PHYSICAL REVIEW RESEARCH **4**, 023061 (2022)

First steps towards achieving both ultranonlocality and a reliable description of electronic binding in a meta-generalized gradient approximation

Timo Lebeda, Thilo Aschebrock, and Stephan Kümmel[*]
Theoretical Physics IV, University of Bayreuth, 95440 Bayreuth, Germany

(Received 27 January 2022; accepted 24 March 2022; published 21 April 2022)

It has been demonstrated that a meta-generalized gradient approximation (meta-GGA) to the exchange-correlation energy of density functional theory can show a pronounced derivative discontinuity and significant ultranonlocality similar to exact exchange, and can accurately predict the band gaps of many solids. We here investigate whether within the meta-GGA form these properties are compatible with a reasonable accuracy for electronic binding energies. With the help of two transparent and inexpensive correlation functional constructions we demonstrate that this is the case. We report atomization energies, show that reliable bond lengths are obtained for many systems, and find promising results for reaction barrier heights, while keeping the strong derivative discontinuity and ultranonlocality, and thus accuracy for band gaps.

DOI: 10.1103/PhysRevResearch.4.023061

I. INTRODUCTION

Density functional theory (DFT) is one of the most widely used many-body theories, with successes in many branches of electronic structure theory [1]. DFT comes in the mathematical structure of a single-particle theory, yet incorporates many-particle effects via the exchange-correlation (xc) energy functional E_{xc}. This allows the practitioner who is interested in electronic structure problems to choose the level of accuracy and computational cost by choosing a certain class of xc approximation. The local density approximation [2,3] (LDA) and generalized gradient approximation [4,5] (GGA) functionals are computationally efficient. Higher accuracy can typically be reached with hybrid functionals [6], where recent years have seen a lot of activity in the development of advanced forms such as local hybrid functionals [7–10], range-separated hybrid functionals [11–20], and local range-separated hybrid functionals [21–23]. Further advanced xc approximations can reach a level at which DFT rivals wave-function-based methods in accuracy [24].

However, much of the success of DFT in applied electronic structure theory rests on its computational efficiency, and a certain dilemma has thus been limiting DFT applications for a long time: The computationally efficient LDA and GGA forms suffer from systematic shortcomings, many of which can be traced back to the very features which make these forms efficient. The sole dependence of their energy density on the electron density $n(\mathbf{r})$ and its gradient $\nabla n(\mathbf{r})$ leads to expressions for the energy and the xc potential that can efficiently be numerically evaluated, but also prevents that important features of the exact xc functional are reflected in these functionals. Especially the lack of a derivative discontinuity in E_{xc}, which leads to a particle-number discontinuity in the corresponding Kohn-Sham xc potential $v_{\mathrm{xc}}(\mathbf{r})$, has seriously detrimental consequences in many applications, e.g., the prediction of band gaps [25] and response properties [26–28]. The derivative discontinuity and the corresponding (ultra-)nonlocality can be incorporated into E_{xc}, e.g., by including exact exchange [29–31] or via self-interaction corrections [32–34]. This, however, increases the computational cost sharply compared to a plain GGA calculation. Model potentials [35–37] and especially tailored GGAs can provide some improvement [38,39], but come with their own set of problems and serious limitations [40–44].

The functional class of the meta-GGAs [45] seems ideally suited to resolve the dilemma between the need for semilocal computational efficiency on the one hand, and the need to capture (ultra-)nonlocality-physics on the other hand. Depending only on the density, its gradient, and the gradient of the orbitals, makes meta-GGAs semilocal in terms of the computational cost. However, as the orbitals are nonlocal functionals of the density, meta-GGAs, in principle, depend nonlocally on the density as well [31]. Unfortunately though, the experience in practice for a long time had been that typical meta-GGAs can improve over GGAs in the accuracy of, e.g., atomization energies [46–50], yet offer little nonlocality [51].

With the recent construction of the meta-GGA by T. Aschebrock, S. Kümmel (TASK) for exchange [52] it has been demonstrated that a meta-GGA can have a pronounced derivative discontinuity and can reproduce many of the (ultra-)nonlocal features that previously could only be reached with exact exchange. This demonstrated that (ultra-)nonlocality is not just, in principle, a feature that meta-GGAs can have, but also in practice. However, the TASK meta-GGA for exchange has so far been combined with plain LDA correlation [52] (LDAc) [53]. This was a preliminary step, taken for the simple reason that LDAc is universal and thus better suited for combining it with a new exchange functional than

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[*] stephan.kuemmel@uni-bayreuth.de; http://tp4.uni-bayreuth.de/en

a correlation functional that has been tailored towards some other specific exchange functional. While the combination of TASK exchange with LDAc yields excellent results for the fundamental band gap of many materials in the generalized Kohn-Sham evaluation [39,52], and promising results for response properties [52,54], the atomization energies are of limited accuracy [52]. Thus, one may speculate whether the meta-GGA form is too limited to yield accurate binding energy physics and pronounced nonlocality at the same time, e.g., whether one and the same meta-GGA can yield both good atomization energies and band gaps.

In this article we take a step towards answering this question. Our focus here is not (yet) on the development of the best possible meta-GGA correlation functional to go with the TASK exchange functional, but on exploring whether having a pronounced derivative discontinuity and having good atomization energies are necessarily mutually exclusive within the meta-GGA functional form. To this end we focus on very transparent forms for a semilocal correlation functional that ensure that the excellent band gaps that are found with TASK exchange and LDAc remain intact, while atomization energies are improved. We specifically discuss the role of one-electron self-interaction, and we also report bond lengths, atomization energies, and reaction barrier heights for TASK exchange in combination with different correlation functionals. We find that it is possible to construct a functional that yields the same accurate band gaps as the combination of TASK and LDAc, while at the same time yielding greatly improved atomization energies.

II. TASK EXCHANGE WITH ISO-ORBITAL CORRECTED LDA CORRELATION

Following usual practice we write the correlation energy in terms of the electron density $n(\mathbf{r})$ and the correlation energy density per particle ε_c,

$$E_c[n_\uparrow, n_\downarrow] = \int n(\mathbf{r})\varepsilon_c(\mathbf{r})d^3\mathbf{r}. \qquad (1)$$

As explained above our aim is to check whether one can find a transparent correlation functional that yields reasonable atomization energies when combined with the TASK exchange functional, without ruining the good features of TASK exchange that result from its pronounced nonlocality. Our first step is to correct for one-electron self-correlation. This is close-lying and natural because first, TASK exchange was designed with a focus on reducing one-electron errors (by ensuring the correct energy for the hydrogen atom), second, TASK exchange mimics exact exchange, which is one-electron self-interaction free, and third, one-electron self-correlation can readily be eliminated within the meta-GGA form by using iso-orbital indicators [55]. One such indicator is

$$z(\mathbf{r}) = \frac{\tau_W(\mathbf{r})}{\tau(\mathbf{r})}, \qquad (2)$$

where

$$\tau(\mathbf{r}) = \frac{\hbar^2}{2m}\sum_{\sigma=\uparrow,\downarrow}\sum_{i=1}^{N_\sigma}|\nabla\varphi_{i\sigma}(\mathbf{r})|^2 \qquad (3)$$

is the noninteracting kinetic energy density that is obtained from the Kohn-Sham or generalized Kohn-Sham spin orbitals $\varphi_{i\sigma}(\mathbf{r})$, respectively, and

$$\tau_W(\mathbf{r}) = \frac{\hbar^2}{8m}\frac{|\nabla n(\mathbf{r})|^2}{8n(\mathbf{r})} \qquad (4)$$

is the von Weizsäcker kinetic energy density. The latter is the single-orbital limit of τ, thus, z approaches one in regions of spatially identical orbitals. By multiplying z with the square of the spin polarization

$$\zeta(\mathbf{r}) = \frac{n_\uparrow(\mathbf{r}) - n_\downarrow(\mathbf{r})}{n_\uparrow(\mathbf{r}) + n_\downarrow(\mathbf{r})}, \qquad (5)$$

one obtains the one-spin-orbital region indicator $z\zeta^2$. This can be used to define a transparent form of an LDA-based self-interaction corrected correlation (CC) functional [10] via

$$\varepsilon^{\mathrm{CC}}(\mathbf{r}) = (1 - z(\mathbf{r})\zeta^2(\mathbf{r}))\varepsilon^{\mathrm{LDAc}}(\mathbf{r}), \qquad (6)$$

where $\varepsilon^{\mathrm{LDAc}}$ is the correlation energy density of LDA, including its spin dependence, which we, however, do not denote explicitly for ease of notation. In our calculations we use the LDA in the parametrization of Perdew and Wang [3].

The CC correlation is a very transparent functional that does not contain any empirical parameter. Combining it with TASK exchange, denoted by TASK+CC in the following, leads to a nonempirical meta-GGA that fulfills many exact constraints. TASK exchange satisfies all exact constraints for exchange that SCAN exchange does [52,56]. Since τ_W and hence z vanish for a uniform density, the CC correlation equals the LDA correlation in the uniform-density limit and for spin-unpolarized densities. On the other hand, the CC correlation vanishes for any fully spin-polarized one-electron density because $(1 - z\zeta^2)$ vanishes in this case, resulting in no one-electron self-interaction error. Thus, TASK+CC is exact for the ground-state energy of the hydrogen atom. The same would hold for $(1 - z|\zeta|^k)$ with $k > 0$, but in our experience $k = 2$ is optimal. Both the LDA correlation energy and the CC correlation energy are nonpositive and vanish under uniform density scaling to the low-density limit, i.e., go to the correct limiting value. The combined functionals TASK+LDAc and TASK+CC are size extensive and satisfy the general Lieb-Oxford bound ($F_{\mathrm{xc}} \leqslant 2.215$) [57] as tightened by Chan and Handy [58], where F_{xc}, the enhancement over local exchange, is defined by $E_{\mathrm{xc}} = \int n\varepsilon_{\mathrm{x}}^{LDA}F_{\mathrm{xc}}d^3r$. Finally, TASK+LDAc and TASK+CC are smooth at fixed electron number, which is beneficial for numerical computations and has recently been suggested as an exact constraint that a meta-GGA can satisfy [59].

By construction, the CC correlation equals the LDA correlation in the uniform-density limit and for spin-unpolarized systems. Therefore, the sizable derivative discontinuity is guaranteed to be preserved for spin-unpolarized systems and it is guaranteed that TASK+CC yields the same good results as TASK+LDAc for the band gaps that were studied in Ref. [52].

For a first impression of the performance of TASK+CC in predicting atomization energies we use a test set of 10 diatomic molecules [63]. For those, very accurate Kohn-Sham all-electron calculations can be done on a real-space prolate spheroidal grid with the all-electron code DARSEC [64].

TABLE I. Atomization energies of diatomic molecules in kcal/ mol. The values for the established nonempirical functionals LDA [3], Perdew-Burke-Ernzerhof (PBE) [5], and the Strongly Constrained and Appropriately Normed meta-GGA (SCAN) [56] are listed for comparison. TASK+LDAc, TASK+CC, and TASK+CCaLDA are TASK exchange with PW92-LDA correlation, CC correlation, and the functional that combines CC and LDA correlation in the form of Eq. (9) (CCaLDA), respectively. The CCaLDA correlation is introduced in Sec. III. The experimental values (with zero point vibration removed) and the experimental bond lengths were taken from Ref. [60] (cf. the Supplemental Material [61] of Ref. [22] for numerical details and comments about numerical stability [52,62]).

Molec.	LDA	PBE	SCAN	TASK +LDAc	TASK +CC	TASK +CCaLDA	Expt.
H_2	112.9	104.6	107.7	117.0	144.8	117.0	109.5
FH	162.0	142.0	138.7	139.2	158.5	144.6	141.1
LiH	60.8	53.5	55.6	58.9	79.4	64.0	58.0
Li_2	23.8	19.9	18.1	11.6	24.8	21.8	24.7
LiF	156.3	139.0	138.1	130.2	142.2	140.7	138.3
F_2	78.0	52.8	37.4	23.4	34.2	34.2	38.4
CO	299.2	269.1	255.2	234.6	265.9	265.9	259.5
N_2	268.0	243.9	220.9	174.9	227.4	227.4	228.3
NO	199.4	170.9	146.5	109.3	147.3	147.3	152.5
O_2	175.1	144.1	127.0	103.8	124.7	124.7	120.5
MSE	26.5	6.9	−2.6	−16.8	7.8	1.7	
MAE	26.7	9.7	3.9	18.5	9.9	4.3	
RMSE	32.7	12.3	4.6	24.8	14.6	4.7	

Table I compares TASK+CC with LDA, the GGA PBE [5], the meta-GGA SCAN [56], and TASK+LDAc [65]. We used the Krieger-Li-Iafrate (KLI) apprximation [29] for the Kohn-Sham calculations with orbital dependent functionals, and information about numerical details, e.g., the advantages of writing the functional derivative in terms of $\sqrt{n}$ instead of n, is given in the Supplemental Material [61]. At the bottom of the table the usual statistical measures summarize the performance of each functional, i. e., the mean absolute error (MAE), the mean signed error (MSE), and the root mean square error (RMSE), cf. the Supplemental Material [61]. The comparison shows that TASK+CC is still significantly less accurate than SCAN, but the CC correlation reduces the MAE by more than a factor of two compared to TASK+LDAc. TASK+CC is thus on a similar level of accuracy as the PBE functional for this test set. This first test therefore already demonstrates that a suitable correlation functional can significantly improve atomization energies while maintaining the pronounced ultranonlocality of the TASK exchange.

A closer look at the data reveals that the error is mostly due to a systematic overestimation of the atomization energy of hydrogen-containing molecules. For molecules that do not contain hydrogen, the MAE is reduced by a further factor of two. Therefore, in the next section we explore how much can be gained by focusing on hydrogen in a specific construction called CCaLDA correlation. However, before doing so we study the main grroup atomization energies (MGAE109) [66] as a large benchmark test set for atomization energies. Table II summarizes the statistical measures for MGAE109. Similar to what we have already seen for the diatomic test set, TASK+CC improves significantly over TASK+LDAc. For detailed data we refer to Table S.II in the Supplemental Material [61]. These show that the performance is systematically different for different classes of molecules. TASK+CC is systematically overbinding all hydrocarbons by about 5 kcal/ mol. For inorganic molecules the situation is different and less systematic. While TASK+CC tends to underbind most of the inorganic molecules that contain elements from the second main group, it overbinds many molecules that contain heavier elements. However, TASK+CC is systematically overbinding all molecules which contain hydrogen. Therefore, one again observes that hydrogen containing systems play a special role for TASK+CC, and we thus take a closer look at them in the following. We mention in passing that in the Supplemental Material [61] we also show results for TASK exchange alone and in combination with SCAN correlation.

III. HEURISTIC FURTHER IMPROVEMENT OF ATOMIZATION ENERGIES

The systematic overestimation of the atomization energies of hydrogen-containing molecules that we observed in the previous section can be improved by changing the isoorbital limit. In the following we show that one can reduce the MAE of TASK+CC by nearly another factor of two by modifying solely this limit. To this end, we interpolate between the CC correlation and LDA correlation by a function of the isoorbital indicator α [69], which is defined by

$$\alpha(\mathbf{r}) = \frac{\tau(\mathbf{r}) - \tau_W(\mathbf{r})}{\tau_F(\mathbf{r})}, \tag{7}$$

where

$$\tau_F(\mathbf{r}) = \frac{3\hbar^2}{10m}(3\pi^2)^{\frac{2}{3}} n(\mathbf{r})^{\frac{5}{3}} \tag{8}$$

TABLE II. MGAE109 [66] main-group atomization energies per bond in kcal/ mol. The calculations were performed with the QZ4P-basis set in the Amsterdam Density Functional (ADF) program of the Amsterdam Modeling Suite (AMS) [67] using the generalized Kohn-Sham scheme.

	LDA	PBE	SCAN	TASK +LDAc	TASK +CC	TASK +CCaLDA [68]
MSE	19.5	3.8	−0.1	−12.1	6.1	−1.4
MAE	19.5	4.2	1.4	12.3	7.3	4.2
RMSE	21.7	6.0	2.2	14.4	9.2	5.2

TABLE III. MGBL20 [71] main-group bond lengths in Å. The calculations were performed with the QZ4P-basis set in AMS/ADF [67].

	LDA	PBE	SCAN	TASKx	TASK +LDAc	TASK +SCANc	TASK +CC
	MGHBL9 [72] (Main-group hydrogenic bond lengths)						
MSE	0.015	0.011	0.002	0.016	0.001	0.009	0.001
MAE	0.015	0.011	0.002	0.016	0.005	0.009	0.005
RMSE	0.015	0.011	0.003	0.018	0.007	0.011	0.007
	MGNHBL11 [71] (Main-group Non-hydrogenic bond lengths)						
MSE	−0.006	0.007	−0.005	0.024	0.006	0.006	0.006
MAE	0.007	0.007	0.006	0.024	0.006	0.009	0.006
RMSE	0.010	0.008	0.007	0.030	0.011	0.012	0.011

is the uniform-density limit of τ. The correlation energy density of the CCaLDA functional reads

$$\varepsilon^{\mathrm{CCaLDA}}(\mathbf{r}) = f(\alpha(\mathbf{r}))\varepsilon^{\mathrm{CC}}(\mathbf{r}) + [1 - f(\alpha(\mathbf{r}))]\varepsilon^{\mathrm{LDAc}}(\mathbf{r}), \quad (9)$$

where we use the function

$$f(\alpha(\mathbf{r})) = (1 + c)\frac{\alpha(\mathbf{r})}{1 + c\alpha(\mathbf{r})} \quad (10)$$

to switch between LDA and CC correlation. The form of $f(\alpha)$ and the parameter $c = 10000$ are chosen such that only the isoorbital limit ($\alpha \approx 0$) of $\varepsilon^{\mathrm{CC}}$ is modified. The changes in the atomization energies that result from the CCaLDA construction can be studied in Table I for the dimer test set. As intended, the atomization energies of TASK+CCaLDA match those of TASK+CC for molecules containing neither hydrogen nor lithium, while the energy of H_2 matches that of TASK+LDAc. Only the energies of the hydrogen atom and the lithium atom are changed compared to TASK+CC, where the latter effect is due to the singly occupied 2s orbital in the lithium atom, which induces a relatively large iso-orbital region. As one can see in Table II, TASK+CCaLDA also significantly improves the prediction of the atomization energies for the large MGAE109 test set and is on par with PBE. Taking into account that the TASK exchange is designed for a qualitatively correct description of ultranonlocality and the derivative discontinuity, this is an encouraging observation. It is noteworthy also from the perspective that the CCaLDA functional does not use any empirical parameters, as c has not been fitted to minimize the deviation from experimental data, but was chosen to provide the desired single-orbital limit. Detailed data for the MGAE109 test can be found in Table S.II in the Supplemental Material [61].

For putting these results into a larger perspective we note that the LDA correlation is far from being the optimal choice for a correlation functional in the iso-orbital limit: Neither is it free from self-interaction, nor is it the best choice for optimizing the atomization energies of TASK for hydrogen-containing molecules. We are therefore convinced that the MAE could be further improved by a more sophisticated choice for the iso-orbital limit. However, we do not pursue a functional construction of the TASK+CCaLDA type any further because of a conceptual shortcoming: Compared to TASK+CC, TASK+CCaLDA gains its increased accuracy for the atomization energies of hydrogen containing systems by making a correction to the energy of the hydrogen atom, which is exact in TASK+CC, instead of the energy of the hydrogen-containing molecules, which would need the correction. The results for TASK+CCaLDA can therefore be seen in the light of Ref. [70], which discusses the limits of judging the quality of an exchange-correlation approximation based on the errors in atomization energies. Nevertheless, the TASK+CCaLDA construction has the value that it demonstrates that atomization energies at least on the level of PBE, and at the same time band gaps on the level of TASK+LDAc, are possible with a meta-GGA. Further efforts for a meta-GGA correlation functional to go along with TASK exchange will therefore be well invested. In such efforts, one will need to pay attention to the form of the orbital dependence of the correlation functional, since this dependence will in general also lead to a correlation contribution to the derivative discontinuity.

TABLE IV. DGH4 [73] bond lengths of diatomic molecules containing heavy elements, in Å. The calculations were performed with the QZ4P-basis set in AMS/ADF [67]. Relativistic effects were included in the ZORA [74] approximation (required for Ag).

Molecule	LDA	PBE	SCAN	TASKx	TASK +LDAc	TASK + SCANc	TASK +CC	Expt.
NaBr	2.471	2.518	2.503	2.678	2.638	2.623	2.638	2.502
HBr	1.430	1.431	1.417	1.432	1.413	1.420	1.413	1.414
ZnS	2.007	2.046	2.017	2.053	2.026	2.006	2.026	2.046
Ag2	2.485	2.571	2.520	2.605	2.564	2.519	2.564	2.530
MSE	−0.025	0.019	−0.008	0.069	0.037	0.019	0.037	
MAE	0.032	0.019	0.011	0.069	0.047	0.045	0.047	
RMSE	0.034	0.024	0.015	0.096	0.071	0.064	0.071	

TABLE V. DBH24/08 [78] barrier heights in kcal/ mol. The calculations were performed with the QZ4P-basis set in AMS/ADF [67].

	LDA	PBE	SCAN	TASKx	TASK +LDAc	TASK +SCANc	TASK +CC
MSE	−13.7	−8.5	−7.2	5.8	4.1	−0.6	2.6
MAE	13.7	8.5	7.3	7.5	5.0	3.1	4.5
RMSE	17.1	10.4	8.3	9.0	6.3	4.0	6.5

IV. BOND LENGTHS AND BARRIER HEIGHTS

In the remaining part of this paper we return to investigating TASK+CC, as this functional is better justified than CCaLDA from a conceptual point of view, and also test TASK+LDAc for further observables, namely bond lengths and reaction barriers. As the functionals TASK+LDAc, TASK+CC, and TASK+CCaLDA are equivalent for spin-unpolarized systems, the bond-length tests done for TASK-LDAc also tell us the bond lengths that one obtains with the other two functionals for spin-unpolarized systems.

We show results for the bond lengths of molecules consisting of main-group atoms (MGBL20) in Table III (details are listed in Table S.III in the Supplemental Material [61]) and for bond lengths of diatomic molecules which contain heavy elements (DGH4) in Table IV. TASK+LDAc has an MAE of about 0.005 Å for the MGBL20 set of main-group bond lengths, i.e., an accuracy on the level of PBE and SCAN. For the DGH4 set which contains heavy elements, TASK+LDAc performs significantly worse. However, the error is at the level of PBE and SCAN except for the NaBr molecule. The bond length of the latter is overestimated by more than 0.1 Å. Table S.I in the Supplemental Material [61] shows that this is a general feature of TASK for systems which contain alkali metals [75]. We have analyzed the interplay of s and α in these systems and found that systems with alkali metal atoms are special because both α and s become rather large in the core-valence separation region. This finding is to some extent in line with earlier, GGA-based observations about the special role of s in alkali-metal atom containing solids [76], and systematic deviations for solids with alkali metal atoms have also been observed with the Haas, Tran, Blaha, Schwarz (HTBS) GGA [77]. The bond length analysis thus revealed an issue that will need to be addressed in future work.

Table V shows results for a test set of diverse barrier heights (DBH24). For detailed data we refer to Table S.IV in the Supplemental Material [61]. The main observation is that TASK in combination with each of the correlation functionals reaches a remarkable accuracy in comparison to LDA, PBE, and SCAN. This observation is interesting also from the perspective that it has been argued that for a good description of barrier heights, true exchange nonlocality is beneficial [56,79]. From this point of view the good results that we find here can be interpreted as a natural consequence of the ultranonlocality that has been one of the main guiding principles in the construction of the TASK functional.

V. SUMMARY AND CONCLUSIONS

Traditionally, there has been a dividing gap in the world of density functional exchange-correlation approximations. They were either computationally efficient like the (semi)local LDA and GGAs, but lacked a derivative discontinuity and ultranonlocality. Or they could show ultranonlocality, like the self-interaction corrections [34] and various forms of hybrid functionals [18], yet at the price of a steeply increased computational cost. It has long been known that meta-GGAs, in principle, can build a bridge over this gap, because being orbital dependent they can incorporate ultranonlocality, while using the density, the orbitals and their gradients only locally, their computational cost remains semilocal [31]. The TASK meta-GGA for exchange is a functional that, different from many previous meta-GGAs, shows pronounced ultranonlocality. Previous work that combined it with LDA correlation demonstrated very good accuracy for the band gaps of many solids, but a limited accuracy for the atomization energies of molecules. This raised the question whether it is at all possible to achieve both, good binding energies on the one hand, and pronounced ultranonlocality and accurate band gaps on the other, within the meta-GGA form. It also was an open question what kind of results would be achieved with the TASK construction for other observables such as bond lengths and reaction barrier heights. These questions were addressed in this work.

We showed that TASK exchange with LDA correlation is accurate for many bond lengths, and we identified alkali metals as difficult cases that will need further work. TASK exchange in combination with different correlation functionals is accurate for reaction barrier heights. One can interpret this as a natural consequence of the exact-exchange-like nonlocality that is guaranteed by the construction principles that were used for the TASK exchange.

We further showed that two correlation functionals of very transparent form and without empirical parameters lead to a considerably improved accuracy for atomization energies. Furthermore, TASK exchange with CC correlation is attractive from a computational perspective because the calculations converge well, in contrast to some other meta-GGAs. It is not as accurate for atomization energies as other functionals, e.g., the SCAN meta-GGA, yet its remarkable accuracy for band gaps in combination with the reasonable description of electronic binding makes it ideal for material science problems, e.g., when one wants to screen the band gap of many compounds from first principles, and thus needs computational efficiency together with qualitative nonempirical reliability. The present paper presents an important step forward by demonstrating that pronounced ultranonlocality and reasonable electronic binding are not mutually exclusive and can well be achieved within the meta-GGA form. Therefore, there is hope that further improved meta-GGAs with pronounced ultranonlocality can be developed in future work.

ACKNOWLEDGMENTS

We acknowledge discussions with F. Tran about the bond lengths obtained with TASK+LDAc. We appreciate financial support from the Deutsche Forschungsgemeinschaft

(KU 1410/4-1), we are grateful for support from the Elite Study Program Biological Physics of the Elite Network of Bavaria, and we appreciate support from the Bavarian Polymer Institute in terms of computing resources. The Open Access publication has partially been funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) - 491183248, and the Open Access Publishing Fund of the University of Bayreuth.

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