Effects of Frustration and Dzyaloshinskii–Moriya Interaction on the Spin-1/2 Anisotropic Heisenberg Antiferromagnet with the Application to La$_2$CuO$_4$

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The magnetic properties of the 2D anisotropic antiferromagnetic spin-1/2 Heisenberg model with Dzyaloshinskii–Moriya interaction and in-plane frustration are studied. The method of spin Green’s functions within the framework of Tyablikov’s random-phase-approximation decoupling scheme is used to derive expressions for the spin-wave spectrum, sublattice magnetization, and transition temperature. Based on these expressions, a detailed analysis of the influence of varying values of model parameters on their magnetic properties is conducted. The model is also applied to the high-$T_c$ superconducting parent compound La$_2$CuO$_4$ and the results compared with available experimental data.

1. Introduction

Though the high-$T_c$ superconducting parent compound La$_2$CuO$_4$ has been thoroughly examined in past few decades, the interest in this system, being a good example of the quasi-2D antiferromagnetic (AFM) Heisenberg model, seems to be inexhaustible. Many models have been proposed to describe the magnetic behavior within the CuO$_2$ planes, having in mind that it may be associated with the mechanism of the high-$T_c$ superconductivity in this compound. Beside the nearest-neighbor (NN) and next-nearest-neighbor (NNN) exchange interaction, it is often suggested that the description without taking into account the Dzyaloshinskii–Moriya (DM) interactions,[1,2] arising from spin–orbit coupling, may be considered as incomplete.[3–11] Namely, in La$_2$CuO$_4$, due to the spin–orbit couplings arising from the small orthorhombic distortion below the structural tetragonal–orthorhombic transition temperature $T_u = 530$ K, CuO$_2$ planes exhibit a weak ferromagnetic moment, i.e., all spins cant out of the CuO$_2$ plane by a small angle $\theta$. However, this is not a common feature of the copper oxide layers as in the absence of the orthorhombic distortion DM interaction does not emerge (in Sr$_2$CuO$_2$Cl$_2$, e.g.).[12–14] Special interest in DM interaction has also grown due to the possibility of measuring its direction and strength by making use of synchrotron radiation, for the

specific class of materials to which La$_2$CuO$_4$ belongs.[15] The anisotropy introduced via DM interaction, together with the NN-exchange interaction anisotropy,[16] is responsible for the existence of the long-range antiferromagnetic order below some nonzero temperature in case of the considered 2D Heisenberg model. Besides these anisotropies, a great impact on the magnetic behavior of the studied low-dimensional system has significant in-plane frustration, which is introduced into the model by taking into account NNN-exchange interaction. Namely, though the system is not geometrically frustrated, within the CuO$_2$ planes, there exists the conflict of spin orientation due to the antiferromagnetic NNN exchange bonds. The intention to thoroughly analyze the influence of in-plane frustration on the magnetic properties of the system is corroborated by the fact that frustration is strongly pronounced in 2D quantum magnets,[17,18] though it is often not taken into account.[15,6,8] Our earlier results[19–22] also suggest that the model with the NNN interaction gives predictions in better agreement with the experimental data. Besides the aforementioned, the choice of the dominant interactions is supported by the fact that these interactions, especially NN, NNN, and DM interaction, but also spin anisotropy to a lesser extent, show a distinctive pressure dependence,[23,24] enabling one to affect the phase transition temperature by applying high pressures on the system. Having this in mind, the purpose of this article will be to study the 2D antiferromagnetic Heisenberg model with the uniaxial $XXZ$ spin anisotropy, DM interaction, and in-plane frustration included. We shall use the spin Green’s function method within the framework of Tyablikov’s random-phase-approximation (RPA) decoupling scheme. To facilitate the calculations, we shall make use of the coordinate frame rotation method where the quantization axis coincides with the classical moment of the ground state.[25–27] The obtained magnetic properties will be studied in detail to describe quantitatively the role of different model parameters. Having at our disposal experimental data for the magnetization temperature dependence in La$_2$CuO$_4$, we shall apply our model to this compound, as an additional check of our results.

The article is organized as follows. In Section 2, we present the model Hamiltonian with the dominant exchange interactions and conduct the coordinate transformation which facilitates the further calculations. In Section 3, the main expressions for the quantities to be analyzed are derived by making use of spin Green’s function method within the RPA scheme.

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In Section 4, the detailed analysis of the numerical results with the emphasis on the comparison of the influence of different system parameters on the studied magnetic properties is given, followed by the application of our results to La$_2$CuO$_4$ and comparison with available experimental data. The conclusions are briefly stated in Section 5.

2. Model Hamiltonian

We consider the $S = 1/2$ anisotropic Heisenberg antiferromagnet on the rectangular lattice (Figure 1a), with the dominant interactions comprising the following Hamiltonian.

$$
\hat{H} = J \sum_{n_x, \delta} \left( \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(b)}_{n_x + \delta} + \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(b)}_{n_x + \delta_1} + \alpha \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(b)}_{n_x + \delta} \right) - D \sum_{n_x, \delta_1} \left( \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(b)}_{n_x + \delta_1} - \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(b)}_{n_x + \delta} \right) + \frac{J_2}{2} \sum_{n_x, \delta_2} \left( \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(a)}_{n_x + \delta_2} + \hat{S}^{(a)}_{n_x} \cdot \hat{S}^{(a)}_{n_x + \delta_2} \right)
$$

(1)

We observe that the Hamiltonian contains the NN DM interaction. The DM vector $D$ is chosen to point along $x$-axis ($D = (D, 0, 0)$), as its $y$-component would yield the term which does not contribute to the gap but only has a slight influence on the spin-wave spectrum, wherefore it can be neglected.\[28,29\] As the DM interaction itself yields continuous symmetry in the ground state and therefore precludes the existence of the long AFM order at nonzero temperatures, in 2D models, it is common to introduce in Hamiltonian the symmetric pseudodipolar interaction (often denoted by $\Gamma$), which lifts that symmetry. However, one can achieve long-range order also in the case of zero pseudodipolar interaction if the spin anisotropy is included in Hamiltonian,\[5\] as we do in Equation (1). Namely, the first term in Equation (1) represents the NN anisotropic exchange interaction characterized by the exchange parameter $J$, with the anisotropy parameter denoted by $\alpha$. We take parameter $\alpha$ to be slightly smaller than unity, meaning that the magnetic moments are ordered in the $XY$ plane (easy-plane antiferromagnetism). Finally, we include the NNN interaction, defined by exchange integral $J_2$. Hereafter we shall define the fundamental energy scale by the NN-exchange interaction $J$ and use dimensionless ratios, namely frustration ratio $\delta = J_2/J$ and DM ratio $d = D/J$, parametrizing the relative strength of DM interaction.

To determine the canting angle $\theta$ we conduct the $180^\circ$ rotation of the $b$ sublattice spins around $Z$-axis, which presents the use of the extended translational symmetry method of effectively lowering the number of sublattices in the system.\[30,31\] Minimizing the classical ground state energy, we obtain

$$
\tan 2\theta = \frac{2d}{1 + \alpha}
$$

(2)

It is interesting to notice that the presence of NNN interaction in the Hamiltonian does not influence the magnitude of the canting angle.

The calculations are easier to be carried out if we pass from the initial crystallographic coordinate system to local coordinate systems in which $Z$-axes coincide with the direction of sublattice magnetization (Figure 1b). For the sake of simplicity, we take that the spins are initially antiferromagnetically aligned along the $Y$-axis.

The transformation matrix for the $a/b$ sublattice reads

$$
\begin{bmatrix}
\hat{\xi}^{a(b)} \\
\hat{\xi}^{a(b)} \\
\hat{\xi}^{a(b)}
\end{bmatrix} =
\begin{bmatrix}
1 & 0 & 0 \\
0 & \sin \theta & \pm \cos \theta \\
0 & \mp \cos \theta & \sin \theta
\end{bmatrix}
\begin{bmatrix}
\hat{S}^{a(b)} \\
\hat{S}^{a(b)} \\
\hat{S}^{a(b)}
\end{bmatrix}
$$

(3)

where the upper (lower) sign stands for $a$ ($b$) sublattice.

The Hamiltonian (Equation (1)) in terms of the new operators $\hat{S}$ reads

$$
\hat{H}/J = \sum_{n_x, \delta_1} \left[ A \left( \hat{S}^{a(b)}_{n_x} \cdot \hat{S}^{a(b)}_{n_x + \delta_1} + H.c. \right) + C \hat{S}^{a(b)}_{n_x} \cdot \hat{S}^{a(b)}_{n_x + \delta_1} \right] + M(\hat{S}^{a(b)}_{n_x} \cdot \hat{S}^{a(b)}_{n_x + \delta_1} + H.c.) + \frac{\lambda}{2} \sum_{n_x, \delta_2} \left[ \frac{1}{2} \left( \hat{S}^{a(b)}_{n_x} \cdot \hat{S}^{a(b)}_{n_x + \delta_2} + H.c. \right) + \hat{S}^{a(b)}_{n_x} \cdot \hat{S}^{a(b)}_{n_x + \delta_2} \right]
$$

(4)

where we use the following notation...
A = \frac{1}{2} - (1 + \alpha) \cos^2 \theta - \frac{d \sin 2\theta}{4} \\
C = \alpha - (1 + \alpha) \cos^2 \theta - d \sin 2\theta \\
M = \frac{(1 + \alpha) \cos^2 \theta}{4} + \frac{d \sin 2\theta}{4}

By making use of the transformation (Equation (3)) we have effectively described the system by the easy-axis model. Transformed Hamiltonian (Equation (4)) will present the starting point in our calculations.

3. Spin-Wave Spectrum, Magnetization, and Related Quantities

To derive the spin-wave spectrum, we use the spin Green’s function method within the framework of Tyablikov’s decoupling approximation. Due to the structure of Hamiltonian, we obtain the following system of four equations for Green’s functions $\langle \langle \hat{S}^+(a) | \hat{S}^-(a) \rangle \rangle_{k,k'} \equiv \hat{G}_{aa}^{+}$ (explicit dependence on wave vector and energy will be hereafter omitted for brevity).

$$(\omega + Q_k) G_{a}^{+} - P_k G_{a}^{-} - R_k G_{a}^{0} = \frac{i}{2 \pi} \langle \langle \hat{S}^{+(a)} | \hat{B} \rangle \rangle$$

$$(\omega - Q_k) G_{a}^{-} + P_k G_{a}^{+} + R_k G_{a}^{0} = \frac{i}{2 \pi} \langle \langle \hat{S}^{-(a)} | \hat{B} \rangle \rangle$$

$$(\omega - Q_k) G_{a}^{+} - P_k G_{a}^{-} + (\omega + Q_k) G_{a}^{0} = \frac{i}{2 \pi} \langle \langle \hat{S}^{+(b)} | \hat{B} \rangle \rangle$$

$$(\omega + Q_k) G_{a}^{-} + P_k G_{a}^{+} + (\omega - Q_k) G_{a}^{0} = \frac{i}{2 \pi} \langle \langle \hat{S}^{-(b)} | \hat{B} \rangle \rangle$$

where

$$Q_k = J[2z_1 C + z_2 \lambda (1 - \gamma_k^{(2)})] \sigma$$

$$P_k = 2z_1 J A_k^{(1)} \sigma$$

$$R_k = 2z_1 J M_k^{(1)} \sigma$$

In Equation (7), $\sigma = \langle \hat{\sigma}^z \rangle$ denotes the sublattice magnetization (in the absence of the external magnetic field, $\sigma^{(0)} = \sigma^{(0)} = \sigma$), $z_{1,2} = 4$ defines the number of NN and NNN, whereas quantities $\gamma_k^{(1,2)}$ present geometric factors for NN and NNN, respectively, and are given by

$$\gamma_k^{(1)} = \cos \frac{k_x a_0}{2} \cos \frac{k_y b_0}{2}$$

$$\gamma_k^{(2)} = \frac{1}{2} (\cos k_x a_0 + \cos k_y b_0)$$

where $a_0$ and $b_0$ denote the lattice parameters.

Spin-wave spectrum obtained from the system of Equation (6) for the in-plane mode is given by

$$\omega_{1k} = z_1 \sigma \sqrt{C + \lambda (1 - \gamma_k^{(2)}) - 2 A_k^{(1)} - 4 M_k^{(1)}}$$

whereas for the out-of-plane mode it reads

$$\omega_{2k} = z_1 \sigma \sqrt{C + \lambda (1 - \gamma_k^{(2)}) + 2 A_k^{(1)} - 4 M_k^{(1)}}$$

The corresponding zone-center ($k = 0$) energy gaps are

$$\omega_{1/2k=0} = z_1 \sigma \sqrt{(C + 2A) - 4M^2}$$

To determine sublattice magnetization from the well-known formula

$$\sigma = \frac{1}{2} \frac{1}{N} \sum_k \langle \langle \hat{S}_z \hat{\mathcal{S}}^+ \rangle \rangle_k$$

we calculate Green’s function $\langle \langle \hat{S}_z \hat{\mathcal{S}}^{+} \rangle \rangle_{k,k'} \equiv \hat{G}_{zz}^{+}$ from the system (Equation (6)), taking for the operator $\hat{B} \equiv \hat{S}^{\pm}$. Using the standard procedure to obtain the correlation function $\langle \langle \hat{S}_z \hat{\mathcal{S}}^+ \rangle \rangle_k$, we derive the following expression for the sublattice magnetization

$$\sigma = \frac{1}{2} \frac{1}{N} \sum_k \left( \frac{A_k - C_k}{\sqrt{(C_k - A_k)^2 - M_k^2}} \coth \frac{\beta \omega_{1k}}{2} \right)^{-1}$$

where

$$A_k = 2J A_k^{(1)}$$

$$C_k = |C + \lambda (1 - \gamma_k^{(2)})|$$

$$M_k = 2J M_k^{(1)}$$

and $\beta = 1/k_B T$. The temperature at which magnetization vanishes is obtained from Equation (12), using the expansion $\coth x \sim 1/x$ as valid at high temperatures, wherefrom

$$k_B T_N = \frac{1}{2N} \sum_k \left( \frac{A_k - C_k}{(C_k - A_k)^2 - M_k^2} - \frac{A_k + C_k}{(C_k + A_k)^2 - M_k^2} \right)^{-1}$$

We shall now analyze these results.

4. Analysis of Results

We shall start the analysis with the numerical calculation of the reduced zero-temperature energy gaps in the long-wavelength limit $\omega_{1/2k=0}/J$ as a function of the introduced model parameters. The range of parameter values is chosen, having in mind the parameter sets based on the experimental measurements describing the real magnetic systems (La$_2$CuO$_4$ e.g., as will be evident later). In Figure 2 we present the energy gaps’ dependence on the DM parameter $d$, for different values of the other two parameters $\alpha$ and $\lambda$. By inspection of Figure 2, we infer that for the nonzero values of $d$ both spin-wave modes possess a gap, whereas when $d$ vanishes, the in-plane mode (almost independent on $d$) possesses a gap of the magnitude
$\omega_{1k=0} \propto z_1 \sigma_1 \sqrt{1 - \alpha}$ and the out-of-plane mode (almost linear in $d$) becomes gapless, i.e., Goldstone mode appears. These observations agree with the ones from a study by Tabunshchyk et al.,\cite{5} suggesting that the NNN interaction does not change qualitatively the energy-gap behavior. However, to describe quantitatively the influence of the frustration parameter $\lambda$ we here conduct a more detailed analysis by the comparison of the influence of the parameters $\lambda$ and $\alpha$ on the studied dependance. As shown, the influence of the NNN interaction parameter $\lambda$ on the nonvanishing gap is much smaller than the influence of the anisotropy parameter $\alpha$. Similar observations can be made based on the energy-gap dependance on anisotropy parameter $\alpha$, shown in Figure 3. Contrary to the previous case, the in-plane mode is the one which, in the case of the isotropic model ($\alpha = 1$), becomes gapless, whereas the out-of-plane gap remains finite: $\omega_{2k=0} \propto z_1 \sigma_1 \sqrt{\cos 3\theta + (d^2 + (d^2 - 1) \cos 2\theta) \sin \theta}$. The influence of parameter $\lambda$ on the nonvanishing gap is less prominent than the influence of DM parameter. Finally, we consider the energy-gap dependance on frustration parameter $\lambda$ (Figure 4). In the considered range of $\lambda$ both energy gaps are finite and almost independent on that parameter.

It is further interesting to examine the dependence of transition temperature on various parameters of the system. In Figure 5, 6, and 7 we present the reduced Néel temperature dependence on parameters $d$, $\alpha$, and $\lambda$, respectively, as a function of several values of the other two parameters. Figure 5 shows that with the increase in DM parameter $d$, the transition temperature also increases. Due to the fact that the Goldstone mode in the spin-wave spectrum appears when $d$ vanishes, transition temperature drops to zero in that limit. In Figure 6, we can see that the decrease in parameter $\alpha$ (i.e., the increase in spin anisotropy)
increases the transition temperature. On the other hand, \( T_N \) becomes zero for \( \alpha = 1 \). From Figure 7, we see that the increase of parameter \( \lambda \) yields a decrease in transition temperature, as the in-plane frustration disorders the system. The transition temperature is however not suppressed to zero, as in the considered parameter range energy gaps remain finite.

We now calculate the zero-temperature antiferromagnetic order parameter \( \sigma_0 \), by making use of Equation (13). The behavior of \( \sigma_0 \) as a function of parameter \( d \) for several value of parameters \( \alpha \) and \( \lambda \) is shown in Figure 8. The analogous dependencies of the zero-temperature magnetization on parameters \( \alpha \) and \( \lambda \) for different values of two other Hamiltonian parameters are shown in Figure 9 and 10. By inspection of Figure 8–10, one can see that the increase in DM parameter tends to increase \( \sigma_0 \), whereas the increase of either the frustration parameter \( \lambda \) or parameter \( \alpha \) tends to decrease the order parameter. It should be noted that though the transition temperature of the isotropic model (\( \alpha = 1 \)) and the model without DM interaction (\( d = 0 \)) equals zero, the sublattice magnetization at \( T = 0 \) K is finite in both cases, i.e., the long-range order exists at zero temperature but is destroyed by the thermal fluctuations for nonzero temperatures. Finally, the analysis of Figure 8 and 9 shows that the influence of the frustration parameter \( \lambda \) on zero-temperature magnetization is more pronounced than in the case of energy gaps.

The influence of the frustration, DM interaction, and spin anisotropy on sublattice magnetization in the wide temperature range \( 0 \leq T \leq T_N \) is shown in Figure 11. Due to the self-consistency of Equation (13), the iterative procedure has to be applied. The value of the NN-exchange interaction \( J \) is chosen to be 100 meV.

To conclude the general discussion, in Figure 12, we show the dependance of the canting angle \( \theta \) on system parameters \( d \) and \( \alpha \).

As it was stated before, the high-\( T_c \) superconducting parent compound La\(_2\)CuO\(_4\) can be considered as a quasi-2D system\(^{[5,6]} \) described by the Hamiltonian (Equation (1)), wherefore, as a test of our results, we shall apply the obtained expressions to that system. To determine model parameters, we use experimental data for the gaps of the in-plane and out-of-plane polarized magnons at 100 K,\(^{[33]} \) with magnitudes \( \omega_{1k=0} = 2.3 \) meV and \( \omega_{2k=0} = 5 \) meV, respectively. Using the self-consistent procedure to determine \( J \) and \( \alpha \), and choosing other two parameters to reproduce the correct value for Néel temperature, we obtain

\[
J = 110.251 \text{ meV}, \quad \alpha = 0.99987 \\
\text{d} = 0.035, \quad \lambda = 0.09
\]  

Figure 7. Reduced transition temperature versus parameter \( \lambda \) for different values of parameters \( \alpha \) and \( d \).

Figure 8. Sublattice magnetization \( \sigma \) at \( T = 0 \) K versus parameter \( d \) for different values of parameters \( \alpha \) and \( \lambda \). Dots emphasize the nonzero values of \( \sigma_0 \) for \( d = 0 \).

Figure 9. Sublattice magnetization \( \sigma \) at \( T = 0 \) K versus parameter \( \alpha \) for different values of parameters \( d \) and \( \lambda \). Dots emphasize the nonzero values of \( \sigma_0 \) for \( \alpha = 1 \).

Figure 10. Sublattice magnetization \( \sigma \) at \( T = 0 \) K versus parameter \( \lambda \) for different values of parameters \( d \) and \( \alpha \).
wherefrom the canting angle in La$_2$CuO$_4$ reads $\theta \approx 0.017$ rad. While the parameters $\alpha$ and $d$ are of the same order of magnitude as those proposed in the study by Tabunshchik et al.,[5] the super-exchange value $J$ is lower than the one quoted in the same study,[5] the latter leading to significant overestimation (over 30%) of the transition temperature.

An important check of our results is the comparison with the magnetization temperature dependence obtained by neutron scattering reported in a study by Keimer et al.[34] As shown in Figure 13, our model compares favorably with the experiment. Finally, we present the temperature dependence of the energy gaps of both modes in the excitation spectrum (Figure 14). It is shown that our results are in agreement with the experimental data given in the study by Keimer et al.[33]

5. Conclusion

We study the 2D anisotropic Heisenberg antiferromagnet, with the DM interaction and in-plane frustration included. By making use of spin Green’s function method within Tyablikov’s decoupling approximation, we obtain the expressions for the spin-wave energy gaps, sublattice magnetization, and transition temperature. A detailed comparison of the influence of model parameters on the magnetic properties of the system is conducted. We conclude that these parameters have the opposite impact on the long-range antiferromagnetic order, whereby the increase of the frustration parameter $\lambda$ and spin anisotropy parameter $\alpha$ destabilizes the system, whereas the increase of the DM parameter $d$ stabilizes the system.

We also apply our results to the high-$T_c$ superconducting parent compound La$_2$CuO$_4$, a layered copper oxide which has been both theoretically and experimentally investigated in the past decades with undiminished interest. The magnetization curve and energy-gap temperature dependance based on our calculations agree favorably with the experimental data. Therefore, though the use of the frame rotation method for the antiferromagnet in the transverse magnetic field is questionable,[33] its application for the antiferromagnet in the absence of the magnetic field, described by the Hamiltonian (Equation (11)), seems justified. However, new experimental results, such as measurements of the strength and direction of DM interaction in La$_2$CuO$_4$ as well as high-pressure-induced variation of the in-plane frustration, would serve as a further check of our results.

Figure 11. Sublattice magnetization $\sigma$ versus temperature $T$, for the NN interaction $J = 100$ meV. The lines in color are denoted by the value of the parameter by which they differ from the black line.

Figure 12. Spins’ tilt angle $\theta$ versus parameters $d$ and $\alpha$.

Figure 13. Square of the relative magnetization $(\sigma/\sigma_0)^2$ versus temperature $T$. Full circles denote the experimental data from a study by Keimer et al.[34] Solid line represents the theoretical result based on Equation (13).

Figure 14. In-plane (dashed curve) and out-of-plane (full curve) zone-center energy gaps $\omega_{1/2k-q}$ versus relative temperature $T/T_N$. Full circles denote the experimental data from a study by Keimer et al.[33] Solid lines represent the theoretical result based on Equation (11).
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Conflict of Interest
The authors declare no conflict of interest.

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