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], AFM1 at large P and AFM2 at small P. Yoshimi et al. suggest that there also exist two distinct zero-temperature SP phases, SP1 and SP2. Here we point out that the occurrence of two distinct SP phases contradicts experiments [2,3], and is found in [1] because of unrealistic model parameters.

Experiments [2,3] emphasize co-operative interaction between the ferroelectric charge order (FCO) and AFM2 phases. In the experimental phase diagram [2,3] TCO and the Neél temperature in the AFM2 phase both decrease with P. Thus charge occupancies in the FCO and AFM2 phases are likely the same. In contrast, P increases the AFM1 phase transition temperature, indicating that FCO and SP2 phases compete. No CO was detected for P > 0.5 GPa in (TMTTF)$_2$SbF$_6$ [2,4], in the P region where the SP2 phase occurs at lower temperature. It is then unlikely that SP2 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 $V_b \approx V_q \ll V_a$ is more appropriate.

We repeated the calculations with more realistic $V_b = V$, $V_b = V_q = 0$, and $4 \leq U \leq 8$. For these parameters, the intra-dimer charge structure factor ($C_{\pm}(q)$ in [1]) peaks at several $q$ values, indicating comparable energies for both FCO and the checkerboard pattern CO, in agreement with experiments [5]. Peaks in $S_{\pm}(q)$ remain at the same $q$ values as in Fig. 2 of [1]. We conclude that the $V_{ij}$ assumed in [1] is not required to explain coexisting FCO/AFM order in the AFM2 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 [1], but with 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 [1], the width of the FCO+SP phase is directly proportional to the strength of the inter-site 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 [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 AFM1 and AFM2. We acknowledge support from the Department of Energy grant DE-FG02-06ER46315.

A. B. Ward¹, R. T. Clay¹, and S. Mazumdar²
¹Department of Physics & Astronomy
²HPC² Center for Computational Sciences
Mississippi State University
Mississippi State, MS 39762-5167
¹Department of Physics
University of Arizona
Tucson, AZ 85721

FIG. 1: (color online) 8×2 phase diagram for $U=6$, $V_b=V_q$, and $K_1=1$. The inset shows the lattice structure assumed by [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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