

Seo *et al.* Reply: Our Letter [1] theoretically studied the origin of the complex variation of phases observed in the molecular TMTTF₂X family summarized as a pressure-temperature phase diagram (Fig. 1 in Ref. [1]). The constructed effective model Hamiltonian successfully reproduced the ground states where the charge order (CO) plays a role in determining the magnetic dimensionality. The Comment by Ward *et al.* [2] criticizes two points: (i) The experiments show no coexistence of CO and spin-Peierls (SP) orders, denoted as SP₂ in the phase diagram; (ii) the coexistence is found in our Letter by choosing unrealistic model parameters.

The first objection is based on two experimental papers on TMTTF₂SbF₆ under pressure [3,4]. In Ref. [3], the metal-insulator transition at $T = 156$ K is shown to be a result of CO using NMR methods, and the attractive relationship between CO and antiferromagnetic (AF) orders was established by studying the pressure dependence of the transition temperatures. At ambient pressure, $T_{CO} = 156$ K and $T_N = 8$ K; both decrease with applied pressure, as does the amplitude of the CO. For pressures $P > 0.5$ GPa, evidence for CO is no longer resolved, but AF ordering was already undetected at a lower pressure. Consistent experimental evidence for this observation is summarized in Fig. 9 of Ref. [4]. Neither result conflicts with the SP phase emerging from a CO state upon cooling. Further, CO + SP coexistence was unambiguously demonstrated for TMTTF₂AsF₆ [5,6], and this is particularly relevant here because of something referred to as the *chemical pressure effect*.

This continuous connection of phases for different members of family compounds is one of the key concepts in the field of molecular conductors. In effect, the Comment is questioning its validity, or at least its applicability to the TMTTF salts. However, its foundations are contained in experiments performed in over three decades of research on the TMTSF and TMTTF salts. In the specific case of TMTTF₂SbF₆, it led the authors of Ref. [3] to propose that the physical properties are smoothly transferred to those of TMTTF₂AsF₆ and TMTTF₂PF₆. Further, the repulsive coupling between CO and SP order parameters is clear from experiments [5], but it is not so strong as to produce a line of first order phase transitions between the two. Returning specifically to the designations SP₁ and SP₂ in Ref. [1], we note that the only distinction is that SP₂ refers merely to the coexistence of the SP phase with the ferroelectric-type CO (denoted FCO in Ref. [1]) symmetry breaking.

In addressing (ii), we first note that our aim was to construct an effective model capturing the essential physics and phases observed experimentally. We thus started from first-principles band calculations and included both electronic and electron-phonon couplings. The suggestion that our choice of on-site Coulomb interaction is too small for

the occurrence of CO, is based on the results from a purely electronic and one-dimensional model. However, our model is two-dimensional and includes electron-phonon couplings, both known to widen the parameter space for the ordered phases compared to the one-dimensional electronic model [7–9]. Moreover, the authors of Ref. [2] pointed out the geometry of interchain Coulomb interactions to be unrealistic in our setup and performed the same calculations with their choice of parameters. Nevertheless, the resultant phase diagrams (Fig. 3 in Ref. [1] and Fig. 1 in Ref. [2]) are analogous: there are four regions, distinguished by the existence or absence of two order parameters, i.e., CO and SP. This naturally leads us to the conclusion that the CO + SP coexistence and the variation among the phases are rather general, without the need of fine-tuning of parameters. Finally, we believe that the speculation in Ref. [2] that a smaller electron-phonon coupling may suppress completely the coexistence phase in the thermodynamic limit is unlikely, considering the previous studies on one-dimensional models showing that the CO + SP coexistence phase becomes stabilized, on the contrary, for larger system sizes [7,8].

H. Seo,¹ S. Ishibashi² and S. E. Brown³

¹Condensed Matter Theory Laboratory and
CEMS Quantum Matter Theory Research Team, RIKEN,
Saitama 351-0198, Japan

²Nanosystem Research Institute “RICS”, AIST,
Ibaraki 305-8568, Japan

³Department of Physics and Astronomy, UCLA,
California 90095, USA

Received 8 May 2014; published 11 July 2014

DOI: 10.1103/PhysRevLett.113.029702

PACS numbers: 71.10.Fd, 71.20.Rv, 71.30.+h, 75.30.Kz

- [1] K. Yoshimi, H. Seo, S. Ishibashi, and S. E. Brown, *Phys. Rev. Lett.* **108**, 096402 (2012).
- [2] A. B. Ward, R. T. Clay, and S. Mazumdar, preceding Comment, *Phys. Rev. Lett.* **113**, 029701 (2014).
- [3] W. Yu, F. Zhang, F. Zamborszky, B. Alavi, A. Baur, C. A. Merlic, and S. E. Brown, *Phys. Rev. B* **70**, 121101(R) (2004).
- [4] F. Iwase, K. Sugiura, K. Furukawa, and T. Nakamura, *Phys. Rev. B* **84**, 115140 (2011).
- [5] F. Zamborszky, W. Yu, W. Raas, S. E. Brown, B. Alavi, C. A. Merlic, and A. Baur, *Phys. Rev. B* **66**, 081103(R) (2002).
- [6] S. Hirose, A. Kawamoto, N. Matsunaga, K. Nomura, K. Yamamoto, and K. Yakushi, *Phys. Rev. B* **81**, 205107 (2010).
- [7] M. Kuwabara, H. Seo, and M. Ogata, *J. Phys. Soc. Jpn.* **72**, 225 (2003).
- [8] R. T. Clay, S. Mazumdar, and D. K. Campbell, *Phys. Rev. B* **67**, 115121 (2003).
- [9] Y. Otsuka, H. Seo, Y. Motome, and T. Kato, *J. Phys. Soc. Jpn.* **77**, 113705 (2008).