Role of nonlocality in exchange correlation for magnetic two-dimensional van der Waals materials

Yongbin Lee  
*Ames Laboratory, yblee@ameslab.gov*

Takao Kotani  
*Tottori University*

Liqin Ke  
*Ames Laboratory, liqinke@ameslab.gov*

Follow this and additional works at: [https://lib.dr.iastate.edu/ameslab_manuscripts](https://lib.dr.iastate.edu/ameslab_manuscripts)

Part of the Condensed Matter Physics Commons, and the Materials Science and Engineering Commons

**Recommended Citation**

Lee, Yongbin; Kotani, Takao; and Ke, Liqin, "Role of nonlocality in exchange correlation for magnetic two-dimensional van der Waals materials" (2020). *Ames Laboratory Accepted Manuscripts*. 710.  
[https://lib.dr.iastate.edu/ameslab_manuscripts/710](https://lib.dr.iastate.edu/ameslab_manuscripts/710)

This Article is brought to you for free and open access by the Ames Laboratory at Iowa State University Digital Repository. It has been accepted for inclusion in Ames Laboratory Accepted Manuscripts by an authorized administrator of Iowa State University Digital Repository. For more information, please contact [digirep@iastate.edu](mailto:digirep@iastate.edu).
Role of nonlocality in exchange correlation for magnetic two-dimensional van der Waals materials

Abstract
To obtain accurate independent-particle descriptions for ferromagnetic two-dimensional van der Waals materials, we apply the quasiparticle self-consistent GW (QSGW) method to VI3, CrI3, CrGeTe3, and Fe3GeTe2. QSGW provides a description of the nonlocal exchange-correlation term in the one-particle Hamiltonian. The nonlocal term is important not only as the U of density functional theory (DFT) + U but also for differentiating occupied and unoccupied states in semiconductors. We show the limitations of DFT + U in mimicking QSGW.

Disciplines
Condensed Matter Physics | Materials Science and Engineering

This article is available at Iowa State University Digital Repository: https://lib.dr.iastate.edu/ameslab_manuscripts/710
Role of nonlocality in exchange correlation for magnetic two-dimensional van der Waals materials

Y. Lee, 1 Takao Kotani, 2 and Liqin Ke 1,*

1Ames Laboratory, U.S. Department of Energy, Ames, Iowa 50011, USA
2Department of Applied Mathematics and Physics, Tottori University, Tottori 680-8552, Japan

(Received 2 January 2020; revised manuscript received 31 March 2020; accepted 21 April 2020; published 5 June 2020)

To obtain accurate independent-particle descriptions for ferromagnetic two-dimensional van der Waals materials, we apply the quasiparticle self-consistent GW (QSGW) method to VI$_3$, CrI$_3$, CrGeTe$_3$, and Fe$_2$GeTe$_2$. QSGW provides a description of the nonlocal exchange-correlation term in the one-particle Hamiltonian. The nonlocal term is important not only as the $U$ of density functional theory (DFT) + $U$ but also for differentiating occupied and unoccupied states in semiconductors. We show the limitations of DFT + $U$ in mimicking QSGW.

DOI: 10.1103/PhysRevB.101.241409

**Introduction.** The recent experimental realization of magnetic two-dimensional (2D) van der Waals (vdW) materials has generated great interest for exploiting novel 2D magnetism and for applications such as energy-efficient ultracom- pact spin-based electronics [1]. Long-range ferromagnetic ordering in the atomically thin systems was first demonstrated in the CrGeTe$_3$ bilayer [2] and CrI$_3$ monolayer [3], albeit only at very low temperatures. Later, Deng et al. [4] showed that an electric field could drastically increase the Curie temperature $T_C$ of a Fe$_2$GeTe$_2$ monolayer up to room temperature. Recently, VI$_3$ has been identified as the first vdW hard ferromagnet with a large coercivity [5–7]. Spurred by these experiments, many theoretical efforts have been published treating magnetic 2D vdW materials (m2Dv) [8–15].

We are also witnessing the recent revolutionary development of materials informatics (MI). For example, Mounet et al. [16] have employed a computational MI to search for 2D exfoliable materials by multilevel screening from the databases of experimentally known compounds. The quality of such work largely depends on the choice of the first-principles method used for the final screening. In the future, such an MI procedure may be applied to m2Dv. Then the first-principles method used in MI should be as reliable as possible and with no adjustable parameters for each material.

Until now, m2Dv has been theoretically treated mostly within density functional theory (DFT) + $U$, with a single Hubbard $U$ applied on the cation-3d orbitals, as in Refs. [17–20]. Phenomenological theories, such as DFT + $U$ and dynamical mean-field theory, are very useful for various material systems. However, it is not clear that one can use DFT + $U$ for the above-mentioned MI, because of the limitation of the single parameter $U$, as we illustrate in the following.

First, the cation-3d bands in m2Dv contain more degrees of freedom than a single $U$ parameter can describe. Although DFT + $U$ may adjust overall splitting between occupied and unoccupied 3d bands for each spin, it ignores the $k$ dependence and frequency dependence of effective interactions and thus the interaction anisotropy regarding in-plane and out-of-plane 3d orbitals in m2Dv cannot be adequately treated. An idea using many parameters for the $U$ term would be hard to implement because of the difficulty in determining the unique parameters.

Second, the relative positions of cation-3d and anion-p bands are not directly controlled by on-site $U$. For example, even in nonmagnetic CdO where we expect no $U$ effect because Cd-4d states are fully occupied, we see the center of occupied 4d states can be pushed down about 2 eV (see Fig. A1 in Ref. [21]) in comparison with DFT. Note that the relative positions of and hybridizations between cation-3d and anion-p can be important to determine the superexchange coupling in m2Dv.

In this Rapid Communication, we apply the quasiparticle self-consistent GW (QSGW) method [21–23] to m2Dv, including VI$_3$, CrI$_3$, CrGeTe$_3$, and Fe$_2$GeTe$_2$. QSGW has been applied to a wide range of materials and shown to be the most reliable method available to determine the one-particle Hamiltonian $H_0$, which defines the independent-particle picture of a particular material. For each material, an accurate $H_0$ is the key to evaluate all of its physical quantities theoretically. We will show that QSGW reasonably describes electronic structures consistent with experiments for all m2Dv treated here. Then we will examine whether DFT + $U$ can mimic the band structures obtained in QSGW. We will demonstrate the serious limitations of DFT in treating m2Dv, corresponding to the two reasons discussed above.

**Methods.** First, let us recall the GW approximation (GWA) [24,25]. GWA can be applied to any one-particle Hamiltonian $H_0$, for example, to the Kohn-Sham Hamiltonian of DFT. In GWA, we calculate the self-energy $\Sigma(r, r', \omega) = \Sigma(1, 2) = iG_0(1, 2)W(1+, 2)$. Here, $G_0 = 1/(\omega - H_0)$ is the Green’s function of $H_0$; $W$ is the dynamically screened Coulomb interaction calculated using $G_0$, usually in the random phase approximation (RPA). Then we can determine the quasiparticle energies with $\Sigma(r, r', \omega)$ in the place of the...
exchange-correlation (xc) potential. The reliability of this one-shot method, so-called $G_0W_0$, depends on the reliability of $H_0$.

Thus, the main theoretical problem of $G_0W_0$ is how to determine $H_0$ to which we apply GWA. For this purpose, various self-consistent schemes have been developed. In practice, a partial self-consistency is often employed due to the demanding nature of computation or the intrinsic problems of the methods [26]. In the so-called energy-only self-consistent GW method [27,28], the eigenfunctions are fixed while only the one-electron particles are iterated to reach consistency. In a $G_0W_0$ method [28], one may calculate $W$ using DFT $G_0$, but iterate $G$ until convergence.

QSGW [22,23,29] is given as a self-consistent perturbation method based on the quasiparticle picture within GWA. The full many-body Hamiltonian $H$ is divided into $H = H_0 + (H - H_0)$, then $(H - H_0)$ is treated as a perturbation in GWA. The self-consistent perturbation requires that we should determine $H_0$ so that the term generated in GWA due to $(H - H_0)$ gives virtually zero.

Based on this idea, we generate the QSGW xc potential $V_{\text{QSGW}}^{\text{xc}}$ from the self-energy $\Sigma (\mathbf{r}, \mathbf{r}', \omega)$ obtained in GWA as

$$V_{\text{QSGW}}^{\text{xc}} = \frac{1}{2} \sum_{ij} |\psi_i| \{ \text{Re}[\Sigma(\epsilon_i)]_{ij} + \text{Re}[\Sigma(\epsilon_j)]_{ij} \} |\psi_j|. \quad (1)$$

Here, $\epsilon_i$ and $|\psi_i\rangle$ are eigenvalues and eigenfunctions, respectively, of Hamiltonian $H_0$. Re denotes the Hermitian part. $\Sigma_{ij}(\omega) = \langle \psi_i | \Sigma(\omega) | \psi_j \rangle = \int d^3 \mathbf{r} \int d^3 \mathbf{r'} |\psi_i^*(\mathbf{r})\Sigma(\mathbf{r}, \mathbf{r'}, \omega)\psi_j(\mathbf{r})\rangle$. With Eq. (1), we have a mapping to generate a new $H_0$, $H_0^{(i)} \rightarrow H_0^{(i+1)}$. This is repeated until $H_0$ is converged. Note that $G_0W_0$ applied to this self-consistent $H_0$ does not cause corrections of the quasiparticle energies because of this self-consistency.

QSGW, as it is, tends to systematically overestimate exchange effects, especially for band gaps [21,22,30]. This can be due to the underestimation of the screening effect in RPA, which neglects electron-hole correlations in the proper polarization function [28,30], and/or the neglect of the screening effect of phonons [31]. Shishkin et al. [28] performed calculations that include the correlation via the vertex correction for $W$ and demonstrated very reliable predictions of band gaps by recovering the screening underestimate. However, their methods are too computationally demanding to apply to the materials treated here. Based on the observation that the underestimations are rather systematic in various systems [32], we here use a hybrid QSGW method, QSGW80 [21,33], which uses an empirical mixing of $V^{\text{xc}} = 0.8V_{\text{QSGW}}^{\text{xc}} + 0.2V_{\text{LDA}}^{\text{xc}}$. QSGW80 is taken to be a substitution of the method by Shishkin et al. to remedy the underestimation quickly and efficiently. Unless specified, all QSGW calculations in this work are carried out in QSGW80, referred to hereafter as QSGW, for simplicity.

The nonlocality of $V_{\text{QSGW}}^{\text{xc}}$ provides a natural description of the correct independent-particle picture. Generally speaking, we can classify this nonlocality into two parts: on site and off site. The on-site nonlocality, which can differentiate five 3$d$ orbitals, can be approximated, to a certain extent, by the Hubbard $U$ in DFT + $U$. The off-site nonlocality is critical to generate band gaps in semiconductors. To illustrate this, let us consider a hydrogen dimer $H_2$. To lowest the highest occupied molecular orbital (HOMO) energy without changing the shape of eigenfunctions, one needs to introduce a projector of HOMO. The corresponding projector is naturally represented by a nonlocal potential, and the screened exchange contribution in $V_{\text{QSGW}}^{\text{xc}}$ works exactly as such a projector.

Furthermore, in contrast to the hybrid functional methods, the electron screening effects on the exchange is calculated explicitly in QSGW. The screened Coulomb interaction $W$, which determines the screened exchange, is spatially dependent and self-consistently determined without any system-dependent parameters. On the other hand, in the hybrid functional methods such as Heyd-Scuseria-Ernzerhof (HSE), the xc functional is obtained by mixing the DFT xc with the Hartree-Fock (HF) exchange, which is calculated using the bare Coulomb interaction kernel. The mixing parameter solely mimics the screening effect. This limits the universality of the hybrid functional methods because the screening effects vary significantly between metals and semiconductors, and their spatial dependence could be important in anisotropic systems, which can be hard to be mimicked by one single parameter. In fact, He and Franchini [34] showed that the mixing could be very material dependent. Thus, the explicit treatment of screened exchange allows QSGW to treat complex subjects such as metal/insulator interfaces, and also m2Dv, which contain both features of semiconductor and anisotropic magnetic materials.

**Computational details.** We use the QSGW method from the ECALI package [23], which is implemented with a mixed basis and allows automatic interpolation of self-energy in the whole Brillouin zone without resorting to the WANNIER90 techniques [35,36]. The spin-orbit coupling (SOC) is included as a perturbation [21] after we attain the self-consistency of QSGW. We employed the experimental lattice parameters [5,37–39] for calculations. As for DFT + $U$, we use both fully-localized-limit (PLL) [40] and around-the-mean-field (AMF) [41] double-counting schemes to investigate the dependence of band structures on the correlation parameter $U$, which is applied on the cation-3$d$ orbitals. All DFT and DFT + $U$ calculations are carried out within the generalized gradient approximation using the functional of Perdew, Burke, and Ernzerhof (PBE) [42].

**Results.** Fe$_2$GeTe$_2$ is a metallic m2Dv and has a higher $T_C$ than its semiconducting counterparts [4]. Figure 1 shows the total density of states (DOS) and partial density of states (PDOS) calculated in QSGW. DOS obtained by DFT is also shown for comparison. Both QSGW and DFT suggest that Fe$_2$GeTe$_2$ is a metal, as found in experiments. DOS are dominated by Fe-3$d$ states in this energy window. The Fermi level $E_F$ is located at a pseudogap of Fe$_1$:3$d$ states in the minority-spin channel. QSGW gives slightly narrower 3$d$ bands than DFT, suggesting a somewhat stronger localization of electron states in QSGW. Indeed, much 3$d$-band narrowing is rather general in QSGW as shown in Refs. [43,44]. Considering the fact that QSGW describes metals such as bcc Fe and also transition-metal oxides such as NiO very well, our result supports the applicability of DFT to Fe$_2$GeTe$_2$. For a band structure comparison between DFT and QSGW, see the Supplemental Material [45].
Note the difficulty of hybrid functionals such as HSE applied to m2Dv without a choice of material-dependent parameters. For example, one usually uses a mixing parameter $a = 0.25$ for semiconductors. However, it was found that $a = 0.15$ [46] is optimum for transition-metal oxides. Furthermore, $a = 0$ is apparently good for bcc Fe while HSE06 gives a magnetic moment of 2.89 $\mu_B$/Fe [47]. Since semiconducting and metallic features coexist with transition metals in m2Dv, we can hardly expect HSE to work well for m2Dv. We think that QSGW is the optimal choice to describe electronic structures of m2Dv along the line of MI.

Table I summarizes the experimental and our calculated $E_g$ values in m2Dv. Unlike DFT, QSGW correctly predicts VI$_3$ as a semiconductor. It is worth noting that $G_0W_0$ applied to DFT does not open the gap in VI$_3$, as it does in VO$_2$, demonstrating the necessity of self-consistency of GW calculations as in QSGW. For CrGeTe$_3$, QSGW gives $E_g = 0.66$ eV, within the range of reported experimental values of $0.20$–$0.74$ eV, while DFT gives a much smaller value of $E_g = 0.19$ eV. On the other hand, in CrI$_3$, QSGW gives $E_g = 1.68$ eV, 35% larger than the only reported experimental value of 1.2 eV. This difference is somewhat larger than expected, considering that QSGW produces $E_g$ within ~10% difference for a wide range of materials [21].

SOC reduces the calculated $E_g$ in all three semiconducting compounds, as shown in Table I, especially within QSGW. The strong SOC effects on $E_g$ are due to the heavy anion atoms in the compounds. I- and Te-5p orbitals have rather large SOC constants, $\xi_p = 0.9$–$1.0$ eV, while V- and Cr-3d orbitals have $\xi_p = 20$–$30$ meV. The contribution of SOC to $E_g$ of CrI$_3$ in QSGW (0.55 eV) is about twice as large as in DFT (0.29 eV).

CrI$_3$. Figures 2(a) and 2(b) show the PDOS of CrI$_3$ calculated in DFT and QSGW, respectively, without SOC. QSGW shifts up the unoccupied states in both spin channels, resulting in a larger $E_g$ than the one we obtain in DFT. In the majority spin, the valence cation-3d states are pushed down relative to the anion-5p states, and the top of valence bands at $\Gamma$ becomes more dominated by anion-p states.

Figure 2(c) compares the QSGW band structures of Cr$_3$ calculated with and without SOC. It clearly shows that SOC pushes up top valence bands around the $\Gamma$ point, resulting in a smaller $E_g$. Within QSGW, the top of majority-spin valence bands become more pure anion-p-like after 3d states are pushed down. As a result, SOC has a stronger effect on decreasing $E_g$ in QSGW than in DFT. Similar SOC effects are also found in VI$_3$ and CrGeTe$_3$.

Table I. Band gaps $E_g$(eV) calculated in DFT and QSGW, with and without SOC. Experimental values are listed to compare. The reported theoretical $E_g$ are in the range of 0.74–1.6, 0–0.43, and 0–1.0 eV for bulk CrI$_3$ [10,48], CrGeTe$_3$ [9,13], and VI$_3$ [6,20], respectively.

| Compound | Experiment | DFT SOC | DFT QSGW | SOC | QSGW |
|----------|-------------|---------|----------|-----|------|
| VI$_3$   | 0.32–0.67$^a$ | 0       | 0.53     | 0   | 0.75 |
| CrI$_3$  | 1.2$^b$     | 0.78    | 1.68     | 1.07| 2.23 |
| CrGeTe$_3$ | 0.20–0.74$^c$ | 0.19    | 0.66     | 0.42| 0.99 |

$^a$Resistivity measurement: 0.32 eV [20]; optical reflectance: 0.6 eV [7]; optical transmittance: 0.67 eV [20].

$^b$Optical transition measurement [49].

$^c$Angle-resolved photoemission spectroscopy (ARPES) measurements: 0.38 eV [50] and 0.2 eV [51]; resistivity measurement: 0.2 eV [52]; scanning tunneling microscopy (STM) measurement: 0.74 eV [18].
To understand the behavior of $E_g$ dependence on $U$, we examine how electronic structures evolve with the increasing of $U$ in AMF. Figure 5 shows the $U$ dependence of the valence band maximum (VBM), the conduction band minimum (CBM), and the band centers of valence and conduction cation-$3d$ states in both spin channels, comparing with QSGW results. For all three compounds, applying $U$ increases the gap and the distance between the centers of occupied and unoccupied $3d$ bands in the majority-spin but not the minority-spin channel. This is clearly shown in Fig. 3 for the case of $V_3I_3$ with $U = 2.7$ eV. In $CrI_3$ and $CrGeTe_3$, a large $U$ pushes up the unoccupied $3d$ bands in the majority spin but lowers them in the minority spin. When $U$ is sufficiently large, the unoccupied $3d$ states in the minority spin are shifted below those in the majority spin, and $E_g$ is determined by the exchange splitting instead of crystal-field splitting. A similar trend is also observed in $V_3I_3$, but it occurs at a much larger $U$. DOS calculated within DFT, DFT $+ U$, and QSGW can be found in the Supplemental Material [45].

Can we mimic QSGW DOS by applying $U$ on cation-$d$ orbitals? Now we compare DFT $+ U$ with QSGW DOS. As shown in Fig. 5, QSGW separates further, in comparison to DFT, the occupied and unoccupied states in both spin channels, while DFT $+ U$ only separates them in one spin channel. Hence, within DFT $+ U$, a single $U$ parameter is not able to mimic the QSGW $3d$ band centers simultaneously in both spin channels.
CBM and VBM are denoted by open circles (DFT $+U$ calculation. SOC is not included."

\[ \text{CBM and VBM are denoted by open circles (DFT } \text{+U calculation. SOC is not included.} \]

\\[2\] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, and X. Xu, Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit, Nature (London) 546, 270 (2017).

\\[4\] Y. Deng, Y. Yu, Y. Song, J. Zhang, N. Z. Wang, Z. Sun, Y. Yi, Y. Z. Wu, S. Wu, J. Zhu, J. Wang, X. H. Chen, and Y. Zhang, Gate-tunable room-temperature ferromagnetism in two-dimensional Fe$_2$GeTe$_2$, Nature (London) 563, 94 (2018).

\\[5\] S. Tian, J.-F. Zhang, C. Li, T. Ying, S. Li, X. Zhang, K. Liu, and H. Lei, Ferromagnetic van der Waals Crystal VI$_3$, J. Am. Chem. Soc. 141, 5326 (2019).
[6] J. He, S. Ma, P. Lyu, and P. Nachtigall, Unusual Dirac half-metallicity with intrinsic ferromagnetism in vanadium trihalide monolayers, J. Mater. Chem. C 4, 2518 (2016).

[7] T. Kong, K. Stolze, E. I. Timmons, J. Tao, D. Ni, S. Guo, Z. Yang, R. Prozorov, and R. J. Cava, V1₁—a new layered ferromagnetic semiconductor, Adv. Mater. 31, 1808074 (2019).

[8] S. Baidya, J. Yu, and C. H. Kim, Tunable magnetic topological insulating phases in monolayer CrI₃, Phys. Rev. B 98, 155148 (2018).

[9] Y. Fang, S. Wu, Z.-Z. Zhu, and G.-Y. Guo, Large magnetooptical effects and magnetic anisotropy energy in two-dimensional Cr₂Ge₂Te₆, Phys. Rev. B 98, 125416 (2018).

[10] P. Jiang, L. Li, Z. Liao, Y. X. Zhao, and Z. Zhong, Spin direction-controlled electronic band structure in two-dimensional ferromagnetic CrI₃, Nano Lett. 18, 3844 (2018).

[11] V. V. Kulish and W. Huang, Single-layer metal halides MX₂(X = Cl, Br, I): Stability and tunable magnetism from first principles and Monte Carlo simulations, J. Mater. Chem. C 5, 8734 (2017).

[12] N. Sivadas, S. Okamoto, X. Xu, C. J. Fennie, and D. Xiao, Stacking-dependent magnetism in bilayer CrI₃, Nano Lett. 18, 7658 (2018).

[13] G. Menichetti, M. Calandra, and M. Polini, Electronic structure and magnetic properties of few-layer Cr₇Ge₇Te₆: The key role of nonlocal electron-electron interaction effects, 2D Mater. 6, 045042 (2019).

[14] D. Torelli and T. Olsen, Calculating critical temperatures for ferromagnetic order in two-dimensional materials, 2D Mater. 6, 015028 (2018).

[15] J. L. Lado and J. Fernández-Rossier, On the origin of magnetic anisotropy in two dimensional CrI₃, 2D Mater. 4, 035002 (2017).

[16] N. Mounet, M. Gibertini, P. Schwaller, D. Campi, A. Merkys, A. Marrazzo, T. Sohier, I. E. Castelli, A. Cepellotti, G. Pizzi, and N. Marzari, Two-dimensional materials from high-throughput computational exfoliation of experimentally known compounds, Nat. Nanotechnol. 13, 246 (2018).

[17] S. W. Jang, M. Y. Jeong, H. Yoon, S. Ryee, and M. J. Han, Microscopic understanding of magnetic interactions in bilayer CrI₃, Phys. Rev. Materials 3, 031001 (2019).

[18] Z. Hao, H. Li, S. Zhang, X. Li, G. Lin, X. Luo, Y. Sun, Z. Liu, and Y. Wang, Atomic scale electronic structure of the ferromagnetic semiconductor Cr₇Ge₇Te₆, Sci. Bull. 63, 825 (2018).

[19] X. Li and J. Yang, CrₓTe₅₋ₓ(X = Si, Ge) nanosheets: Two dimensional intrinsic ferromagnetic semiconductors, J. Mater. Chem. C 2, 7071 (2014).

[20] S. Son, M. J. Coak, N. Lee, J. Kim, T. Y. Kim, H. Hamidov, H. Cho, C. Liu, D. M. Jarvis, P. A. C. Brown, J. H. Kim, C.-H. Park, D. I. Khomskii, S. S. Saxena, and J.-G. Park, Bulk properties of the van der Waals hard ferromagnet V₁₁, Phys. Rev. B 99, 041402(R) (2019).

[21] D. Deguchi, K. Sato, H. Kino, and T. Kotani, Accurate energy bands calculated by the hybrid quasiparticle self-consistent GW method implemented in the eCALJ package, Jpn. J. Appl. Phys. 55, 051201 (2016).

[22] T. Kotani, M. van Schilfgaarde, and S. V. Faleev, Quasiparticle self-consistent GW method: A basis for the independent-particle approximation, Phys. Rev. B 76, 165106 (2007).

[23] T. Kotani, Quasiparticle self-consistent GW method based on the augmented plane-wave and muffin-tin orbital method, J. Phys. Soc. Jpn. 83, 094711 (2014).

[24] L. Hedin and S. Lundqvist, Effects of Electron-Electron and Electron-Phonon Interactions on the One-Electron States of Solids, Vol. 12 (Oxford University Press, New York, 1969).

[25] F. Aryasetiawan and O. Gunnarsson, The GW method, Rep. Prog. Phys. 61, 237 (1998).

[26] A. L. Kutepov, Electronic structure of Na, K, Si, and LiF from self-consistent solution of Hedin’s equations including vertex corrections, Phys. Rev. B 94, 155101 (2016).

[27] M. S. Hybertsen and S. G. Louie, Electron correlation in semiconductors and insulators: Band gaps and quasiparticle energies, Phys. Rev. B 34, 5390 (1986).

[28] M. Shishkin, M. Marsman, and G. Kresse, Accurate Quasiparticle Spectra from Self-Consistent GW Calculations with Vertex Corrections, Phys. Rev. Lett. 99, 246403 (2007).

[29] H. Sakakibara, T. Kotani, M. Obata, and T. Oda, Finite electricfield approach to evaluate the vertex correction for the screened Coulomb interaction in the quasiparticle self-consistent GW method, Phys. Rev. B 101, 205120 (2020).

[30] M. van Schilfgaarde, T. Kotani, and S. Faleev, Quasiparticle Self-Consistent GW Theory, Phys. Rev. Lett. 96, 226402 (2006).

[31] S. Botti and M. A. L. Marques, Strong Renormalization of the Electronic Band Gap due to Lattice Polarization in the GW Formalism, Phys. Rev. Lett. 110, 226404 (2013).

[32] C. Bhandari, M. van Schilfgaarde, T. Kotani, and W. R. L. Lambrecht, All-electron quasiparticle self-consistent GW band structures for SrTiO₃ including lattice polarization corrections in different phases, Phys. Rev. Mater. 2, 013807 (2018).

[33] A. N. Chantis, M. van Schilfgaarde, and T. Kotani, Ab Initio Prediction of Conduction Band Spin Splitting in Zinc Blende Semiconductors, Phys. Rev. Lett. 96, 086405 (2006).

[34] J. He and C. Franchini, Screened hybrid functional applied to 3d⁰ → 3dⁿ transition-metal perovskites LaM̅O₃(M = Sc–Cu): Influence of the exchange mixing parameter on the structural, electronic and magnetic properties, Phys. Rev. B 86, 235117 (2012).

[35] A. A. Mostofi, J. R. Yates, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, wannier90: A tool for obtaining maximally-localised Wannier functions, Comput. Phys. Commun. 178, 685 (2008).

[36] A. A. Mostofi, J. R. Yates, G. Pizzi, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, wannier90: An updated version of wannier90: A tool for obtaining maximally-localised wannier functions, Comput. Phys. Commun. 185, 2309 (2014).

[37] M. A. McGuire, H. Dixit, V. R. Cooper, and B. C. Sales, Coupling of crystal structure and magnetism in the layered, ferromagnetic insulator CrI₃, Chem. Mater. 27, 612 (2015).

[38] V. Carteaux, D. Brunet, G. Ouvrard, and G. Andre, Crystallographic, magnetic and electronic structures of a new layered ferromagnetic compound Cr₇Ge₇Te₆, J. Phys.: Condens. Matter 7, 69 (1995).

[39] H.-J. Deiseroth, K. Aleksandrov, C. Reiner, L. Kienle, and R. K. Kremer, Fe₃Ge₆Te₂ and Ni₃Ge₆Te₂ - two new layered transition-metal compounds: Crystal structures, HRTEM investigations, and magnetic and electrical properties, Eur. J. Inorg. Chem. 2006, 1561 (2006).
[40] A. I. Liechtenstein, V. I. Anisimov, and J. Zaanen, Density-functional theory and strong interactions: Orbital ordering in Mott-Hubbard insulators, Phys. Rev. B 52, R5467 (1995).

[41] A. G. Petukhov, I. I. Mazin, L. Chioncel, and A. I. Lichtenstein, Correlated metals and the LDA + $U$ method, Phys. Rev. B 67, 153106 (2003).

[42] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized Gradient Approximation Made Simple, Phys. Rev. Lett. 77, 3865 (1996).

[43] T. Kotani and H. Kino, Re-examination of half-metallic ferromagnetism for doped LaMnO$_3$ in a quasiparticle self-consistent GW method, J. Phys.: Condens. Matter 21, 266002 (2009).

[44] S. W. Jang, T. Kotani, H. Kino, K. Kuroki, and M. J. Han, Quasiparticle self-consistent GW study of cuprates: Electronic structure, model parameters, and the two-band theory for $T_C$, Sci. Rep. 5, 12050 (2015).

[45] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.101.241409 for more detailed band structures and density of states calculated using various methods.

[46] P. Janthon, S. A. Luo, S. M. Kozlov, F. Viñes, J. Limtrakul, D. G. Truhlar, and F. Illas, Bulk properties of transition metals: A challenge for the design of universal density functionals, J. Chem. Theory Comput. 10, 3832 (2014).

[47] Y. Meng, X.-W. Liu, C.-F. Huo, W.-P. Guo, D.-B. Cao, Q. Peng, A. Dearden, X. Gonze, Y. Yang, J. Wang, H. Jiao, Y. Li, and X.-D. Wen, When density functional approximations meet iron oxides, J. Chem. Theory Comput. 12, 5132 (2016).

[48] H. Wang, V. Eyert, and U. Schwingenschlägl, Electronic structure and magnetic ordering of the semiconducting chromium trihalides CrCl$_3$, CrBr$_3$, and CrI$_3$, J. Phys.: Condens. Matter 23, 116003 (2011).

[49] J. F. Dillon and C. E. Olson, Magnetization, resonance, and optical properties of the ferromagnet CrI$_3$, J. Appl. Phys. 36, 1259 (1965).

[50] Y. F. Li, W. Wang, W. Guo, C. Y. Gu, H. Y. Sun, L. He, J. Zhou, Z. B. Gu, Y. F. Nie, and X. Q. Pan, Electronic structure of ferromagnetic semiconductor CrGeTe$_3$ by angle-resolved photoemission spectroscopy, Phys. Rev. B 98, 125127 (2018).

[51] M. Suzuki, B. Gao, K. Koshiishi, S. Nakata, K. Hagiwara, C. Lin, Y. X. Wan, H. Kumigashira, K. Ono, S. Kang, S. Kang, J. Yu, M. Kobayashi, S.-W. Cheong, and A. Fujimori, Coulomb-interaction effect on the two-dimensional electronic structure of the van der Waals ferromagnet Cr$_2$Ge$_2$Te$_6$, Phys. Rev. B 99, 161401(R) (2019).

[52] H. Ji, R. A. Stokes, L. D. Alegria, E. C. Blomberg, M. A. Tanatar, A. Rejinders, L. M. Schoop, T. Liang, R. Prozorov, K. S. Burch, N. P. Ong, J. R. Petta, and R. J. Cava, A ferromagnetic insulating substrate for the epitaxial growth of topological insulators, J. Appl. Phys. 114, 114907 (2013).

[53] C. Ricca, I. Timrov, M. Cococcioni, N. Marzari, and U. Aschauer, Self-consistent DFT + $U$ + $V$ study of oxygen vacancies in SrTiO$_3$, arXiv:2001.06540.

[54] A. L. Kutepov, Self-consistent GW method: $O(n)$ algorithm for the polarizability and the self energy, arXiv:1911.05633.