Non-adiabatic Landau-Zener transitions in low spin molecular magnet $V_{15}$

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1. Introduction

The $V_{15}$ polyoxovanadate molecule is made of 15 spins 1/2 with antiferromagnetic couplings. It belongs to the class of molecules with very large Hilbert space dimension ($2^{15}$ in $V_{15}$, $10^8$ in $Mn_{12}$-ac). It is a low spin/large molecule with spin $S = 1/2$. Contrary large spins/large molecules of the $Mn_{12}$-ac type, $V_{15}$ has no energy barrier against spin rotation. Magnetization measurements have been performed and despite the absence of a barrier, magnetic hysteresis is observed over a timescale of several seconds. This new phenomenon characterized by a “butterfly” hysteresis loop is due to the effect of the environment on the quantum rotation of the entangled 15 spins of the molecule, in which the phonon density of states is not at its equilibrium (phonon bottleneck).

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2. The $V_{15}$ magnetic moment evolution in low sweeping rates.

The $V_{15}$ complex of formula $K_6[V^{IV}_1A_{80}O_{42}(H_2O)] \cdot 8H_2O$ was prepared as described in [12–14]. It is made of a lattice of molecules with fifteen $V^{IV}$ ions of spin $S = 1/2$, placed in a quasi-spherical structured formed of a triangle, sandwiched by two hexagons (Fig. 1). All the interactions being antiferromagnetic, the resultant (collective spin) is $S = 1/2$. The symmetry is trigonal (space group $R3c$, $a = 14.029\,\AA$, $\alpha = 79.26^\circ$, $V = 2632\,\AA^3$). The unit-cell contains two $V_{15}$ clusters and it is large enough so that dipolar interactions between spin 1/2 molecules are negligible (few mK). Each hexagon contains three pairs of strongly coupled spins ($J \cong -800\,\text{K}$) and each spin at a corner of the inner triangle is coupled to two of those pairs (one belonging to the upper hexagon and one belonging to the lower hexagon) ($J' \cong J_1 \cong -150\,\text{K}$, $J'' \cong J_2 \cong -300\,\text{K}$ $\text{[3]}$). Thus, the $V_{15}$ molecule can be seen as formed by three groups of five $V^{IV}$ ions with resultant spin 1/2 and assembled on each corner of the inner triangle. These three spins 1/2 interact to each others through two main paths, one passing by the upper hexagon and one passing by the lower one. This is a typical example of frustrated molecular, where the exchange $J_0$ between the spins 1/2, is much smaller than the exchange interactions between two spins.

Two types of single crystal magnetization measurements have been performed: (i) characterization of the general thermodynamical properties of the system at equilibrium and (ii) study of the non-equilibrium to equilibrium transition.

In the first case a small dilution refrigerator allowing measurements above 0.1 K, was inserted in an extraction magnetometer providing fields up to 16 T, with low sweeping rates. Below 0.9 K we observed one jump in zero field and two others at $B_1 \cong \pm 2.8\,\text{T}$ (Fig. 2). They correspond to the $|1/2, -1/2| \leftrightarrow |1/2, 1/2|$ and $|1/2, 1/2| \leftrightarrow |3/2, 3/2|$ spin transitions, with respective saturations at $0.50 \pm 0.02\,\mu_B$ and $2.95 \pm 0.02\,\mu_B$ per $V_{15}$ molecule, in agreement with the level scheme given in Fig. 2 inset. For all temperatures the magnetization curves are reversible and do not show any anisotropy,
showing that eventual energy barrier preventing spin reversal must be quite small (less than 50 mK).

The magnetization curves measured at equilibrium (Fig. 2) are fitted using the Heisenberg Hamiltonian for three frustrated spins $S = 1/2$:

$$H = -\sum_{\alpha=XYZ} J_{\alpha}(S_{\alpha 1} S_{\alpha 2} + S_{\alpha 2} S_{\alpha 3} + S_{\alpha 3} S_{\alpha 1}) - g\mu_B B_0 (S_1 \cdot S_2 + S_2 \cdot S_3)$$

with $S_1 = S_2 = S_3 = 1/2$, $J_X = J_Y < 0$, $J_Z < 0$ (antiferromagnetic coupling). $g = 2$, $B_0$ the applied field and $\mu_B$ the Bohr magneton. In the isotropic case ($J_{X,Y,Z} = J_0$) the configuration $S = 3/2$ becomes favourable against the $S = 1/2$ one for $B_1 = -3 J_0/(2 g\mu_B)$. The measured value $B_1 \approx 2.8\, T$ becomes $-2.5\, K$. The comparison of the calculated curves ($S_{21} = \langle |S_{21}| + S_{22} + 2 S_{23} \rangle$ averaged over the different eigenstates $E_{\alpha i} |i\rangle$, $i = 1..8$) with the measured ones is quite good at any temperature. The width of the transition at $B_1 \approx \pm 2.8\, T$ is nevertheless broader in the experiments (Fig. 2). This broadening of about 0.7 T cannot be inferred to dipolar or hyperfine field distributions (about 1 mT and 40 mT respectively). The observed anisotropy of the electronic $g$-factor ($g_x = 1.95$ and $g_y = 1.98$) is also too small.

Antisymmetrical Dzyaloshinsky-Moriya interactions (see inset) allowed by symmetry and coupling the states $S = 1/2$ and $S = 3/2$, could explain this broadening.

In low fields, susceptibility measurements give the following effective paramagnetic moments: $\mu_{eff} = g\sqrt{S(S+1)} = 1.75 \pm 0.02\, \mu_B$ corresponding to $S = 1/2$ below 0.5 K, and $\mu_{eff} = 3 \pm 0.02\, \mu_B$ corresponding to 3 independent spins $S = 1/2$ above 100 K. It confirms clearly that the ground state of the molecule is $S = 1/2$ below 0.5 K.

3. Spin-phonon coupling in $V_{15}$ in low fields and high sweeping rates: a dissipative spin 1/2 two-level system.

We showed that, between -2.8 T and 2.8 T, the total spin of this molecule is $S = 1/2$. Such a small spin leads to vanishingly small energy barrier and relatively large splitting in zero-field ($\sim 10^{-2}$ K). Resonant spin-phonon transitions between the symmetric and antisymmetric states at resonance are then possible in sub-Kelvin experiments, contrary to the case of big molecules in zero field, where these states are much too close.

The sensitivity and time resolution of the micro-SQUID magnetometer allowed to study very small $V_{15}$ crystals in good contact with the thermal bath. Down to 50 mK non-equilibrium behavior was nevertheless observed at fast sweeping rate (up to $d B_0 / d t = 0.7\, T/s$). Few hysteresis loops are represented Fig. 3a and Fig. 4a (only the positive part, the other one being rigorously symmetrical). When the field increases, coming from negative values, the magnetization passes (i) in negative fields the system is colder than the bath, (ii) the spin system remains colder until the magnetization curve intersects the equilibrium curve, (iii) after the intercept the spin temperature overpass the bath temperature, (iv) at sufficiently high fields (about 0.5 T) the system reaches the equilibrium.

In order to interpret this magnetic behavior of the $V_{15}$ molecules, we will analyse how the level occupation numbers vary in this two level system (see Fig. 3a inset) when sweeping an external field. In the bare Landau-Zener (LZ) model [18,19] the probability for the $[1/2,-1/2] \leftrightarrow [1/2,1/2]$ transition is:

$$P = 1 - \exp \left( -\frac{\pi \Delta_0^2}{4 h \mu_B r} \right).$$

Taking the typical value $r = 0.1\, T/s$ and the zero-field splitting $\Delta_0 \approx 0.05\, K$, one gets a ground state switching probability very close to unity: the spin occupancy of the two level system is not modified when the field is reversed. In such an LZ transition, the plateaus of Fig. 3 should decrease if the sweeping rate increases, which is contrary to the experiments. Not surprisingly the $S = 1/2$ spin system is not isolated: LZ transitions are non-adiabatic due to spin couplings to the environment.

The mark of the $V_{15}$ system is that this coupling is acting also near zero-field because $h \omega \approx \Delta_0$ is of the order of the bath temperature. The spin temperature $T_S$ is such that $n_1/n_2 = \exp(\Delta_0/k_B T_S)$, where $\Delta_0$ is the two levels field-dependent separation, and $n_1,2(\{n_1,2 eq\})$ the out of equilibrium (equilibrium) level occupation numbers. In the magnetization curves at 0.1 K (Fig. 3a, inset), the spin temperature is significantly lower than the bath temperature $T$ ($n_1 > n_{1 eq}$, $T_S < T$) between $-0.3\, T$ (when the magnetization curve departs from the equilibrium one) and 0.15 T (the field at which the magnetization curve intersects the equilibrium one). After this intercept $T_S$ is larger than the bath temperature ($n_1 < n_{1 eq}$, $T_S > T$), and at sufficiently high fields (about 0.5 T) it reaches the equilibrium value ($n_1 = n_{1 eq}$, $T_S = T$). Note that the magnetization curves measured between $-0.7\, T$ and 0.7 T...
and 0.02 T at fast sweeping rates (0.07 and 0.14 T/s) are nearly the same, suggesting weak exchange with the bath, i.e. nearly adiabatic demagnetization.

Indeed, as we will show below, the silver sample holder is not sufficient to maintain the phonon temperature equal to the temperature of the bath. This is because in V15 below 0.5 K, the heat capacity of the phonons \( C_{ph} \) is very much smaller than that of the spins \( C_S \), so that the energy exchanged between spins and phonons will very rapidly adjust the phonons temperature \( T_{ph} \) to the spin one \( T_S \). Thus, we can see the spin system and the phonons as a single coupled system (quantum non-adiabatic LZ transitions) in weak exchange with the external bath (thermodynamical adiabatic demagnetization). The spins energy is transferred from the spins only to those phonons with \( h\omega = \Delta H \) (within the resonance line width). The number of such lattice modes being much smaller than the number of spins, energy transfer between the phonons and the sample holder must be very difficult, a phenomenon known as the phonon bottleneck [1]. Following [10], the number of phonons per molecule available for such resonant transitions is \( n_T = \int_{\Delta\omega} \sigma(\omega)d\omega/\exp(h\omega/kT) - 1 \), where \( \sigma(\omega)d\omega = 3V\omega^2d\omega/(2\pi^2v^3) \) is number of phonon modes between \( \omega \) and \( \omega + d\omega \) per molecule of volume \( V \), \( v \) is the phonon velocity and \( \Delta\omega \) is the transition linewidth due to fast hyperfine field fluctuations (they broaden both energy levels). Taking the typical values \( v \approx 3000 \text{ m/s}, T \approx 10^{-1} \text{ K} \) and \( \Delta\omega \approx 10^2 \text{ MHz} \) [2] we find \( n_T \) of the order of \( 10^{-6} \) to \( 10^{-8} \) phonons/molecule. Such a small number of phonons is very rapidly absorbed, burning a hole of width \( \Delta\omega \) in the spin-phonon density of states at the energy \( h\omega = \Delta H \) [11]. If this spin-phonon density of states does not equilibrate fast enough, the hole must persist and move with the sweeping field, leading to a phonon bottleneck.

4. Quantitative approach and numerical calculations.

Now this description will be made quantitative. For a given splitting \( \Delta H \), the time evolution of the two levels populations \( n_{1,2} \) and of the phonon numbers \( n_{T_{ph}} \) at \( T_{ph} \) obeys the set of two differential equations [10]:

\[
\begin{align*}
\dot{n}_1 &= \dot{n}_2 = P_{12}n_1 - P_{21}n_2 \\
\dot{n}_{T_{ph}} &= -(n_{T_{ph}} - n_T)/\tau_{T_{ph}} - P_{12}n_1 + P_{21}n_2, \\
\end{align*}
\]

(0.3)

where \( P_{12,21} \) are the transition probabilities between the two levels (they are themselves linear functions of \( n_{T_{ph}} \) and \( \tau_{T_{ph}} \approx L/2v \) is the phonon-bath relaxation time (\( L \) is the sample size). Using the notations \( x = (n_1 - n_2)/(n_{eq1} - n_{eq2}); y = (n_{T_{ph}} - n_T)/(n_T + n/2) \) with \( n = \int_{\Delta\omega} \sigma(\omega)d\omega \) we get:

\[
\begin{align*}
\dot{x} &= \left(1 - x - xy\right)/\tau_1 \\
\dot{y} &= -y/\tau_{T_{ph}} + b\dot{x},
\end{align*}
\]

(0.4)

where \( b = C_S/C_{ph} \) and \( 1/\tau_1 = P_{12} + P_{21} \) the direct spin-phonon relaxation time. By solving numerically this system for typical values, e.g. \( \tau_1 = 10^{-2} \text{ s}, \tau_{T_{ph}} < 10^{-6} \text{ s}, b > 10^5 \), we can see that \( T_{ph} \rightarrow T_S \neq T \) (phonon bottleneck) very rapidly, as expected. This leads to \( y = 1/x - 1 \) and the second equation of the differential system becomes \( \dot{x} = (x - x^2)/(1 + bx^2)/\tau_{T_{ph}} \). In the limit \( b >> 1 \) (in our case \( b \approx 10^8 - 10^{10} \)) this equation has the solution:

\[
-x/\tau_{T_{ph}} = x - x_0 + \ln((x - 1)/(x_0 - 1)),
\]

(0.5)

where \( x = x(t = 0) \) and \( \tau_{T_{ph}} \) is the spin-phonon relaxation time \( (T_{ph} = T_S \rightarrow T) \). When the system is not far from equilibrium \( (x \sim 1) \), we get an exponential decay of the magnetization, with the same time constant \( \tau_H = b\tau_{T_{ph}} \). For a spin 1/2 system [10]:

\[
\tau_H = \frac{\alpha\tanh^2(\Delta H/2k_BT)}{\Delta H},
\]

(0.6)

with \( \alpha = 2\pi^2h^2v^3N_{T_{ph}}/3\Delta\omega \) (\( N \) the molecule density).

The dynamical magnetization curves calculated in this model are given Fig. 3a and Fig. 4a. We started from equilibrium \( (x_0 = 1) \) in large negative fields. Then we let the system relax for a very short time \( \delta t \) and we calculated \( x(\delta t) \) using Eq. 0.5. This value was taken as the initial value for the next field (the field step is \( r\delta t \)). The parameters have been chosen to mimic the measured curves of Fig. 3a and Fig. 4a [23]. The obtained similarity supports the possibility of the phonon bottleneck effect at the timescale of a few 0.1 s. In the Fig. 3a inset, we show the variation of the calculated spin-phonon temperature \( T_S \) for \( T = 0.1 \text{ K} \) and \( r = 0.14 \text{ T/s} \). After a cooling in negative fields, we can note a linear variation in the plateau region (small positive fields), where \( n_1/n_2 \approx \text{cst.} \).

The slope of this quasi-adiabatic linear region gives the plateau position and varies with the bath temperature and sweeping rate. In the Fig. 3a inset we show the calculated field evolution of the number of phonons at energy \( h\omega = \Delta H \) at equilibrium \( (T_{ph} = T_S = T) \), dashed line) and out-of-equilibrium \( (n_{T_{ph}} = n_{T_{ph}} = T_S); r = 0.14 \text{ T/s} \), black line). The difference between the two curves (thick segment \( \Delta\omega \)) suggests the moving hole in the phonon distribution, while their intersection gives the plateau intercept of the equilibrium magnetization curve (above which the hole dissapears and \( T_{ph} = T_S \) becomes small enough).

Furthermore, we measured the relaxation of the magnetization of our crystal at different fields and temperatures, along the plateau region. The relaxation curves compared well to exponential decay and the obtained relaxation times are presented in Fig. 3a. The comparison
with those calculated (Fig. 5b) is acceptable. But we noted that a direct fit to Eq. 0.3 would necessitate larger values for $\alpha$ and $\Delta_0$ ($\approx 0.4 - 0.6$ sK$^2$ and $\approx 0.2 - 0.3$ K).

5. Conclusions

In conclusion, this dynamical study of a single crystal of non-interacting $V_{15}$ molecules with spin 1/2 is an example of a non-adiabatic Landau-Zener model with dissipation. Due to relatively large splitting $\Delta_0$ at the $S = 1/2$ anticrossing in $V_{15}$ molecules (i) the probability for ground-state adiabatic transitions is nearly equal to one and (ii) the spin system absorbs phonons during the Landau-Zener transition creating a hole in their distribution. The time and field evolution of this hole generates a “butterfly” hysteresis loop quite different from the one of high spin molecular magnets with large barrier and infinitesimal $\Delta_0/k_BT$ ratio (no phonons are available at the anticrossing). The effects presented in this paper seem to be a mark of molecules with low spin.

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[20] For an isolated spin 1/2, $\Delta_0 = 0$, but different couplings could generate a splitting, such as the hyperfine one $A\vec{I} \cdot \vec{S}, I = 7/2, S = 1/2, A \approx 10$ mK, $\Delta_0 \approx 10$ mK. Also, the molecule symmetry allows the Dzyaloshinsky-Moriya interactions that can generate $\Delta_0$ as large as 0.1 K. A value $\Delta_0 \approx 50$ mK is strongly supported by the experiment-to-model comparison in Figs. 3 and 4.
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$\alpha = 0.15$ sK$^2$ (2.9 $\cdot 10^{-47}$ sJ$^2$). $A \approx 10$ mK. Taking $L \sim 30 - 50 \mu m, N \sim (277 m^{-3}), \Delta_0 \sim 5 \cdot 10^8$ s$^{-1}$, one gets a phonon velocity $v \approx 2800 - 3600$ m/s which is a quite reasonable value.
FIG. 1. The geometry and the spin coupling scheme in V$_{15}$ complex. There are five antiferromagnetic exchange constants leading to a ground state $S = 1/2$ in fields below 2.8 T and $S = 3/2$ for greater fields (here such a field is pointing down).

FIG. 2. The agreement between the calculated curve at thermodynamical equilibrium (line) and the experimental magnetization data is quite good for an isotropic exchange $J_0 = -2.445$ K. For $J_x = J_y = -2.75$ K, $J_z = -2.2$ K and the field applied in the triangle plane (dashed line) one observes that the calculated second flip is slightly displaced but its slope is still greater than the experimental one. In inset are represented the energy levels vs. applied field in the effective triangle approach. When the applied field reaches $-3J_0/(2g\mu_B)$ the ground state switch from $S = 1/2$ to $S = 3/2$.

FIG. 3. Measured (a-top) and calculated (b-bottom) hysteresis loops for three temperatures and for a given field sweeping rate 0.14 T/s. The plateau is more pronounced at low T. The inset is a schematic representation of a two-level system $S_Z = \pm 1/2$ with repulsion due to non-diagonal matrix elements. In a swept field the switching probability $P$ is given by the Landau-Zener formulae (see text). The two levels are broadened by the hyperfine fields and the absorption or the emission of phonons can switch the polarization state of spins.
FIG. 4. Measured (a—top) and calculated (b—bottom) hysteresis loops for three field sweeping rates at $T = 0.1$ K. The observed plateau is more pronounced at high sweeping rate. The equilibrium curve can be approximated by the median of the two branches of the low sweeping rate hysteresis loop (dotted curve). In the top inset is plotted the spin and phonon temperature $T_S = T_{ph}$ for $T = 0.1$ K and $r = 0.14$ T/s, when the field is swept from negative values. $T_S$ decreases until zero-field and then increases linearly within the plateau region. Then it overpasses the bath temperature to finally reach the equilibrium. In the bottom inset the calculated number of phonons with $\hbar \omega = \Delta H$ is plotted vs. the sweeping field modulus (note the arrows) at equilibrium ($T_{ph} = T_S = T$, dashed line) and out-of-equilibrium ($n_{T_{ph}} = n_{T=T_S}$, $r = 0.14$ T/s, black line). The difference between the two curves (thick segment $\Delta \omega$) suggests the moving hole in the phonon distribution, while their intersection gives the plateau intercept of the equilibrium magnetization curve.

FIG. 5. The relaxation times $\tau_H$, measured (a—top) and calculated (b—bottom, same parameters as in Fig. 3b).