Spin relaxation in Mn$_{12}$-acetate

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Abstract. – We present a comprehensive theory of the magnetization relaxation in a Mn$_{12}$-acetate crystal based on thermally assisted spin tunneling induced by quartic anisotropy and weak transverse magnetic fields. The overall relaxation rate as function of the magnetic field is calculated and shown to agree well with data including all resonance peaks. The Lorentzian shape of the resonances is also in good agreement with recent data. A generalized master equation including resonances is derived and solved exactly. It is shown that many transition paths with comparable weight exist that contribute to the relaxation process. Previously unknown spin-phonon coupling constants are calculated explicitly.

The molecular magnet Mn$_{12}$-acetate—a spin 10 system with large easy-axis anisotropy—has attracted much recent interest[1, 2, 3] since several experiments on the magnetization relaxation have revealed pronounced peaks[4, 5, 6] in response to a varying magnetic field $H_z$ applied along the easy axis of the crystal. These peaks occur at field values where spin states become pairwise degenerate. Following earlier suggestions[7, 8], this phenomenon has been interpreted as a manifestation of resonant tunneling of the magnetization. However, in a critical comparison between model calculations[9, 10, 11, 12, 13, 14] and experimental data[4, 5, 15] Friedman et al.[15] point out that a consistent explanation of the relaxation is still missing. One of the main challenges for theory is to explain the overall shape of the relaxation curve as well as the Lorentzian shape of the measured resonance peaks[15].

In this work we shall present a model calculation of the magnetization relaxation which is based on thermally assisted tunneling. We find for the first time reasonably good agreement both with the overall relaxation rate (including the resonances) measured by Thomas et al.[3] (see fig.1) and with the Lorentzian shape of the first resonance peaks (see figs.2,3) measured by Friedman et al.[15] for four different temperatures.

The model introduced below contains five independent parameters: three anisotropy constants $A \gg B \gg B_4$, a misalignment angle $\theta$ (angle between field direction and easy axis), and sound velocity $c$. Previously unknown spin-phonon coupling constants are determined for the first time; we find that, quite remarkably, they can be expressed in terms of $A$ only.
The constants $A, B, B_4$ are known within some experimental uncertainty from independent measurements\[14, 17\], $\theta$ is typically about $1^\circ$\[13\], while $c$ is not measured yet (to our knowledge). We achieve optimal agreement between our theory and data as follows. In accordance with \[4\] we set $\theta = 1^\circ$, while the constants $A, B, B_4$ are fitted to the relaxation data by observing, however, the constraints that $A, B, B_4$ are allowed to vary only in the range of their experimental uncertainties. The sound velocity $c$, on the other hand, is a free fit parameter, for which we obtain $c = (1.45 - 2.0) \cdot 10^3$ m/s, which is within expected range. Thus, in contrast to previous calculations\[4, 10, 11, 12, 13, 18\] our theory is in reasonably good agreement not only with the relaxation data of refs.\[11, 12\] but with all experimental parameter values known so far. In addition, a new prediction is made (sound velocity $c$) which can be tested experimentally.

Extending previous work\[3, 4, 10, 11, 12, 13, 21\] we make use of a generalized master equation which treats phonon-induced spin transitions between nearest and next-nearest energy levels as well as resonant tunneling due to quartic anisotropies and transverse fields on the same footing. We solve the derived master equation by exact diagonalization. In addition, we identify for the first time the dominant transition paths (see fig.4) and show that the magnetization reversal is not dominated by just one single term but rather by several paths of comparable weight. Finally we note that our model is minimal in the sense that it is sufficient to explain the first time the dominant transition paths (see fig.4) and show that the magnetization reversal is not dominated by just one single term but rather by several paths of comparable weight. Finally we note that our model is minimal in the sense that it is sufficient to explain the.

\begin{equation}
\mathcal{H}_a = -AS_z^2 - BS_z^4
\end{equation}

describes the magnetic anisotropy with easy axis ($A > B > 0$) along the z-direction. $S$ is the spin operator with $s = 10$, and $A/k_B = 0.52 - 0.56$ K\[16, 17\], and $B/k_B = (1.1 - 1.3) \cdot 10^{-3}$ K\[14, 7\] are the anisotropy constants ($k_B$ is the Boltzmann constant). The Zeeman term through which the external magnetic field $H_z$ couples to the spin $S$ is given by $H_z = -g\mu_B H_z S_z$, while the tunneling between $S_z$-states is described by

\begin{equation}
\mathcal{H}_T = -\frac{1}{2} B_4 \left( S_+^4 + S_-^4 \right) - g\mu_B H_z S_x,
\end{equation}

where $H_x = |H| \sin \theta (\ll H_z)$ is the transverse field, with $\theta$ being the misalignment angle. Here, $B_4/k_B = (4.3 - 14.4) \cdot 10^{-5}$ K\[14\], and $g = 1.9$\[22\]. Finally, the most general spin-phonon coupling\[22\] is described by

\begin{equation}
\mathcal{H}_{sp} = \frac{g_1}{2} (\epsilon_{xx} - \epsilon_{yy}) \otimes \{S_x^2 - S_y^2\} + \frac{1}{2} g_2 \epsilon_{xy} \otimes \{S_x, S_y\} + \frac{1}{4} g_3 (\omega_{xz} \otimes \{S_x, S_z\} + \omega_{yz} \otimes \{S_y, S_z\}) + \frac{1}{4} g_4 (\omega_{xz} \otimes \{S_x, S_z\} + \omega_{yz} \otimes \{S_y, S_z\}),
\end{equation}

where $g_i$ are the spin-phonon coupling constants, and $\epsilon_{\alpha\beta}$ ($\omega_{\alpha\beta}$) is the (anti-)symmetric part of the strain tensor defined through the displacement $u$ as $(\partial u_\alpha / \partial x_\beta \pm \partial u_\beta / \partial x_\alpha)/2$. To determine $g_i$ occurring in \[4\] we follow ref.\[24\] (full details will be given elsewhere\[25\]). From the displacement resulting from rotation only, $u = \delta \phi \times x$ (in leading order), we obtain the infinitesimal rotation angle $\delta \phi = \frac{1}{2} \nabla \times u = (\omega_{yz}, \omega_{zx}, \omega_{xy})$. Rotating then the spin vector $S$ we find (to leading order in $\omega_{\alpha\beta}$) that the easy axis term, $-AS_z^2$, is transformed into $A(\omega_{xz} \{S_x, S_z\} + \omega_{yz} \{S_y, S_z\})$. Comparison with the last term in \[4\] then yields $g_4 = 2A$. Next,
expanding the rotation matrices and \( u \) up to second-order, we find\[^{25} \] \(-\delta \phi_{x}^{2} = \varepsilon_{yy} + \varepsilon_{zz} - \varepsilon_{xx} ,\) and cyclic. After rotating the rhs of \(-AS_{x}^{2} = -A(S_{x}^{2} - S_{y}^{2} - S_{z}^{2})\) we obtain a term of the form \( A(\varepsilon_{xx} - \varepsilon_{yy})(S_{x}^{2} - S_{y}^{2})\). Comparing this with \(^{4}\) we see that \( g_{1} = A\), and thus \( g_{1} = g_{4}/2 = A\). Finally, we note that the terms \( \propto g_{1,2} \) produce second-order transitions, while the ones \( \propto g_{3,4} \) produce first-order transitions. Thus, it is very plausible, in accordance with \(^{26}\), to adopt the approximations, \( |g_{2}| \approx g_{1} = A\), and \( |g_{3}| \approx g_{4} = 2A \) (the sign is irrelevant for the transition rates calculated below).

We denote by \(|m\rangle\), \(-s \leq m \leq s\), the eigenstate of the unperturbed Hamiltonian \( \mathcal{H}_{a} + \mathcal{H}_{Z} \) with eigenvalue \( \varepsilon_{m} = -Am^{2} - Bm^{4} - g_{\mu_{B}}H_{z}m\). If the external magnetic field \( H_{z} \) is increased one gets doubly degenerate spin states whenever a level \( m \) coincides with a level \( m' \) on the opposite side of the well (separated by the barrier \( \approx A \)). The resonance condition for double degeneracy, \( i.e. \varepsilon_{m} = \varepsilon_{m'}\), leads to the resonance field

\[
\gamma_{mm'} = -(m + m') [A + B (m^{2} + m'^{2})] .
\]

As usual, we refer to \( m + m' = \text{even (odd) as even (odd) resonances.} \)

**Master equation.** We describe the relaxation of the magnetization in terms of a master equation for the reduced density matrix \( \rho(t) \) which includes off-diagonal terms due to resonances. Using the notation \( \rho_{mm'} = \langle m | \rho | m' \rangle , \) \( \rho_{m} = \langle m | \rho | m \rangle , \) we start from the generalized master equation\[^{27}\]

\[
\dot{\rho}_{m'm} = \delta_{m'm} \sum_{m(\neq m)} W_{mnm} \rho_{n} - \gamma_{m'm} \rho_{m'm} ,
\]

where in the white-noise approximation \( \gamma_{m'm} = \gamma_{mm'} = (W_{m} + W_{m'})/2 , \) with \( W_{m} = \sum_{m(\neq m)} W_{nm} \).

The phonon induced transition rates \( W_{nm} \) are calculated below. Eq. \(^{4}\) holds for correlation times \( \tau_{c} \) much smaller than the spin relaxation time \( \tau \) — being satisfied here\[^{28}\]. Next we incorporate resonant tunneling into \(^{4}\). Let \( |m\rangle \) and \( |m'\rangle \) be two unperturbed eigenstates on the rhs and lhs of the barrier, resp., which are degenerate when \( \delta H_{z} = H_{z}^{mm'} - H_{z} \) vanishes, but get detuned otherwise. In the presence of tunneling, induced by \( \mathcal{H}_{T} , \) the two states form (anti-\) symmetric levels split by \( E_{mm'} \) (for \( \delta H_{z} = 0 \)).

\( E_{mm'} \) is obtained by standard means\[^{29}\]. The 2-state Hamiltonian including a bias field then reads

\[
\mathcal{H}_{T} = \xi_{m} |m\rangle \langle m| + \frac{E_{mm'}}{2} |m\rangle \langle m'| + (m \leftrightarrow m') ,
\]

with \( \xi_{m} = g_{\mu_{B}} \delta H_{z}m , \) \( \mathcal{H}_{T} \) provides a valid description as long as the level splitting \( \Delta = \sqrt{\xi_{m} - \xi_{m'}} \) remains smaller than \( |\varepsilon_{m} - \varepsilon_{m \pm 1}| , \) and \( |\varepsilon_{m'} - \varepsilon_{m' \pm 1}| \). Next we consider the quantum dynamics of the associated density matrix adopting standard arguments\[^{27}\].

From the von Neumann equation \( \dot{\rho} = i [\rho , \mathcal{H}_{T}] / \hbar \), we get \( \dot{\rho}_{m'} = iE_{mm'} \langle \rho_{mm'} - \rho_{m'm} \rangle / 2\hbar \).

\[
\dot{\rho}_{mm'} = \left( \frac{i}{\hbar} \xi_{mm'} + \gamma_{mm'} \right) \rho_{mm'} + \frac{iE_{mm'}}{2\hbar} (\rho_{m} - \rho_{m'}) ,
\]

where \( \xi_{mm'} = \xi_{m} - \xi_{m'} \), and likewise for \( m \leftrightarrow m' \). Here we have allowed for damping due to phonons \( i.e. \) finite life-time of the levels \( m, m' \) according to \(^{4}\). Inserting the stationary solution of \(^{4}\) into the \( \rho_{m} \)–equation, we get \( \dot{\rho}_{m} = \Gamma_{m} (\rho_{m'} - \rho_{m}) , \) where

\[
\Gamma_{m'} = E_{mm'}^{2} \frac{W_{m} + W_{m'}}{\xi_{mm'} + \hbar^{2} (W_{m} + W_{m'})^{2} / 4} ,
\]

is the transition rate from \( m \) to \( m' \) (induced by tunneling) in the presence of phonon-damping.

We note that \( \Gamma_{m'} \) has a Lorentzian shape with respect to the external magnetic field \( \delta H_{z} \) (the
$H_z$-dependence of $W_m$ around the resonances turns out to be much weaker); it is this $\Gamma_m^{m'}$ that will determine the peak shape of the magnetization resonances (see below and figs.1-3)\[30\].

To obtain the complete master equation we include also the transitions involving non-resonant levels and get

$$\dot{\rho}_m = -W_m \rho_m + \sum_{n \neq m, m'} W_{mn} \rho_n + \Gamma_m^{m'} (\rho_{m'} - \rho_m),$$

(9)

and the same with $m \leftrightarrow m'$ (see ref.\[33\] for details). A similar equation has been used in ref.\[10\], whose $\Gamma_m^{m'}$ has been obtained through a different and rather lengthy derivation \[31\]. For levels $k \neq m, m'$, eq. (1) reduces to $\dot{\rho}_k = -W_k \rho_k + \sum_n W_{kn} \rho_n$.

As in ref.\[12\] we evaluate the spin-phonon rates via Fermi golden rule (with thermal averaging over phonons) and find\[25\]

$$W_{m \pm 1, m} = \frac{A^2 s_{\pm 1}}{12\pi \rho \hbar^4} \frac{(\varepsilon_{m+1} - \varepsilon_m)^3}{e^{\beta(\varepsilon_{m+1} - \varepsilon_m)} - 1},$$

(10)

$$W_{m \pm 2, m} = \frac{17A^2 s_{\pm 2}}{192\pi \rho \hbar^4} \frac{(\varepsilon_{m+2} - \varepsilon_m)^3}{e^{\beta(\varepsilon_{m+2} - \varepsilon_m)} - 1},$$

(11)

where $s_{\pm 1} = (s \pm m)(s \pm m + 1)(2m + 1)^2$, and $s_{\pm 2} = (s \pm m)(s \pm m + 1)(s \pm m - 1)(s \pm m + 2)$. The mass density $\rho$ is given by $1.83 \times 10^3 \text{kg/m}^3$\[12\]. Here, $c$ is the sound velocity of the Mn-crystal, which is the only free parameter in our theory. It has not been considered before and we are not aware of any measurements of $c$.

Relaxation time. We solve now the master equation exactly to find the largest (i.e. dominant) relaxation time. To this end it is convenient to rewrite (1) as $\dot{\rho}(t) = \hat{W} \rho(t)$, where $\rho_n$ are the components of $\rho$. The rate matrix $\hat{W}$ has 21 eigenvalues $w_i$ found by exact diagonalization. The eigenvalues $w_i$ turn out to be non-degenerate with the smallest one being far separated from the remaining ones. The relaxation time is then obtained from $\tau = \max_i \{ -1/\text{Re } w_i \}$. The result is plotted in fig.1, where the overall relaxation rate $\tau$ is shown as function of $H_z$ and at $T = 1.9$K. We note that in our model the even resonances are induced by the quartic $B_4$-anisotropy, whereas the odd resonances are induced by product-combinations of $B_4 S_4^\perp$- and $H_z S_4^\perp$-terms. In accordance with the experimental uncertainty\[13\] we keep $-1^\circ \leq \theta \leq 1^\circ$ for all plots. Then the relaxation time $\tau$ at an even resonance peak is about 3 times smaller than the one at a subsequent odd resonance peak.

The relevant tunnel splitting energies of the even and odd resonances are about the same: $E_{4,-4} \approx E_{2,-3} \approx E_{3,-5} \approx E_{1,-4} \approx k_B \cdot 20$mK. ($E_{2,-2}/k_B \approx 0.5$K.) For comparison we also include in fig.1 the data reported by Thomas et al.\[3, 34\]. We have optimized the fit (as explained in the introduction) in such a way that the fits of the model parameters, given by $A/k_B = 0.54$K, $B/k_B = 1.1 \cdot 10^{-3}$K, and $B_4/k_B = 4.3 \cdot 10^{-5}$K, $\theta = 1^\circ$, are roughly within the reported experimental uncertainties\[16, 17\] (see above), while the fit of the sound velocity yields $c = 1.45 \cdot 10^3$ m/s. Almost identical plots are obtained for $0.5^\circ \lesssim \theta \lesssim 3^\circ$,\[25, 33\].

In figs.2,3 we plot the peaks of the first resonance at $H_z = 0$ (induced only by the $B_4$-anisotropy) for four different temperatures, $T = 2.5, 2.6, 2.7, 2.8$K. The peaks (like all others) are of single Lorentzian shape as a result of the 2-state transition rate $\Gamma_m^{m'}$ given in \[8\] (see also \[30\]). For comparison we plot in figs.2, 3 the data reported by Friedman et al.\[14\] for the same temperatures (no error bars, however, are given). The optimal fit values are: $A/k_B = 0.56$K, $B/k_B = 1.3 \cdot 10^{-3}$K, $B_4/k_B = 10.2 \cdot 10^{-5}$K, for all $-1^\circ \leq \theta \leq 1^\circ$, and $c = 2.0 \cdot 10^3$m/s. Note that these values are the same for all four temperatures. These fitting parameters turn out to be somewhat larger than in fig.1, which could be caused by differences e.g. in volume-to-surface ratio and/or in shape anisotropy, etc. Indeed, the sample of ref.\[15\] consists of many small
crystallites in contrast to the single crystal used in ref. [5]. In any case, the differences are small, and the sound velocity c seems to be within the expected order of magnitude. Clearly, it would be highly desirable to check this prediction by an independent measurement of c. To sum up, the agreement between theory and experiment is reasonably good, and our model and its evaluation contains the essential physics responsible for the magnetization relaxation.

Relaxation paths. It is instructive to determine the dominant transition paths via which the spin can relax into its ground state. For this we derive an approximate analytic expression for the relaxation time denoted by \( \tau^* \) (to distinguish it from the exact \( \tau \) obtained above). First, we solve the master equation for one particular transition path \( n \) which does not intersect with other paths. For \( H_z \geq 0 \) we find (details will be given elsewhere [25]):

\[
\tau_n = \frac{1}{1 + e^{\beta (\epsilon_f - \epsilon_i)}} \sum_{m} \frac{e^{\beta (\epsilon_m - \epsilon_f)}}{\gamma_m},
\]

where \( \gamma_m = W_{mm'} \) or \( \Gamma_{m'} \), depending on the particular path \( n \) characterized by the sum over the levels \( m \) (see fig.4). Eq. (12) holds for arbitrary initial \( (\epsilon_i) \) and final \( (\epsilon_f) \) energies; for \( H_z = 0 \) (i.e. \( \epsilon_i = \epsilon_f \)) eq. (12) reduces to the result found by Villain et al. [12]. If there is more than one dominant path (which is typically the case) we have to account for intersections at vertices. For this we associate with each path a probability current \( J_n = \rho_n \), and interpret eq. (12) as a serial circuit with the summands playing the role of “resistances”. This allows us then to set up flow diagrams for \( J_n \) (see fig.4), which obey the analog of Kirchhoff’s rules:

(K1) \( \sum_n J_n = 0 \): The sum over all incoming and outgoing currents vanishes at a vertex (current conservation). (K2) \( \sum_n J_n \tau_n = \Delta N \): The sum over all voltage drops \( (J_n \tau_n) \) is equal to the source-drain voltage \( \Delta N = \rho_s - \rho_{-s} \) for any closed path (probability conservation). The total probability current is given by \( J = \Delta N \). Fig.4 shows the complete (a) and its serially reduced (b) flow diagram for \( 0 \leq H_z \leq A/g \mu_B \). From (K1) we get \( J = J_1 + J_2, J_2 + J_5 = J_6, J_1 = J_3 + J_4, J_3 + J_6 = J_7, J_4 = J_5 + J_8, \) and \( J_7 + J_8 = J \), while from (K2) we get \( \Delta N = J_1 \tau_1 + J_2 \tau_2 + J_3 \tau_3 + J_7 \tau_7, J_1 \tau_3 = J_4 \tau_4 + J_5 \tau_5 + J_6 \tau_6, J_2 \tau_2 = J_1 \tau_1 + J_4 \tau_4 + J_5 \tau_5, J_6 \tau_6 = J_5 \tau_5 + J_6 \tau_6 + J_7 \tau_7, \) From these equations we obtain the relaxation time defined by \( \tau^* = \Delta N/J \) [23]. Finally, when plotted as function of \( H_z \) there is no visible difference between the exact \( \tau \) and this approximate \( \tau^* \), which confirms that the diagram in fig.4 contains the physically relevant relaxation paths close to the first resonance. Similar results are obtained for the other resonances [23].

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Additional Remark. After submission of this work, we were notified by J.R. Friedman that our prediction of the sound velocity \( c \) is in very good agreement with recent specific heat measurements [22] (see [25] for details).

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Fig. 1. – Full line: semilogarithmic plot of calculated relaxation time $\tau$ as function of magnetic field $H_z$ at $T = 1.9$K. The optimal fit values are: $A/k_B = 0.54$K, $B/k_B = 1.1 \cdot 10^{-3}$K, and $B_4/k_B = 4.3 \cdot 10^{-5}$K, $\theta = 1^\circ$, and $c = 1.45 \cdot 10^3$m/s. Dots and error bars: data[5].

Fig. 2. – Full line: plot of calculated relaxation rate $\Gamma = 1/\tau$ as function of $H_z$ for the first resonance peak at $T = 2.6$K. The Lorentzian shape is due to $\Gamma_m$ in (6). The optimal fit values are: $A/k_B = 0.56$K, $B/k_B = 1.3 \cdot 10^{-3}$K, $B_4/k_B = 10.2 \cdot 10^{-5}$K, $-1^\circ \leq \theta \leq 1^\circ$, and $c = 2.0 \cdot 10^3$m/s. Dots: data[5].
Fig. 3. – Full lines: semilogarithmic plots of calculated relaxation rate $\Gamma = 1/\tau$ as function of $H_z$ for the first resonance peak at (a) $T = 2.5\text{K}$, (b) $T = 2.6\text{K}$, (c) $T = 2.7\text{K}$, and (d) $T = 2.8\text{K}$. The optimal fit values are the same as in fig.2. Dots: data.$^{[13]}$

Fig. 4. – (a) Spin relaxation paths (from $s = 10$ to $s = -10$) for $0 \leq g\mu_B H_z \leq A$. Dashed lines: tunneling transitions due to $B_{3}$- and $H_x$-terms. (b) Associated serially reduced diagram with relaxation times $\tau_n$ given in $^{[12]}$. For $|H_z| \lesssim 0.05\text{T}$ only the path $\tau_1 \to \tau_3 \to \tau_7$ is relevant.