Ultranonlocality and accurate band gaps from a meta-generalized gradient approximation

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The proper description of step structures in the exchange correlation potential, of charge localization, and a reasonable prediction of band gaps have been long-standing, serious challenges for semilocal density functionals. In practice, obtaining all of these properties from the functional derivative of an energy functional was possible only at the price of incorporating exact exchange. We here show that they can be achieved at significantly lower, semilocal computational expense by using kinetic-energy density-dependent functionals. The key to obtaining these features is a functional construction strategy that focuses on the derivative discontinuity and the density response.

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I. INTRODUCTION

The success of density functional theory (DFT) is based on its favorable ratio of accuracy to computational cost. Semilocal functionals such as generalized gradient approximations (GGAs) [1–5] are to this day popular in solid-state physics and material science because of their moderate computational cost. They also continue to play a role, especially for large-scale applications, in the molecular sciences, where hybrid functionals [6] have become the standard functionals for systems of small to moderate size. However, DFT based on semilocal functionals has also faced notorious problems, with prime examples known since the early days of DFT. In solid-state physics, the prediction of the fundamental (band) gap is an intrinsic limitation of explicit density-dependent semilocal functionals [7,8]. In material science, the proper description of charge localization and charge distributions between subsystems is a major challenge [9,10] that affects many problems, e.g., surface adsorption [11,12]. In molecular physics and chemistry, missing field-counteracting terms have serious, detrimental consequences for the calculation of polarizabilities, hyperpolarizabilities, and charge transfer [13–15]. These problems originate from several fundamental shortcomings that are closely interconnected: a missing derivative discontinuity [16–18], a lack of step structures in the exchange correlation (xc) potential [19–22], and the delocalization error [23,24]. Density-driven errors [25,26] characterize many of these problems.

The standard approach to overcome the limitations is turning to functionals that are nonlocal. Self-interaction corrections can restore nonlocality [27–31], but the dominant

and highly successful strategy of incorporating nonlocality has been the use of exact (Fock) exchange. By exploiting the adiabatic connection, yet higher accuracy can be reached [32], with DFT starting to rival wave-function-based methods in accuracy. However, the accuracy comes at a steep increase in computational cost. A standard hybrid functional is computationally already much more demanding than a GGA. Therefore, calculations incorporating Fock exchange are restricted to much smaller systems than semilocal calculations. This restriction can have serious consequences because often the interesting features of real-world systems, especially in the realm of nanomolecular and supramolecular science, stem from an intrinsic complexity that requires the explicit treatment of a large number of particles. Understanding light-harvesting systems [33–35] is a paradigm example: it requires calculating energy and charge transfer through arrays of dozens of chromophores, where each chromophore typically has hundreds of electrons. The chromophores in turn are typically embedded in a protein matrix, at least parts of which should also be taken into account explicitly [33,35–37]. However, semilocal functionals are currently at their limits for such systems due to their inability to properly describe charge transfer [38–40]. Therefore, there is a serious need for functionals that do not suffer from the large, qualitative errors that traditional semilocal functionals plague, yet come at a comparable computational price.

Here we demonstrate that one can achieve this with functionals that depend on the noninteracting kinetic energy density

$$\tau(\mathbf{r}) = \frac{\hbar^2}{2m} \sum_{i=1}^N |\nabla \varphi_i(\mathbf{r})|^2. \quad (1)$$

These so-called meta-GGAs are well established and their development has become an impressive success story [41–56]. They are the natural candidates for curing the above-mentioned deficiencies because the explicit use of the (occupied) orbitals creates nonlocality since each $\varphi_i(\mathbf{r})$ depends on the density $n(\mathbf{r}')$ at all points $\mathbf{r}'$. Yet, their computational cost is manifestly semilocal. In the following, we show how

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meta-GGA nonlocality can be led to unfold by focusing on the potential response and derivative discontinuity features.

Focusing the xc functional development less on the energy and also on the density or potential has already been advocated in principle [57–59] in the past, but in practice relied on expensive functionals [58,60,61]. The development of model potentials [62–66] has demonstrated that beneficial properties can be achieved semilocally. However, model potentials lack a corresponding energy functional and are not functional derivatives [63,67–69]. Furthermore, many of these constructions, as well as the attempt to restore the functional derivative property within the GGA form [70], lead to divergences [71–73]. Moreover, such purely multiplicative potentials are formally inappropriate to yield the fundamental gap [74].

The meta-GGA form allows to bring these earlier approaches to a unifying success. To this end, we write the exchange energy in the form

$$E_x^{\text{mGGA}}[n] = A_x \int n^{4/3} F_x(s, \alpha) d^3r, \quad (2)$$

where as usual $A_x = -(3e^2/4)(3/\pi)^{1/3}$, thereby satisfying the correct uniform coordinate density-scaling behavior [75]. Here the enhancement factor $F_x(s, \alpha)$ is parametrized by the dimensionless variables

$$s = \frac{|\nabla n|}{2(3\pi^2)^{1/3} n^{4/3}} \quad (3)$$

and [51–54,76,77]

$$\alpha = (\tau - \tau^W)/\tau^{\text{unif}}, \quad (4)$$

where the von Weizsäcker kinetic energy density

$$\tau^W = \frac{\hbar^2}{8m} \frac{|\nabla n|^2}{n} \quad (5)$$

is the single-orbital limit of τ and $\tau^{\text{unif}} = A_s n^{5/3}$ with $A_s = (3\hbar^2/10m)(3\pi^2)^{2/3}$ its uniform-density limit. Spin polarization can be accounted for via the spin-scaling relation for exchange [78].

II. ORIGIN OF ULTRANONLOCALITY

In order to demonstrate that crucial (ultra)nonlocal features of exact exchange (EXX) can be captured by a meta-GGA, we recall that the EXX Kohn-Sham (KS) potential for a spin-saturated N -electron system

$$\begin{aligned} v_x^{\text{EXX}}(\mathbf{r}) = & e^2 \int \frac{n_x^{\text{EXX}}(\mathbf{r}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d^3r' \\ & + \frac{1}{n(\mathbf{r})} \sum_{i=1}^{N/2} \left\{ |\varphi_i(\mathbf{r})|^2 (\bar{v}_{xi}^{\text{EXX}} - \bar{v}_{xi}^{\text{EXX}}) \right. \\ & \left. - \frac{\hbar^2}{m} \nabla \cdot [\psi_i^*(\mathbf{r}) \nabla \varphi_i(\mathbf{r})] \right\} + \text{c.c.} \end{aligned} \quad (6)$$

can be decomposed [79] into the smooth Coulomb potential of the EXX hole n_x^{EXX} (first term) and the response potential (second term). Here, a short overscore with index i denotes

taking the expectation value with the i th orbital. Furthermore,

$$u_{xi}^{\text{EXX}}(\mathbf{r}) = \frac{1}{\varphi_i^*(\mathbf{r})} \frac{\delta E_x^{\text{EXX}}[n]}{\delta \varphi_i(\mathbf{r})} \quad (7)$$

is the associated orbital-specific potential and $\psi_i(\mathbf{r})$ denotes the usual orbital shift of optimized effective potential (OEP) theory [80,81]. Hallmark nonlocal features of EXX result from the response potential [13,15,79,82,83], in particular from the orbital-average terms on the right of Eq. (6). Note that the KS potential retains the very same mathematical structure [84] when the EXX energy is replaced by another orbital-dependent energy functional, e.g., a meta-GGA. The only difference is that EXX quantities are replaced by the corresponding meta-GGA quantities, e.g.,

$$u_{xi}^{\text{mGGA}}(\mathbf{r}) = \frac{1}{\varphi_i^*(\mathbf{r})} \frac{\delta E_x^{\text{mGGA}}[n]}{\delta \varphi_i(\mathbf{r})}. \quad (8)$$

Thus, it is clear that, in principle, a meta-GGA can incorporate ultranlocal features because the ultranlocality of the potential stems from the orbital-average terms. For EXX, this nonlocality is enhanced by the Fock integrals. As meta-GGAs lack such integrals, one has to take special care to embed the nonlocality in the $\bar{v}_{xi}^{\text{mGGA}} - \bar{u}_{xi}^{\text{mGGA}}$ terms with proper magnitude and sign.

Guidance on how this can be achieved can be found by recalling that the ultranlocality in the KS potential is closely connected [15,85] to the derivative discontinuity

$$\Delta_x = v_x(\mathbf{r})|_+ - v_x(\mathbf{r})|_- = \frac{\delta E_x[n]}{\delta n(\mathbf{r})} \Big|_+ - \frac{\delta E_x[n]}{\delta n(\mathbf{r})} \Big|_-, \quad (9)$$

where the positive and negative signs denote evaluation of the functional derivative approaching the integer particle number from above and below, respectively. All experience to date is in line with $\Delta_x \geq 0$ [7,86,87]. When evaluating the functional derivative of a meta-GGA,

$$\begin{aligned} \frac{\delta E_x^{\text{mGGA}}[n]}{\delta n(\mathbf{r})} = & \frac{\delta}{\delta n(\mathbf{r})} \int e_x(n, \nabla n, \tau) d^3r' \\ = & \frac{\partial e_x}{\partial n}(\mathbf{r}) - \nabla \cdot \left[\frac{\partial e_x}{\partial \nabla n}(\mathbf{r}) \right] \\ & + \int \frac{\partial e_x}{\partial \tau}(\mathbf{r}') \frac{\delta \tau(\mathbf{r}')}{\delta n(\mathbf{r})} d^3r', \end{aligned} \quad (10)$$

only the τ dependence contributes to the derivative discontinuity as the first two terms correspond to the ones found in a usual GGA potential and originate from the explicit density dependence [85]. Therefore, and since $n(\mathbf{r}) = n(\mathbf{r})|_- = n(\mathbf{r})|_+$ as well as $\tau(\mathbf{r}) = \tau(\mathbf{r})|_- = \tau(\mathbf{r})|_+$,

$$\Delta_x^{\text{mGGA}} = \int \frac{\partial e_x}{\partial \tau}(\mathbf{r}') \left[\frac{\delta \tau(\mathbf{r}')}{\delta n(\mathbf{r})} \Big|_+ - \frac{\delta \tau(\mathbf{r}')}{\delta n(\mathbf{r})} \Big|_- \right] d^3r'. \quad (11)$$

In order to simplify this into a transparent expression, we assume (similar to previous work [88]) that the first factor can be approximated by its average over the energetically relevant region of space. Thus,

$$\Delta_x^{\text{mGGA}} \approx \overline{\frac{\partial e_x}{\partial \tau}} \left[\frac{\delta T_s[n]}{\delta n(\mathbf{r})} \Big|_+ - \frac{\delta T_s[n]}{\delta n(\mathbf{r})} \Big|_- \right] = \overline{\frac{\partial e_x}{\partial \tau}} \Delta_s, \quad (12)$$

where $T_s[n]$ is the usual noninteracting kinetic energy functional and Δ_s the corresponding non-negative KS gap. Equation (12) thus establishes a link between the magnitude and sign of $\partial e_x/\partial\tau$ and Δ_x^{mGGA} .¹

A meta-GGA with a sizable discontinuity of proper sign can therefore be constructed by requesting

$$\partial e_x/\partial\tau > 0, \quad (13)$$

as this ensures

$$\overline{\partial e_x/\partial\tau} > 0 \quad (14)$$

for any system. For a parametrization in s and α [cf. Eq. (2)],

$$\frac{\partial e_x}{\partial\tau} = \frac{A_x}{A_s} \frac{1}{n^{1/3}} \frac{\partial F_x}{\partial\alpha}. \quad (15)$$

With A_s and n positive and $A_x < 0$, a positive Δ_x^{mGGA} follows from

$$\partial F_x/\partial\alpha < 0, \quad (16)$$

and a larger negative slope in the energetically relevant regions of space can be expected to lead to a larger Δ_x^{mGGA} . We further note that the sign in Eq. (16) goes well along with the conjectured strongly tightened bound

$$F_x(s, \alpha) \leq F_x(s, \alpha = 0) \leq 1.174 \quad (17)$$

for all α [89]. It is also in line with the observations of ultranonlocality for the polarization dependence of solids [90] in the meta-GGA kernel [88] (see the detailed discussion in Appendix C). Furthermore, as α is a well-established measure of electron localization [51,91,92], Eq. (16) can be interpreted as a condition to energetically favor electron localization via the exchange energy, as EXX does.

III. PROOF OF CONCEPT

In order to demonstrate how these findings can be used in actual meta-GGA construction, we proceed in two steps. First, as a proof of concept (PoC) we discuss the simple, conceptual enhancement factor

$$F_x^{\text{PoC}}(\alpha) = c_H - c_1\alpha/(1 + c_2\alpha) \quad (18)$$

which uses only α and three parameters. This will not yield a generally useful functional, but allows to clearly demonstrate the connection between Eq. (16) and practically important manifestations of meta-GGA (ultra)nonlocality. We chose

$$c_H = \frac{40}{81} \left(\frac{4\pi^2}{3} \right)^{1/3} \quad (19)$$

to obtain the correct hydrogen atom energy (cf. Appendix B for a derivation). The parameters c_1 and c_2 determine the slope and the curvature of $F_x^{\text{PoC}}(\alpha)$ and will be discussed in the following.

¹Note that the above arguments do not exclude the possibility that $\Delta_x^{\text{mGGA}} \neq 0$ for $\Delta_s = 0$; the former is in principle possible when ones go beyond the averaging approximation of Eq. (12). Further note that the argument formulated here for Δ_x can readily be extended to Δ_{xc} .

FIG. 1. Difference between the $x(c)$ potentials (OEP) with and without a homogeneous external electric field for a chain of 12 hydrogen atoms at alternating distances of $2a_0$ and $3a_0$ (cf. Appendix D for computational details).

As a challenging, paradigm test of nonlocality we analyze the response potential of hydrogen chains [13,30,97–103]. Figure 1 shows the potential response obtained from LDA and EXX for a H_{12} chain in a static electric field, demonstrating the well-known field-counteracting term of EXX [13] and its absence in LDA. The striking observation in Fig. 1 is that F_x^{PoC} can achieve a potential response that is very close to the one of EXX with a suitable choice of parameters, namely, $c_1 = 27/40$ and $c_2 = c_1/(3 + c_H)$. Table I shows that the polarizabilities for hydrogen chains obtained with this F_x^{PoC} are close to the coupled-cluster (CCSDT) reference values for all chain lengths. Hence, this meta-GGA exhibits the correct trend with increasing chain length that so far has not been accessible with any semilocal functional due to the lack of the field-counteracting term. This is a proof of concept that even a simple meta-GGA can yield strong ultranonlocality.

Furthermore, this simple meta-GGA allows to transparently and explicitly demonstrate the relation between the derivative $\partial F_x/\partial\alpha$ and the field-counteracting term in the KS potential. By choosing different values for the parameters c_1 and c_2 , different slopes in α can easily be realized. For a first alternative (1st alt.) PoC meta-GGA we opt for the opposite slope at $\alpha = 0$ compared to the original (orig.) PoC

TABLE I. Polarizability calculated from the dipole moment in a_0^{-3} for hydrogen chains of different length and $x(c)$ energy functionals. CCSD(T) values from Ref. [94]. DFT calculations were performed self-consistently in a locally modified version of PARSEC [95,96], orbital-dependent functionals treated in full OEP unless noted otherwise (cf. Appendix D).

	H ₄	H ₆	H ₈	H ₁₂	H ₁₈
LDA (xc) [93]	37.6	72.8	115.0	211.4	369
PKZB (xc) [45]	35.5	65.8	105.1	189.1	323
τ -HCTH (xc) [46]	35.7	68.6	96.9	194.3	335
MVS (x) [54]	34.0	63.6	102.3	181.5	308
SCAN (xc) [52]	35.4	67.8	106.6	192.3	329
TASK (x)	34.0	62.9	96.2	169.8	286
PoC (x)	30.3	51.6	74.4	121.7	194
EXX (x)	32.1	56.5	83.0	138.7	225
CCSD(T)	28.8	50.5	74.1	123.6	200

FIG. 2. Explicit demonstration of the relation between $\partial F_x/\partial\alpha$ and the field-counteracting term in $v_x(\mathbf{r})$. Left: original and alternative PoC enhancement factors. Right: difference between the exchange response potentials (KLI) with and without a homogeneous external electric field of strength $\mathcal{F} = 5 \times 10^{-4} E_h/a_0$ for the H_{12} chain evaluated with the respective enhancement factors.

and use the Lieb-Oxford bound [104] as a natural limiting value for $\alpha \rightarrow \infty$ and thus as the upper bound. This leads to $c_1 = -27/40$ and $c_2 = -c_1/(1.804 - c_H)$. The left panel of Fig. 2 shows the resulting 1st alt. PoC enhancement factor as a function of α . It has a strictly positive slope, contrary to the strictly negative one of the orig. PoC meta-GGA. The right panel of Fig. 2 displays the difference between the exchange response potential [in Krieger-Li-Iafrate (KLI) approximation [82]] [cf. decomposition in hole and response potential as in Eq. (6), with and without a homogeneous external electric field for the H_{12} chain]. While the orig. PoC meta-GGA exhibits a pronounced field-counteracting behavior, the 1st alt. PoC meta-GGA also displays substantial ultranonlocality in the potential, but with the opposite sign and thus unphysically enhances the external field. This clearly demonstrates the link between $\partial F_x/\partial\alpha < 0$ and the field-counteracting term in the KS potential. In similar fashion one can transparently alter the slope $\partial F_x/\partial\alpha$ in the PoC meta-GGA form to show that the amplitude of $\partial F_x/\partial\alpha$ is directly correlated to the amplitude of the field-counteracting term in the KS potential. To this end, we construct a second alternative (2nd alt.) PoC meta-GGA with $c_1 = c_H(c_H - 1)$ and $c_2 = c_H - 1$, where these parameters are determined by the homogeneous electron gas limit, $F_x(\alpha = 1) = 1$, and by the negativity of exchange energy density, i.e., $\lim_{\alpha \rightarrow \infty} F_x(\alpha) = 0$. Figure 2 shows that the restricted slope of $F_x(\alpha)$ of this 2nd alt. PoC meta-GGA also limits the amplitude of the field-counteracting term in the KS potential.

Another stringent test of discontinuity features that probes the delocalization problem is defined by the H_8 - H_8 system of Ref. [9]. To the best of our knowledge, no semilocal functional to date has passed this test and even regular and local hybrids fail it [9,105]. Two aligned, but well-separated hydrogen chains are exposed to a constant external electric field of increasing strength in a series of ground-state calculations. As the field strength increases, charge eventually transfers from one chain to the other. Due to their large separation of 8 Å, integer preference [106] must be preserved, i.e., charge transfer should only occur via integer electron jumps at specific field strengths. However, as shown in the upper panel of Fig. 3, the local density approximation (LDA) leads to a gradual transfer of fractional charges, and usual semilocal

FIG. 3. Top: integrated (unsigned) charge on the H_8 acceptor as a function of the external field for LDA, as well as the PoC and TASK meta-GGAs. Bottom: plots of the exchange potential revealing the step structure in the PoC meta-GGA, by which the principle of integer preference is enforced. See Appendix E for computational details.

functionals and hybrids show similar behavior [9]. The upper panel of Fig. 3 shows that F_x^{PoC} (with the same choice of parameters that lead to good hydrogen chain polarizabilities) also correctly restores integer preference. If a step function would be used as an external field and the chains would be infinitely far apart, the critical step size associated with an integer electron transfer would be given directly [17] by the fundamental gap

$$I - A = \Delta_s + \Delta_{xc} \quad (20)$$

of a single H_8 chain. The situation considered here is more complex due to the finite distance as well as the linear field that induces a considerable polarization of both chains. Nevertheless, the Δ_{xc} contribution remains pivotal for a correct description of this charge transfer within KS theory. It manifests itself in the KS potential [21] as depicted in the lower panel of Fig. 3, which demonstrates that F_x^{PoC} leads to the right properties for the right reason: At $\mathcal{F} = 3.6 \times 10^9$ eV/m, which would lead to a fractional charge transfer for any conventional semilocal functional, a pronounced step structure that counteracts the charge transfer is observed. Similarly, after the jump a potential step with opposite sign stabilizes the charge transfer. This is exactly the behavior that EXX exhibits and by which a nonlocal self-interaction correction cures the charge-transfer problem [21].

IV. TASK: A GENERAL PURPOSE META-GGA

After this proof of concept as to how nonlocality can be achieved in meta-GGA construction, we take a second step and devise a more generally useful meta-GGA for exchange that features nonlocality. The outer framework of this functional, named TASK for the initials of the authors, is adopted

from the exchange part of the SCAN meta-GGA [52] and reads as

$$F_x^{\text{TASK}}(s, \alpha) = h_x^0 g_x(s) + [1 - f_x(\alpha)] [h_x^1(s) - h_x^0] [g_x(s)]^d. \quad (21)$$

We likewise choose

$$F_x \leq h_x^0 = 1.174 \quad (22)$$

as an upper bound, thereby obeying the rigorous strongly tightened bound [89] for $\alpha = 0$, as well as the more strict conjectured bound given by Eq. (17). By implying $f_x(\alpha = 0) = 1$ this meta-GGA is identical to SCAN for one-orbital systems ($\alpha = 0$), with

$$g_x(s) = 1 - \exp(-cs^{-1/2}), \quad (23)$$

where $c = 4.9479$ was obtained to recover the exact hydrogen atom energy. Moreover, $g_x(s)$ governs $F_x(s, \alpha)$ for $s \rightarrow \infty$ and vanishes like $s^{-1/2}$ [89], making the exchange energy per particle scale correctly to a negative constant under nonuniform coordinate scaling to the true two-dimensional limit [107,108].

We chose $h_x^1(s)$ to only depend on s . In this way, we can readily satisfy the construction principle of a negative slope in α of sizable magnitude [cf. Eq. (16)] to preserve the desired field-counteracting properties. Furthermore, we determine $f_x(\alpha)$ and $h_x^1(s)$ by recovering the fourth-order gradient expansion (GE4) for exchange [109], valid for slowly varying densities with small s and $\alpha \approx 1$. We do this in a way, however, that deviates decisively from SCAN, where the GE4 is solely recovered by h_x^1 and without a leading-order contribution of α , as $f_x(\alpha)$ is chosen to be flat, i.e., to vanish in any power of ∇n . Our aim, however, is to fulfill Eq. (16) with an appreciable slope, and therefore we require nonvanishing GE4 contributions of $f_x(\alpha)$ and thus α to all relevant orders. To this end, we start from the general expression for the gradient expansion around $s = 0$ and $\alpha = 1$,

$$F_x^{\text{GE4}}(s, \alpha) \sim 1 + \mu_s s^2 + \mu_\alpha (\alpha - 1) + C_s s^4 + C_{s\alpha} s^2 (\alpha - 1) + C_\alpha (\alpha - 1)^2 + \mathcal{O}(\nabla^6), \quad (24)$$

and then make use of the GE4 of τ [110,111] as detailed in Appendix A. Comparison with the usual GE4 for exchange then leads to the coefficients

$$\mu_s = \frac{10 + 60\mu_\alpha}{81}, \quad C_s = -\frac{1606 - 50\mu_\alpha}{18225}, \quad (25)$$

$$C_{s\alpha} = -\frac{511 - 50\mu_\alpha}{13500}, \quad C_\alpha = \frac{73 - 50\mu_\alpha}{5000} \quad (26)$$

and one degree of freedom, the choice of μ_α . However, due to the separation of variables with respect to s and α in our ansatz for the enhancement factor of Eq. (21), the mixed fourth-order term in the GE4 of Eq. (24) is generated by multiplication and is thereby not independent of the second-order terms. Therefore, in our construction, the corresponding mixed coefficient $C_{s\alpha}$ is directly linked to the second-order coefficients via

$$\mu_s \mu_\alpha = (1 - h_x^0) C_{s\alpha}, \quad (27)$$

which implies two possible values for μ_α :

$$\mu_\alpha^\pm = -\frac{97 + 3h_x^0 \pm \sqrt{9(h_x^0)^2 + 74166h_x^0 - 64175}}{1200}. \quad (28)$$

To obtain a sizable derivative discontinuity we choose the more negative solution

$$\mu_\alpha = \mu_\alpha^+ \approx -0.209897, \quad (29)$$

and, thus, a pronounced negative slope in α in accordance with Eq. (16).

To satisfy the listed constraints while obtaining a smooth $F_x(s, \alpha)$, we choose the following ansatz:

$$h_x^1(s) = \sum_{v=0}^2 a_v R_v(s^2), \quad f_x(\alpha) = \sum_{v=0}^4 b_v R_v(\alpha), \quad (30)$$

which is based on Chebyshev rational functions [112,113] $R_v(x)$ of degree v . The eight coefficients are determined by

$$h_x^1(0) = 1, \quad f_x(0) = 1, \quad f_x(1) = 0 \quad (31)$$

as well as the values of

$$\left. \frac{\partial^2 h_x^1}{\partial s^2} \right|_{s=0}, \quad \left. \frac{\partial^4 h_x^1}{\partial s^4} \right|_{s=0}, \quad \left. \frac{\partial f_x}{\partial \alpha} \right|_{\alpha=1}, \quad \left. \frac{\partial^2 f_x}{\partial \alpha^2} \right|_{\alpha=1} \quad (32)$$

that follow from imposing the GE4 as given by Eq. (24). The final condition to determine the coefficients can be formulated as choosing the limiting value of $f_x(\alpha)$ as $\alpha \rightarrow \infty$. In order to ensure $\partial F_x / \partial \alpha < 0$ everywhere as well as negativity of the exchange energy density, $f_x(\alpha \rightarrow \infty)$ has to be limited to the interval $]-4.4, -2.5[$ approximately. We here choose

$$f_x(\alpha \rightarrow \infty) = -3 \quad (33)$$

as a good balance between a pronounced negative slope for small values of α and minimizing undesired curvature in α . We find $a_0 = 0.938719$, $a_1 = -0.076371$, $a_2 = -0.0150899$, $b_0 = -0.628591$, $b_1 = -2.10315$, $b_2 = -0.5$, $b_3 = 0.103153$, $b_4 = 0.128591$.

Remarkably, the central premise of Eq. (16) follows naturally from the ansatz (21) when one opts for recovering the GE4 with a nonvanishing leading-order α contribution. Finally, we choose $d = 10$ to bundle $F(s, \alpha)$ quickly for $s \rightarrow \infty$, thereby restricting numerical issues that may arise as $\mathbf{r} \rightarrow \infty$ in the asymptotic region of finite systems, as well as to enforce negativity of the exchange energy density. In summary, our TASK functional fulfills all of the exact constraints for exchange that were imposed on SCAN (yet only the hydrogen atom norm), and in addition achieves a more pronounced negative slope in Eq. (16) that is crucial for field-counteracting properties. The resulting enhancement factor of TASK is depicted in Fig. 4. The constraints force the TASK functional toward a field-counteracting term that is somewhat less pronounced than the one of F_x^{PoC} and EXX, as seen in Fig. 1. However, as seen in Table I the polarizabilities are still substantially improved compared to LDA and SCAN. Also, the upper panel of Fig. 3 depicts a clear improvement over LDA for the H₈-H₈ charge-transfer system.

The potential that we obtain is relatively smooth and we did not experience difficulties in the numerical representation. By fulfilling Eq. (16), one guarantees that as functions of

FIG. 4. TASK exchange enhancement factor $F_x^{\text{TASK}}(s, \alpha)$ as a function of s for fixed values of α .

s curves for different α do not cross, a condition that has been associated with numerical stability [52]. Several meta-GGAs that have previously been constructed have numerical features that require special attention in their computational evaluation, and the underlying reasons have been studied with a new iso-orbital indicator [114].

We here deliberately focused on the exchange functional because exchange is decisive for curing charge-transfer errors. A recent study [118] furthermore demonstrated that correlation contributes to the field-counteracting term in a similar fashion as exchange. Therefore, it is not unreasonable to cover the field-counteracting terms by meta-GGA exchange. Ground-state energetics, on the other hand, usually benefit from a correlation functional which is especially tailored toward the corresponding semilocal exchange. However, constructing such a correlation functional is a task in itself, to be addressed in future work. Therefore, we presently combine the TASK exchange with PW92-LDA correlation [93], as LDA is universal and not tied to a specific exchange functional.

We calculated atomization energies for a set of diatomic molecules. The set is small, but comprises single, double, and triple bonds and thus gives a first impression of binding properties. The mean absolute error for the atomization energy are reported in Table II. Without further context, TASK+PW92 accuracy for atomization energies is not impressive. However, one can here see a parallel between meta-GGAs and hybrid functionals: Hybrids with a small (ca. 20%) fraction of Fock exchange are good for atomization energies. A reliable description of charge transfer and charge localization, however, requires much larger (up to 75%) fractions [125] and correspondingly leads to a considerably poorer description of atomization energies [126]. Therefore, we see it as a sign of hope that TASK+PW92 atomization energies are at least significantly better than LDA while at the same time successfully tackling the charge-transfer errors. Furthermore, as the TASK functional is constructed in purely nonempirical fashion, it can be expected to be reliable over a wide range of systems and situations.

TABLE II. Atomization energies in kcal/mol of diatomic molecules based on self-consistent KS calculations at experimental bond lengths and mean absolute error (MAE) across the test set. The calculations were performed on a real-space prolate spheroidal grid with DARSEC [115], an all-electron code for single atoms or diatomic molecules. SCAN, EXX, and TASK (here in combination with PW92-LDA correlation) potentials were evaluated in the KLI approximation [82]. The experimental values (with zero point vibration removed) and the experimental bond lengths are taken from Ref. [116] (cf. the Supplemental Material of Ref. [117] for computational details).

Molecule	EXX	LDA	PBE	SCAN	TASK	Expt.
H ₂	83.8	112.9	104.6	107.7	117.0	109.5
LiH	34.1	60.8	53.5	55.6	58.9	58.0
Li ₂	3.6	23.8	19.9	18.1	11.6	24.7
LiF	90.5	156.3	139.0	138.1	130.2	138.3
CO	172.3	299.2	269.1	255.2	248.2	259.5
N ₂	112.4	268.0	243.9	220.9	174.9	228.5
NO	47.6	199.4	170.9	146.5	122.9	152.5
OH	65.4	123.2	105.0	94.9	97.6	106.4
O ₂	30.0	175.1	144.1	126.8	131.0	120.5
FH	96.6	162.0	142.0	140.4	139.2	141.1
F ₂	-41.7	78.0	52.8	37.4	23.4	38.4
MAE	62.1	25.7	9.1	4.6	14.6	

Further evidence that the TASK functional indeed incorporates correct physics and yields a sizable Δ_x^{mGGA} is seen when calculating band gaps. For the study of gaps, we resorted to general KS (gKS) theory [127], as it gives direct access to the

FIG. 5. Experimental band gaps compared to band gaps computed by PBE [4], SCAN (gKS) [52], and TASK+PW92 (gKS). See Table III for values and further details.

TABLE III. Structure, experimental geometrical parameters, and fundamental band gaps of the solids considered in this work. All band gaps are obtained from band structures that are sampled with a step size of $0.02a_0^{-1}$.

Solid	Structure	Lattice parameter (Å)	Fundamental band gaps (eV)			
			PBE	SCAN	TASK+PW92	Expt.
Ge	Diamond	5.652 [119]	0.03	0.19	0.85	0.74 [120]
CdO	Rock salt	4.704 [121]	0.00	0.05	0.89	0.84 [122]
Si	Diamond	5.430 [119]	0.68	0.93	1.12	1.17 [120]
GaAs	Zinc blende	5.648 [119]	0.55	0.83	1.72	1.52 [120]
SiC	Zinc blende	4.358 [119]	1.43	1.75	2.06	2.42 [120]
ZnS	Zinc blende	5.409 [120]	2.10	2.63	3.84	3.66 [120]
MgS	Zinc blende	5.622 [120]	3.55	4.25	5.74	5.4 [120]
C	Diamond	3.567 [119]	4.21	4.60	4.36	5.48 [120]
BN	Zinc blende	3.603 [120]	4.54	5.03	5.50	6.22 [120]
MgO	Rock salt	4.207 [119]	4.78	5.68	7.33	7.22 [120]
LiCl	Rock salt	5.106 [119]	6.37	7.32	9.55	9.4 [123]
Kr	fcc	6.130 [123]	7.17	8.01	11.45	11.6 [123]
LiF	Rock salt	4.010 [123]	9.17	10.14	12.93	13.6 [124]
Ar	fcc	5.310 [123]	8.65	9.52	13.28	14.3 [124]

fundamental gap [74,124,128,129], i.e.,

$$\Delta_s^{\text{gKS}} \approx \Delta_s + \Delta_{\text{xc}}. \quad (34)$$

All calculations were performed at the experimental geometry with a modified version of the periodic all-electron code BAND [130], a $9 \times 9 \times 9$ Monkhorst-Pack $\mathbf{k}$ grid [131], and the TZ2P [132] basis set (except for Ar and Kr, for which QZ4P is used). The scalar relativistic effects are included by the ZORAMethod [133]. Figure 5 shows that gaps calculated with TASK in combination with PW92-LDA correlation are uniformly and significantly improved compared to usual semilocal results, here represented by PBE [4], for systems ranging from traditional semiconductors to wide-gap insulators (see Table III for detailed numbers). For most systems studied, TASK also performs significantly better than SCAN, which was reported [124] to already yield considerably more

realistic band gaps. It is noticeable that TASK band gaps are remarkably close to experiment for a wide range of materials, reaching from the semiconductors Ge, CdO, Si, and GaAs to minerals such as MgO and LiCl. The functional also gives significantly improved values for the large-gap insulators Kr, LiF, and Ar. Most notably, TASK opens the band gap with a magnitude close to the experimental value for Ge and CdO, whereas PBE and SCAN incorrectly predict both systems to be metallic (see Figs. 6 and 7 for the corresponding band structures).

V. SUMMARY AND CONCLUSIONS

In summary, we laid out an exchange-correlation functional construction strategy that takes into account properties of the KS potential such as the derivative discontinuity and

FIG. 6. Band structure of Ge calculated with PBE [4] (blue), SCAN [52] (green), and TASK+ PW92 (red) at the experimental geometry (meta-GGAs calculated in gKS scheme). The top of the valance band is the zero of the energy scale. The experimental fundamental band gap is visually indicated by the hatched region.

FIG. 7. Band structure of CdO calculated with PBE [4] (blue), SCAN [52] (green), and TASK+ PW92 (red) at the experimental geometry (meta-GGAs calculated in gKS scheme). The top of the valance band is the zero of the energy scale. The experimental fundamental band gap is visually indicated by the hatched region.

the density response in addition to the ground-state energy. We derived a transparent general criterion for obtaining a sizable derivative discontinuity from a meta-GGA. In a proof-of-concept construction we demonstrated that this criterion directly governs the response potential and the ultranonlocality of a meta-GGA. Taking into account known exact constraints, we further developed a nonempirical, general-purpose meta-GGA for exchange that yields improved response properties and band gaps. This suggests that the ground-state energy need not be the only focus of meta-GGA construction, but construction strategies can also target the density and the potential. Our work shows that meta-GGAs can live up to the orbital functional promise of yielding ultranonlocal features without requiring costly exchange integrals. The meta-GGA form thus continues to hold great promise for future functional development.

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APPENDIX A: FOURTH-ORDER GRADIENT EXPANSION FOR EXCHANGE IN s AND α

We here describe in detail how $F_x(s, \alpha)$ is able to recover the GE4 for exchange [47,109] with one degree of freedom. Typically, the GE4 is written in s^2 and

$$q = \frac{\nabla^2 n}{4(3\pi^2)^{2/3} n^{5/3}} \quad (\text{A1})$$

as

$$F_x^{\text{GE4}}(s, q) \sim 1 + \frac{10}{81}s^2 + \frac{146}{2025}q^2 - \frac{73}{405}s^2q + 0 \cdot s^4 + \mathcal{O}(\nabla^6) \quad \text{as } s, q \rightarrow 0 \quad (\text{A2})$$

assuming a vanishing s^4 coefficient as the best numerical estimate. As $\alpha \rightarrow 1$ in the homogeneous electron gas limit, the GE4 for exchange in s and α has the general form of Eq. (24) with its five coefficients $\mu_s, \mu_\alpha, C_s, C_{s\alpha}$, and C_α to be determined as follows: First, $\alpha = \tau/\tau^{\text{unif}} - 5s^2/3$ is expanded by utilizing the GE4 of τ [110,111] to obtain

$$\alpha \sim 1 - \frac{40}{27}s^2 + \frac{20}{9}q + \frac{1}{81} \left[-14q^2 + \frac{140}{3}s^2q - 48s^2 + 12 \frac{\nabla^2(\nabla^2 n)}{2^4(3\pi^2)^{4/3}n^{7/3}} - 30 \frac{\nabla n \cdot \nabla(\nabla^2 n)}{2^4(3\pi^2)^{4/3}n^{10/3}} - 7 \frac{\nabla^2|\nabla n|^2}{2^4(3\pi^2)^{4/3}n^{10/3}} + \frac{92}{3} \frac{\nabla n \cdot \nabla|\nabla n|^2}{2^4(3\pi^2)^{4/3}n^{13/3}} \right] + \mathcal{O}(\nabla^6). \quad (\text{A3})$$

Inserting this expansion into Eq. (24) gives

$$F_x^{\text{GE4}}(s, \alpha) \sim 1 + \left(\mu_s - \frac{40}{27}\mu_\alpha \right) s^2 + \left(\frac{20}{9}\mu_\alpha \right) q + \left(-\frac{14}{81}\mu_\alpha + \frac{400}{81}C_\alpha \right) q^2 + \left(\frac{140}{243}\mu_\alpha + \frac{20}{9}C_{s\alpha} - \frac{1600}{243}C_\alpha \right) s^2q + \left(-\frac{48}{81}\mu_\alpha + C_s - \frac{40}{27}C_{s\alpha} + \frac{1600}{729}C_\alpha \right) s^4 + \mu_\alpha \left[\frac{4}{27} \frac{\nabla^2(\nabla^2 n)}{2^4(3\pi^2)^{4/3}n^{7/3}} - \frac{10}{27} \frac{\nabla n \cdot \nabla(\nabla^2 n)}{2^4(3\pi^2)^{4/3}n^{10/3}} - \frac{7}{81} \frac{\nabla^2|\nabla n|^2}{2^4(3\pi^2)^{4/3}n^{10/3}} + \frac{92}{243} \frac{\nabla n \cdot \nabla|\nabla n|^2}{2^4(3\pi^2)^{4/3}n^{13/3}} \right] + \mathcal{O}(\nabla^6). \quad (\text{A4})$$

Note that, under integration by parts,

$$\int n^{4/3} s^2 d^3r = 3 \int n^{4/3} q d^3r. \quad (\text{A5})$$

Therefore, and as we only consider $F_x^{\text{GE4}}(s, \alpha)$ under the exchange energy integral $E_x = A_x \int n^{4/3} F_x^{\text{GE4}}(s, \alpha) d^3r$, the separate occurrences of q in Eq. (A4) can be recast to $s^2/3$. Moreover, one can transform the remaining fourth-order terms to the variables s and q in similar fashion via integration by parts:

$$\int n^{4/3} \frac{\nabla^2(\nabla^2 n)}{2^4(3\pi^2)^{4/3}n^{7/3}} d^3r = \int n^{4/3} (2s^2q - q^2) d^3r, \quad (\text{A6a})$$

$$\int n^{4/3} \frac{\nabla n \cdot \nabla(\nabla^2 n)}{2^4(3\pi^2)^{4/3}n^{10/3}} d^3r = \int n^{4/3} (2s^2q - q^2) d^3r, \quad (\text{A6b})$$

$$\int n^{4/3} \frac{\nabla^2|\nabla n|^2}{2^4(3\pi^2)^{4/3}n^{10/3}} d^3r = \int n^{4/3} (-2s^2q + 6s^4) d^3r, \quad (\text{A6c})$$

$$\int n^{4/3} \frac{\nabla n \cdot \nabla|\nabla n|^2}{2^4(3\pi^2)^{4/3}n^{13/3}} d^3r = \int n^{4/3} (-s^2q + 3s^4) d^3r. \quad (\text{A6d})$$

This allows us to write Eq. (A4) as

$$F_x^{\text{GE4}}(s, \alpha) \sim 1 + \left(\mu_s - \frac{20}{27} \mu_\alpha \right) s^2 + \left(\frac{4}{81} \mu_\alpha + \frac{400}{81} C_\alpha \right) q^2 + \left(-\frac{2}{27} \mu_\alpha + \frac{20}{9} C_{s\alpha} - \frac{1600}{243} C_\alpha \right) s^2 q + \left(\frac{2}{81} \mu_\alpha + C_s - \frac{40}{27} C_{s\alpha} + \frac{1600}{729} C_\alpha \right) s^4 + \mathcal{O}(\nabla^6). \quad (\text{A7})$$

Finally, comparison with the GE4 of Eq. (24) yields the values for the coefficients already given by Eqs. (25) and (26) with one degree of freedom, the choice of μ_α . When choosing $\mu_\alpha = 0$ the resulting GE4 is identical to the one given in the Supplemental Material of Ref. [52].

APPENDIX B: HYDROGEN ATOM NORM

A meta-GGA for exchange defined by an enhancement factor $F_x(\alpha)$ that only depends on α reduces for any single-orbital system to a multiple of LDA,

$$E_x^{\text{mGGA}}[n] = A_x \int F_x(\alpha(\mathbf{r})) n^{4/3}(\mathbf{r}) d^3r = F_x(0) A_x \int n^{4/3}(\mathbf{r}) d^3r = F_x(0) E_x^{\text{LDA}}[n] \quad (\text{B1})$$

since $\alpha(\mathbf{r}) \equiv 0$ in the single-orbital limit. Given the completely spin-polarized exact hydrogen ground-state density

$$n_H(\mathbf{r}) = \frac{1}{\pi a_0^3} \exp(-2r/a_0), \quad (\text{B2})$$

the associated exchange energy of the meta-GGA is obtained by virtue of the spin-scaling relation for exchange [78] to be

$$E_x^{\text{mGGA}}[n_H, 0] = F_x(0) E_x^{\text{LDA}}[2n_H]/2 = -F_x(0) \frac{81}{128} \frac{e^2}{a_0} \left(\frac{3}{4\pi^2} \right)^{1/3}. \quad (\text{B3})$$

Consequently, the exchange energy of the meta-GGA matches the exact value at the hydrogen ground-state density

$$E_x[n_H, 0] = -E_H[n_H] = -\frac{e^2}{2} \iint \frac{n_H(\mathbf{r}) n_H(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d^3r d^3r' = -\frac{5e^2}{16a_0}, \quad (\text{B4})$$

if one chooses

$$F_x(0) = c_H := \frac{40}{81} \left(\frac{4\pi^2}{3} \right)^{1/3} \approx 1.16588 \quad (\text{B5})$$

[cf. Eq. (19)].

APPENDIX C: ULTRANONLOCALITY IN RECIPROCAL SPACE

Having explicitly demonstrated that and how a meta-GGA can achieve ultranonlocality associated with the derivative discontinuity in real space and for finite systems, a comparison with previous work on ultranonlocality in reciprocal space and extended systems is worthwhile. Nazarov and Vignale [88] showed via a reciprocal space analysis that the xc kernel f_{xc} of a meta-GGA is capable of long-rangedness, which also is an aspect of ultranonlocality. When thinking about f_{xc} ,

different aspects of ultranonlocality are associated [90,134] with a singularity of the type

$$f_{xc,00}(\mathbf{q}) \sim \frac{\beta}{q^2} \quad (\text{C1})$$

in the optical limit $\mathbf{q} \rightarrow 0$. Using an averaging approximation similar to the one that we use in Eq. (12) to demonstrate the relation between Δ_x and $\partial e_x / \partial \tau$, they extract the singularity to be

$$f_{xc,GG'}(\mathbf{q}) \approx -\frac{\partial e_{xc}}{\partial \tau} \chi_{s,GG'}^{-1}(\mathbf{q}), \quad (\text{C2})$$

where χ_s^{-1} is the inverse of the noninteracting KS response function. Using this expression as an approximation to the full kernel, they calculate the excitation spectra of semiconductors with a special focus on the excitonic peak. These spectra and comparison with the work of Reining *et al.* [134] then suggest $\beta < 0$, and hence $\partial e_{xc} / \partial \tau < 0$ in order to capture the electron-hole interaction. Their analysis thus leads to the opposite sign of $\partial e_{xc} / \partial \tau$ that we reach based on the functional derivative of Eq. (10) and the fact that Δ_x is determined by its last term.

To resolve this seemingly contradiction, we first note that Ref. [88] focuses on a different physical problem than we do. Nazarov and Vignale [88] study the binding of excitons. As argued in Ref. [134], this merely requires a static long-range contribution to the kernel,

$$\Delta f_{xc}(\mathbf{r}, \mathbf{r}') = \frac{\beta}{4\pi |\mathbf{r} - \mathbf{r}'|} \quad (\text{C3})$$

with $\beta < 0$. We further note that obtaining properties of f_{xc} and v_{xc} that are long range in real space correctly with semilocal approximations is only possible within narrow conditions and typically leads to divergences of the KS potential for many finite systems [71,72].

In our work here, we focus on the ultranonlocality that is associated with potential step structures and the derivative discontinuity. Our tests were done for finite systems, but Ghosez *et al.* [90] demonstrated that this type of ultranonlocality is also present in solids. When calculating the polarization dependence of solids, which is in close analogy to the ultranonlocality effects in the electrical response of molecular chains, this ultranonlocality is connected to a positive ($\beta > 0$) singularity of the form of Eq. (C1) (see, e.g., Fig. 1 of Ref. [90]). Within the approximation of Eq. (C2) this singularity can in turn be associated with $\partial e_{xc} / \partial \tau > 0$. Therefore, the analysis of Ghosez *et al.* [90] is in line with our arguments.

Finally, we note that also Ref. [134] finds a positive contribution to the singularity in f_{xc} that stems from the energy shift between KS and quasiparticle eigenvalues. Within KS

TABLE IV. Dimensions of ellipsoidal boundaries used for the hydrogen chain polarizability calculation in PARSEC in a_0 .

Semiaxis	H ₄	H ₆	H ₈	H ₁₂	H ₁₈
Parallel	15.0	17.5	20.0	25.0	32.5
Perp.	10.0	10.0	10.0	10.0	13.0

theory this energy shift is accounted for by the derivative discontinuity Δ_{xc} . In the calculations of Ref. [134], however, it is absorbed into an energy shift of the starting χ_s .

APPENDIX D: HYDROGEN CHAIN POLARIZABILITY CALCULATION DETAILS

The polarizabilities of hydrogen chains were calculated with a locally modified version of the publicly available electronic structure program PARSEC [95] on a real-space grid with a grid spacing of $0.27a_0$. The linear chains with alternating H–H distances of $2a_0$ and $3a_0$ are centered in an ellipsoid with dimensions as listed in Table IV. All calculations were performed self-consistently with a functional independent pseudopotential [135], an energy convergence criterion for the KS iterations of $5 \times 10^{-7} E_h$, and a field strength of $5 \times 10^{-5} E_h/a_0$. To obtain the KS potential based on the OEP scheme via S -Iterations [81], S was converged to at least $3 \times 10^{-7} E_h$. The implementation of all meta-GGAs with the exception of PoC, TASK, and SCAN is built on an interface to LIBXC version 4.3.4 [136]. We found that for several of the meta-GGAs that are available in LIBXC [136], e.g., TPSS [47], revTPSS [50], TM [55], and the Minnesota meta-GGAs [49], the grid-based calculations are difficult to converge as their potentials show rapidly varying features. This type of observation is in line with the arguments of, e.g., Ref. [114]. Therefore, the results reported here are restricted to numerically benevolent meta-GGAs. For the calculations with the MVS functional we found that the OEP calculations converged very slowly for the longer hydrogen chains.

Therefore, for the MVS functional [54], Table I reports OEP polarizabilities for H₄ and H₆, but the values reported for H₈, H₁₂, and H₁₈ are based on the KLI approximation [82]. All other values are OEP based.

APPENDIX E: H₈–H₈ CHARGE-TRANSFER CALCULATION DETAILS

All calculations for the H₈–H₈ charge-transfer system were performed spin polarized, self-consistently on a real-space grid with BT-DFT [40] using Troullier-Martins LDA pseudopotentials [137], ellipsoidal boundaries with semiaxis of $20a_0$ perpendicular to and $32a_0$ along the chains, and a grid spacing of $0.3a_0$. For the KS iterations, an energy convergence criterion of $5 \times 10^{-6} E_h$ was used. The two hydrogen chains, each containing eight hydrogen atoms separated by 1 Å, are centered in the ellipsoid with a mutual distance of 8 Å. Due to the nature of the situation with nearly degenerate ground-state solutions, the calculations are numerically very challenging (cf. Refs. [9,105]). The LDA calculations were performed with occupation numbers following from a Fermi-Dirac distribution with a temperature of 800 K, as zero-temperature calculations cannot be converged once the external field strength leads to (a fractional) transfer of charge. The meta-GGA calculations in the KLI approximation [82] were performed at 0 K. For the PoC meta-GGA, convergence was possible for any considered field strength. We can carefully converge two nearly degenerate ground states with an integer charge of 8 and 9 electrons, respectively, even for field strengths below and above the critical field strength of 3.7×10^9 V/m, at which an integer electron transfer occurs. Consequently, the associated Fig. 3 only shows the solution of lower total energy at each field strength and the critical field strength is given by the energy crossing point of both solutions. While the TASK functional does not allow for a numerical stable solution from field strengths of 3.0×10^9 V/m to 5.8×10^9 V/m, it still improves considerably over LDA as it allows to counteract the charge transfer up to 2.8×10^9 V/m and yields a broad integer electron plateau starting at 6.0×10^9 V/m.

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