) \frac{1}{\mu_B^2}.$$  \hspace{1cm} (3)

A self-consistent treatment involving the 4 Er moments, each with its 3 components, is performed, the only parameters being thus the 2 components of the $\lambda$-tensor. The dipolar coupling is not included in the calculation.

We apply first this model to the paramagnetic susceptibility and to the zero-field AF phase. In the paramagnetic phase, the $\chi_\parallel(T)$ and $\chi_\perp(T)$ data measured in Ref.11 are very well reproduced using the above model with the axially symmetric AF molecular field tensor: $\lambda_\parallel = -0.06(3)$ T$/\mu_B$ and $\lambda_\perp = -0.51(4)$ T$/\mu_B$ (see Fig.5). These values are close to those obtained in Ref.11 using a simpler single sublattice model ($\lambda_\parallel = -0.15$, $\lambda_\perp = -0.45$ T$/\mu_B$). The planar anisotropy of the $\lambda$ tensor is strong ($\lambda_\perp/\lambda_\parallel \approx 10$) and reinforces that of the crystal field. In the AF phase in zero-field, the 4 Er moments in the $v_2$ ground state comply with the rule: $\sum_i m_i = 0$, whence, for instance, $m_2 + m_3 + m_4 = -m_1$. Therefore, the molecular field acting on ion $i$ is, according to Equ.(2): $H_{mol}^i = -\frac{1}{6} \lambda_\perp m_i$, since the spontaneous moments lie in the easy plane. According to
In the magnetic phase, the value of the critical or “spin-flip” field $\lambda$ depends on the spin-flip field in the presence of anisotropic structure with moments perpendicular to the field, the rotation of the Er moments as the field is increased. Re-match the data, but it captures the main trends of the data. In order to obtain a transition temperature of 1.2 K, one needs the value $\lambda_\perp = -0.435 \text{T}/\mu_B$, which is remarkably close to that derived in the paramagnetic phase. This coherence gives confidence in the applicability of the molecular field approximation in the magnetic phase. This coherence gives confidence in the applicability of the molecular field approximation in the paramagnetic and AF phases of Er$_2$Ti$_2$O$_7$. The associated transverse exchange integral is $J_\perp = -1.75 \text{K}$, and the $T = 0$ molecular field $H_{\text{mol}} = 0.5 \text{T}$. The zero-field spontaneous moment is $m_0 = 3.52 \mu_B$, close to the experimental value $3.25(20) \mu_B$, but somewhat higher than $3.00(05) \mu_B$ measured in Ref.6.

We then used the self-consistent exchange and crystal field model described above to calculate the evolution of the magnetic structure with increasing field. In a first step, we chose the two components of the axially symmetric $\lambda$ tensor as determined from the fit of the paramagnetic susceptibility ($\lambda_\perp = -0.51 \text{T}/\mu_B$ and $\lambda_\parallel = -0.06 \text{T}/\mu_B$). The results are displayed in Figs.4a, b and c as solid lines. The calculation does not exactly match the data, but it captures the main trends of the transition temperature of 1.2 K. In order to obtain a transition temperature of 1.2 K, one needs the value $\lambda_\perp = -0.435 \text{T}/\mu_B$, which is remarkably close to that derived in the paramagnetic phase. This coherence gives confidence in the applicability of the molecular field approximation in the magnetic phase.

In Fig.6, we show the evolutions of the four moments versus the magnetic field, calculated within this last model. Finally, the high field configuration is the following: the $\beta$ moments lie along the field and the $\alpha_1$ and $\alpha_2$ moments are on either side of the field, at an angle of about 20°. This can be understood since the [110] direction lies within the easy plane for the $\beta$ moments, whereas it is not a principal axis for the $\alpha$ moments.

It is worth emphasizing here that the data cannot be

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**FIG. 5.** [Color on line] Low temperature region of the thermal variation of the longitudinal and transverse susceptibilities in Er$_2$Ti$_2$O$_7$. The data are taken from Ref.11 and the lines are calculated using the self-consistent model described in the text.

**FIG. 6.** [Color on line] Evolution of the Er magnetic moments with the magnetic field at 0.3K, as calculated from the self-consistent calculation with with $\lambda_\alpha = -0.54 \text{T}/\mu_B$, $\lambda_\gamma = -0.48 \text{T}/\mu_B$ and $\lambda_\parallel = -0.06 \text{T}/\mu_B$. Arrows are proportional to the moment values. Each field step corresponds to 0.48 T.

The strong dependence of $H_{sf}$ on $\lambda_\perp$ since $g_\perp/g_\parallel > 1$ in Er$_2$Ti$_2$O$_7$. However, as seen in Fig.4c, there is a discrepancy concerning the rotation of the $\beta$ moments: the calculation yields different variations of the angle with the field for the two sites, whereas the data present identical variations within experimental uncertainties. This is due to a moment reorientation occurring in the calculation at a very small field (about 0.03 T): the moments jump from the $\psi_2$ state, with moments along the local OX axis of the $<211>$ family, to a configuration where each moment lies at about 38° from OX, which probably corresponds to the (small) potential well created by the crystal field interaction. Since this reorientation is not observed in the data, one has to devise a mechanism which would reinforce the potential well along the local OX axis, at least at low field. For this purpose, in a second step, we lifted the in-plane degeneracy of the molecular field tensor by about 5%, through the introduction a slightly higher (absolute) value for $\lambda_\alpha$ than for $\lambda_\gamma$. The result of the calculation, with $\lambda_\alpha = -0.54 \text{T}/\mu_B$ and $\lambda_\gamma = -0.48 \text{T}/\mu_B$, is shown for the angle $\theta_B = (m, H)$ in Fig.4d (the other calculated quantities remaining practically unchanged). The agreement is better for the $\beta$ moments, i.e. the $\psi_2$ configuration evolves smoothly with increasing field, to a field of 1.2 T. For $1.2 < H < 2 \text{T}$, the calculation presents an anomalous moment jump, still unexplained. Therefore, in Fig.4d, we do not consider the calculated curves in this field interval and we have drawn dashed lines interpolated between the lower and higher field regions where the model reproduces the data reasonably well. In Fig.6 we show the evolutions of the four moments versus the magnetic field, calculated within this last model.

Since its moment configuration is more complicated than a two-sublattice AF structure, this expression cannot be directly applied to Er$_2$Ti$_2$O$_7$, but it accounts for

\[
H_{sf} = \frac{1}{2} g_\parallel \mu_B |\lambda_\parallel + \left(\frac{g_\perp}{g_\parallel}\right)^2 \lambda_\perp|.
\]
reproduced with an isotropic molecular field constant \( \lambda = -0.51 \, T/\mu_B \). In this case, the calculation yields moments which remain in their easy plane up to the critical field of 2 T, and this field is not a spin-flip field since the \( \alpha \) moments do not align along its direction. The \( \alpha \) moment magnitude then shows no minimum at 2 T. Therefore, our calculation, within its limits of validity, demonstrates the existence of an anisotropic molecular field tensor in \( \text{Er}_2\text{Ti}_2\text{O}_7 \).

V. DISCUSSION

In this section, we show that the model described above also accounts relatively well for other experimental results related to the spin excitations in \( \text{Er}_2\text{Ti}_2\text{O}_7 \), as previously measured by inelastic neutron scattering and heat capacity. In Refs.\textsuperscript{4} and \textsuperscript{13}, a dispersionless mode was observed at about 0.4 meV for \( H=0 \), which further splits into higher energy modes when the field increases above the critical field. In first approximation, mostly valid in the middle of the Brillouin zone, such flat modes may be attributed to the splitting of the ground Kramers doublet under the coupled influence of the external and exchange fields. In this picture, the splitting into several modes induced by the field can be understood by the different molecular fields experienced by the \( \alpha \) and \( \beta \) moments. In Fig.\textsuperscript{7}, the excitation energies obtained by our model are compared with the energies of the flat modes determined in Ref.\textsuperscript{13} (square dots in Fig.\textsuperscript{7}). Below 2 T, the calculated energies are almost field independent around 0.25 meV, and above this threshold field, they increase linearly with slightly different slopes. The data match reasonably well the calculated values, except at zero and low field, where the energy of the non-dispersive mode (0.4 meV) is significantly above the calculation. One expects that our single mean field energy calculation holds at high field, where exchange is small with respect to the Zeeman energy, but fails at low field. Indeed, at low field, one should consider, for instance, the system of 4 exchange coupled Er moments on a tetrahedron and calculate the whole spin-wave spectrum, taking altogether crystal field, exchange and Zeeman interactions into account.

In Ref.\textsuperscript{13}, the variation with field of the heat capacity in \( \text{Er}_2\text{Ti}_2\text{O}_7 \) was attributed to a crossover towards a quantum high field paramagnetic state reminiscent of that observed in the quantum critical magnet \( \text{LiHoF}_4 \). We calculated the in-field heat capacity \( C_p(T) \) in \( \text{Er}_2\text{Ti}_2\text{O}_7 \) below 10 K, using the crystal field and Zeeman energies of all the levels (see inset in Fig.\textsuperscript{7}). Whereas this simple Schottky-type calculation cannot capture the anomaly at the phase transition for low fields, it reproduces correctly the main features of the high field heat capacity data of Ref.\textsuperscript{13}. Especially, the smearing of the temperature dependence of \( C_p(T) \) with increasing field can be correctly reproduced in this simple approach, which does not involve quantum fluctuations explicitly, besides the crystal field interaction.

Finally, we also applied our model to the calculation of the ordering temperature as a function of the applied field, i.e. to the \( (H,T_N) \) phase diagram. With the \( \lambda \)-tensor determined above \( (\lambda_x = -0.54, \lambda_y = -0.48 \) and \( \lambda_\parallel = -0.06 \, T/\mu_B) \), we calculated the thermal variation of the Er moments for different field values in the temperature range 0.2 - 2 K, in the field range below 1.3 T, where the model yields good agreement with experiment. As can be seen in Fig.\textsuperscript{8} which displays the thermal variation of the \( \alpha_1 \) moment magnitude, the ordering temperature, defined as the temperature where the moment variation
shows a jump, decreases with increasing field. This reflects the competition between the Zeeman and exchange energies, which leads to the quantum critical point when the Zeeman coupling overwhelms exchange and prevents a spontaneous moment configuration to set in. The low field part of the \((H, T_N)\) phase diagram is shown in the insert of Fig.8. Our calculation is in qualitative agreement with the diagram obtained from the heat capacity data of Ref.13. The curve \(H = f(T_N)\) is seen to follow a power law (see dashed line in the insert of Fig.8, with an exponent \(n = 2\). Extrapolation of this law to zero yields a critical field \(H_c = f(T_N = 0) = 1.85\,\text{T}\), close to the experimental value.

VI. CONCLUSION

Using neutron diffraction in the AF phase of the planar pyrochlore \(\text{Er}_2\text{Ti}_2\text{O}_7\) with a magnetic field applied along [110], we performed a quantitative study of the field evolution of the magnetic structure throughout the quantum critical point at \(H_c \approx 2\,\text{T}\). For this critical field value, the AF magnetic structure has “flipped”, i.e. all the Er moments are aligned along the field and their magnitude reaches a minimum. The strong decrease of the moment magnitude at \(H_c\) reflects the spin wave damping and enhanced quasi-elastic scattering previously observed in neutron experiments. A four sublattice self-consistent calculation, taking into account exchange, Zeeman and crystal electric field interactions, accounts for most of the characteristics of the field induced magnetic structure, from zero field up to well above \(H_c\). It also explains semi-quantitatively the field dependence of the heat capacity, of the dispersionless inelastic mode and of the transition temperature. The comparison between model and experiment brings out the strongly anisotropic exchange interaction as a necessary ingredient to explain all the features of the Quantum Critical Point. The critical field can then be defined as the field value for which the Zeeman energy overcomes the exchange energy, taking the anisotropic exchange tensor into account. The anisotropic exchange, already outlined by local susceptibility measurements in the paramagnetic phase, should also influence the spin-wave excitation spectrum in the AF ordered phase.

ACKNOWLEDGMENTS

We acknowledge very fruitful discussions with C. Lacroix (Institut Louis Néel, Grenoble) and J. Robert (LLB Saclay). Huibo Cao acknowledges supports from the Triangle de la Physique during his post-doctoral training.

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15 Actually, for an isolated Kramers doublet, if the field is at an angle \(\theta\) from the symmetry axis, then the angle \(\gamma\) of the magnetic moment with the axis is given by: \(\tan \gamma = (\frac{\lambda_{\perp}}{\lambda})^2 \tan \theta\). Inserting the zero-field g-values in this expression leads to an asymptotic angle of the moment with respect to \(\textbf{H}\) of \(\approx 40^\circ\), instead of the actual \(20^\circ\). This discrepancy arises from the mixture of the excited crystal field states into the ground state by the Zeeman effect, which modifies the g-values.
16 With the mean value \(\lambda_{\perp} = -0.51\,\text{T}\), the calculated \(T_N\) value in zero field is 1.48 K, rather than the experimental value 1.2 K. This probably reflects a small imperfection of the model, which requires a 15% higher \(\lambda_{\perp}\) value to account for the in-field properties than for the zero-field \(T_N\) value.