Temperature dependence of sublattice magnetization in quasi-two-dimensional $S=\frac{1}{2}$ cuprate antiferromagnets: Green's function approach

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The study of temperature dependence of the sublattice magnetization in the quasi-two-dimensional $S=\frac{1}{2}$ cuprate antiferromagnets was inspired by the existing experimental evidence concerning sublattice magnetization temperature dependence, both in the vicinity of absolute zero for the high-T_C parent compound La_2CuO_4 and in the whole temperature region from absolute zero to the Néel temperature (T_N) for La_2CuO_4 and $YBa_2Cu_3O_{6.15}$. Using the spin Green's functions method for the quantum Heisenberg antiferromagnetic model, we obtain the results which show excellent agreement with the experimental data, proving that in the wide temperature region (from 0 to T_N), this method is more appropriate than the harmonic spin-wave theory and Schwinger-boson mean-field theory.

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I. INTRODUCTION

Since their discovery,[1] high-T_C superconductors have been in the highlight of the scientific interest. Assuming that the knowledge of the behavior of these compounds in undoped regime could help in finding a satisfactory explanation of the mechanism of their transition to superconductive state, great attention has also been paid to the examination of the parent compounds of this type of superconductors. Experiments performed have shown that their common feature, and thus potentially crucial for the understanding of the transition mechanism, is the presence of one or more adjacent CuO_2 planes, within which there exists strong superexchange interaction, establishing the antiferromagnetic long-range order. Hence, much of the theoretical study has been devoted to the attempt of giving a valid picture of the dominant interactions within and among these planes, leading to the possibility of determining many other magnetic and thermodynamic properties of these compounds.[2,3]

Our research has been primarily focused on the magnetic properties of the high-T_C parent compounds, especially La_2CuO_4, being a remarkable example of the two-dimensional (2D) $S=\frac{1}{2}$ Heisenberg antiferromagnet and therefore of a high theoretical importance.[4] As yet, we have studied in detail the spectrum of the elementary excitations and calculated Néel temperature for this compound,[5] obtaining the results in excellent agreement with the experimental spin-wave spectrum[6] and the measured value of the transition temperature.[7–9] Study of the Néel temperature (T_N) dependence on the in-plane spin anisotropy in La_2CuO_4 yielded an interesting observation concerning the existence of the long-range order in this compound. Namely, while in both orthorhombic and tetragonal phases of La_2CuO_4 the two-dimensional spin isotropic model shows zero T_N as expected due to Mermin-Wagner theorem,[10] in three dimensions, significant differences between the two phases occur. In orthorhombic phase, the spin isotropic model possesses finite T_N as expected, while in tetragonal phase, Néel temperature appears to be zero (Fig. 3. of Ref. 5). This fact is due to the cancellation of interactions among the ferro- and antiferromagnetically coupled nearest-neighbor spins in two adjacent planes in La_2CuO_4 and may be proven analytically using Bogoliubov's inequality,[11] as done by the authors in Ref. 12.

In addition, it is interesting to notice that this result concerning the absence of the long-range order in three-dimensional spin isotropic tetragonal La_2CuO_4 also follows from the Tyablikov's approximation[13] (Eq. (35) of Ref. 5), showing that this approximation, though simple, is sensitive even to the type of the structure of the compound and gives satisfying results in wide temperature region, wherefore is going to be used also throughout this Brief Report.

This Brief Report, being the continuation of our research of the magnetic properties of high-T_C parent compounds, is devoted to the study of the temperature dependence of magnetization in cuprate antiferromagnets La_2CuO_4 and $YBa_2Cu_3O_6$. This research was encouraged by the existing experimental evidence concerning sublattice magnetization temperature dependence, both in the vicinity of absolute zero for the high-T_C parent compound La_2CuO_4 (Ref. 14) and in the whole temperature region from absolute zero to the Néel temperature (T_N) for La_2CuO_4 and $YBa_2Cu_3O_{6.15}$.[9] Starting from the quantum Heisenberg $S=\frac{1}{2}$ antiferromagnetic model and using the spin Green's functions method, we obtain the results which show excellent agreement with the experimental data, proving that in the wide temperature region (from 0 to T_N), this method is more appropriate than the harmonic spin-wave theory and Schwinger-boson mean-field theory.

This Brief Report is organized as follows. The examination of the behavior of the La_2CuO_4 sublattice magnetization in the vicinity of absolute zero and its comparison to the experiment is given in Sec. II. In Sec. III, we present the sublattice magnetization temperature dependence for La_2CuO_4 and $YBa_2Cu_3O_6$ in the whole temperature region from absolute zero to the corresponding Néel temperatures and compare our results to those obtained by experiment as well as by other theories. Main conclusions are presented in Sec. IV.

II. BEHAVIOR OF MAGNETIZATION IN La_2CuO_4 IN THE VICINITY OF $T=0$ K

In order to examine the behavior of magnetization in La_2CuO_4 at the temperatures close to the absolute zero, we shall make use of the expression which we have already derived in Ref. [see Eq. (26), Ref. 5]. This expression, which was obtained by the spin Green's function method, using for the decoupling of the higher-order Green's functions the Tyablikov's approximation defined by the relation $\langle\langle \hat{S}^z_g \hat{S}^{\pm}_f | \hat{B} \rangle\rangle \overset{g\neq f}{\rightarrow} \langle \hat{S}^z_g \rangle \langle\langle \hat{S}^{\pm}_f | \hat{B} \rangle\rangle$, reads

$$\frac{1}{2\sigma} = \frac{1}{N}\sum_{\vec{k}} \frac{\epsilon^T(\vec{k})}{\omega^T(\vec{k})}\left(1 + \frac{2}{e^{E^T(\vec{k})/\theta} - 1}\right), \quad \theta = k_B T, \tag{1}$$

with limiting value

$$\sigma_0 \equiv \lim_{\theta\to 0} \sigma = \frac{1}{2}\left(\frac{1}{N}\sum_{\vec{k}} \frac{\epsilon^T(\vec{k})}{\omega^T(\vec{k})}\right)^{-1}, \tag{2}$$

where

$$E^T(\vec{k}) = J\sigma\omega^T(\vec{k}), \quad \omega^T(\vec{k}) = \sqrt{[\epsilon^T(\vec{k})]^2 - [I^T(\vec{k})]^2}, \tag{3}$$

$$\epsilon^T(\vec{k}) = z_2(1+\eta) + \frac{z_\perp}{2}(\lambda_{\perp_1} - \lambda_{\perp_2}) - \lambda_2\{z_2[1-\gamma_2(\vec{k})] + z_3[1-\gamma_3(\vec{k})]\} + \frac{z_\perp}{2}\lambda_{\perp_2}\gamma_\perp^{aa}(\vec{k}), \tag{4}$$

$$I^T(\vec{k}) = z_2\gamma_1(\vec{k}) + \frac{z_\perp}{2}\lambda_{\perp_1}\gamma_\perp^{ab}(\vec{k}). \tag{5}$$

Here, η represents the in-plane spin anisotropy; λ_2 $\left(\lambda_2 = \frac{J_2}{J}\right)$ describes next-nearest-neighbor in-plane exchange coupling; $\lambda_{\perp_{1/2}}$ $\left(\lambda_{\perp_{1/2}} = \frac{J_{\perp_{1/2}}}{J}, i=1,2\right)$ explicitly describe different interlayer couplings in the orthorhombic phase of La_2CuO_4; and J, J_2, and $J_{\perp_{1/2}}$ denote the corresponding exchange integrals. Taking into account experimental data suggesting that the excitation gap appearing in the spin-wave spectrum is very small,[6,14] we shall consider the isotropic magnet, i.e., in the previous expression, we shall put $\eta=0$. Quantities $\gamma_i(\vec{k})$ represent geometrical factors, which are in the orthorhombic phase given by Eqs. (A1)–(A5) of Ref. 5. The number of the second (third) nearest neighbors in the plane is denoted by $z_2=z_3=4$, while the number of the nearest neighbors in the two adjacent planes is denoted by $z_\perp=8$.

When $T\to 0$, the second term in the denominator of Eq. (1) is much less than the first one, so using expression (2) for the magnetization at $T=0$ K, we can write Eq. (1) in the following form:

$$\sigma = \sigma_0\left(1 + \frac{4\sigma_0}{N}\sum_{\vec{k}} \frac{\epsilon^T(\vec{k})}{\omega^T(\vec{k})} \frac{1}{e^{E^T(\vec{k})/\theta} - 1}\right)^{-1}. \tag{6}$$

Since the fraction in the denominator is much smaller than unity, the previous expression can be expanded into series. Keeping only the first two terms in the expansion, we obtain

$$\sigma(T) = \sigma_0 - \Delta\sigma(T), \quad \Delta\sigma(T) = \frac{4\sigma_0^2}{N}\sum_{\vec{k}} \frac{\epsilon^T(\vec{k})}{\omega^T(\vec{k})} \frac{1}{e^{E^T(\vec{k})/\theta} - 1}. \tag{7}$$

By inspection of Eq. (7), we observe that the most important contribution to the sum is due to the terms with small k (since for the spin isotropic magnets the Goldstone theorem is satisfied). Therefore, we can expand the functions appearing in the general term into series in powers of k, retaining only the terms of the zeroth and first orders, in order to obtain for the magnetization only the correction of the lowest power of temperature. Taking into account that the first correction in the expansion of $\epsilon^T(\vec{k})$ and $I^T(\vec{k})$ is proportional to k^2, we shall keep only the terms of the zeroth order. Thus, we obtain the expressions

$$\epsilon^T(\vec{k}=0) = I^T(\vec{k}=0) = z_2 + \frac{z_\perp}{2}\lambda_\perp^{(1)}, \tag{8}$$

$$\omega^T(\vec{k}\approx 0) = \sqrt{\epsilon^T(\vec{k}=0)}\sqrt{Ak_x^2a^2 + Bk_y^2b^2 + Ck_z^2c^2}, \tag{9}$$

where we use the following notation: $A=1+\lambda_{\perp_1}-6\lambda_2$, $B=1-\lambda_{\perp_2}-6\lambda_2$, and $C=\lambda_{\perp_1}-\lambda_{\perp_2}$. Substituting these expressions into Eq. (7), we obtain the correction to the magnetization:

$$\Delta\sigma(T) \approx \frac{4\sigma_0^2}{N}\sum_{\vec{k}} \frac{\epsilon^T(\vec{k}=0)}{\omega^T(\vec{k}\approx 0)} \frac{1}{e^{J\sigma_0\omega^T(\vec{k}\approx 0)/\theta} - 1}. \tag{10}$$

If we transform the sum into the integral, after shorter calculation, we arrive at the following expression:

$$\Delta\sigma(T) = \frac{2\int_0^\infty \frac{r}{e^r - 1}dr}{\pi^2\sqrt{\epsilon^T(\vec{k}=0)ABC}}\left(\frac{k_BT}{J}\right)^2, \tag{11}$$

where $r = \frac{J\sigma_0\epsilon^T(\vec{k}=0)\sqrt{Ak_x^2a^2+Bk_y^2b^2+Ck_z^2c^2}}{k_BT}$. Taking into account that by performing the integration in the numerator (the upper limit of the integral is infinite since we deal with very low temperatures) we obtain the Riemann zeta function $\sum_{n=1}^{\infty}\frac{1}{n^2} = \frac{\pi^2}{6}$, the final expression for the magnetization at low temperatures reads

$$\sigma(T) = \sigma_0 - DT^2, \quad D = \frac{1}{3\sqrt{\epsilon^T(\vec{k}=0)ABC}}\left(\frac{k_B}{J}\right)^2. \tag{12}$$

Thus, the first correction to the magnetization is proportional to T^2. The proportionality coefficient D is calculated for the following values of parameters J and λ_2 (Ref. 5): $J=141$ meV(±5%), $\lambda_2=0.0942$(±10%), and for several values of the interplanar interaction. The results of this calculation are presented in Table I.

Matsumara *et al.*[14] present the results of the experiment performed in order to obtain the temperature dependence of the magnetization in La_2CuO_4 in the vicinity of absolute zero. By using the nuclear magnetic resonance, it has been shown that in the temperature interval 10 K $< T <$ 100 K, *re-*

TABLE I. Dependence of quantity D on the difference of interplane couplings $\lambda_{\perp_1}-\lambda_{\perp_2}$.

$\lambda_{\perp_1}$	6×10^{-5}	2×10^{-4}	6×10^{-4}
$\lambda_{\perp_2}$	10^{-5}	10^{-4}	10^{-4}
$\lambda_{\perp_1}-\lambda_{\perp_2}$	5×10^{-5}	10^{-4}	5×10^{-4}
$D(K^{-2})$	20×10^{-6}	14×10^{-6}	6×10^{-6}

duction of the magnetization follows the power law AT^{α}, where $A=(6.9\pm0.5)\times10^{-6}$ and $\alpha=2.00\pm0.01$. If we compare this result to ours, we observe the best agreement for $\lambda_{\perp_1}-\lambda_{\perp_2}=5\times10^{-4}$. Though earlier calculations (Table III of Ref. 5) show that the best value for Néel temperature is obtained for $\lambda_{\perp_1}-\lambda_{\perp_2}=5\times10^{-5}$, the data from the same table also suggest that the deviation from the experimentally obtained T_N for $\lambda_{\perp_1}-\lambda_{\perp_2}=5\times10^{-4}$ is smaller than 10%, wherefore it is acceptable to take also this value for the difference of the interplanar exchange interaction integrals, since Table I shows good agreement with experimental data.

III. SUBLATTICE MAGNETIZATION FOR La_2CuO_4 AND $YBa_2Cu_3O_6$

In order to verify our result for the sublattice magnetization in the orthorhombic La_2CuO_4, as well as the expression for the sublattice magnetization in $YBa_2Cu_3O_6$ derived in Ref. 15, we shall plot these dependences in the temperature interval $0<T<T_N$ and compare them to the experimentally obtained plots published in the paper of Keimer *et al.*[9]

La_2CuO_4. We shall determine first the magnetization at absolute zero using expression (2) in Tyablikov's approximation [Eqs. (3)–(5)]. Adopting the following set of parameters. $J=141$ meV, $\lambda_2=0.0942$, $\eta=10^{-3}$, $\lambda_{\perp_1}=2\times10^{-4}$, and $\lambda_{\perp_2}=10^{-4}$, we obtain $\sigma_0=0.3127$. Taking into account the fact that by neglecting the next-nearest-neighbor interaction ($\lambda_2=0$) we would arrive at the value $\sigma_0=0.3663$, we infer that the magnetization at absolute zero is considerably lower than 1/2 not only due to the quantum spin fluctuations but also due to the frustration resulting from the fact that both nearest- and next-nearest-neighbor in-plane exchange interactions are antiferromagnetic, as shown in Ref. 5.

Temperature dependence of the magnetization is plotted using expression (1) in Tyablikov's approximation. Since this expression is self-consistent, we have used the iterative procedure. Afterward, we repeated the calculation for the 2D model and found the results to be almost identical to those obtained in three dimensions.

$YBa_2Cu_3O_6$. The temperature dependence of the sublattice magnetization will be plotted according to expressions (17), (18), and (19), quoted in Ref. 15, obtained also by the spin Green's function method in Tyablikov's approximation.

For the parameters appearing in the expression for the magnetization, we shall take the following set of values from Table I. from Ref. 15, since this set gives the best agreement with the experimentally obtained value for T_N: nearest-neighbor in-plane exchange integral $J=100$ meV, in-plane anisotropy $\eta=10^{-4}$, and exchange integral ratio

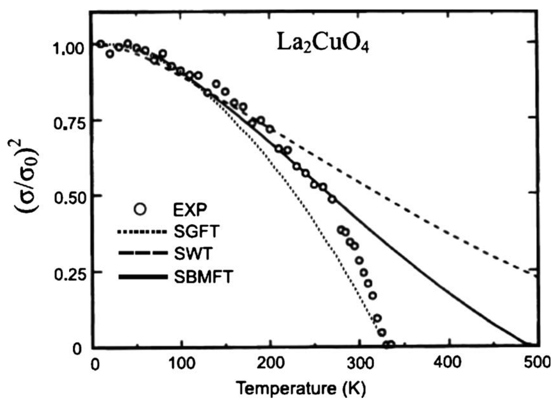

FIG. 1. Sublattice magnetization for La_2CuO_4. Experimental data are represented by circles. Spin Green's functions theory (in Tyablikov's approximation) predictions are denoted by dotted line. The broken line is the prediction of harmonic spin-wave theory. The solid line represents the predictions of Schwinger-boson mean-field theory.

$\lambda_b=J_b/J=5\times10^{-2}$, where J_b is the interplane exchange integral within the bilayer and exchange integral ratio $\lambda'=J'/J=10^{-5}$, where J' is the exchange integral between two bilayers. Using this set of parameters, we obtain for the magnetization at absolute zero $\sigma_0=0.3764$, which is the value higher than the one obtained for La_2CuO_4, confirming the absence of the frustration in $YBa_2Cu_3O_6$. Similar to what we have noticed in the case of La_2CuO_4, the plot for the 2D model of $YBa_2Cu_3O_6$ almost coincides to the one obtained in three dimensions.

To facilitate the comparison of our results to those published in Ref. 9, we plot $(\sigma/\sigma_0)^2$ vs temperature for La_2CuO_4 (Fig. 1) and $YBa_2Cu_3O_6$ (Fig. 2), together with the experimental data taken from Ref. 9 and spin-wave theory (SWT) and Schwinger-boson mean-field theory (SBMFT) results for La_2CuO_4 as well as SWT results for $YBa_2Cu_3O_6$, also taken from Ref. 9.

By inspection of these plots, we see that the spin-wave theory gives a satisfying description of the behavior of mag-

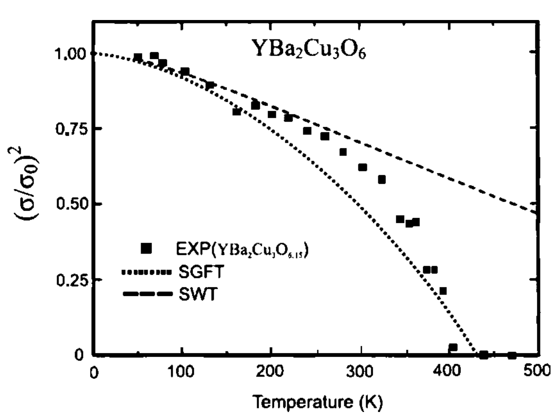

FIG. 2. Sublattice magnetization for $YBa_2Cu_3O_6$. Experimental data are represented by full squares. Spin Green's functions theory (in Tyablikov's approximation) predictions are denoted by dotted line. The broken line is the prediction of harmonic spin-wave theory.

netization at low temperatures but shows a significant discrepancy with the experiment close to and above the Néel temperature. In order to fit the experimental curve at higher temperatures, in Ref. 9, Schwinger-boson mean-field theory is used, giving excellent agreement with the experiment up to $T \approx 0.8 T_N$. However, neither this theory gives valid values for the Néel temperature.

On the other hand, Figs. 1 and 2 show that the spin Green's function theory gives good agreement with the experiments, both at low and high (close to T_N) temperatures. The agreement is better for La_2CuO_4, since for $YBa_2Cu_3O_6$, we compare our theory with the data for slightly doped material.

IV. CONCLUSION

Having at our disposal the experimental results concerning the behavior of the magnetization in quasi-two-dimensional cuprate antiferromagnets La_2CuO_4 and $YBa_2Cu_3O_{6.15}$, we were inspired to try to obtain this behavior theoretically. By making use of the expressions derived starting from the quantum Heisenberg $S=\frac{1}{2}$ antiferromagnetic model and using the method of the spin Green's functions, we were able to calculate the behavior of the magnetization in La_2CuO_4 in the vicinity of absolute zero, reproducing the power law obtained experimentally.

We have also obtained the magnetization curves for the compounds mentioned above, for the whole temperature region from absolute zero to the corresponding Néel temperatures. For both compounds, the plots obtained for the two- and three-dimensional models coincide, suggesting that it is justified to consider these cuprate antiferromagnets quasi-two-dimensional. The comparison of these plots to those obtained experimentally shows excellent agreement, considerably better than the one obtained by the spin-wave or Schwinger-boson mean-field theory.

Though, it would be fair to stress that if we tried to calculate the behavior of the magnetization in the vicinity of Néel temperature using Tyablikov approximation, we would obtain molecular-field theory result, i.e., the critical exponent for the magnetization would take on the value $\beta=\frac{1}{2}$, which is the result widely known from the literature, showing that in the vicinity of the critical temperature, more sophisticated methods (as renormalization group method, for example) have to be applied.

One important justification of our approach is the agreement of our results with the Mermin-Wagner theorem. Furthermore, as mentioned in Sec. I, we have shown that even three-dimensional isotropic tetragonal La_2CuO_4 shows the absence of long-range order.[12] This seems to be not a unique case, but a general result common for the tetragonal quasi-two-dimensional cuprate antiferromagnets can be confirmed also by the results of Ref. 16. In this paper, the authors analyze $SrCuO_2Cl_2$, which is isostructural to tetragonal La_2CuO_4, retaining this structure down to at least 10 K. Due to the cancellation of interplane exchange interactions in tetragonal phase and the absence of any orthorhombic distortion which would lead to the structure characterized by the interplane exchange interactions responsible for the antiferromagnetic state experimentally detected in this compound, the authors, in order to explain the existence of the long-range order, were forced to introduce some kind of anisotropy, namely, the magnetic dipole-dipole interaction. It is precisely this interaction that leads to the long-range order in $SrCuO_2Cl_2$, although T_N is substantially lower in La_2CuO_4 which transits to orthorhombic phase.

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