Comment on "Experimental Observation of the 1/3 Magnetization Plateau in the Diamond-Chain Compound $Cu_3(CO_3)_2(OH)_2$"

Recently, Kikuchi *et al.* [1] have performed an elegant measurement on a spin-1/2 diamond-chain compound $Cu_3(CO_3)_2(OH)_2$. They have nicely observed the 1/3 magnetization plateau, unambiguously confirming the previous theoretical prediction, and the two broad peaks both in the magnetic susceptibility and the specific heat. By using exact diagonalization (ED), density matrix renormalization group (DMRG), and high temperature series expansion methods, they have fitted the experimental data, and concluded that (i) the compound is a model substance for the frustrating diamond spin chain, with the isotropic antiferromagnetic exchange couplings estimated as $J_1{:}J_2{:}J_3 = 1{:}1.25{:}0.45$ where $J_1 = 19$ K, (ii) the two broad peaks of the susceptibility and specific heat are induced by the spin frustration effect, and (iii) the magnetic anisotropy observed can be explained if the Dzyaloshinskii-Moriya (DM) interaction of $D/J_2 = 0.3$ is present.

We note that in Ref. [1], the experimental data at finite temperatures are fitted by the zero-temperature theoretical results obtained by the ED and DMRG methods (Fig. 5), while the result of the high temperature series expansion fails to fit the low-temperature behavior of the susceptibility (Fig. 3). These deficiencies could likely lead to some misinterpretation on the experimental results. By overcoming it, we have attempted to reanalyze the experimental data presented in Ref. [1] by using the finite-temperature transfer matrix renormalization group (TMRG) method (see Ref. [2] for a review) to fit the experiments for the whole available temperature region. Surprisingly, in contrast to the conclusion drawn in Ref. [1], we have found that (i) the compound $Cu_3(CO_3)_2(OH)_2$ is a diamond-chain antiferromagnet without spin frustration, because the exchange couplings J_1 and J_2 are observed to be antiferromagnetic (AF), while J_3 is ferromagnetic (F), (ii) our fittings show that J_1 and J_2 are isotropic, and J_3 is anisotropic, satisfying $J_1{:}J_2{:}J_{3z} = 1{:}1.9{:}-0.3$ with $J_1 = 23$ K, and the anisotropic ratio for the F interaction $J_{3x}/J_{3z} = J_{3y}/J_{3z} = 1.7$, where the z axis is defined as perpendicular to the b axis, (iii) the two round peaks observed in the susceptibility χ and specific heat C cannot thus be explained by the spin frustration effect, which may be attributed to the two kinds of gapless and gapful excitations owing to the competition of the AF and F interactions in the system. Someone might argue that the condition $J_2 \gg J_1$, $|J_3|$ may be necessary to explain the double peak behavior of the susceptibility for the diamond chain. In fact, such an argument is not necessarily true, as we find by a TMRG calculation that the double peak structure of χ for small J_2 such as $J_1{:}J_2{:}J_3 = 1{:}0.5{:}-0.1$ can also be observed. Unlike the assertion in Ref. [1] that the two round peaks are not common in one-dimensional magnets, in the AF-AF-F trimerized spin chain, the double peaks in χ and C are obviously observed (see Ref. [3]), and (iv) the magnetic anisotropy observed experimentally may come from the anisotropy of the F exchange interaction on J_3 bond, in spite of the DM interaction.

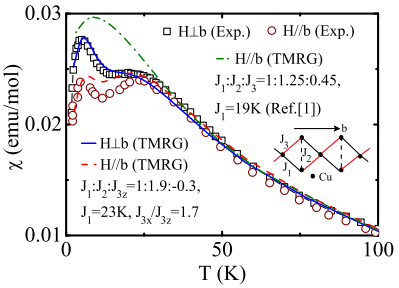

FIG. 1 (color online). A fitting of the magnetic susceptibility for $Cu_3(CO_3)_2(OH)_2$ with the TMRG results, where the experimental data (open squares and circles) are taken from Ref. [1]. The solid (blue) and dashed (red) lines are theoretical results calculated by the TMRG method in the transverse and longitudinal fields, respectively. The dash-dot (green) line is obtained also by the TMRG method but with the parameters given in Ref. [1] for a comparison.

In our TMRG calculations, the number of kept optimal states is taken as $N = 64$ for the susceptibility, where the width of the imaginary time slice is taken as $\varepsilon = 0.1$. The Trotter-Suzuki error is less than 10^{-3}, and the truncation error is smaller than 10^{-6}. The physical quantities are calculated down to $T = 0.025$ (in units of J_1). Our fitting result for the susceptibility χ is presented in Fig. 1 as an example. For a comparison, we have also included the TMRG result calculated by using the parameters given in Ref. [1]. Obviously, our TMRG results with $J_1{:}J_2{:}J_{3z} = 1{:}1.9{:}-0.3$ fit very well the experimental data of χ, and the two round peaks at low temperatures are nicely reproduced, while the result with $J_1{:}J_2{:}J_3 = 1{:}1.25{:}0.45$ obtained in Ref. [1] cannot fit the low-temperature behavior of χ. We have also uncovered that J_3 can be anisotropic, but J_2 cannot.

In conclusion, our TMRG calculations reproduce well the main characteristics of the experimental results of the susceptibility, indicating that the compound $Cu_3(CO_3)_2 \times (OH)_2$ may not be a spin frustrated magnet.

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