

Research Article

Quadrupole Excitations in Magnetic Susceptibility of Magnetic Nanoparticle Fe_8

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Magnetic susceptibility dependence on temperatures in different magnetic fields will be discussed. Until today, to calculate magnetization and magnetic susceptibility, only dipole excitations have been considered, but, due to the symmetry of operators in Hamiltonian and also to achieve more accuracy, other multiple excitations must be taken into account too. To this end, here, both dipole and quadrupole excitations are considered and then the resulting curves will be plotted in presence of different magnetic fields. Finally, seen that the graphs obtained using the multipole excitations more accurately with results taken by experimental data.

1. Introduction

Today about 100 magnetic clusters are known that show some behaviors or characteristics which are interesting from physics science point of view. These molecules are intermediate between simple paramagnetic salts and superparamagnetic particles of submicrometer size [1].

The magnetic cluster Fe_8 , Figure 1, has a spin ground state $S = 10$, which arises from competing antiferromagnetic interactions among the eight $S = 5/2$ Fe spins whose dynamics are quite similar. Spin-orbit and spin-spin interactions destroy complete rotational invariance and give rise to anisotropy with respect to the crystal lattice directions [2, 3].

A variety of experimental techniques (electron spin resonance, ac susceptibility, magnetic relaxation, Mossbauer spectroscopy, and neutron scattering) indicates [3–6] that experimental data are in agreement with the following model Hamiltonians:

$$\mathcal{H} = -DS_z^2 + ES_x^2 + g\mu_B \mathbf{S} \cdot \mathbf{B}, \quad (1)$$

where S_x and S_z are the two components of spin operator and D and E are the anisotropy constants and are known through variety of experimental evidence with $D = 0.33k$, $E = 0.092k$ [7, 8]. The last term of the Hamiltonian describes the Zeeman energy associated with and applied field \mathbf{H} . This

Hamiltonian defines hard and easy axes of magnetization in x and z direction, respectively.

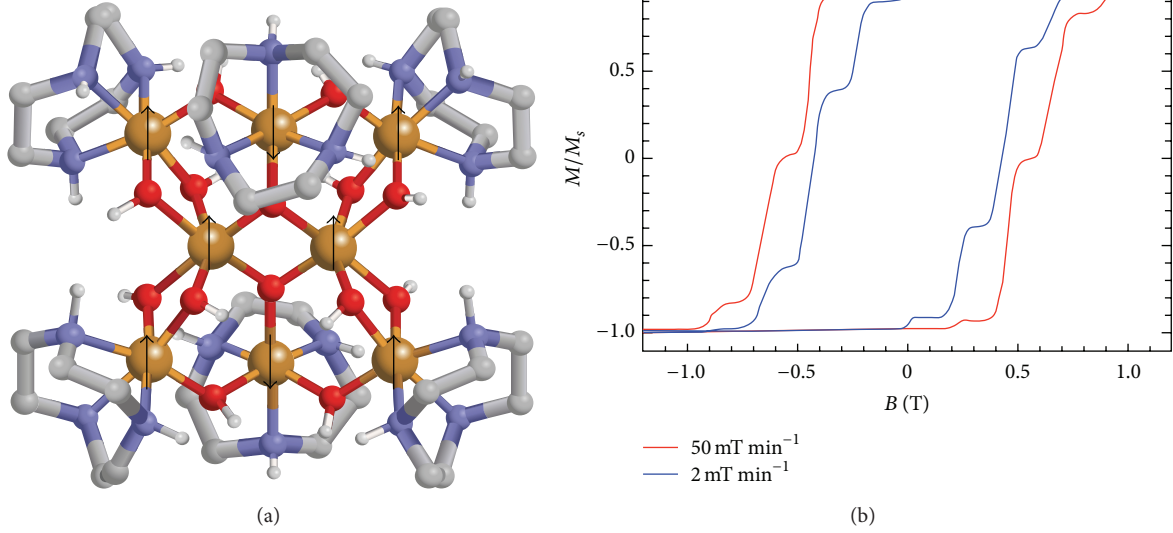
In physics, every system could be characterized by its response to external stimuli. In our study, nanoparticles could be characterized by a response function. We are mainly concerned with the response of such a system to a magnetic field. In this case, the output is the magnetization and the response function is the magnetic susceptibility.

The magnetic susceptibility of a material, commonly symbolized by χ_m , is equal to the ratio of the magnetization \mathbf{M} within the material to the applied magnetic field strength B and can be obtained from the following equation [7]:

$$\chi = -\frac{1}{B} \frac{(\sum_i (\partial E_i / \partial B) e^{-E_i/kT})}{\sum_i e^{-E_i/kT}}, \quad (2)$$

where E_i is the energy of the system.

In this study, firstly, the magnetic susceptibility dependence of high spin molecule Fe_8 (exact chemical formula: $[\text{Fe}_8\text{O}_2(\text{OH})_{12}(\text{tacn})_6]^{8+}$) on temperature in the presence of different magnetic fields in $SU(2)$ group is calculated. In other words, only dipole excitation in Hamiltonian is utilized. In order to be more accurate and due to the symmetry and power of spin operators in Hamiltonian, other multipole excitations are introduced.

FIGURE 1: Chemical structure and hysteresis loop of Fe_8 .

2. Magnetic Susceptibility Calculation in SU(2) Group

Expectation values of different spin operators in SU(2) group are in the following form:

$$\begin{aligned} S^+ &= e^{i\varphi} \sin \theta, \\ S^- &= e^{-i\varphi} \sin \theta, \\ S^z &= \cos \theta. \end{aligned} \quad (3)$$

If only dipole excitations in calculation of magnetization are considered, classical energy of the system from Hamiltonian (1) is obtained through the following form:

$$E_d = -D\cos^2\theta + E\sin^2\theta\cos^2\varphi + g\mu_B SB \cos \theta. \quad (4)$$

Substituting this relation into (2), magnetic susceptibility is obtained in the following form:

$$\begin{aligned} \chi &= \left(\int_0^{\pi/2} (g\mu_B S \cos \theta) \right. \\ &\quad \cdot \exp \left[- \left(-D\cos^2\theta + E\sin^2\theta\cos^2\varphi \right. \right. \\ &\quad \quad \left. \left. + g\mu_B SB \cos \theta \right) \right. \\ &\quad \left. \cdot (kT)^{-1} \right] d\theta \Big) \\ &\quad \cdot \left(\int_0^{\pi/2} \exp \left[- \left(-D\cos^2\theta + E\sin^2\theta\cos^2\varphi \right. \right. \right. \end{aligned}$$

$$\begin{aligned} &\quad \left. \left. + g\mu_B SB \cos \theta \right) \right. \\ &\quad \left. \cdot (kT)^{-1} \right] d\theta \Big)^{-1}. \end{aligned} \quad (5)$$

For calculation of this integral, we used this approximation:

$$e^x = \sum_{n=0}^{\infty} \frac{x^n}{n!} \approx 1 + x. \quad (6)$$

Then

$$\chi = \frac{20\mu_B \left(1 + (1/kT) (0.21 - 0.03\cos^2\varphi - 15.6\mu_B B) \right)}{\left(1 + (1/kT) (0.24 - 0.07\cos^2\varphi - 20\mu_B B) \right)}. \quad (7)$$

Magnetic susceptibility along easy axis ($\varphi = 0$) and hard axis ($\varphi = \pi/2$) is obtained in the following forms:

$$\chi = \begin{cases} \frac{20\mu_B \left(1 + (1/kT) (0.21 - 15.6\mu_B B) \right)}{\left(1 + (1/kT) (0.24 - 20\mu_B B) \right)} & \text{for } \varphi = \frac{\pi}{2} \\ \frac{20\mu_B \left(1 + (1/kT) (0.18 - 15.6\mu_B B) \right)}{\left(1 + (1/kT) (0.17 - 20\mu_B B) \right)} & \text{for } \varphi = 0. \end{cases} \quad (8)$$

The plot of the numerical calculation of magnetic susceptibility versus temperature is obtained as shown in Figure 2.

As it is seen from Figure 2, in low magnetic field, an increase in the temperature causes an increase in the magnetic susceptibility. But as the magnetic field increases, magnetic susceptibility becomes less dependent on temperature and eventually it almost reaches a constant value. Moreover, for magnetic field along the easy and hard axes, both of these curves are similar.

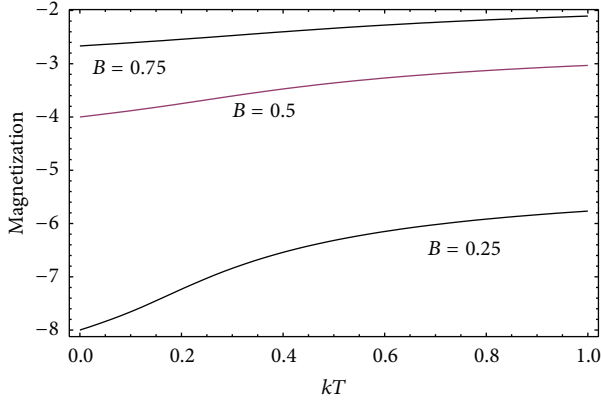


FIGURE 2: In this case, only dipole excitation is considered and magnetic susceptibility curves along easy axes ($\varphi = 0$) and hard axes ($\varphi = \pi/2$) are similar.

3. Magnetic Susceptibility Calculation in SU(3) Group

If dipole and quadrupole excitations are considered, the expectation values of spin operators are obtained in the following form [9]:

$$\begin{aligned} S^+ &= e^{i\varphi} \cos 2g \sin \theta, \\ S^- &= e^{-i\varphi} \cos 2g \sin \theta, \\ S^z &= \cos 2g \cos \theta. \end{aligned} \quad (9)$$

Classical energy of the system is obtained from Hamiltonian (1) in the following form:

$$\begin{aligned} E_{cl} &= -D\cos^2 2g\cos^2 \theta + E\cos^2 2g\sin^2 \theta\cos^2 \varphi \\ &\quad - g\mu_B B \cos 2g \cos \theta. \end{aligned} \quad (10)$$

With substitution of this relation into (2), magnetic susceptibility is obtained:

$$\begin{aligned} \chi &= \left(\int_0^{\pi/2} \int_0^{\pi/4} (g\mu_B S \cos 2g \cos \theta) \right. \\ &\quad \cdot \exp \left[- \left(-D\cos^2 2g\cos^2 \theta \right. \right. \\ &\quad \quad \left. \left. + E\cos^2 2g\sin^2 \theta\cos^2 \varphi \right. \right. \\ &\quad \quad \left. \left. + g\mu_B B \cos 2g \cos \theta \right) \right. \\ &\quad \left. \cdot (kT)^{-1} \right] d\theta dg \end{aligned}$$

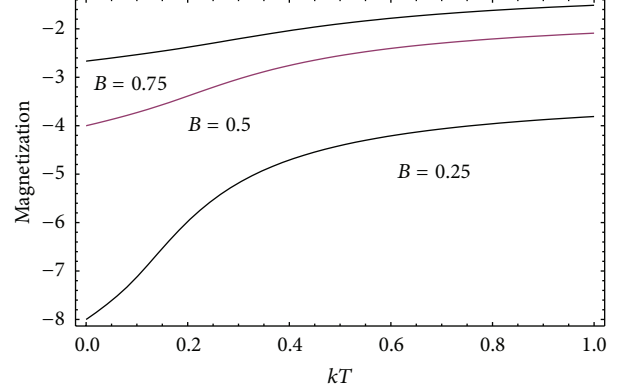


FIGURE 3: In this case, both dipole and quadrupole excitations are considered and magnetic susceptibility curve along easy axes ($\varphi = 0$) and along hard axes ($\varphi = \pi/2$).

$$\begin{aligned} &\cdot \left(\int_0^{\pi/2} \int_0^{\pi/4} \exp \left[- \left(-D\cos^2 2g\cos^2 \theta \right. \right. \right. \\ &\quad \left. \left. + E\cos^2 2g\sin^2 \theta\cos^2 \varphi \right. \right. \\ &\quad \left. \left. + g\mu_B B \cos 2g \cos \theta \right) \right. \\ &\quad \left. \cdot (kT)^{-1} \right] d\theta dg \end{aligned} \quad (11)$$

If used from relation (6), magnetic susceptibility is obtained in the following form:

$$\chi = \frac{20\mu_B (0.5 + (1/kT) (0.07 - 0.01\cos^2 \varphi - 6\mu_B B))}{(1 + (1/kT) (0.09 - 0.04\cos^2 \varphi - 10\mu_B B))}. \quad (12)$$

Magnetic susceptibility in different magnetic fields for the easy axis direction ($\varphi = 0$) and the hard axis direction ($\varphi = \pi/2$) is obtained in the following form:

$$\chi = \begin{cases} \frac{20\mu_B (0.5 + (1/kT) (0.07 - 6\mu_B B))}{(1 + (1/kT) (0.09 - 10\mu_B B))} & \text{for } \varphi = \frac{\pi}{2} \\ \frac{20\mu_B (1 + (1/kT) (0.06 - 6\mu_B B))}{(1 + (1/kT) (0.05 - 10\mu_B B))} & \text{for } \varphi = 0. \end{cases} \quad (13)$$

If dipole and quadrupole excitations are considered, the plot of the numerical calculation of magnetic susceptibility is obtained as shown in Figure 3.

As it is clear from Figure 3, similar to the previous section, in low magnetic field, if temperature increases, magnetic susceptibility increases too. But, because of the quadrupole excitation, this dependency is very dramatic. Also, with increasing magnetic field, magnetic susceptibility becomes less dependent and it approaches a constant value, but this is different from the case in which only dipole excitation was considered. In addition, like the previous results, both

of these curves are similar for both magnetic fields along the easy and hard axes.

4. Conclusion

In this paper, we have discussed magnetic susceptibility dependence on temperatures in presence of different magnetic fields. If only dipole excitation in calculation is considered, for the magnetic susceptibility, Figure 2 is obtained. As we see, in low magnetic field and in low temperatures, this dependency is very clear. As temperature increases or magnetic field increases, magnetization almost becomes independent of temperatures and this is due to all magnetic dipoles that all of them along with the magnetic field (saturate magnetization).

If quadrupole excitation is added, Figure 3 is obtained. At low temperature, this dependency is very dramatic and this is because of the quadrupole excitations that have not been considered. As magnetic field increases, in comparison to the previous section, there are some dependencies due to quadrupole excitations.

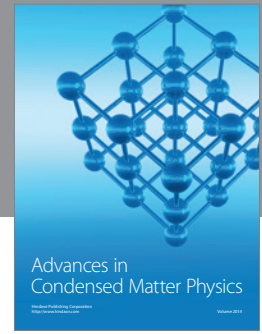
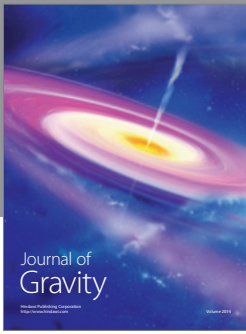
Conflict of Interests

The author declares that there is no conflict of interests regarding the publication of this paper.

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