Workhorse minimally-empirical dispersion-corrected density functional, with tests for weakly-bound systems: $r^2\text{SCAN}+r\text{VV10}$

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SCAN+rVV10 has been demonstrated to be a versatile van der Waals (vdW) density functional that delivers good predictions of both energetic and structural properties for many types of bonding. Recently, the $r^2\text{SCAN}$ functional has been devised as a revised form of SCAN with improved numerical stability. In this work, we refit the rVV10 functional to optimize the $r^2\text{SCAN}+r\text{VV10}$ vdW density functional, and test its performance for molecular interactions and layered materials. Our molecular tests demonstrate that $r^2\text{SCAN}+r\text{VV10}$ outperforms its predecessor SCAN+rVV10 in both efficiency (numerical stability) and accuracy. This good performance is also found in lattice-constant predictions. In comparison with benchmark results from higher-level theories or experiments, $r^2\text{SCAN}+r\text{VV10}$ yields excellent interlayer binding energies and phonon dispersions for layered materials.

I. INTRODUCTION

Quantum fluctuations in the electronic density give rise to instantaneous dipole moments, making van der Waals (vdW) or London dispersion interactions ubiquitous in electronic matter. Despite its relative small strength, the ubiquitous vdW force plays a fundamental role in diverse fields of both science and industry: from structural biology and polymer science, to nanotechnology and surface science. It participates in the structural evolution of DNA, proteins, and many other complex molecules and their interactions, and hence the origination and physical activities of living beings. The vdW forces are also crucial for the surface and interfacial reactions controlling artificial and natural catalytic and corrosion reactions on alloy surfaces. The vdW interactions are even found to be necessary for accurate descriptions of some densely packed systems, suggesting that vdW forces are not as negligible for normal solids as commonly thought.

While vdW interactions are fully captured in the exact density functional theory (DFT) their non-local nature means they (or at least their most long-ranged parts) are missed by semi-local exchange-correlation (XC) density functional approximations (DFAs) like the local density approximation (LDA), generalized gradient approximation (GGA), or meta-GGA. Despite this limitation, semi-local DFAs are the mainstay of modern first-principles electronic structure modelling, achieving useful accuracy at reasonable cost. While higher-level methods that fully account for vdW forces, such as quantum Monte Carlo (QMC), coupled-cluster singles and doubles with perturbative triples CCSD(T), and the adiabatic-connection fluctuation-dissipation theorem within the random-phase approximation (RPA) can provide benchmark references, their poor scaling with system size prohibits large-scale applications. Instead, a practical choice for improving accuracy is to include vdW interactions in the DFT framework as a modification or correction to a semi-local XC approximation. Common approaches include the DFT+D series, the Tkatchenko-Scheffler (TS) methods, the Rutgers-Chalmers vdW-DF family, Vydrov-van Voorhis (VV10), rVV10 density functionals, and the Becke-Johnson exchange hole model. We should also mention the damped-Zaremba-Kohn (dZK) correction, which requires many material-dependent input parameters.

The performance of the vdW-corrected DFA depends upon both the semi-local XC and vdW functionals. A good example for this case is the SCAN+rVV10 vdW functional. The strongly constrained and appropriately normed (SCAN) meta-GGA satisfies all known 17 exact constraints applicable to a meta-GGA, and has shown good accuracy for diverse bonding environments. It has been demonstrated that SCAN includes a portion of the intermediate range of vdW interactions, which rationalizes its excellent predictions of structural and energetic properties of water. The rVV10 non-local vdW density functional requires only the electron density and its first derivatives as inputs, and contains two empirical parameters, $C$ and $b$. The final SCAN+rVV10 vdW density functional has been demonstrated to work for general geometries, and achieves an accuracy comparable to that of higher-level methods like RPA and CCSD(T) for various vdW benchmark systems, while scaling more favorably with system size.

Despite these successes, SCAN exhibits undesirable numerical problems that harm its computational efficiency and can prevent the self-consistent field process
from converging. To achieve high accuracy for diverse systems, SCAN interpolates between single-orbital and slowly-varying energy densities using a variable $\alpha$ (defined in Ref. 30) that is sensitive to the local chemical environment. $\alpha$ partly contributes to the numerical instability of SCAN.33

Moreover, SCAN’s inclusion of intermediate vdW interactions can be a hindrance when combined with non-local dispersion corrections. SCAN predicts quantitatively correct lattice parameters for the layered solid PPTA, whereas SCAN+rVV10 strongly overbinds within layers, yielding a much too-small $\alpha$-parameter.45 SCAN’s tendency to overbind hydrogen-bound molecules is worsened in both SCAN+rVV10 and SCAN+D425,55. When evaluated on the Hartree-Fock density (a kind of “density correction”), SCAN provides a chemically accurate description of liquid water, whereas dispersion-corrected variants of SCAN still overbind59.

The rSCAN meta-GGA33 modifies SCAN to successfully improve numerical stability, but at the price of reduced accuracy10,12. To remove the divergence in the derivatives of $\alpha$ in single orbital regions ($\alpha \to 0$)54 rSCAN uses a regularized $\alpha'$ that breaks exact coordinate scaling condition13,14 and the uniform density limit obeyed by SCAN. To remove oscillations in the exchange-correlation potential of SCAN induced by the function of $\alpha$ that interpolates between energy densities, rSCAN uses a smooth polynomial for the chemically-relevant range $0 \leq \alpha \leq 2.5$. This choice introduces spurious terms in the slowly-varying ($\alpha \approx 1$) density-gradient expansion that deviate from the exact expansion16,17 recovered by SCAN.

These shortcomings are remedied by the $r^2$SCAN meta-GGA53, which modifies the rSCAN regularizations to obey almost every exact constraint SCAN does. (A higher-order density-gradient expansion for exchange is recovered by SCAN52.) The satisfaction of exact constraints and greater smoothness of $r^2$SCAN preserves the accuracy of SCAN and numerical efficiency of rSCAN19,24,25, permitting construction of meta-GGA pseudopotentials53. Therefore, we expect $r^2$SCAN to be a better candidate for the rVV10 correction.

It should be noted that a variant of $r^2$SCAN with a long range D4 dispersion correction was recently published19. $r^2$SCAN+D4 more realistically describes non-covalent and hydrogen-bound systems than SCAN+D419, suggesting that $r^2$SCAN includes less of the intermediate vdW interaction than SCAN. Reference 19 presented a fitted value $b = 12.3$ for $r^2$SCAN+VV10 (not rVV10). rVV10 was designed to perform like VV10, but at a lower computational cost in plane-wave basis set codes. We now motivate why an $r^2$SCAN+rVV10 is needed when a highly-accurate $r^2$SCAN+D4 exists.

The D and VV10 series of vdW corrections are complementary approaches for describing long-range vdW interactions in real systems. Both corrections have empirical parts, with the VV10 series requiring two material-independent empirical parameters, and D4 requiring three parameters in its damping function. The D4 dispersion coefficients are computed on-the-fly from tabulated material-dependent data like the atomic polarizabilities and Mulliken partial charges.13 rVV10 is conceptually simpler than D4 and its reliance on fewer empirical parameters makes it an appealing alternative to D4 for solid-state physics, though both methods find common use. In a comparison22 of 243 non-covalent cluster interactions, SCAN-D3 and SCAN+D4+rVV10 had comparable root mean square deviations from reference values.

The original VV1023 and subsequent rVV1025 vdW corrections differ in subtle ways. The VV10 kernel is a two-point function, and its evaluation requires a double integral over real space. Such a correction is challenging to implement in plane-wave codes because of the high numeric cost of this double integral. The rVV10 kernel approximates the VV10 kernel by interpolation over a set of grid points, drastically reducing the computational overhead in plane-wave basis set codes.

When rVV10 is a good approximation to VV10, the $b$ parameters should not differ substantially. We confirm this interpretation here. However, a VV10-corrected DFA which tends to overbind molecules is expected40 to further overbind when combined with rVV10 using the same $b$ parameter. When using the same $b$ parameter, the most pronounced differences between VV10 and rVV10 occur in low-density regions54. However, the dispersion correction to a meta-GGA like SCAN or $r^2$SCAN should be most meaningful in these low-density regions.

A limitation of the VV10 and rVV10 long-range dispersion corrections is that they can describe only two-body interactions between volume elements, ignoring the three-body Axilrod-Teller55 effects. Here we fit the $b$ parameters in those corrections to the binding energy curve of the Ar dimer, in which the conventional many-body expansion stops at the two-body term.

The vdW interactions are crucial in shaping the structure and properties of 2D/layered materials. Such materials have seen renewed interest since the exfoliation of graphene in 200426, and have nurtured new applications promising the next generation of information technology devices57. As such, we test the newly determined $b$ parameter for $r^2$SCAN+rVV10 on standard sets, with a focus on layered materials properties.

II. METHODS

A. Parameters in $r^2$SCAN+rVV10

The rVV1024,25 non-local correlation functional is similar in construction to the Rutgers–Chalmers vdW-DF family23,

$$E_{\text{el}} = \int d\mathbf{r} n(\mathbf{r}) \left[ \frac{\hbar}{2} \int d\mathbf{r}' \phi(\mathbf{r}, \mathbf{r}') n(\mathbf{r}') + \beta \right]. \quad (1)$$
\[
E_{xc} = E_{xc}^0 + E_{xc}^{nl}. \tag{2}
\]

Here, \(n(r)\) is the electron density, \(\phi(r, r')\) is the density-density interaction kernel, and \(E_{xc}^0\) is the semi-local exchange correlation functionals to be corrected. \(\beta = (3/5)^{3/4}/32\) in Hartree is required for zero \(E_{xc}^{nl}\) for the uniform electron gas. Two empirical dimensionless parameters \(C\) and \(b\) appear in the kernel \(\phi(r, r')\): \(C\) is adjusted to recover the accurate \(-C_6/R^6\) asymptotic vdW interaction between molecules at large separation \(R\), and \(b\) controls the damping of \(E_{xc}^{nl}\) at short range.

The original VV10 and rVV10 functionals were combined with the semi-local XC functional \(E_{xc}^0 = E_{xc}^{PW86} + E_{xc}^{PBE}\), partly due to the near absence of vdW in rPW86 exchange. (For a discussion of how intermediate-range vdW can arise from semilocal exchange, see Ref. [32].) For a semi-local \(E_{xc}^0\), \(C = 0.0093\) was recommended, and the \(b\) parameter was determined as 5.9 and 6.3 by fitting to the interaction energies of the S22 set for the original VV10 and rVV10, respectively. Increasing \(C\) or \(b\) generally results in a smaller vdW correction. There is a conventional many-body expansion\(^{29}\) of the dispersion interaction within a collection of bodies (atoms or molecules) that includes two-body and higher-order many-body effective interactions. By construction, the VV10 and rVV10 long-range corrections explicitly account for only pairwise interactions between volume elements. The Ar dimer has only conventional two-atom interactions, whereas the S22 has many-atom interactions. Fitting rVV10 to systems with many-atom interactions would average over the two- and many-atom interaction.\(^{65}\)

Here, we refit \(b = 11.95\) for \(r^2\)SCAN+rVV10 by adjusting it to best recover the binding-energy curve of the argon dimer with bond lengths between 3.5 and 6.0 Å, as shown in Fig. 1(a). Using the \(r^2\)SCAN+VV10 (MAE 0.32 kcal/mol for S22\(^{29}\)) value \(b = 12.3\), the mean absolute error (MAE) in the binding energy curve of Ar\(_2\) increases negligibly by 0.2 meV (0.0046 kcal/mol). Note that \(r^2\)SCAN-D4 makes a 0.29 kcal/mol MAE on the S22 set\(^{29}\), virtually identical to both variants of \(r^2\)SCAN+rVV10.

### B. Computational details

The DFT calculations in this work were performed using the Vienna Ab initio Simulation Package (VASP)\(^{66}\) version 5.4.4, with user corrections for the meta-GGA correlation potential in spin-unrestricted calculations, and to the rVV10 stress tensor.\(^{49}\) For computational details of the Ar dimer binding-energy curve, the S22 molecular interaction energies,\(^{49,61,69}\) and the L28 layered material database,\(^{70}\) refer to the original publication of SCAN+rVV10\(^{32}\) with the following adjustments. We follow the practice of using hard pseudopotentials for the Rutgers–Chalmers vdW-DFs, and the XC functional reads as

Error statistics for the inter-layer binding energies and lattice parameters of the L28 set are presented in Tables I and III. Values for individual solids are presented in Tables VII and IX of the Appendix. The same methods used to validate SCAN+rVV10\(^{32}\) are used here for calculation of the L28 binding energies (compared to reference RPA\(^{72}\) calculations): the intra-layer lattice constants were fixed to their experimental values, and the inter-layer lattice constants were relaxed only for the bulk structures. Only atomic coordinates were relaxed for the mono-layer model, as in the RPA calculations. The calculated lattice constants in Tables I, III, VII and IX are from full relaxations.

Non-magnetic ground states were used in the current calculations for these compounds, except the three vanadium-based compounds, where a ferromagnetic ordering was used instead. For the SCAN+rVV10 and \(r^2\)SCAN+rVV10 results in Table IV, intra-layer lattice constants were also relaxed for the bulk and mono-layer models, although the difference in binding energy was negligible. For the phonon calculations of graphite and MoS\(_2\), we used the Phonopy code\(^{70}\) to obtain the harmonic force constants from VASP atomic force calcula-
tions within the finite displacement method (0.015 Å).
For solid poly(p-phenylene terephthalamide) (PPTA),
\( k\) -point spacing of 0.15 Å\(^{-1} \) (yielding a \( 6 \times 9 \times 4 \) \( k\)-grid),
and a plane-wave cutoff of 900 eV were used.

### III. RESULTS AND DISCUSSIONS

#### A. Dispersion interactions in molecules

To evaluate the performance of \( r^2\)SCAN+rVV10 with the newly fit \( b = 11.95 \), we tested it on both molecular systems (S22 data set) and layered materials. We are especially interested in the efficiency and accuracy of \( r^2\)SCAN+rVV10, in comparison with its predecessor SCAN+rVV10.

We assessed the accuracy of SCAN+rVV10 and \( r^2\)SCAN+rVV10 predicted interaction energies for the S22 molecular complexes data set. The S22 set includes seven hydrogen-bonded, eight dispersion-bound, and seven mixed-binding complexes. Table I presents the error statistics of SCAN+rVV10 and \( r^2\)SCAN+rVV10 for the S22 set, relative to CCSD(T) benchmark. Table VII of the Appendix complements Table I, presenting values for each molecule in the S22 set, and comparing our SCAN+rVV10 and \( r^2\)SCAN+rVV10 results with Perdew-Burke-Ernzerhof (PBE) \( 22 \) SCAN, and vdW-DF \( 23 \) predictions.

To further demonstrate the improved numeric stabil-
FIG. 2. The binding energy curves (solid lines) and forces (dashed lines) for the T configuration of benzene dimer from (a) \( r^2\text{SCAN}+\text{rVV10} \) and (b) \( \text{SCAN}+\text{rVV10} \) compared to the CCSD(T) results as the reference, as a function of their separation \( R \) in Å. As in Ref. 75, forces are computed using a spline interpolation of the binding energy data.

A previous study demonstrated that \( \text{SCAN}+\text{rVV10} \) produces significant oscillations in the interaction energy and force curves of the benzene dimer, which persist even with a large energy cutoff. In this work, we consider the T benzene dimer and confirm that removing such oscillations requires denser real space grids. Specifically for VASP users, we recommend using a high ENAUG (∼1500) at certain ENCUT with PREC=High, instead of increasing ENCUT with PREC=Accurate. The \( r^2\text{SCAN}+\text{rVV10} \) binding energy and force curves don’t show oscillations even with low accuracy settings, as shown in Fig. 2. However, \( r^2\text{SCAN} \) as a meta-GGA is still much more complicated than LDA and PBE, and thus may still need dense real space grids for certain applications. To ensure stable convergence behavior, we recommend using dense real space grids (PREC=High; ENAUG=1500 or 2000) for SCAN-like metaGGAs’s and functionals based on them.

### B. Layered materials

We also tested the predictions of \( \text{SCAN}+\text{rVV10} \) and \( r^2\text{SCAN}+\text{rVV10} \) for geometry and inter-layer binding properties for 28 layered materials (L28). As shown in Tables II and III, \( r^2\text{SCAN}+\text{rVV10} \) more accurately predicts lattice constants than \( \text{SCAN}+\text{rVV10} \) for this test set, and converges quicker with respect to plane-wave ba-

### TABLE II. Unsigned layer-layer binding energy \( E_b \) in meV/Å\(^2\), lattice constants \( c \) and \( a \) in Å, for 28 layered materials (L28) from \( \text{SCAN}+\text{rVV10} \) and \( r^2\text{SCAN}+\text{rVV10} \). Mean deviations (MDs) and mean absolute deviations (MADs) are taken with respect to the RPA (an uncertain reference; see Table IV) for \( E_b \), and experimental for the lattice constants \( c \) and \( a \). Table VIII in the Appendix presents values for each material in the set, the reference values, and values for other density functional approximations.

|                     | \( \text{SCAN}+\text{rVV10} \) | \( r^2\text{SCAN}+\text{rVV10} \) |
|---------------------|-------------------------------|----------------------------------|
| \( E_b \)          | MD 0.476 0.132 0.019           | 2.670 0.108 0.009                |
| \( c \)            | MAD 1.527 -0.167 0.179         | 2.786 0.139 0.018                |
| \( a \)            | MAD 0.024 -0.010 0.019         | 0.009 0.000 0.008                |

### FIG. 3. Phonon dispersion in (a) graphite, (b) MoS\(_2\), highlighting the improvements on the phonon branches along \( \Gamma-A \) (along the \( c \)-axis, or interlayer direction) from vdW corrections, compared with available experimental data from Refs. 79–81 for graphite and Refs. 82–85 for MoS\(_2\). Calculations were performed at the relaxed lattice parameters. For an analogous figure using PBE and PBE-D4, see Fig. 5 of the Appendix.
and black phosphorus, calculated from SCAN+rVV10 and r²SCAN+rVV10 compared with available data from experiments and other computational methods. A high-level, finite cluster CCSD(T) calculation benchmark studies with expanded comparison to high binding energies than the RPA, and r²SCAN underbinds bulk graphite, MoS₂, and TiS₂ than those found experimentally. We expect this may be due to the complicated electronic ground states of these materials, featuring charge density wave or superconductive phases, which were not considered in the present calculations. The effect of vdW functional corrections on these properties warrants further examination, but is beyond the scope of the current work.

To assess inter-layer binding energies for the L28 set in Table IV, we must use RPA reference values as those from more sophisticated methods [like the CCSD(T) references for S22] are unavailable. Select exceptions will be discussed further. While the RPA includes long-range vdW interactions, it lacks an accurate description of short-range correlation and tends to underestimate C₆ vdW coefficients. RPA may tend to underbind layered materials.

Table V presents inter-layer binding energies for a few solids where high-level QMC and silver-standard RPA values are available. No gold-standard correlated wavefunction calculations [such as CCSD(T)] for these solids have been undertaken at the time of writing. The QMC and experimental benchmarks show that RPA underbinds bulk graphite, MoS₂, and TiS₂ by 5–10 meV/Å². SCAN+rVV10 and r²SCAN+rVV10 are slightly more accurate than RPA for these three bulk materials, but overestimate the bilayer binding energies of graphite and MoS₂. SCAN+rVV10 often predicts larger binding energies than the RPA, and r²SCAN+rVV10 often predicts larger binding energies than SCAN+rVV10.

With these findings, we may tentatively say that r²SCAN+rVV10 is more accurate than RPA and SCAN+rVV10 for layered materials, though further benchmark studies with expanded comparison to high accuracy QMC calculations would be beneficial.

Alongside accurate static structural properties, dynamical lattice properties are also essential for materials design applications. We have recently shown that while SCAN gives accurate static structural properties, its accuracy for dynamical properties is limited by its numerical sensitivity, while r²SCAN maintains good performance for both static and dynamical properties. With this in mind, phonon dispersion in graphite and MoS₂ are presented in Fig. 3. For both systems, our r²SCAN+rVV10 results are in excellent agreement with the experimental data, especially for the lowest longitudinal acoustic (LA), longitudinal optical (LO), transverse acoustic (TA), and transverse optical (TO) phonon branches along the Γ–A (interlayer or c-axis) direction. The calculated strengths of these branches are dominated by the inter-layer binding forces, and are thus sensitive to vdW corrections. Without the rVV10 correction, the uncorrected r²SCAN severely underestimates these phonon branches.

### TABLE IV. Unsigned layer-layer binding energy Eᵦ in meV/Å² of graphite, hexagonal boron nitride (h-BN), MoS₂, TiS₂ and black phosphorus, calculated from SCAN+rVV10 and r²SCAN+rVV10 compared with available data from experiments and other computational methods.

|          | Expt. | QMC | RPA | rev-vdW-DF2 | SCAN+rVV10 | r²SCAN+rVV10 |
|----------|-------|-----|-----|-------------|------------|-------------|
| Graphite |       |     |     |             |            |             |
| Bulk     | 23.28 ± 1.94 | 22.91 ± 1.94 | 18.32 | 23.43 | 20.01 | 22.85 |
| Bilayer  | 13.51 ± 0.65 |             |       |            | 17.64 | 20.13 |
| h-BN     |       |     |     |             |            |             |
| Bulk     | 15.02 ± 0.46 | 14.49 | 21.15 | 20.62 | 22.55 |
| Bilayer  |             |       |       |            | 17.03 | 19.70 |
| MoS₂     |       |     |     |             |            |             |
| Bulk     | 34.33 ± 8.14 | 20.53 | 23.56 | 20.15 | 23.07 |
| Bilayer  |             |       |       |            | 17.71 | 20.06 |
| TiS₂     |       |     |     |             |            |             |
| Bulk     | 27.2 ± 0.8  | 18.88 | 24.8 | 18.97 | 21.49 |
| Bilayer  | 24.9 ± 1.6  | 23.8  | 17.71 | 20.06 |
| Black P  |       |     |     |             |            |             |
| Bulk     | 22.4 ± 1.0  | 22.59 |      |            | 25.46 |
| Bilayer  | 16.6 ± 2.2  | 21.28 |      |            | 23.97 |

### TABLE V. Equilibrium lattice constants of PPTA, found by stress minimization within the VASP code. Computed and experimental values from Ref. 35 are included for comparison.

|          | a(Å) | b(Å) | c(Å) | α(deg) |
|----------|------|------|------|--------|
| Expt.    | 7.87 | 5.18 | 12.9 | 90     |
| SCAN     | 7.75 | 5.10 | 12.96| 90.2   |
| SCAN+rVV10 | 7.21 | 5.08 | 12.95| 90     |
| r²SCAN+rVV10 | 7.86 | 5.09 | 12.96| 90.3   |
| This Work |        |      |      |        |
| SCAN     | 7.43 | 5.10 | 12.96| 90.1   |
| r²SCAN   | 7.99 | 5.14 | 12.96| 90.2   |
| r²SCAN+rVV10 | 7.35 | 5.15 | 12.99| 90.1   |
C. Complex materials: PPTA

Last, we present calculations for poly(p-phenylene terephthalamide) (PPTA), a layered material that is challenging for standard DFAs. PPTA, as shown in Fig. 4, is primarily vdW-bonded along its a-axis, hydrogen-bonded along its b-axis, and covalently bonded along its c-axis— a robust test for general-purpose DFAs. Table V presents the equilibrium structure of PPTA determined by SCAN, \( r^2 \)SCAN, and their rVV10 variants. The SCAN+rVV10 lattice parameters computed in Ref. 35 (and included in Table V) used an older version of VASP where the rVV10 stress tensor elements were not correctly computed. The calculations performed here use a corrected version of VASP and different computational parameters than those of Ref. 35. We used a \( 6 \times 9 \times 4 \) k-point grid (corresponding to \( k \)-point spacing of 0.15 Å\(^{-1} \)) and a plane-wave cutoff of 900 eV, whereas Ref. 35 used a \( 6 \times 6 \times 6 \) k-point grid and plane-wave cutoff of 520 eV. The number of grid points along c is well-converged at 4 points.

The effects of incorrect stress tensor elements are pronounced: the minima in the energy curves as a function of strained lattice parameters in Fig. 2 of Ref. 35 do not coincide with the values in their Table 1. As their relaxed values of \( b \) and \( c \) for SCAN and SCAN+rVV10 are similar to ours, we refit their energy data as a function of \( a \) at fixed \( b = 5.10 \) Å and \( c = 12.96 \) Å for SCAN, and \( b = 5.08 \) Å and \( c = 12.95 \) Å for SCAN+rVV10. We find \( a = 7.92 \) Å for SCAN, and \( a = 7.42 \) Å for SCAN+rVV10, more comparable to our values in Table V.

Although the \( a \) axis is the vdW-bonded axis in PPTA, the uncorrected SCAN provides the most correct description of inter-layer binding in PPTA. SCAN+rVV10 and \( r^2 \)SCAN+rVV10 severely overbind along the \( a \) axis, and do not provide substantive corrections to the parent meta-GGA along the \( b \) and \( c \) axes.

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Appendix: Additional data sets and figures

TABLE VI. Mean errors (ME, kcal/mol), mean absolute errors (MAE, kcal/mol), mean percentage errors (MPE), and mean absolute percentage errors (MAPE) in the unsigned interaction energies of the S22 data set, taken with respect to CCSD(T) results\textsuperscript{[5]}\textsuperscript{[6]}\textsuperscript{[7]}. Different (ENCUT, ENAUG) settings (described in Section IIIA) are tested for r\textsuperscript{2}SCAN+rVV10 and SCAN+rVV10; both values are in eV. Users who need less accuracy can use lower settings. For a concise presentation of this data, refer to Table I. Table VII in the Appendix presents interaction energies for each molecule in the S22 set, the CCSD(T) reference values, as well as values for other density functional approximations.

|                | SCAN+rVV10 |        | r\textsuperscript{2}SCAN+rVV10 |        |        | b = 11.95 |        | b = 12.3 |
|----------------|------------|--------|--------------------------------|--------|--------|----------|--------|----------|
|                | (600, 600) | (600, 1000) | (900, 2000) | (600, 600) | (600, 1000) | (900, 2000) | (900, 2000) |
| **Total**      |            |        |                                |        |        |
| MAE            | 0.58       | 0.46   | 0.43                           | 0.30   | 0.30   | 0.32     | 0.32   |
| ME             | 0.27       | 0.25   | 0.21                           | 0.20   | 0.20   | 0.22     | 0.22   |
| MAPE           | 10.85      | 7.31   | 6.34                           | 4.39   | 4.21   | 4.42     | 4.76   |
| MPE            | 1.65       | 2.05   | -0.29                          | 1.85   | 1.63   | 1.21     | 0.11   |
| **7 hydrogen-bonded complexes** |            |        |                                |        |        |
| MAE            | 0.99       | 0.95   | 0.89                           | 0.54   | 0.56   | 0.62     | 0.58   |
| ME             | 0.99       | 0.95   | 0.89                           | 0.54   | 0.56   | 0.62     | 0.58   |
| MAPE           | 7.78       | 7.54   | 6.64                           | 3.90   | 3.99   | 4.38     | 4.11   |
| MPE            | 7.78       | 7.54   | 6.64                           | 3.90   | 3.99   | 4.38     | 4.11   |
| **8 dispersion-bound complexes** |            |        |                                |        |        |
| MAE            | 0.42       | 0.17   | 0.22                           | 0.14   | 0.13   | 0.18     | 0.18   |
| ME             | -0.11      | -0.11  | -0.16                          | 0.09   | 0.08   | 0.08     | 0.00   |
| MAPE           | 14.00      | 6.17   | 6.65                           | 3.01   | 2.56   | 3.65     | 4.66   |
| MPE            | -3.93      | -2.11  | -6.02                          | 0.14   | -0.10  | -1.06    | -3.00  |
| **7 mixed complexes** |            |        |                                |        |        |
| MAE            | 0.36       | 0.31   | 0.20                           | 0.23   | 0.23   | 0.18     | 0.20   |
| ME             | -0.02      | -0.04  | -0.06                          | -0.01  | -0.03  | -0.02    | -0.06  |
| MAPE           | 10.32      | 8.37   | 5.69                           | 6.44   | 6.32   | 5.32     | 5.54   |
| MPE            | 1.88       | 1.32   | -0.69                          | 1.75   | 1.24   | 0.64     | -0.33  |

\textsuperscript{[5]} Different (ENCUT, ENAUG) settings (described in Section IIIA) are tested for r\textsuperscript{2}SCAN+rVV10 and SCAN+rVV10; both values are in eV. Users who need less accuracy can use lower settings. For a concise presentation of this data, refer to Table I. Table VII in the Appendix presents interaction energies for each molecule in the S22 set, the CCSD(T) reference values, as well as values for other density functional approximations.
TABLE VII. Positive interaction energy errors (approximate minus the CCSD(T) reference), in kcal/mol, for the molecular dimers in the S22 data set from PBE, rVV10, vdW-DF2 (numerical results from Ref. [24], SCAN results from Ref. [32], SCAN+rVV10 and r²SCAN+rVV10 with respect to the CCSD(T) results. Different (ENCUT, ENAUG) settings are tested for r²SCAN+rVV10 and SCAN+rVV10; both values are in eV. Absolute errors that are greater than twice the corresponding MAD are bold-faced.

|                | CCSD(T) | PBE rVV10 | vdW-DF2 | SCAN | SCAN+rVV10 | r²SCAN+rVV10 | SCAN+rVV10 |
|----------------|---------|-----------|---------|------|------------|-------------|-----------|
| **7 hydrogen-bound complexes** |         |           |         |      |            |             |           |
| NH₃ dimer (C2h) | 3.13    | -0.32     | 0.28    | -0.16| -0.01      | 0.04         | 0.05      |
| H₂O dimer (Cs) | 4.99    | -0.05     | 0.52    | -0.21| 0.44       | 0.34         | 0.35      |
| Formic acid dimer (C2h) | 18.75 | -0.51     | 1.22    | -1.98 | 2.18       | 1.84         | 1.87      |
| Formamide dimer (C2h) | 16.06 | -1.28     | 0.66    | -1.63| 0.48       | 0.35         | 0.39      |
| Uracil dimer (C2h) | 20.64   | -2.10     | 0.48    | -1.05| -0.15      | 0.27         | 0.32      |
| 2-pyridone–2-aminopyridine (C1) | 16.93 | -1.56     | 1.13    | -1.56 | 0.66       | 0.66         | 0.68      |
| Adenine–thymine WC (C1) | 16.66 | -2.31     | 0.76    | -1.92| -0.67      | 0.30         | 0.26      |
| **8 dispersion-bound complexes** |         |           |         |      |            |             |           |
| CH₄ dimer (D3d) | 0.53    | -0.43     | -0.04   | 0.15 | -0.18      | -0.02        | -0.01     |
| C₂H₄ dimer (D2d) | 1.47    | -1.14     | -0.06   | -0.15| -0.45      | -0.06        | -0.06     |
| Benzene–CH₄ (C3) | 1.45    | -1.40     | -0.01   | -0.16| -0.58      | -0.00        | -0.03     |
| Benzene dimer (C2h) | 2.65    | -4.50     | 0.07    | -0.50| -1.58      | 0.12         | 0.05      |
| Pyrazine dimer (Cs) | 4.25    | -4.93     | -0.22   | -0.96| -1.60      | -0.11        | -0.10     |
| Uracil dimer (C2) | 9.80    | -7.07     | -0.08   | -1.04| -1.84      | 0.45         | 0.50      |
| Indole–benzene (C1) | 4.52    | -6.69     | 0.01    | -1.08| -2.40      | 0.05         | 0.00      |
| Adenine–thymine (C1) | 11.73   | -10.31    | -0.31   | -2.15| -3.08      | 0.31         | 0.31      |
| **7 mixed complexes** |         |           |         |      |            |             |           |
| C₂H₄–C₂H₂ (C2v) | 1.50    | -0.32     | 0.17    | 0.03 | -0.16      | 0.07         | 0.06      |
| Benzene–H₂O (Cs) | 3.27    | -1.25     | 0.04    | -0.48| 0.00       | 0.43         | 0.40      |
| Benzene–NH₃ (Cs) | 2.31    | -1.38     | -0.04   | -0.32| -0.32      | 0.21         | 0.20      |
| Benzene–HCN (Cs) | 4.54    | -1.71     | -0.27   | -0.99| -0.48      | 0.07         | 0.06      |
| Benzene dimer (C2v) | 2.72    | -2.59     | -0.17   | -0.66| -1.24      | -0.17        | -0.19     |
| Indole–benzene (Cs) | 5.63    | -3.57     | -0.35   | -1.43| -1.56      | -0.19        | -0.19     |
| **Total** |         |           |         |      |            |             |           |
| MAE [REF CCSD(T)] | 2.67    | 0.32      | 0.94    | 0.94 | 0.30       | 0.30         | 0.32      |
| ME [REF CCSD(T)] | -2.67   | 0.17      | -0.92   | -0.66| 0.20       | 0.20         | 0.22      |
| STD DEV [REF CCSD(T)] | 2.55   | 0.43      | 0.71    | 1.09 | 0.44       | 0.45         | 0.49      |
| MAE [REF (900,2000)] | 0.10    | 0.08      | 0.00    | 0.36 | 0.36       | 0.36         | 0.36      |
| Material | $E_b$ | $c$ | $\Delta E_b$ | $\Delta c$ | $\Delta \alpha$ | $\text{MD}$ | $\text{STD DEV}$ |
|----------|------|----|-------------|----------|-------------|--------|-----------|
| h-BN     | 14.49 | 6.54 | 2.51 | 6.66 | 0.00 | 0.00 | -7.20 | 0.30 | -0.01 | 4.96 | 0.00 | -0.01 | 8.00 | -0.04 | -0.01 |
| Graphite | 18.32 | 6.70 | 2.46 | 5.13 | -0.11 | 0.00 | -10.40 | 0.16 | -0.01 | 1.63 | -0.05 | -0.01 | 4.53 | -0.07 | -0.00 |
| HiS$_2$ | 16.13 | 5.84 | 3.63 | 3.77 | -0.01 | -0.02 | -10.94 | 0.20 | 0.00 | -0.09 | 0.04 | -0.02 | 2.09 | 0.04 | -0.01 |
| HiSe$_2$ | 17.09 | 6.16 | 3.75 | 3.33 | 0.02 | -0.01 | -11.66 | 0.24 | 0.00 | -0.82 | 0.06 | -0.01 | 1.30 | 0.05 | 0.00 |
| HfTe$_2$ | 18.68 | 6.65 | 3.96 | 4.48 | 0.04 | -0.03 | -11.68 | 0.28 | 0.01 | -0.50 | 0.13 | -0.01 | 1.37 | 0.13 | 0.02 |
| MoS$_2$ | 20.53 | 12.30 | 3.16 | 3.00 | 0.04 | 0.01 | -14.86 | 0.52 | 0.01 | -0.32 | 0.18 | 0.01 | 2.71 | 0.13 | 0.02 |
| MoSe$_2$ | 19.63 | 12.93 | 3.29 | 3.45 | 0.12 | 0.01 | -14.01 | 0.61 | 0.02 | 0.08 | 0.24 | 0.01 | 2.91 | 0.19 | 0.02 |
| MoTe$_2$ | 20.80 | 13.97 | 3.52 | 3.30 | 0.11 | 0.01 | -13.95 | 0.66 | 0.00 | -0.18 | 0.30 | -0.01 | 2.22 | 0.25 | 0.03 |
| NbS$_2$ | 17.58 | 17.91 | 3.33 | 7.58 | 0.24 | -0.01 | -10.65 | 0.93 | 0.01 | 2.94 | 0.46 | 0.00 | 5.65 | 0.40 | 0.01 |
| NbSe$_2$ | 19.57 | 12.55 | 3.44 | 7.82 | -0.06 | 0.01 | -11.93 | 0.50 | 0.03 | 0.45 | 0.49 | 0.02 | 3.04 | 0.44 | 0.02 |
| NbTe$_2$ | 23.03 | 6.61 | 3.68 | 4.14 | 0.20 | -0.01 | -14.37 | 0.57 | -0.02 | -1.24 | 0.33 | -0.03 | 1.03 | 0.27 | 0.02 |
| PbO      | 20.25 | 5.00 | 3.96 | -3.30 | 0.05 | 0.07 | -8.43 | 0.10 | 0.03 | 3.08 | 0.07 | 0.03 | 1.40 | 0.01 | 0.03 |
| PdTe$_2$ | 40.17 | 5.11 | 4.02 | 3.44 | 0.05 | 0.05 | -14.98 | 0.07 | 0.03 | 2.25 | 0.08 | 0.02 | -0.25 | 0.06 | 0.04 |
| PtS$_2$  | 20.55 | 5.04 | 3.54 | 2.85 | -0.13 | 0.05 | -15.14 | 0.50 | -0.01 | -1.39 | 0.17 | -0.01 | 1.46 | 0.09 | 0.01 |
| PtSe$_2$ | 19.05 | 5.11 | 3.73 | 5.86 | -0.13 | 0.06 | -13.14 | 0.62 | -0.04 | 0.34 | 0.29 | -0.03 | 3.06 | 0.20 | 0.00 |
| TaS$_2$  | 17.68 | 5.90 | 3.36 | 8.29 | 0.00 | -0.01 | -10.30 | 0.24 | 0.00 | 3.74 | 0.06 | 0.01 | 6.43 | 0.05 | 0.01 |
| TaSe$_2$ | 19.44 | 6.27 | 3.48 | 6.37 | 0.02 | -0.01 | -12.12 | 0.25 | 0.00 | 2.69 | 0.06 | -0.01 | 5.24 | 0.04 | 0.01 |
| TiS$_2$  | 18.88 | 5.90 | 3.41 | 5.47 | -0.25 | -0.02 | -11.98 | -0.02 | 0.01 | 0.14 | -0.14 | -0.00 | 2.66 | -0.16 | 0.00 |
| TiSe$_2$ | 17.39 | 6.27 | 3.54 | 7.38 | -0.29 | -0.02 | -10.50 | 0.01 | 0.01 | 1.42 | -0.16 | 0.00 | 3.86 | -0.17 | 0.01 |
| TiTe$_2$ | 19.76 | 6.50 | 3.78 | 7.11 | 0.02 | -0.03 | -12.06 | 0.32 | -0.01 | 0.19 | 0.15 | -0.02 | 2.35 | 0.11 | 0.01 |
| VS$_2$   | 25.61 | 5.75 | 3.22 | 1.17 | 0.06 | -0.05 | -18.40 | 0.32 | -0.03 | -4.30 | 0.30 | 0.03 | -1.37 | 0.00 | 0.04 |
| VSe$_2$  | 22.26 | 6.11 | 3.36 | 3.26 | 0.05 | -0.04 | -15.02 | 0.38 | -0.03 | -2.64 | 0.08 | 0.04 | 0.14 | 0.03 | 0.05 |
| VT$_2$   | 20.39 | 6.58 | 3.64 | 6.27 | 0.01 | -0.05 | -12.89 | 0.55 | -0.09 | -0.56 | 0.10 | 0.02 | 1.35 | 0.01 | 0.06 |
| WS$_2$   | 20.24 | 12.32 | 3.15 | 3.69 | 0.09 | 0.02 | -12.15 | 0.32 | 0.03 | 0.56 | 0.21 | 0.01 | 3.60 | 0.16 | 0.02 |
| WSe$_2$  | 19.98 | 12.96 | 3.28 | 3.45 | 0.13 | 0.02 | -13.29 | 0.44 | 0.03 | 0.25 | 0.25 | 0.01 | 3.06 | 0.22 | 0.03 |
| ZrS$_2$  | 16.98 | 5.81 | 3.66 | 3.09 | 0.02 | -0.01 | -11.55 | 0.21 | 0.03 | -0.85 | 0.06 | 0.02 | 1.35 | 0.05 | 0.01 |
| ZrSe$_2$ | 18.53 | 6.13 | 3.77 | 2.55 | 0.02 | 0.00 | -12.66 | 0.24 | 0.03 | -1.84 | 0.08 | 0.02 | 0.34 | 0.06 | 0.02 |
| ZrTe$_2$ | 16.34 | 6.66 | 3.95 | 8.84 | 0.01 | -0.02 | -8.33 | 0.26 | 0.05 | 3.33 | 0.08 | 0.03 | 5.23 | 0.07 | 0.05 |

**Note:** Bold-faced values indicate absolute errors that are greater than twice the MAD. The deviations are reported in meV/Å. The table entries are absolute errors from RPA calculations and the corresponding MADs are also presented.
TABLE IX. Lattice constants $c$ and $a$ in Å for 28 layered materials (L28 data set) from SCAN+$r$VV10 and $r^2$SCAN+$r$VV10. Deviations are reported under $\Delta c$ and $\Delta a$ columns. Different (ENCUT, ENAUG) settings are presented; both values are in eV. The experimental values of $c$ and $a$ are included for comparison. Mean deviations (MDs) and mean absolute deviations (MADs) are taken with respect to the largest ENCUT, 800 eV, and ENAUG, 2000 eV, setting. $r^2$SCAN+$r$VV10 approaches its converged values more rapidly than does SCAN+$r$VV10.

|        | Expt. | SCAN+$r$VV10 | $r^2$SCAN+$r$VV10 |
|--------|-------|--------------|-------------------|
|        | $c$   | $a$          | $\Delta c$ | $\Delta a$ | $c$ | $a$ | $\Delta c$ | $\Delta a$ | $c$ | $a$ |
| h-BN   | 6.54  | 2.51         | $-0.08$    | $-0.00$    | -0.07 | -0.00 | 6.54 | 2.50 | 6.50 | 2.50 |
| Graphite | 6.70  | 2.46         | $0.03$     | $0.00$     | $0.02$ | $0.00$ | 6.65 | 2.45 | 6.63 | 2.46 |
| HS$_2$ | 5.84  | 3.63         | $0.04$     | $0.00$     | $-0.01$ | $-0.00$ | 5.87 | 3.61 | 5.88 | 3.62 |
| HSSe$_2$ | 6.16  | 3.75         | $-0.01$    | $-0.00$    | $0.00$ | $0.00$ | 6.22 | 3.74 | 6.21 | 3.75 |
| HTTe$_2$ | 6.65  | 3.96         | $-0.01$    | $-0.00$    | $0.00$ | $0.00$ | 6.79 | 3.95 | 6.78 | 3.98 |
| MoS$_2$ | 12.30 | 3.16         | $0.02$     | $-0.00$    | $-0.00$ | $0.00$ | 12.47 | 3.17 | 12.43 | 3.18 |
| MoSe$_2$ | 12.93 | 3.29         | $-0.02$    | $-0.00$    | $0.00$ | $0.00$ | 13.17 | 3.30 | 13.12 | 3.31 |
| MoTe$_2$ | 13.97 | 3.52         | $0.04$     | $-0.00$    | $-0.00$ | $0.00$ | 14.26 | 3.51 | 14.22 | 3.55 |
| NbS$_2$ | 17.91 | 3.33         | $-0.01$    | $-0.00$    | $-0.03$ | $-0.00$ | 18.34 | 3.33 | 18.31 | 3.34 |
| NbSe$_2$ | 12.55 | 3.44         | $0.00$     | $-0.00$    | $-0.01$ | $-0.00$ | 13.01 | 3.46 | 12.99 | 3.46 |
| NbTe$_2$ | 6.61  | 3.68         | $0.00$     | $-0.00$    | $-0.00$ | $0.00$ | 6.95 | 3.65 | 6.88 | 3.70 |
| PbO    | 5.00  | 3.96         | $-0.04$    | $0.01$     | $-0.02$ | $-0.00$ | 4.94 | 3.99 | 5.01 | 3.99 |
| PbTe$_2$ | 5.11  | 4.02         | $0.00$     | $-0.00$    | $-0.00$ | $0.00$ | 5.03 | 4.04 | 5.17 | 4.06 |
| Pt$_2$S$_2$ | 5.04  | 3.54         | $0.06$     | $-0.00$    | $-0.01$ | $0.00$ | 5.19 | 3.53 | 5.13 | 3.55 |
| PtSe$_2$ | 5.11  | 3.73         | $-0.01$    | $0.00$     | $-0.01$ | $0.00$ | 5.40 | 3.70 | 5.31 | 3.73 |
| TaS$_2$ | 5.90  | 3.36         | $-0.03$    | $-0.00$    | $-0.01$ | $0.00$ | 5.96 | 3.35 | 5.95 | 3.37 |
| TaSe$_2$ | 6.27  | 3.48         | $-0.01$    | $-0.00$    | $-0.00$ | $0.00$ | 6.34 | 3.47 | 6.31 | 3.49 |
| TiS$_2$ | 5.90  | 3.41         | $0.00$     | $0.00$     | $-0.01$ | $-0.00$ | 5.77 | 3.41 | 5.74 | 3.41 |
| TiSe$_2$ | 6.27  | 3.54         | $0.02$     | $-0.01$    | $-0.01$ | $0.00$ | 6.12 | 3.55 | 6.10 | 3.55 |
| TiTe$_2$ | 6.50  | 3.78         | $0.01$     | $-0.00$    | $-0.00$ | $0.00$ | 6.64 | 3.77 | 6.61 | 3.79 |
| VS$_2$ | 5.75  | 3.22         | $0.05$     | $-0.00$    | $-0.01$ | $-0.00$ | 5.76 | 3.25 | 5.75 | 3.26 |
| VSe$_2$ | 6.11  | 3.36         | $-0.02$    | $-0.00$    | $-0.00$ | $0.00$ | 6.19 | 3.40 | 6.14 | 3.41 |
| VT$_2$ | 6.58  | 3.64         | $0.07$     | $-0.01$    | $-0.05$ | $0.02$ | 6.68 | 3.66 | 6.59 | 3.70 |
| WS$_2$ | 12.32 | 3.15         | $0.01$     | $-0.00$    | $-0.01$ | $-0.00$ | 12.51 | 3.16 | 12.48 | 3.17 |
| WSe$_2$ | 12.96 | 3.28         | $-0.02$    | $-0.00$    | $-0.01$ | $-0.00$ | 13.22 | 3.29 | 13.18 | 3.31 |
| ZrS$_2$ | 5.81  | 3.66         | $0.01$     | $-0.00$    | $-0.01$ | $-0.00$ | 5.88 | 3.68 | 5.86 | 3.67 |
| ZrSe$_2$ | 6.13  | 3.77         | $-0.01$    | $-0.00$    | $0.00$ | $0.00$ | 6.21 | 3.79 | 6.19 | 3.79 |
| ZrTe$_2$ | 6.66  | 3.95         | $0.02$     | $0.00$     | $0.00$ | $0.00$ | 6.74 | 3.98 | 6.73 | 4.00 |
| MAD    | 0.024 | 0.002        | 0.010      | 0.001      | 0.000 | 0.000 | 0.009 | 0.003 | 0.008 | 0.000 | 0.000 |
| MD     | 0.006 | -0.001       | -0.008     | 0.000      | 0.000 | 0.000 | -0.007 | -0.001 | -0.006 | -0.000 | 0.000 |
| STD DEV| 0.032 | 0.003        | 0.017      | 0.000      | 0.000 | 0.000 | 0.015 | 0.003 | 0.013 | 0.000 | 0.000 |
FIG. 5. Phonon dispersion in graphite and MoS$_2$ analogous to Fig. 3 but using PBE$^{58}$ and PBE-D4$^{18}$ instead of $r^2$SCAN. Just as for $r^2$SCAN, adding a dispersion correction to PBE produces a more realistic phonon dispersion, especially along the inter-layer direction A-Γ. In MoS$_2$, PBE-D4 is in good accord with available experimental phonon dispersion data, however $r^2$SCAN+rVV10 provides a more realistic description of phonons in graphite along the inter-layer direction.
ENCUT = 900 eV, ENAUG = 2000 eV

FIG. 6. Box and whisker plot of the S22 errors (kcal/mol) for $r^2$SCAN+rVV10 with the presently-fitted value $b = 11.95$, with the VV10-fitted $b = 12.3$, and SCAN+rVV10. See Tables II and VII for tabulated errors.
FIG. 7. Box and whisker plot of the L28 set binding energy $E_b$ errors (meV/Å$^2$) for rev-vdW-DF2, $r^2$SCAN+rVV10 with the presently-fitted value $b = 11.95$, and SCAN+rVV10. See Tables II and VIII for tabulated errors.

FIG. 8. Box and whisker plot of the L28 set out-of-plane lattice constant $c$ errors (Å) for rev-vdW-DF2, $r^2$SCAN+rVV10 with the presently-fitted value $b = 11.95$, and SCAN+rVV10. See Tables II and VIII for tabulated errors.
FIG. 9. Box and whisker plot of the L28 set in-plane lattice constant $a$ errors (Å) for rev-vdW-DF2, $r^2$SCAN+rVV10 with the presently-fitted value $b = 11.95$, and SCAN+rVV10. See Tables II and VIII for tabulated errors.