Letter

Examining the justification for the introduction of a fermion localization function

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Becke and Edgecombe suggested in 1990 a theoretical tool to describe electron localization in atoms and molecules, an idea which was borrowed by a large number of nuclear theorists since 2011 to describe nucleon localization in nuclear systems. I argue here that these arguments are highly questionable and cannot be used in interacting systems where effects beyond the naive mean field or the simple Hartree-Fock framework are important and the inclusion of correlations induced by particle interactions is necessary in order to introduce such a localization function. I also describe several aspects of the exchange and irreducible two-body density matrices, which depend on the character and strength of the two-particle interaction and which can be useful in justifying the derivation of an appropriate energy density functional.

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In nuclei lighter than ^{40}Ca molecular-like states have been studied for a long time [1,2]. The nuclear molecular clusters are in reality microcrystals, as the relative separation between the clusters varies very little. In the crust of neutron stars the formation of the so-called pastalike phase, another type of matter clusterization, or matter crystallization more accurately, is known for decades [3]. Obviously, the formation of fission fragments in nuclear fission [4] is another example of dynamical formation of nuclear clusters. In order to easily identify theoretically the formation of "clumps" of electronic matter in atoms and molecules Becke and Edgecombe [5] advocated the use in the Hartree-Fock approximation of the quantity

$$D_{\tau,\sigma}(\boldsymbol{r}) = \tau_{\tau,\sigma}(\boldsymbol{r}) - \frac{1}{4}\frac{|\nabla n_{\tau,\sigma}(\boldsymbol{r})|^2}{n_{\tau,\sigma}(\boldsymbol{r})} - \frac{|\boldsymbol{j}_{\tau,\sigma}(\boldsymbol{r})|^2}{n_{\tau,\sigma}(\boldsymbol{r})}, \quad (1)$$

where the subscripts $\tau = n$, p, and σ stand for isospin and spin respectively in the case of nuclear systems and $\tau_{\tau,\sigma}(\boldsymbol{r})$, $n_{\tau,\sigma}(\boldsymbol{r})$, and $\boldsymbol{j}_{\tau,\sigma}(\boldsymbol{r})$ are the kinetic energy, nucleon and current number densities, respectively. The current density term is required by Galilean invariance [6–12], the form used by nuclear theorists [13–30] (this is very likely an incomplete list of references). Readers will recognize that $D_{\tau,\sigma}(\boldsymbol{r})$, related to the kinetic energy density without the last term, was known to von Weizsäcker [31] (here in the original form, without the current number density),

$$\tau_{\tau,\sigma}(\boldsymbol{r}) = \frac{3}{5}(6\pi^2)^{2/3} n_{\tau,\sigma}^{5/3}(\boldsymbol{r}) + \frac{1}{4}\frac{|\nabla n_{\tau,\sigma}(\boldsymbol{r})|^2}{n_{\tau,\sigma}(\boldsymbol{r})}. \quad (2)$$

The accuracy of second term in Eq. (2) was questioned many times over the years [32–35], and gradient expansions lead to a prefactor $1/36$ instead of $1/4$, and currently Padé approximants and other parametrizations are also considered. There are several reasons for this intense interest in the gradient term, which originates from the definition of the two-body density matrix (see below), since in the density functional theory (DFT) [34] the exchange and correlation energies, arising from interactions, are treated on a equal footing, unlike in the Hartree-Fock approximation, which was used in introducing $D_{\tau,\sigma}(\boldsymbol{r})$ in Eq. (1). Equation (1) is derived from the Hartree-Fock approximation of the two-body number density for spin pairs with $S = 1, S_z = \pm 1, T = 1, T_z = \pm 1$ only, for which Pauli correlation exists

$$n_2(\xi, \zeta) = \tfrac{1}{2}[n_1(\xi, \xi) n_1(\zeta, \zeta) - n_1(\xi, \zeta) n_1(\zeta, \xi)], \quad (3)$$

where $n_1(\xi, \zeta) = \sum_k \phi_k(\xi)\phi_k^*(\zeta)$ is the Hartree-Fock density matrix expressed through the single-particle wave functions $\phi_k(\xi)$ and $\xi = (\boldsymbol{r}_1, \sigma, \tau), \zeta = (\boldsymbol{r}_2, \sigma', \tau')$. In the limit $\boldsymbol{s} = \boldsymbol{r}_1 - \boldsymbol{r}_2 \to \boldsymbol{0}$ [5–7,9,11–13]

$$n_2(\xi, \zeta) = \tfrac{1}{3} n_{\tau,\sigma}(\boldsymbol{r}) D_{\tau,\sigma}(\boldsymbol{r}) \boldsymbol{s}^2 + \mathcal{O}(\boldsymbol{s}^4)$$

and the conditional probability to find a particle with coordinate $\boldsymbol{r}'$ from a particle with coordinate $\boldsymbol{r}$ and the same spin and isospin is proportional to $D_{\tau,\sigma}(\boldsymbol{r})$. The fermion localization function (FLF) $C_{\tau,\sigma}(\boldsymbol{r})$ was introduced in Ref. [5,13] basically as a measured of the accuracy of the zeroth-order Thomas-Fermi approximation for the kinetic energy density

$$C_{\tau,\sigma}(\boldsymbol{r}) = \left\{1 + \left[\frac{D_{\tau,\sigma}(\boldsymbol{r})}{\frac{3}{5}(6\pi^2)^{5/3} n_{\tau,\sigma}^{2/3}(\boldsymbol{r})}\right]^2\right\}^{-1}. \quad (4)$$

It appears that in atomic and molecular systems, where the role of correlation energy is significantly less important than in nuclei and the Hartree-Fock approximation is sufficiently accurate [21], the spatial profiles of the electronic shells more pronounced in $C_\sigma(\boldsymbol{r})$ than in number density profiles $n_\sigma(\boldsymbol{r})$. As the term proportional to $|\nabla n_{\tau,\sigma}(\boldsymbol{r})|^2$ has a subdominant role in DFT [32–35], likely it can be neglected, and in the case of static systems the quantity $C_{\tau,\sigma}(\boldsymbol{r})$ is defined basically by the ratio of the actual kinetic energy density to its Thomas-Fermi approximation. It is obvious that neither this ratio nor the quantity $C_{\tau,\sigma}(\boldsymbol{r})$ defines an observable and this is merely a somewhat arbitrary measure of the accuracy of the Thomas-Fermi approximation to the kinetic energy density alone.

From many studies performed in nuclear physics over the years, it is clear that the extended Thomas-Fermi approximation, including corrections up to $\mathcal{O}(\hbar^4)$ for the kinetic energy density provides a pretty good approximation for densities and total energies of nuclear systems, see Section 4.4 in Ref. [35], where references to many more complete studies can be found. The (extended) Thomas-Fermi approximation, however, does not describe shell effects or, more generally, the quantization of the single-particle motion in various geometries.

Does the quantity $C_{\tau,\sigma}(\boldsymbol{r})$ indeed help us better visualize clustering effects? Cluster formation are typically due to the existence of two-, three-, and four-body and higher interactions between particles and not the result of the quantization of the single-particle motion in a finite system. The main part of the interaction between electrons is repulsive, and in any physical system (except hard balls at high density) clusters would form only if there will be an effective attractive interaction between the electrons. One might argue that the Fock exchange Coulomb energy between electrons is attractive, a result which is, however, accurate only for nonrelativities systems [34]. However, it is hard to make the argument that the "attractive" electron Coulomb exchange energy between electrons with the same spin, or in spin-polarized electron systems, leads to electron clusterization or electron shells. On the other hand, in nuclear systems, which are typically bound and therefore the interparticle interaction is mainly attractive, and volume and symmetry energies favor spin- and isospin unpolarized systems, the exchange interaction is repulsive, and again, one can hardly make the case that the spin- and isospin-polarized nuclear systems can clusterize or lead to quantization of the single-particle motion. One might bring as a counterexample the Wigner crystal [36] of a very low density electron gas, which, however, is basically a classical system, where exchange effects are negligible, see also below. The Wigner crystal is similar to a system of hard spheres, which does not clusterize, but the negligible effects of the exchange energy could lead to a disorder state of spins. Clearly, the quantity $C_{\tau,\sigma}(\boldsymbol{r})$, which is sensitive to the quantization of the single-particle motion and is by definition a one-body quantity, cannot describe the formation of clusters, which implies a very strong spatial correlation between various particles and which therefore should be described by a two-body or many-body number density.

In nuclei and in cold fermionic atomic systems, which are qualitatively similar to dilute neutron matter [37], the situation is much more complex. It makes sense to discuss at first the cold atom systems, where the interaction is very simple and both experimental and *ab initio* theoretical approaches are in complete agreement to a very high degree of accuracy. For a Fermi gas with the zero-range interaction the properties of the system are controlled by a single dimensionless parameter $k_F a$, where a is the s-wave scattering length, k_F is the average Fermi momentum, and where there is a complete understanding of both infinite homogeneous systems as well as systems in external traps [10,12]. Around unitarity, where the scattering length $|a| \gg 1/\sqrt[3]{n}$ only two fermions interact, one with spin up and the other with spin down, and in the limit $|a| \to +\infty$ the entire system is a gas of barely overlapping pairs with total spin $S = 0$, thus a clustered system. In such a Fermi system Cooper pairs are formed, with size ranging from extremely large to smaller than the average interparticle separation, depending on the actual value of a, and these Cooper pairs freely collide with each other without being destroyed. The two-body number density for a particle with spin-up and the other with spin-down has a universal behavior $n_2(\boldsymbol{r}_1, \boldsymbol{r}_2) \propto 1/|\boldsymbol{r}_1 - \boldsymbol{r}_2|^2$ at any energy or temperature for $|\boldsymbol{r}_1 - \boldsymbol{r}_2| < |a|$. This behavior is related to a one-body momentum distribution for large momenta $n_p \propto 1/p^4$ [10,12,37–49]. This system is very similar to the dilute neutron matter appearing in neutron stars [50–52]. These essentially independent "Cooper pairs" are clusters.

The method suggested by Becke and Edgecombe [5] and used in nuclear studies cannot capture this kind of clustering in either dilute neutron matter, nor in nuclei [13–30], since it considers the correlations or clustering between fermions with the same spin only. An *ad hoc* alternative was, however, adopted in nuclei to describe clustering or localization on nucleons, and one uses the product $C_{n,\sigma}(\boldsymbol{r})C_{p,\sigma}(\boldsymbol{r})$. It is obvious, on the other hand, that a product of probabilities describes independent events and therefore the proton and neutron distributions cannot be correlated, and therefore this measure actually points to the absence of clustering. The clustering in nuclear systems cannot be explained through exchange effects but through the interplay among volume energy, surface tension, and symmetry energy in a region with a nuclear matter with both protons and neutrons; see for a classical example the case of the nuclear pasta phase, which actually is a quantum crystal [3], as are the nuclear molecular states [1,2] as well. A cluster has a well-defined surface, which is characterized by a repulsive surface energy, due to nuclear surface tension. Surface tension can be counteracted by a stronger, typically proton-neutron attraction, iff proton and neutron densities are (almost) spin saturated.

The kinetic energy density for fermions with the same spin in a large momentum interval exhibits a power-law behavior $n_p \propto 1/p^4$ in the presence of either pairing correlations and/or SRCs [10,12,38–43,46–49] and that would lead to a false signal using Eq. (4). The SRCs in nuclei are due to several effects, the tensor interaction between protons and neutrons [45,53–58] and pairing correlations [10,12,37–44,46–49], the latter also leading to long-range correlations. It is important to appreciate that in the case of zero-range interactions the SRCs between two fermions, which for a dilute atomic gas are between fermions of the same kind, but with opposite spins, are present at any excitation energy [37,41–43], even when a pairing condensate, characterized by long-range phase order, does not even exist for example in nuclear systems [47,48,59–64].

In order to describe such clustering or SRCs using Eq. (4) one has to consider the generalization of the FLF,

$$C_\tau(\boldsymbol{r}) = \left\{ 1 + \left[\frac{\mathcal{D}_\tau(\boldsymbol{r})}{\frac{3}{5}(3\pi^2)^{2/3} n_\tau^{2/3}(\boldsymbol{r})} \right]^2 \right\}^{-1}, \tag{5}$$

$$\mathcal{D}_\tau(\boldsymbol{r}) = \tau_\tau(\boldsymbol{r}) - \Delta_\tau(\boldsymbol{r})\kappa_\tau(\boldsymbol{r}) - \frac{1}{4}\frac{|\nabla n_\tau(\boldsymbol{r})|^2}{n_\tau(\boldsymbol{r})} - \sum_\tau \frac{|\boldsymbol{j}_\tau(\boldsymbol{r})|^2}{n_\tau(\boldsymbol{r})}, \tag{6}$$

with the very critical important correction arising from the pairing interaction, here for *nn*- or *pp* pairs only. Here $n_\tau(\boldsymbol{r})$, $\tau_\tau(\boldsymbol{r})$, $\kappa_\tau(\boldsymbol{r})$, and $\Delta_\tau(\boldsymbol{r})$ are the regularized number, kinetic energy, anomalous densities, and the renormalized corresponding pairing potentials. In the case of zero-range pairing interaction the kinetic energy density and anomalous number density both diverge, but the combination $\tau_\tau(\boldsymbol{r}) - \Delta_\tau(\boldsymbol{r})\kappa_\tau(\boldsymbol{r})$ in Eq. (5) is free of divergencies [10,39,40,65]. Since in nuclei the SRCs are mostly due to the presence of the tensor interaction between the protons and neutrons, their effect can be described by introducing an effective *pn*-pairing field [47]. For nuclear systems, as experiments also amply demonstrate [53,55–58] there is always a momentum interval where the nucleon momentum distribution has power-law behavior $n(p) \propto 1/p^4$ as obtained in various *ab initio* studies [45,63,66] and references therein. The correction due the presence of pairing or/and SRCs has never been considered in discussing nucleon localization in previous studies and it is clear that the definition of the function $\mathcal{D}(\boldsymbol{r})$ then depends on arbitrary cutoffs used in theoretical calculations [13–30]. The use of finite short-range interactions, such as Gogny interaction, might superficially mask the presence of divergences, as there is always a relatively large momentum interval contributing to the kinetic energy and anomalous densities controlled by the short radius of the interaction and where the momentum distribution has a power-law behavior.

Even after introducing the renormalized quantity $\mathcal{D}_\tau(\boldsymbol{r})$, see Eq. (6), it is still not obvious that $C_\tau(\boldsymbol{r})$ is satisfactory measure of clusterization in nuclear physics, since $C_\tau(\boldsymbol{r})$ is not an observable. As $C_\tau(\boldsymbol{r})$ is *a measure of how good the Thomas-Fermi approximation only*, and that since in DFT the gradient term of the nucleon density $\frac{|\nabla n_\tau(\boldsymbol{r})|^2}{n_\tau(\boldsymbol{r})}$ can likely be neglected [32–35], and in the absence of any currents, the only thing left in Eq. (4) is the ratio of the renormalized kinetic energy density to its Thomas Fermi approximation, which again, it is not an observable, unlike the presence of a cluster.

The tensor interaction plays a very important role in nuclei; in particular it leads to a bound proton-neutron system. The role of SRCs in nuclei, due mainly to the tensor interaction between protons and neutrons has been known for a long time. Levinger [53,54] pointed out more than 70 years ago that SRCs are critical to describe the nuclear photo-effect. Since photons are a weak probe of nuclear properties, the fact that a pair of neutron and proton is predominantly emitted clearly points to the presence of short-range quasideuteron pairs in nuclei prior to the photon striking a nucleus. The presence of strong SRCs in nuclei has been persuasively demonstrated in the JLAB experiment in the past decade [55–58]. Obviously, SRCs due predominantly to tensor interaction lead to the formation of clusters, as one can clearly see in light nuclei where α-like molecular clusters are routinely observed and in which case both proton and neutron subsystems are spin saturated and symmetry energy effects dominate, leading to clusters with mainly equal proton and neutron numbers. A simple evaluation of the magnitude of the symmetry energy shows that half of its magnitude is controlled by minimizing the kinetic energy and thus equalizing the spin-up and spin-down occupation probabilities of same type of nucleons, and the other half is due to the proton-neutron interaction. As the initial study of localization effects in nuclei [13] clearly shows, see Fig. 2 in this reference, the FLF $C_{\tau,\sigma}(\boldsymbol{r})$ simply very accurately predicts where the gradient of the number density $n_{\sigma,\tau}$ for a specific σ and τ is largest. The volume and symmetry energy ensure that a subsystem is spin- and/or isospin unpolarized, unless Coulomb effects become relevant, as in the case of the pasta phase in neutron star crust [3]. The volume energy and the nuclear surface tension are largely spin and isospin independent. When an "internal" surface appears the single-particle quantization effects start playing a subdominant role and they appear to be amplified by the FLF $\mathcal{C}_{\sigma,\tau}(\boldsymbol{r})$, however, leading to unrealistic images of the nuclear matter distribution. Consider the example of ^{16}O discussed in Ref. [13], where the FLF $C_{n,\sigma}(\boldsymbol{r})$ allegedly points to the existence of "spatial shell-like" structure of ^{16}O with an average radius of about 3 fm or larger and an very pronounced "inner density depression" with a radius of about 1.25 fm (estimated at half-density). One might surmise that the proton localization should be very similar. At the same time no conceivable density probe of ^{16}O ever revealed the existence of such a pronounced clustering effect, specifically the existence of a well-defined spherical shell structure in the number density. Is there any other type of probe to reveal the reality of this type of clustering? The FLF $C_{\tau,\sigma}(\boldsymbol{r})$ is at best some rather arbitrary measure of the accuracy of the Thomas-Fermi approximation to the spin and isospin kinetic energy density and has a very tenuous relation with possible clustering effects, which are controlled by the interplay of the surface tension, the local spin, and isospin saturation in a given nucleus. In larger nuclei and particularly in neutron start crust there are significant effects due to Coulomb interaction, which only indirectly reflect on the accuracy of the Thomas-Fermi approximation of the spin-isospin kinetic energy density "measured" by the FLF $C_{\tau,\sigma}(\boldsymbol{r})$.

The exact two-body density matrix $n_2(\xi,\zeta,\zeta',\xi')$ can be represented as a Hartree-Fock like contribution due to the one-body density matrix $n_1(\xi,\zeta)$, plus an irreducible two-body part $n_{\rm corr}(\xi,\zeta,\zeta',\xi')$,

$$\begin{aligned} n_2(\xi,\zeta,\zeta',\xi') &= \langle\Phi|\psi^\dagger(\xi)\psi^\dagger(\zeta)\psi(\zeta')\psi(\xi')|\Phi\rangle \\ &= \frac{1}{2}[n_1(\xi,\xi')n_1(\zeta,\zeta') \\ &\quad - n_1(\xi,\zeta')n_1(\zeta,\xi')] \\ &\quad + n_{\rm corr}(\xi,\zeta,\zeta',\xi'), \end{aligned} \tag{7}$$

$$\begin{aligned} n_1(\xi,\zeta) &= \langle\Phi|\psi^\dagger(\zeta)\psi(\xi))|\Phi\rangle \\ &= \sum_k n_k\phi_k(\xi)\phi_k^*(\zeta), \quad 0 \leqslant n_k \leqslant 1, \end{aligned} \tag{8}$$

$$\sum\!\!\!\!\!\!\int_\xi n_1(\xi,\xi) = \sum_k n_k = N,\ \langle\phi_k|\phi_l\rangle = \delta_{kl}, \tag{9}$$

$$n_{\rm ex}(\xi,\zeta,\zeta',\xi') = -\frac{1}{2}n_1(\xi,\zeta')n_1(\zeta,\xi'), \tag{10}$$

$$\sum\!\!\!\!\!\!\int_{\xi,\zeta} n_2(\xi,\zeta,\zeta,\xi) = \frac{N(N-1)}{2}, \tag{11}$$

$$\sum\!\!\!\!\!\!\int_{\xi,\zeta} n_{\rm corr}(\xi,\zeta,\zeta,\xi) = -\frac{1}{2}\sum_k n_k(1-n_k) \leqslant 0, \tag{12}$$

where N is the particle number, n_k and $\phi_k(\xi)$ are the canonical occupation probabilities and canonical single-particle wave functions [48], also known as natural orbitals [67,68], and ξ, ζ, ξ', ζ' are the particle coordinates $\xi = (\boldsymbol{r}, \sigma, \tau)$ and so forth. The irreducible two-body part of the two-body density matrix vanishes only in the case of a pure Hartree-Fock wave function, when $n_k = 0$ or 1 only, and it is only the second rank of the general many-body Born-Bogoliubov-Green-KirkwoodYvon (BBGKY) hierarchy of many-body reduced density matrices [69]. Obviously the solution for the entire chain of these different rank density matrices is more complicated than the solution of the corresponding many-body Schrödinger equation and in practice the BBGKY hierarchy of equations is truncated in practice at the one-, two-, or three-body levels at most [70–78]. The trace of the exchange + two-body irreducible density matrices is independent of the particle interaction

$$\sum\!\!\!\!\!\!\int_{\xi,\zeta} [n_{\rm ex}(\xi,\zeta,\zeta,\xi) + n_{\rm corr}(\xi,\zeta,\zeta,\xi)] \equiv -\frac{N}{2}, \qquad (13)$$

which justifies the Kohn-Sham introduction of the exchange-correlation energy density [79] and which naturally follows form the Kohn-Hohenberg theorem [80], which states that there is a one-to-one correspondence between the many-body wave function and the one-body density distribution. There are many methods suggested over the years to go the beyond the Hartree-Fock approximation, and perhaps the most common is the shell model in different incarnations, the most recent version being the valence-space in-medium similarity renormalization group method (VS-IMSRG) in nuclear physics, recently extended to atomic systems, see Ref. [81] where comparison with other approaches, such as coupled clusters and configuration interaction, many-body perturbation theory, and earlier references are available. IMSRG framework requires, however, the construction of operators for observables in a reduced space, in this case for the density matrix $n_2(\xi, \zeta, \zeta', \xi')$, which is not a simple and very transparent procedure [82]. The total, mean field, and correlation energies of a system with two-body interactions V_{2b} only are given by

$$E_{\rm tot} = {\rm Tr}(T n_1) + {\rm Tr}(n_2 V_{2b}), \qquad (14)$$

$$E_{\rm mf} = {\rm Tr}(n_{2,\rm mf} V_{2b}), \quad n_{2,\rm mf} = n_2 - n_{\rm corr}, \qquad (15)$$

$$E_{\rm corr} = {\rm Tr}(n_{\rm corr} V_{2b}), \qquad (16)$$

where T is the kinetic energy and the trace is an integral over all spatial coordinates and a sum over spin-isospin coordinates. The correlation energy is negative for a repulsive V_{2b} interaction and, together with the effect of the Fock contribution, leads to a bigger "Fermi hole," particularly in the case of a short-ranged repulsive interaction, as one would expect (assuming the single-particle occupation probabilities n_k do not change). The opposite happens in the case of an attractive interaction. Equation (13) suggests that the exchange and correlation effects act in opposite directions, a trend partially confirmed by microscopic calculations in the case of the homogeneous electron gas [78,83–87].

For the sake of the following argument I introduce the coupling constant λ of the two-body interaction λV_{2b}, which is negative for attractive and positive for repulsive particle-particle interaction. In general an arbitrary two-body interaction can have both attractive and repulsive parts, and for simplicity of the argument I define here a given two-body interaction λV_{2b} to be attractive if the interaction energy ${\rm Tr}(\lambda V_{2b} N_{\rm corr}) < 0$ in the limit $\lambda \to -\infty$ and repulsive otherwise. (For interactions with both repulsive and attractive parts this criterion should be applied with care.) The derivative of the correlation interaction energy with respect to the coupling constant (fixed n_k, thus first order perturbation in $\delta\lambda$)

$$\frac{dE_{\rm corr}}{d\lambda} = {\rm Tr}(V_{2b} n_{\rm corr}) \leqslant 0, \qquad (17)$$

describes the effect of presence of two-body correlations alone on the two-particle distributions in the presence of two-body interactions beyond the mean field. Equation (17) shows that with increasing strength λ, from very strong attractive to very strong repulsive interaction, the correlation energy $E_{\rm corr}$ decreases, which in the case of short-range interactions implies that the "Fermi hole" for two identical nucleons becomes bigger. The extreme values of the trace of ${\rm Tr}(n_{\rm corr})$, under the constraint $\sum_k n_k = N$, are

$$-\frac{N}{2} < -\frac{N}{2}\left(1 - \frac{N}{N_{\rm sp}}\right) \leqslant \sum\!\!\!\!\!\!\int_{\xi,\zeta} n_{\rm corr}(\xi,\zeta,\zeta,\xi) \leqslant 0, \qquad (18)$$

which are achieved for

$$n_k = \frac{N}{N_{\rm sp}}, \quad \text{for the minimum,} \qquad (19)$$

$$n_k = 0 \quad \text{or} \quad n_k = 1 \quad \text{for the maximum,} \qquad (20)$$

where $N_{\rm sp}$ is the number of single-particle states and which theoretically is infinite. The maximum value for ${\rm Tr}(n_{\rm corr}) = 0$ is achieved in the case of a Wigner crystal [36] for long-range very strong repulsive interactions (electron gas) [78,83–85] or in the case of a gas for short-ranged very strong repulsive interactions. The minimum value is attained for a Bardeen-Cooper-Schrieffer superconductor with very large attraction and zero spin polarization. In this limit, known as the Bore-Einstein condensate (BEC) state, the unpolarized Fermi system is a gas of highly bound dimers/Cooper pairs) with sizes much smaller than the average interparticle separation and these dimers repel each other [88,89]. In the limit of a Wigner gas (infinitely repulsive V_{2b}) ${\rm Tr}(n_{\rm ex}) \equiv 0$ and in the opposite BEC limit (infinitely attractive V_{2b}) ${\rm Tr}(n_{\rm ex}) = -N^2/2N_{\rm sp}$) is basically vanishing as well. In the case of attractive short-range interactions and finite spin polarization a wide range of phases are possible [39,62,90–97].

Since the main argument presented by Becke and Edgecombe [5] is based on the behavior of the two-body number density in the limit when $\boldsymbol{r}_1 - \boldsymbol{r}_2 \to \boldsymbol{0}$ and equal spin and isospins, it becomes obvious that merely the presence of Pauli correlations is not a sufficient argument to judge the probability to find another particle nearby in a nuclear medium, in particularly when they have different spins and/or isospins. Correlations induced by the strong particle interactions are crucial and their character depends of whether the interaction

is attractive or repulsive. The same rule in Eq. (12) for the two-body irreducible density matrix is expected to be satisfied for any accurate many-body wave function $|\Phi\rangle$ and since in the presence of interactions $\sum_k n_k(1-n_k) \neq 0$ it becomes obvious that the procedure suggested in Ref. [5] and used quite extensively in nuclear physics [13–30] cannot describe the clusterization of matter. The *ad hoc* procedure adopted in these nuclear studies of the FLF as $C_{n,\sigma}(\boldsymbol{r})C_{p,\sigma}(\boldsymbol{r})$ describes independent neutron and proton spin-number densities and "does not describe correlated neutron-proton subsystems." The recent study [29], pointing to the formation of α-like structures during nuclear fission, is a clear example why the FLF is such an inadequate measure, as the authors of this study where unable to even determine whether either two ^3H, ^{4}He, or ^{6}He are present in the neck region. At the same time proton and neutron density distributions presented in the study [29] fail to show the presence of any cluster either before, during, or after scission. One can fairly well decide that the clusterlike structures in the FLFs observed so far in these studies is fictitious or simply coincidental at best, since the FLF, which is a product $C_{n,\sigma}(\boldsymbol{r})C_{p,\sigma}(\boldsymbol{r})$, cannot and does not describe correlations between proton and neutron subsystems, unlike the irreducible two-body number density.

The *nn*- and *pp*-pairing correlations (with $S=0$ and $T=1$) alone may lead to a significant correction the FLF. Similarly, the role of the *np* SRCs in localization effects is another aspect (summed over isospin), which cannot be described with the nuclear FLF $C_{n,\sigma}(\boldsymbol{r})C_{p,\sigma}(\boldsymbol{r})$, but which can be simulated in a DFT framework by introducing a dynamic proton-neutron pairing [47,49]. The presence of pairing correlations leads to another complication, due to the fact that the gauge or particle conservation symmetry is broken. In the Hartree-Fock-Bogoliubov (HFB) approximation the two-body density matrix has the structure

$$\begin{aligned} n_2(\xi,\zeta,\zeta',\xi') &= \langle\Phi_{\rm HFB}|\psi^\dagger(\xi)\psi^\dagger(\zeta)\psi(\zeta')\psi(\xi')|\Phi_{\rm HFB}\rangle \\ &= \tfrac{1}{2}[n_1(\xi,\xi')n_1(\zeta,\zeta') - n_1(\xi,\zeta')n_1(\zeta,\xi')] \\ &\quad + \tfrac{1}{2}\kappa(\xi,\zeta)\kappa^*(\xi',\zeta'), \end{aligned} \tag{21}$$

where $\kappa(\xi,\zeta) = \langle\Phi_{\rm HFB}|\psi(\zeta)\psi(\xi)|\Phi_{\rm HFB}\rangle$ is the anomalous density-matrix and the corresponding irreducible two-body density matrix satisfies now the incorrect condition

$$\frac{1}{2}\sum\!\!\!\!\!\!\int_{\xi,\zeta} |\kappa(\xi,\zeta)|^2 \geqslant 0, \tag{22}$$

with an the opposite sign to Eq. (12) and as a result the corresponding two-body density matrix does not satisfy anymore the expected sum rule defined in Eq. (11). This aspect can be corrected only after the particle projection of the HFB many-wave function $\Phi_{\rm HFB}$ is performed. Consequently, the improved $\mathcal{D}_\tau(\boldsymbol{r})$ introduced in Eq. (6) cannot be expected to lead to a correct outcome in the presence of pairing correlations, unless particle projection is performed when evaluating the two-body density matrix. The particle projected occupation probabilities n_k and the corresponding particle projected HFB two-body density matrix $n_2(\xi,\zeta,\zeta',\xi')$ can be easily evaluated [98]. Pairing correlations are particular example when the irreducible density matrix $n_{\rm corr}(\xi,\zeta,\zeta',\xi')$ plays a large role and the role of exchange effects is reduced.

In summary, I have shown that the use of the FLF introduced by Becke and Edgecombe [5] and widely used in theoretical nuclear studies [13–30] is ill justified and cannot correctly describe clusterization effects, which require the knowledge of the irreducible two-body number density $n_{\rm corr}$ if a similar approach is adopted. The cases of the Wigner crystal and of the unitary Fermi gas in the BEC limit discussed above are clear examples where FLF fails to disentangle the "clusters." Another example would be a system of two dimers, one a relatively strongly bound "$S=1$ neutron dimer" and the other a relatively strongly bound "$S=1$ proton dimer," when either these two dimers repel in a trap or are weakly bound. Particularly large errors can be incurred when including pairing correlations within the energy density functional, which are ubiquitous nuclear systems discussed so far in literature, and which have a significant impact on various nuclear properties. The homogeneous electron gas [78,83–87], neutron matter with chiral effective two- and three-body interactions [50–52] and the dilute Fermi gas, particularly in the unitary and BEC regime, are examples of microscopic well-studied systems [10,12,37,39,62,88–97], where the role of both exchange, correlations, and temperature effects are important. The case of quarks localized inside hadrons is the most notable example of the dominant role of strong correlations effects.

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