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High-field magnetization of V_{15} cluster at ultra-low temperatures: Importance of antisymmetric exchange and its precise estimation

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Abstract

The low temperature adiabatic magnetization of the nanoscopic V_{15} cluster exhibiting a triangular spin-frustrated V_3^{IV} array is analyzed within the model that includes isotropic exchange interactions and antisymmetric (AS) exchange. Along with the absolute value of AS exchange the orientation of the AS vector is shown to play an important physical role in the spin-frustrated systems. We were able to reach a perfect fit to the experimental data on the stepwise magnetization vs. field in the whole temperature range and for the first time to estimate precisely two components of the AS vector, namely, in-plane and the perpendicular parts.

1. Introduction

The cluster anion present in $K_6[V_{15}^{IV}As_6O_{42}(H_2O)]]\cdot 8H_2O$ (hereafter V_{15} cluster) was discovered more than 15 years ago [1] and since that time this system attracts continuous and increasing attention as a unique molecular magnet based on an unique structure [2–6]. The reason is that there are three layers with different magnetization containing six, three, and six ions, respectively. Whereas the coupling between the spins in the middle layer causing spin frustration is very weak the spins in the hexagons are coupled strongly. That has, e.g. been thoroughly discussed in a recently published important textbook about molecular magnetism [6]. Studies of the adiabatic magnetization and quantum dynamics of the V_{15} cluster with $S = 1/2$ ground state showed that this system exhibits the hysteresis loop of magnetization of molecular origin and can be referred to as a mesoscopic system [5–11]. As magnetic properties of the V_{15} cluster are inherently related to spin frustration as mentioned above the manifestations of the AS exchange interaction – introduced by Dzyaloshinsky [12] and Moria [13] – are especially important for detailed studies. The understanding of a special role of the AS exchange in spin-frustrated systems dates back to the 70s [14]. AS exchange was shown to result in a zero-field splitting of the frustrated ground state of the half-integer triangular spin systems, magnetic anisotropy and specific EPR spectra [14–26].

In this article we focus on the three-spin model of the V_{15} cluster that takes into account isotropic and AS exchange [27] for the study of the low lying spin excitations. The energy pattern of the system is analyzed and the approximate expressions for the energy levels are deduced. Afterwards, it is shown how different components of the AS exchange affect the dependence of magnetization of the V_{15} cluster at low temperatures. Finally, the experimental data [28] on the magnetization of V_{15} vs. field at ultra-low temperature are discussed while it is shown that the model provides a perfect fit to the experimental data. It has been concluded that AS exchange interaction is cru-

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cially important for understanding the peculiarities of the observed low temperature magnetization of the V_{15} cluster.

2. The model, AS exchange

The molecular cluster V_{15} has a distinctly layered structure within which fifteen V^{IV} ions ($S_i = 1/2$) are placed in an arrangement of a central triangle sandwiched by two hexagons [1,2]. The overall structure of V_{15} consists of two hexanuclear V_6^{IV} spin-paired (at low temperatures) systems with a frustrated V_3^{IV} triangle in between, resulting in an $S = 1/2$ ground state. The three-spin model proposed and substantiated in Refs. [3,4,6] gives accurate results for the low lying set of the levels that are populated at $T < 20$ K. The isotropic exchange can be described by the following Hamiltonian:

$$H_0 = -2J_0(\mathbf{S}_1\mathbf{S}_2 + \mathbf{S}_2\mathbf{S}_3 + \mathbf{S}_3\mathbf{S}_1) \tag{1}$$

with $S_i = 1/2$ and J_0 being the parameter of the antiferromagnetic exchange ($J_0 < 0$); we will use a positive parameter $J = -J_0$ and the spin functions will be labeled as $|S_1S_2(S_{12})S_3SM\rangle \equiv |(S_{12})SM\rangle$. The energy pattern $\varepsilon_0(S) = J[S(S+1) - 9/4]$ includes two degenerate spin doublets with $\varepsilon_0(1/2) = -(3/2)J$ and a spin quadruplet with $\varepsilon_0(3/2) = (3/2)J$.

The three vectors $\mathbf{D}_{ij}(ij = 12,23,31)$ of the AS exchange interaction associated with the sides ij can be expressed as $\mathbf{D}_{ij} = D_l\mathbf{x}_{ij} + D_t\mathbf{y}_{ij} + D_n\mathbf{z}_{ij}$ where $\mathbf{x}_{ij}$ and $\mathbf{y}_{ij}$ are the unit vectors along and perpendicular to the sides (in plane of the triangle) and $\mathbf{z}_{ij}$ is perpendicular to the plane [27]. The Hamiltonian of AS exchange that is invariant in trigonal symmetry can be represented as:

$$\begin{aligned} H_{AS} = {} & D_n([\mathbf{S}_1 \times \mathbf{S}_2]_Z + [\mathbf{S}_2 \times \mathbf{S}_3]_Z + [\mathbf{S}_3 \times \mathbf{S}_1]_Z) \\ & + D_l\left([\mathbf{S}_1 \times \mathbf{S}_2]_X - \frac{1}{2}[\mathbf{S}_2 \times \mathbf{S}_3]_X + \frac{\sqrt{3}}{2}[\mathbf{S}_2 \times \mathbf{S}_3]_Y \right. \\ & \left. - \frac{1}{2}[\mathbf{S}_3 \times \mathbf{S}_1]_X - \frac{\sqrt{3}}{2}[\mathbf{S}_3 \times \mathbf{S}_1]_Y\right) \\ & + D_t\left([\mathbf{S}_1 \times \mathbf{S}_2]_Y - \frac{\sqrt{3}}{2}[\mathbf{S}_2 \times \mathbf{S}_3]_X - \frac{1}{2}[\mathbf{S}_2 \times \mathbf{S}_3]_Y \right. \\ & \left. + \frac{\sqrt{3}}{2}[\mathbf{S}_3 \times \mathbf{S}_1]_X - \frac{1}{2}[\mathbf{S}_3 \times \mathbf{S}_1]_Y\right). \end{aligned} \tag{2}$$

Here, the parameter D_n is associated with the 'normal' component of AS exchange, D_l and D_t are those for the 'in-plane' contributions see Fig. 1.

The AS exchange acts within the $(S_{12})S = (0)1/2, (1)1/2$ manifold and gives rise to two Kramers doublets [14,15]. The matrix of the full Hamiltonian $H_0 + H_{AS}$ including also the Zeeman term can be calculated with the aid of the irreducible tensor operator technique [29–31]. It should be noted that the normal part of the AS exchange operates only within four $(S_{12})S = (0)1/2, (1)1/2$ states while the in-plane part of AS exchange leads only to a mixing of $S = 1/2$ and $S = 3/2$ levels.

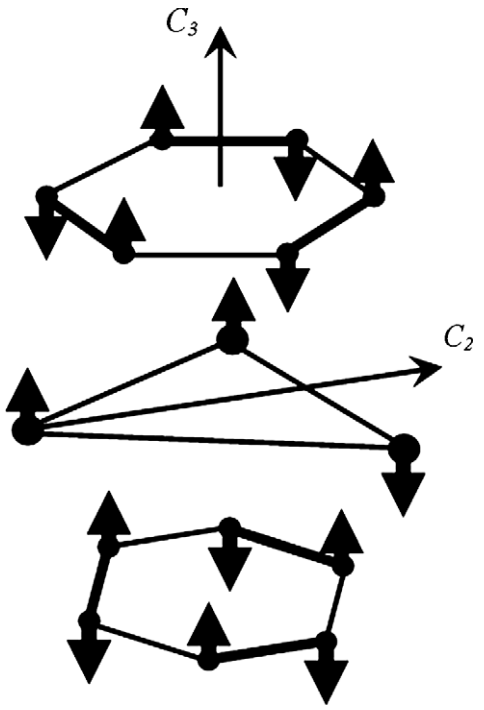

Fig. 1. Scheme of the V_{15} metal network and spin arrangement in the low energy region.

3. Zeeman levels in a perpendicular field

In absence of the field and in the parallel field ($\mathbf{H} \| C_3$ axis) the system possesses actually axial symmetry that allows to find the analytical expressions for the energy levels [27]. The low temperature single crystal magnetization experiments [28] are performed in the perpendicular ($\mathbf{H} \perp C_3$) field so that the axial symmetry is broken and the numerical analysis is required. Nevertheless, in the important particular case when only normal contribution of AS exchange is taken into account ($D_n \neq 0, D_l = D_t = 0$) the exact solution can be found:

$$\begin{aligned} \varepsilon_1 = \varepsilon_3 &= -\frac{3}{2}J - \frac{1}{2}\sqrt{(g\beta H)^2 + 3D_n^2}, \\ \varepsilon_2 = \varepsilon_4 &= -\frac{3}{2}J + \frac{1}{2}\sqrt{(g\beta H)^2 + 3D_n^2}, \\ \varepsilon_{5,6} &= \frac{3}{2}J \mp \frac{1}{2}g\beta H, \quad \varepsilon_{7,8} = \frac{3}{2}J \mp \frac{3}{2}g\beta H, \end{aligned} \tag{3}$$

where H is the field in any direction in the plane, say $H = H_X$ and correspondingly g-factor $g \equiv g_\perp$. The levels $\varepsilon_i(H)$ with $i = 1, 2, 3, 4$ are related to $S = 1/2$ while $i = 5, 6, 7, 8$ are the numbers of Zeeman sublevels for $S = 3/2$ (with $M = \mp 1/2$ and $M = \mp 1/2$) as shown in Fig. 2a in the case of the isotropic model. The energy pattern for the case $D_n \neq 0$, $D_l = D_t = 0$ is shown in Fig. 2b. Three peculiarities of the energy pattern that are closely related to the magnetic behavior should be mentioned: (1) the

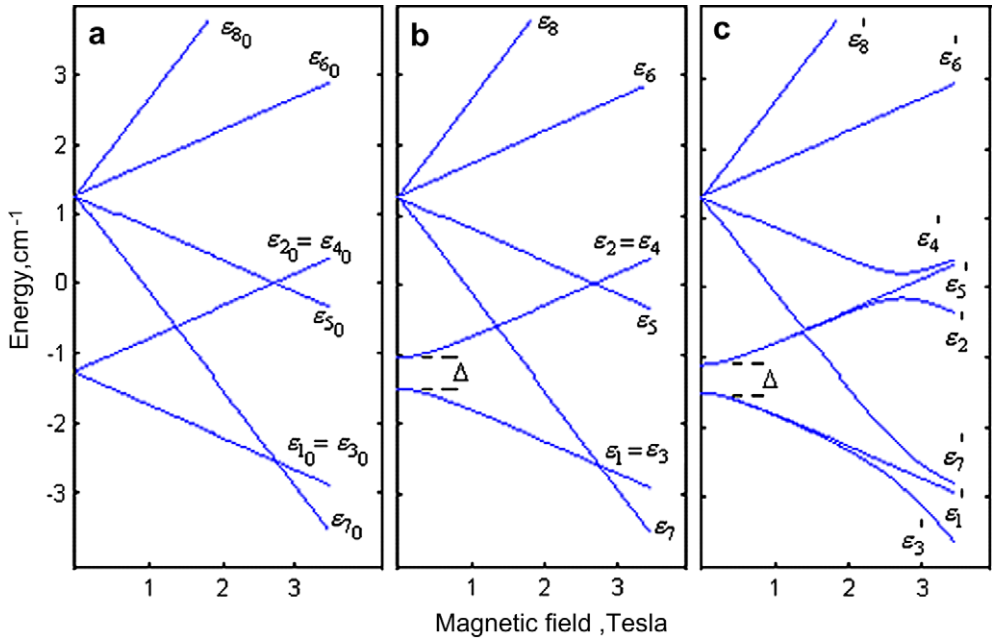

Fig. 2. Energy pattern of the triangular vanadium unit in the magnetic field applied in the plane ($\mathbf{H}\perp C_3$), $J = 0.847\ \mathrm{cm}^{-1}$, $g = 2$: (a) $D_n = 0$, $D_\perp = 0$ (the energies are denoted as ε_{i0}), (b) $D_n = 0.3J$, $D_\perp = 0$, and (c) $D_n = 0.3J$, $D_\perp = 0.6J$.

ground state involving two $S = 1/2$ levels shows zero-field splitting into two Kramers doublets separated by the gap $\Delta = \sqrt{3}D_n$; (2) at low fields $g\beta H \leqslant \Delta$ the Zeeman energies are double degenerate and show quadratic dependence on the field:

$$\begin{aligned}\varepsilon_1 = \varepsilon_3 &= -\sqrt{3}D_n/2 - (g\beta H)^2/4\sqrt{3}D_n,\\ \varepsilon_2 = \varepsilon_4 &= -\sqrt{3}D_n/2 + (g\beta H)^2/4\sqrt{3}D_n.\end{aligned} \tag{4}$$

This behavior is drastically different from that in the isotropic model and from the linear magnetic dependence in a parallel field [27] and can be considered as a breaking of the normal AS exchange by the perpendicular field (the effect mentioned in [14], see also references therein); (3) the magnetic sublevels arising from $S = 3/2$ ($M = -1/2$ and $M = -3/2$) cross the sublevels belonging to $S = 1/2$ spin levels, no avoided crossing points are observed. At high field the levels $\varepsilon_{1,3}$ and $\varepsilon_{2,4}$ exhibit again linear magnetic dependence:

$$\begin{aligned}\varepsilon_1 = \varepsilon_3 &= -\frac{3}{2}J - \frac{3D_n^2}{g\beta H} - \frac{1}{2}g\beta H,\\ \varepsilon_2 = \varepsilon_4 &= -\frac{3}{2}J + \frac{3D_n^2}{g\beta H} + \frac{1}{2}g\beta H.\end{aligned} \tag{5}$$

One can see that a strong perpendicular field restores linear Zeeman splitting but without zero-field splitting so that the perpendicular field reduces the normal part of AS coupling.

When the AS exchange in its general form is involved ($D_n \neq 0, D_l \neq 0, D_t \neq 0$) the energy pattern shows new peculiarities (Fig. 2c). The low field part of the spectrum is not affected by the in-plane part of AS exchange and is very close to that in Fig. 2b due to the fact that the in-plane part of AS exchange is not operative within the ground manifold and the effect of $S = 1/2 - S = 3/2$ mixing is small at low fields due to the large gap $3J \gg |D_\perp|$. At the same time in the vicinity of the crossing points the effect of the normal AS exchange is negligible but the in-plane AS exchange acts as a first order perturbation giving rise to the avoided crossings as shown in Fig. 2c.

In order to obtain three closely spaced low lying levels in the region of anticrossing field we will use the perturbation theory respectively to the in-plane part of AS exchange and the basis formed by three eigen-functions of H_0 whose eigen-values have crossing points at $g\beta H = 3J$, namely $|(0)1/2,-1/2\rangle$, $|(1)1/2,-1/2\rangle$, $|(1)3/2,-3/2\rangle$ (Fig. 2a). One can find the following expressions for the energy levels $\varepsilon_i'(H)$ in this region of the field:

$$\varepsilon_1' = -\frac{3}{2}J - \frac{1}{2}g\beta H, \tag{6}$$

$$\varepsilon_3' = -g\beta H - \frac{1}{8}\sqrt{(4g\beta H - 12J)^2 + 18D_\perp^2}, \tag{7}$$

$$\varepsilon_7' = -g\beta H + \frac{1}{8}\sqrt{(4g\beta H - 12J)^2 + 18D_\perp^2}, \tag{8}$$

where the notation $D_\perp^2 = D_t^2 + D_l^2$ is introduced.

4. Temperature and field dependence of magnetization-effects of AS exchange

Let us consider some general features of the field dependence of magnetization related to the AS exchange by plotting the results of sample calculations. The most spectacular aspect is the low temperature limit for which one can find the following expressions for magnetization

(per molecule) that work well at low field (Eq. (9)) and high field in the vicinity of anticrossing of the low lying levels (Eq. (10)).

$$\mu(H) = \frac{g^2\beta^2 H}{2\sqrt{3D_n^2 + g^2\beta^2 H^2}}, \tag{9}$$

$$\mu(H) = g\beta + \frac{2g\beta(g\beta H - 3J)}{\sqrt{2(g\beta H - 3J)^2 + 18D_\perp^2}}. \tag{10}$$

Due to the effect of the reduction of the Zeeman interaction at low field by the normal AS exchange the magnetization in this region of the field proves to be induced by the field as follows from Eq. (9). In Fig. 3 we show the field dependence of magnetization at $T = 0$ within the triangular model.

The parameter $J = 0.847\ \mathrm{cm}^{-1}$ [28] is used; to make the illustration more clear we increased the parameters of the AS exchange.

Fig. 3a shows the field dependence of magnetization calculated in the framework of the isotropic model when magnetization exhibits two sharp non-broadened steps, one at zero field and the second one at the field $H = 3J/g\beta$. This is just the field at which the level with $S = 3/2$, $M = -3/2$ crosses the degenerate pair of levels $S = 1/2$, $M = -1/2$ ($S_{12} = 0$ and $S_{12} = 1$) so that the $S = 3/2$ spin state becomes favorable against $S = 1/2$, this can be clearly seen in Fig. 3a. In Fig. 3b the effect of the normal part of the AS exchange is illustrated. As one can see the normal part of AS exchange results in the broadening of the low field step in $\mu(H)$ meanwhile the high-field step remains non-broadened. The broadening of the first step is closely related to the magnetic anisotropy of the AS exchange that gives rise to a quadratic Zeeman effect in the low perpendicular field (see Fig. 2b). It can be said that the normal part of AS exchange reduces perpendicular magnetization at low field and allows only second order magnetic splitting and Van Vleck type paramagnetism [32]. Since the magnetization is induced by the field and does depend on the field even at $T = 0$ the jump of $\mu(H)$ acquires a peculiar shape (Fig. 3b). At the same time the normal AS exchange leaves the exact crossing of $S = 3/2$, $M = -3/2$ level with the lowest component of $S = 1/2$ that results in absence of the broadening of the high-field step (Fig. 3b). Finally when both normal and in-plane parts of the AS exchange are taken into account we obtain broadening of both steps (Fig. 3c). Now the anticrossing in the region $H = 3J/g\beta$ appears due to coupling of $S = 1/2$, $M = -1/2$ $S = 3/2$, $M = -3/2$ levels through in-plane AS exchange that obviously gives rise to a smoothed switch from $S = 1/2$ to $S = 3/2$ as shown in Fig. 3c.

5. Temperature and field dependence of magnetization of V_{15} cluster

The low-temperature adiabatic magnetization vs. field applied in the plane of the V_3 triangle ($\mathbf{H} \perp C_3$) exhibits steps whose broadening and shapes are temperature dependent (Fig. 4). Analysis of the experimental data in [28] has been performed in the framework of an isotropic model supplemented by quadrupolar anisotropy ($J_{XX} = J_{YY} \neq J_{ZZ}$). Agreement between the calculated curves and experimental magnetization data proved to be quite good for $T = 0.9$ K and 4.2 K but the model fails to explain the data at ultra-low temperature. In this view the AS exchange has been mentioned in [28] as a possible origin of the broadening of the steps in the magnetization curve.

Modeling of the magnetization curves so far performed shows that the low temperature behavior of the magnetization vs. field is just most sensitive to the AS exchange. In order to avoid artificial fitting we adopt the following initial set of parameters: $J = -0.847\ \mathrm{cm}^{-1}$, $g = 1.952$, and $D_\perp = 0.22\ \mathrm{cm}^{-1}$. This set of parameters gives the best fit to the experimental data at very low temperature (0.1 K)

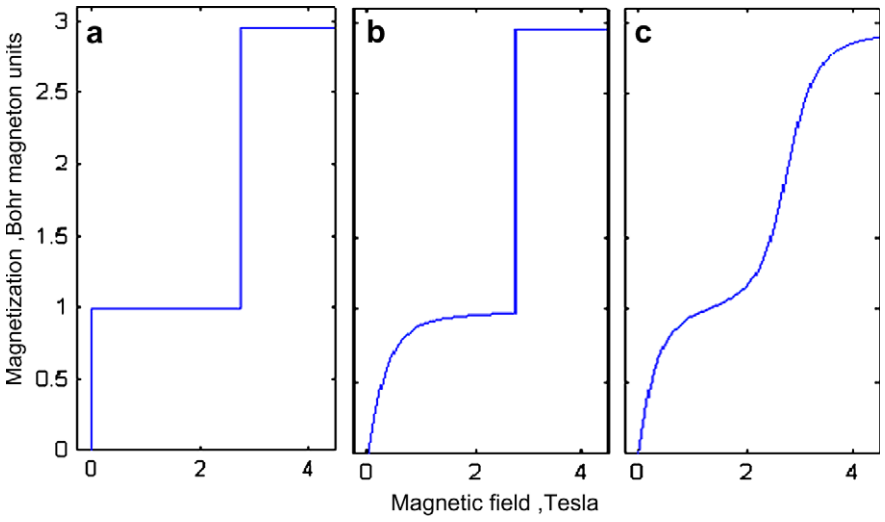

Fig. 3. Magnetization at $T = 0$ K in the perpendicular field for the cases: (a) $J = 0.847\ \mathrm{cm}^{-1}$, $D_n = 0$, $D_\perp = 0$; (b) $J = 0.847\ \mathrm{cm}^{-1}$, $D_n = 0.3J$, $D_\perp = 0$; and (c) $J = 0.847\ \mathrm{cm}^{-1}$, $D_n = 0.3J$, $D_\perp = 0.6J$.

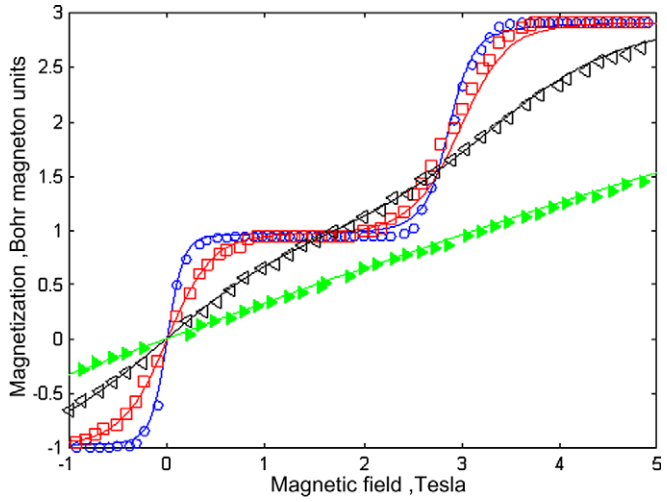

Fig. 4. Experimental data (from Ref. [25]) and theoretical curves of static magnetization calculated taking into account the isotropic and AS exchange interactions ($\mathbf{H}\perp C_3$). Experimental data: circles, $T = 0.1$ K, squares, $T = 0.3$ K, unfilled triangles, $T = 0.9$ K, filled triangles, $T = 4.2$ K. Solid lines, calculated curves with the best fit of the parameters (see text).

in the shape of the high-field step at $H \approx 2.8$ Tesla corresponding to $S = 3/2 - S = 1/2$ crossing region. In order to preserve the zero-field splitting of spin doublets $\Delta = 0.14\ \mathrm{cm}^{-1}$ [33,34] we have used the expression for this value containing second order correction $\Delta = \sqrt{3}D_n - D_\perp^2/8J$ (see [27]). Taking this into account we can get a reasonable initial value of $D_n = 0.0851\ \mathrm{cm}^{-1}$. The best fit procedure gives the following set of parameters: $J = -0.855\ \mathrm{cm}^{-1}$, $g = 1.94$, $D_\perp = 0.238\ \mathrm{cm}^{-1}$, $D_n = 0.054\ \mathrm{cm}^{-1}$. The in-plane parameter $D_\perp$ brings a dominating contribution to the overall AS exchange. This parameter is just responsible for the behavior of the levels in the anticrossing region $H \approx 2.8$ Tesla. Fig. 4 shows that the model that includes AS exchange interaction gives a perfect fit of field dependence of magnetization in the whole range of fields for all temperatures including extremely low temperature.

The least mean square error (MSE) (in μ_B^2) calculated for the whole sets of the $\mu(H)$ curves is found to be 2.3×10^{-3}. The data at 4.2 K are only slightly affected by AS exchange so this fit is almost insensitive to the model. In fact, the best fit for 4.2 K in the isotropic model (at $J = 0.86\ \mathrm{cm}^{-1}$, $g = 1.945$) can be obtained with MSE $= 8.38 \times 10^{-4}$ which does not exceed significantly the corresponding value (6.97×10^{-4}) in the isotropic model. The best fit parameters J and g are very close in both models, which demonstrates low sensitivity of the theoretical curves to the AS exchange at 4.2 K. On the contrary, the data at 0.1 K can not be fitted within the isotropic model (that has been indicated in [28]), but they are fitted perfectly with the significant reduction of MSE from 6.3×10^{-3} to 1.7×10^{-3} when AS exchange is taken into account.

6. Conclusion

The experimental data of the low temperature adiabatic magnetization of the nanoscopic low spin cluster anion V_{15} present in $K_6[V_{15}^{IV}As_6O_{42}(H_2O)] \cdot 8H_2O$ are analyzed as a function of applied field and temperature within the three-spin model that includes isotropic exchange interaction and AS exchange. It was shown that the AS exchange plays a crucial role in the understanding of the field and temperature dependence of the adiabatic magnetization of the V_{15} single crystals. Furthermore, it was demonstrated that the orientation of the AS exchange vector plays a special physical role in the magnetic behavior of spin-frustrated systems but not only its absolute value. In fact, the normal part of the AS exchange affects the low field part of the magnetization but also the in-plane components gives rise to a peculiar shape of magnetization vs. field in the vicinity of the crossing point of the magnetic sublevels. The influence of the AS exchange is most pronounced at the low temperature ($T = 0.1$ K). We could reach a perfect fit to the experimental data on the adiabatic magnetization vs. applied field in the whole temperature range including ultra-low temperature and for the first time to precisely estimate two components of the AS vector coupling constant. Furthermore, we can conclude that we understand more about frustrated systems integrated in complex systems. Therefore, we intend to extend this to other complex systems, e.g. where frustrated systems are interacting like in $\{Mo_{57}\}\{V_3\}_2$ [35].

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