Monte Carlo study of the magnetic properties of spin liquid compound $NiGa_2S_4$*

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The magnetic properties of two-dimensional antiferromagnet $NiGa_2S_4$ have attracted much attention and yet some problems are far from being solved. We investigate the magnetic properties of $NiGa_2S_4$ by Monte Carlo simulations. A new spin-interacting model is proposed to describe the system, and the specific heat together with the doping effect of nonmagnetic impurity is studied by simulations. The double peaks of the specific heat as well as other behaviors are well reproduced. We also compare our results with those of other models, and the underlying physics is discussed.

Keywords: spin liquid state, specific heat, magnetization, correlation length

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1. Introduction

Generally, when a spin glass system is cooled down to the freezing temperature, its spins gradually become frozen, which means that the relative direction between spins becomes fixed.[1–6] However, for a geometrically frustrated system, i.e., $NiGa_2S_4$, which is a two-dimensional (2D) antiferromagnet with a triangular lattice, the case is very different. Experiments found no long-range order for the compound $NiGa_2S_4$, and the spins remained disordered even at a very low temperature, which is called the spin liquid state.[7] It was once believed that the geometrical frustration could lead to the spin liquid state. However, numerical studies revealed that the spins form a spiral ground state with 120° rotation from each other if the nearest-neighbor coupling is considered. The studies also revealed that $NiGa_2S_4$ has a third nearest neighbor coupling with an abnormally large magnitude, which is comparable to the first nearest neighbor coupling, and the second nearest neighbor coupling is very small compared to the two other couplings.[8] Experiments have shown that the specific heat of $NiGa_2S_4$ has two rounded peaks. One peak is broad and has a Curie–Weiss temperature of 80 K, and the other locates at 10 K.[7] Furthermore, the specific heat measurement shows no sign of phase transition at both temperatures. Below 10 K, the specific heat decreases proportionally to T^2 as the temperature approaches to zero, which indicates a gapless energy spectrum.[7] Although the spins do not become frozen, an incommensurate order emerges and the correlation length increases to a finite length as the temperature decreases to zero.[7,9,10] Moreover, the T^2 law of the specific heat and the low-temperature peak are more robust and do not change significantly as nonmagnetic impurities are gradually introduced.[11–13] Theoretically, several studies attributed the low-temperature T^2 law to the formation of a quadrupolar state, which could be stabilized by the biquadratic interaction and solved by the mean-field method.[14–16] Although the mean-field method can describe the low-temperature thermodynamics, it fails to give the double peaks of the specific heat. Monte Carlo simulations also verified that only one peak emerges for the specific heat when the first and the third nearest neighbor couplings are considered.[17] Actually, the double peaks should be attributed to the competition between two kinds of magnetic interactions. Recently, Stoudenmire *et al.* proposed that the biquadratic exchange between the nearest neighbor spins played an important role in the double peaks of the specific heat.[18] Their studies cast some light on the puzzles of the magnetic properties of $NiGa_2S_4$, however more investigations and careful inspections are still needed. Besides, the theoretical study also proposed that the double peaks arose from the proliferation of vortices and antivortices which led to the Berezinskii–Kosterlitz–Thouless (BKT) transition.[19] This model can give some proper explanations for the behavior of $NiGa_2S_4$. However, a more quantitative analysis is still absent.

In this article, we report our numerical study on the mag-

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netic properties of $NiGa_2S_4$. A new spin-interacting model is proposed, and the specific heat and the correlation lengths are calculated by using the Monte Carlo method. Our results reveal that the double peaks of the specific heat and the magnetization behavior can be well reproduced by the model. The specific heat of nonmagnetic-impurity doped $NiGa_2S_4$ is also studied, and the results agree with the experimental ones quite well. We also compare our results with those of other models, and the underlying physics is discussed.

2. Methods and calculation details

A triangular lattice with a size of $L \times L$ is used to simulate the magnetic properties of two-dimensional antiferromagnet $NiGa_2S_4$, and L takes 18, 30, and 42 respectively. The Heisenberg model of the spin system in an external field can be written as

$$H = J_{ij} \sum_{i,j} \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j - k_u \sum_{i,j} (\boldsymbol{\sigma}_i \cdot \boldsymbol{n})(\boldsymbol{\sigma}_j \cdot \boldsymbol{n}) - \sum_i \boldsymbol{\sigma}_i \cdot \boldsymbol{h}. \quad (1)$$

The first term contains the exchange couplings between the first, the second, and the third nearest neighbors. The second term is the inter-spin anisotropic coupling between the nearest neighbors, where $\boldsymbol{n}$ is a unit vector and set to be along the x axis. Recently, Mazin studied the neighbor coupling parameters by first-principles calculations.[8] The first nearest neighbor coupling is found to be easily suppressed by the on-site Coulomb repulsion, while the third nearest neighbor coupling is very robust against the impact of the Coulomb repulsion. The second nearest neighbor coupling is negligible compared to the first and the third nearest neighbor couplings. In our simulations, we use the neighbor coupling parameters in Ref. [8] and set them to be $J_1 = 8.4$ meV, $J_2 = 0.3$ meV, and $J_3 = 4.1$ meV. We use the heat bath method to update the spin configurations.[20,21] This method directly generates new spin orientations according to the Boltzmann distribution and hence updates the configurations effectively. Further, we use the parallel tempering method[22,23] to speed up the relaxation of the frustrated systems at low temperatures. The system is assumed to reach an equilibrium state when the correlation length becomes saturated with increasing Monte Carlo sweeps. The specific heat of the magnetic system can be written as

$$C_M = (E - \langle E \rangle)^2 / k_B T^2. \quad (2)$$

Furthermore, we calculate the correlation lengths of both spin-glass (SG) and chiral-glass (CG) phases for the system. In most simulations, the SG order is defined as the spin overlaps between two replicas which have the same parameters but evolve independently. The components of the k-dependent spin overlaps can be written as[24,25]

$$q_{\mu\nu}(\boldsymbol{k}) = \frac{1}{N} \sum_i \sigma_\mu^{(\alpha)}(\boldsymbol{r}_i) \sigma_\nu^{(\beta)}(\boldsymbol{r}_i) \, e^{i\boldsymbol{k}\cdot\boldsymbol{r}_i}, \quad (3)$$

where μ and ν are the spin components, α and β are the indices of the replicas, and N is the total number of spins. The SG order can be written as

$$q_s(\boldsymbol{k}) = \sqrt{\sum_{\mu,\nu} |q_{\mu\nu}(\boldsymbol{k})|^2}. \quad (4)$$

Similarly, the chirality can be written as $\chi_i = \boldsymbol{\sigma}_i \times \boldsymbol{\sigma}_j$,[26] and the k-dependent overlaps of the chirality can be written as

$$q_\chi(\boldsymbol{k}) = \frac{1}{N} \sum_i \chi_i^{(\alpha)}(\boldsymbol{r}_i) \chi_i^{(\beta)}(\boldsymbol{r}_i) \, e^{i\boldsymbol{k}\cdot\boldsymbol{r}_i}. \quad (5)$$

Since the chirality defined above is along the z direction, $|q_\chi(\boldsymbol{k})|$ is just the k-dependent CG order. For the system with a finite size, the correlation length can be written as[27,28]

$$\xi_L = \frac{1}{2\sin(k_m/2)} \sqrt{\frac{[\langle q(0)^2 \rangle]}{[\langle q(\boldsymbol{k}_m)^2 \rangle]} - 1}, \quad (6)$$

where $\boldsymbol{k}_m = (2\pi/L, 0)$ or its permutation.

3. Results and discussion

The specific heat of $NiGa_2S_4$ is experimentally found to have double peaks at two distinct temperatures. The two peaks have nearly the same magnitude.[7] Moreover, the field-cooled magnetization in a small external field is observed to rise gradually when the system is cooled down from a high temperature. After the system is cooled below the freezing temperature, a maximum is observed and the magnetization becomes flat.[7] By simulation, we calculate the specific heat and the magnetization from 5 K to 190 K and show them in Fig. 1.

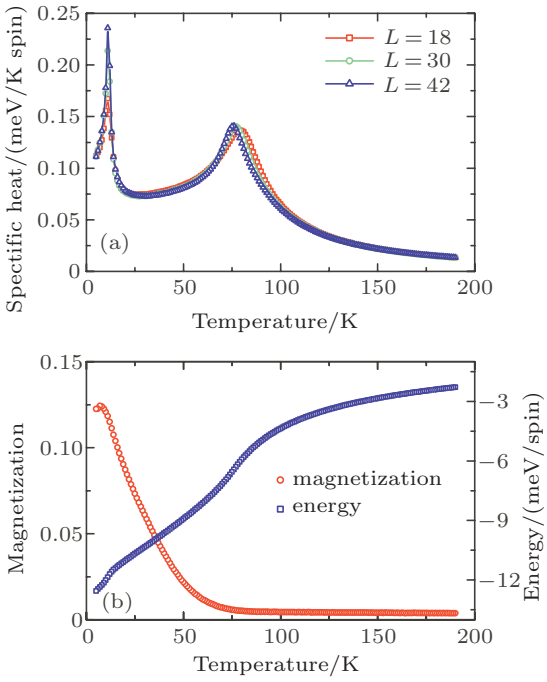


Fig. 1. (color online) (a) Specific heat versus temperature for the systems with different sizes. The J_1, J_2, and J_3 are set to be 8.4 meV, 0.3 meV, and 4.1 meV, respectively, and k_u is 8.0 meV. (b) Magnetization and average spin energy versus temperature for a lattice of $L = 42$. An external field ($h = 0.5$ meV) is applied along the y axis.

As can be seen from Fig. 1(a), double peaks emerge in the specific heat as the temperature decreases. The high-temperature peak locates at $T_h = 79$ K, while the low temperature peak locates at $T_l = 11$ K. These values agree well with the experimental results. Both peaks shift to lower temperatures as the system size increases, indicating a finite size effect. Note that the BKT model also gives two specific heat peaks with the same magnitude at $T = 0.5J$ and $1.3J$, respectively.[19] Figure 1(b) shows the magnetization behavior with decreasing temperature. Our results reveal that the magnetization arises gradually as the temperature decreases, then it reaches the maximum at the freezing temperature T_l, and becomes flat after that. This is consistent with the experimental results.[7] The energy curve is found to have two upward curvatures at T_h and T_l, which lead to the maxima of the specific heat. The energy descents at T_h and T_l are caused by the three neighbor couplings and the anisotropic interaction, respectively.

It should be mentioned that the specific heat below T_l decreases more rapidly than the T^2 law. This is because our simulations are based on the canonical Monte Carlo method and the spin reversal obeys the Boltzmann distribution. However, at low temperatures, the theoretical studies show that the collective spin-wave excitation is responsible for the T^2 law.[29] Thus, the quantitative study on the low temperature behavior of the specific heat is beyond the scope of this research.

Our result shows that the height of the low-temperature peak is significantly higher than that of the high-temperature peak, while the experimental result reveals that they have nearly the same height. However, our study reveals that the ratio of the two peak heights is highly dependent on the coupling parameters. For a small J_1, the height of the low-temperature peak decreases more compared to that of the high-temperature peak. Thus better coupling parameters from experiments would possibly improve the behavior.

It is interesting to study the correlation lengths of the SG and the CG phases of the model. Experiments have shown that the spins gradually become frozen and the correlation length grows when the temperature is below the freezing temperature.[7,9,10] In our study, both the SG and the CG correlation lengths are calculated and shown in Fig. 2. It is noticed that the SG correlation lengths are negligible when the temperature is above T_h. As the temperature decreases below T_h, they increase very slowly. When the temperature is below T_l, they increase rapidly. Note that the curves cross each other at T_h, which means that an SG transition occurs. However, the SG correlation length increases rapidly only when the temperature is below the freezing temperature T_l. It is also noticed that the CG correlation length increases very slowly until the temperature is below T_l. Below T_l, the CG correlation length increases rapidly, indicating a CG transition. The previous studies also revealed that a CG transition occurred in the system with biquadratic interaction.[26] This means that a CG phase may exist in the spin liquid state of $NiGa_2S_4$ at low temperatures.

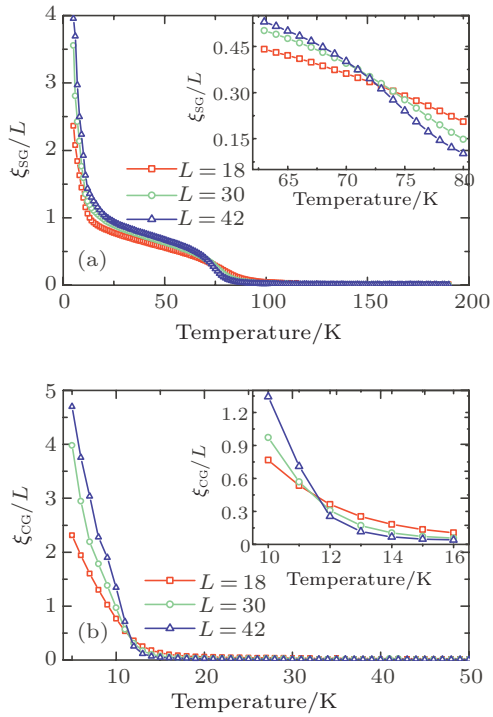


Fig. 2. (color online) (a) Spin glass correlation length versus temperature for the systems with different sizes. The inset shows the crossing details near the transition temperature. (b) Chiral glass correlation length versus temperature. The inset shows the crossing details.

Furthermore, we use the finite-size scaling method to obtain the critical temperatures and the exponents of both SG and CG transitions. It is suggested that the correlation lengths satisfy the following scaling function $\xi_L/L = F[(TL)^{1/\nu}(1 - T_c/T)]$, where T_c and ν are the critical temperature and the exponent, respectively.[24,27,28] We calculate the CG correlation lengths for the systems with $L = 12, 18, 24, 30, 42, 48, 60$. For the system with $L > 60$, the Monte Carlo sweeps needed to relax the system to the equilibrium state significantly increase, which is beyond the computing power of our computers. The scaled SG and CG correlation lengths are shown in Fig. 3. Obviously, the correlation lengths well obey the scaling function, and our scaling result gives $T_c = 11.0$ K, $\nu_c = 1.10$ and $T_g = 73.0$ K, $\nu_g = 1.40$ for the CG and the SG transitions, respectively. The critical exponent ν_c is larger than $\nu_c = 0.83$ for the system with only the nearest neighbor coupling.[30] Moreover, we calculate the critical temperatures and the exponents of both SG and CG transitions for the systems with $k_u = 0, 2.0, 4.0, 6.0, 8.0$, and list them in Table 1 together with T_l and T_h obtained from the specific heat calculation. It is noticed that both critical temperatures coincide with each other, which manifests that the low-temperature peak of the specific heat is caused by the CG transition. For $k_u = 0$, the critical temperatures for both SG and CG transitions are very close. As k_u increases, they gradually separate from each other.

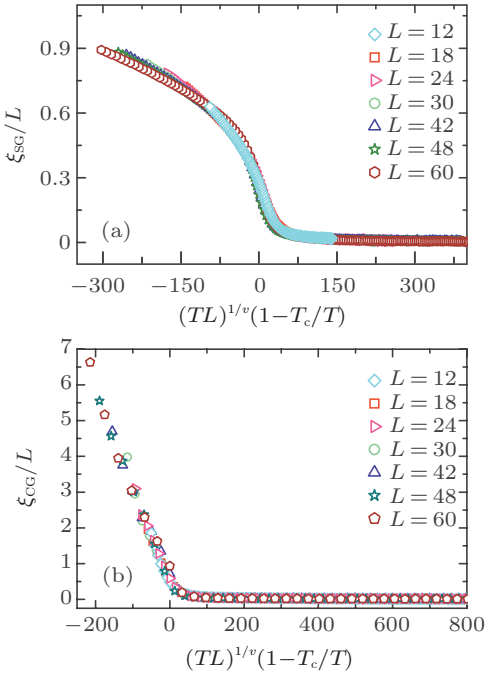


Fig. 3. (color online) The scaled (a) SG and (b) CG correlation lengths for the systems of different sizes.

Table 1. Critical temperature T_c (T_g) and exponent ν_c (ν_g) for the CG (SG) transition of the systems with $k_u = 0, 2.0, 4.0, 6.0, 8.0$. The T_c and T_g are obtained by finite-size analysis, while T_l and T_h are obtained from the specific heat calculation. The ranges of errors are given in the brackets.

k_u/meV	T_c/K	T_l/K	ν_c	T_g/K	T_h/K	ν_g
0	62.0 (±0.5)	62.0	1.10 (±0.01)	60.0 (±0.4)	62.0	1.40 (± 0.02)
2.0	53.0 (±0.5)	53.0	1.10 (±0.01)	53.0 (±0.8)	64.0	1.40 (±0.04)
4.0	40.0 (±0.4)	40.0	1.10 (±0.01)	69.0 (±0.5)	71.0	1.40 (±0.03)
6.0	26.0 (±0.5)	26.0	1.10 (±0.01)	72.0 (±0.6)	75.0	1.40 (±0.02)
8.0	11.0 (±0.3)	11.0	1.10 (±0.01)	73.0 (±0.5)	76.0	1.40 (±0.02)

The underlying physics of the CG transition can be interpreted as follows. The chirality represents the handedness of two neighbor spins, and it is easily affected by the anisotropy interaction since the chirality is the cross product of two vectors. Only when the relative direction between spins is frozen does the chirality have a nonvanishing value. When the competition between anisotropy and exchange interactions becomes steady, the chirality is nonvanishing and hence the CG phase transition occurs. That is why two critical temperatures coincide with each other. For $k_u = 0$, the chirality is only dominated by the neighbor couplings. When the spins become frozen, the cross product of two spin vectors also becomes nonvanishing. Therefore, the critical temperatures of the CG and the SG transitions nearly coincide. For larger k_u, the chirality is more easily affected by anisotropy compared with the spins, therefore, the CG critical temperature is much lower than the SG one. It is noticed that both the CG and the SG correlation lengths increase exponentially only when the temperature is below T_l, which is consistent with the results of the BKT model.[19]

As we have mentioned, the double peaks of the specific heat arise from the competition of magnetic interactions. One may wonder how the anisotropic interaction affects the specific heat. Figure 4(a) shows the specific heat as a function of temperature for different k_u. For $k_u = 0$, the specific heat only peaks at $T = 62$ K, indicating the existence of the Curie–Weiss temperature. As k_u increases, the single peak splits into double peaks. The peak at low temperature significantly shifts to lower temperature while the high-temperature peak remains nearly unchanged. This indicates that the specific heat peak at low temperature is mainly caused by the anisotropic interaction.

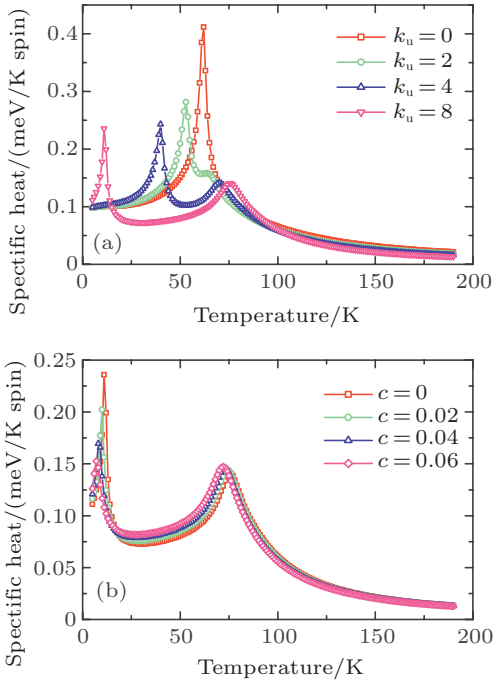


Fig. 4. (color online) (a) Specific heat versus temperature for different k_u. (b) Specific heat versus temperature for the systems with different concentrations of nonmagnetic impurities. The lattice size L is 42.

In experiments, researchers have observed that the specific heat peaks shift slightly to lower temperatures with less magnitudes as the nonmagnetic Zn impurities substitute Ni.[11–13] To test whether our model can describe this behavior, we calculate the specific heat of the systems with different concentrations (c) of nonmagnetic impurities. Figure 4(b) shows that the specific heat peak slightly shifts to lower temperatures compared to that of the undoped system. Note that the magnitude of the specific heat peak at low temperature is slightly reduced as the impurity concentration increases. These properties agree well with the results of the experiments.[11–13]

It would be interesting to envisage the spin configurations below the freezing temperature for the system with anisotropic interaction. Figure 5 shows the snapshots of the spin configurations at 5 K. For the spin system with $k_u = 0$, the spins rotate away from each other and form a 120° spiral ground

state. This is consistent with the results of the previous studies. However, for the system with $k_u = 8.0$, the regular spirals are distorted by the anisotropic interaction and a spin disordered state emerges.

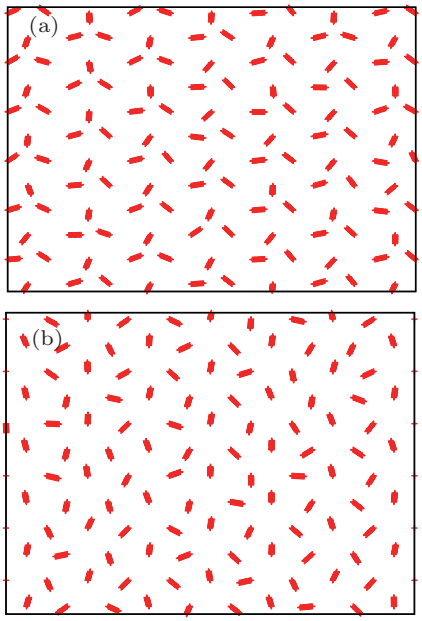


Fig. 5. (color online) Snapshots of spin configurations at 5 K: (a) $k_u = 0$, (b) $k_u = 8.0$.

Experimentally, the first nearest neighbor coupling was suggested to be ferromagnetic and considerably smaller than the third nearest neighbor coupling, which was antiferromagnetic.[7,31] The anisotropy of single spin $D(S_i^z)^2$ was suggested to be very small. However, this does not invalidate our model since an anisotropy of the inter-spin interaction is included in our model. Therefore, we calculate the specific heat for ferromagnetic J_1 and antiferromagnetic J_3 and show it in Fig. 6. In our simulations, J_3 is considerably larger than J_1 and J_2 is set to be zero, which is close to the case suggested by the experiments. As can be seen, two peaks emerge clearly in the specific heat. The low-temperature peak shifts to lower temperatures as J_1 decreases. Therefore, our model can describe the specific heat qualitatively.

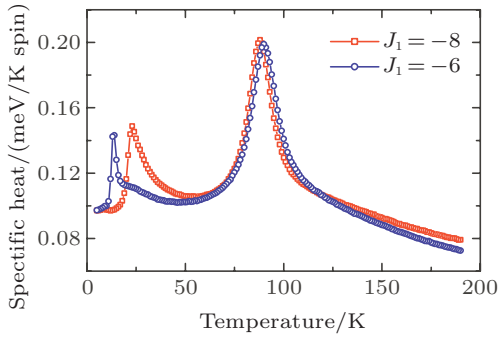


Fig. 6. (color online) Specific heat versus temperature for ferromagnetic J_1 and antiferromagnetic J_3, where J_2, J_3, and k_u are set to be 0 meV, 13.0 meV, and 14.0 meV, respectively.

3.1. Comparison with other models

Generally, the double peaks of the specific heat can be caused by two kinds of interactions with competition. Numerical studies have verified that only one peak emerges when only the neighbor couplings are considered.[17] So the second kind of interaction must be different from the Heisenberg type of $J_{ij}\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j$. Recently, Stoudenmire *et al.* proposed that a biquadratic exchange could induce a competition between interactions and hence resulted in the double peaks of the specific heat.[18] Using the heat bath and the parallel tempering methods, we simulate the specific heat for the spin system with the biquadratic interaction. The Hamiltonian of the system in an external field can be written as

$$H = J_{ij}\sum_{i,j}\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j - k_u\sum_{ij}(\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j)^2 - \sum_i \boldsymbol{\sigma}_i \cdot \boldsymbol{h}. \quad (7)$$

Figure 7(a) shows the specific heat versus temperature for the frustrated spin system with a different magnitude of the biquadratic interaction. Clearly, double peaks emerge in the specific heat. However, the low temperature peak is very susceptible to the biquadratic interaction. For large k_u, the peak is rounded and low. As k_u decreases, the peak shifts to lower temperatures and becomes very sharp. The full width at half maximum for the low-temperature peak is less than 2 K. The sharp transition is caused by the abrupt descent of the spin energy, as shown in Fig. 7(b). Moreover, the specific heat increases again when the temperature is below the freezing temperature. This means that the biquadratic interaction is too powerful to compete with the neighbor couplings. Experimentally, the specific heat measurement on $NiGa_2S_4$ reveals that the two peaks have nearly the same magnitude.[7] However, for $k_u = 5.5$, the magnitude ratio of the low-temperature peak to the high-temperature one exceeds 20, which is unreasonable. Furthermore, the calculated magnetization reveals that the system has a sharp transition at T_l and the corresponding plateau is absent. This is not consistent with the gradual increase and subsequent plateau of magnetization observed in the experiments. We also calculate the specific heat for the model of biquadratic interaction with ferromagnetic J_1 and antiferromagnetic J_3 suggested by the experiments. However, the specific heat with double peaks cannot be reproduced well.

The double peaks of the specific heat have also been observed experimentally in a dipolar-interacting system.[32] The dipolar interaction is very different from the superexchange interaction and may cause the competition between magnetic interactions. We calculate the specific heat of the system with both neighbor exchange and dipolar interactions. Although the double peaks emerge for the system, they depend strongly on the size of the system, and the magnitude of the dipolar interaction required is comparable to the superexchange interactons, which is unreasonable, hence, the contribution of the dipolar interaction can be ruled out.

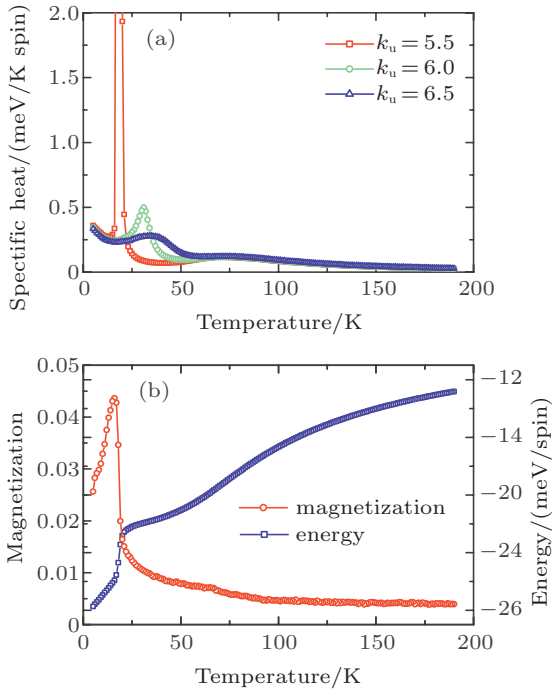


Fig. 7. (color online) Simulation results of the model with biquadratic interaction, where J_1, J_2, and J_3 are set to be 8.4 meV, 0.3 meV, and 4.1 meV, respectively, and the lattice size L is 42. (a) Specific heat versus temperature for different magnitudes of the biquadratic interaction, (b) magnetization and average energy versus temperature for the system with $k_u = 5.5$. The magnetic field is applied along the y axis with a magnitude of $h = 0.5$ meV.

Can the Dzyaloshinsky–Moriya interaction lead to the double peaks of the specific heat? As it is assumed to cause the spiral spin structure in the chiral magnets,[33] i.e., MnSi and FeSi. Our results reveal that only one peak of specific heat arises for the system with the Dzyaloshinsky–Moriya interaction. The peak shifts to higher temperature as the magnitude of the Dzyaloshinsky–Moriya interaction increases. This manifests that the Dzyaloshinsky–Moriya interaction cannot compete effectively with the superexchange interaction and therefore, does not lead to the double peaks of the specific heat.

4. Conclusion

We have investigated the magnetic properties of the 2D geometrical frustrated system $NiGa_2S_4$ by Monte Carlo simulations. By adding an inter-spin anisotropic interaction term, the double peaks of the specific heat and the magnetization behavior are well reproduced in the simulations. Both SG and CG correlation lengths increase as the temperature decreases. The nonmagnetic impurity effect is also investigated by simulations, and the results are consistent with the experimental ones. We hope that our study can be helpful for the future investigations of $NiGa_2S_4$.

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