LETTER

On the origins of antiferromagnetic order in a frustrated spin system at high pressure

In this letter, I would like to complement the discussion on the antiferromagnetic (AFM) order in the model-frustrated quantum magnet $SrCu_2(BO_3)_2$ at high pressure. The understanding of the Shastry–Sutherland model (SSM) phase diagram and of its only spin S = 1/2 realization $SrCu_2(BO_3)_2$ is regarded to be of particular interest in the field quantum magnetism (1). I therefore comment here on the possible origins of AFM order in $SrCu_2(BO_3)_2$.

In PNAS, Haravifard et al. (2) observe AFM ordering in $SrCu_2(BO_3)_2$ at 5.5 GPa below 122 K. As reported by the authors, the compound has, at this pressure, entered a structural monoclinic phase and is thus no longer exactly described by the orthogonal dimer SSM. The original SSM model (equation 1 in ref. 2) describes the low pressure phase of $SrCu_2(BO_3)_2$ and is complemented for more accurate treatment by 3D and Dzyaloshinskii–Moriya interaction terms (1). Studies of this model indicate that $SrCu_2(BO_3)_2$ could become AFM ordered when the next nearest neighbor to nearest neighbor coupling ratio J'/J reaches a value of about 0.9 (1). Conversely, AFM ordering can also be expected to occur when the compound distorts to the monoclinic phase. There, dimers are not constrained to remain orthogonal any longer, and the four next nearest neighbor couplings J' may become different, relieving the frustration.

It seems that in $SrCu_2(BO_3)_2$, the AFM ordering occurs at 4.0 GPa just before the structural transition at ∼4.5 GPa (3, 4). The boundary between the tetragonal and monoclinic phase (4) also shows a curvature to higher pressures around 120 K, which could be related to the appearance of AFM ordering and thus supports the existence of AFM ordering inside the tetragonal region.

It is therefore possible that in $SrCu_2(BO_3)_2$, the AFM order occurs first within the original SSM orthogonal dimer model, in the tetragonal phase, and is then stabilized by the structural distortions and the 3D effects described in ref. 2.

Mohamed Ezzat Zayed[1]
Department of Mathematics, Statistics and Physics, College of Arts and Science, Qatar University, Doha, Qatar

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[1]Email: mohamed.zayed@qu.edu.qa.

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