Toroidal moment in the molecular magnet V_{15}

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Quantum-mechanical calculations predict the existence of toroidal spin structure in the molecular magnet V_{15}. It is shown that nonzero toroidal moment arises from symmetry violation of the exchange interactions between spins of the base triangle of the V_{15} molecule due to the Jahn-Teller effect. It is established that the value of the toroidal moment is connected with value of total spin projection of V_{15} on z axis. It enables to induce the toroidal moment by external magnetic field and (or) to induce the magnetic moment by variable electric field or by current.

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Triangular antiferromagnets offer interesting quantum systems, the spin states of which can be controlled and manipulated by external fields.[1–3] This opens perspectives for the development of quantum devices for information handling. Recently it was shown (see Ref. 4) that electric polarization of a Cu_3 antiferromagnet could induce a spin-electric effect under certain conditions: a relativistic Hamiltonian is used, containing Dzyaloshinskii-Moriya (DM) spin-exchange, spin-chirality, and spin-orbit interactions. In addition a low-symmetry external electric field, e.g., provided by an approaching scanning tunnel microscope tip, is required in this Brief Report to induce asymmetry. However as we will demonstrate here, isotropic exchange coupling in a spin triangle will give rise to spontaneous spin-electric effects, due to symmetry breaking. In the present case the asymmetry is an intrinsic feature of the magnetoelastic interaction. The carrier of this spin-electric effect is an intrinsic toroidal magnetic moment, as our subsequent quantum-mechanical analysis will reveal. The resulting quantum structure provides ways to control and manipulate the qubit state for use in quantum computation.[5,6] In particular, the existence of a toroidal moment provides the interaction between spin and external current. It opens an important possibility for the control of the qubit state.[4] On the more fundamental level the treatment clearly shows the presence of a parity nonconservation effect in a molecule.

At the present time the search for the toroidal moment is on in chemistry and condensed-matter physics, particularly in multiferroics.[7] In works[8,9] it has been experimentally found that spin structure of some magnets with antiferromagnetic interaction has a toroidal type of symmetry. Recently toroics with a toroidal domain structure were discovered and investigated.[10] The toroidal moment changes sign upon both time and spatial reversal. This symmetry condition for the appearance of toroidal moment in molecules can be achieved by application of crossed electric and magnetic fields.[11,12] But intrinsic mechanisms of its spontaneous appearance at zero field are not known. In work[13] it has been proposed that toroidal moment can exist in some mesoscopic molecules. But these ideas were not realized. Recently an interesting hypothesis for the construction of toroidal moment in magnetic nanoclusters has been put forward.[14] The required specific magnetic anisotropy is realized in triangular Dy_3 complexes[2,3] but the question on its toroidal moment demands further study. In present work the molecular magnet V_{15} is considered. According to the quantum-mechanical analysis (see below) it may exhibit a toroidal moment. The latter is caused by the Jahn-Teller (JT) activity of the frustrated ground state.

Molecular magnet $K_6[V_{15}^{IV}As_6O_{42}H_2O]\cdot 8H_2O$ (hereafter V_{15}) contains 15 ions V^{4+} with $S=1/2$ (Ref. 15) which are arranged in the apices of two hexagons and the triangle layered between them. Thus we are dealing with three magnetic subsystems which are formed by vanadium ions. The scheme of exchange interaction between V^{4+} ions is shown in Fig. 1. The spin Hamiltonian of the molecular magnet (see, e.g., Ref. 15) has D_3 symmetry. All exchange interactions between vanadium ions are antiferromagnetic leading to the total spin $S_\Sigma=1/2$ in the ground state. Nearly all contributions to the total spin are due to the spins arranged in the apices of triangle. The contribution of ions arranged in the apices of hexagons is rather small.[16] It is ~2.8% of the triangle contribution.

The exchange interaction between spins of triangle can be described by the Hamiltonian

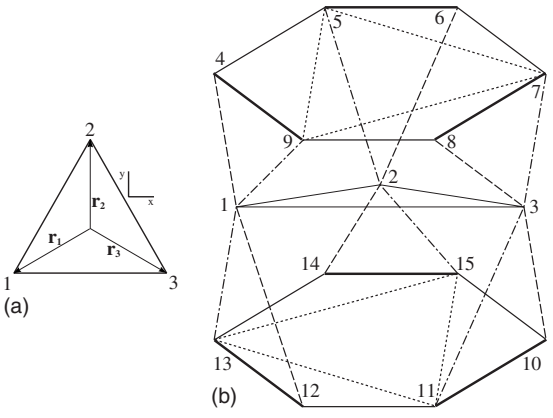

FIG. 1. The structure of exchange interactions in the molecular magnet V_{15}.

$$\hat{\mathcal{H}}_t = J_e(\hat{\mathbf{S}}_1\hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_1\hat{\mathbf{S}}_3 + \hat{\mathbf{S}}_2\hat{\mathbf{S}}_3), \quad (1)$$

where $J_e \sim 2.9$ K (Ref. 17) is the effective exchange parameter arising from the projection of spin Hamiltonian onto the subspace of spin states $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$. The energy spectrum of triangle subsystem, which is determined by Hamiltonian (1), consists of two degenerate Kramers doublets with energy $E=-3/4J_e$ and overlaying quartet with energy $E=3/4J_e$ (see Ref. 15). The four eigenfunctions of the ground state can be written as

$$\chi_1(m) = \frac{1}{\sqrt{2}}[\tau_1(m)\tau_2(-m) - \tau_1(-m)\tau_2(m)]\tau_3(m),$$

$$\chi_2(m) = \frac{1}{\sqrt{6}}[2\tau_1(m)\tau_2(m)\tau_3(-m) - \tau_1(m)\tau_2(-m)\tau_3(m) - \tau_1(-m)\tau_2(m)\tau_3(m)], \quad (2)$$

where the magnetic quantum number of the molecule $m = \pm 1/2$, $\tau(m_i)$ is the spin function of the ith ion ($m_i = \pm 1/2$). The wave functions, Eq. (2), assume the "(12)+3" spin coupling scheme.

There have been suggested two mechanisms that may be operative for removing the degeneracy of the ground state. The first one is concerned with the Jahn-Teller concept[17,18] and the second one is the result of antisymmetric exchange interaction between ions of triangle.[19] In the present case the first mechanism is operative. The inelastic neutron-scattering study clearly demonstrates that the low-energy properties of V_{15} are accurately described by a triangle model with distortion.[20] The DM interaction model is incompatible with these experimental results.[20] The measured by NMR values of spin moment $-0.6\mu_B$, $-0.6\mu_B$, and $0.33\mu_B$ (see Ref. 21) are close to the values given by JT model $-0.67\mu_B$, $-0.67\mu_B$, $0.33\mu_B$ and contradict the DM interaction model calculation $-0.33\mu_B$, $-0.33\mu_B$, and $-0.33\mu_B$.

Naturally the exchange integrals between the ions of the triangle depend on the mutual arrangement of vanadium ions in the molecular magnet V_{15}. In linear approximation with respect to the normal modes of displacements Q_x and Q_y (see Ref. 17) of vanadium ions

$$\hat{\mathcal{H}}_{me} = V_0(Q_x\hat{\sigma}_x + Q_y\hat{\sigma}_y), \quad (3)$$

where $\hat{\sigma}_{x,y}$ are Pauli matrices. V_0 is a reduced matrix element of the magnetoelastic operator, which is proportional to space derivatives of exchange integral, i.e., $V_0 \sim dJ_e/dQ$.[17] The diagonalization of Eq. (3) yields the following eigenvalues and eigenvectors

$$E_q = q|V_0|\rho, \quad \rho = \sqrt{Q_x^2 + Q_y^2}, \quad e^{i\alpha} = (Q_x + iQ_y)/\rho,$$

$$\varphi_q(m) = \frac{1}{2}(1 + qe^{i\alpha})\chi_1(m) + \frac{i}{2}(1 - qe^{i\alpha})\chi_2(m), \quad (4)$$

where $q = \pm 1$. At this level of approximation, taking into account the elastic energy of vanadium ion displacements, the nuclear motion is localized near the bottom of the trough at radius $\rho = V_0/k$, where k is the coefficient of elastic "rigidity" of triangle. In addition the motion of the nuclei is concerted such that they form an isosceles triangle. Its axis of symmetry is rotated by an arbitrary angle α around original triangle. The motion of the nuclei becomes more complex if we take into account the terms of the second order with respect to the normal modes of the displacements. In this case three minima of potential appear. These minima are defined by the angle $\alpha = \pi n/3$, where $n=1,3,5$ or $n=(0,2,4)$ (see Ref. 17 for details). Periodically the lag of deformations occurs near these minima. The peculiarities of distortion due to the vibronic interactions in the spin clusters can appear in the electromagnetic properties of the molecular crystal.

Under the JT symmetry breaking the spins of triangle are directed in such a way as to form a circular magnetic moment. The toroidal moment of vanadium ions placed at the apices of triangle is defined by the expression[22,23] $\hat{\mathbf{T}} = \frac{1}{2}g\mu_B\Sigma\mathbf{r}_i \times \hat{\mathbf{S}}_i$, where $g=2$ is the Lande g factor, $\mathbf{r}_i$ is the radius vector which connects the center of the undeformed triangle with apex (see the Fig. 1). Let us define the normalized toroidal moment

$$\hat{\mathbf{t}} = \frac{2\hat{\mathbf{T}}}{gl\mu_B} = \sum \mathbf{e}_i \times \hat{\mathbf{S}}_i, \quad (5)$$

where l is the distance between the center of triangle and its apex, $\mathbf{e}_i = \mathbf{r}_i/l$ are the unit vectors. Let us direct the x axis from 1 ion to 3 ion and the y axis is perpendicular to the x axis in the plane of the triangle.

In the Hilbert space based on the ground-state functions $\varphi_{1,2} = \varphi_{\mp 1}(\mp\frac{1}{2})$, $\varphi_{3,4} = \varphi_{\mp 1}(\pm\frac{1}{2})$, where $\varphi_q(m)$ ($q = \pm 1$, $m = \pm 1/2$) are defined in Eq. (4), the operators of the toroidal moment components are the 4×4 matrices, which are given by the formulas

$$\hat{t}_z(\alpha) = \frac{1}{2}\cos\tilde{\alpha}\begin{pmatrix} -\hat{\sigma}_x & \hat{\sigma}_z \\ \hat{\sigma}_z & \hat{\sigma}_x \end{pmatrix} + \frac{1}{2}\sin\tilde{\alpha}\begin{pmatrix} \hat{\sigma}_y & i\hat{I} \\ -i\hat{I} & -\hat{\sigma}_y \end{pmatrix},$$

$$\hat{t}_x(\alpha) = -\frac{1}{2}(\hat{\gamma}_1 \sin\tilde{\alpha} + \hat{\gamma}_4 \cos\tilde{\alpha}),$$

$$\hat{t}_y(\alpha) = \hat{t}_x\left(\alpha - \frac{\pi}{2}\right), \quad (6)$$

where $\tilde{\alpha} = \alpha + \pi/3$, $\gamma_1 = i\begin{pmatrix} 0 & -\hat{\sigma}_x \\ \hat{\sigma}_x & 0 \end{pmatrix}$, $\gamma_4 = \begin{pmatrix} \hat{I} & 0 \\ 0 & -\hat{I} \end{pmatrix}$ are the Hermitian matrices of Dirac and $\hat{I}$ is the unit matrix 2×2.

The ground state of the system as well as the excited one is degenerate in the magnetic quantum number m. The diagonal matrix elements of the toroidal moment change sign at $m \to -m$. A magnetic field directed along the z axis removes the degeneracy and induces a toroidal moment. In this case the corresponding energy levels are given by $E_{qm}(H) = E_q + mg\mu_B H$, where E_q are defined by Eq. (4). The scheme of the level splitting is shown in Fig. 2.

The quantum numbers $q=-1$, $m=-1/2$ correspond to the ground state. The toroidal moment lies in the xy plane and its components are given by $t_x^0 = -\frac{1}{2}\cos(\alpha + \frac{\pi}{3})$, $t_y^0 = -\frac{1}{2}\sin(\alpha + \frac{\pi}{3})$. The mean values of the toroidal moment in the three other states, which arise from the splitting of ground

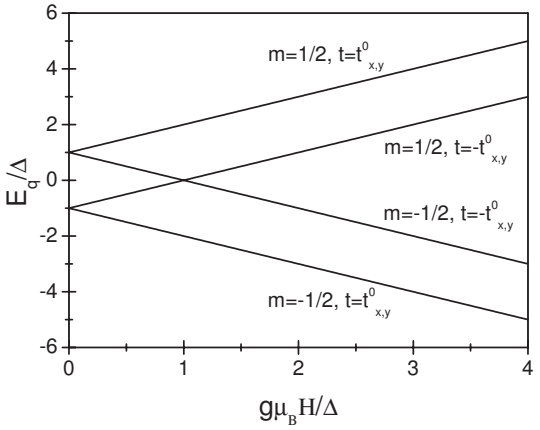

FIG. 2. The level splitting of the V_{15} ground state in the external magnetic field directed parallel to the z axis.

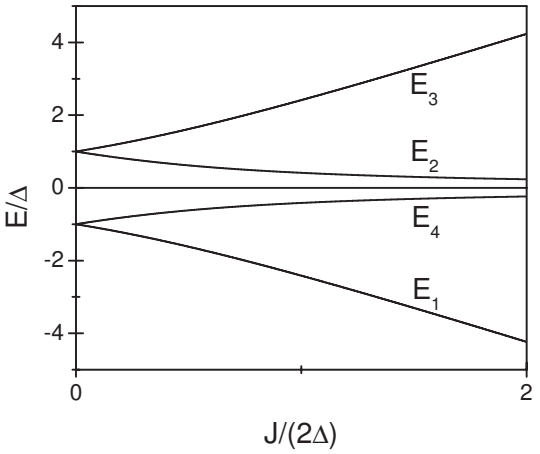

FIG. 3. The level splitting of the V_{15} ground state due to the interaction of the toroidal moment with the current applied along the z axis.

doublets in magnetic field, are defined by Eq. (6). The above quartet ($S=3/2$) is separated from ground doublets by energy $\delta E=3/2J_e$ (at $H=0$). The toroidal moment is zero in each of these states.

At low temperature ($kT \ll \delta E$) the averaged components of toroidal moment $\langle \hat{t}_i \rangle = \mathrm{Tr}[\hat{t}_i \exp(-\hat{\mathcal{H}}/kT)]/\mathrm{Tr}\exp(-\hat{\mathcal{H}}/kT)$ are given by

$$\frac{\langle \hat{t}_{x,y} \rangle}{t^0_{x,y}} = M(H,T)\tanh\left(\frac{\Delta}{kT}\right), \tag{7}$$

where $\Delta=\rho|V_0|$ is the half width of the energy gap between the ground doublet and exited doublet, $M(H,T)=\tanh(\frac{\mu_B H}{kT})$ is the magnetic moment of the molecular magnet normalized to μ_B. The expression (7) gives the dependence of the toroidal moment on the external magnetic field, temperature, and energy gap. Note that $\langle t_{x,y} \rangle$ tends to zero at $\Delta \to 0$.

Let us consider the case when magnetic field lies in plane of triangle. Then the interaction of vanadium ions with external field is given by Hamiltonian $\hat{\mathcal{H}}_z = g\mu_B \Sigma_i (H_x \hat{S}_{ix} + H_y \hat{S}_{iy})$. The eigenfunctions are given by

$$\psi_{1,2} = \frac{1}{\sqrt{2}}\left[\exp\left(\frac{i\gamma}{2}\right)\varphi_{-1}\left(-\frac{1}{2}\right) \mp \exp\left(-\frac{i\gamma}{2}\right)\varphi_{-1}\left(\frac{1}{2}\right)\right],$$

$$\psi_{3,4} = \frac{1}{\sqrt{2}}\left[\exp\left(\frac{i\gamma}{2}\right)\varphi_{1}\left(-\frac{1}{2}\right) \mp \exp\left(-\frac{i\gamma}{2}\right)\varphi_{1}\left(\frac{1}{2}\right)\right], \tag{8}$$

where $\tan\gamma = H_y/H_x$. In this case magnetic field induces the toroidal moment directed along the z axis. For the ground state

$$t^0_z = \langle \psi_2 | \hat{t}_z | \psi_2 \rangle = \frac{1}{2}\cos\left(\alpha + \frac{\pi}{3} - \gamma\right). \tag{9}$$

From Eq. (9) it is evident that the value of the toroidal moment depends on orientation of external field in the plane of triangle. In the present case the dependence of averaged toroidal moment $\langle \hat{t}_z \rangle$ on the external field, temperature, and gap value is also given by Eq. (7).

Along with the appearance of the toroidal moment under the action of external magnetic field, which is described above, the inverse effect can be observed: variable electric field can produce the magnetic moment $\mathbf{M}$. To consider this effect, the interaction of toroidal moment with current should be taken into account. It is given by the term

$$\delta\hat{\mathcal{H}} = \frac{4\pi}{c}\mathbf{j}\hat{\mathbf{T}} = \mathbf{J}\hat{\mathbf{t}}, \tag{10}$$

where $\mathbf{j}=\mathbf{j}_e+\mathbf{j}_d$, $\mathbf{j}_e$ is the electric current, $\mathbf{j}_d=\partial\mathbf{E}/\partial t/4\pi$ is the displacement current, $\mathbf{E}$ is external electric field, which depends linearly on time, $\mathbf{J}=(2\pi l g\mu_B/c)\mathbf{j}$. Let us consider the case when the current is applied along the z axis. The energy levels E_i and eigenfunctions ψ_i are determined by diagonalization of Hamiltonian $\hat{\mathcal{H}}=\hat{\mathcal{H}}_{me}+\delta\hat{\mathcal{H}}$ [see Eqs. (3) and (10)]. Then one can find $E_{1,2}=-\frac{1}{2}(J \pm \sqrt{J^2+4\Delta^2})$, $E_{3,4}=\frac{1}{2}(J \pm \sqrt{J^2+4\Delta^2})$. The scheme of the level splitting is shown in Fig. 3. The eigenfunctions are rather cumbersome and they are not shown here. With the expression $\mathbf{M}=\mathrm{Tr}[\hat{\boldsymbol{\mu}}\exp(-\beta\hat{\mathcal{H}})]/\mathrm{Tr}[\exp(-\beta\hat{\mathcal{H}})]$, where $\hat{\boldsymbol{\mu}}=-g\mu_B\Sigma_{i=1,2,3}\hat{\mathbf{S}}_i$ is the magnetic-moment operator one can easily define the components of $\mathbf{M}$. Note the main features of this phenomenon. First, the current applied along the z axis induces magnetic moment in the plane of triangle. Second, $M_{x,y}$ diminishes with increasing J and $M_{x,y} \to 0$ at $J \gg \Delta$. At small J ($J < \Delta$) the components $M_{x,y}$ take simple form

$$\begin{pmatrix} M_x \\ M_y \end{pmatrix} = -\frac{1}{2}g\mu_B\begin{pmatrix} \cos\tilde{\alpha} \\ \sin\tilde{\alpha} \end{pmatrix}\tanh\frac{J_z}{kT},$$

where J_z is the projection of $\mathbf{J}$ on the z axis. Thus an electric field which is linear in time induces a constant magnetic moment in V_{15}. The Jahn-Teller splitting of levels causes this effect, which thus will vanish when $\Delta \to 0$.

Thus we show that the external magnetic field directed perpendicular to the plane of triangle induces toroidal moment in the plane of triangle along with magnetic moment directed perpendicular to the plane. It should be noted that this effect is responsible for the redistribution of spin density

among vanadium ions, which arranged in the apices of triangle. This redistribution is caused by the vibronic interactions. For the ground state $\langle \hat{S}_{1z,2z} \rangle = -\frac{1}{6} - \frac{1}{3}\cos(\alpha \mp \frac{\pi}{3})$, $\langle \hat{S}_{3z} \rangle = -\frac{1}{6} + \frac{1}{3}\cos\alpha$, i.e., $\langle \hat{S}_{1z} \rangle \neq \langle \hat{S}_{2z} \rangle \neq \langle \hat{S}_{3z} \rangle$, though $\Sigma_{i=1,2,3} \langle \hat{S}_{iz} \rangle = -1/2$. Note an interesting feature of the spectrum in the perpendicular magnetic field. In the field exceeding some critical value the ground state and the neighboring excited level have the same quantum number $m=-1/2$. The values of toroidal moment $t_{x,y}$ have opposite sign in these states. In accordance with it the direct magnetodipole absorption of electromagnetic radiation at frequency corresponding to transition between these splitted states, is forbidden. On the other hand, the existence of nonzero off-diagonal elements of toroidal moment $\langle \varphi_q(m) | \hat{t}_{x,y} | \varphi_{-q}(m) \rangle$ can allow the optical transition at frequency $2\pi f = \Delta/\hbar$ due to the perturbation $\delta\hat{\mathcal{H}} = (4\pi/c)\mathbf{j}(t)\hat{\mathbf{T}}$, where $\mathbf{j}(t) \sim (f\mathbf{E}/2)\cos(2\pi f t)$, $\mathbf{E} \perp \mathbf{z}$ is the amplitude of electric field. At that the intensity of the absorption is proportional to the transition probability per unit time $w_{q,-q}$, that is, determined by the off-diagonal components of the toroidal moment operator, i.e.,

$$w_{q,-q} \sim \frac{g^2 l^2 \mu_B^2}{\hbar c^2} 2\pi^2 f^2 E^2 \sin^2(\alpha + \pi/3 - \beta), \qquad (11)$$

where $\tan\beta = j_y/j_x$. It follows that the absorption of electromagnetic radiation depends on time due to the time evolution of the Jahn-Teller deformation of cluster determining the phase parameter $\alpha(t)$. In the case of incoherent distortions of clusters (or for long time) the averaged value of absorption will be observed.

There are more complicated interactions of the toroidal moment with external fields **E** and **H**: $V_1 \sim (\mathbf{T}[\mathbf{S},\mathbf{E}])$ and $V_2 \sim (\mathbf{T}[\mathbf{P},\mathbf{H}])$, where **S** and **P** are the spin and the dipole moment of molecule. They can be of interest in context of molecular magnetoelectricity.[4]

In conclusion, the present analysis reveals that a toroidal moment arises spontaneously in a triangular antiferromagnet due to intrinsic symmetry breaking as a result of the Jahn-Teller effect. The resulting quantum structure is thus richer and more versatile than originally assumed. This is quantified by the appearance of the four-component operators of Dirac's linear wave equation in the description of the qubits. We have indicated how these states may be manipulated by an external magnetic field and how inversely a magnetic moment can be induced by variable electric field or current. This feature can be useful for quantum computations in molecular clusters. At the present time the possibility of using V_{15} in quantum computers is actively studied.[24]

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