Comment on "Tuning the Magnetic Dimensionality by Charge Ordering in the Molecular TMTTF Salts"

Yoshimi *et al.* [1] have attempted to explain the pressure (P)-dependent behavior of Fabre salts which exhibit charge order (CO), antiferromagnetic (AFM), and spin-Peierls (SP) phases. Experiments find two AFM phases [2,3], AFM_1 at large P and AFM_2 at small P. Yoshimi *et al.* suggest that there also exist two distinct zero-temperature SP phases, SP_1 and SP_2. Here we point out that the occurrence of two distinct SP phases contradicts experiments [2,3], and is found in Ref. [1] because of unrealistic model parameters.

The experiments of Refs. [2,3] emphasize *cooperative interaction* between the ferroelectric charge order (FCO) and AFM_2 phases. In the experimental phase diagram [2,3] T_{CO} and the Néel temperature in the AFM_2 phase both decrease with P. Thus charge occupancies in the FCO and AFM_2 phases are likely the same. In contrast, P *increases* [2,3] the SP transition temperature, indicating that FCO and SP_2 phases *compete*. No CO was detected for $P > 0.5$ GPa in $(TMTTF)_2SbF_6$ [2,3], in the P region where the SP_2 phase occurs at lower temperature. It is then unlikely that SP_2 and FCO coexist at zero temperature.

The hopping parameters used by the authors in their model calculations are realistic. Their choice of Coulomb interactions is, however, unrealistic. The onsite Coulomb interaction assumed, $U/t_{a2} = 4$, is too small—in the purely electronic one-dimensional model no CO occurs for this U [4,5]. The assumed intersite Coulomb interactions $V_b = 0$ and $V_q = V_a$, are also unrealistic. Given the lattice geometry (see Fig. 1) it is highly unlikely that $V_b \ll V_q$, and with large interchain separation $V_q = V_a$ is equally unrealistic. $4 \lesssim U \lesssim 8$ and $V_b \simeq V_q \ll V_a$ is more appropriate.

We repeated the calculations with more realistic $V_a = V$, $V_b = V_q = 0$, and $4 \leq U \leq 8$. For these parameters, the intradimer charge structure factor ($C_-(\mathbf{q})$ in Ref. [1]) peaks at several $\mathbf{q}$ values, indicating comparable energies for both FCO and the checkerboard pattern CO, in agreement with experiments [6]. Peaks in $S_\pm(\mathbf{q})$ remain at the same $\mathbf{q}$ values as in Fig. 2 of [1]. We conclude that the V_{ij} assumed in Ref. [1] is *not* required to explain coexisting FCO and AFM order in the AFM_2 state.

We also repeated (see Fig. 1) the 8×2 calculations with these parameters. We have three main observations. (i) For $V_a = V$, $V_b = V_q = 0$, we find a phase diagram similar to that in Ref. [1], but with the FCO entering at larger V as expected [4,5]. The choice $V_q = V$, $V_b = 0$ is also *not* required to realize the FCO phase; FCO can be stabilized by antiferromagnetic superexchange along the t_b bonds. (ii) As U increases the FCO + SP phase narrows. (iii) For both these and the parameters assumed in Ref. [1], the width of the FCO + SP phase is directly proportional to the strength of the intersite electron phonon coupling (larger K_1 gives weaker coupling). Unconditional transitions in the thermodynamic limit occur in the limit of 0^+ phonon coupling. Importantly, point (iii) was not discussed in Ref. [1], and together with (ii) suggests that in the thermodynamic limit the FCO + 2DAFM and DM + SP phases may share a common border.

To understand the phase diagram one must consider thermodynamics. For large Coulomb interactions the free energy is dominated by spin excitations. We have previously shown that the same DM + SP ground state can have two kinds of soliton spin excitations: (i) with local CO, or (ii) with uniform charge but local bond distortion [7]. In this picture, to the left of the line bisecting the SP phase [2], soliton excitations with local CO dominate at finite T; to the right occur excitations with uniform site charges. A unique SP *ground state* is expected at all pressures between AFM_1 and AFM_2.

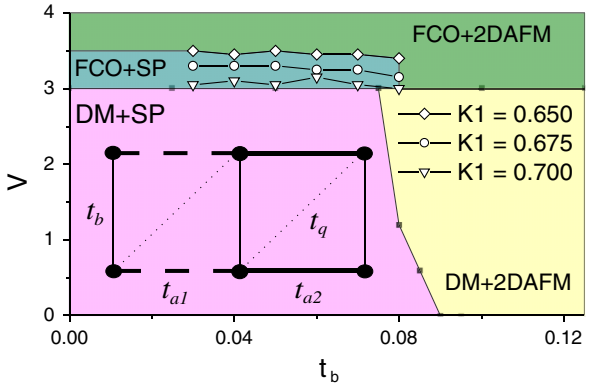

FIG. 1 (color online). 8×2 phase diagram for $U = 6$, $V_a = V$, and $K_2 = 1$. The inset shows the lattice structure assumed by Ref. [1]. As K_1 increases, the size of the FCO + SP phase shrinks. Other points do not significantly change with K_1.

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