Magnetic susceptibility of effective two-level systems: insights from a Heisenberg isosceles triangle

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Abstract

We derive a general description of the low-temperature magnetic susceptibility for systems effectively represented by two energy levels. In analogy with the Schottky theory of the specific heat of two-level systems, the position and height of the magnetic susceptibility maximum are shown to be determined solely by the energy gap, the relative degeneracy of the two levels, and their spin difference. The theory predicts that increasing the relative degeneracy enhances the susceptibility peak and shifts it to lower temperatures. The usefulness of this theory is illustrated on the spin-1/2 Heisenberg isosceles triangle, which serves as a paradigmatic example of the proposed two-level description.

Keywords: magnetic susceptibility; two-level system; Heisenberg triangle

1. Introduction

Frustrated quantum spin systems often exhibit a highly degenerate low-energy spectrum, giving rise to unconventional magnetic properties and field-induced quantum phase transitions. In the vicinity of a level-crossing field, their low-temperature thermodynamics is frequently governed by only the ground state and the first excited state, allowing for an effective two-level description [1, 2]. Although the magnetic specific heat of effectively two-level systems is well established within the Schottky theory [3–5], a general analytical description of the low-temperature magnetic susceptibility has not been reported so far.

Magnetic susceptibility is one of the most widely used experimental probes of magnetic materials, providing direct information on exchange interactions, spin states, and low-energy excitations. In the present work, we derive a general analytical description of the susceptibility maximum in effective two-level systems and demonstrate that its position and height are fully determined by the energy gap, the relative degeneracy, and the spin difference of the two lowest energy levels. The theory is illustrated using the exactly solvable spin-1/2 Heisenberg isosceles triangle and further demonstrated on experimental susceptibility data for a linear trinuclear copper(II) complex [6].

2. Susceptibility of a two-level system

Let us consider an effective two-level system consisting of a ground state and a first excited state with energies

$$\begin{aligned} E_0 &= E_0^{h=0} - hS_0, \\ E_1 &= E_1^{h=0} - hS_1, \end{aligned} \tag{1}$$

where $E_0^{h=0}$ and $E_1^{h=0}$ are the zero-field energies of the ground and first excited states, respectively, $h = g\mu_{\rm B}B$ denotes the magnetic field term, with g being the Landé g factor and $\mu_{\rm B}$ the Bohr magneton, while S_0 (S_1) is the total spin of the ground (first excited) state. Defining the energy gap as

$$\Delta_g = E_1 - E_0 = \Delta E^{h=0} - h\Delta S,$$

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where $\Delta E^{h=0} = E_1^{h=0} - E_0^{h=0}$ and $\Delta S = S_1 - S_0$, the partition function of the two-level system reads

$$\begin{aligned} \mathcal{Z} &= d_0 \exp(-\beta E_0) + d_1 \exp(-\beta E_1) \\ &= \exp(-\beta E_0) \left[d_0 + d_1 \exp(\beta \Delta_g) \right]. \end{aligned} \tag{2}$$

Here, d_0 and d_1 represent the degeneracy factors of the ground state and the first excited state, respectively. The corresponding Helmholtz free energy then reads

$$F = -k_B T \ln \mathcal{Z} = E_0 - k_B T \ln[d_0 + d_1 \exp(\beta \Delta_g], \tag{3}$$

from which the magnetization follows as

$$M = -\left(\frac{\partial F}{\partial B}\right)_T = g\mu_B \left[S_0 + \Delta S \frac{d_1}{d_1 + d_0 \exp(\beta \Delta_g)} \right]. \tag{4}$$

The magnetic susceptibility is then obtained as

$$\chi = \left(\frac{\partial M}{\partial B}\right)_T = \frac{d_1}{d_0} \frac{(g\mu_B \Delta S)^2 \beta}{\left[\exp\left(\frac{\beta \Delta_g}{2}\right) + \frac{d_1}{d_0} \exp\left(-\frac{\beta \Delta_g}{2}\right)\right]^2}. \tag{5}$$

Since Eq. (5) exhibits a maximum as a function of temperature, its position can be determined from the stationary condition $\left(\frac{\partial \chi}{\partial T}\right)_h = 0$, which leads to the transcendental equation

$$\beta_{max} \Delta_g = \frac{\exp\left(\frac{\beta_{max}\Delta_g}{2}\right) + \frac{d_1}{d_0} \exp\left(-\frac{\beta_{max}\Delta_g}{2}\right)}{\exp\left(\frac{\beta_{max}\Delta_g}{2}\right) - \frac{d_1}{d_0} \exp\left(\frac{\beta_{max}\Delta_g}{2}\right)}. \tag{6}$$

The position and height of the susceptibility maximum are obtained by numerically solving Eq. (6) and substituting the resulting solution into Eq. (5). As follows from Eqs. (5)–(6), the position of the susceptibility maximum depends on the energy gap and the relative degeneracy of the first excited and ground states. By contrast, the height of the susceptibility maximum also depends on the spin difference ΔS between the two levels.

Typical temperature dependences of the low-temperature susceptibility are shown in Fig. 1(a) for selected degeneracy ratios d_1/d_0. The susceptibility and temperature are normalized with respect to the energy gap Δ_g and the spin difference ΔS to isolate the effect of the degeneracy ratio. With increasing d_1/d_0, the susceptibility peak becomes higher, sharper, and shifts to lower temperatures. The corresponding positions and heights of the susceptibility maxima are summarized in Table 1. This trend is analogous to the Schottky-type specific-heat maximum, whose height also increases and its position shifts to lower temperatures with increasing relative degeneracy [5].

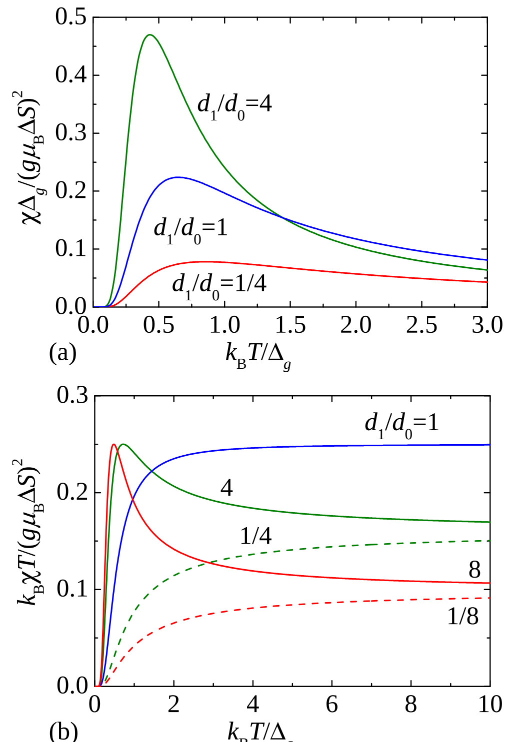

Figure 1: (a) Low-temperature susceptibility peak as a function of temperature for selected degeneracy ratios $d_1/d_0 = 1/4$, 1, and 4. (b) Susceptibility times temperature product χT for degeneracy ratios $d_1/d_0 = 1/8$, 1/4, 1, 4, and 8.

The susceptibility times temperature product is shown in Fig. 1(b) for several values of the degeneracy ratio d_1/d_0. Three main features can be identified. First, the χT product increases monotonically with temperature for $d_1/d_0 \leq 1$, whereas a low-temperature maximum appears for $d_1/d_0 > 1$. Second, the emergence of this maximum

d_1/d_0	$\frac{k_B T_{max}}{\Delta_g}$	$\frac{\chi_{max}\Delta_g}{(g\mu_B\Delta S)^2}$	d_1/d_0	$\frac{k_B T_{max}}{\Delta_g}$	$\frac{\chi_{max}\Delta_g}{(g\mu_B\Delta S)^2}$
1/4	0.856	0.078	3/2	0.579	0.287
1/2	0.763	0.137	5/3	0.562	0.305
2/3	0.717	0.170	9/5	0.549	0.318
1	0.648	0.224	2	0.532	0.337

Table 1: Position and height of the susceptibility maximum for selected values of the relative degeneracy d_1/d_0 in a two-level system.

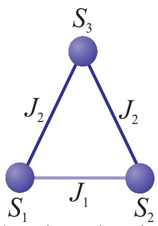

Figure 2: Spin-1/2 Heisenberg isosceles triangle with two distinct coupling constants J_1 and J_2.

is solely determined by the relative degeneracy of the two energy levels. Finally, the high-temperature limit of the χT product reaches its maximum at $d_1/d_0 = 1$, while it is identical for reciprocal degeneracy ratios d_1/d_0 and d_0/d_1, reflecting the equal Boltzmann weights of the two levels in the $T \to \infty$ limit.

3. Spin-1/2 Heisenberg isosceles triangle

To illustrate the applicability of this theory, we consider the exactly solvable spin-1/2 Heisenberg isosceles triangle schematically shown in Fig. 2, which is described by the Hamiltonian

$$\hat{\mathcal{H}} = J_1 \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 + J_2 \left(\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_3 + \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_3 \right) - h \hat{S}_T^z, \tag{7}$$

where $\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$ denotes the scalar product between two spin-1/2 operators, $J_1 > 0$ and $J_2 > 0$ are two antiferromagnetic coupling constants, and $h = g\mu_B B$ is the Zeeman's term. Introducing the composite spin operator $\hat{\mathbf{S}}_{12} = \hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2$ and the total spin operator $\hat{\mathbf{S}}_T = \hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_3$, the Hamiltonian can be diagonalized using the conserved quantum spin numbers S_{12}, S_T, and S_T^z. The corresponding eigenenergies are

$$\begin{aligned} E_{S_T, S_{12}, S_T^z} = &\frac{J_1}{2} \left[S_{12}(S_{12}+1) - \frac{3}{2} \right] \\ &+ \frac{J_2}{2} \left[S_T(S_T+1) - S_{12}(S_{12}+1) - \frac{3}{4} \right] - h S_T^z. \end{aligned} \tag{8}$$

The allowed values of the quantum spin numbers are $S_{12} = 0, 1$, $S_T = 1/2, 3/2$, and $S_T^z = -S_T, -S_T + 1, \ldots, S_T$. The partition function then takes the form

$$\begin{aligned} \mathcal{Z} = &\; 2\exp\left(\frac{3\beta J_1}{4}\right) \cosh\left(\frac{\beta h}{2}\right) + 2\exp\left(\frac{\beta J_1}{4} + \frac{\beta J_2}{2}\right) \cosh\left(\frac{\beta h}{2}\right) \\ &+ 2\exp\left(-\frac{\beta J_1}{4} - \frac{\beta J_2}{2}\right) \left[\cosh\left(\frac{3\beta h}{2}\right) + \cosh\left(\frac{\beta h}{2}\right) \right]. \end{aligned} \tag{9}$$

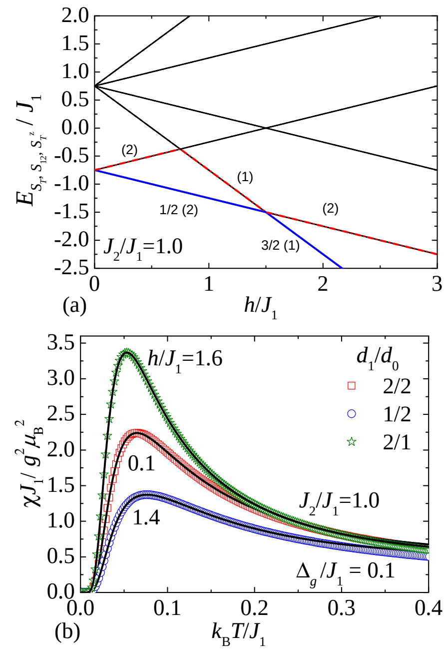

Figure 3: The results for the spin-1/2 Heisenberg equilateral triangle with $J_2/J_1 = 1$: (a) energy spectrum with blue (red) lines denoting ground state (first excited state); (b) temperature dependence of the susceptibility for selected magnetic fields. Lines show the exact solution, whereas symbols represent the effective two-level theory.

4. Results and discussions

The energy spectrum of the spin-1/2 Heisenberg equilateral triangle corresponding to the particular case with $J_2/J_1 = 1$ is shown in Fig. 3(a). Ground (first excited) states are shown as blue solid (red dashed) lines and labeled by the corresponding total spin and degeneracy. Two level crossings occur at $h/J_1 = 0$ and 1.5. Notably, the Zeeman splitting does not completely lift the degeneracy, leaving two-fold degenerate levels that may constitute either the ground or the first excited state.

The temperature dependence of the susceptibility is shown in Fig. 3(b) for three representative magnetic fields, $h/J_1 = 0.1$, 1.4, and 1.6, chosen to correspond to the same energy gap $\Delta_g/J_1 = 0.1$. The susceptibility peaks nevertheless exhibit different positions and heights owing to different relative degeneracies of the two lowest energy levels, namely $d_1/d_0 = 1$, 1/2, and 2. As predicted by the two-level theory, increasing the relative degeneracy makes the susceptibility peak higher, sharper, and shifts it to lower temperatures, in close analogy with the Schottky-type specific-heat peak discussed in Ref. [5]. The two-

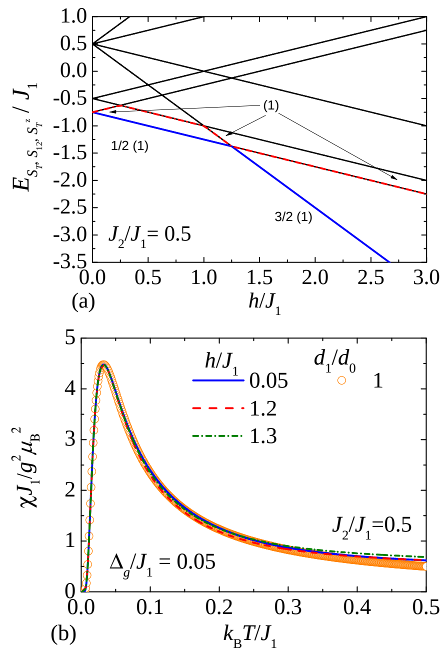

Figure 4: Spin-1/2 Heisenberg isosceles triangle with $J_2/J_1 = 0.5$: (a) energy spectrum with blue (red) lines denoting ground state (first excited state); (b) temperature dependence of the susceptibility for selected magnetic fields. Lines show the exact solution, whereas symbols represent the effective two-level theory.

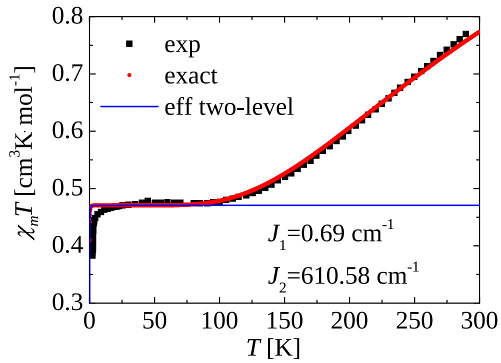

Figure 5: Experimental χT data for trinuclear complex Cu_3 from Ref. [6] together with the exact solution of the spin-1/2 Heisenberg triangle and the effective two-level approximation.

level description remains in excellent agreement with the exact results up to $k_B T/J_1 \lesssim 0.3$.

The energy spectrum of the spin-1/2 Heisenberg isosceles triangle with $J_2/J_1 = 0.5$ is shown in Fig. 4(a). In contrast to the equilateral triangle, all relevant ground and first excited states are nondegenerate ($d_1 = d_0 = 1$). Temperature dependence of the susceptibility per spin is shown in Fig. 4(b) for several magnetic fields selected in the vicinity of the level-crossing fields $h/J_1 = 0$ and 1.25. All fields were chosen to yield the same energy gap $\Delta_g/J_1 = 0.05$. Since all relevant states are nondegenerate ($d_1/d_0 = 1$), the susceptibility curves exhibit identical maxima, in agreement with the two-level theory. The theoretical prediction is in excellent agreement with the exact results up to $k_B T/J_1 \lesssim 0.3$, while deviations at higher temperatures arise from the thermal population of higher-energy states beyond the two-level approximation.

Finally, we demonstrate the applicability of the two-level description to experimental data reported for the linear trinuclear copper(II) complex $Cu_3(bhbd)_2(CH_3OH)_2(ClO_4)_2$ from Ref. [6] further abbreviated as Cu_3. Here, $\chi_m = N_A \chi$ denotes the molar magnetic susceptibility (N_A is Avogadro's constant). The coupling constants were obtained from a fit of the exact solution of the spin-1/2 Heisenberg isosceles triangle to the measured susceptibility. As shown in Fig. 5, the effective two-level model accurately reproduces the low-temperature plateau, while deviations at elevated temperatures originate from the thermal population of higher-energy states neglected within the two-level approximation.

5. Concluding remarks

We have derived a general analytical description of the low-temperature magnetic susceptibility for effective two-level systems. The proposed analytical framework can be readily applied to a broad class of magnetic systems exhibiting an effective two-level low-energy spectrum. The theory shows that the position of the susceptibility maximum is determined by the energy gap and the relative degeneracy of the two levels, whereas its height additionally depends on their spin difference. These general predictions were illustrated using the exactly solvable spin-$1/2$ Heisenberg isosceles triangle, where different relative degeneracies lead to qualitatively different susceptibility maxima. Finally, the applicability of the proposed approach was demonstrated by describing the low-temperature susceptibility plateau of a linear trinuclear copper(II) complex. The present results provide a simple framework for interpreting low-temperature susceptibility data in a broad class of magnetic systems.

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