King's Research Portal

DOI:
10.1103/PhysRevB.95.041112

Document Version
Publisher's PDF, also known as Version of record

Link to publication record in King's Research Portal

Citation for published version (APA):

Citing this paper
Please note that where the full-text provided on King's Research Portal is the Author Accepted Manuscript or Post-Print version this may differ from the final Published version. If citing, it is advised that you check and use the publisher's definitive version for pagination, volume/issue, and date of publication details. And where the final published version is provided on the Research Portal, if citing you are again advised to check the publisher's website for any subsequent corrections.

General rights
Copyright and moral rights for the publications made accessible in the Research Portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognize and abide by the legal requirements associated with these rights.

•Users may download and print one copy of any publication from the Research Portal for the purpose of private study or research.
•You may not further distribute the material or use it for any profit-making activity or commercial gain
•You may freely distribute the URL identifying the publication in the Research Portal

Take down policy
If you believe that this document breaches copyright please contact librarypure@kcl.ac.uk providing details, and we will remove access to the work immediately and investigate your claim.

Download date: 22. Dec. 2018
Self-energies in itinerant magnets: A focus on Fe and Ni

Lorenzo Sponza, Paolo Pisanti, Alena Vishina, Dimitar Pashov, Cedric Weber, and Mark van Schilfgaarde
King’s College London, London WC2R 2LS, United Kingdom

Swagata Acharya
Indian Institute of Technology, Kharagpur 721302, India
and King’s College London, London WC2R 2LS, United Kingdom

Julien Vidal
EDF R&D, Department EFESÉ, 6 Quai Watier, 78401 Chatou, France

Gabriel Kotliar
The State University of New Jersey, 136 Frelinghuysen Road, Piscataway, New Jersey 08854, USA
(Received 21 March 2016; revised manuscript received 6 December 2016; published 26 January 2017)

We present a detailed study of local and nonlocal correlations in the electronic structure of elemental transition metals carried out by means of the quasiparticle self-consistent GW (QSGW) and dynamical mean field theory (DMFT). Recent high resolution ARPES and Haas-van Alphen data of two typical transition metal systems (Fe and Ni) are used as a case study. (i) We find that the properties of Fe are very well described by QSGW. Agreement with cyclotron and very clean ARPES measurements is excellent, provided that final-state scattering is taken into account. This establishes the exceptional reliability of QSGW also in metallic systems. (ii) Nonetheless QSGW alone is not able to provide an adequate description of the Ni ARPES data due to strong local spin fluctuations. We surmount this deficiency by combining nonlocal charge fluctuations in QSGW with local spin fluctuations in DMFT. (iii) Finally we show that the dynamics of the local fluctuations are actually not crucial. The addition of an external static field can lead to similarly good results if nonlocal correlations are included through QSGW.

DOI: 10.1103/PhysRevB.95.041112

High-resolution spectroscopy is limited in transition metals, in part because it is difficult to make sufficiently high quality samples. Fe and Ni are elements of which high quality films have been grown, and high-resolution angle-resolved photoemission spectroscopy (ARPES) performed [1]. These experiments provide a good reference to test the validity of different approximations of the electronic structure.

There are also not many calculations of spectral functions in these materials. Fe has been studied in the local-density approximation (LDA) [2] and with corrections through dynamical-mean field theory (DMFT) [3]. It is not surprising that the LDA does not track the ARPES experiment well [4], but it has been found that LDA+DMFT also fails to properly account for ARPES data [3]. The GW approximation [5] is widely applied to many kinds of insulators, but how well it describes 3d transition metals is much less established.

Through quasiparticle self-consistency (QSGW) one determines the noninteracting Green’s function \( G_0 \) which is minimally distant from the true Green’s function \( G \) [6–8]. Within QSGW many electronic properties are in excellent agreement with experiment [6], most notably the quasiparticle band structures. Moreover, at self-consistency the poles of QSGW \( G_0(k,\omega) \) coincide with the peaks in \( G(k,\omega) \). This means that there is no many-body “mass renormalization” of the noninteracting Hamiltonian, which allows for a direct association of QSGW energy bands \( E(k) \) with peaks in the spectral function \( A(k,\omega) \). Thus, QSGW provides an optimum framework to test the range of validity and the limitations to the GW approximation.

In this Rapid Communication, we compare QSGW results to various experimental data in elemental 3d materials in the Fermi liquid (FL) regime, with a heavy focus on Fe because of the high quality of ARPES [1] and de Haas-van Alphen (dHvA) [9,10] data available. We will show that QSGW and ARPES spectral functions agree to within experimental resolution, with the proviso that the final state scattering is properly accounted for in interpreting the experimental data. By contrast, discrepancies appear in Ni—a classical itinerant ferromagnet. This can be attributed to the lack of spin fluctuations in GW diagrams. However we find out that there is no need to include finite-energy spin fluctuations, instead a static correction to the QSGW self-energy is sufficient to correct for the size of the local moment. This finding opens up an avenue to test the validity of a similar argument for other transition metals. The LDA or LDA+DMFT should be problematic, as nonlocality in the self-energy can be important (see Supplemental Material).

I. FE IN THE FERMI LIQUID REGIME

Figure 1 compares the calculated QSGW band structure of Fe to peaks in ARPES spectra of Ref. [1], along with some inverse photoemission data [11]. While agreement appears to be very good, there are some discrepancies, particularly along the \( \Gamma \)–H line [see also Fig. 2(a)]. As noted earlier, the QSGW band structure reflects the peaks of \( A(k,\omega) \) with no renormalizations from the \( \omega \) or \( k \) dependence of \( \Sigma \).

In the FL regime, ARPES spectra \( I(k,\omega) \) are generally thought to be a fairly direct measure of \( A(k,\omega) \). But the two
be written as normal to the film surface. (d) dispersion in the QS

FIG. 1. Left: QSWG band structure of Fe (solid lines), LSDA
(gray dashed), k-point averaged QSWG (black dotted, see text),
ARPES spectra [1] (diamonds), and inverse photoemission spec-
reported in Ref. [1]. Red and green depict majority and minority d
character, respectively.

are not identical even in the FL regime, independently of
the precision of the experimental setup. Assuming a one-step
model [12] for the photoemission process (initial and final
state coupled through Fermi’s golden rule [12,13]) I(k,ω) can
be written as

\[ I(k,\omega) \propto \int dk_\perp |T_{f} f|^2 M_{f}(k_\perp)^2 A_{f}(k_\perp)A(k,\omega), \]

where \( A_{f}(k_\perp) = \frac{\Delta k_\perp / 2\pi}{(\Delta k_\perp / 2)^2 + (k_\perp - k_0^B)^2} \)

(1)
is the spectral function of the final state, broadened by scat-
tering of the photoelectron as it approaches the surface [14].
\( T_{f} f \) is the final-state surface transmission amplitude and \( M_{f} \)
the photoexcitation matrix element (taken to be constant and
\( k \) independent [15]). Thus the final state is considered to
be a damped Bloch wave, taking the form of a Lorentzian
distribution centered in \( k_0^B \) and broadened by \( \Delta k_\perp \) [14],
while the initial state is an undamped Bloch function with an
energy broadening \( \Delta E \), obtained through the QSWG spectral
function. This approximation is reasonable since in the FL
regime \( A(k,\omega) \) is sharply peaked around the QP level. \( \Delta k_\perp \)
directly related to the inverse of the electron mean free
path. For photon energy in the range 100–130 eV, \( \Delta k_\perp \approx
0.2 \text{ Å}^{-1} [16,17] \).

The final-state scattering broadens \( I(\omega) \), but it also can
shift the peak \( \bar{\omega} \) in \( I(\omega) \). The most significant discrepan-
cy between ARPS and QSWG is found in the \( V_m \) band, Fig. 2(a)
between \( k = 0 \) and 0.4 × \( H \). Figure 2(b) shows \( A(k,\omega) \)
calculated by QSWG, and the corresponding \( I(k,\omega) \) calculated
from Eq. (1). Estimating the peak shift change from \( \delta \bar{\omega} = f d\omega I f d\omega I - f d\omega A f d\omega A \), we find \( \delta \bar{\omega} \approx 0.01 \text{ eV} \)
at \( \Gamma \), increasing to \( \delta \bar{\omega} \approx 0.06 \text{ eV} \) for \( k \) between 0.1H
and 0.3H. \( \delta \bar{\omega} = 0.06 \text{ eV} \) tallies closely with the discrepan-
cy between the \( V_m \) band and the measured ARPS peak for
0.1H < \( k < 0.3H \). There is also a significant discrepancy in
the \( I_{\text{II}} \) band near \( k = 0.77 \times \text{H} \). Where it crosses \( E_F \),
the QSWG bands deviate from the ARPS peak by nearly 0.15 eV.
But ARPS simulated by Eq. (1) is much closer to experiment
[Fig. 2(c)]. This is understood from Fig. 2(d), which plots the
QSWG dispersion along a line \( \Delta k_\perp \) normal to the film surface,
passing through [0,0,0.77H]. A measurement that includes
contributions from this line biases the ARPS peak in the
direction of \( E_F \) since \( E_{\text{QP}} \) is minimum at \( k_{\perp} = 0 \). Thus we
attribute most of the discrepancy in the Fermi surface crossing
[red star in Fig. 1(b)] to an artifact of final-state scattering.

To better pin down the errors in QSWG, we turn to de
Haas-van Alphen (dHvA) measurements. Extremal areas of
the FS cross sections can be extracted to high precision from
dHvA and magnetoresistance experiments. Areas normal to
[110] and [111] are given in Table I, along with areas calculated
by QSWG. Figure 1 shows the QSWG Fermi surface, which
closely resembles the one inferred by Lonzarich (version B) [19].
There is some ambiguity in resolving the small \( V_{\text{III}} \)
pocket at \( N \) because its tiny area is sensitive to computational
details. Discrepancies in the extremal areas are not very
meaningful: It is more sensible to determine the change \( \Delta E_F \)
in Fermi level needed to make the QSWG area agree with
dHvA measurements. This amounts to the average error in
the QSWG QP levels, assuming that the bands are rigidly. This
assumption is well verified for all pockets, except for the small
VI one owing to strong electron-phonon coupling [20].

Some limited cyclotron data for effective masses are also
available [18], which are expected to be more reliable than
ARPES data. It is seen that agreement is excellent (Table I,
bottom panel) except for the small VI pocket. We get a better
comparison by accounting for the electron-phonon coupling
with a simple model [20,21]. From the model, \( v_F \) is renor-
malized by a factor \( 1 + \lambda = 1.6 \), which reasonably accounts
for discrepancy between the QSWG and the cyclotron mass in
pocket VI. The other pockets are much larger [Fig. 1(b)], mak-
ing \( v_F \) much larger on average and the renormalization smaller.

Such a perfect agreement with experiments could not
be possible without the accurate description of nonlocal
components in the QSWG self-energy. To prove this statement

FIG. 2. (a) The \( \Gamma-H \) line of Fig. 1 in high resolution. Labels
correspond to traditional assignments of Fermi surface pockets [1,9].
(b) Dashed line is QSWG spectral function \( A(k,\omega) \) for various points
on the \( \Gamma-H \) line, with \( k = 0 \) and \( k = 1 \) denoting \( \Gamma \) and \( H \). Solid
line is \( A(\omega) \) modified according to Eq. (1). (c) The analog of (b)
for \( k = 0.77 \times \text{H} \). Where \( I_{\text{II}} \) band crosses \( E_F \). \( E_{\text{QP}} \) indicates
the QSWG QP level, and \( E_{\text{ARPES}} \) the experimental ARPES peak at 0.77H.
(d) dispersion in the QSWG \( I_{\text{II}} \) band on a line \( k_\perp = [0,0.77H] \)
normal to the film surface.
TABLE I. de Haas-van Alphen measurements of extremal areas $A$ on the [110] and [111] Fermi surfaces, in $\AA^{-2}$. $\Delta E_F$ is an estimate of the error in the QP level (eV), as described in the text. Bottom panel: cyclotron mass, $m^*/m = (\hbar^2/2\pi m) \partial A/\partial E$.

<table>
<thead>
<tr>
<th>FS pocket</th>
<th>$d\text{HvA}$ [110]</th>
<th>$d\text{HvA}$ [111]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\text{QSGW}$</td>
<td>$\text{expt.} [9]$</td>
</tr>
<tr>
<td>I</td>
<td>3.355</td>
<td>3.3336</td>
</tr>
<tr>
<td>II</td>
<td>2.5</td>
<td>2.0</td>
</tr>
<tr>
<td>III</td>
<td>0.2138</td>
<td>0.3190</td>
</tr>
<tr>
<td>IV</td>
<td>0.0897</td>
<td>0.1175</td>
</tr>
<tr>
<td>VI</td>
<td>0.3176</td>
<td>0.5559</td>
</tr>
<tr>
<td>VII</td>
<td>0.0148</td>
<td>0.0405</td>
</tr>
</tbody>
</table>

---

we computed the band structure with a local potential obtained from a $k$-point average of the QSGW self-energy. The result is reported as a dotted black curve in Fig. 1, to be compared with the pale gray lines of LSDA and the solid lines of QSGW. The $k$-averaged potential reproduces a band structure that is much closer to the LSDA one than to the QSGW results. This results in the overestimation of the binding energy, e.g., of most states close to Fermi (for instance at $\Gamma$), or in the range between $-2$ and $-3$ eV (see at $\Gamma$, $P$, and $H$). An additional verification that local physics is not relevant in the description of the quasiparticle structure of Fe can be found in the Supplemental Material [20].

**II. Ni: An Archetypical Itinerant Magnet**

Less detailed information is available for other elemental transition metals. We have extracted some experimental bandwidths and also the exchange splitting $\Delta E_s$ in the magnetic elements. Figure 3(a) shows that both seem to be very well described by QSGW, except that $\Delta E_s$ deviates strongly from experiment in Ni. QSGW significantly improves not only on the LSDA, but also on fully self-consistent GW [22] because of loss of spectral weight in fully self-consistent $G$ that is avoided in QSGW [6].

Figure 4(a) compares the QSGW band structure of Ni to ARPES data [23]. Agreement is excellent in the minority channel, but $\Delta E_s$ is uniformly too large on the symmetry lines shown. Also the band near $-1$ eV at L (consisting of $s$ character there) is traditionally assumed to be a continuation of the $d$ band denoted as white and green diamonds, but the calculations show that at it is a continuation of the Ni $s$ band. The corresponding LSDA band (light dotted lines) crosses L at $E_F = 0.44$ eV; also the $d$ bands are much wider. $\Delta E_s$ is about twice too large in both QSGW and the LSDA, and for that reason spin wave frequencies are also too large [24]. Spin fluctuations ($M^2$) are important generally in itinerant magnets but they are absent in both LSDA and QSGW. One important property they have is to reduce the average magnetic moment $\langle M \rangle$ and hence to quench $\Delta E_s$ [25,26]. Figure 3(b) shows this trend quite clearly: Systems such as Fe, Co, and NiO are very well described by QSGW, but $M$ is always overestimated in itinerant magnets such as FeAl, Ni$_3$Al, and Fe based superconductors such as BaFe$_2$As$_2$. Ni is

---

FIG. 3. (a): $d$ bandwidth (top panel) and exchange splitting $\Delta E_s$ (bottom panel) in the 3$d$ elemental metals. (b): Magnetic moment of several compounds.
also itinerant to some degree (unlike Fe, its average moment probably disappears as $T \to T_C$), and its moment should be overestimated. This is found to be the case for QSGW, as Fig. 3(b) shows.

Local spin fluctuations are well captured by localized nonperturbative approaches such as DMFT. We can reasonably expect that the addition of spin-flip diagrams to QSGW would be sufficient to incorporate these effects. A $G_0W_0$+DMFT study of ferromagnetic Ni can be found in Ref. [28], but the dependency of $G_0W_0$ on the starting point, together with all the advantages mentioned at the beginning, motivated us to devise a QSGW-based approach.

Here we adopt an implementation merging QSGW with DMFT. We adapted Haule’s continuous time quantum Monte Carlo solver [29,30] with the projection and embedding schemes described in Ref. [31], and which are outlined in the Supplemental Material [20]. In this sense, this method is close to the one introduced by some of us in [32], but based on a different self-consistent scheme for the solution of the weakly correlated part.

The fully consistent QSGW+DMFT calculation is composed by alternately repeated DMFT and QSGW loops. First the QSGW Green’s function is converged at fixed density, then it is projected on the Ni $d$ orbitals and finally, within the DMFT loop, the local self-energy is obtained. Updating the total density with the locally corrected Green’s function and repeating the procedure leads to complete self-consistency. This method fully takes into account the dynamics of the local spin fluctuations included in the DMFT diagrams. Results are reported in Table II and they confirm that DMFT adds the correct local diagrams missing in the QSGW theory. Moreover by carefully continuing the resulting Green’s function on the real-frequency axis, we find an exchange splitting of ~0.3 eV and a satellite at ~5 eV [20].

In order to investigate the importance of the dynamics in the local spin-spin channels, we carry out a QSGW+DMFT calculation by retaining only the static limit of the DMFT loop (we call it QSGW+“SLDMFT”). Once the DMFT loop converges, we take the zero frequency limit of the magnetic part of the DMFT self-energy and add it to the spin-averaged QSGW Hamiltonian [20]. As it is clearly shown in Table II and Fig. 4, this static Hamiltonian reproduces very accurately magnetic moment and details of the band structure. This is a strong indication that for Ni the dynamics of local spin fluctuations is not crucial. This will be the case if the quasiparticle picture is a reasonable description of Ni, even if QSGW alone does not contain enough physics to yield an optimum quasiparticle approximation.

To verify this further, we model spin fluctuations by carrying out the QSGW self-consistent cycle in the presence of a magnetic field $B^{\text{eff}}$, and tuning $B^{\text{eff}}$ to reduce $M$ (see inset, Fig. 4). Our key finding is that when $B^{\text{eff}}$ is tuned to make $M$ agree with experiment, $\Delta E_s$ does also, reproducing ARPES spectra to high precision in the FL regime, as clearly reported in Fig. 4 and Table II. Both the QSGW and LSDA overestimate $M$ for itinerant systems, but the latter also underestimates it in local-moment systems [Fig. 3(b)]. In the LSDA treatment of Ni, these effects cancel and render the moment fortuitously good. When spin fluctuations are folded in through $B^{\text{eff}}$, the LSDA moment becomes too small. This finding must be interpreted as a sign of the superior level of internal consistency in the QSGW theory with respect to LSDA. Without such a degree of consistency spin fluctuations could not be approximated by a static field.

### III. CONCLUSIONS

We have performed detailed QSGW calculations of the electronic band structure of several 3$d$ metallic compounds to assess the reliability of this theory in the Fermi liquid regime and the importance of the nonlocal terms in the self-energy.

---Fe: Through de Haas-van Alphen and cyclotron measurements we established that QSGW QP levels at $E_F$ have an error of ~0.05 eV, and effective masses are well described. Comparable precision is found below $E_F$ by comparing to ARPES data, provided final state scattering is taken into account. The QSGW $d$ bandwidth falls in close agreement with ARPES and is approximately 0.75 times that of the LDA (Fig. 1).

If $\Sigma$ is $k$-averaged to simulate a local self-energy, the QSGW band structure changes significantly and resembles the LDA. Thus nonlocality in the self-energy is important in transition metals, and its absence explains why LDA+DMFT does not yield good agreement with ARPES [3].

---Ni: QSGW $d$ bandwidths, the $t_2g-e_g$ splitting, the $s$-$d$ alignment, are all in excellent agreement with experiment, while $\langle M \rangle$ and $\Delta E_s$ are too large. However through the addition of a uniform static external field QSGW can give both in good agreement, indicating a high level of consistency in the theory, contrary to LSDA in which it is not possible to have both quantities correct at the same time.

To account for spin fluctuations in an *ab initio* framework, we constructed a QSGW+DMFT implementation and we utilized it at different degrees of approximations demonstrating that in itinerant magnets as Ni (i) the dynamics of fluctuations is irrelevant and (ii) their effect can be very well approximated by a static field as long as the nonlocal correlation part is treated accurately.

### ACKNOWLEDGMENTS

This work was supported by the Simons Many-Electron Collaboration and EPSRC Grant No. EP/M011631/1. The authors gratefully acknowledge computer resources from the Gauss Centre for Supercomputing e.V. (www.gauss-centre.eu). We also acknowledge the Partnership for Advanced Computing in Europe (PRACE Project No. 2010PA2762).
for awarding us access to the following resources: Curie FN and TN based in France at the Très Grand Centre de Calcul (TGCC), and SuperMUC, based in Germany at Leibniz Supercomputing Centre.

[28] S. Biermann, F. Aryasetiawan, and A. Georges, Phys. Rev. Lett. 90, 086402 (2003). The main accomplishment of this work was to reproduce within GW+DMFT the satellite at $\sim -6$ eV. This is also captured by LDA+DMFT [3].