

**Zhao *et al.* Reply** In the first sentence of the Comment [1], the authors refer to four systems: GaN(10 $\bar{1}$ 0)-1H, ZnO(10 $\bar{1}$ 0)-1H, GaN(10 $\bar{1}$ 0)-Li, and ZnO(10 $\bar{1}$ 0)-1Li. In the second sentence, the authors claim to demonstrate that “a charge-density-wave (CDW) phase is due to the artifact of the generalized gradient approximation (GGA), while the antiferromagnetic (AFM) ground state is predicted by the hybrid DFT calculation...” The Comment contains a single table, showing the total energy of the GaN(10 $\bar{1}$ 0)-1H system only. No numerical results were presented for the other three systems. It is not clear how the authors arrived at their very general conclusion.

We have checked the validity of Kim *et al.*'s claim by performing new calculations using two popular hybrid functionals, HSE06 [2,3] and B3LYP [4], on the four systems mentioned in the Comment. These two hybrid functional methods are generally considered to be at the same level of accuracy. All hybrid functional calculations are performed on a 12-layer slab separated by a 12 Å vacuum. The two-dimensional  $k$  grid of the Brillouin zone is  $8 \times 6$  for the  $p(1 \times 1)$  cell and the same density is used for larger cells. The results are listed below.

Based on these results, we draw a few observations: (i) For the GaN(10 $\bar{1}$ 0)-1H system, different hybrid functionals produce different ground states. The conclusions are not consistent. Our Table I shows that the B3LYP hybrid method favors the CDW state by 0.21 eV per  $p(2 \times 2)$  unit cell, and that this preference is reversed with the HSE06 hybrid method, which instead favors the AFM state by 0.06 eV. This is similar to Kim's HSE results, which also favored the AFM state by 0.07 eV. Importantly, Kim *et al.* also perform RPA calculations, which favor the AFM state by 122 meV per  $p(2 \times 2)$  unit cell in GaN-1H. We have confirmed this value for RPA. For the GaN(10 $\bar{1}$ 0)-1Li system, however, both hybrid functionals B3LYP and HSE06 favor the CDW ground state, by 0.5 and 0.25 eV, respectively. For GaN(10 $\bar{1}$ 0)-1Li, we have also carried out RPA calculations [5–8] using a four-layer slab. The RPA results favor the CDW state over the AFM state by 0.16 eV. Thus, for GaN(10 $\bar{1}$ 0)-1Li, all results using B3LYP, HSE06 and RPA support our findings [9], and disagree with the claim of Kim *et al.* (ii) For ZnO(10 $\bar{1}$ 0) with 1H and 1Li, both HSE06 and B3LYP functionals favor the CDW ground state, according to our new results shown

in Table I. The new results again disagree with the claim of Kim *et al.* and support our initial findings [9].

There are other contradictory statements in the Comment with which we disagree. For example, in the last paragraph, Kim *et al.* speculated that the AFM ground state is favored because of the narrow band width at the Fermi level. Since both GaN(10 $\bar{1}$ 0)-1H and GaN(10 $\bar{1}$ 0)-1Li have similar bandwidths at the Fermi level, and the latter clearly prefers the CDW, no general statement can be made based on bandwidth. In the same paragraph the authors stated that “more localized dangling-bond electrons in GaN(10 $\bar{1}$ 0) are likely to favor the AFM order over the CDW formation, contrasting with the case of ZnO(10 $\bar{1}$ 0)-1H.” This sentence contradicts the claim they make in their first paragraph, i.e., that in all four systems including ZnO(10 $\bar{1}$ 0)-1H, the AFM phase is favored.

In summary, only one system out of four proposed in our original Letter is predicted to have an AFM order ground state, and is lower than the CDW state only by a very small amount. Similar systems will have to be checked individually in future calculations, but the mechanism proposed in our Letter is robust and generic, if not universal. We conclude that care must be taken with AFM instabilities and semilocal  $xc$  functionals. In delicate cases where hybrid and semilocal functionals disagree, full RPA, or better approximations, may be necessary to discern the true ground state.

This work is supported by the National Natural Science Foundation of China (NSFC, Grants No. 11674148, No. 11334003, and No. 11404159) and the Basic Research Program of Science, Technology and Innovation Commission of Shenzhen Municipality (Grant No. JCYJ20160531190054083). Z. Z. acknowledges financial support by the Deutsche Forschungsgemeinschaft Grant No. ZA780/3-1.

J. Z. Zhao,<sup>1,2</sup> W. Fan,<sup>3</sup> M. J. Verstraete,<sup>4</sup> Z. Zanolli,<sup>5,6</sup>  
J. Fan,<sup>1</sup> X. B. Yang,<sup>7</sup> H. Xu<sup>1,\*</sup> and S. Y. Tong<sup>1,8,†</sup>

<sup>1</sup>Department of Physics

South University of Science and Technology of China  
518055 Shenzhen, People's Republic of China

<sup>2</sup>Dalian Institute of Chemical Physics

Chinese Academy of Sciences

116023 Dalian, People's Republic of China

TABLE I. Total energy (in eV) per  $p(2 \times 2)$  cell of each phase with respect to the  $p(1 \times 1)$  nonmagnetic phase. The numbers in bold correspond to the lower energy state.

	Hybrid function	GaN(10 $\bar{1}$ 0)-1H		GaN(10 $\bar{1}$ 0)-1Li		ZnO(10 $\bar{1}$ 0)-1H		ZnO(10 $\bar{1}$ 0)-1Li	
		CDW	AFM	CDW	AFM	CDW	AFM	CDW	AFM
$\Delta E$	B3LYP	<b>-0.61</b>	-0.40	<b>-0.90</b>	-0.40	<b>-0.18</b>	-0.02	<b>-0.36</b>	-0.02
	HSE06	-0.87	<b>-0.93</b>	<b>-0.72</b>	-0.47	<b>-0.24</b>	-0.20	<b>-0.58</b>	-0.40
	HSE(Kim)	-1.04	<b>-1.11</b>						

<sup>3</sup>Computational Condensed Matter Physics Laboratory  
RIKEN, Wako, Saitama 351-0198, Japan

<sup>4</sup>Département de Physique and European Theoretical  
Spectroscopy Facility, Université de Liège  
B-4000 Sart Tilman, Liège, Belgium

<sup>5</sup>Peter Grünberg Institute (PGI-1) and Institute for Advanced  
Simulation (IAS-1), Forschungszentrum Jülich  
D-52425 Jülich, Germany

<sup>6</sup>Institute for Theoretical Solid State Physics and European  
Theoretical Spectroscopy Facility (ETSF)  
RWTH Aachen University  
D-52056 Aachen, Germany

<sup>7</sup>Department of Physics  
South China University of Technology  
Guangzhou 510640, People's Republic of China

<sup>8</sup>School of Science and Engineering  
The Chinese University of Hong Kong (Shenzhen)  
518172 Shenzhen, People's Republic of China

\*xu.h@sustc.edu.cn

†tong.sy@sustc.edu.cn

- [1] S.-W. Kim *et al.*, preceding Comment, *Phys. Rev. Lett.* **118**, 239601 (2017).
- [2] J. Heyd, G. E. Scuseria, and M. Ernzerhof, *J. Chem. Phys.* **118**, 8207 (2003).
- [3] A. V. Krukau, O. A. Vydrov, A. F. Izmaylov, and G. E. Scuseria, *J. Chem. Phys.* **125**, 224106 (2006).
- [4] A. D. Becke, *J. Chem. Phys.* **98**, 5648 (1993).
- [5] D. Bohm and D. Pines, *Phys. Rev.* **92**, 609 (1953).
- [6] M. Gell-Mann and K. A. Brueckner, *Phys. Rev.* **106**, 364 (1957).
- [7] D. C. Langreth and J. P. Perdew, *Phys. Rev. B* **15**, 2884 (1977).
- [8] X. Ren, A. Tkatchenko, P. Rinke, and M. Scheffler, *Phys. Rev. Lett.* **106**, 153003 (2011).
- [9] J. Z. Zhao, W. Fan, M. J. Verstraete, Z. Zanolli, J. Fan, X. B. Yang, H. Xu, and S. Y. Tong, *Phys. Rev. Lett.* **117**, 116101 (2016).

Received 29 March 2017; published 9 June 2017

DOI: [10.1103/PhysRevLett.118.239602](https://doi.org/10.1103/PhysRevLett.118.239602)