Convergence of the regularized Kohn–Sham iteration in Banach spaces

Markus Penz
Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany
and
Basic Research Community for Physics, Leipzig, Germany

Andre Laestadius
Hylleraas Centre for Quantum Molecular Sciences, Department of Chemistry, University of Oslo, Norway

The Kohn–Sham iteration of generalized density-functional theory on Banach spaces with Moreau–Yosida regularized universal Lieb functional and an adaptive damping step is shown to converge to the correct ground-state density. This result demands state spaces for (quasi)densities and potentials that are uniformly convex with modulus of convexity of power type. The Moreau–Yosida regularization is adapted to match the geometry of the spaces and some convex analysis results are extended to this type of regularization. Possible connections between regularization and physical effects are pointed out as well.

I. INTRODUCTION

Density-functional methods are a major tool in electronic-structure calculations that underlie the whole field of quantum chemistry. Within these methods, a complex, interacting many-particle system with ground-state density as the basic state variable is compared against a simpler, non-interacting reference system that shares the same ground-state density. The internal differences between the systems are compensated by an auxiliary potential called the Kohn–Sham potential. This includes the exchange-correlation (xc) potential as the main, unknown part and acts as an external force on the reference system. From approximations to this potential the unknown density is then determined within the self-consistent Kohn–Sham (KS) iteration scheme (see Section III).

This work can be seen as part of a series [1–4] that aims at constructing a general and well-defined mathematical framework for the formulation of this KS iteration scheme and at proving convergence. Density-functional theory (DFT) has been previously formalized by Lieb [5] using tools from convex analysis in Banach spaces and we will strictly remain within this setting. Therein, the underlying state space of densities is formed by a Banach space $X$, with the potentials acting on them as elements of the dual space $X^\ast$. The pairing $\langle x^\ast, x \rangle$ then equals the potential energy of a given state $x \in X$ within a potential $x^\ast \in X^\ast$. That the ground-state problem of quantum mechanics, usually formulated as an eigenvalue problem in linear operator theory on Hilbert spaces, can be translated into a more general variational problem in terms of the total energy is customary. The replacement of the original interacting problem by one that contains no interactions but adds an auxiliary potential that already depends on the solution, makes the resulting equations non-linear. Employing tools from convex optimization thus seems suitable.

A few extra steps are required in order to translate the method of DFT into a convex optimization problem. The total energy is modeled as consisting of an internal part, given by the expectation value of the universal Hamiltonian $H_0$, and an external part from the linear pairing of potential and density. But the density functional $\tilde{F} : \tilde{X} \rightarrow \mathbb{R} \cup \{+\infty\}$ for the universal internal part, defined by a constrained search over all wave-functions yielding a given physical density from $\tilde{X} \subset X$, is neither convex [5, Theorem 3.4(i)] nor differentiable [6], revealing it as unsuitable for methods of convex analysis. Thus a first extension, performed also in Lieb [5], was to take the convex hull of $\tilde{F}$ to get a lower-semicontinuous, convex $F$ defined on all of $X$. While $X$ contains only physical densities, the quasidensities in $X$ can be non-normalized or negative. They received no further interpretation in Lieb [5] but moved to the center of interest in the next step, the Moreau–Yosida (MY) regularization of $F$, to generate a differentiable functional $F_\varepsilon$ [1]. This allows to finally set up a well-defined KS iteration scheme that has been generalized from the setting of Hilbert spaces on bounded domains in Kvaal et al. [1] to reflexive and strictly convex Banach spaces in Laestadius et al. [2]. The latter is not limited to the

---

a)Electronic mail: markus.penz@mpsd.mpg.de
standard formulation of DFT, where probability densities are linked to scalar electrostatic potentials, but was shown to be general enough for current-density-functional theory (CDFT), where additionally the probability-current density and a vector potential are taken into account. A host of other physical settings, even such that are outside the usual domain of quantum chemistry, can be included as well into this generalized KS iteration scheme on Banach spaces that was baptized MYKSODA. Already in Laestadius et al. 2017, the algorithm contained another important ingredient, namely the damping of the iteration-step length. This procedure is known from the optimal damping algorithm (ODA) and occurred prior to our treatment in Wagner et al. 2020, where it was used to guarantee a strictly descending sequence of energies in the KS iteration.

The final building block to date has been added in Penz et al. 2020 and presented a proof of convergence in the limited setting of finite lattice systems, $X = l^2(M), M \in \mathbb{N}$. The obvious next step and aim of this work is a generalization of this proof to infinite-dimensional Banach spaces, a non-trivial task because of the lacking Hilbert-space structure and the resulting lack of simple geometry of $X$. In order to complete the task, the MY regularization will be generalized to an infimal convolution with a function $\varepsilon_p$ on $X$ (Definition 17), called $p$MY regularization, where the case $p = 2$ corresponds to the standard definition. This does not change the usual properties of the regularization and carries over to a corresponding definition of the proximal mapping (Definition 18) that assumes an important role in relating the regularized and unregularized functionals by shifting the quasidensities in $X$. But in order to make full use of the properties of the $p$MY regularized functional $E_x$, mainly a strong monotonicity property of the differential of the associated energy functional $E_x$ in Lemma 20(iii), the involved spaces $X$ and $X^*$ have to exhibit a less common convexity constraint. This feature is called $p$-uniform convexity or uniform convexity (with the modulus of convexity) of power type $p$, termed here simply $p$-convexity (Definition 2). It is stronger than the usual uniform convexity of spaces but featured, for example, by $L^p, 1 < p < \infty$. Apart from our main result (Theorem 23) that provides a full proof of convergence of the KS iteration within the described setting, this work also contains some probably new results relating to $(p)$MY regularized functionals.

Convergence of the KS iteration and similar self-consistent-field methods is known to be an issue, since the iteration can easily get stuck in, or oscillate between local minima due to the lack of convexity. Previous solutions to this problem include, e.g., the mentioned optimal damping and random perturbations of the erroneously converged density [12]. The present approach attacks the problem at its conceptual basis and delivers the mathematical structures needed for guaranteed convergence. It is our hope that such structures can also be translated to practical implementations for electronic structure calculations in order to achieve better and more reliable results.

The structure of the article is as follows. In Section II, we make all necessary preparations for our main results on a purely mathematical level. The introduction to $p$-convex spaces includes Lemma 14 that is directly derived from results in Xu 2016 and offers a norm-estimate as a replacement for the polarization identity. The part about $p$MY regularization includes some results from the domain of convex analysis tailored to $p$MY regularized functionals that might also be of interest in other areas. In particular, Lemma 20 establishes a strong equivalence between (i) $p$MY regularization, (ii) strong concavity of the conjugate functional, and (iii) a strong monotonicity property that is then linked to differentiability in Lemma 22. Section II presents the whole setting of regularized DFT and the idea behind the KS iteration. The respective energy functionals are introduced and the effect of regularization, that establishes differentiability of the density functional, is presented as a sufficient condition for a Hohenberg–Kohn theorem. The central result is presented in Section III where the KS iteration is formulated as a theorem and its convergence is proven. Section IV shows that the proof is applicable to density-functional theories in their reflexive Banach space formulation, including standard ground-state DFT and (paramagnetic) CDFT. Section V links the regularization effects to physical principles and even gives a physical interpretation for quasidensities.

II. CONVEXITY ANALYSIS PREREQUISITES

A. Convexity and Duality

Our basic setting is that of a real, reflexive Banach space $X$ with the special properties of $p$-convexity and $q$-smoothness given in Definition 3 below. It should be recalled that $X$ uniformly smooth means that its dual $X^*$ is uniformly convex. By the Milman–Pettis theorem, uniform convexity of $X^*$ already implies reflexivity. Typical uniformly convex spaces are the Lebesgue spaces $L^p, 1 < p < \infty$, yet not $L^1, L^\infty$ that are not even strictly convex, nor reflexive. Following this, $p$ will fulfill, if not otherwise stated, $1 < p < \infty$. Let $p^*$ always denote the Hölder conjugate of $p$, i.e., the number $1 < p^* < \infty$ that obeys $1/p + 1/p^* = 1$. For $x \in X, x^* \in X^*$,
the dual pairing \((x^*, x)\) simply means applying \(x^*\) on \(x\), \((x^*, x) = x^*(x)\). The norm of these spaces will usually be denoted as \(\| \cdot \|\), only in a few cases we will explicitly add information about the space, \(\| \cdot \|_X\), or write \(\| \cdot \|_2\) for the \(L^2\)-norm. The main instrument to identify various degrees of convexity of spaces is given by the \emph{modulus of convexity}.

**Definition 1.** The \emph{modulus of convexity} of a normed space \(X\) is defined as the function \(\delta_X : (0, 2] \to [0, 1]\),

\[
\delta_X(\varepsilon) = \inf \left\{ 1 - \frac{1}{2} \| x + y \| : \| x \| = \| y \| = 1, \| x - y \| = \varepsilon \right\}.
\]

On the dual side the \emph{modulus of smoothness} of a normed space \(X\) is the function \(\rho_X : (0, \infty) \to [0, \infty)\),

\[
\rho_X(\tau) = \inf \left\{ \frac{1}{2} \left( \| x + y \| - \| x - y \| \right) - 1 : \| x \| = 1, \| y \| = \tau \right\}.
\]

**Remark 1.** In Chidume [14, Sec. 1.4 and 2.2] the equivalence of various different definitions of \(\delta_X\) and \(\rho_X\) that can be found in the literature is demonstrated. The characterizations of uniform convex and uniform smoothness are then: \(X\) is uniformly convex iff \(\delta_X(\varepsilon) > 0\) for all \(\varepsilon \in (0, 2]\). \(X\) is uniformly smooth iff \(\lim_{\tau \to 0} \rho_X(\tau)/\tau = 0\).

**Definition 2.** Let \(p, q > 1\) be real numbers. We call \(X\) a \(p\)-convex space (also: \(p\)-uniformly convex; uniformly convex (with the modulus of convexity) of power type \(p\)) if it has \(\delta_X(\varepsilon) \geq c\varepsilon^p\) for some \(c > 0\). Respectively, \(X\) is called a \(q\)-smooth space (also: \(q\)-uniformly smooth) if it has \(\rho_X(\tau) \leq c\tau^q\) for some \(c > 0\).

**Proposition 3** (Chidume [14], Proposition 5.6, and Kazimierski [15], Theorems 2.41–43 and Corollary 2.34). A space \(X\) is \(p\)-convex \((q\)-smooth\) iff \(X^*\) is \(p^*\)-smooth \((q^*\)-convex\). Any \(p\)-convex space always has \(p \geq 2\), while a \(q\)-smooth space has \(q \leq 2\). Moreover a \(p\)-convex \((q\)-smooth\) space is also \(p^*\)-convex \((q^*\)-smooth\) for all \(p < p^* < \infty\) \((1 < q^* < q)\).

That such \(p\)-convex \((q\)-smooth\) spaces are by no means rare is demonstrated by the following theorem and the examples given right after.

**Theorem 4** (Kazimierski [15], Theorem 2.39). For any uniformly convex or uniformly smooth space \(X\) there always exists an equivalent norm such that \(X\) equipped with this norm is \(p\)-convex and \(q\)-smooth for some \(p \geq 2\), \(q \leq 2\). (This statement can even be generalized to super-reflexive spaces [16, Theorem 3.2] that will not be of further interest here.)

**Remark 2** (Prus and Smarzewski [17], Chidume [14], Example 4.13, and Kazimierski [15], Examples 2.37 and 2.38). For \(1 < p < \infty\) the sequence spaces \(\ell^p\), the Lebesgue spaces \(L^p\), and the Sobolev spaces \(W^{k,p}\) for a domain \(\Omega \subseteq \mathbb{R}^n\) are all \(\max[\{2, p\}\)-convex and \(\min[\{2, p\}\)-smooth. Hilbert spaces are 2-convex and 2-smooth.

This establishes the basic setting in terms of special classes of Banach spaces on which we define functions, \textit{functionals}, \(X \to \mathbb{R} \cup \{\pm \infty\}\) including those from which DFT derives its name.

**Definition 5.** A convex function \(f : X \to \mathbb{R} \cup \{+\infty\}\) is said to be proper if not identical to \(+\infty\). Let \(\Gamma_0(X)\) denote the set of proper, convex, lower semi-continuous functions \(X \to \mathbb{R} \cup \{+\infty\}\). On the dual side, \(\Gamma_0(X^*)\) is the set of proper, convex, weak-* lower semi-continuous functions \(X^* \to \mathbb{R} \cup \{+\infty\}\). Later we will mostly use the sign-flipped \(-\Gamma_0(X^*)\) for concave functions on \(X^*\).

**Lemma 6.** If \(X\) is reflexive, then \(\Gamma_0(X^*) = \Gamma_0(X^*)\).

**Proof.** Weak-* semi-continuity is equivalent to weak semi-continuity for \(X\) reflexive. But weak (semi-)continuity always implies strong (semi-)continuity, so indeed \(\Gamma_0(X^*) = \Gamma_0(X^*)\).

**Definition 7.** Sub- and superdifferential of a function \(f : X \to \mathbb{R} \cup \{\pm \infty\}\) are defined as

\[
\partial f(x) = \{x^* \in X^* \mid \forall y \in X : f(x) - f(y) \leq \langle x^*, x - y \rangle\},
\]

\[
\overline{\partial} f(x) = \{x^* \in X^* \mid \forall y \in X : f(x) - f(y) \geq \langle x^*, x - y \rangle\}.
\]
Theorem 11. A mapping $T : X \to X^*$ is called monotone if either
\begin{align}
\langle T(x) - T(y), x - y \rangle &\geq 0 \quad \text{or} \\
\langle T(x) - T(y), x - y \rangle &\leq 0
\end{align}
holds for all $x, y \in X$. If $x \neq y$ and the inequalities are strict, the property is accordingly called strict monotonicity. The same definitions are applicable to set-valued mappings if $T(x)$ is replaced by all $x^* \in T(x)$ when $T(x)$ is non-empty and the same for $y$.

Remark 3. It is well known that sub- and superdifferentials are always monotone. For $f \in \Gamma_0(X)$ they are even maximal monotone [19], which means they cannot be extended any further in a monotonous fashion. Strict convexity of a function yields strict monotonicity of its subdifferential and we will introduce a special, even stronger form of monotonicity in Lemma 20 below.

**Definition 8.** A mapping $T : X \to X^*$ is called monotone if either
\begin{align}
\langle T(x) - T(y), x - y \rangle &\geq 0 \quad \text{or} \\
\langle T(x) - T(y), x - y \rangle &\leq 0
\end{align}

**Lemma 9** (finiteness lemma). For $f \in \Gamma_0(X)$ the following statements are equivalent:
(i) The function $f$ is finite everywhere (equivalent to saying, $f$ has effective domain $X$).
(ii) The function $f$ is continuous everywhere.
(iii) The subdifferential $\partial f$ is non-empty everywhere (equivalent to saying, $\partial f$ has domain $X$).
(iv) For any (pre)compact set $A \subset X$ the set $\{x^* \in \partial f(x) \mid x \in A\} \subset X^*$ is bounded.

Proof. (i) ⇒ (ii). Continuity everywhere follows directly from Proposition 2.16 in Barbu and Precupanu [19], since the algebraic interior of the effective domain is just the whole $X$ in this case.
(ii) ⇒ (iii). Continuity at a point leads to a non-empty subdifferential at that point by Proposition 2.36 in Barbu and Precupanu [19].
(iii) ⇒ (iv). The subdifferential as a monotonous operator is locally bounded at every interior point of its domain, which is all of $X$ by (iii) [19, Theorem 1.144]. This means one can find an upper bound that holds for all $x \in A$ since there is a finite subcover for the (pre)compact $A$.
(iv) ⇒ (i). Take $x \in X$ arbitrary and any $y \in X$ where $f$ is finite. Then from the definition of the subdifferential in [3],
\begin{align}
|f(x)| &\leq \sup_{x^* \in \partial f(x)} |(x^*, x - y)| + |f(y)| \leq \|x - y\| \cdot \sup_{x^* \in \partial f(x)} \|x^*\| + |f(y)|.
\end{align}
Since the supremum is bounded by assumption from (iv), $f$ is finite at $x$. \qed

We use a non-standard definition of the convex conjugate (Legendre–Fenchel transform) adopted to the setting of DFT [1; 5].

**Definition 10.** For any $f : X \to \mathbb{R} \cup \{\pm \infty\}$ and $g : X^* \to \mathbb{R} \cup \{\pm \infty\}$, we define the convex conjugates,
\begin{align}
f^\vee(x^*) &= \inf \{f(x) + \langle x^*, x \rangle \mid x \in X\}, \\
g^\vee(x) &= \sup \{g(x^*) - \langle x^*, x \rangle \mid x^* \in X^*\}.
\end{align}

With respect to the convex conjugates the sets $\Gamma_0(X)$ and $-\Gamma_0(X^*)$ have a special relevance: Firstly, application of the convex conjugate always yields functionals of this type, and secondly, the convex conjugate acts as a bijection on those sets with the inverse map just given by the dual convex conjugate.

**Theorem 11.** Let $f : X \to \mathbb{R} \cup \{+\infty\}$ non-identical to $+\infty$ and $g : X^* \to \mathbb{R} \cup \{-\infty\}$ non-identical to $-\infty$, then [19, Proposition 2.19(i) and Theorem 2.22]
\begin{align}
f^\vee \in -\Gamma_0^\circ(X^*), \quad g^\vee \in \Gamma_0(X), \\
f = (f^\vee)^\vee &\iff f \in \Gamma_0(X), \\
g = (g^\vee)^\vee &\iff g \in -\Gamma_0^\circ(X^*).
\end{align}

\footnote{Lieb [3], Theorem 3.1(iii), even shows local Lipschitz continuity of the concave ground-state energy functional in the setting of standard DFT.}
Double application of the convex conjugates, \((f^\wedge)^\wedge\), always gives a \(\Gamma_0(X)\)-type functional that is called the convex hull, convex envelope, or closure of the generally non-convex \(f\). We further find an important theorem about the connection between the existence of a minimizer to (8) and the sub- and superdifferentials of \(f\) and \(f^\wedge\), respectively.

**Theorem 12.** Let \(f \in \Gamma_0(X)\), then the following are equivalent \cite[Proposition 2.33]{14}:

1. \(-x^* \in \partial f(x)\)
2. \(x \in \partial f^\wedge(x^*)\)
3. \(f^\wedge(x^*) = f(x) + \langle x^*, x \rangle\)

**Remark 4.** Note that a direct consequence of (i) and (ii) from the previous theorem is that the subdifferential of \(f\) is the negative inverse of the superdifferential of the conjugate of \(f\), i.e., \(\partial f = -[\partial f^\wedge]^{-1}\). Additionally, (i) yields the expected possibility of translating the condition for \(x\) being a minimizer of \(f + x^*\) from (iii) into finding a zero subdifferential, \(0 \in \partial(f + x^*)(x)\).

**Definition 13.** The generalized duality map \(J_p : X \rightarrow X^*\) is

\[
J_p(x) = \{x^* \in X^* \mid \|x^*\| = \|x\|^{p-1}, \langle x^*, x \rangle = \|x\|^p\}.
\]  

\(\text{(13)}\)

**Remark 5.** If \(p = 2\) then \(J_p = J_2 = J\) is the (normalized) duality map. The generalized duality map is just the subdifferential of the functional \(\phi_p = \| \cdot \|^p/p\) on \(X\). \(\partial \phi_p = J_p\) \cite[Proposition 4.9]{14}. Since further \(J_p(x) = \|x\|^{p-2} J(x)\) for \(x \neq 0\), which follows directly from the definition, many of the properties of \(J\) translate directly to \(J_p\). Most importantly, for \(X\) \(p'\)-convex (any \(p' \geq 2\)) and \(X^*\) strictly convex, \(J_p\) \((p \neq p'\) possible\) is single-valued, demicontinuous (norm-to-weak continuous), bijective, and has a single-valued inverse that is given by \(J_p^{-1} = J_{p'} : X^* \rightarrow X\) \cite[Theorem 2.44(4)]{14}. Since the same notation \(J_p\) for a map \(X \rightarrow X^*\) and a map \(X^* \rightarrow X\) could cause confusion, the duality map \(X^* \rightarrow X\) will always be denoted \(J_p^{-1}\). The generalized duality map \(J_p\) can be better suited than \(J\) to reflect the geometry of the underlying Banach space, as it is the case with \(J_p\).

**Lemma 14.** Let \(X\) be \(p\)-convex and \(X^*\) be \(p^*\)-convex, then there are \(\varepsilon, \varepsilon' > 0\) such that for all \(x, y \in X\)

\[
\varepsilon\|y\|^p \leq \|x + y\|^p - \|x\|^p - p(J_p(x), y) \leq \varepsilon'\|y\|^p.
\]  

\(\text{(14)}\)

If \(X\) is a real Hilbert space then \(p = p^* = 2\), the constants are \(\varepsilon = \varepsilon' = 1\), and the relation above reduces to the familiar polarization identity \(\|x + y\|^2 = \|x\|^2 + \|y\|^2 + 2\langle x, y \rangle\).

**Proof.** The statement follows directly from Corollary 1(iii) and Corollary 1(iii)’ in Xu \cite{13}, where we have chosen \(q = p\) and used that the duality map \(J_p\) is single-valued for smooth spaces \(X\).

**B. Moreau–Yosida regularization**

**Lemma 15.** For all \(p > 1\), the function \(\phi_p : X \rightarrow \mathbb{R}, \phi(x) = \|x\|^p/p\) is strictly convex iff the space \(X\) is strictly convex.

**Proof.** For strict convexity of \(X\) we draw upon the definition in Barbu and Precupanu \cite[Proposition 1.103(ii)]{13}, where also the above statement for \(p = 2\) is shown: A space \(X\) is strictly convex if the connecting line between two points \(x \neq y\) on the unit sphere lies completely within the unit sphere.

Now assume \(X\) strictly convex. Take any \(x, y \in X, \lambda \in (0, 1)\), then by the triangle inequality \(\|\lambda x + (1-\lambda)y\|^p \leq (\lambda\|x\| + (1-\lambda)\|y\|)^p\). Since \(t \rightarrow t^p\) is clearly strictly convex on \(\mathbb{R}\) it follows \(\|\lambda x + (1-\lambda)y\|^p < \lambda\|x\|^p + (1-\lambda)\|y\|^p\) if \(\|x\| \neq \|y\|\). In the case \(\|x\| = \|y\|\), set \(\|x\| = \|y\| = 1\) without loss of generality and then resort to strict convexity of \(X\) to show \(\|\lambda x + (1-\lambda)y\|^p < \lambda\|x\|^p + (1-\lambda)\|y\|^p\) for \(x \neq y\) from \(\|\lambda x + (1-\lambda)y\| < 1\).

The strict convexity of \(X\) follows directly from the defining inequality of strict convexity of \(\phi_p\) when setting \(\|x\| = \|y\| = 1\).

\(\square\)
Lemma 16. If $X^*$ is strictly convex then $\phi_p$ is Gâteaux differentiable. If $X^*$ is uniformly convex then $\phi_p$ is Fréchet differentiable. In both cases, the derivative of $\phi_p$ is the generalized duality map $J_p$.

Proof. That the subdifferential of $\phi_p$ is identical to the generalized duality map $J_p$ was already mentioned in Remark 3. If $X^*$ is strictly convex then the normalized duality map is single-valued [19, Proposition 1.117(iii)], which because of $J_p(x) = \|x\|^{-2}J(x)$ also means that $J_p$ is single-valued. This in turn already establishes Gâteaux differentiability [19, Proposition 2.40]. If furthermore $X^*$ is uniformly convex then the normalized duality map is also continuous [19, Proposition 1.117(vi)]. Continuity of $J_p$ is not lost close to the origin, as can be seen using $J$ homogeneous by writing $J_p(x) = \|x\|^{-2}J(\|x\|^{-1}x/y/\|x\|) = \|x\|^{-1}J(x/\|x\|)$. Finally, continuity of the differential implies Fréchet differentiability [21, Lemma 34.3].

Definition 17. For $\varepsilon > 0$ the $p$-Moreau–Yosida (pMY) regularization of $f \in \Gamma_0(X)$ is defined as the infimal convolution of $f$ with $\varepsilon^{-1}\phi_p$, $f_{\varepsilon}(x) = \inf\{f(y) + \varepsilon^{-1}\phi_p(x - y) \mid y \in X\}$. (15)

Remark 6. The case $p = 2$ recovers the standard MY regularization. Since the $p$ will later be fixed by the underlying space $X$ we will not include it in the notation for the pMY regularization. This generalized form of the MY regularization can be found in the textbook Bauschke and Combettes [22, Proposition 12.15], where it is defined on Hilbert spaces. Kuwae [23] and Bacák and Kohlenbach [24] present it with the MY regularization can be found in the textbook Bauschke and Combettes [22, Proposition 12.15], where it is defined on Hilbert spaces. Kuwae [23] and Bacák and Kohlenbach [24] present it with $\varepsilon^{-1}$ replaced by $\varepsilon^{1-p}$ under the name Hopf–Lax formula that is related to solutions of a Hamilton–Jacobi equation and consequently $f_{\varepsilon}$ is also called a Hamilton–Jacobi semigroup (in $t = \varepsilon^{1/(p-1)}$). Penot and Ratsimahalo [20] and Bacák and Kohlenbach [24] allow for the even more general Young functions instead of just $\phi_p$. Further, in Penot and Ratsimahalo [20] a Yosida approximation for multimappings from $X$ into $X^*$ is discussed alongside its relation to the MY regularization of functionals, a relation that is also established here with (29). Another form of generalization of the standard MY regularization, by replacing $\phi_p(x)$ with $\|Ax\|_2^2/2$, is briefly discussed in the conclusions in Kvaal et al. [1]. Therein, some interesting possible connections to actual DFT approaches that resemble the regularization technique are noted as well. The definition of regularization as an infimal convolution has the benefit of yielding convenient relations between the convex conjugates of regularized and unregularized functional, expressed through equations like (21) and (22).

Remark 7. Penot and Ratsimahalo [20] show that $f_{\varepsilon} \in \Gamma_0(X)$, more specifically that the regularized $f_{\varepsilon}$ is even a continuous, convex function [22, Proposition 5.6]. From the definition it follows directly that $f_{\varepsilon} \leq f$ everywhere and that for all $x, y \in X$ it holds $f_{\varepsilon}(x) \leq f(y) + \varepsilon^{-1}\phi_p(x - y)$, which in the context of Fig. 1 means that the regularization paraboloids are always above $f_{\varepsilon}$. This feature will be used in the proof of convergence in Section IV. Further, for any minimum $x \in X$ of $f$ it holds that also $f_{\varepsilon}$ has a minimum at the same $x$. The infimum in (15) above is always uniquely attained, because the added $\phi_p$ is strictly convex if $X$ is strictly convex by Lemma 13 [19, Proposition 1.103(iv), Theorem 2.11, Remarks 2.12 and 2.13]. This unique minimizer for any given $x$ allows for the definition of the proximal mapping.
Lemma 20. Let $f \in \Gamma_0(X)$, and $\varepsilon > 0$. For all $x \in X$ the proximal mapping is
\[
\text{prox}(x) = \arg\min_{\varepsilon f} \{ f(y) + \varepsilon^{-1} \phi_p(x - y) \mid y \in X \}
= \arg\min_{\varepsilon f} \{ \varepsilon f(y) + \phi_p(x - y) \mid y \in X \}.
\]

Remark 8. The above definition leads, by seeking the minimum in (15), to the following useful relations,
\[
\frac{\partial f(\text{prox}(x))}{\varepsilon f} - \varepsilon^{-1} J_p(x - \text{prox}(x)) \ni 0,
\]
\[
f_\varepsilon(x) = f(\text{prox}(x)) + \varepsilon^{-1} \phi_p(x - \text{prox}(x)).
\]

Remark 9 (proximal-point algorithm). For $f \in \Gamma_0(X)$ a sequence defined by $x_{i+1} = \text{prox}_f(x_i)$ gradually approaches the infimum of $f$ like depicted in Fig. II see also Bauschke and Combettes [22, Theorem 27.1]. Anticipating Lemma [22] this amounts exactly to a gradient-descent algorithm for the regularized $f_\varepsilon$. The KS iteration itself, as described in Theorem [23], is not a gradient-method because the correct gradient is deemed in calculable as a solution of the inverse Schrödinger problem. Instead an auxiliary reference problem, that is in a sense thought close to the original Schrödinger problem, is used to design a different algorithm with the Hartree-exchange-correlation potential as a new ingredient.

Definition 19. For $p > 1$ a function $f : X \to \mathbb{R} \cup \{\pm \infty\}$ is called $p$-strongly convex with parameter $\varepsilon > 0$ if $f - \varepsilon \phi_p$ is still convex. It is called $p$-strongly concave with parameter $\varepsilon > 0$ if $f + \varepsilon \phi_p$ is still concave. Choosing $p = 2$ recovers the usual definition of strong convexity and strong concavity.

The following Lemma of equivalences includes a crucial step of the proof of convergence and is inspired by a finite-dimensional treatment in Rockafellar and Wets [23, Proposition 12.60].

Lemma 20. Let $X$ be strictly convex and $X^*$ be $p^*$-convex, then the following statements are equivalent for $f_\varepsilon \in \Gamma_0(X)$:

(i) $f_\varepsilon$ is the MY regularization with parameter $\varepsilon$ of some $f \in \Gamma_0(X)$.

(ii) $(f_\varepsilon)^\wedge$ is $p^*$-strongly convex with parameter $\varepsilon$.

(iii) There is a $\zeta > 0$ such that for all $x^*, y^* \in X^*$ and all $x \in (f_\varepsilon)^\wedge(x^*)$, $y \in (f_\varepsilon)^\wedge(y^*)$ with non-empty superdifferentials it holds (strong monotonicity property)
\[
(x^* - y^*, x - y) \leq -\varepsilon \zeta \| x^* - y^* \|^{p^*}.
\]

Proof. In the proof we will use, as noted in Remark 5 that under the given assumptions on $X$ the duality map $J_p : X \to X^*$ is a bijection and thus has a well-defined inverse.

(i) $\Rightarrow$ (ii). We note by definition
\[
(f_\varepsilon)^\wedge(x^*) = \inf \{ f(y) + \varepsilon^{-1} \phi_p(x - y) + \langle x^*, y \rangle \mid y \in X \}
= \inf \{ f(y) + \varepsilon^{-1} \phi_p(x - y) + \langle x^*, y - x \rangle \mid x, y \in X \}
= \inf \{ f(y) + \varepsilon^{-1} \phi_p(x - y) + \varepsilon^{-1} \phi_p(x - y) \mid y \in X \}
= f^\wedge(x^*) + \varepsilon^{-1} \phi_p(x^*)
\]

From $\phi_p(x) = \|x\|^{p^*}/p$ it follows by definition that $\phi_p(x^*) = \inf \{ \|x\|^{p^*}/p + \langle x, x \rangle \mid x \in X \}$, where the minimum is uniquely attained because of strict convexity. Seeking the minimum by subdifferentiation yields $J_p(x) = -x^*$. The definition of the generalized duality map then provides the relations $\|x^*\| = \|x\|^{p^*}$ and $-(x^*, x) = \|x\|^{p^*}$ that must hold for $\lambda > 0$. Inserting these two equalities into the expression of the convex conjugate of $\phi_p$ gives $\phi_p(x^*) = -\|x^*\|^{p^*}/p^* = -\phi_{p^*}(x^*)$. Additionally, the scaling relation $\lambda f^\wedge(x^*) = \lambda f^\wedge(x^*/\lambda)$, $\lambda > 0$, yields $\varepsilon^{-1} \phi_p(x^*) = -\varepsilon \phi_{p^*}(x^*)$ and thus $f^\wedge = (f_\varepsilon)^\wedge + \varepsilon \phi_{p^*}$, which means that $(f_\varepsilon)^\wedge$ is $p^*$-strongly concave and thus proves (ii).

(ii) $\Rightarrow$ (iii). Because of $p^*$-strong concavity of $(f_\varepsilon)^\wedge$ we know there is a $f \in \Gamma_0(X)$ such that
\[
(f_\varepsilon)^\wedge = f^\wedge + \varepsilon(-\phi_{p^*}),
\]
Proof. The definition of convex conjugation in (8) applied to \( \partial f \) on \( X^* \) by \( \partial(-\phi_{p'}) = -\partial \phi_{p'} = -J_p^{-1} \) from Remark 5. Note that the sum rule for the superdifferentials only holds because the involved functionals are all concave. Now for any \( \tilde{x} \in \partial f^*(x^*) \), \( \tilde{y} \in \partial f^*(y^*) \) by definition of the superdifferential it holds

\[
\begin{aligned}
f^*(x^*) - f^*(y^*) &\geq \langle x^* - y^*, \tilde{x} \rangle, \\
f^*(y^*) - f^*(x^*) &\geq \langle y^* - x^*, \tilde{y} \rangle = \langle x^*-y^*,-\tilde{y} \rangle.
\end{aligned}
\]

Adding those two inequalities gives (monotonicity of the superdifferential)

\[
\langle x^*-y^*,\tilde{x}-\tilde{y} \rangle \leq 0.
\]

Equation (22) results in the relations \( x = \tilde{x} - \varepsilon J_p^{-1}(x^*) \) and \( y = \tilde{y} - \varepsilon J_p^{-1}(y^*) \) for \( x \in \partial f^*(x^*) \), \( y \in \partial f^*(y^*) \), and inserting this into (25) yields

\[
(x^* - y^*, x - y) \leq -\varepsilon \langle x^* - y^*, J_p^{-1}(x^*) - J_p^{-1}(y^*) \rangle.
\]

Corollary 1(iii) in Xu [13], but with the roles of \( X \) and \( X^* \) interchanged, shows that for \( X^* \) \( p^* \)-convex there is a \( \zeta > 0 \) such that

\[
(x^* - y^*, J_p^{-1}(x^*) - J_p^{-1}(y^*)) \geq \zeta \|x^* - y^*\|^p.
\]

This together with (26) establishes (iii).

\[\text{(iii) } \Rightarrow (i).\] The strong monotonicity property of (iii) implies that \( T = \partial(f^*)^\sim + \varepsilon J_p^{-1} \) is still monotone, \( \langle x^* - y^*, T(x^*) - T(y^*) \rangle \leq 0 \). Also, the domain for the monotone \( \partial(f^*)^\sim \) cannot be extended (Remark 5) and thus it is maximal monotone alongside \( T \). It then holds that \( R(T) = X \) [13, Theorem 1.141] so \( \pm T^{-1} \) is well-defined on \( X \) and maximal monotone as the inverse of a maximal monotone operator [25, Exercise 12.8(a)].

This makes \( -T^{-1} \) the subdifferential of an \( f \in \Gamma_0(X) \) [13, Theorem 2.46, Example 2.49] (note the negative sign because the monotonicity is \( \leq 0 \) instead of \( \geq 0 \)). By inversion we get from Theorem 12 that

\[
\partial f_{\varepsilon} = -[\partial(f^*)^\sim]^{-1} = [-T + \varepsilon J_p^{-1}]^{-1},
\]

which is a generalization to the Yosida approximation of \( -T^{-1} = \partial f \) [24, Example 12.13]. In the last step we will demonstrate that this Yosida approximation of \( \partial f \) indeed coincides with the subdifferential of the \( p^* \)MY regularization of \( f \) (up to an additive constant). For this we take (22) that has been derived as the subdifferential of a \( p^* \)MY regularization and invert it using Theorem 12 once more,

\[
\partial f_{\varepsilon} = [(\partial f)^\sim - \varepsilon J_p^{-1}]^{-1}.
\]

This is exactly the same as (25) from before and since the subdifferential uniquely defines \( f_{\varepsilon} \) (up to an additive constant) [13, Theorem 2.46], we have established that \( f_{\varepsilon} \) must be the \( p^* \)MY regularization of some \( f \in \Gamma_0(X) \), which corresponds to (i).

**Lemma 21.** Let the spaces be like in Lemma 24 and \( f_{\varepsilon} \) be the \( p^* \)MY regularization with parameter \( \varepsilon \) of some \( f \in \Gamma_0(X) \), then for any \( w^* \in X^* \) the function \( f_{\varepsilon} + w^* \) is also a \( p^* \)MY regularization with parameter \( \varepsilon \).

Furthermore, let \( x_{\varepsilon} \) be a minimizer of \( f_{\varepsilon} + w^* \), then \( x = x_{\varepsilon} + \varepsilon J_p^{-1}(w^*) \) is a minimizer of \( f + w^* \) and the functional minima are related by

\[
f_{\varepsilon}(x_{\varepsilon}) + (w^*, x_{\varepsilon}) = f(x) + (w^*, x) - \varepsilon \phi_{p^*}(w^*). \tag{30}
\]

**Proof.** The definition of convex conjugation in (8) applied to \( f_{\varepsilon} + w^* \) gives when using also (24)

\[
(f_{\varepsilon} + w^*)^\sim(x^*) = \inf \{ f_{\varepsilon}(x) + (x^* + w^*, x) \mid x \in X \} = (f_{\varepsilon})^\sim(x^* + w^*) = f^\sim(x^* + w^*) - \varepsilon \phi_{p^*}(x^* + w^*). \tag{31}
\]

The right-hand side of (31) is a concave functional \( f^\sim \) plus the strictly concave \( -\varepsilon \phi_{p^*} \), thus \( (f_{\varepsilon} + w^*)^\sim \) is \( p^* \)-strongly concave with parameter \( \varepsilon \). Lemma 20(ii) \( \Rightarrow (i) \) then says that if \( f_{\varepsilon} + w^* \) is \( p^* \)-strongly concave with parameter \( \varepsilon \) then \( f_{\varepsilon} + w^* \) is a \( p^* \)MY regularization with parameter \( \varepsilon \).

Furthermore, if \( x \) is an infimum of \( f + w^* \) then by (8) it has the infimal value \( f(x) + (w^*, x) = f^\sim(w^*) \), whereas an infimum \( x_{\varepsilon} \) of \( f_{\varepsilon} + w^* \) gives \( f_{\varepsilon}(x_{\varepsilon}) + (w^*, x_{\varepsilon}) = (f_{\varepsilon})^\sim(w^*) \). Using again (24) we see that \( (f_{\varepsilon})^\sim(w^*) = f^\sim(w^*) - \varepsilon \phi_{p^*}(w^*) \), so the values differ by \( \varepsilon \phi_{p^*}(w^*) \). For the location of the minima we use Theorem 12(ii) \( x \in \partial f^\sim(w^*), x_{\varepsilon} \in \partial(f_{\varepsilon})^\sim(w^*) \), and then (22) gives the desired relation \( x_{\varepsilon} = x - \varepsilon J_p^{-1}(w^*) \).
The following result finally gives the differentiability of regularized functionals and was already stated for the usual MY regularization in Laestadius et al. [2] Theorem 9. Here we extend it to pMY-regularized functionals and include a fully contained proof for the sake of completeness.

**Lemma 22.** Let $X$ be $p'$-convex for any $p' \geq 2$, $X^*$ be strictly convex, and $f \in \Gamma_0(X)$ with pMY regularization $f_\varepsilon$ ($p \neq p'$ possible). Then $f_\varepsilon$ is Gâteaux differentiable on $X$ and

$$
\nabla f_\varepsilon(x) = \varepsilon^{-1} J_p(x - \text{prox}_\varepsilon(x)).
$$

(32)

If $X^*$ is uniformly convex, then $f_\varepsilon$ is even Fréchet differentiable. If $X$ is a Hilbert space and $p = 2$, then $\nabla f_\varepsilon$ is Lipschitz-bounded with constant $\varepsilon^{-1}$.

**Proof.** In the following we set $p_x = \text{prox}_{\varepsilon f}(x), p_y = \text{prox}_{\varepsilon f}(y)$. We choose $p^*_y = \varepsilon^{-1} J_p(y - p_y)$, which by [17] means $p^*_y \in \partial f(p_y)$. We draw upon [13] to write

$$
f_\varepsilon(y) - f_\varepsilon(x) = f(p_y) - f(p_x) + \varepsilon^{-1} \phi_p(y - p_y) - \varepsilon^{-1} \phi_p(x - p_x)
$$

$$
\leq (p^*_y - p_y) - \varepsilon^{-1} \phi_p(y - p_y) + (p^*_y - p_x) + \varepsilon^{-1} \phi_p(y - p_y) - \varepsilon^{-1} \phi_p(x - p_x).
$$

(33)

From Chidume [14] Theorem 4.10 we have that $\phi_p(y) - \phi_p(x) - (J_p(y), y - x) \leq 0$ and so the above can be further estimated by $f_\varepsilon(y) - f_\varepsilon(x) \leq (p^*_y, y - x)$. If we analogously work with $p^*_x = \varepsilon^{-1} J_p(x - p_x)$, the result is $f_\varepsilon(y) - f_\varepsilon(x) \geq (p^*_x, x - y)$ and together they yield

$$
0 \leq f_\varepsilon(y) - f_\varepsilon(x) - (p^*_y, y - x) \leq (p^*_y - p^*_x, y - x).
$$

(34)

Now set $y = x + tu$, $t \in \mathbb{R}$, divide by $t$, and take the limit $t \to 0$. Then

$$
0 \leq \lim_{t \to 0} \frac{f_\varepsilon(x + tu) - f_\varepsilon(x)}{t} - (p^*_x, u) \leq \lim_{t \to 0} (p^*_y - p^*_x, u).
$$

(35)

On the right hand side, the demicontinuity of $J_p$ from Remark [5] for $X$ $p'$-convex ensures $\lim_{t \to 0} (p^*_y - p^*_x, u) = 0$ if $\lim_{t \to 0} (y - p_y) - (x - p_x) = p_x - \lim_{t \to 0} p_y = 0$. This is indeed true for uniformly convex $X$ due to a recent continuity result for the given generalized proximal mapping by Bacák and Kohlenbach [24] Theorem 3.10. So

$$
\lim_{t \to 0} \frac{f_\varepsilon(x + tu) - f_\varepsilon(x)}{t} = (p^*_x, u),
$$

(36)

which shows that $f_\varepsilon$ is Gâteaux differentiable and $\nabla f_\varepsilon(x) = p^*_x = \varepsilon^{-1} J_p(x - p_x)$. Proposition 1.146(i) in Barbu and Precupanu [19] further states that if $X^*$ is uniformly convex, then $\nabla f_\varepsilon : X \to X^*$ is continuous, which implies Fréchet differentiability [21] Lemma 54.3.3]. Finally, if $X$ is a Hilbert space and $p = 2$, the duality map is additive [10] Proposition 1.117(ii) and $x \mapsto x - p_x$ is firmly non-expansive [22] Proposition 12.27] which gives an overall Lipschitz-constant of $\varepsilon^{-1}$.

**Remark 10.** In the Hilbert space case, the equivalence of the above result to the propositions of Lemma 20 can be established like in Rockafellar and Wets [25] Proposition 12.60]. Note that the Lipschitz-bound of $\varepsilon^{-1}$ is the reason why one might talk about a curvature bound for $f_\varepsilon$. Equation (32) has an easy interpretation that will be used in the proof of convergence in Section IV. Since $J_p$ is just the differential of $\phi_p$, we rewrite $\nabla f_\varepsilon(x) = \varepsilon^{-1} \nabla \phi_p(x - \text{prox}_{\varepsilon f}(x))$ and see that at any point the regularized functional is aligned tangentially to its regularization function.

### III. REGULARIZED DENSITY-FUNCTIONAL THEORY

A basic task of quantum chemistry is to find the ground-state density (and related observables) of an interacting $N$-electron system in a fixed external potential. This problem is hard because of the possibly huge number of internal degrees of freedom expressed by the enormous size of the underlying wave-function Hilbert space. It can, however, be reformulated as the variational problem of finding a density in $\tilde{X}$ that minimizes the total energy. This energy given by the ground-state energy functional $E : X^* \to \mathbb{R} \cup \{-\infty\}$,

$$
E(x^*) = \tilde{F}^\wedge(x^*) = \inf_{x \in \tilde{X}} \{\tilde{F}(x) + \langle x^*, x \rangle\}.
$$

(37)
The ground-state energy \( E \) is defined as the infimum of the internal energy, \( \tilde{F}(x) \), plus the potential energy expressed as the linear pairing with the external potential \( x^* \in X^* \), taken over all physical densities \( \tilde{X} \subset X \) that are assumed to form a closed convex subset. No special assumptions are imposed on the internal energy functional \( \tilde{F} : \tilde{X} \to \mathbb{R} \cup \{+\infty\} \), although in (standard) DFT it can always be thought of as a constrained-search functional over pure-state \( N \)-representable densities \([3, 26]\) that form the closed convex set described in \((67)\). If the constrained search is performed over density matrices instead of pure states like in \((69)\) later, one even has \( \tilde{F} = F \) on \( \tilde{X} \) \([4.5]\), where \( F \) is the convex hull of \( \tilde{F} \) given in \((38)\) below. This identity holds in the standard DFT setting, but it is unclear if it remains true in other DFT settings like CDFT or its simplified version restricted to uniform magnetic fields \([27]\).

As stressed before, for densities and potentials we work in a setting of a Banach space \( X \) and its (topological) dual \( X^* \). Note that this extends the space of densities far beyond the usual physical densities that are collected in the subset \( \tilde{X} \subset X \), since naturally \( X \) also contains elements that are not normalized to the particle number \( N \) or take negative values. This is why a general element of \( X \) will be referred to as \textit{quasidensity} instead of just density. Although the energy functional defined like in \((37)\) can take in principle minus-infinity values, its finiteness will later be an important restriction for the KS algorithm.

The density functional \( F \) (universal Lieb functional) on the whole of \( X \) is then defined as the \textit{biconjugate} of \( \tilde{F} \). This corresponds to the \textit{convex hull} of \( \tilde{F} \), defined as the pointwise supremum over all convex, lower-semicontinuous functions with majorant \( \tilde{F} \). Then, by Theorem \([11]\) \( E \) on \( X^* \) is linked back to \( F \) by the convex conjugate again,

\[
F(x) = E^\vee(x) = \sup_{x^* \in X^*} \{E(x^*) - \langle x^*, x \rangle\} = (\tilde{F}^\vee)^\vee(x), \quad F \in \Gamma_0(X) \tag{38}
\]

\[
E(x^*) = F^\vee(x^*) = \inf_{x \in X} \{F(x) + \langle x^*, x \rangle\} = (E^\vee)^\vee(x^*), \quad E \in -\Gamma_0(X^*). \tag{39}
\]

In a reflexive Banach space setting, which will become important in the proof of convergence, we even have \( E \in -\Gamma_0(X^*) \) by Lemma \([6]\). Further, it holds \( F(x) = +\infty \) for all \( x \in X \setminus \tilde{X} \) and \( F(x) = \tilde{F}(x) \) for all minimizers of \((37)\) by definition as a convex hull. More generally, since the space of physical densities \( \tilde{X} \) was assumed closed convex, the extension to \( X \) cannot introduce new minimizers in \((39)\) that by Theorem \([12]\) are found as solutions \( x \in \tilde{X} \) of \( x \in \partial E(x^*) \) or \( -x^* \in \partial \tilde{F}(x) \), equivalently. We will thus write and later use \( \partial E(x^*) \subset \tilde{X} \).

Now, the central idea of DFT is to invert the process of finding a ground-state density in a given potential by going from an attained minimum \( x \) in \((39)\) back to the corresponding \( x^* \) in order to be able to map from solutions of interacting systems to non-interacting systems that are much cheaper to solve. By Theorem \([12]\) this is equivalent to determining all elements in the subdifferential of \( F \) at \( x \). The notion of a subdifferential already indicates that there might be many solutions, thus in principle different potentials can produce the same ground-state density. In DFT such a situation is undesirable, since to any given density the corresponding potential from a non-interacting reference (KS) system should be assigned. That this mapping is given uniquely is usually the content of the Hohenberg–Kohn theorem, but this statement is by far not secured in different density-functional settings \([23]^{*} - [32]^{*}\).

A first step towards a guaranteed density-potential mapping is to make possibly coinciding densities that come from different potentials different. The idea is to mark or mix them with the respective potential, so that they differ subsequently. In other words, the unknown quantity is not the density alone, but the density and the potential. This change in strategy is discussed as an abstract axiomatic approach for the KS iteration in Lammert \([33]\). Here, we follow a more direct approach that turns out to be equivalent to regularization. Say \( x_1, x_2 \in \tilde{X} \) come from different potentials \( x_1^*, x_2^* \in X^* \), then one way to mix them is by creating \textit{quasidensities} \( x = x_1 - \varepsilon J_{p}^{-1}(x_1^*) \) and \( x_2 = x_2 - \varepsilon J_{p}^{-1}(x_2^*) \) that differ for sure. In order to work with these quasidensities we need the corresponding energy functional, let us call it \( E_\varepsilon \), that fulfills

\[
x = \tilde{x} - \varepsilon J_{p}^{-1}(x^*) \in \partial E_\varepsilon(x^*) \tag{40}
\]

when \( \tilde{x} \in \partial E(x^*) \). Since \( \partial(-\phi_\varepsilon) = -J_{p}^{-1} \) on \( X^* \), this functional must be \( E_\varepsilon = E - \varepsilon \phi_\varepsilon \). Comparing these relations to \((21)\) and \((22)\), we see that \( \tilde{E} = F^\vee \) and \( E_\varepsilon = (F_\varepsilon)^\vee \), where \( F_\varepsilon \) is the pMY regularization of \( F \). In this way we have just reinvented pMY regularization in order to have a well defined density-potential mapping. That we can indeed invert the mapping \( \partial E_\varepsilon : X^* \to X \) is guaranteed by its strict monotonicity (Lemma \([24]\ iii))

---

2 This important restriction was not explicitly mentioned in our previous works on the topic \([3,4]\). It is always fulfilled in the DFT and current DFT settings discussed in Section \([5]\) here.
that makes $\partial E_\varepsilon$ an injective (set-valued) map. It equivalently means that its inverse $-\partial F_\varepsilon$ always maps to singletons, a fact that is also guaranteed by the (Gateaux) differentiability of $F_\varepsilon$ (Lemma 22 and Barbu and Precupanu [14, 2.40]). Phrased differently once more, in order to go from densities to potentials, we must find the maximizer of $\sup_{\varepsilon \in X} \{ E_\varepsilon(x^*) - \langle x^*, x \rangle \}$, which must be unique since $E_\varepsilon$ is strictly concave (Lemma 24 ii)).

To summarize, $F_\varepsilon$ is defined as the $\phi$MY regularization of $F$ and $E_\varepsilon$ as its convex conjugate,

$$E_\varepsilon(x^*) = (F_\varepsilon)\wedge (x^*) = \inf_{x \in X} \{ F_\varepsilon(x) + \langle x^*, x \rangle \}. \quad (41)$$

This transforms the in general non-differentiable $F$ into a proper, lower semi-continuous, convex, and differentiable functional $F_\varepsilon$, for which the results of Lemma 20 are applicable. As noted before, differentiability automatically yields a unique density-potential mapping (as asserted by the Hohenberg–Kohn theorem, which closes the circle). The condition for a minimizer of (41) is commonly translated into

$$-x^* = \nabla F_\varepsilon(x) \quad (42)$$

by Theorem 12. We have here replaced the subdifferential by a unique gradient “$\nabla$” because of the achieved differentiability, so any ground-state quasidensity is mapped to a corresponding potential. Thus, regularization avoids both the problem of $\nu$-representability (which densities are ground-state densities of an external potential) and the not generally warranted availability of a Hohenberg–Kohn theorem (unique mapping from such $\nu$-representable ground-state densities to their associated potentials). Using the other implication of Theorem 12 a minimizer of (41) can equivalently be found as

$$x \in \partial E_\varepsilon(x^*). \quad (43)$$

As has been noted before, for the energy functional we have the relations already derived in (21) and (22),

$$E_\varepsilon = E - \varepsilon \phi_p, \quad (44)$$

$$\partial E_\varepsilon = \partial E - \varepsilon \nabla^{-1} \phi_p, \quad (45)$$

which give a useful link between the regularized and the original setting that is also expressed in Lemma 21.

From this point on we will always talk about two systems and their associated functionals: The full system represented by $(\tilde{F}^1, F^1, E^1)$, and the reference system with $(\tilde{F}^0, F^0, E^0)$. The KS iteration scheme is then set up as follows. Choose such a reference system defined by $\tilde{F}^0$ on $\tilde{X} \subset X$ that is reasonably close to the full, interacting system with $F^1$. Here close means that similar terms appear in the expressions of the internal energy, so that some parts effectively cancel out when comparing the two systems. The other desired property for the reference system is that it is easy to solve, which means finding the ground-state density $x \in \partial E^0(x^*)$ for a chosen potential $x^* \in X^*$ should be computationally cheap. For the given $\tilde{F}^0$, $\lambda \in \{0, 1\}$, we define $E^\lambda, F^\lambda, E^\lambda_\varepsilon, F^\lambda_\varepsilon$ as above. Fixing the external potential $w^* \in X^*$ of the full problem, one can ask which KS potential $z^* \in X^*$ must be chosen such that both potentials lead to the same ground-state quasidensity $z \in X$,

$$-w^* = \nabla F^\lambda_\varepsilon(z), \quad -z^* = \nabla F^\lambda_\varepsilon(z). \quad (46)$$

We see that for the regularized setting this construction is always possible, although the ground-state (quasi)density does not have to be unique (in quantum mechanics this is due to possible degeneracy of the ground-state) and subsequently also the assigned KS potential will, in general, be non-unique. Subtracting the two equations of (40) and further introducing sequences $\{x_i\}_i \subset X$, $\{x^*_i\}_i \subset X^*$ instead of $z, z^*$ leads to

$$z^* = w^* + \nabla F^\lambda_\varepsilon(z) - \nabla F^0_\varepsilon(z) = w^* + \nabla (F^\lambda_\varepsilon(z) - F^0_\varepsilon(z)), \quad (47)$$

$$x^*_{i+1} = w^* + \nabla F^\lambda_\varepsilon(x_i) - \nabla F^0_\varepsilon(x_i) = w^* + \nabla (F^\lambda_\varepsilon(x_i) - F^0_\varepsilon(x_i)), \quad x^*_{i+1} \in E^\lambda_\varepsilon(x^*_{i+1}). \quad (48)$$

This manipulation captures the whole KS iteration scheme. The unknown quantities $z, z^*$ get replaced by sequences that hopefully converge like $x_i \rightarrow z, x^*_i \rightarrow z^*$, after starting from some initial guess. That the correct stopping condition is fulfilled can be immediately checked by setting $x_i = z$ or $x^*_{i+1} = z^*$ and using (40). On the other hand, the linearity of the gradient allows the introduction of an energy functional $F^\lambda_\varepsilon - F^0_\varepsilon$ (Hartree-exchange-correlation (Hxc) functional), which is the centerpiece of DFT’s machinery. At least some internal effects are expected to cancel which makes good approximations to this expression feasible. Note carefully that this introduction of the Hxc functional depends critically on the differentiability of $F^\lambda_\varepsilon$. If such differentiability is tacitly assumed, as it is the case in most presentations of DFT, see for example Engel and Dreizler [34, (2.105)], then it is as if we put ourselves already into a regularized setting.
With the KS iteration a direct solution of the full, interacting system is completely avoided and only the ground-state density of the non-interacting, reference system must be determined in the second step \[13\]. In order to show convergence, a further ingredient that is known from other self-consistent-field methods \[7, 8\] called optimal damping has to be added to the algorithm \[15\]. It means that not the full step from \(x_i\) to \(x_{i+1}\) is taken, but only a fraction of it. The final algorithm, called MYKSODA, is presented in the next section including its proof of convergence.

IV. MYKSODA PROOF OF CONVERGENCE

We now state our main result, the convergence of the MYKSODA algorithm described within the following theorem.

Theorem 23. Let the real Banach space \(X\) be \(p\)-convex and \(X^*\) be \(p^*\)-convex. For \(\tilde{X} \subset X\) a closed convex subset, and given internal energy functionals \(F^1 : \tilde{X} \rightarrow \mathbb{R} \cup \{+\infty\}\) of the full system and \(F^0 : X \rightarrow \mathbb{R} \cup \{+\infty\}\) of the reference system, define the functionals \(F^\lambda, F^\lambda_0 \in \Gamma^0(X)\) and \(E^\lambda, E^\lambda_0 \in -\Gamma^0(X^*), \lambda \in \{0, 1\},\) like in \[37, 38\]. Further, assume \(E^0\) finite everywhere and fix a \(w^* \in X^*\) as external potential. The algorithm then proceeds as follows: Choose an initial guess \(x_0 \in X\) and iterate \(i = 0, 1, 2, \ldots\) according to:

(a) Set \(x_{i+1} = x_i + w^* + \nabla (F_1^0(x_i) - F_0^0(x_i))\) and stop if \(x^*_{i+1} = -\nabla F_0^0(x_i) = z^*\) (KS potential).

(b) Select \(x_{i+1}^* \in \partial E_0^0(x_{i+1}^*)\) and set the iteration direction to \(y_i = (x_{i+1}^* - x_i)/\|x_{i+1}^* - x_i\|\).

(c) Choose the step length \(\tau_i > 0\) as a solution to \((J_p(x_i + \tau_i y_i - \text{prox}_J(x_i)), y_i) = 0\) where \(f_\varepsilon = F_1^1 + w^*\) and set \(x_{i+1} = x_i + \tau_i y_i\).

This algorithm guarantees convergence of \(x_i^* \rightarrow z^* \in X^*\) (KS potential) and \(x_i \rightarrow z \in \partial E_0^0(z^*) \cap \partial E_1^0(w^*)\), which is the ground-state quasidensity of both the reference system with \(z^*\) and the full system with \(w^*\). The ground-state energy is then defined by \(E_1^1(w^*) = F_1^1(z) + \langle w^*, z \rangle\).

Remark 11. Both, \(E_1^1(w^*)\) and the quasidensity \(z \in X\) are still solutions of the regularized problem, but with \[14\] and \[15\] a transformation back to the unregularized setting is easily achieved,

\[
E_1^1(w^*) = E_1^1(z^*) + \varepsilon \phi_{p}(w^*),
\]

\[
z = z + \varepsilon J_p^{-1}(w^*) \in \partial E_0^0(w^*) + \varepsilon J_p^{-1}(w^*).
\]

Here \(z \in \tilde{X}\) is the desired ground-state density of the full system with given external potential \(w^*\). It is guaranteed to be a physical density as an element of \(\partial E_0^0(w^*) \subset \tilde{X}\). If we transform instead to the ground-state density of the reference system using the KS potential, \(z + \varepsilon J_p^{-1}(z^*)\), we reach a different physical density. This is because in this version of the KS algorithm, the link between reference system and full system is implemented via the quasidensitites and not the physical densities.

Proof. First, we demonstrate that the superdifferential of \(E_0^0\) is always non-empty such that step (b) can always be performed. By Lemma 3, \(E_0^0 \in -\Gamma_0^0(X^*) = \Gamma_0^0(X^*)\). The assumed local boundedness below of \(E^0\) also gives \(E_0^0\) locally bounded below by \[14\]. Together this means that \(\partial E_0^0\) is non-empty everywhere by Lemma 3 (i) \(\Rightarrow \) (iii).

We use (a) and \(x_{i+1}^* \in \partial E_0^0(x_{i+1}^*)\) from (b), which is equivalent to \(x_{i+1}^* = -\nabla F_0^0(x_{i+1})\) by Theorem 23 (ii) to write

\[
\langle \nabla F_1^0(x_i) + w^*, x_{i+1}^* - x_i \rangle = \langle x_{i+1}^* + \nabla F_0^0(x_i), x_{i+1} - x_i \rangle = \langle -\nabla F_0^0(x_{i+1}), \nabla F_0^0(x_i), x_{i+1}^* - x_i \rangle.
\]

We now apply Lemma 29 specifically \[31\] to \[33\] with \(x^* = -\nabla F_0^0(x_{i+1}), y^* = -\nabla F_0^0(x_i), x = x_{i+1}, y = x_i\) and the regularized functional \(f_\varepsilon = F_0^1\) (note that \(x \in \partial f_\varepsilon(x^*)\) since here \(f_\varepsilon^* = E_0^0\) and similarly for \(y\) and \(y^*\),

\[
\langle \nabla F_1^0(x_i) + w^*, x_{i+1}^* - x_i \rangle \leq -\varepsilon \| \nabla F_0^0(x_{i+1}) + \nabla F_0^0(x_i) \| + \varepsilon \| \nabla F_0^1(x_i) + w^* \| \leq -\varepsilon \| \nabla F_0^1(x_i) + w^* \|.
\]

The right hand side is strictly smaller than zero unless \(\| \nabla F_1^0(x_i) + w^* \| = 0\), which is equivalent to \(\| x_{i+1}^* + \nabla F_0^0(x_i) \| = 0\) by (a) and means that we have already converged to the ground-state quasidensity. We thus
infer that we always have a negative directional derivative of $F^1_\varepsilon + w^*$ at $x_i$ in the step direction $y_i$ which is parallel to $x_{i+1}' - x_i$.

$$\langle \nabla F^1_\varepsilon(x_i) + w^*, y_i \rangle < 0. \quad (53)$$

Now, the energy functional $F^1_\varepsilon + w^*$ is again a pMY regularization of some $f \in \Gamma_0(X)$ by Lemma 21. This means that the regularization function is aligned tangentially to the (differentiable) energy functional $F^1_\varepsilon + w^*$ at $x_i$, like it is expressed in Lemma 22. Evaluated on the step direction $y_i$ this leads to

$$\langle \nabla F^1_\varepsilon(x_i) + w^*, y_i \rangle = \varepsilon^{-1} \langle \nabla \phi_p(x_i + \tau y_i - p_i), y_i \rangle = \varepsilon^{-1} (J_p(x_i - p_i), y_i) < 0, \quad (54)$$

where we used the abbreviation $p_i = \text{prox}_\varepsilon f(x_i)$. In order to find the step length $\tau_i$, we determine the minimum of the section $\tau \mapsto \varepsilon^{-1} \phi_p(x_i + \tau y_i - p_i)$ through the regularization function, like it is depicted in Fig. 2. Note that only in a Hilbert space setting, like previously considered in Penz et al., this is again a parabola with the same curvature $\varepsilon^{-1}$. Since from $X$ strictly convex it follows $\phi_p$ strictly convex (Lemma 15) and thus trivially also for every section through it, this choice of $\tau_i > 0$ will be unique. This, then, defines the next iteration step $x_{i+1}' = x_i + \tau_i y_i$, where the energy $e_i = F^1_\varepsilon(x_i) + \langle w^*, x_i \rangle$ is always larger than the energy value $m_i$ at the minimum, see Fig. 2. Setting the derivative with respect to $\tau$ of the section to zero gives the condition

$$\frac{d}{d\tau} \phi_p(x_i + \tau y_i - p_i) \bigg|_{\tau = \tau_i} = \langle J_p(x_i + \tau y_i - p_i), y_i \rangle = (J_p(x_{i+1} - p_i), y_i) = 0 \quad (55)$$

for a determination of $\tau_i$. In a general Banach space, where $J_p$ is not simply the identity, we are not able to solve this directly, but we can get an estimate from Lemma 14 by setting $x = x_{i+1} - p_i + \tau_i y_i = x_{i+1} - p_i, y = -\tau_i y_i$,

$$\xi \tau_i^p \leq \|x_i - p_i\|^p - \|x_{i+1} - p_i\|^p + p \tau_i \langle J_p(x_{i+1} - p_i), y_i \rangle \leq \xi' \tau_i^p. \quad (56)$$

Since we can set the term involving $J_p$ to zero for our choice of step length according to (55), this reduces to

$$\xi \tau_i^p \leq \|x_i - p_i\|^p - \|x_{i+1} - p_i\|^p \leq \xi' \tau_i^p. \quad (57)$$

Clearly $\xi \tau_i^p > 0$, so the step from $x_i$ to $x_{i+1}$ always moves closer to the proximal point $p_i$ which would be itself the outcome of a gradient-descent method (see Remark 9). But (57) also gives direct access to the energy difference between the points $x_i$ and $x_{i+1}$ on the regularization function,

$$e_i - m_i = \varepsilon^{-1} \phi_p(x_i - p_i) - \varepsilon^{-1} \phi_p(x_{i+1} - p_i) = \frac{1}{p\varepsilon}(\|x_i - p_i\|^p - \|x_{i+1} - p_i\|^p). \quad (58)$$

Substituting the energy difference, we have

$$\frac{\xi \tau_i^p}{p\varepsilon} \leq e_i - m_i \leq \frac{\xi' \tau_i^p}{p\varepsilon}. \quad (59)$$
Since the section through the regularization function lies fully above the energy functional $F^1 + w^*$ by construction (Remark 7), the energy $\epsilon_{i+1}$ at $x_{i+1}$ must obey $\epsilon_{i+1} \leq m_i < \epsilon_i$. The strictly decreasing sequence $\{\epsilon_i\}$ is now by definition bounded below by $E^2_0(w^*)$ from (11) and thus converges. With the estimate for $\epsilon_i - m_i$ from (59) this gives

$$\frac{\xi \tau^p_i}{p \epsilon} \leq \epsilon_i - m_i \leq \epsilon_i - \epsilon_{i+1} \rightarrow 0,$$

and we can infer convergence of $\tau_i = \|x_{i+1} - x_i\| \rightarrow 0$. This implies convergence in quasidensities $x_i \rightarrow z \in X$. Step (a) then defines an associated potential from a convergent potential sequence, and we can infer convergence of $w^*$ since the gradients are continuous as Fréchet-derivatives (Lemma 22). Convergence is thus secured, like it holds in case of the absolute minima but not along an arbitrary direction.

Remark 12 (Hilbert space setting and erratum to previous article). In a Hilbert space setting where one identifies $X^* = X$ the proof simplifies considerably through $J = \text{id}$ and $p = p^* = 2$. Then (53) can be used together with (54) to solve for $\tau_i$ directly which yields the step length $\tau_i = -\varepsilon(\nabla F^1(x_i) + w^*, y_i)$. Such a version of the proof was given in Penz et al. [4] where also only a finite number of dimensions was considered in order to have boundedness of $X^0$. This last restriction was avoided here with the use of Lemma 9(iv). At this point we would also like to point out a misleading feature of Fig. 1 in [4] where the impression is raised that the minima of the one-dimensional sections through $F^1 + w^*$ and $F^1 + w^*(F + v$ and $F_v + v$ in the notation there) coincide, which holds in case of the absolute minima but not along an arbitrary direction.

3 Our previous proof in Penz et al. [4] relied on the additional assumption that $X^0 \subset X$ is bounded, which could be overcome here by Lemma 9(iv).
Remark 13 (Connection to different reference systems). Instead of the non-interacting system a strongly interacting reference system with $F_{λ}^λ$, $λ = ∞$, could be chosen, which means the proof is directly applicable to the strong-interaction limit of DFT [8, 39]. Other reference points along (or beyond) the “adiabatic connection” $F_{λ}^λ$, $λ ∈ [0, 1]$, can be equally well chosen. Another method that fits the given formalism is “range separated DFT” [57, 58], where a parameter $μ ∈ [0, ∞]$ interpolates between non-interacting and interacting system and in-between a system with only long-range interactions is realized. In any practical application the exact Hxc functional $F_{λ}^λ − F_{0}^λ$ is not attainable, but we might think about some approximate Hxc functional as the gradient difference between the non-interacting $F_{λ}^0$ and some different $F_{λ}^0$ that itself only approximates the interacting system. If indeed $F_{λ}^λ$ is the pMY regularization of a $Γ_0(λ)$ functional (which must not be obvious), then the whole proof works out as presented, just that the ground-state (quasi)density of the approximative system is approached.

V. APPLICATION TO DENSITY-FUNCTIONAL THEORIES

Finally it should be demonstrated that the whole machinery is indeed applicable to common DFT settings, most prominently standard DFT where the system variables are the probability density $ρ$ and the external scalar potential $v$. Firstly, the general structure must be along the lines of Section III with the total energy given by an internal energy term that depends exclusively on the density plus a linear pairing between potential and density. Secondly, all the conditions of Theorem 23 have to be fulfilled, which means the reference energy expression $E^0$ must be finite for all potentials and the Banach spaces of the system variables must be of $p$-convex and $p^*$-convex type. We will check all these properties for the case of standard DFT and paramagnetic CDFT that additionally deals with probability currents and vector potentials.

A. Standard DFT

The standard DFT setting for regularized DFT was previously discussed in Laestadius et al. [2, Section IV.A] and is based on the classical work of Lieb [2]. The state space is that of (quasi)densities $x ∈ X = L^3 ∩ L^1$ on any spatial domain $Ω ⊆ \mathbb{R}^3$ and the dual space is that of scalar potentials $v ∈ X^* = L^{3/2} + L^∞$. Actual probability densities $ρ$ are limited to the space of physical densities

$$\tilde{X} = \{ρ | ρ ≥ 0, ∫ ρ \in H^1, ∥ρ∥_1 = N\} \subset L^3 ∩ L^1,$$

(67)

which is the set of $N$-representable densities with finite kinetic energy that is closed and convex in $X$. The Levy–Lieb functional $\tilde{F}$ is then defined on $\tilde{X}$ as the constrained search for infimal internal energy over all wave functions or density matrices that give a fixed density $ρ ∈ \tilde{X}$, $ψ ↦ ρ$ or $Γ ↦ ρ$. The internal energies are hereby given as the expectation value of the universal part of the usual many-body Hamiltonian,

$$H^λ = -\frac{1}{2} \sum_{i=1}^{N} \nabla_i^2 + λ \sum_{i<j}^{N} \frac{1}{r_{ij}},$$

(68)

$$\tilde{F}^λ(ψ) = \inf_{ψ ↦ ρ} \langle ψ, H^λ ψ \rangle \quad \text{or} \quad \tilde{F}^λ(ρ) = \inf_{Γ ↦ ρ} \text{Tr}(H^λ \Gamma).$$

(69)

Here $λ = 1$ corresponds to the fully interacting system, whereas $λ = 0$ represents the non-interacting reference system. The ground-state energy functional from (67) is then

$$E^λ(v) = \inf_{ρ ∈ \tilde{X}} \left\{ \tilde{F}^λ(ρ) + ∫ v ρ \, dx \right\}.$$  

(70)

The density functionals $F^λ$ and the regularized versions are defined just like in Section III. Yet, the above defined Banach spaces for densities and potentials do not have the necessary properties, most importantly the inclusion of the $L^1 - L^∞$ pair breaks reflexivity and uniform convexity. In order to overcome this we just enlarge the state space to $X = L^3$ with the dual space $X^* = L^{3/2}$ that are both $p$-uniformly convex with $p = 3$ in case of $L^3$ and $p^* = 2$ in case of $L^{3/2}$ (Remark 13). This might first look like dropping the normalizability condition for densities by not restricting them to $L^1$ any more, but note that the space of physical densities $X ⊆ X$ did not change in the process and that the ground-state density from a KS iteration (Theorem 23) is always in $X$. 

Corollary 24 (MYKSODA for standard DFT). Take $X = L^3$ on a spatial domain $\Omega \subseteq \mathbb{R}^3$ as the space of quasidensities and the dual $X^* = L^{3/2}$ for potentials. For the internal energy functionals given by (71) on the set of physical densities $\tilde{X}$ from (67), the MYKSODA algorithm from Theorem 23 is applicable and thus converges.

B. Paramagnetic CDFT

MY regularization of the CDFT case received a separate, thorough treatment in Laestadius et al. [3], where a reflexive Banach space setting was established that generalized a previous first attempt of a convex analysis formulation (without regularization) in Laestadius [40]. Here it must be noted that the state variables are the probability density and the paramagnetic probability current, $(\rho, j)$. One then uses the paramagnetic constrained-search functional of Vignale and Rasolt [41],

$$\tilde{F}^\lambda(\rho, j) = \inf_{\psi \mapsto (\rho, j)} \langle \psi, H^\lambda \psi \rangle$$

$$\tilde{X} = \{ (\rho, j) | \rho \geq 0, \sqrt{\rho} \in H^1, \| \rho \|_1 = N, j \in \tilde{L}^1 \cap \tilde{L}^{3/2}, \| j \|^2/\rho \|_1 < +\infty \}.$$  \hfill (72)

The notation $\tilde{L}^p$ just means that each of the three components of a vector field is in $L^p$, and $\psi \mapsto j$ says that $j$ is the paramagnetic probability current associated with $\psi$. $\tilde{F}^\lambda$ is defined on the pure-state $N$-representable densities $(\rho, j) \in \tilde{X}$ discussed in Lieb and Schrader [42] and Laestadius [40] (the ensemble version can be found in Tellegren et al. [43]). The paramagnetic current must be carefully distinguished from the total (or physical) current $j + \rho A$ that cannot be determined from the wave function $\psi$ alone since it also includes the external vector potential $A$. For more details on the choice of variables in CDFT see Tellegren et al. [31] and Laestadius and Benedicks [32]. The vector potential on the other hand is part of the pair of potentials in the dual space, $(v, A) \in X^*$. The spaces $X, X^*$ are cartesian products of Banach spaces, and a special relation termed compatibility in Laestadius et al. [3] Section 2.4 must hold between their components in order to make a convex formulation possible. In particular, a Hilbert space formulation (as previously employed in the standard DFT setting in Kvaal et al. [44]) cannot be used, since then the new effective potential $v + |A|^2/2$ that ensures convexity of the functionals does not belong to the space of scalar potentials any more. A possible choice of $p$-convex spaces is then

$$X = L^3 \times \tilde{L}^{3/2}, \quad X^* = L^{3/2} \times \tilde{L}^3.$$  \hfill (73)

Finiteness of the ground-state energy functional $E^0$ on this potential space can then be demonstrated [3, Section 2.5]. The choice of function spaces in (73) is motivated by the fact that $\tilde{X} \subset X$, as demonstrated for $\rho$ in Lieb [3] and for $j$ in Laestadius et al. [3].

Corollary 25 (MYKSODA for paramagnetic CDFT). Take $X = L^3 \times \tilde{L}^{3/2}$ on a spatial domain $\Omega \subseteq \mathbb{R}^3$ as the space of quasidensities and currents and the dual $X^* = L^{3/2} \times \tilde{L}^3$ for effective scalar and vector potentials. For the internal energy functionals given by (71) on the set of physical densities $\tilde{X}$ from (72), the MYKSODA algorithm from Theorem 23 is applicable and thus converges.

VI. REGULARIZATION EFFECTS FROM PHYSICAL CONSIDERATIONS

Different physical effects, like coupling to internal magnetic fields [45] or finite temperatures [46, 47], can have an effect analogous to regularization, most importantly the establishment of a Hohenberg–Kohn result. Here we want to go the other direction and derive a possible physical interpretation for pMY regularization from just the chosen Banach space setting. For this, in a standard DFT setting, choose $X = W^{-1,2} = H^{-1}$ on a spatial domain $\Omega \subseteq \mathbb{R}^3$ (distributional; see Adams and Fournier [48, Theorem 3.12] for a definition) and
\( X^* = W_0^{1,2} = H_0^1 \) (zero boundary conditions). The dual pairing between \( f \in X^*, g \in X \) is given in terms of an \( L^2 \)-inner product, \( (f,g) = \int f g \, dx \) in order to match the expression for the potential energy in (70). This potential space \( X^* \) fits exactly to the one considered previously in a formalization of time-dependent DFT [49], where it was chosen this way in order to give unique solutions to a linearized version of the divergence of local-force equation [50]. The zero boundary conditions also do not come completely unexpected. Such a boundary condition fixes the gauge of the scalar potential, since all potentials that differ by only a constant are physically equivalent. On the other hand, a gauge-fixing in order to establish uniqueness of the density-potential mapping is not needed in the regularized DFT formulation, since the differentiability of \( F_e \) already secures this. Moreover, the spaces \( H_0^1, H^{-1} \) are Hilbert spaces and are thus naturally 2-convex as well as 2-smooth.

The usual norm of \( v \in X^* = H_0^1 \) is given by \( \|v\|_{X^*} = (\|v\|_2^2 + \|\nabla v\|_2^2)^{1/2} \) (here \( \nabla \) is the spatial gradient and \( \| \cdot \|_2 \) the \( L^2 \)-norm) but for bounded \( \Omega \) the Friedrichs’s inequality gives an equivalent norm \( \|v\|_{X^*} = \|\nabla v\|_2 \).

Recall Definition 13 for the (inverse) duality map \( J^{-1} : X^* \to X, \)

\[
J^{-1}(v) = \{ g \in X \mid \|g\|_X = \|v\|_{X^*}, (v, g) = \|v\|_{X^*}^2 \}. \tag{74}
\]

This corresponds to the Riesz map since we are in a Hilbert space setting. For the chosen equivalent norm the second condition means \( (v, g) = \int v g \, dx = \|\nabla v\|_2^2 \) which via partial integration is easily fulfilled by \( g = -\Delta v \), the Poisson equation. The norm on the space \( X \) can be defined like an operator norm [48, 3.13],

\[
\|g\|_X = \sup \left\{ \frac{|(u, g)|}{\|u\|_{X^*}} \mid u \in X^* \right\}, \tag{75}
\]

so the first condition \( \|g\|_X = \|v\|_{X^*} \) is also fulfilled for \( g = -\Delta v \), since the supremum will always be reached with \( u = v \) by saturation of the Cauchy–Schwarz inequality. Hence the inverse duality map \( J^{-1} : H_0^1 \to H^{-1} \) (\( \Omega \) bounded) is \( -\Delta \) and [15] translates to

\[
\mathcal{J}E_\epsilon = \mathcal{J}E + \epsilon \Delta, \tag{76}
\]

or, for a choice of \( g \in \mathcal{J}E_\epsilon(v), \rho \in \mathcal{J}E(v) \) of densities, to \( g = \rho + \epsilon \Delta v \). This gives the connection between densities \( \rho \) and their quasidensities \( g \) a physical underpinning: Since \( -\Delta \epsilon v \) is the electrostatic density for a potential \( \epsilon v \) (Poisson’s equation with permittivity \( \epsilon \)), \( g \) is the combined internal and external density. Here we see \( \rho \) as (positive) internal electron density and \( +\epsilon \Delta v \) as (negative) external density creating (minus) the given potential \( -v \). Since both densities have opposite signs, it is clear that \( g \) can be negative and it will also not be normalized to the number of particles. The origin \( g = 0 \) within the space \( X \) is then at \( \rho = -\epsilon \Delta v \), exactly when the density is at equilibrium with the external potential. In this construction, the quasidensity is per se a mixed quantity and the denomination with “quasi” can equally well remind of quasiparticles.

If we decide not to switch to the equivalent norm \( \|\nabla v\|_2 \) (thus allowing \( \Omega \) unbounded), then \( J^{-1} = \id - \Delta \) and \( g = \rho + \epsilon \Delta v - \epsilon v \), so the Poisson-equation connection is lost. But it still remains possible to switch to the equivalent norm \( (\lambda^2 \|\epsilon v\|_2^2 + \|\nabla v\|_2^2)^{1/2} \) and then get the screened Poisson equation \( g = -\Delta (\lambda^2 - 1) v \) as a connection.

ACKNOWLEDGMENTS

We are grateful for helpful discussions with Simen Kvaal, who pointed out the misleading labelling in Penz et al. [4] noted in Remark 12. Erik I. Tellgren, and Michael Ruggenthaler. A.L. has received funding from the Research Council of Norway (RCN) under CoE Grant Nos. 287906 and 262695 (Hylleraas Centre for Quantum Molecular Sciences).
REFERENCES

1. S. Kvaal, U. Ekström, A. M. Teale, and T. Helgaker, J. Chem. Phys. 140, 18A518 (2014).
2. A. Laestadius, M. Penz, E. I. Tellgren, M. Ruggenthaler, S. Kvaal, and T. Helgaker, J. Chem. Phys. 149, 164103 (2018).
3. A. Laestadius, E. I. Tellgren, M. Penz, M. Ruggenthaler, S. Kvaal, and T. Helgaker, J. Chem. Theory Comput. 15, 4603 (2019).
4. M. Penz, A. Laestadius, E. I. Tellgren, and M. Ruggenthaler, Phys. Rev. Lett. 123, 037401 (2019).
5. E. Lieb, Int. J. Quantum Chem. 24, 243 (1983).
6. E. Lammert, Int. J. Quantum Chem. 107, 1943 (2007).
7. M. C. Zerner and M. Hehenberger, Chem. Phys. Lett. 550 (1979).
8. E. Cancès and C. Le Bris, Int. J. Quantum Chem. 14, 230 (1983).
9. P. E. Lammert, Int. J. Quantum Chem. 107, 1943 (2007).
10. M. C. Zerner and M. Hehenberger, Chem. Phys. Lett. 62, 550 (1979).
11. E. Cancès and C. Le Bris, ESAIM Math. Model. Numer. Anal. 34, 749 (2000).
12. A. Laestadius and M. Benedicks, Int. J. Quantum Chem. 114, 782 (2014).
13. H.-K. Xu, Nonlinear Anal. Theory Methods Appl. 16, 1127 (1991).
14. C. Chidume, Geometric Properties of Banach Spaces and Nonlinear Iterations, Vol. 1965 (Springer, 2009).
15. K. S. Kazmiernski, Aspects of Regularization in Banach Spaces (Logos Verlag Berlin GmbH, 2010).
16. G. Pisier, Isr. J. Math. 20, 326 (1975).
17. B. Prus and R. Smarzewski, J. Math. Anal. Appl. 121, 10 (1987).
18. E. I. Tellgren, A. Laestadius, T. Helgaker, S. Kvaal, and A. M. Teale, J. Chem. Phys. 148, 024101 (2018).
19. K. Capelle and G. Vignale, Phys. Rev. Lett. 86, 5546 (2001).
20. K. Capelle and G. Vignale, Phys. Rev. A 65, 113106 (2002).
21. K. Capelle, C. A. Ullrich, and G. Vignale, Phys. Rev. A 76, 012508 (2007).
22. E. I. Tellgren, S. Kvaal, E. Sagvolden, U. Ekström, A. M. Teale, and T. Helgaker, Phys. Rev. A 86, 062506 (2012).
23. A. Laestadius and M. Benedicks, Int. J. Quantum Chem. 114, 782 (2014).
24. E. Lammert, arXiv:1807.06125.
25. Engel and R. M. Dreizler, Density Functional Theory: An Advanced Course (Springer, 2011).
26. M. Reed and B. Simon, II: Fourier Analysis, Self-adjointness, Vol. 2 (Elsevier, 1975).
27. M. Reed and B. Simon, II: Fourier Analysis, Self-adjointness, Vol. 2 (Elsevier, 1975).
28. A. Laestadius, Int. J. Quantum Chem. 14, 1445 (2014).
29. G. Vignale and M. Rasolt, Phys. Rev. B 37, 10685 (1988).
30. E. Lieb and R. Schrader, Phys. Rev. A 88, 032516 (2013).
31. E. I. Tellgren, S. Kvaal, and T. Helgaker, Phys. Rev. A 89, 012515 (2014).
32. E. I. Tellgren, Phys. Rev. A 97, 012504 (2018).
33. J. Chayes, L. Chayes, and M. B. Ruskai, J. Stat. Phys. 38, 497 (1985).
34. K. J. Giesbertz and M. Ruggenthaler, Phys. Rep. 806, 1 (2019).
35. L. Garrigue, J. Stat. Phys. 177, 415 (2019).
36. R. A. Adams and J. J. Fournier, Sobolev Spaces (Elsevier, 2003).
37. M. Penz and M. Ruggenthaler, J. Phys. A Math. Theor. 44, 335208 (2011).
38. M. Ruggenthaler, M. Penz, and R. Van Leeuwen, J. Condens. Matter Phys. 27, 203202 (2015).