Quasiparticle self-consistent *GW* method applied to localized 4*f* electron systems

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Rare-earth pnictides possess unique electronic and magnetic properties that make them interesting for both fundamental and practical reasons. We apply the quasiparticle self-consistent *GW* (QS*GW*) method to the rare-earth metals Gd and Er and to the rare-earth monopnictides GdN, EuN, YbN, GdAs, and ErAs. QS*GW* is unique in that it automatically takes into account both *spd* and *f* electrons in a single, unified framework without any materials-dependent parameters or special treatment of the *f* electrons. It enables us to examine approximations behind the standard local density approximation+*U* and self-interaction corrected approaches. QS*GW* reproduces the experimental occupied 4*f* levels well, though the unoccupied levels are a little overestimated. Properties of the Fermi surface responsible for electronic properties—governed by the *spd* electrons—are in good agreement with available experimental data. GdN is predicted to be very near a critical point of a metal-insulator transition.

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I. INTRODUCTION

Most 4*f* compounds belong to a class of materials whose electronic structure can be approximately described in terms of the coexistence of two subsystems—a localized *f* subsystem and an itinerant *spd* subsystem. States near the Fermi energy E_F predominantly consist of the latter; 4*f* electrons largely play a passive role except to spin polarize the *spd* subsystem through an indirect exchange mechanism. Describing both subsystems in the framework of *ab initio* electronic structure methods, however, poses a rather formidable challenge. The most widely used method, the local density approximation (LDA), fails catastrophically for open *f*-shell systems, leaving *f* electrons at E_F. To surmount these failures, a variety of strategies to extend the LDA have been developed. These include exact exchange (EXX)[1] self-interaction correction (SIC),[2] LDA+*U*,[3–5] and more recently LDA+dynamical mean-field theory (DMFT).[6] Because they are dependent on and entangled with the LDA, they have serious problems, both formal and practical. SIC, LDA+*U*, and LDA+DMFT add nonlocal potentials to certain localized electrons in a special manner, leaving some ambiguity about how a localized electron state is defined and how the double-counting term should be subtracted. Further, such approaches are specialized; they cannot remedy the LDA's inadequate description of itinerant *spd* subsystems (e.g., its well-known underestimation of semiconductor band gaps), which is the relevant one for transport properties. Thus, they are problematic for 4*f* materials such as the rare-earth monopnictides we study here. EXX has less ambiguity but suffers from many other failures. When random phase approximation (RPA) correlation is added to the EXX potential, essentially the LDA result is recovered in Si.[1] (The improvement in EXX semiconductor band gaps is artifact of fortuitous cancellation between the discontinuity in the exchange potential and neglect of correlation.) The standard *GW* (i.e., one-shot *GW* as perturbation to the LDA or $G^{\mathrm{LDA}}W^{\mathrm{LDA}}$) significantly improves on the LDA's description of itinerant *spd* subsystems, but it has many shortcomings,[7] and it fails qualitatively in open *f* systems in much the same way as the LDA fails.[7]

Rare-earth (RE) pnictides such as GdAs and GdN are interesting materials for potential spintronics applications[8] because they can be grown on conventional semiconductors such as GaAs. GdAs is believed to be a semimetallic antiferromagnet, which can be rendered ferromagnetic with slight changes in stoichiometry or an applied magnetic field. GdN is a similar compound with a ferromagnetic ground state. It is thought that GdN (or its modification) can be a half-metal and therefore an ideal spin injector. However, the electrical properties of GdN are not well established experimentally.[9–11] While the optical absorption edge of ~1 eV measured in GdN by Kaldis and Zurcher[9] suggests that it is a semiconductor, the large carrier concentration of 1.9×10^{21} cm^3 measured by Wachter and Kaldis[10] indicates that it is a semimetal. On the other hand, Xiao and Chien performed direct measurements of resistivity and concluded that GdN is an insulator.[11] Duan *et al.* recently used LDA+*U* to predict the metal-insulator transition in GdN.[3] However, such a method cannot reliably describe the relevant electronic structure near E_F, any more than the LDA can reliably predict whether Ge or ScN is an insulator or not. ErAs has also attracted interest because it has been grown epitaxially on GaAs.[4,12] Thin layers of ErAs sandwiched between AlAs and GaAs layers provide an excellent system for the study of resonant tunneling diodes based on semiconductor and/or semimetal heterostructures.[5] These possible applications are based on their unique electronic and magnetic properties originating from the interplay of the magnetic, localized 4*f* electrons with the delocalized *spd* valence electrons.

Here, we apply a recently developed quasiparticle self-consistent *GW* (QS*GW*) theory[13–16] to study the electronic structure of GdAs, GdN, and ErAs. Because it is not known whether *GW* theory—a perturbative treatment based on the random phase approximation—can reasonably describe *f* systems at all, we also consider elemental Gd and Er. We note in passing that self-consistency is essential for these studies; the customary one-shot *GW* approach, where *G* and *W* are computed from the LDA, is practically meaningless for *f* systems.[14] In addition, this work brings out some fun-

damental points. First, the position of unoccupied f levels is systematically too high. Second, we show that the shifts in f levels, which QSGW determines relative to LDA, lie outside the degrees of freedom inherent in the standard LDA+U method. The method enables us, in principle, to reconstruct parameters used in the LDA+U theory, as we will show. We show that we can use a subset of the potential that mimics LDA+U, which enables us—in principle—to reconstruct optimum choices for parameters such as U and J in LDA+U. Last, we show that GdN is near the critical point of metal-insulator transition.

II. METHOD

The method is currently implemented with a variant of the full-potential linear muffin-tin orbital (LMTO) method.[14,17] While QSGW was outlined in Ref. 13 and is described in detail elsewhere,[18] we present a minimum description below. In the usual GW approximation,[19] the self-energy $\Sigma(\mathbf{r},\mathbf{r}',\omega)$ can be computed from the eigenvalues $\{\varepsilon_i\}$ and eigenfunctions $\{\psi_i\}$ for any Hermitian one-body Hamiltonian H^0 (H^0 can be nonlocal). In QSGW, we generate a static, Hermitian, nonlocal potential $V^{\mathrm{xc}}(\mathbf{r},\mathbf{r}')$ from $\Sigma(\mathbf{r},\mathbf{r}',\omega)$ as follows:

$$V^{\mathrm{xc}} = \frac{1}{2}\sum_{ij} |\psi_i\rangle\{\mathrm{Re}[\Sigma(\varepsilon_i)]_{ij} + \mathrm{Re}[\Sigma(\varepsilon_j)]_{ij}\}\langle\psi_j|. \qquad (1)$$

"Re" signifies the Hermitian part of $\Sigma(\varepsilon_i)_{ij} = \frac{1}{2}\langle\psi_i|\Sigma(\varepsilon_i)+\Sigma^{\dagger}(\varepsilon_i)|\psi_j\rangle$. This V^{xc} together with the Hartree potential (also calculable from H^0) defines a different H^0. It is used for the input to an iteration; we repeat the procedure until H^0 is converged. Thus, QSGW is a self-consistent perturbation theory, where the self-consistency condition is constructed to minimize the size of the perturbation. QSGW is parameter free, independent of basis set and of the LDA.[18] It contains LDA+U kinds of effects, but no subsystem is singled out for specialized treatment; there are no ambiguities in double-counting terms or in what is included and what is left out of the theory. We showed that QSGW reliably describes a wide range of *spd* systems.[13] Its success in describing f systems is important because it is not known whether the GW method can reasonably describe correlated f electrons at all.

Reference 18 gives some formal justification as to why QSGW should be preferred to conventional self-consistent GW. The orbital basis and our all-electron GW are described in Ref. 7. Local orbitals (e.g., 5f states) are essential for reliable description of these systems in QSGW. It is also important not to assume time-reversal symmetry in the open f systems, as Larson *et al.* noted.[20] Also, we cannot assume that the crystal symmetry is cubic. By not making this assumption, we find that the occupation of 4f states rather closely follows Hund's rules for the atom. Unfortunately, the required computational effort increases significantly.

III. RESULTS

We considered the following 4f systems: Gd, Er, EuN, GdN, ErAs, YbN, and GdAs. Gd and Er are metals, while the rest are narrow-gap insulators or semimetals. QSGW always shifts 4f levels away from E_F. The electronic structure around E_F is dominated by *spd* electrons, which we will consider later. Figure 1 shows energy bands and quasiparticle density of states (DOS), for some cases, together with x-ray photoemission (XPS) and bremsstrahlung isochromat (BIS) spectroscopies.

In all cases, stable ferromagnetic solutions were found with the 4f element in the 3+ state: that is, 6, 7, 11, and 31f levels are occupied in Eu, Gd, Er, and Yb, respectively; the remainder are unoccupied. [Antiferromagnetic solutions were also found, but ferromagnetic solutions are presented here to compare with Shubnikov–de Haas (SdH) experiments.] Occupied 4f levels were always dispersionless, as Fig. 1 shows, while unoccupied states show some dispersion, reflecting their hybridization with the *spd* subsystem.

Gd is the only 4f element that the LDA and $G^{\mathrm{LDA}}W^{\mathrm{LDA}}$ set no f states at E_F.[26] This is because they form a closed shell (filled $f^{\uparrow}$ levels, empty $f^{\downarrow}$ levels), separated by an exchange splitting (U-J in LDA+U terminology). For the remaining 4f elements either the $4f^{\uparrow}$ or the $4f^{\downarrow}$ level is partially filled. QSGW predicts large exchange splittings *within* the partially filled channel (controlled by different combinations of U and J) (see, e.g., the ErAs DOS in Fig. 1). *Occupied* f levels are generally in reasonable agreement with available XPS data (see Table I). In the two Er compounds (Er and ErAs), occupied $f^{\uparrow}$ and $f^{\downarrow}$ levels are fairly well separated. The shallower $4f^{\downarrow}$ levels likely correspond to the XPS peak between −4.5 and −5 eV, and the $4f^{\uparrow}$ levels to the broad XPS peak between −8 and −10 eV, as shown in Fig. 1. In YbN, the separation between occupied $f^{\uparrow}$ and $f^{\downarrow}$ is small, and the XPS peak (whose width is ~3 eV) probably corresponds to some average of them. More precise identification is not possible because multiplet effects are not included. In contrast, the *unoccupied* 4f levels are systematically higher than observed BIS peaks, typically by ~3–4 eV. The only exception is ErAs, where the overestimate is closer to 1 eV. (This may well be an artifact of final-state effects in ErAs, as suggested in Ref. 4.)

Overall, the $4f^{\uparrow}$-$4f^{\downarrow}$ splitting is 16.2 eV in Gd and ~18 eV in GdN and GdAs. This *change* in the splitting is reflected in the BIS-XPS data (12.2 eV for Gd and 14 eV for GdN and GdAs). The carrier concentration at E_F is larger in Gd than in GdAs, which results in a larger dielectric response and more strongly screened U.

The *orbital moments* follow what is expected from Hund's rule. Spin moments are a little overestimated in the metals Er and Gd, following the trend observed in 3d magnetic systems such as MnAs.[13]

We think that the main reason for the deviation from the experiments can be attributed to omission of screening contributions to W. In QSGW, W is calculated from the irreducible polarization function in the RPA, which neglects the interaction between electron-hole pairs in its intermediate states (excitonic effects). Because of this, band gaps are too large by ~0.2 eV (Ref. 13) in sp semiconductors and by ~1.3 eV in NiO,[16,18] where the gap occurs between localized 3d bands and somewhere in between $\lesssim$1 eV for $SrTiO_3$ and TiO_2. The gap overestimate systematically increases with localization of the orbitals. Yet, the long-range part of W, rep-

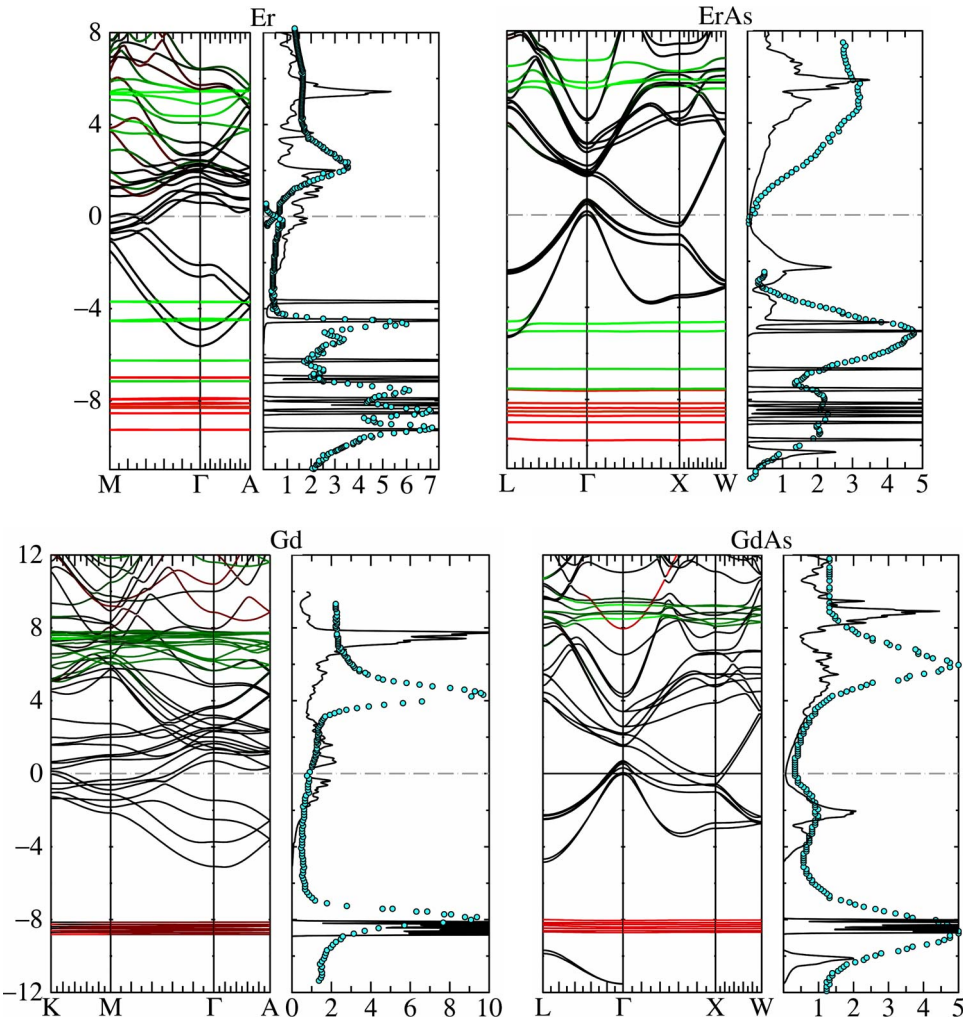

FIG. 1. (Color online) The QS*GW* energy band structure of Er, ErAs, Gd, and GdAs. Right panels show DOS, together with experimental XPS and BIS data (Refs. 4 and 21–23) (circles). E_F =0 eV. Colors indicate the spin character of the band (blue for majority and red for minority). Lattice constants were taken to be 5.80 Å (GdAs), 5.73 Å (ErAs) in the NaCl structure, and 3.64 Å (Gd) and 3.56 Å (Er) in hcp.

resented by $\epsilon(\mathbf{q}=0,\omega\rightarrow 0)$, is uniformly ≈80% of experiment[18] in all cases. Taken together, these facts indicate that the short-range part of W is more strongly affected by excitonic effects in localized systems. This is reasonable because excitonic effects in NiO should be rather atomiclike internal d-d transitions (or charge-transfer-like). As 4f electrons are more localized than 3d electrons, the short-ranged excitonic effects are still more pronounced and should reduce splittings between the occupied and unoccupied 4f states more than in, e.g., NiO. Independently of this, QS*GW* omits higher order processes, such as the effect of a third electron in a different state on an electron-hole pair excitation. Such kind of correlation is attractive to both occupied and unoccupied states, shifting both kinds to lower energy. The combination of these two effects is necessary to explain why unoccupied 4f states are too high while the occupied states are reasonably well described.

The *spd* subsystem comprises the states at E_F, which control electronic transport properties. Table II presents two of the de Haas–van Alphen (dHvA) frequencies observed in Gd. By comparing them to the calculated ones as a function of E_F, we can determine the shift in E_F required to match the dHvA data[27] and thus assess the error in those bands at E_F. Table II shows that the QS*GW* $\gamma 1$ and $\alpha 1$ should be shifted by ∼−0.2 and −0.1 eV, respectively, consistent with precision of QS*GW* for itinerant systems.[13] The QS*GW* DOS at E_F (1.84 states/eV atom) is slightly overestimated [1.57 states/eV atom (Ref. 28)].

ErAs and GdAs may be viewed as slightly negative-gap insulators (semimetals), with an electron pocket at X compensated by a hole pocket at Γ (see Fig. 1). Several experiments address the *spd* subsystem near E_F:

(i) The As-p-like Γ_{15} band dispersion between Γ and X in ErAs. As seen in Fig. 1, the X point has states at −1.2 and −0.8 eV and split-off bands at −3.52 eV. A single band from Γ to X of ∼1.5 eV width was observed by photoemission (Fig. 7 of Ref. 4).

(ii) SdH frequencies f and cyclotron masses m^* (see Table III). SdH measurements in $Er_{0.68}Sc_{0.32}As$ (Ref. 29) agree reasonably well with QS*GW* calculations for ErAs. Allen *et al.*[30] estimated $m^*=0.17$ from dc field measurements for ErScAs, which is consistent with the QS*GW* values $m^*(e_{\bar{A}\uparrow})=0.16$ and $m^*(e_{\bar{A}\downarrow})=0.13$. Nakanishi *et al.*[31] identified two branches in the [100] direction from dHvA measurements in GdAs: $m^*=0.2$ ($f=246$ T) for $m^*(e_{\bar{A}})$ and $m^*=0.26$ ($f=439$ T) for $m^*(h_2)$. These are in good agreement with QS*GW* values in Table III. Koyama *et al.*[32] obtained $m^*=0.48$ from a broad peak in cyclotron experiments. This may be understood as some kind of average of masses in Table III.

TABLE I. QSGW spin and orbital moments (bohr), average position of $4f$ levels relative to E_F (eV), and corresponding peaks in XPS and BIS data (where available). When $f^{\downarrow}$ or $f^{\uparrow}$ states are split between occupied and unoccupied levels, average positions for both occupied and unoccupied are given (top and bottom numbers). When the occupied or unoccupied part of $f^{\downarrow}$ or $f^{\uparrow}$ levels consist of multiple states split about the average (see, e.g., ErAs bands in Fig. 1), the range of splitting is denoted in parentheses.

	μ_{spin}	μ_{orb}	μ_{expt}	$f^{\uparrow}$	XPS	$f^{\downarrow}$	BIS
Gd	7.8	⋯	7.6[a]	−8.5	−8.0	7.7	4.2
				−8.1(2)	−8.4, −4.6	−4(1)	
Er	3.5	6.0	9.1[b]				
						5.1	2.1
				−6(2)			
EuN	6.0	−2.8					
				3.1		9	
GdN	7.0	⋯		−8.3	−8.5	9.5	5.8
GdAs	7.0	⋯		−7.0	−8.0	10.7	6
				−8.5(2)	−9, −4.8	−5	
ErAs	3.0	6.0					
						6	5
						−6	
YbN	1.0	3.0			−6.5		
				−7(1)		4.5	0.2

[a]Reference 24.
[b]Reference 25.

(iii) The electron concentration in the pocket at X is controlled by the (negative) gap between Γ and X. The QSGW result agrees with experiment to within the reliability of the calculated band gap ($\sim$0.2 eV): for ErAs, 3.5×10^{20} cm^{-3} QSGW, $3\pm1\times10^{20}$ cm^{-3} experiment from Ref. 31; for GdAs, 3.3×10^{20} cm^{-3} QSGW, 2.3×10^{20} cm^{-3} experiment from Ref. 12.

GdN is qualitatively similar to GdAs and ErAs, but there is some confusion as to whether GdN is an insulator or semimetal. QSGW predicts GdN to be semiconductor with a small indirect (Γ-X) band gap of 0.22 eV (Fig. 1). Except for spin polarization from the f electrons, the electronic structure for GdN around E_F is similar to ScN, with the valence band near Γ mainly of N p character and the conduction near X of cation d character. The LDA predicts the Γ-X gap to be negative in ScN, well below the experimental value of $E_g=0.9\pm0.1$ eV (Ref. 33) [see panel (a) of Fig. 2]. (The direct X-X gap is 2.15 eV.) A similarly negative gap is predicted for GdN by LDA+U: electron pockets at X are compensated by the hole pocket at Γ in the minority channel.[3] However, as we will show, LDA+U is essentially LDA-like for the spd subsystem. QSGW predicts a positive band gap in both GdN and ScN, as panel (b) shows, with $E_g\sim0.22$ eV in GdN. For the best theoretical prediction of E_g, it is necessary to account for the systematic tendency for QSGW to overestimate band gaps slightly. To do this, we adopt the "scaled Σ" approach as we did in Ref. 15. We take a hybrid of the LDA and QSGW potentials: $H^0(\alpha)=(1-\alpha)H^{0,\mathrm{QS}GW}+\alpha H^{\mathrm{LDA}}$. This accounts in a simple way for the underestimation of ϵ by QSGW. We found that $\alpha=0.2$ rather accurately reproduces experimental band gaps for a wide variety of semiconductors, insulators, and nitrides, as we will show in detail elsewhere (see Ref. 15 for III-V and II-VI semiconductors). Energy bands with $\alpha=0.2$ are shown in panel (c) of Fig. 2. Then, the ScN Γ-X and X-X gaps agree to within $\sim$0.1 eV of experiment, consistent with our general experience. It is reasonable to expect that GdN will be similarly accurate. Panel (c) shows $E_g\sim0.05$ eV. Thus, it appears likely that GdN is right on the cusp of a metal-insulator transition. The spin-averaged X-X gap (1.48 eV for majority and 0.46 eV for minority) is in close agreement with 0.98 eV (Ref. 9) measured in paramagnetic GdN.

TABLE II. Gd dHvA frequencies (T) for a magnetic field oriented along the [0001] direction. $\gamma1$ originates from the majority $6s$ band and $\alpha1$ from the majority $5d$ band. Both lie in the ΓKM plane; they are depicted in Ref. 27.

	$E_F-0.2$ eV	$E_F-0.1$ eV	E_F	Expt.[a]
$\alpha1$	4934	4260	3585	4000
$\gamma1$	7209	6099	5177	6900

[a]Reference 27.

TABLE III. QSGW cyclotron masses m^*, in units of the free electron mass m, and frequencies f (Tesla) for GdAs and ErAs. Three bands cross E_F near Γ (see Fig. 1): the heavy hole ($h1$), light hole ($h2$) and split-off hole (sh). Ellipsoids at X have two inequivalent axes, $e_{\bar{\mathrm{BC}}}$ and $e_{\bar{A}}$. Also shown are SdH frequencies measured for $Er_{0.68}Sc_{0.32}As$ (Ref. 29). Petukhov *et al.* showed that Sc doping has a modest effect on the Fermi surface, at least within the LDA (corelike $4f$) approximation (Ref. 5). $sh\uparrow$ and $sh\downarrow$ are not distinguished in experiments. Notation follows Ref. 29 except that $\uparrow$ and $\downarrow$ are exchanged.

	GdAs		ErAs		ErScAs
	m^*/m	f (T)	m^*/m	f (T)	f (T)
$e_{\bar{A}\uparrow}$	0.17	392	0.16	452	386
$e_{\bar{A}\downarrow}$	0.15	95	0.13	301	328
$e_{\bar{\mathrm{BC}}\uparrow}$	0.51	1589	0.49	1317	1111
$e_{\bar{\mathrm{BC}}\downarrow}$	0.31	386	0.44	887	941
$h1\uparrow$	0.34	1575	0.43	1642	1273
$h1\downarrow$	0.40	1433	0.45	1368	1222
$h2\uparrow$	0.23	712	0.26	726	612
$h2\downarrow$	0.26	571	0.24	590	589
$sh\uparrow$	0.12	191	0.06	174	150
$sh\downarrow$	0.08	9	0.07	25	

Connection between quasiparticle self-consistent GW and local density approximation +U

The U in LDA+U theory focuses on corrections to a particular subsystem, the f subsystem in this case. Writing H^0

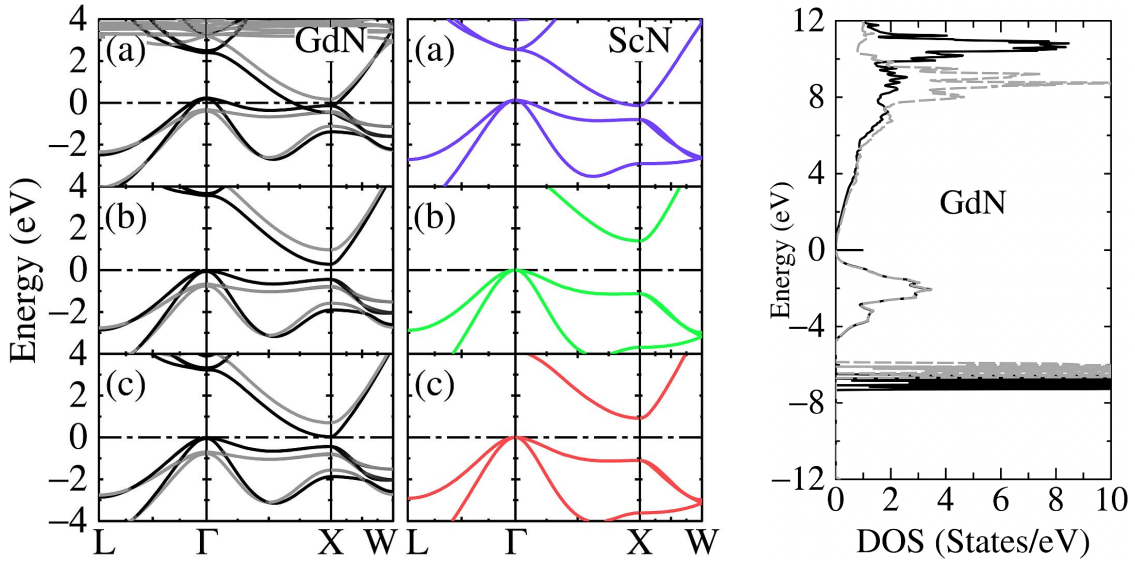

FIG. 2. (Color online) Energy bands near E_F (set at 0 eV) in GdN and ScN. For GdN, majority bands drawn as solid (red) lines and minority bands as broken (blue) lines. (a) LDA+U for GdN (U=6.7 eV and J=0.69 eV) and LDA for ScN. In both cases, the conduction band at X falls slightly below the valence band at Γ, resulting in an electron pocket at X and a hole pocket at Γ. (b) QSGW bands. A band gap is seen in both GdN and ScN. (c) scaled $\Sigma(\alpha=0.2)$ bands (see text). The ScN Γ-Γ and Γ-X gaps fall close to the measured values. The GdN gap is predicted to be ~0.05 eV. Right panel: density of states in QSGW (blue) and scaled $\Sigma(\alpha=0.2)$ (green). Scaling reduces the $4f^{\downarrow}$-$4f^{\uparrow}$ splitting, but the *spd* subsystem around E_F is little affected because the f levels are still far removed from E_F.

$=\frac{-\nabla^2}{2m}+V^{\text{eff}}$, V^{eff} can be divided as $V^{\text{eff}}=V^{\text{LDA}}+\Delta V$. In LDA+$U$, ΔV is limited to matrix elements of f orbitals at the same site. The onsite f-f block is usually specified through parameters U and J, which specifies ΔV.[34] Usually, U and J are determined empirically. We can compare the effect of QSGW on the f subsystem as follows. Matrix elements of V^{xc} can be rotated into the (Bloch-summed) orbital basis by performing an inverse Bloch sum. This yields V^{xc}, and consequently H^0, in the original real-space basis of atom-centered orbitals where the LDA+U potential is constructed. In QSGW, $\Delta V=V^{\text{xc}}-V^{\text{xc,LDA}}$; but now, ΔV connects *all pairs* of orbitals. An analog of the LDA+U potential is obtained by restricting ΔV to the onsite f-f block (QSGW[f-f]).

The result is shown in Fig. 3 (without further self-consistency), together with QSGW and LDA+U calculations. QSGW[f-f] does not reproduce *spd* subsystem around E_F, but instead is in almost perfect agreement with LDA+U. This is reasonable because *spd* subsystem is described essentially by the LDA in both cases. On the other hand, QSGW[f-f] reproduces the position of f levels in QSGW very well. Significantly, *no choice* of the standard LDA+U parameters can reproduce the QSGW ($f^{\downarrow}$,$f^{\uparrow}$) levels simultaneously because their average position is essentially determined from the LDA:[34] $\varepsilon^{(\downarrow,\uparrow),\text{LDA}+U}=\varepsilon^{(\downarrow,\uparrow),\text{LDA}}\pm(U-J)/2$.

The QSGW[f-f] construction suggests an alternate way to address a long-standing problem, namely, how to determine U. Following standard arguments,[34] U and J can be computed from the self-consistent $W(\mathbf{r},\mathbf{r}',\omega=0)$. An alternate way is to choose U and J directly so as to reproduce ΔV by QSGW as much as possible (density matrix with U and J determine ΔV). Though there are limitations of LDA+U as discussed above, we will be able to determine the optimum U with QSGW as a reference.

IV. CONCLUSION

In conclusion, we have applied QSGW theory to several rare-earth pnictides. We showed how QSGW theory connects

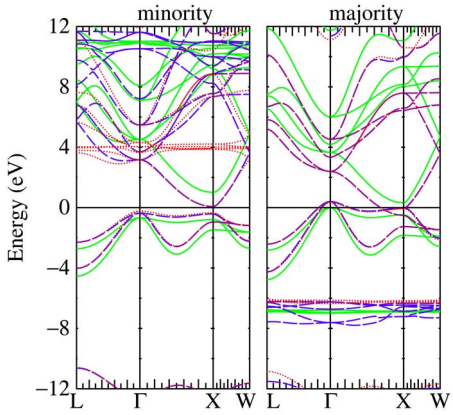

FIG. 3. (Color online) Energy bands of GdN computed by three different methods. Solid (green): QSGW; dotted (red): LDA+U, with U=6.7 and J=0.69 eV; broken (blue): QSGW[f-f], where $V^{\text{xc}}-V^{\text{xc,LDA}}$ is added only for matrix elements between f orbitals centered at the same site. Spin orbit was omitted. LDA+U and QSGW[f-f] are very similar for the *spd* subsystem, particularly around E_F. The majority-spin conduction band at X falls below the valence band maximum at Γ in both cases, whereas QSGW shows a small gap. QSGW and QSGW[f-f] are very similar for the f subsystem except for some unphysical dispersion in the QSGW[f-f] case. Restricting the QSGW potential to a particular orbital block is a basis-dependent operation. Here, it can be traced to tails of the f LMTO's, as they are not restricted to the Gd site where they are centered. The dispersion is thus an artifact of the choice of basis.

to the standard LDA+U approach. As in LDA+U, large on-site exchange splittings of the f levels sweep states of f character away from E_F (self-consistency is essential to obtain this result). Occupied f levels are in reasonable agreement with experiment, but unoccupied f levels were somewhat overestimated. We showed how contributions beyond LDA+U are necessary to properly describe states of *spd* character and how the QS*GW* potential has degrees of freedom beyond standard LDA+U even for the f states. QS*GW* contains such effects automatically in a simple and unified manner, without any tunable parameters. We compared a variety of electronic properties associated with states near E_F and obtained reasonable agreement with available experiments (detailed comparison is difficult because of experimental limitations). We predicted that GdN is near the critical point of a metal-insulator transition.

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