Reverse engineering in many-body quantum physics: What many-body system corresponds to an effective single-particle equation?

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The mapping, exact or approximate, of a many-body problem onto an effective single-body problem is one of the most widely used conceptual and computational tools of physics. Here, we propose and investigate the inverse map of effective approximate single-particle equations onto the corresponding many-particle system. This approach allows us to understand which interacting system a given single-particle approximation is actually describing, and how far this is from the original physical many-body system. We illustrate the resulting reverse engineering process by means of the Kohn-Sham equations of density-functional theory. In this application, our procedure sheds light on the non-locality of the density-potential mapping of DFT, and on the self-interaction error inherent in approximate density functionals.

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One of the most widely used and successful approaches to many-particle physics is to map the many-body problem onto an effective single-body problem. Innumerable concepts and methods of theoretical physics derive from this general idea. The mean-field approximation, one of the most widely used approximation schemes in all fields of physics, is of this type, as well as the (in principle exact) mapping onto Kohn-Sham (KS) equations, used in density-functional theory (DFT). Here we introduce the concept of reverse engineering of single-particle equations \cite{1}, as a tool for discovering for which many-body system a given single-particle approximation becomes exact. The idea is developed below in the framework of DFT, but the concept is completely general, and can be extended to other single-particle methods.

DFT \cite{2} in the Kohn-Sham formulation \cite{3} is a tool for calculating properties of many-body systems by means of a one-to-one mapping between the interacting system and a fictitious non-interacting one, with the same ground-state density $n(r)$. All ground-state properties can in principle be expressed as functionals of this density. The success of DFT depends on the quality of the approximation to the exchange-correlation (xc) energy functional $E_{xc}[n]$, which enters the KS equations through its functional derivative, the xc potential $v_{xc}(r)$. $E_{xc}$ stems from the interactions in the original many-body system, and its functional form for Coulomb interacting systems, such as electrons in atoms, molecules, nanostructures and solids, is unknown. In order to construct viable approximations, a great deal of work has therefore been devoted to the derivation of exact properties of the exchange-correlation functional and potential \cite{4,5}. The performance of a functional is judged by how close the density, and the observables calculated from it, are to that of the many-body system under study.

The idea of reverse engineering suggests a different mode of analysis, namely to ask: for what system does a given approximation become exact? This question can be interpreted in two ways, one very common, the other being proposed here. To exemplify both, consider the local-density approximation (LDA). By construction, the LDA becomes exact for uniform densities. But in practice we rarely apply density functionals to uniform systems. Instead, we apply DFT to realistic inhomogeneous many-body systems, for which the LDA density is $r$-dependent and approximate. The question we ask is: for which alternative many-body system is this approximate density the exact ground-state density? In this context, we call this alternative many-body system the interacting-LDA (i-LDA) system \cite{6}. Reverse engineering of DFT refers to the process of constructing this alternative many-body system.

The aim of this construction is not to simulate large systems, for which nothing is gained by mapping a many-body system on another, equally complex, one. Rather, it shifts attention from the density predicted by the LDA to the external potential predicted by the LDA (i.e., the i-LDA potential), allowing us to investigate the LDA (or any other single-particle approximation to the many-particle Hamiltonian) in a novel and particularly detailed way. For example, we can now ask: how close is the corresponding i-system external potential to the true external potential? Which artificial features is the chosen approximation building into the system? As in other parts of science, reverse engineering enables one to understand the functionality and structure of the engineered device on a different level, opening up new pathways for improvement.

To illustrate the basic idea, we start from the LDA densities of the helium atom and of Hooke’s atom, and invert the many-body Schrödinger equation to construct that external potential for which the LDA densities are exact ground-state densities. Comparison to the true external potentials of the helium atom ($\sim 1/r$) and Hooke’s atom ($\sim r^2$) reveals the errors inherent in the approximate density and functional. By calculating explicitly the external potential for the i-LDA system, the pro-

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posed method provides the exact i-LDA system Hamiltonian. This implies that all properties of the i-LDA system (which themselves depend on the accuracy of the LDA) can, in principle, be directly calculated and compared to the ones of the exact many-body system.

Studying relatively simple systems, such as helium or Hooke’s atoms, to understand approximations has proved fruitful in the past [7–9]. Earlier inversion schemes in DFT were used to find the \( v_{xc}(r) \) that reproduces a given exact density, by inverting the single-particle Kohn-Sham equations [10]. This approach provides information on the exact \( v_{xc} \). The reverse engineering procedure, by contrast, aims at reproducing a given approximate density by inverting the many-body Schrödinger equation, providing information on the approximate \( v_{xc} \). Inversion of the many-body equation is a much harder task, which up to now had only been attempted for a one-dimensional model system within the adiabatic approximation to time-dependent DFT [9]. To achieve this, we developed inversion schemes that are substantially more accurate than previous ones (see discussion below).

We consider the N-body Hamiltonian

\[
H = \hat{T} + \hat{V}_{ee} + \hat{V}_{ext},
\]

where \( \hat{T} \) is the kinetic energy operator, \( \hat{V}_{ee} \) is the electron-electron interaction and \( \hat{V}_{ext} = \sum_{i=1}^{N} v_{ext}(r_i) \) is the external potential. By multiplying the Schrödinger equation \( H \Psi = E \Psi \) from the left by \( \Psi^* \) and integrating over all but one of the coordinates we obtain

\[
\int \Psi^* (r_1 \ldots r_N) \left( \hat{T} + \hat{V}_{ee} + \hat{V}_{ext} \right) \Psi (r_1 \ldots r_N) d^3r_2 \ldots d^3r_N = E \frac{n(r_1)}{N}.
\]

From this we find, for the term concerning the external potential,

\[
\int \Psi^* \sum_{i=1}^{N} v_{ext}(r_i) \Psi d^3r_2 \ldots d^3r_N = v_{ext}(r_1) \frac{n(r_1)}{N} + (N-1) \int \Psi^* \frac{v_{ext}(r_2)}{N} \Psi d^3r_2 \ldots d^3r_N
\]

\[
= v_{ext}(r_1) \frac{n(r_1)}{N} + \frac{2}{N} \int \gamma(r_1, r_2; r_1, r_2) v_{ext}(r_2) d^3r_2,
\]

where \( \gamma(r_1, r_2; r_1, r_2) = \frac{N(N-1)}{2} \int \Psi^* \Psi d^3r_3 \ldots d^3r_N. \)

We combine these results to obtain an iterative relation for the external potential \( v_{ext}(r_1) \) that reproduces the target density \( n_{\text{target}}(r_1) \),

\[
v^{i+1}_{\text{ext}}(r_1) = \frac{1}{n_i(r_1)} \left[ E_i n_{\text{target}}(r_1) - E_i n_i(r_1) \right] - N \int \Psi_i^* (\hat{T} + \hat{V}_{ee}) \Psi_i d^3r_2 \ldots d^3r_N
\]

\[
-2 \int \gamma_i(r_1, r_2; r_1, r_2) v^{i}_{\text{ext}}(r_2) d^3r_2,
\]

To avoid having to calculate the integrals, we use the identity

\[
\frac{1}{n_i(r_1)} \left[ E_i n_{\text{target}}(r_1) - E_i n_i(r_1) \right] = -N \int \Psi_i^* (\hat{T} + \hat{V}_{ee}) \Psi_i d^3r_2 \ldots d^3r_N
\]

\[
-2 \int \gamma_i(r_1, r_2; r_1, r_2) v^{i}_{\text{ext}}(r_2) d^3r_2,
\]

and obtain the simple iterative relation

\[
v^{i+1}_{\text{ext}}(r_1) = \frac{1}{n_i(r_1)} E_i [n_{\text{target}}(r_1) - n_i(r_1)] + v^i_{\text{ext}}(r_1),
\]

which is to be iterated with the Schrödinger equation \( (\hat{T} + \hat{V}_{ee} + \hat{V}_{\text{mol}}) \Psi = E \Psi \). At convergence, \( n_i(r_1) = n_{\text{target}}(r_1) \) and \( v^{i+1}_{\text{ext}}(r_1) \) is the external potential that reproduces this density via the many-body Schrödinger equation.

We note that if \( n_i(r_1) \) is larger (smaller) than \( n_{\text{target}}(r_1) \) then the potential must increase (decrease) at this point to bring \( n_{i+1}(r_1) \) closer to \( n_{\text{target}}(r_1) \). Therefore, iteration of Eq. (6) is expected to converge if \( E_i < 0 \). If \( E_i > 0 \), we replace Eq. (6) by

\[
v^{i+1}_{\text{ext}}(r_1) = \frac{1}{n_i(r_1)} E_i [n_i(r_1) - n_{\text{target}}(r_1)] + v^i_{\text{ext}}(r_1).
\]

Scheme (6) converges relatively easily for the helium atom (where \( E < 0 \)) and scheme (7) for Hooke’s atom (where \( E > 0 \)). We aid convergence in both cases by mixing \( v^{i+1}_{\text{ext}} \) with 80% of \( v^i_{\text{ext}} \), and iterate until the relative error \( \int d^3r |n_i(r) - n_{\text{target}}(r)|/ \int d^3r n_{\text{target}}(r) \) has reached a desired level.

At convergence, we obtain that external potential whose many-body ground state has the same density as was predicted by the approximate density functional for the true external potential. As this interacting system ground-state reproduces the LDA density then by the Hohenberg-Kohn theorem [11] this interacting system must be the exact interacting system corresponding to the LDA. We check this property by independently solving the many-body Schrödinger equation with the converged alternative external potential, using a larger basis set than that used in the iterations. This additional consistency procedure allows our inversion scheme to be precise even in regions of space where the density is just \( 10^{-7} \) a.u. (helium) and \( 10^{-12} \) a.u. (Hooke’s atom), i.e. orders of magnitudes smaller than previous inversion schemes, which, according to Ref. [9] attain an accuracy of up to \( 10^{-2} \) a.u.

We now illustrate both inversion schemes, and the additional consistency check, by applying our procedure to the helium atom and Hooke’s atom, generating the approximate (target) density from the LDA. For two electrons in a spherically symmetric potential the ground state can only be a function of the distance of each electron from the origin and the angle between the electron vectors. Hence we employ the basis \( \phi_{i\ell l} = R_i(r_1) R_j(r_2) \sqrt{2l+1} P_l(\cos(\theta))/\sqrt{4\pi} \). For Hooke’s atom \( R_i(r) = Q_i(r)e^{-\alpha r^2} \) is a harmonic oscillator-like wavefunction while for the helium atom it is a hydrogen-like wavefunction \( R_i(r) = Q_i(r)e^{-\alpha r} \). The \( Q_i(r) \) are polynomials of degree \( i \) created via the Gram-Schmidt procedure such that the \( R_i \) are orthonormal.
Hooke’s atom is an interacting system of two electrons in the harmonic confining potential $v_{\text{ext}} = \omega r^2/2$. We use the LDA in the parametrization of Perdew and Wang [12] for $v_{\text{ext}}$, and solve the KS equations for $\hbar \omega = 0.0365$ Hartree. For comparison we also calculate the exact density from the exact interacting wave function using the method of Taut [13]. We then apply Eq. (7) to find the external potential of the interacting system that reproduces the LDA density, i.e. the i-LDA system [6].

In practice, an external potential found with a basis of e.g. $6^3$ functions may not reproduce the LDA density when we counter-check the scheme by solving the many-body Schrödinger equation of the i-LDA system using a larger basis of size e.g. $7^3$. Hence, we repeat the procedure with increasing basis size until we find an external potential that reproduces the LDA density even with a larger basis.

In Fig. 1 (a) the exact and LDA densities are plotted. The LDA and i-LDA densities are indistinguishable, but the difference between the LDA and exact densities is large, and consequently the potentials of the true and the i-LDA system are rather different. Fig. 1(c) shows a substantial difference between the LDA and exact densities is large, and consequently the potentials of the true and the i-LDA system are rather different. Fig. 1(c) shows a substantial difference between the i-LDA potential and the true external potential at large $r$, where the i-LDA potential grows more rapidly than the true potential. The LDA density is very different from the true density in this region (Fig. 1 (a) inset), with a relative error $(n(r) - n^{\text{LDA}}(r))/n^{\text{true}} \sim 60\%$ for $r = 28$ [14]. By contrast, near the origin the i-LDA potential is slightly weaker than the true one. In between the two limiting regions, the i-LDA potential and the true external potential cross various times. These crossings manifest themselves in a complex way in the behavior of the corresponding densities: Fig. 1 shows that a crossing in the potentials around $r = 8$ (Fig. 1 (c) inset) corresponds to a crossing in the densities (Fig. 1 (b)), whereas the crossings at $r \sim 2$ and $r \sim 25$ are not accompanied by a crossing in the densities.

Next we consider the helium atom, for which we use the iterative scheme of Eq. (6) to calculate the i-LDA external potential. Here a basis of $7^3$ functions is required to find the external potential that satisfies our consistency check, i.e. reproduces the density when solving the Schrödinger equation with a larger basis ($8^3$ functions).

As Fig. 2 shows, the LDA reproduces relatively well the exact radial probability density, but an underestimate at small $r$ and overestimate at large $r$ is evident. A closer view of large $r$ (inset of Fig. 2(b)) shows that there is a larger discrepancy (49% on the range $4 \leq r/\omega_0 \leq 5.5$ compared to 3.68% overall) between the LDA density and the exact density here. However we still find that the i-LDA system density is almost indistinguishable from the LDA density on this scale (0.78% error) and its overall error at 0.037% is comparable to that of 0.0063% which we achieved for Hooke’s atom. In Fig. 2(c) we again note that the external potential of the i-LDA system is substantially different from the true one for large values of $r$, where it is weaker than the true potential, causing the LDA density to be spread out slightly more than the exact one. We also observe a crossing of both potentials and densities for $r \sim 1.2$ (Fig. 2(b)) where the radial probability density is high.

Both Hooke’s and the helium atom thus exhibit a relationship between crossings in densities and crossings in potentials, but this relation is local only in regions where the radial probability density is high. The systems considered here have spherical symmetry, hence the radial probability density $4\pi n(r)r^2$ indicates the probability of finding a particle at distance $r$ from the origin. Thus, a single marked peak of the radial probability density at $r$ indicates a high probability of finding a particle at that distance and a very low probability of finding the particle elsewhere. In this sense, the system is “almost classical” in the region of the peak. The charge density itself does not have this property. The regions of high radial probability density can be interpreted as almost classical, so that a local relationship between the ‘particle position’ and the external potential can be expected.

By contrast, crossings in the potentials in regions with low values of the radial probability density do not necessarily correspond to crossings in the densities. These non-local regions are the ones in which the quantum nature of the system is more apparent. The identification of such regions is particularly interesting in view of recent investigations of semiclassical approximations to DFT [15]. The LDA density can even be higher than the exact density in regions where the i-LDA
confining at very large $r$. Hooke’s atom [17], the i-LDA potential becomes much too weak in the asymptotic region, so that the i-LDA potential and exchange-correlation (xc) potential diverges to infinity even faster than the exact potential. This dramatically highlights the non-local nature of the density-potential mapping in DFT and, more generally, in all many-body methods that introduce an effective single-particle potential. This approach allows one to judge the performance and failures of, e.g., an approximate density functional or single-particle equation, by comparing two external potentials and densities. We speculate that this feature is caused by a less investigated consequence of self-interaction, causing the electron density to spread out more at smaller $r$ in an attempt to minimize the self-Coulomb energy. As this starts to occur before the asymptotic region is reached, it involves many electrons, and can thus be interpreted as a consequence of the many-electron self-interaction error [18, 19].

The self-interaction problem in a single-particle framework has been studied for many years. Much more recently, it has become apparent that this single-particle self-interaction correction does not fully remove the self-interaction error in many-particle systems. The search for a clear indicator and a remedy for this problem has recently taken center stage as one of the main unsolved problems of DFT [18, 19]. Our reverse engineering prescription is a way to shed light on this complex problem, since the many-electron self-interaction error contributes to the difference between the true and the i-LDA potentials at intermediate distances.

These considerations illustrate the concept and use of the i-LDA system. To construct this system we have developed two iterative schemes for inverting the many-body Schrödinger equation. Previous inversion schemes within DFT either were concerned with inverting the much simpler single-particle KS equation, or directed at a one-dimensional model many-body system. Our schemes attain orders of magnitude higher accuracy than previous schemes and are applicable to three-dimensional many-body systems.

Building on this technical advance, we have introduced the concept of reverse engineering in DFT and, more generally, in all many-body methods that introduce an effective single-body potential. This approach allows one to judge the performance and failures of, e.g., an approximate density functional or single-particle equation, by comparing two external potentials, thus revealing spatially resolved information on properties such as self-interaction errors and non-locality, in a physically transparent way. Another possible application of quantum reverse engineering is to design that external potential that reproduces a desired density distribution in a given spatially inhomogeneous many-body system, which is an exciting prospect for the design of nanostructured devices.

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[1] The usual challenge in engineering is to design a device with a given functionality in mind. Reverse engineering refers to the inverse process where one tries to understand, and then improve, the construction and operation of a given device whose functionality and design principles are a priori unknown.

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