

Bridging Constrained Random-Phase Approximation and Linear Response Theory for Computing Hubbard Parameters

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The predictive accuracy of popular extensions to density-functional theory (DFT) such as DFT + U and DFT plus dynamical mean-field theory (DFT + DMFT) hinges on using realistic values for the screened Coulomb interaction U . Here, we present a systematic comparison of the two most widely used approaches to compute this parameter, i.e., linear response theory (LRT) and the constrained random-phase approximation (cRPA), using a unified framework based on the use of maximally localized Wannier functions. We show that, even in cases of partially filled well-separated interacting subspaces, for which there is no ambiguity in the application of both techniques, the U in LRT and cRPA can differ as much as 30%. We demonstrate that this discrepancy arises from two main differences: neglecting the response of the exchange-correlation potential in cRPA and additional excitation channels in LRT. By taking these differences into account, we can achieve near perfect agreement between the two techniques. Moreover, we show that in cases with strong hybridization between interacting and screening subspaces, the application of cRPA becomes ambiguous and can lead to unrealistically small U values, while LRT remains well-behaved. Our Letter formally connects both methods, sheds light on their strengths and limitations, and emphasizes the importance of using a consistent set of Wannier orbitals to ensure transferability of U values between different implementations.

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An accurate description of many quantum materials [1] relies on correctly capturing the strong electron-electron interaction among a subset of electrons inhabiting the states around the Fermi level. The method of choice is to divide the electrons into strongly and weakly interacting subspaces, where the latter can be treated within common approximations to density-functional theory (DFT) [2,3], while the strongly interacting subspace is augmented with a more accurate treatment of the local electron-electron interaction, resulting in the widely used DFT + U [4–7] or DFT plus dynamical mean-field theory (DFT + DMFT) methods [8–10].

The accuracy and predictive capabilities of these techniques depend on a realistic estimation of the screened Coulomb interaction within the strongly interacting subspace, which is typically parametrized in terms of an on-site

Hubbard U . Consequently, several first-principles approaches to computing Hubbard parameters have been developed, including constrained DFT [11–19], linear response theory (LRT) [20–25], Hartree-Fock-based methods [26–31], and the constrained random-phase approximation (cRPA) [32–36]. In particular, LRT and cRPA are widely used in practical applications. Notably, LRT is used almost exclusively for DFT + U calculations, in particular for high-throughput applications of DFT + U [7,37–39], with few exceptions [40], while cRPA is utilized more often in the context of DFT + DMFT calculations [41–43].

Crucially, it has not been established to what extent LRT and cRPA can be viewed as equivalent. While some attempts were made to compare different methods for computing Hubbard parameters [35,44–46], there is still lack of understanding and consensus regarding the similarities and differences between them. So far, an explicit quantitative comparison of the calculated U parameters has been hindered by the fact that specific implementations generally use different basis orbitals to represent the strongly interacting subspace, with the U parameter depending very sensitively on the specific choice of these orbitals [7,37,47,48]. Many DFT codes implementing the LRT method use internally defined localized orbitals (e.g., orthogonalized atomic orbitals [47] or partial wave

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projectors [49]), while cRPA calculations are often based on Wannier functions [50–53].

In this letter, we systematically compare U values obtained from LRT and cRPA. We specifically address the need for a consistent definition of the interacting subspace and a consistent choice of *Hubbard projectors*, which represent the corresponding localized basis orbitals. To this end, we extend the LRT calculation of the U parameter using density-functional perturbation theory [21,23] within QUANTUM ESPRESSO [54–56] by interfacing it with WANNIER90 [57] in order to employ maximally localized Wannier functions (MLWFs) [58] as Hubbard projectors. This enables a direct quantitative comparison between LRT and cRPA, the latter being evaluated using QUANTUM ESPRESSO in combination with RESPACK [59,60]. We first show mathematically that the definitions of U in the two approaches are not strictly equivalent, but that, for cases where the interacting and screening subspaces correspond to isolated or nearly isolated sets of bands, one can clearly identify the corresponding differences. We then demonstrate numerically, that LRT and cRPA can be brought in remarkable quantitative agreement, if these differences are properly accounted for. For cases where the two subspaces hybridize strongly, the application of cRPA is problematic, leading to unrealistically small U values, whereas LRT remains largely unaffected.

An essential quantity in both methods is the charge susceptibility, or *response tensor*, which characterizes how a system of interacting electrons reacts to external perturbations. Thereby, one distinguishes between the *bare* susceptibility, χ_0 , which does not consider the change in the electronic interactions caused by the charge rearrangement, and the *full* susceptibility, χ , which considers both the external perturbation and the effect of the resulting electron redistribution.

Within RPA, the bare susceptibility can be written as a sum over all possible electron-hole excitations in an independent particle basis,

$$\chi_0(\omega, \mathbf{r}, \mathbf{r}') = \sum_{m,n} \frac{f_n - f_m}{\omega + \epsilon_n - \epsilon_m} \langle \mathbf{r} | \psi_m \rangle \langle \psi_n | \mathbf{r} \rangle \langle \mathbf{r}' | \psi_n \rangle \langle \psi_m | \mathbf{r}' \rangle, \quad (1)$$

where $|\psi_n\rangle$, ϵ_n , and f_n are the Kohn-Sham (KS) states, their energies, and occupations, respectively. The cRPA formalism [34,35] then emphasizes the division of the electronic Hilbert space into the *interacting subspace*, $\mathcal{D}$, and the *screening subspace*, mirroring the corresponding division within DFT + U and DFT + DMFT. [61] The bare susceptibility is then split into two parts, $\chi_0 = \chi_0^{\mathcal{D}} + \chi_0^{\mathcal{R}}$, where $\chi_0^{\mathcal{D}}$ contains only electron-hole excitations within the interacting subspace, and $\chi_0^{\mathcal{R}}$ contains all other transitions. The partially screened Coulomb interaction acting within the $\mathcal{D}$ subspace is then obtained as $W^{\mathcal{R}} = [1 - V\chi_0^{\mathcal{R}}]^{-1}V$,

where $V = 1/|\mathbf{r} - \mathbf{r}'|$ is the unscreened Coulomb interaction. For the following it is also useful to define the full susceptibility within the interacting subspace, $\chi^{\mathcal{D}}$, through the Dyson equation $W^{\mathcal{R}} = (\chi_0^{\mathcal{D}})^{-1} - (\chi^{\mathcal{D}})^{-1}$.

Static interaction parameters are computed as matrix elements of $W^{\mathcal{R}}(\omega = 0)$ with the Hubbard projector functions $\phi_i^I(\mathbf{r})$, i.e., the specific basis orbitals used to represent $\mathcal{D}$, corresponding to orbital i on site I [34],

$$(W^{\mathcal{R}})_{ijkl}^{IJKL} = \int \int \phi_i^I(\mathbf{r})^* \phi_j^I(\mathbf{r}) \times W^{\mathcal{R}}(\omega = 0, \mathbf{r}, \mathbf{r}') \phi_k^J(\mathbf{r}')^* \phi_l^J(\mathbf{r}') d\mathbf{r} d\mathbf{r}'. \quad (2)$$

Within the Slater parametrization of the local interaction, which is often employed in calculations using the full d shell [5], U is defined as average over both inter- and intraorbital matrix elements (see, e.g., Ref. [62]),

$$U_{\text{cRPA}}^I = \frac{1}{(N^I)^2} \sum_{ij} (W^{\mathcal{R}})_{iijj}^{IIII}, \quad (3)$$

where N^I is the number of interacting orbitals on site I .

On the other hand, in LRT, the relevant susceptibilities are obtained by directly calculating the response of the KS system to small localized perturbations, with $\tilde{\chi}^{IJ} = \partial n^I / \partial \lambda^J$ defined in terms of the change in occupation of the Hubbard projector orbitals on site I due to a local potential shift on site J , λ^J , acting only within the interacting subspace. The Hubbard parameter for site I is then defined as the corresponding diagonal element of the difference of the inverse susceptibility matrices [7,20],

$$U_{\text{LRT}}^I = (\tilde{\chi}_0^{-1} - \tilde{\chi}^{-1})^{II}, \quad (4)$$

where $\tilde{\chi}_0$ is calculated without considering the change in the KS potential resulting from the charge rearrangement. Note that $\tilde{\chi}_0$ and $\tilde{\chi}$ describe the density-density response only on a per site basis, without any orbital dependence, i.e., they describe a purely *monopolar* response. However, as shown in Ref. [7], these “coarse-grained” susceptibilities can be related to a more general, site- and orbital-dependent, four-index response function. In Sec. I.F of Supplemental Material [63] we show that for cases where the interacting subspace $\mathcal{D}$ constitutes an isolated set of bands, the cRPA susceptibilities $\chi_0^{\mathcal{D}}$ and $\chi^{\mathcal{D}}$ can be identified with this general four-index response function if one neglects the exchange-correlation (xc) contribution to the susceptibility (the latter is included in LRT but typically neglected in cRPA). We emphasize that a strict separation between the interacting and screening subspaces is a necessary prerequisite for establishing the mathematical equivalence between the techniques. In this case the coarse grained response can be written in terms of the orbital averages of the bidiagonal components,

$\tilde{\chi}_0^{IJ} = \sum_{ij} (\chi_0^{\mathcal{D}})_{ijij}^{IIJJ}$ and $\tilde{\chi}^{IJ} = \sum_{ij} (\chi^{\mathcal{D}})_{ijij}^{IIJJ}$ [63]. U_{LRT} can then be related to the cRPA interaction tensor, Eq. (2), through,

$$U_{\text{LRT}}^I = \sum_{zt} \sum_{opqs}^{OPQS} (\tilde{\chi}_0^{-1})^{tZ} \times \left[(\chi_0^{\mathcal{D}})_{zzpo}^{\text{ZZPO}} (W^{\mathcal{R}})_{opqs}^{\text{OPQS}} (\chi^{\mathcal{D}})_{sqtt}^{\text{SQTT}} \right] (\tilde{\chi}^{-1})^{tI}, \quad (5)$$

where all indices correspond to $\mathcal{D}$.

If we separate the cRPA susceptibilities into their locally “isotropic” component (i.e., the purely monopolar part that gives rise to $\tilde{\chi}_0$ and $\tilde{\chi}$) and a locally “nonisotropic” part, see Eq. (42) in Supplemental Material [63], the isotropic contributions cancel with the corresponding coarse-grained susceptibilities in Eq. (5), resulting in the orbitally averaged U_{cRPA}^I from Eq. (3) plus all remaining terms containing nonisotropic parts. Thus, we can write $U_{\text{LRT}}^I = U_{\text{cRPA}}^I + \Delta U^I$, where ΔU^I contains all nonisotropic contributions. These additional terms correspond to excitations completely inside the interacting subspace, and are thus excluded from the screening within cRPA. They represent processes that alter the “shape” of the charge density on an atomic site (orbital reorganizations, bond fluctuations) and also include local exchange terms containing matrix elements of the form $(W^{\mathcal{R}})_{oppo}^{\text{OOOO}}$.

Thus, in the case of well separated interacting and screening subspaces, we can identify two differences in the definition of the U parameter between LRT and cRPA: (1) the xc contribution to the response, which is typically neglected in cRPA but included in LRT, and (2) the coarse-grained nature of LRT introduces additional terms ΔU^I related to nonisotropic excitations within $\mathcal{D}$.

We now investigate how these differences manifest numerically within realistic calculations, using two different materials as benchmarks: KCuF_3 and Sr_2FeO_4 . Section V of Supplemental Material [63] contains results for two additional materials: NiO and CrO_2 .

KCuF_3 has been studied using both DFT + U and DFT + DMFT [5,64], due to its Jahn-Teller-distorted structure and associated orbital order. For the purpose of this Letter, we consider KCuF_3 in the high symmetry cubic perovskite structure [shown in Fig. 1(a)], both for ease of computation and since it exhibits a prototypical band structure of a transition metal (TM) perovskite, with a clear separation between bands with dominant Cu d and F p character [see Fig. 1(b)]. Sr_2FeO_4 crystallizes in a layered perovskite structure [shown in Fig. 1(a)] [65,66], and is of interest due to its structural and electronic similarity to the unconventional superconductor Sr_2RuO_4 [43]. The corresponding KS bands [see Fig. 1(c)] exhibit a small overlap between bands with dominant Fe d and dominant O p character. As reported recently, the computed value of U_{cRPA} for the Fe d states is too small to reproduce the experimentally observed insulating state of Sr_2FeO_4 within DFT + DMFT [43].

For both materials, we compare three different choices of the interacting subspace. In each case we represent the corresponding basis orbitals as MLWFs constructed from the KS band structures shown in Fig. 1, obtained for the nonmagnetic, i.e., spin-degenerate, state. In the first case, we consider all bands with dominant TM d and ligand p character [see the projected densities of states in Figs. 1(b) and 1(c)], which are completely separated from other bands at lower and higher energies for both materials, and construct a complete MLWF basis for these bands consisting of five TM centered orbitals and three ligand centered orbitals per F or O within the unit cell.

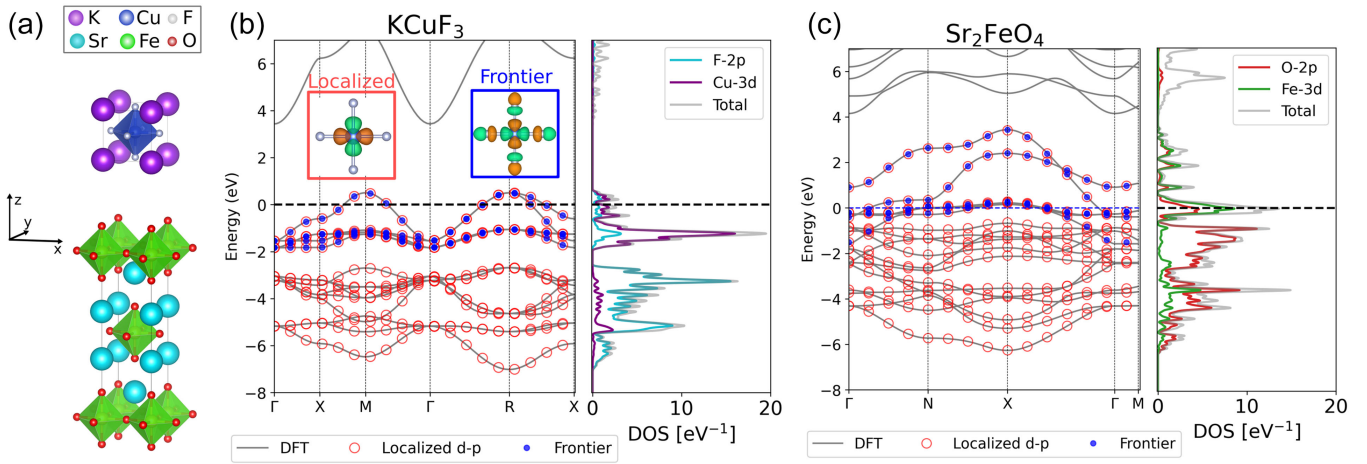


FIG. 1. (a) Crystal structures of cubic KCuF_3 and tetragonal Sr_2FeO_4 . (b) and (c) Band structure and projected densities of states (DOS) of (b) KCuF_3 and (c) Sr_2FeO_4 . Zero energy corresponds to the Fermi level. Open and closed circles indicate the bands recalculated from the localized d - p and frontier orbital basis sets, respectively. The red and blue insets in (b) show isosurface plots of the Cu $d_{x^2-y^2}$ MLWF in the localized d - p and frontier orbital basis sets, respectively.

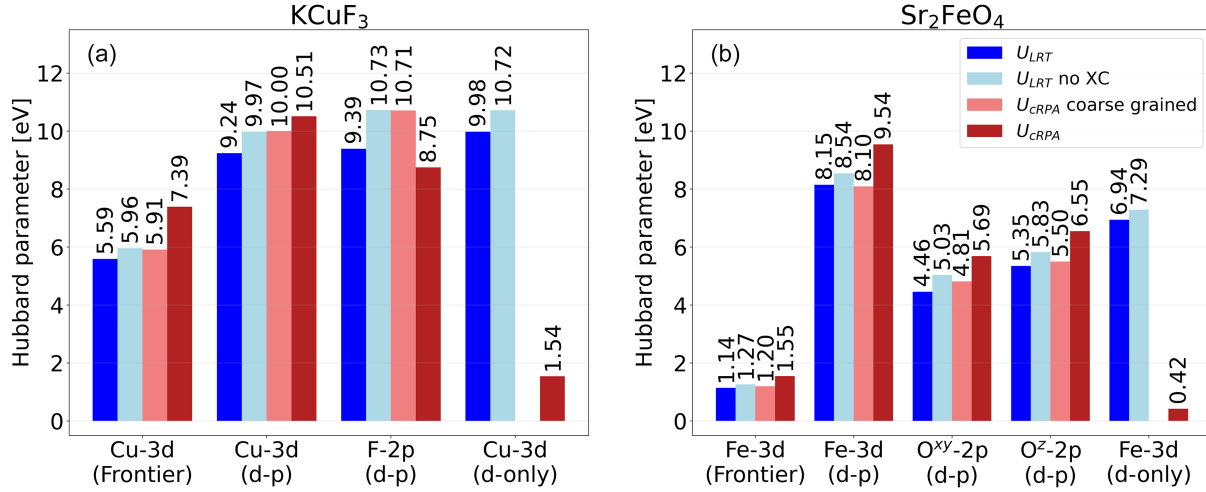


FIG. 2. Interaction parameters obtained from LRT and cRPA for (a) KCuF_3 and (b) Sr_2FeO_4 using different choices for the Hubbard projectors (frontier, d - p , and d -only). Dark blue and dark red bars correspond to interaction parameters computed from a standard LRT and cRPA calculation, respectively. The light blue bar corresponds to a modified LRT calculation, i.e., by removing the contribution of the xc kernel, while the light red bar corresponds to the modified cRPA calculation, i.e., obtained by using the coarse graining. For Sr_2FeO_4 , O^{xy} refers to the oxygen within the Fe planes, while O^z refers to the apical oxygen. For the d -only case, the formal equivalence between cRPA and LRT is lacking and thus no coarse graining is performed.

The resulting MLWFs closely resemble atomic d and p orbitals, respectively [see inset in Fig. 1(b) for the example of the Cu $d_{x^2-y^2}$ -like orbital in KCuF_3]. We call this the *localized d - p* basis. In the second case, we consider the same set of bands but construct only five MLWFs centered at the TM sites. These MLWFs again closely resemble atomic d orbitals but are slightly more localized compared to the d - p basis. We call this second case *localized d -only*. Finally, in the third case, we restrict the energy window for the calculation of the MLWFs to include only the “frontier” bands with dominant TM d character immediately around the Fermi level. This results in five more delocalized d -like MLWFs, centered at the TM sites but exhibiting p -like tails at the surrounding ligand sites, which reflects the hybridized character of the corresponding bands [see the blue inset of Fig. 1(b) for the Cu $d_{x^2-y^2}$ -like orbital in KCuF_3]. We call this the *frontier orbital* basis. Such orbitals are often used in DFT + DMFT calculations.

In Fig. 2, we summarize the calculated values for various definitions of U for both KCuF_3 (a) and Sr_2FeO_4 (b) and the three different choices for the interacting subspace. We first focus on the localized d - p and the frontier orbital basis. In these two cases, the MLWFs completely span the KS bands within the respective energy windows [see Figs. 1(b) and 1(c)], resulting in a clear separation between the interacting and screening subspace. The interaction parameters obtained from LRT (shown in dark blue) are of comparable magnitude to those obtained from cRPA (in dark red), with U_{cRPA} being larger than U_{LRT} by around 10%–30%. The only exception is the F p states in KCuF_3 in the d - p basis, for which U_{LRT} is about 7% larger. This last result can be explained by the known tendency of LRT to

overestimate U in cases with essentially completely full (or completely empty) shells, where the applicability of the linear response approach becomes problematic [7,44,67]. Indeed, the occupation of the F p states in KCuF_3 is nearly 1.0 electrons per orbital in our d - p basis, while in Sr_2FeO_4 , the stronger hybridization with the TM d states reduces the average occupation of the O p states to ~ 0.9 , such that the applicability of LRT is not problematic.

To relate these quantitative differences between U_{LRT} and U_{cRPA} to our mathematical considerations, we first analyze the effect of removing the response of the xc potential from the LRT calculation (light blue bars in Fig. 2), as explained in Sec. II in the Supplemental Material [63]. This generally increases U_{LRT} by 5–10%, thus bringing it closer to the cRPA value. To analyze the remaining difference to U_{cRPA} , we then perform a coarse-graining of the cRPA results by computing $\tilde{\chi}_0$ and $\tilde{\chi}$ from the four-index tensors χ_0^D and χ^D (see Sec. I in Supplemental Material [63]), and extract a corresponding U via Eq. (4). The resulting value (light red bars in Fig. 2) is mathematically the same quantity as U_{LRT} with the xc response neglected. Indeed, the corresponding values show very good quantitative agreement. They are nearly identical for KCuF_3 , while for Sr_2FeO_4 they exhibit residual discrepancies of about 5%. These small numerical differences can be assigned to the finite convergence threshold used in the calculations and the fact that RESPACK computes, in addition to all density-density terms, $(W^R)_{ijji}^{IIJJ}$ across all cells, only the exchange integrals, $(W^R)_{ijji}^{IIJJ}$, in the first unit cell. Other elements are thus missing from our coarse-graining procedure. Overall, the remarkable quantitative agreement between modified

LRT and cRPA for both the frontier and d - p bases, despite using completely different implementations, strongly supports our analytical result.

We now turn to the localized d -only case (last set of bars in both panels of Fig. 2), for which the interacting subspace does not form an isolated set of bands, and thus a formal equivalence of cRPA and LRT cannot be established. Nevertheless, several methods have been suggested to obtain χ_0^R in such cases [50,68–70]. We use the *weighted method* introduced in Ref. [68]. While U_{cRPA} is very small in this case, U_{LRT} is much larger and comparable to the values obtained for the localized d - p basis. The very small U_{cRPA} for the d -only case is due to the strong screening from transitions with very low energy, related to bands with mixed character that are close to or even crossing the Fermi level. The d -only case thus represents a case where the applicability of the cRPA approach becomes questionable, and the results can depend sensitively on the specific way the two subspaces are separated. [71] On the other hand, the d -only case is quite relevant for practical applications, since in DFT + U and DFT + DMFT calculations for TM compounds, one typically applies the U correction only on the TM sites, while the ligand states are considered as weakly interacting.

Within LRT, apart from the fact that the d -only MLWFs are slightly more localized, the main difference to the d - p case is the absence of TM-ligand off-diagonal elements in the susceptibility matrices $\tilde{\chi}^{IJ}$ and $\tilde{\chi}_0^{IJ}$, which affect U_{LRT}^I due to the inversion in Eq. (4). These off-diagonal elements are larger in Sr_2FeO_4 than in KCuF_3 , due to the stronger d - p hybridization in the oxide compared to the more ionic fluoride, which results in a larger difference of U_{LRT} between the two localized basis sets for Sr_2FeO_4 .

We note that, since the generalized susceptibility $(\chi_0)_{ijkl}^{IJKL}$ is equivalent to the bare cRPA susceptibility [63] (for clearly separable $\mathcal{D}$), the well known overscreening effects identified for cRPA when interacting and screening subspaces are close in energy or overlap [73–77], can be expected to also affect U_{LRT} in such cases. As noted recently, such overscreening can also be due to an incorrect energy separation between the frontier and ligand states in the underlying band structure [78]. This could explain why in the Sr_2FeO_4 frontier basis, for which $\mathcal{D}$ overlaps slightly with the lower-lying screening bands, U_{LRT} and U_{cRPA} are too low to reproduce the experimentally observed insulating state [43].

In summary, we have introduced a unified framework based on Wannier functions that enables a *direct* comparison between LRT and cRPA for the calculation of Hubbard parameters. We have shown that, for an isolated interacting subspace, both approaches can be placed on the same theoretical footing, and we have identified the origin of the remaining differences: namely, the neglect of exchange-correlation contributions in cRPA and the inclusion of additional screening channels in LRT. Once these are

consistently accounted for, the two methods yield quantitatively equivalent results, resolving a long-standing discrepancy. Beyond this formal connection, our Letter clarifies that the widely reported differences in U (often amounting to several eV) can arise not only from variations in computational setups, but also from inconsistent definitions of interacting and screening subspaces. Our results also highlight the advantage of using MLWFs as local basis orbitals, which enables a consistent and *transferable* definition of Hubbard parameters across different implementations. Thus, by removing several ambiguities, we establish conditions under which U becomes a well-defined quantity, and consequently our Letter advances the reproducibility and predictive power of DFT + U and DFT + DMFT calculations. Furthermore, for systems with strong hybridization between interacting and screening subspaces, we find that cRPA can become ambiguous and yield unphysically small U values, while LRT remains well behaved. Conversely, LRT in its current formulation is not suitable for closed-shell systems, where it can produce unphysically large U values. These results provide practical guidance on the applicability of each method. By placing both methods on a common footing and explicitly identifying their underlying approximations, our work provides a basis for the systematic improvement of Hubbard parameter calculations in future DFT + U and DFT + DMFT studies.

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Data availability—The data that support the findings of this article are openly available on the Materials Cloud [79].

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