

Erratum: Effect of charge self-consistency in DFT+DMFT calculations for complex transition metal oxides [Phys. Rev. Research 2, 033088 (2020)]

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The results presented in our original article were affected by a bug in the workflow used for our density functional theory plus dynamical mean-field theory (DFT+DMFT) calculations. Within this workflow, which was based on the implementation outlined in Ref. [1], a correction to the occupation matrix within the *correlated subspace* is obtained after each DMFT step, which is then used to recalculate the charge density. This new charge density defines the effective potential for the DFT Kohn-Sham (KS) Hamiltonian. Solving the KS Hamiltonian in turn provides the input for the next DMFT step. This procedure is then repeated until self-consistency is obtained. For comprehensive details we refer to Ref. [1].

In short, the bug caused only the diagonal components of the DMFT-corrected occupation matrix in the KS basis to be properly included when updating the charge density for the next DFT step, while the off-diagonal components of the occupation matrix were essentially ignored. As a result, the DFT+DMFT calculations in our work achieved charge self-consistency only with respect to the diagonal occupations.

More specifically, the bug originated from the simultaneous update of charge density and KS wave functions during the process of determining the electronic ground state within the Vienna *ab initio* simulation package (VASP) [2,3]. In this procedure, the KS eigenvalues and eigenstates are obtained through an iterative diagonalization method, where each iteration also updates the charge density based on the current KS states. Within the DFT+DMFT workflow, the charge density is computed using both the updated KS states and the DMFT-corrected occupations. These corrected occupations are computed using the *projector functions* [$P_{mv}(\mathbf{k})$, see Ref. [1]], which encode the transformation between the KS states, $|\psi_{v\mathbf{k}}\rangle$, and the correlated subspace basis, $|\chi_m\rangle$. Importantly, in the original workflow used in our article, the projector functions were associated with the KS states from the previous DFT iteration, and a few iterative optimization steps were done between each DMFT step to converge the new KS states after the charge density has been updated from DMFT. However, during the numerical diagonalization, the updated KS states will gain arbitrary phase factors, which do not match the phases of the projector functions obtained from

the KS states of the previous iteration. These arbitrary phase factors cancel out when evaluating the diagonal components of the charge density correction, but affect the off-diagonal components in an uncontrolled way, thereby essentially rendering the off-diagonal components to random noise.

This bug can in principle be corrected either by recalculating both the projector functions and the charge density correction using the updated KS wave functions in each iterative diagonalization step, or by using the exact diagonalization algorithm in VASP. However, the latter makes the calculation extremely costly. Hence, in practice, this means that one has to recalculate the joined DFT+DMFT charge density after each update to the Kohn-Sham states with the updated projector functions. After detecting the bug and discussing with the authors of Ref. [1], we now updated the corresponding TRIQS/DFTTools [4] interface and online documentation. Furthermore, the implementation of DFT+DMFT utilized in our calculations [5] has also been updated and corrected accordingly in version 3.2.0 [6].

The severity of this bug depends highly on the investigated system, and thus has not been detected in the original implementation of Ref. [1], where only cases with high symmetry have been tested. Even for the example of a monolayer of SrVO₃ with tetragonal symmetry investigated in Ref. [1], the occupations within the t_{2g} shell are purely diagonal, and thus the recalculation of the charge density is completely unaffected by this bug. However, for systems with lower symmetry, such as strained orthorhombic CaVO₃ and the rare-earth nickelate compounds studied in our article, off-diagonal occupations appear in the charge density update, and thus the corresponding calculations were affected by the bug. As a result, the effect of the charge density update on the results was generally underestimated in our article, rendering these calculations much closer to “one-shot” calculations, while the overall qualitative trends discussed in our article remain valid.

Specifically, for the case of the orbital polarization in CaVO₃, discussed in Sec. III A of our article, the correct charge self-consistent calculation leads to a somewhat stronger suppression of the orbital polarization, as can be seen in the upper panel of Fig. 1 for the case of strained CaVO₃. Furthermore, the correct charge density update now also leads to a slightly larger shift of the critical U value for the metal-insulator transition by 0.1 eV. For comparison, we also include corresponding data obtained from charge-self-consistent calculations using a different implementation based

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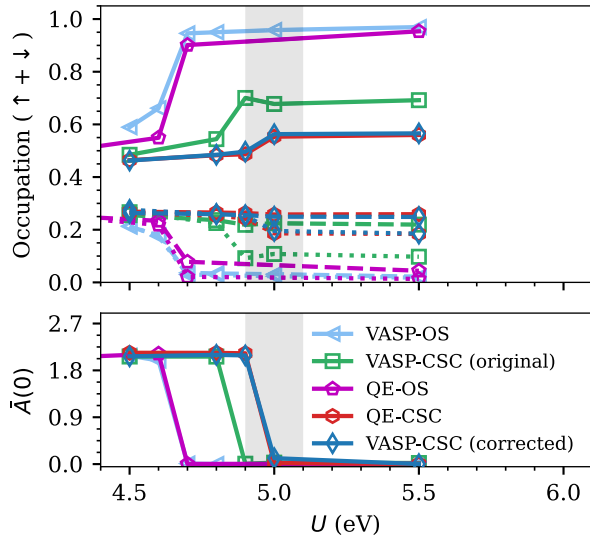


FIG. 1. Update to Figs. 2(c) and 2(d) in the original article. Evolution of orbital occupation (top) and spectral weight at the Fermi level $\tilde{A}(0)$ (bottom) as a function of interaction strength U in strained CaVO_3 . Comparison between “one-shot” (OS) and charge self-consistent (CSC) calculations between VASP and QE, showing the original and corrected VASP CSC calculations. Solid, dashed, and dotted lines in the top panel refer to the three different t_{2g} orbitals.

on QUANTUM ESPRESSO [7] (QE) that was not available at the time of publication of our original article, which agrees very well with the corrected VASP implementation.

For the charge ordered (or charge disproportionated) state in LuNiO_3 , we had identified and discussed two competing effects related to charge self-consistency in our original article. First, the use of DMFT occupations to evaluate the double-counting corrections strongly favors a charge disproportionated state, since the double-counting correction effectively lowers the energy of the more occupied site relative to the less occupied site, and thus provides a strong positive feedback mechanism supporting a charge redistribution. While this is not strictly an effect of charge self-consistency, the introduction of charge self-consistency does remove any ambiguity of whether to use DFT or DMFT occupations to evaluate the double-counting correction compared to one-shot calculations. In the charge-self-consistent case, only the

DMFT occupations carry any physical meaning since they determine the charge density for which the KS potential is obtained. Therefore, all conclusions related to the use of DMFT occupations to evaluate the double-counting correction remain valid and are unaffected by the bug in our original calculations.

The second effect discussed in our article was the effect of the actual charge density update, which was found to suppress the emergence of charge disproportionation compared to the corresponding one-shot calculation (with DMFT occupations used for the double-counting correction). This finding also remains valid qualitatively, but was significantly underestimated quantitatively by the bug related to the off-diagonal components of the DMFT-corrected occupation matrix. The full charge density update, with all off-diagonal components correctly taken into account, leads to a significantly stronger suppression of the corresponding charge redistribution. For the values of $U = 1.85$ eV and $J = 0.42$ eV used for most LuNiO_3 calculations in our article, the charge disproportionated state even becomes energetically completely unfavorable. This has by now been demonstrated and discussed in Ref. [8], and is also similar to what has been observed for the related case of potential charge disproportionation in SrCrO_3 in Ref. [9]. A charge disproportionated state in LuNiO_3 , in agreement with experimental observations, can nevertheless be obtained by increasing U to values of $U \gtrsim 2.3$ eV, which has been explicitly demonstrated in Ref. [8] [see Fig. 4(a) in that work, which can directly be compared with Fig. 5(a) of our original article]. Furthermore, Ref. [8] also argues that such a larger value of $U \approx 2.7$ eV is in fact more realistic for LuNiO_3 and can be obtained from calculations using the constrained random phase approximation with the correct charge transfer energy, i.e., the energetic distance between the transition metal d and ligand p bands.

In view of the new results and insights now available in Ref. [8], we refrain from explicitly recalculating the corresponding data presented in Figs. 4 and 5 of our original article and instead refer to Ref. [8]. We further emphasize that the results presented in our article should be interpreted as obtained from a partial self-consistency with respect to only the diagonal components of the occupations matrix.

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- [1] M. Schüler, O. E. Peil, G. J. Kraberger, R. Pordzik, M. Marsman, G. Kresse, T. O. Wehling, and M. Aichhorn, Charge self-consistent many-body corrections using optimized projected localized orbitals, *J. Phys.: Condens. Matter* **30**, 475901 (2018).
- [2] G. Kresse and J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [3] G. Kresse and D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Phys. Rev. B* **59**, 1758 (1999).
- [4] M. Aichhorn, L. Pourovskii, P. Seth, V. Vildosola, M. Zingl, O. Peil, X. Deng, J. Mravlje, G. Kraberger, C.

- Martins, M. Ferrero, and O. Parcollet, TRIQS/DFTTools: A TRIQS application for *ab initio* calculations of correlated materials, *Comput. Phys. Commun.* **204**, 200 (2016).
- [5] A. Hampel, S. Beck, and C. Ederer, soliDMFT, <https://github.com/materialtheory/soliDMFT> (2019).
- [6] A. Hampel, N. Wentzell, D. Simon, M. Merkel, S. Beck, A. Carta, O. Parcollet, H. U. R. Strand, P. Dumitrescu, T. Hahn, and H. LaBollita, TRIQS/solid_dmft: Version 3.2.0, Zenodo, doi:10.5281/zenodo.8308203 (2023).
- [7] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I.

- Dabo, A. D. Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos *et al.*, QUANTUM ESPRESSO: A modular and open-source software project for quantum simulations of materials, *J. Phys.: Condens. Matter* **21**, 395502 (2009).
- [8] A. Carta, A. Panda, and C. Ederer, Explicit demonstration of the equivalence between DFT+ U and the Hartree-Fock limit of DFT+DMFT, *Phys. Rev. Res.* **7**, 013289 (2025).
- [9] A. Carta, A. Panda, and C. Ederer, Emergence of a potential charge-disproportionated insulating state in SrCrO₃, *Phys. Rev. Res.* **6**, 023240 (2024).