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Spectral statistics and the Dzyaloshinsky–Moriya interaction of nanomagnet V_{15}

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Abstract

The spectral statistics of nanomagnet V_{15} is studied. We study the dependence of the Dzyaloshinsky–Moriya interaction in V_{15} on the spectral statistics.

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Nanoscale molecular magnets, or nanomagnets, have been studied intensively in recent years. The novel structure of discrete energy levels of nanomagnets stimulates not only fundamental research such as quantum dynamics but also applied research of quantum device. Indeed, a quantum computation by nanomagnets with the help of the ESR technique is proposed [1].

Nanomagnet V_{15} is the complex of formula $K_6[V^{IV}_{15}As_6O_{42}(H_2O)] \cdot 8H_2O$ [2,3]. In the cluster of the complex, 15 vanadium ions, each of which has a $\frac{1}{2}$ spin, are placed almost on a sphere. The three spins forming a triangle are sandwiched by 12 spins forming 2 hexagons.

The existence of the Dzyaloshinsky–Moriya (DM) interaction [4–6] in the Hamiltonian of V_{15} is implied by the following experiments. If we sweep the magnetic field adiabatically at low temperatures, the magnetization changes smoothly at zero field from $-\frac{1}{2}$ to $\frac{1}{2}$ [7]. This fact implies that there exists a finite gap between the ground state and the lowest excited state at zero field. The origin of this gap is attributed to the DM interaction [8]. In addition, the ground state magnetization changes smoothly from $\frac{1}{2}$ to $\frac{3}{2}$ at the field around 2.8 T [9]. This broadness of the change is also considered to be caused by the DM interaction [10]. However, the DM vectors of the DM interaction are not fully understood yet. In

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this paper, we study the DM interaction of V_{15} from the viewpoint of the spectral statistics.

We consider V_{15} in the magnetic field applied parallel to the c-axis of the molecule. The total Hamiltonian $\mathscr{H}$ is given by

$$\mathscr{H} = \mathscr{H}_0 - H_S \sum_i S_i^z$$
$$\mathscr{H}_0 = -\sum_{\langle i,j\rangle} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \mathscr{H}_{DM}$$
$$\mathscr{H}_{DM} = \sum_{\langle i,j\rangle} \boldsymbol{D}_{ij} \cdot [\boldsymbol{S}_i \times \boldsymbol{S}_j]. \tag{1}$$

We adopt the same notation as Ref. [12] for numbering the sites in the cluster, spins, and DM vectors. The interactions J_{ij} in $\mathscr{H}_0$ take three different values J, J_1, and J_2 ($|J|>|J_2|>|J_1|$) [11,12]. Here we take $J = -800\,\mathrm{K}$, $J_2 = -350\,\mathrm{K}$, and $J_1 = -225\,\mathrm{K}$ [10]. We also take $H_S = 4\,\mathrm{T}$. We note that the Hamiltonian $\mathscr{H}$ has the C_3 symmetry.

Let us determine the DM vectors in the DM interaction $\mathscr{H}_{DM}$. Since the system has C_3 symmetry, $\mathscr{H}_{DM}$ is invariant with the unitary transformation

$$U = T_L e^{-i\frac{2\pi}{3}S^z_{tot}}, \quad S^z_{tot} = \sum_i S_i^z, \tag{2}$$

where T_L is an operator acting on the spatial degrees of freedom and it rotates the position of spin $\boldsymbol{S}_i$ by $2\pi/3$. Let us assume that position 5 is different from position 1 by $2\pi/3$ rotation in the x–y plane. Then, $\boldsymbol{S}_1$ is transformed by U as

$$US_1^x U^{-1} = S_5^x \cos\frac{2\pi}{3} + S_5^y \sin\frac{2\pi}{3},$$
$$US_1^y U^{-1} = -S_5^x \sin\frac{2\pi}{3} + S_5^y \cos\frac{2\pi}{3},$$
$$US_1^z U^{-1} = S_5^z. \tag{3}$$

The DM vectors are considered to exist in the bonds with interaction J on the upper and lower hexagons. That is, we consider $\boldsymbol{D}_{1,2}$, $\boldsymbol{D}_{3,4}$, and $\boldsymbol{D}_{5,6}$ on the upper hexagon and $\boldsymbol{D}_{10,11}$, $\boldsymbol{D}_{12,13}$, and $\boldsymbol{D}_{14,15}$ on the lower hexagon.

In order to satisfy $[\mathscr{H}_{DM}, U] = 0$, the DM vectors on the upper hexagon are obtained by rotating the reference DM vector $\boldsymbol{D}_{1,2}$ by $2\pi/3$ and $4\pi/3$. For example, $\boldsymbol{D}_{3,4}$ is given as

$$\begin{pmatrix} D^x_{3,4} \\ D^y_{3,4} \end{pmatrix} = \begin{pmatrix} \cos\frac{2\pi}{3} & \sin\frac{2\pi}{3} \\ -\sin\frac{2\pi}{3} & \cos\frac{2\pi}{3} \end{pmatrix} \begin{pmatrix} D^x_{1,2} \\ D^y_{1,2} \end{pmatrix},$$
$$D^z_{3,4} = D^z_{1,2}. \tag{4}$$

Since the V_{15} cluster itself has the D_3 symmetry [2], $\boldsymbol{D}_{14,15}$ on the lower hexagon is obtained by rotating $\boldsymbol{D}_{1,2}$ by $-\pi/6$ in the x–y plane and changing the sign of $D^z_{1,2}$. The other two DM vectors on the lower hexagon are obtained by rotating $\boldsymbol{D}_{14,15}$ by $2\pi/3$ and $4\pi/3$.

We obtain the states $|\phi\theta\rangle$ which are eigenstates of U by applying the projection operator $\hat{P}_\theta$ on the basis vectors $|\phi\rangle$ [13].

$$|\phi\theta\rangle = \frac{1}{\|\hat{P}_\theta|\phi\rangle\|}\hat{P}_\theta|\phi\rangle, \tag{5}$$

where the projection operator is defined by

$$\hat{P}_\theta = \sum_{n=0,\pm1} \exp\left[-i\frac{2\pi}{3}\left(kn + S^z_{tot}\right)\right] T^n_L,$$
$$\theta = k + S^z_{tot}, \quad k = 0, \pm1. \tag{6}$$

We classify subspaces of the total Hilbert space by θ. The dimension of the total Hilbert space is $2^{15} = 32\,768$. For example, the space of $\theta = \frac{3}{2}$ contains 4821 levels.

We study the spectral statistics of V_{15} in the subspace of $\theta = \frac{3}{2}$. We use from the 1500th to 2500th energy levels. We consider the following five cases of the reference DM vector $\boldsymbol{D}_{1,2}$:

(i) Only $D^x_{1,2}$ and $D^y_{1,2}$ exist ($D^x_{1,2} = D^y_{1,2} = 40.0\,\mathrm{K}$, $D^z_{1,2} = 0$).
(ii) Only $D^z_{1,2}$ exists ($D^x_{1,2} = D^y_{1,2} = 0$, $D^z_{1,2} = 40\,\mathrm{K}$).
(iii) All of $D^x_{1,2}$, $D^y_{1,2}$, and $D^z_{1,2}$ exist ($D^x_{1,2} = D^y_{1,2} = D^z_{1,2} = 40\,\mathrm{K}$).
(iv) Same as (iii) but the magnitude is half ($D^x_{1,2} = D^y_{1,2} = D^z_{1,2} = 20\,\mathrm{K}$).
(v) Same as (iii) but the magnitude is twice larger ($D^x_{1,2} = D^y_{1,2} = D^z_{1,2} = 80\,\mathrm{K}$).

We obtain the energy levels E_i by diagonalizing the Hamiltonian in the subspace. The normalized

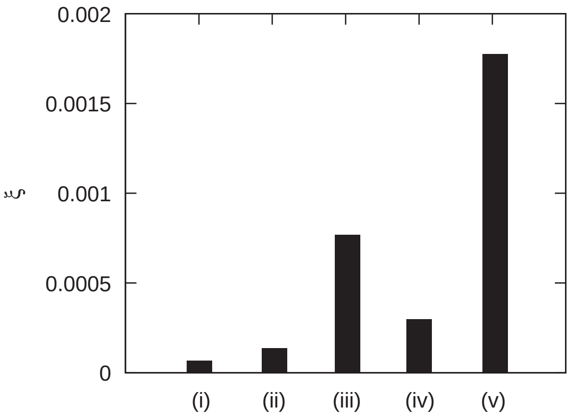

Fig. 1. The values of ξ are shown with respect to the five cases.

nearest-neighbor spacing S_i is given by

$$S_i = \rho\left(\frac{E_{i+1} - E_i}{2}\right)(E_{i+1} - E_i), \tag{7}$$

where $\rho(E)$ is the density of states. Let us denote by $P(S)\,\mathrm{d}S$ the probability that S_i is in the range between S and $S + \mathrm{d}S$. Note that $\int_0^\infty P(S)\,\mathrm{d}S = 1$ and $\int_0^\infty SP(S)\,\mathrm{d}S = 1$. The cumulative spacing distribution $I(S)$ is written as

$$I(S) = \int_0^S P(S')\,\mathrm{d}S'. \tag{8}$$

If there is no correlation between energy levels, the cumulative spacing distribution is given by [14]

$$I_0(S) = 1 - \mathrm{e}^{-S}. \tag{9}$$

If there exists the effect of level repulsion, $I(S)$ will deviate from $I_0(S)$. In order to study this deviation quantitatively, we introduce a parameter ξ.

$$\xi \equiv \int_0^1 [I(S) - I_0(S)]^2\,\mathrm{d}S. \tag{10}$$

The parameter ξ measures *distance* between $I(S)$ and $I_0(S)$. Fig. 1 shows ξ for each case. We conclude that ξ reflects the DM interaction in the Hamiltonian, and the five cases align according to

$$\text{(i)} < \text{(ii)} < \text{(iv)} < \text{(iii)} < \text{(v)} \tag{11}$$

in the order of the strength of level repulsion.

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