Coarse-Grained V-Representability

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The unsolved problem of determining which densities are ground state densities of an interacting electron system in some external potential is important to the foundations of density functional theory. A coarse-grained version of this ensemble V-representability problem is shown to be thoroughly tractable. Averaging the density of an interacting electron system over the cells of a regular partition of space produces a coarse-grained density. It is proved that every strictly positive coarse-grained density is coarse-grained ensemble V-representable: there is a unique potential, constant over each cell of the partition, which has a ground state with the prescribed coarse-grained density. For a system confined to a box, the (coarse-grained) Lieb functional is also shown to be Gâteaux differentiable. All results extend to open systems.

1. INTRODUCTION

Four decades ago, Hohenberg and Kohn launched modern density functional theory with their famous theorem\cite{HK}, stating that a density function for a system of interacting particles can be a ground state density of at most one external potential. A density is said to be \textit{ensemble V-representable} (EV-representable) \textit{if there is a mixed ground state (i.e., a density matrix) for some potential which has that density}. The V-representability problem, of determining which densities are EV-representable not only remains unresolved to this day, but has seen almost no progress.

Optimistically, one might have hoped that any density which can arise from a state with finite energy is EV-representable. But there are known counterexamples\cite{KCR, P} based on nonanalyticity. It is also easy to construct densities, by modifying the short wavelength parts, which can come only from states with infinite kinetic energy, and thus certainly cannot be ground state densities. These observations point to the potential usefulness of suppressing short length-scale degrees of freedom, or otherwise rendering them innocuous. Formulating the theory on a lattice achieves that by eliminating short wavelengths. The lattice program was initiated by Kohn\cite{K} and Chayes, Chayes and Ruskai\cite{CCR} later proved that all densities on a lattice are ensemble V-representable\cite{KCR, P}.

Unfortunately, working on a lattice fundamentally alters the problem, and it is difficult to relate the results to standard continuum quantum mechanics. In this paper, I propose a way to make the short length-scale degrees of freedom harmless without altering the underlying quantum mechanics. Instead, by coarse-graining, we impose a bound on the precision with which we permit ourselves to specify a desired density. Specifically, we divide our space, either all of $\mathbb{R}^3$ or a parallelepiped, into a regular array of parallelepipeds cells of volume $\Omega$. The cells are labelled $B_1, B_2, \ldots$ (a countable infinity), or $B_1, \ldots, B_M$, according to the case. Denoting the operator corresponding to the number of particles in cell $B_k$ by $N_k$, we get a coarse-grained density operator $\hat{\rho}$, with components $\hat{\rho}_k = N_k/\Omega$.

Now we can ask about states of our system of particles which have a specified coarse-grained density $\rho$. In particular, we want to know whether there is a potential which has a (mixed) ground state with coarse-grained density $\rho$. If so, $\rho$ is said to be \textit{coarse-grained EV-representable} (cg-EV-representable). As will be shown, \textit{all} coarse-grained densities, except those which put zero particles in one or more cells, are coarse-grained EV-representable. Moreover, among the potentials which are constant on each cell, there is one and only one having a ground-state coarse-grained density $\rho$. The proof of coarse-grained EV-representability is the central result of this paper. It should be clear, but bears emphasizing, that among all the states yielding the coarse-grained density $\rho$, the one which is guaranteed to be V-representable by this result minimizes the kinetic-plus-interaction energy. In the case of a system of particles confined to a box, we show that $F_L$, viewed as a function on the affine hull of the set where it is finite, is Gâteaux differentiable. In what sense $F_L$ can be differentiable at EV-representable densities is a question of continuing interest\cite{P}. In the unconfined case, $F_L$ is not Gâteaux differentiable anywhere, at least in the ordinary sense.

Coarse-graining allows short length-scale degrees of freedom to do as they like, without artificially removing them. Arguably, it is also more harmonious with practical computational limitations than the traditional approach.

In the standard (fine-grained) theory\cite{Lieb}, it is known that the densities which are EV-representable are dense in the effective domain of $F_L$ (see Thm. 3.13 of Lieb\cite{Lieb} or Sec. I.6 of Ekeland and Témam\cite{ET}). The usefulness of this observation is limited by the absence of any criterion to pick out the good densities, and the fact (relevant for Kohn-Sham theory\cite{KS}) that they may depend on the particle interaction. Within its scope, the coarse-grained theory does not suffer from these problems. One might suspect that the coarse-grained EV-representability statements follow trivially from the ex-
istence of that dense set of densities, but it is not so be-
cause the set of fine-grained densities belonging to a given
coarse-grained density does not contain any $L^1$ neigh-
borhood.

The body of this paper unfolds as follows. Section 2
describes, in a rough way, the main ideas involved and
the problems to be grappled with. Details of the confined
case (in a box) are given in Section 3. Technically, this
case is much easier than the unconfined case. Thus, al-
though it stands on its own, it also serves as a warm-up
exercise for the unconfined case, which is treated in Sec-
ction 4. In Section 5, the extension to open systems with
indefinite total particle number is discussed, in somewhat
less detail. All the results carry over, and the requisite
modifications of the proofs are sketched. Finally, in sec-
ction 6 the results are put into perspective and directions
for future work indicated.

In the body of this paper, the word density without
modifier will carry the usual meaning, namely, “fine-
grained density.” Frequently, coarse-grained will be
abbreviated as cg. Coarse-grained density and number are
usually expectation values; when used in their operator
aspect, a caret will be added (\( \hat{\rho} \)) to help avoid con-
fusion. This practice is not extended to other operators,
for which ambiguity is not a danger.

2. MAIN IDEAS

This section is an impressionistic sketch of the main
ideas involved in the technical work of the following three
sections, concentrating on the unconfined case with fixed
particle number. As described in the introduction, we
cover $\mathbb{R}^3$ with identical cells. All (fine-grained) densi-
ties with the same average density in each cell ($\rho_i$ in $B_i$)
are gathered together into an equivalence class called a
coarse-grained density. A cg-density $\rho = (\rho_1, \rho_2, \ldots)$ sat-
sifies $\rho_i \geq 0$ and $\sum \rho_i = N$. Dual to the cg-densities,
we introduce a space $V$ of external potentials which are
constant on each cell; other restrictions will be imposed
later. For such a potential, the integral $\int \nu \rho \, dx$ depends
only on the coarse-grained class of the density $\rho$.

The Lieb functional is a functional of coarse-grained
density which represents the minimal intrinsic energy
(kinetic plus Coulomb interaction) consistent with the
specified cg-density:

$$F_{L}(\rho) = \inf \{ \text{Tr} \, \Gamma H_0 : \Gamma \to \rho \},$$

where $H_0$ is the kinetic-plus-interaction Hamiltonian and
the customary notation $\Gamma \to \rho$ indicates that mixed state
\( \Gamma \) has cg-density $\rho$. $F_{L}$ is not defined as a minimum be-
cause it is not obvious that there is any state which re-
alizes the infimum; finite-dimensional intuition is a bad
guide here. But only a state with minimal intrinsic energy
in its class can possibly be a ground state for a poten-
tial from $V$, so it is crucial for cg-EV-representability that we show $F_{L}$ really is a minimum.

To illustrate the kind of problem which can arise in
ensuring the existence of a minimum in infinite-
dimensional situations, consider trying to minimize the
function $f(\psi) = \sum_{n=1}^{\infty} (1/n) |\langle \psi | \phi_n \rangle|^2$ over the unit vec-
tors in a Hilbert space, where $\{ \phi_n \}$ is an orthonormal
basis. No matter how small $\epsilon > 0$, $S_\epsilon = \{ f < \epsilon \}$ is
infinite-dimensional (and yet $\cap S_\epsilon$ is empty). Absent de-
tailed knowledge about the intrinsic energy, the way to
show that the Levy search implicit in the definition of $F_{L}$ has a solution is to show that the search is “al-
most finite-dimensional.” For $F_{L}$, the gist of the argu-
ment may be phrased semiclassically: Rough knowledge
about localization of the density approximately confines
the search to a bounded region of configuration space,
while considering only states with relatively low intrin-
sic energy gives localization in momentum. The two to-
gether approximately confine the search to a bounded
volume of phase space. Since a volume $2\pi \hbar^3 N$ of phase
space corresponds to one dimension in Hilbert space, the
search is almost-finite-dimensional.

The ground state energy $E(\nu)$ of $\nu \in V$ is the lower
bound on energies of states in the presence of $\nu$, which
can be expressed as

$$E(\nu) = \inf_{\rho} \left\{ F_{L}(\rho) + \int v \rho \, dx \right\}.$$ (Note that, despite the name, $E(\nu)$ may fail to be at-
tained, as for the potential which is everywhere zero.)
This equation shows that $E$ is the Legendre transform of
$F_{L}$. It also shows that, for given $\rho$,

$$F(\rho) + \int v \rho \, dx \geq E(\nu)$$

holds for all $\nu$. Now, if the cg-density $\rho$ is cg-EV-
representable, then this inequality must be an equality
for the realizing potential, and conversely, if it is equality
for some $\nu$, then $\rho$ is clearly cg-EV-representable.

So, the task is to show that equality is achieved in in-
equality (1). To that end, more structure is useful. We
embed the cg-densities in the vector space $X = \ell^1$ of real
sequences $\rho$ such that $\| \rho \| = \sum |\rho_i| < \infty$, which is a Ba-
anch space equipped with the norm $\| \rho \|$. Then we declare
that we consider only potentials in $X^*$, the topological
dual of $X$, which consists of $\nu$ which are bounded. The
double Legendre transform of $F_{L}$ is its lower semicontin-
uous convex envelope. Therefore, by showing that $F_{L}$ is
lower semicontinuous and convex, we establish that $F_{L}$
is (up to signs) the Legendre transform of $E$:

$$F(\rho) = \sup_{\nu} \left\{ E(\nu) - \int v \rho \, dx \right\}.$$ Convexity is immediate from the definition, but lower
semicontinuity requires some work.

In the case of a system confined to a box, $X$ and $X^*$
are finite-dimensional. In that case, existence of a po-
tential which gives equality in Eq. (1) is an automatic
consequence of the mutual Legendre transform relation of $E$ and $F_L$.

The unconfined case is harder. We attack it by noting that the second Legendre transform relation says that equality in Eq. (1) can be approached as closely as desired. That is, a sequence of $v \in X^*$ can be found such that

$$E(v_n) - \int v_n \rho \, dx \to F(\rho).$$

The trick is then to show that this sequence $v_n$ converges in some sense and that the limit $v_\infty$ satisfies equality in (1). In carrying that out, it is necessary, in general, to locate the limit outside of $X^*$ entirely. This may seem surprising, at first, but was entirely to be expected. After all, the harmonic oscillator ground state is $v$-representable, but the harmonic-oscillator potential is not bounded, hence not in $X^*$.

This, in brief, is how cg-EV-representability is established. We also prove a coarse-grained version of the Hohenberg-Kohn Theorem, with only a little more work than for the usual version. With that, we can see that, not only is every everywhere-positive $\rho$ cg-EV-representable, but that the representing potential is unique.

3. CONFINED SYSTEMS

In this section, we treat confined systems. Standard tools of convex analysis will be used, without much comment apart from reminders of definitions. The reader for whom the material is unfamiliar can find it in a number of textbooks, for example, chapter I of Ekeland and Témam\[9\], or van Tiel\[11\]. Rockafellar\[12\] treats the unconfined case is harder. We attack it by noting that the total interaction is nonnegative. The Hamiltonian $H_0$ includes kinetic energy and the interaction between the electrons, with periodic, Dirichlet or Neumann boundary conditions. We assume the interaction is the repulsive Coulomb interaction, possibly modified by a bounded interaction, such as would be required to adapt it to periodic boundary conditions. The bounded addition may even be a multiparticle interaction, but we assume it has a constant adjusted so that the total interaction is nonnegative. $H_0$ is then unbounded, but bounded below by zero.

There is a lower limit to the kinetic plus interaction energy of states having a given cg-density, the Lieb functional

$$F_L(\rho) := \inf \{ \text{Tr} \, \Gamma H_0 \geq \rho \in S(\rho) \}. \quad (4)$$

If $S(\rho)$ is empty, $F_L(\rho) = \infty$. It may appear at first sight that the trace in the definition is potentially not well-defined, since $H_0$ is unbounded. However, since $H_0 > 0$ and $\Gamma$ is a positive operator, there is no real difficulty. We can, for example, replace $H_0$ by $H_0 P H_0$, where $P H_0$ is the spectral projection onto $H_0 \leq \lambda$, and take the limit $\lambda \to \infty$. The set of states which realize the infimum in Eq. (4) is denoted $S(\rho)$:

$$S(\rho) := \{ \Gamma \in S(\rho) \geq \text{Tr} \, \Gamma H_0 = F_L(\rho) \}. \quad (5)$$

We will show later that this set is nonempty. $F_L$ is bounded both above and below: $0 < F_L \leq F_L^{\text{max}}$. The lower bound is simple, and the upper bound follows by using the fact (N-representability) that a smooth density with any desired cg-density can be realized by an appropriate mixed state, and then using the bounds on $F_L$ in terms of $\nabla \rho$ as derived in Lieb\[3\] (see Thms. 3.8 and 3.9 there). The largest of these (finite) bounds is for the case of all particles in a single cell, so $F_L$ is bounded above.

Having dealt with densities, we now turn to potentials. The potentials we want to consider take the constant bounded, but bounded below by zero.

$\ldots$
value $v_i$ on cell $B_i$, information which is encoded in the vector $v = (v_1, \ldots, v_M) \in \mathbb{R}^M$. We will write $V$ for this space of potentials. Adding the potential $v$ to $H_0$ gives

$$H(v) = H_0 + \sum_{i=1}^{M} \Omega v_i \hat{\rho}_i.$$  

When convenient, we use the alternate, and suggestive, notations $\int v \hat{\rho}(x) \, dx$ or $v \cdot \hat{\rho}$ for the potential term.

The ground state energy for potential $v \in V$ is

$$E(v) := \inf \{ \text{Tr} \, \Gamma H(v) : \Gamma \in S \}.$$  

This really is the energy of some state; the infimum is sure to be realized in the present setting, as follows from the fact that our system is in a box (cf. Thm. 3.2). We can rewrite $E(v)$ in terms of the Levy constrained search as

$$E(v) = \inf_{\rho} \{ F_L(\rho) + v \cdot \rho \}$$  

That is, $-E(-v)$ is the Legendre-Fenchel transform (also known as convex conjugate, conjugate, or polar) of $F_L(\rho)$. Thus, $E(v)$ is concave and upper semicontinuous where it is finite. But, the infimum in Eq. (7) is certain to lie between $\min \{v_i\}$ and $F_L^{\max} + \max \{v_i\}$, so that $E(v)$ is finite everywhere on $\mathbb{R}^M$, and therefore continuous. The Legendre-Fenchel transform of $-E(-v)$ is the closed convex hull of $F_L$. $F_L$ is easily seen to be convex from its definition, so continuity on $X_N^+$ is automatic because of finite dimensionality (ET: Cor. I.2.3, VT: Thm. 5.23, R: Thm. 10.1), but lower semicontinuity at the relative boundary of $X_N^+$ is not. In fact, $F_L$ is lower semicontinuous there as well. Rather than provide the proof (see Thm. 4.3), we will here simply assume that $F_L$ is defined on the relative boundary so that it is lower semicontinuous. This is harmless because the cg-densities there are not of interest. We summarize the foregoing discussion.

**Proposition 3.1 (convex duality).** The functions $F_L(\rho)$ and $E(v)$ on $\mathbb{R}^M$ are in duality, i.e., they are (up to signs) Legendre transforms of each other. Hence, $F_L$ and $-E$ are convex and lower semicontinuous.

One aspect of this relationship between $F_L$ and $E$ which will be important later is that $v \in V$ is regarded as a linear functional on $\mathbb{R}^M$ via $\rho \mapsto \int v \rho \, dx$. Since $\mathbb{R}^M$ is finite-dimensional, $v$ is automatically a continuous functional (though $v(x)$ is not a continuous function of $x$ unless it is constant).

Now, we are ready for a coarse-grained version of the Hohenberg-Kohn theorem. If potentials $v$ and $v'$ differ by an overall constant, write $v \sim v'$, and denote the equivalence class of $v$ in $V/\mathbb{R}$ by $[v]$.

**Theorem 3.1 (cg Hohenberg-Kohn).** Given $S(\rho)$, there is at most one equivalence class in $V/\mathbb{R}$ which has a ground state in $S(\rho)$.

**Proof.** First, $v \sim v'$ implies that $H(v)$ and $H(v')$ have disjoint ground state manifolds in $\mathcal{H}$. This is a result about partial differential equations and is proven just as for the conventional Hohenberg-Kohn theorem, being just a specialization to potentials in $V$.

If $v \in V$ has a ground state in $S(\rho)$, then that state is in $S(\rho)$, and all elements of $S(\rho)$ are ground states for $v$, since all such states have the same value of Tr $H_0 \Gamma$. Thus, if both $v$ and $v'$ have ground states in $S(\rho)$, then they share a ground state, which contradicts the previous paragraph.

This proof carries over to the unconfined setting of Section 4 and the Fock space setting of Section 5, with no essential changes.

As just observed, the only states in $S(\rho)$ which possibly can be ground states are those in $S(\rho)$. The next task is therefore to show that $S(\rho)$ is nonempty.

**Theorem 3.2 (Existence of Levy search solutions).** For every $\rho \in X_N^+$, $S(\rho) \neq \emptyset$. That is, there is a normalized mixed state $\Gamma$ such that $F(\Gamma) = \text{Tr} \, H_0 \Gamma$.

We prove this by means of a couple of lemmas. It will be useful to write the intrinsic energy in a functional notation:

$$E(\Gamma) := \text{Tr} \, \Gamma H_0,$$  

regarded as a function on $S$, so that $E^{-1}(0, M]$ consists of normalized mixed states with intrinsic energy not exceeding $M$.

**Lemma 3.1.** $E$ is lower semicontinuous with respect to trace norm on $S$.

**Proof.** Suppose $\Gamma_n \in E^{-1}(0, M]$ for $n = 1, 2, \ldots$ and $\Gamma_n \rightarrow \Gamma$ in trace norm. We need to show that $E(\Gamma) \leq M$. Suppose instead that $E(\Gamma) > M$.

If $P_{H_0}(\epsilon)$ denotes the spectral projection onto $H_0 \leq \epsilon$, then $\Gamma \text{Tr} \, H_0 P_{H_0}(\epsilon)$ is monotonic in $\epsilon$ so that there is some finite $\epsilon$ for which $\text{Tr} \, H_0 P_{H_0}(\epsilon) > M$. But, trace-norm convergence implies weak convergence and $H_0 P_{H_0}(\epsilon)$ is a bounded operator. Therefore, $\text{Tr} \, \Gamma_n H_0 P_{H_0}(\epsilon)$ exceeds $M$ for large enough $n$ since it converges to $\text{Tr} \, H_0 P_{H_0}(\epsilon)$. This, however, is impossible because $M \geq E(\Gamma_n) \geq \text{Tr} \, \Gamma_n H_0 P_{H_0}(\epsilon)$.

This result is very general, depending as it does only on the positivity of mixed states and the semiboundedness of the Hamiltonian. The theorem and proof apply to the unconfined case, and even in Fock space.

For the next lemma, we recall the concept of total boundedness. A set in a metric space is called totally bounded if, given $\epsilon > 0$, it can be covered by a finite number of balls of radius $\epsilon$. For a subset of a complete metric space, total boundedness is equivalent to relative compactness, i.e., having a compact closure. A Banach space is a complete metric space.

**Lemma 3.2.** $E^{-1}(0, M]$ for $M < \infty$ is relatively compact with respect to trace norm.
Proof. $\mathcal{E}^{-1}(0, M]$ is certainly bounded since it is contained in the unit ball of $L_1^+(\mathcal{H})$. The strategy is to show that, given $\varepsilon$, there is a finite dimensional space $W$, such that all points of $\mathcal{E}^{-1}(0, M]$ are within distance $\varepsilon$ of $W$. Since a bounded subset of $W$ is totally bounded and $\varepsilon$ is arbitrary, this will show that $\mathcal{E}^{-1}(0, M]$ is totally bounded, hence relatively compact.

The interaction energy is nonnegative, so $\mathcal{E}^{-1}(0, M]$ can only get bigger if we drop the interaction. Thus, it suffices to prove the lemma for the case that $H_0 = T$ contains only kinetic energy.

Due to our assumptions, the single-particle eigenstates $\varphi_m$ of $T$ can be written down explicitly, and the spectrum is certainly discrete. Up to energy $M/\varepsilon$, there are only a finite number, $\varphi_1, \ldots, \varphi_K$, and the set of Slater determinants made from these likewise span a finite-dimensional space, $W$.

Now, any state $\Gamma$ can be written as a linear combination of Slater determinants of the $\varphi_m$. But, if $\mathcal{E}(\Gamma) \leq M$, the terms containing $\varphi_m$ for $m > K$ must, all together, have norm less than $\varepsilon$. That is, $\Gamma$ is the sum of something in $W$ and something with norm less than $\varepsilon$.

The box played a crucial role in this proof; its counterpart in the unconfined case will be much more difficult.

Proof. (of Theorem 3.2) Since the $\hat{N}_i$ are bounded operators, the condition $\text{Tr} \Gamma \mathcal{N} = \mathcal{N}$ defines a closed linear subspace of $L_1^+(\mathcal{H})$. The intersection of this subspace with $\mathcal{E}^{-1}(0, M]$ is therefore compact. $\mathcal{E}$ is lower semicontinuous by (Lemma 3.1). Since a lower semicontinuous function on a compact space is guaranteed to have a minimizer, the theorem is proven.

A cg-density $\rho_0$ is cg-EV-representable if and only if some $\Gamma \in \mathcal{S}(\rho_0)$ is a ground state of some $\psi \in \mathcal{V}$. Since Theorem 3.2 shows that $\mathcal{S}(\rho_0)$ is nonempty, the condition for cg-EV-representability is existence of $\psi \in \mathcal{V}$ such that $F_L(\rho_0) + \int v \rho_0 dx \leq F_L(\rho) + \int v \rho dx$, for all $\rho$. This condition rearranges to the statement that the hyperplane $\rho \mapsto F_L(\rho_0) + \int v(\rho - \rho_0) dx$ lies below the graph of $F_L$ and touches it at $\rho_0$. That is, $\psi$ is a tangent functional to $F_L$ at $\rho_0$ and, since it is continuous, a subgradient of $F_L$ at $\rho_0$. Recall that the set of all subgradients at $\rho$ [denoted $\partial F_L(\rho)$], is called the subdifferential of $F_L$ at $\rho$, and $F_L$ is said to be subdifferentiable at $\rho$ if $\partial F_L(\rho) \neq \emptyset$. It is a basic fact (ET: Prop. 1.5.2, vT: Thm. 5.35) that a convex function on a finite-dimensional space is subdifferentiable everywhere on the relative interior of its effective domain. This observation completes the proof of the central result of this section:

Theorem 3.3 (Ensemble $\psi$-representability). Every $\rho \in \mathcal{X}_N^+$ is cg-EV-representable.

Proof. See preceding discussion.

Combining this theorem with the cg-Hohenberg-Kohn Theorem gives Gâteaux differentiability of the restriction of $F_L$ to $\mathcal{X}_N$.

Theorem 3.4 (Differentiability of $F_L|_{\mathcal{X}_N}$). $F_L$, as a function on $\mathcal{X}_N$, is Gâteaux differentiable on $\mathcal{X}_N^+$.

Proof. Given $\rho \in \mathcal{X}_N^+$, Thm. 3.3 says that there is at least one $\psi \in \mathcal{V}$ which represents $\rho$. On the other hand, Theorem 3.1 asserts that there is at most one. So, there is precisely one. Uniqueness of the subgradient at $\rho$ together with local boundedness of $F_L$ shows (ET: Prop. 1.5.3) that $F_L|_{\mathcal{X}_N}$ is Gâteaux differentiable at $\rho$.

Note in passing that we have no comparable theorem for $E(\psi)$. If $\psi$ has distinct ground states which lie in different strata of $\mathcal{S}$, then $E$ is certainly not Gâteaux differentiable there.

4. UNCONFINED SYSTEMS

Of course, the box used in the previous section is a rather artificial element, so it is desirable to prove analogous results with all space accessible to the particles. We refer to this as the unconfined case, and in this section we will show cg-EV-representability of all normalized, strictly positive, cg-densities for the unconfined case.

Similarly to before, $\mathbb{R}^3$ is partitioned into a regular, countably infinite, array of cells $B_1, B_2, \ldots$, each of volume $\Omega$. This time, the regularity serves the important purpose of ensuring uniform upper and lower bounds on the cell sizes. As in Section 3 $S$ denotes the set of normalized mixed states, $\mathcal{X}_N^+$ the set of normalized cg-densities, and $\mathcal{X}_N^{++}$ those which are nowhere zero. Now, however, $\hat{\chi}$ is $\ell^1$, the space of sequences $(\rho_1, \rho_2, \ldots)$ such that $\|\rho\| = \sum |\rho_i| < \infty$. This space is a Banach space when equipped with the norm $\|\rho\|$. The Lieb functional is defined as before, and has the same upper bound $F_L^{max}$ on $\mathcal{X}_N^{++}$, outside of which it is $+\infty$. $\mathcal{X}_N^+$ is contained in the closed subspace $\mathcal{X}_N = \{\rho \in \ell^1 : \Omega \sum \rho_i = N\}$; however, the interior of $\mathcal{X}_N^+$ relative to $\mathcal{X}_N$ is empty, not $\mathcal{X}_N^{++}$.

Naturally, we then consider potentials in the dual space to $\ell^1$, which is the space $\ell^\infty$ of sequences $(v_1, v_2, \ldots)$ which are bounded: $\sup |v_i| < \infty$. However, we shall eventually find ourselves looking for potentials in the larger set

$$\mathcal{V} = \{v = (v_1, v_2, \ldots) : \inf v_i > -\infty\},$$

of potentials which are required only to be bounded below. Note that the cg-Hohenberg-Kohn Theorem 3.1 works for potentials in $\mathcal{V}$.

From the experience of the box case, one might expect the work here to split into two principal parts: proof of the existence of Levy search solutions, by demonstration that $F_L$ is lower semicontinuous and that the search is taking place in a compact space, followed by proof of subdifferentiability using general results from the duality theory of convex analysis. The first expectation is correct. Lemma 3.1 goes through unaltered, but Lemma 3.2 works for potentials in $\mathcal{V}$. Theorem 3.3 goes through unaltered, but Theorem 3.4 requires some adjustments.
will require a more sophisticated counterpart. The run-up to Theorem 4.2 uses some ideas from Lieb\cite{16} (particularly Thm. 4.4 there), but the proof given here is self-contained. However, what convex analysis gives us in this case is limited to showing that $F_{\epsilon}(\rho)$ and $E(\upsilon)$ are a Legendre-Fenchel transform pair (up to signs). Establishing existence of a potential $\upsilon$ which has ground state with density $\rho \in L_{\infty}^\ast$ takes an ad hoc argument following the pattern of the proof by Chayes, Chayes and Ruskai\cite{18} of EV-representability on a lattice. The basic reason for this is that some of the potentials needed do not lie in the dual space to $l^1$. In fact, they are not even linear functionals, because they take the value $+\infty$ on some densities.

The immediate goal is the general-purpose Thm. 4.1 below, which deals with (fine-grained) densities. Call a set $\Delta$ of densities tight if, given $\epsilon > 0$, there exists a bounded set $\Delta$, such that no $\rho \in \Delta$, puts more than $\epsilon$ into $l^1$. In fact, they are not even linear functionals, because they take the value $+\infty$ on some densities.

Some notation that will be used is gathered here: For a bounded region, denote the Hilbert space projection corresponding to all particles being in $\Lambda$ by $P_\Lambda$. $H_0 + 1$ is bounded below by 1, and therefore has a unique positive square root denoted by $h$ which is also bounded below by 1. $h^{-1}$ is a bounded operator.

The next lemma is key. It is followed by three auxiliary results, the proofs of which can be skipped without detriment to understanding of the main results.

**Lemma 4.1.** For $\Lambda$ a bounded region of space, $P_\Lambda \mathcal{E}^{-1}(0, M)P_\Lambda$ is compact.

**Proof.** Let $(P_\Lambda \Gamma_n P_\Lambda)_{n=1}^\infty$ be a bounded sequence in $P_\Lambda \mathcal{E}^{-1}(0, M)P_\Lambda$. We will show that there is a norm convergent subsequence. Since $\Gamma_n \in \mathcal{E}^{-1}(0, M)$, $\gamma_n = h \Gamma_n h$ is also a bounded sequence in $P_\Lambda \mathcal{L}^+(\mathcal{H})$. Now, $\mathcal{L}^+(\mathcal{H})$ is the topological dual to the space of compact operators on $\mathcal{H}$, and any bounded sequence in $\mathcal{L}_1(\mathcal{H})$, in particular $(\gamma_n)$, has a weak-* convergent subsequence. [This is a slightly subtle point. It requires both the Banach-Alaoglu theorem and the fact that since $\mathcal{H}$ is separable, the space of compact operators on $\mathcal{H}$ is also separable. See, e.g., Thm. 3.17 of Rudin\cite{4}]

By relabeling, write the weak-* convergent subsequence again as $\gamma_n$, so for any compact operator $A$, $\operatorname{Tr} \gamma_n A \to \operatorname{Tr} \gamma A$, where $\gamma$ is the limit. But, the Hamiltonian $H_0$ is locally compact\cite{13}, i.e., $P_\Lambda h^{-1}$ and $h^{-1}P_\Lambda$ are compact operators (Lemma 4.4). Since the product of a bounded operator and a compact operator is compact, for any bounded operator $B$, $\operatorname{Tr} BP_\Lambda \Gamma_n P_\Lambda = \operatorname{Tr} \{(BP_\Lambda h^{-1})\gamma_n(h^{-1}P_\Lambda)\} \to \operatorname{Tr} BP_\Lambda h^{-1} \gamma h^{-1}P_\Lambda$. Defining $\Gamma := h^{-1} \gamma h^{-1}$, this says $P_\Lambda \Gamma_n P_\Lambda \to P_\Lambda \Gamma P_\Lambda$ with respect to the weak topology (not weak-*!). Now apply Lemma 4.3 to deduce $P_\Lambda \Gamma_n P_\Lambda \to P_\Lambda \Gamma P_\Lambda$. So, $P_\Lambda \mathcal{E}^{-1}(0, M)P_\Lambda$ is relatively compact; but it is also closed, by Lemma 4.1.

**Lemma 4.2.** For $\Gamma \in \mathcal{L}_1^+(\mathcal{H})$ and $P$ an orthogonal projection on $\mathcal{H}$, if $\|PTP\|_{tr} > (1 - \epsilon^2/9)\|\Gamma\|_{tr}$, and $\epsilon < 3$, then $\|\Gamma - PTP\|_{tr} < \epsilon\|\Gamma\|_{tr}$.

**Proof.** Without loss, assume $\Gamma$ is normalized, so $\Gamma = \sum c_m|\phi_m\rangle\langle\phi_m|$, where $c_m > 0$, $\sum c_m = 1$ and $\phi_m$ are unit vectors. Write $\Gamma = P\Gamma P + P\Gamma P\perp + P\perp P\Gamma P + P\perp P\perp$, with $P\perp = 1 - P$, and bound the last three terms in trace norm as follows.

$$\|P\Gamma P\perp\|_{tr} = Tr P\Gamma P\perp = 1 - Tr PTP < \epsilon^2/9,$$

where the final inequality is by hypothesis. Using the triangle inequality followed by the Cauchy-Schwarz inequality and then the above bound on $\|P\perp\Gamma P\perp\|_{tr}$, we get $\|PTP\|_{tr} = \|P\perp\Gamma P\perp\|_{tr} \leq \sum c_m\|P\perp\phi_m\|\|P\phi_m\| \leq (\sum c_m\|P\perp\phi_m\|^2)^{1/2}\sum c_m\|P\phi_m\|^2)^{1/2} \leq \epsilon/3$. Gathering the pieces, and using $\epsilon < 3$, gives the result.

**Lemma 4.3.** If $(\Gamma_{\alpha})_{\alpha=1}^\infty$ is a sequence in $\mathcal{L}_1^+(\mathcal{H})$, then $\Gamma_{\alpha} \overset{w}{\to} \Gamma$ if and only if $\Gamma_{\alpha} \overset{tr}{\to} \Gamma$, and in that case $\Gamma \in \mathcal{L}_1^+(\mathcal{H})$.

**Proof.** The implication from norm convergence to weak convergence is trivial, so suppose $\Gamma_{\alpha} \to \Gamma$ weakly. The cone $\mathcal{L}_1^+(\mathcal{H}) \subset \mathcal{L}_1(\mathcal{H})$ is norm closed and convex, hence weakly closed. Thus, any weak limit of $(\Gamma_{\alpha})_{\alpha=1}^\infty$ is also in $\mathcal{L}_1^+(\mathcal{H})$. Since $\|\Gamma_{\alpha}\|_{tr} = Tr \Gamma_{\alpha} \to Tr \Gamma = \|\Gamma\|_{tr}$, the case $\Gamma = 0$ is easy, and we may as well assume the $\Gamma_{\alpha}$ and $\Gamma$ are normalized.

By the triangle inequality,

$$\|\Gamma_{\alpha} - \Gamma\|_{tr} \leq \|\Gamma_{\alpha} - PT_{\alpha}P\|_{tr} + \|PT_{\alpha}P - P\Gamma P\|_{tr} + \|P\Gamma P - \Gamma\|_{tr}. \quad (10)$$

Here, $P$ is a finite-dimensional projection, which can be chosen (depending on $\Gamma$) to make the last term smaller than $\epsilon^2/72$. By weak convergence, $\|PT_{\alpha}P - P\Gamma P\|_{tr} < \epsilon^2/72$ for $n$ greater than some $M$. By Lemma 4.2, $n > M$ then implies $\|\Gamma_{\alpha} - PT_{\alpha}P\|_{tr} < \epsilon/2$, since $\|\Gamma_{\alpha} - PT_{\alpha}P\|_{tr} \leq \|PT_{\alpha}P - P\Gamma P\|_{tr} + \|P\Gamma P - \Gamma\|_{tr}$. Now plugging into Eq. (10) completes the proof.

The next lemma is not new\cite{10}, but proofs in the literature are sketchy.

**Lemma 4.4.** $H_0 + 1$ is locally compact. That is, if $\Lambda$ is a bounded region of $\mathbb{R}^{3N}$, and $Q_{\Lambda}$ denotes projection onto $\Lambda$, then $Q_{\Lambda}(\mathbb{R}^{3N} + 1)^{-1}$ is a compact operator.

**Proof.** First, note that $(1 + T + V_{ee})^{-1} = (1 + T)^{-1} - (1 + T)^{-1}V_{ee}(1 + T + V_{ee})^{-1}$. Since the interaction is relatively bounded with respect to the kinetic energy, i.e., $V_{ee}(1 + T + V_{ee})^{-1}$ is bounded, it suffices to prove this result for $H_0 = T$. 

1 + T = (1 − ∇^2) = h^2, in appropriate units. The inverse of h^{2m} has integral kernel
\[ G_m(x, y) = \int \frac{e^{ik(x-y)}}{(1 + k^2)^m (2\pi)^d} \, dk. \]

\[ G_m(x, y) \text{ is bounded by } c|x-y|^{-2m-3N} \text{ as } |x-y| \to 0, \text{ as follows by scaling, and for large } |x-y|, G_m(x, y) \text{ falls off exponentially.} \]

Thus, for \( m \) large enough, the Hilbert-Schmidt norm of \( Q_\lambda(x)G_m(x, y) \), given by \( \int_{\mathbb{R}^d} |G_m(x, y)|^2 \, dx \, dy \) is finite, so \( Q_\lambda h^{-2m} \) is Hilbert-Schmidt and a fortiori compact. Thus, \( h^{2m} \) is locally compact.

Now, if \( A \geq 1 \) is locally compact, then so is \( A^{1/2} \).

To see this, let \( u_n \) be a sequence of vectors converging weakly to zero. To show that \( A^{-1/2}Qu_n \to 0 \), note that \( \|A^{-1/2}Qu_n\|^2 = \langle Qu_n | A^{-1}Qu_n \rangle \). By assumption, \( A^{-1}Qu_n \to 0 \), so also \( \|A^{-1/2}Qu_n\|^2 \to 0 \). Thus, local compactness of positive operators is preserved by taking square roots. Starting from the locally compact large power \( h^{2m} \), repeated application shows also to be locally compact.

**Theorem 4.1.** If \( \Delta \) is a tight set of densities, and \( M < \infty \), then \( \mathcal{S}(\Delta) \cap \mathcal{E}^{-1}(0, M) \) is relatively compact.

**Proof.** Choose sets \( A_{1/n} \) corresponding to \( \Delta \) as described before Lemma 4.1. Take \( \Gamma \in \mathcal{S}(\Delta) \cap \mathcal{E}^{-1}(0, M) \), and note that \( \Gamma - P_{A_{1/n}}\Gamma P_{A_{1/n}} \) is the part of the state which puts at least one particle outside \( A_{1/n} \), so that its norm is less than \( 1/n: \|\Gamma - P_{A_{1/n}}\Gamma P_{A_{1/n}}\| = 1 < 1/n \). This shows, via Lemma 4.1 that \( \mathcal{S}(\Delta) \cap \mathcal{E}^{-1}(0, M) \) is within \( 1/n \) of the totally bounded set \( P_{A_{1/n}}\mathcal{E}^{-1}(0, M)P_{A_{1/n}} \). Since \( n \) is arbitrary, \( \mathcal{S}(\Delta) \cap \mathcal{E}^{-1}(0, M) \) is itself relatively compact.

With Thm. 4.1 and Lemma 6.1 in hand, it is now easy to prove the main theorems.

**Theorem 4.2 (Existence of Levy search solutions).** For every \( \rho \in \mathcal{X}_N^+ \), \( \mathcal{S}(\rho) \neq \emptyset \). That is, there is a mixed state \( \Gamma \in \mathcal{S}(\rho) \) such that \( \text{Tr} \ H_\rho \Gamma = F_\Gamma(\rho) \).

**Proof.** The set of densities leading to cg-density \( \rho \) is a tight set. Apply Thm. 4.1 to see that \( \mathcal{S}(\rho) = \mathcal{S}(\rho) \cap \mathcal{E}^{-1}(0, F_\Gamma^{\text{max}}) \) is compact. Since \( \mathcal{E} \) is lower semicontinuous (Lemma 6.1), it attains a minimum on \( \mathcal{S}(\rho) \).

**Theorem 4.3 (Lower semicontinuity of \( F_\Gamma \)).** \( F_\Gamma \) is weak lower semicontinuous on \( \mathcal{X}_N^+ \).

**Proof.** It is required to show that \( U_M = \{ \rho : F_\Gamma(\rho) \leq M \} \) is weak closed for any \( M \). Since \( U_M \) is convex (because \( F_\Gamma \) is), it suffices to show that \( U_M \) is norm closed.

Suppose that the sequence \( \{\rho_n\}_{n=1}^\infty \) is contained in \( U_M \). Choose \( \Gamma_n \in \mathcal{S}(\rho_n) \) and apply Thm. 4.1 with \( \Delta = \{\rho_1, \rho_2, \ldots\} \) to see that \( \{\Gamma_n\} \) is contained in a compact set, hence contains a convergent subsequence: \( \Gamma_n \to \Gamma \). Since the map from states to cg-densities is norm continuous (each \( \rho_n \) is a bounded operator), this means that \( \Gamma \in \mathcal{S}(\rho) \). By lower semicontinuity of \( \mathcal{E} \) (Lemma 6.1), \( \mathcal{E}(\Gamma) \leq M \). Thus, \( F_\Gamma(\rho) \leq M \).

The ground state energy for \( v \in \ell^\infty \) is the infimum of the spectrum of \( H(v) \), also given by

\[ E(v) = \inf \{ F_\Gamma(\rho) + v \cdot \rho : \rho \in \ell^1 \}. \]

It can happen that there is no state which actually achieves the infimum. This definition actually makes sense for \( v \in \mathcal{V} \). Also, \( v \) is bounded below in terms of \( E(v) \):

\[ \inf_i v_i \geq E(v) - F_{\Gamma_{\text{max}}}^{\text{max}}. \]

Since \( F_\Gamma \) is clearly convex, and weak lower semicontinuous by Thm. 4.3 it follows (ET: Prop. I.4.1, vT: cor. to lemma 6.12) that \( F_\Gamma \) (on \( \ell^1 \)) and \( E \) (on \( \ell^\infty \)) are a Legendre-Fenchel transform pair, up to signs. That is,

\[ F_\Gamma(\rho) = \sup \{ E(v) - v \cdot \rho : v \in \ell^\infty \}. \]

Again, as in the confined case, \( \rho \) is cg-EV-representable if and only if there is \( v \in \mathcal{V} \) such that such that \( F_\Gamma(\rho_0) + \int v \rho_0 \, dx \leq F_\Gamma(\rho) + \int v \rho \, dx \), for all \( \rho \). Note that this is not the same thing as saying that the supremum on the right-hand side of Eq. (12) is attained. That would mean that the cg-density in question was a ground state cg-density of a potential in \( \ell^\infty \); not all are so.

It is this last observation that requires the method of proof of cg-EV-representability to differ substantially from that employed in Section 3. In a remarkable paper, Chayes, Chayes and Ruskai[3] investigated the representability problem on a lattice and showed that all densities are EV-representable. Surprisingly, their method of proof can be adapted to the current situation with only minor modifications. The general idea is that the potential which represents \( \rho \) should be the one which maximizes \( E(v) - \rho \cdot v \). Find a maximizing sequence \( v^* \), and, using a diagonal construction and the countability of the set of cells, extract a subsequence which converges on each cell, to a candidate potential \( v \). Cell-wise convergence is too vague to ensure much about \( H(v) \), so the second part of the proof establishes that \( H(v^*) \) actually converges to \( H(v) \) in strong resolvent sense, which is enough to show that it has a ground state in \( \mathcal{S}(\rho) \).

Specific results from Reed and Simon[17] or Weidmann[18] are indicated by RS or W.

**Theorem 4.4.** If \( \rho \in \mathcal{X}_N^{++} \), then \( \rho \) is cg-EV-representable by a potential in \( \mathcal{V} \).

**Proof.** A constant can always be added to \( v \) to make \( E(v) = 0 \), so

\[ F_\Gamma(\rho) = \sup \{ -v \cdot \rho : E(v) = 0 \}. \]

Let \( v^* \) be a maximizing sequence for the right-hand side. That is,

\[ \int_{v^* < 0} v^* \, dx - \int_{v^* > 0} v^* \, dx \geq F_\Gamma(\rho). \]

(13)
Since $E(v^\alpha) = 0$, $v^\alpha < 0$ somewhere. Define, for each $\alpha$, a cg-density
\[
\rho^\alpha = \frac{N\theta(-v^\alpha)\rho}{\theta(-v^\alpha)\rho} \geq \theta(-v^\alpha)\rho.
\]
Then,
\[
F_{\rho}^{\alpha} = \int_{v^\alpha < 0} \rho^\alpha |v^\alpha| \, dx \geq \int_{v^\alpha < 0} \rho |v^\alpha| \, dx.
\]
So, $\int_{v^\alpha < 0} \rho |v^\alpha| \, dx$ is bounded, uniformly in $\alpha$, hence, by Eq. (13), $\int_{v^\alpha > 0} \rho |v^\alpha| \, dx$ is, too. Thus, $\int \rho |v^\alpha| \, dx$ is bounded by some constant, and
\[
|v_i^\alpha| \leq \frac{c}{\rho_1}.
\]
(14)
Since each $v_i^\alpha$ thus lies in a bounded interval, a diagonal construction will yield a subsequence $v_i^\alpha$ which converges on each cell, though not in general with any uniformity. By relabeling, we can just write $v_i^\alpha$ for this subsequence, so $v_i^\alpha \to v_i$ for each $i$. This defines our candidate potential $v$ to have $\rho$ as ground state cg-density.

$H(v)$ makes sense because $v_i$ is everywhere bounded below by Eq. (11), and everywhere finite by Eq. (13), so $v \in V$. Let $D$ be the set of smooth pure states with density nonzero only in a finite number of cells; since the Coulomb repulsion is positive and locally $L^2$, $D$ is a common domain of essential self-adjointness for all $H(v')$ with $v' \in V$ (RS: Thm. X.28, Hislop and Sigal: Thm. 8.14). For $\phi$ a unit vector in $D$, $0 \leq \langle \phi | H(v^\alpha) \phi \rangle$ since $E(v^\alpha) = 0$, so
\[
0 \leq \lim_{\alpha \to \infty} \langle \phi | H(v^\alpha) \phi \rangle = \langle \phi | H(v) \phi \rangle < \infty,
\]
because the convergence of the potential is uniform on the support of $\phi$. Thus, since $D$ is a core, $H(v) \geq 0$. Further, $\|H(v) - H(v^\alpha)\| = \|v^\alpha - v\| \to 0$, which shows convergence of $H(v^\alpha)$ to $H(v)$ on $D$. This suffices (RS.VIII.25 or W:Thm. 9.16) to establish strong resolvent convergence.

Now, find $\Gamma_0 \in \mathcal{S}(\rho)$, so that
\[
\mathcal{E}(\Gamma_0) = F_{\rho}(\Gamma_0) = \lim_{\alpha \to \infty} \int -\rho v^\alpha \, dx.
\]
A $\Gamma_0$ which satisfies the first equality exists by Theorem 4.2 and the second equality holds by Eq. (13). Since $e^{-x}$ is a bounded function on $x > 0$, $e^{-H(v^\alpha)} \to e^{-H(v)}$ in strong operator topology (RS.VIII.20, or W:Thm. 9.17). Then $H(v) \geq 0$ implies that
\[
1 \geq \text{Tr} \Gamma_0 e^{-H(v)} = \lim_{\alpha \to \infty} \text{Tr} \Gamma_0 e^{-H(v^\alpha)} \geq e^{-\mathcal{E}(\Gamma_0)} \lim_{\alpha \to \infty} \exp\left[-\int \rho v^\alpha \, dx\right] = 1,
\]
(15)
by use of Jensen’s inequality and the fact that $(v^\alpha)$ is a maximizing sequence for $F_{\rho}(\Gamma)$. Thus, $\text{Tr} \Gamma_0 e^{-H(v)} = 1$, which, since $H(v) \geq 0$, shows that $\Gamma_0$ is concentrated on the ground state manifold of $H(v)$.

5. INDEFINITE PARTICLE NUMBER

Open systems[10] are also of interest for density functional theory. In this case, the total particle number can fluctuate – the system is in equilibrium with a particle reservoir at some chemical potential, which we take to be zero without loss of generality. The theory of the previous two sections extends in full to this case of indefinite particle number. We will explicitly discuss only the unconstrained case.

The relevant state space is the Fock space
\[
\mathcal{F} = \bigoplus_{m=0}^{\infty} \mathcal{H}_m,
\]
where $\mathcal{H}_m$ denotes the $m$-particle Hilbert space. The set of states which interest us is not the entirety of $L^2_1(\mathcal{F})$, but the subset $\mathcal{S}$ of states which are diagonal in particle number, i.e., those of the form $\sum_{m=0}^\infty a_m \Gamma(m)$, where $\Gamma(m)$ is a normalized $m$-particle mixed state, and $\sum a_m = 1$. Below, we write $\Gamma(m)$ for the normalized $m$-particle part of any mixed state $\Gamma$. The following are natural generalizations of previous definitions to the Fock-space setting. The subset of states in $\mathcal{S}$ which have density $\rho$ is denoted $S(\rho)$, and the grand canonical density functional is defined
\[
F(\rho) := \inf \{ \text{Tr} \Gamma H_0 : \Gamma \in S(\rho) \},
\]
(16)
where $H_0$ is simply the direct sum of the kinetic-plus-interaction Hamiltonians for all particle numbers. Notice that the mixed states in Eq. (16) are not restricted to two consecutive total particle numbers, in contrast to the procedure introduced by Perdew, Parr, Levy and Balduz[19]. The ground state energy for potential $v$ is the infimum of the spectrum of $E(v)$ and a state which realizes it is a ground state. As before, we can also write
\[
E(v) := \inf \{ F(\rho) + v \cdot \rho : \rho \in \ell^1 \},
\]
and a ground state is a state which realizes the infimum. Now, the additive constant is important. Decreasing it is likely to result in a ground state with more particles. For some potentials, even though bounded below, $E(v) = -\infty$ because the energy can be lowered indefinitely by adding particles.

The proofs of Lemma 3.1 and of the Hohenberg-Kohn Theorem apply to the current situation with almost no essential changes. The density which is everywhere zero is the exception. It is certainly EV-representable, but not uniquely.

We now sketch the required alterations to the proofs of Sec. 4. Consider generalizing Thm. 4.1 to a tight set of densities $\Delta$ all with the same total particle number $N$. $S(\Delta)$ now refers to the corresponding set of $N$-diagonal Fock-space mixed states. Choose $\epsilon > 0$. If $\text{Tr} \Gamma N_{\text{tot}} = N$, then clearly $\sum_{m>N/\epsilon} a_m < \epsilon$. Similarly, if $\mathcal{E}(\Gamma) \leq \epsilon$, then $\sum_{m \in \ell} a_m < \epsilon$, where
\[
I = \{ m \in \mathbb{N} : \mathcal{E}(\Gamma(m)) > M/\epsilon \}.
\]
According to Thm. 4.1, the set $A_m$ of $m$-particle mixed states with energy less than or equal to $M/\epsilon$ and density less than or equal to some density in $\Delta$ is compact. Since $S(\Delta)$ is within $2\epsilon$ of the convex hull of $\cup_{m \leq N/\epsilon} A_m$, and $\epsilon$ is arbitrary, $S(\Delta) \cap E^{-1}(0, M]$ is itself totally bounded, i.e., relatively compact.

With Lemma 3.1 and Thm. 4.1 thus extended to Fock space, the proofs that $S(\rho) \neq \emptyset$ and that $F$ is lower semicontinuous proceed just as in Thms. 4.2 and 4.3. Thus, we can write, as before,

$$F_L(\rho) = \sup\{E(v) - v \cdot \rho : v \in \ell^\infty\}. \quad (17)$$

What is different about the situation now is that the effective domain of $F_L$ is all cg-densities: positivity is still required, but normalization is not.

The final stage is to adapt the proof of Thm. 4.4 to show that any cg-density, everywhere strictly positive, is Fock-space cg-EV-representable. The trick of adjusting the additive constant in the potential to obtain $E(v) = 0$ had a significant part in the proof of Thm. 4.3 where it was both a labor-saving device and a means of assuring that the potential did not drift off to infinity. In the Fock space setting, it is neither possible nor needed, and we proceed as follows.

Given $\rho$, find a sequence of potentials $v^\alpha$ such that

$$E(v^\alpha) - v^\alpha \cdot \rho \nearrow F(\rho).$$

Since $F(\rho) > 0$, for $\alpha$ large enough

$$E(v^\alpha) > v^\alpha \cdot \rho \geq v^\alpha_{min}N, \quad (18)$$

where $v^\alpha_{min} = \inf_i v^\alpha_i$. But also, by considering a state with any number $M$ of particles all located in a cell where the potential is favorable, $E(v^\alpha) \leq c'M^3 + v^\alpha_{min}M$. Combining these inequalities,

$$v^\alpha_{min} \geq -\frac{c'M^3}{M-N}, \quad \text{if } M > N.$$  

Choosing $M$ to be the next integer larger than $N$, for example, this shows that $v^\alpha_{min}$ is bounded below independently of $\alpha$ for $\alpha$ large enough. And, inserting this bound back into inequality (18) yields a lower bound for the $E(v^\alpha)$ as well. Thus, by a diagonal argument as before, a subsequence (written $v^\alpha$ without loss) exists for which $v^\alpha_i$ converges to $v_i$ for each $i$, and $E(v^\alpha)$ to $E_\infty$ as $\alpha \to \infty$.

The rest of the proof proceeds largely as before, with a few $E_\infty$ inserted. The appropriate domain of essential self-adjointness $D$ is now the set of states with density supported in a finite number of cells, and also with components in only a finite number of $H_m$. The inequality showing positivity of $H(v)$ becomes

$$E_\infty \leq \langle \phi | H(v) | \phi \rangle < \infty.$$  

Since $\{E(v^1), E(v^2), \ldots, E_\infty\}$ is bounded below, $\{H(v^1), H(v^2), \ldots, H(v)\}$ is also, so $e^{-H(v^\alpha)} \to e^{-H(v)}$ in strong operator topology. With $\Gamma_0 \in S(\rho)$, as before, the counterpart of Eq. (15) is

$$e^{-E_\infty} \geq \text{Tr} \Gamma_0 e^{-H(v)} = \cdots = e^{-E_\infty}. \quad (19)$$

6. CONCLUSION

At the simplest level, the results presented in this paper merely increase our knowledge about the V-representability problem. Within every coarse-grained equivalence class of densities, there is at least one which is EV-representable. In addition, there is one such which is representable by a unique potential which is constant on cells, and bounded below, though not necessarily (in the unconfined case) bounded above. Extensions of these results to spin density functional theory and multicomponent systems is straightforward.

Valuable as those observations may be, that way of putting it misses the bigger idea that a coarse-grained point of view may be a generally superior way to look at the foundations of density functional theory. From that perspective, what has been achieved is support for the idea in that EV-representability is shown to not be a problem at all in a coarse-grained framework. Little is lost by this change, since the coarse-graining scale can be chosen as the size of an atomic nucleus, for example, and most knowledge we may have of the fine-grained level can be incorporated relatively easily.

Establishing EV-representability is the first step toward controlling the local structure of the Lieb functional. Because, if $F_L$ is differentiable at $\rho$, in any sense, the derivative is given by the representing potential. With regard to this question, the coarse-grained theory has taken one step. We have shown that $F_L$ is differentiable in the confined case, by taking advantage of the finite-dimensionality. The most immediate outstanding problem at this point is to demonstrate some sort of differentiability in the unconfined situation. General principles avail us nothing because $X_N^+$ has empty interior relative to $X_N$. (The fine-grained theory also has this problem.) A widespread view that $F_L$ is Gâteaux differentiable at EV-representable densities is in error.) Hopefully, it will be possible to show that if $\eta_i \neq 0$ for only finitely many $i$ and $\sum \eta_i = 0$, then $F_L(\rho + s\eta)$ is differentiable as a function of $s$ at $s = 0$. This property is weaker than Gâteaux differentiability, but probably not in any important way.

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