LETTER

Reply to Zayed: Interplay of magnetism and structure in the Shastry–Sutherland model

The connection between electronic and structural degrees of freedom—whether successive, coincident, or causal—suffuses the study of phase transitions. The Shastry–Sutherland model of a planar network of coupled spin dimers (1) and its physical realization in $SrCu_2(BO_3)_2$ (SCBO) provide a fundamental quantum mechanical test of this connection at the onset of antiferromagnetic order. We summarize in Fig. 1 the current understanding of SCBO's phase diagram for $T < 200$ K and an intermediate pressure range of 3.5–6 GPa (2–4). At pressures below ~4–5 GPa, SCBO has a tetragonal structure that hosts several low-temperature magnetic phases. Above this pressure, monoclinic distortions reduce the symmetry of the lattice. In ref. 2, we performed full structural refinements of X-ray and neutron scattering measurements to identify a change in space group at 5.5 GPa as a function of temperature (red circles in Fig. 1). This structural change coincides with the onset of antiferromagnetic ordering as a function of temperature, and we argue that this is not a coincidence but instead represents a cooperative effect between distortions of the lattice, the dimers tilting out of the plane, and the emergence of long-range magnetic order. In his comment on our work, Zayed (5) proposes an alternative scenario, in which the antiferromagnetic ordering onsets at lower pressure, within the tetragonal phase, and is then stabilized by the structural distortion associated with the monoclinic phase. He further speculates that this earlier onset may be associated with the dome in the phase boundary reported in ref. 4 (dark red region, Fig. 1).

Zayed's suggestion is an intriguing one. However, there is not yet evidence in the literature to support it. To tease apart competing effects, one requires a sufficiently detailed measurement to allow a full refinement of both the structural and magnetic diffraction patterns. To the best of our knowledge, a measurement of this sort has not been performed in the dome region. The technique used in ref. 4 to measure the phase boundary, of examining the splitting of the degeneracy of the (211) Bragg reflection, is sufficient for tracking the general transition between tetragonal and monoclinic ordering, but it does not yield sufficient information to either specify the space group within the monoclinic phase or to identify the potential onset of Néel ordering of spins within the tetragonal phase. In the absence of high-resolution powder X-ray and neutron measurements in this region, we believe a more likely scenario is that the dome is due to the onset of magnetic ordering associated with the P121/C121 distortion stabilizing the overall tetragonal to monoclinic transition.

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The authors declare no conflict of interest.

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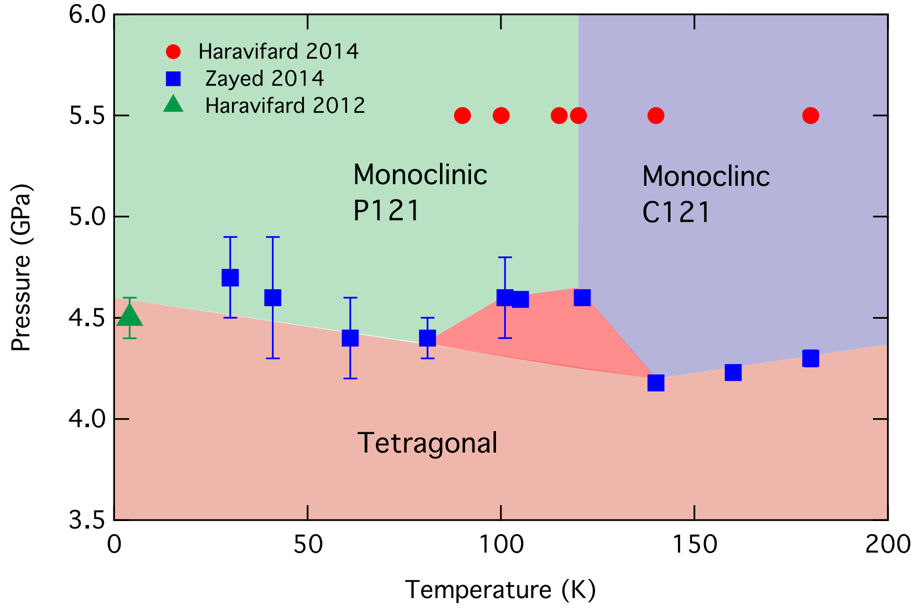

Fig. 1. Structural phase diagram of $SrCu_2(BO_2)_3$ as a function of temperature and pressure. Red circles mark the (P,T) locations at which full structural refinements were performed to identify and locate the C121/P121 structural transition and accompanying onset of magnetic ordering (2). The blue squares and green triangle mark the measured tetragonal to monoclinic transition determined from properties of individual Bragg reflections (3, 4). Background shading identifies different structural phases, with dark red marking the curvature region mentioned in ref. 5.