Local-Scaling Transformation version of Density Functional Theory

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I. ZERO-TEMPERATURE

A. Local-scaling transformations

The well-known *uniform* scaling transformation

$$\mathbf{r} \to \lambda \mathbf{r}\,, \tag{1}$$

where λ is a constant, is frequently used in physics. The *local-scaling* transformation[1,3] (LST) is a generaliztaion of the *uniform* transformation defined by Eq. (1). *Isotropic* LSTs modify the vector $\mathbf{r}$ at each point of space $\mathcal{R}^3$ according to

$$\mathbf{r} \xrightarrow{\hat{f}} \mathbf{f}(\mathbf{r}) = \lambda(\mathbf{r})\mathbf{r} = \left(\lambda(\mathbf{r})x, \lambda(\mathbf{r})y, \lambda(\mathbf{r})z\right), \tag{2}$$

where $\hat{f}$ is an operator associated with this transformation. *Isotropic* transformations keep the direction of the transformed vector $\mathbf{f}(\mathbf{r})$ constant.

LST relates two densities, an initial or "generating" density $\rho_g(\mathbf{r})$ and a final density $\rho(\mathbf{r})$:

$$\rho(\mathbf{r}) = J\big(\mathbf{f}(\mathbf{r});\mathbf{r}\big)\rho_g\big(\mathbf{f}(\mathbf{r})\big) \tag{3}$$

where $J\big(\mathbf{f}(\mathbf{r});\mathbf{r}\big)$ is the Jacobian.

For isotropic transformations the Jacobian is

$$\begin{aligned} J(\lambda(\mathbf{r})\mathbf{r};\mathbf{r}) &= \begin{bmatrix} \frac{\partial\lambda(\mathbf{r})x}{\partial x} & \frac{\partial\lambda(\mathbf{r})y}{\partial x} & \frac{\partial\lambda(\mathbf{r})z}{\partial x} \\ \frac{\partial\lambda(\mathbf{r})x}{\partial y} & \frac{\partial\lambda(\mathbf{r})y}{\partial y} & \frac{\partial\lambda(\mathbf{r})z}{\partial y} \\ \frac{\partial\lambda(\mathbf{r})x}{\partial z} & \frac{\partial\lambda(\mathbf{r})y}{\partial z} & \frac{\partial\lambda(\mathbf{r})z}{\partial z} \end{bmatrix} \\ &= \lambda^3(\mathbf{r})[1 + \mathbf{r}\cdot\nabla_{\mathbf{r}}\ln\lambda(\mathbf{r})]. \end{aligned} \tag{4}$$

From Eqs. (3) and (4) we obtain the following expression for $\lambda(\mathbf{r})$

$$\lambda(\mathbf{r}) = \left[\frac{\rho(\mathbf{r})}{\rho_g\big(\lambda(\mathbf{r})\mathbf{r}\big)\big(1 + \mathbf{r}\cdot\nabla_{\mathbf{r}}\ln\lambda(\mathbf{r})\big)}\right]^{1/3}. \tag{5}$$

Equation (5) is a first order differential equation for the transformation function $\lambda(\mathbf{r})$. Introducing the function $f(\mathbf{r})$ by means of the following equivalence,

$$\lambda(\mathbf{r}) = \frac{f(\mathbf{r})}{r}, \tag{6}$$

we can rewrite Eq. (5) as:

$$\frac{\rho(\mathbf{r})}{\rho_g\big(\mathbf{f}(\mathbf{r})\big)} = \frac{1}{r^3}\mathbf{r}\cdot\nabla_{\mathbf{r}}f^3(\mathbf{r}). \tag{7}$$

B. LST version of DFT

Consider application of the LST operator $\hat{f}$ Eq. (2) to each of the coordinates of the N-electron (general antisymmetric or just simple Slater determinant) wave function $\Phi_g(\mathbf{r}_1,...,\mathbf{r}_N)$. Equivalently, with every coordinate transformation operator $\hat{f}$ we can associate N-particle unitary operator $\widehat{\mathcal{U}}_f$ which acts on the Hilbert space $\mathcal{H}$ of N-particle wave functions. The resulting wave function $\Phi_\rho(\mathbf{r}_1,...,\mathbf{r}_N)$ (the reason of using subindex ρ will be clear after next equation) is given by

$$\begin{aligned} \Phi_\rho(\mathbf{r}_1,...,\mathbf{r}_N) &\equiv \prod_{i=1}^{\mathrm{N}}[J\big(\mathbf{f}(\mathbf{r}_i);\mathbf{r}_i\big)]^{1/2}\ \underbrace{\hat{f}...\hat{f}}_{\mathrm{N-times}}\ \Phi_g(\mathbf{r}_1,...,\mathbf{r}_N) \\ &= \widehat{\mathcal{U}}_f\Phi_g(\mathbf{r}_1,...,\mathbf{r}_N) \\ &= \prod_{i=1}^{\mathrm{N}}[J\big(\mathbf{f}(\mathbf{r}_i);\mathbf{r}_i\big)]^{1/2}\Phi_g(\mathbf{f}(\mathbf{r}_1),...,\mathbf{f}(\mathbf{r}_N)) \end{aligned} \tag{8}$$

The density corresponding to the transformed wave function Φ_ρ may be evaluated as usual

$$\mathrm{N}\int d\mathbf{r}_2...\int d\mathbf{r}_N|\Phi_\rho(\mathbf{r}_1,...,\mathbf{r}_N)|^2 = J\big(\mathbf{f}(\mathbf{r}_1);\mathbf{r}_1\big)\underbrace{\mathrm{N}\int d\mathbf{f}(\mathbf{r}_2)...\int d\mathbf{f}(\mathbf{r}_N)|\Phi_g(\mathbf{f}(\mathbf{r}_1),...,\mathbf{f}(\mathbf{r}_N))|^2}_{\rho_g(\mathbf{f}(\mathbf{r}_1))} = \rho(\mathbf{r}_1)\,, \tag{9}$$

where we used Eq. (3). What we have? For given ρ there are infinite number of Φ_g (and corresponding ρ_g) which may be used in LST defined by Eqs. (3) and (8). As a result an infinite number of functions Φ_ρ yielding the same density ρ will be constructed. Function Φ_ρ is an implicit functional of the density ρ, i.e.

$$\Phi_\rho(\mathbf{r}_1, ..., \mathbf{r}_N) = \Phi([\rho]; \mathbf{r}_1, ..., \mathbf{r}_N)\,. \tag{10}$$

Function Φ_ρ may be used in Levy-Lieb constrained search

$$F[\rho] = \min_{\Phi_\rho} < \Phi_\rho | \hat{T} + \hat{V}_{ee} | \Phi_\rho >\,, \tag{11}$$

where $F[\rho]$ searches all Φ_ρ, which is equivalent to search all Φ_g in LST defined by Eqs. (3)-(8). Then the ground state energy is expressed as

$$\begin{aligned} E_{\mathrm{GS}} &= \min_\rho \; E[\rho] \qquad\qquad (12) \\ &= \min_\rho \left[\min_{\Phi_\rho} < \Phi_\rho | \hat{T} + \hat{V}_{ee} | \Phi_\rho > + \int v(\mathbf{r}) \rho(\mathbf{r}) d\mathbf{r} \right], \end{aligned}$$

which is a search over all N-representable densities.

Noninteracting (in case if Φ is Slater determinant) kinetic energy functional is

$$T_{\mathrm{s}}[\rho] = \min_{\Phi_\rho} < \Phi_\rho | \hat{T} | \Phi_\rho >\,, \tag{13}$$

C. LST of one-particle orbitals and reduced density matrices

LSTs can be used to generate from an initial orbital set $\{\phi_{g,k}(\mathbf{r})\}_{k=1}^N$ a new set of transformed orbitals:

$$\begin{aligned} \phi_{\rho,k}(\mathbf{r}) = \widehat{\mathcal{U}}_f \phi_{g,k}(\mathbf{r}) &= \big[J(\mathbf{f}(\mathbf{r}); \mathbf{r}) \big]^{1/2} \phi_{g,k}\big(\mathbf{f}(\mathbf{r})\big) \\ &= \left[\frac{\rho(\mathbf{r})}{\rho_g\big(\mathbf{f}(\mathbf{r})\big)} \right]^{1/2} \phi_{g,k}\big(\mathbf{f}(\mathbf{r})\big), \end{aligned} \tag{14}$$

where new transformed orbitals have following properties, ortho-normality (if generating set is ortho-normal)

$$< \phi_{\rho,i} | \phi_{\rho,j} > = < \phi_{g,i} | \phi_{g,j} > = \delta_{ij}\,, \tag{15}$$

electron density for transformed orbitals by construction is equal ρ

$$\sum_{k=1}^{\mathrm{N}} |\phi_{\rho,k}(\mathbf{r})|^2 = \rho(\mathbf{r})\,. \tag{16}$$

Relation between generating one-particle reduced density matrix (1-RDM)

$$D_g^{(1)}(\mathbf{r}; \mathbf{r}') = \sum_{k=1}^{\mathrm{N}} \phi_{g,k}(\mathbf{r}) \phi_{g,k}(\mathbf{r}')\,, \tag{17}$$

and transformed 1-RDM may be found from Eq. (14)

$$D_\rho^{(1)}(\mathbf{r}; \mathbf{r}') = \left[\frac{\rho(\mathbf{r})}{\rho_g\big(\mathbf{f}(\mathbf{r})\big)} \right]^{1/2} \left[\frac{\rho(\mathbf{r}')}{\rho_g\big(\mathbf{f}(\mathbf{r}')\big)} \right]^{1/2} D_g^{(1)}(\mathbf{f}(\mathbf{r}); \mathbf{f}(\mathbf{r}'))\,. \tag{18}$$

Similar relation is hold for two-particle RDM. For simplicity consider only diagonal parts of 2-RDMs

$$D_\rho^{(2)}(\mathbf{r}, \mathbf{r}'; \mathbf{r}, \mathbf{r}') = \frac{\rho(\mathbf{r})}{\rho_g\big(\mathbf{f}(\mathbf{r})\big)} \frac{\rho(\mathbf{r}')}{\rho_g\big(\mathbf{f}(\mathbf{r}')\big)} D_g^{(2)}(\mathbf{f}(\mathbf{r}), \mathbf{f}(\mathbf{r}'); \mathbf{f}(\mathbf{r}), \mathbf{f}(\mathbf{r}'))\,, \tag{19}$$

where $D_g^{(2)}(\mathbf{f}(\mathbf{r}), \mathbf{f}(\mathbf{r}'); \mathbf{f}(\mathbf{r}), \mathbf{f}(\mathbf{r}'))$ is 2-RDM constructed from generating set of orbitals $\{\phi_{g,i}(\mathbf{f}(\mathbf{r})\}_{i=1}^N$.

D. Approximate kinetic energy functionals

Kinetic energy corresponding to the transformed 1-RDM Eq. (18) is an inexplicit functional of the density ρ and has the following structure

$$\begin{aligned} T_{\mathrm{s}}[\rho, \{\phi_{g,i}\}_{i=1}^N] &= \int \frac{1}{2} \nabla_{\mathbf{r}} \nabla_{\mathbf{r}'} D_\rho^{(1)}(\mathbf{r}; \mathbf{r}') \Big|_{\mathbf{r}'=\mathbf{r}} d\mathbf{r} \\ = T_W[\rho] &+ \int \rho^{5/3}(\mathbf{r}) \bigg[\Big(1 + \mathbf{r} \cdot \nabla \ln \lambda(\mathbf{r}) \Big)^{4/3} \tau_N([\{\phi_{g,i}\}]; \mathbf{r}) \\ &+ \Big(1 + \mathbf{r} \cdot \nabla \ln \lambda(\mathbf{r}) \Big)^{-2/3} \kappa_N([\{\phi_{g,i}\}]; \mathbf{r}) d\mathbf{r} \bigg]\,, \end{aligned} \tag{20}$$

where τ_N and κ_N are modulating factors which have a compact expression in the terms of generating orbital set $\{\phi_{g,i}(\mathbf{f}(\mathbf{r}))\}_{i=1}^N$. Dependence on the generating orbitals $\phi_{g,i}$ in Eq. (20) may be considered as parametric. Once the generating set chosen, Eq. (20) is transformed into an approximate kinetic energy functional of the density ρ.

Unfortunately the LST differential equation Eq. (7) for the transformed variable $\mathbf{f}(\mathbf{r})$ can be solved in analytical form only for simplest forms of generating density ρ_g. As an example, consider a set of one-dimensional plane-waves, $\psi_{g,k}(r) \sim \exp(ikf(r))$, as generating orbitals. Transformed orbitals in polar coordinates can be expressed as

$$\psi_{\rho,k}(r, \theta, \phi) = \sqrt{\frac{\rho(r, \theta, \phi)}{N}} \exp\left[i \Big(k - \frac{N+1}{2} \Big) f(r) \right], \tag{21}$$

which are known as "equidensity" orbitals (see[4]). 1-RDM corresponding to the transformed set Eq. (21) is[1]

$$D_\rho^{(1)}(\mathbf{r}; \mathbf{r}') = \frac{1}{N} \sqrt{\rho(\mathbf{r}) \rho(\mathbf{r}')} \frac{\sin \frac{N}{2} F(r, r')}{\sin \frac{1}{2} F(r, r')}\,, \tag{22}$$

Kinetic energy evaluated with transformed 1-RDM is an explicit functional of the density ρ

$$T_{\mathrm{s}}[\rho] = T_{\mathrm{W}}[\rho] + \Big(\frac{3}{4\pi} \Big)^{1/3} 2\pi^3 \Big(1 - \frac{1}{N^2} \Big) \int \rho^{5/3} d\mathbf{r}\,. \tag{23}$$

II. FINITE-TEMPERATURE

A. Quantum statistical variational principle

Quantum statistical variational principle for an N-particle system with Hamiltonian $\widehat{H}$ and number operator $\widehat{N}$, chemical potential μ at temperature T states that the Gibbs free energy

$$\mathcal{G}[\hat{\rho}] = Tr\Big(\hat{\rho}[\widehat{H} - \mu\widehat{N} + \beta^{-1}\ln\hat{\rho}]\Big)\,, \tag{24}$$

is minimized by the grand canonical density matrix operator

$$\hat{\rho}_0 = \frac{\exp(-\beta[\widehat{H} - \mu\widehat{N}])}{Tr(\exp(-\beta[\widehat{H} - \mu\widehat{N}]))}\,, \tag{25}$$

where $\beta = 1/(k_{\rm B}{\rm T})$.

All variational quantum statistical techniques are obtained by means of different trial density operators on which the grand canonical potential Eq. (24) is minimized[2]. As an example, the temperature-dependent Hartree-Fock (THF) method uses trial density operator of the following form

$$\hat{\rho} = \frac{\exp(-\sum_{ij} a_i^+\gamma_{ij}a_j)}{Tr(\exp(-\sum_{ij} a_i^+\gamma_{ij}a_j))}\,, \tag{26}$$

where a_i^+, a_i are particle creation and annihilation operators respectively, and γ_{ij} is an arbitrary Hermitian matrix which always can be diagonalized, $\gamma_{ij} = \gamma_i\delta_{ij}$.

B. Finite temperature LST

Consider generating (a given) density matrix operator, $\hat{\rho}_g$. Generating (initial) density operator uniquely defines the corresponding initial 1- and 2- RDMs

$$D_g^{(1)}(\mathbf{r};\mathbf{r}') = Tr\Big(\hat{\rho}_g\psi_g^+(\mathbf{r})\psi_g(\mathbf{r}')\Big) = \sum_\alpha n_\alpha\phi_{g,\alpha}^*(\mathbf{r})\phi_{g,\alpha}(\mathbf{r}')\,, \tag{27}$$

and

$$\begin{aligned} D_g^{(2)}(\mathbf{r},\mathbf{r}';\mathbf{r},\mathbf{r}') &= Tr(\hat{\rho}_g\psi_g^+(\mathbf{r})\psi_g^+(\mathbf{r}')\psi_g(\mathbf{r})\psi_g(\mathbf{r}')) \qquad (28)\\ &= \sum_{\alpha\beta}\Gamma_{\alpha\beta}\phi_{g,\alpha}^*(\mathbf{r})\phi_{g,\beta}^*(\mathbf{r}')\phi_{g,\alpha}(\mathbf{r})\phi_{g,\beta}(\mathbf{r}')\,, \end{aligned}$$

where $\psi_g^*(\mathbf{r}) = \sum_\alpha \phi_{g,\alpha}^*(\mathbf{r})a_\alpha^+$ and $\psi_g(\mathbf{r}) = \sum_\alpha \phi_{g,\alpha}(\mathbf{r})a_\alpha$ are particle field creation and annihilation operators respectively, $\phi_{g,\alpha}(\mathbf{r})$ is the orthonormal generating (initial) set of one-electron states. The local density distribution is

$$\rho_g(\mathbf{r}) = D_g^{(1)}(\mathbf{r};\mathbf{r}) = \sum_\alpha n_\alpha|\phi_{g,\alpha}(\mathbf{r})|^2\,. \tag{29}$$

Let further consider density matrix operator transformed by $\widehat{\mathcal{U}}_{f[\rho]}$, where $f \equiv f[\rho]$ is defined by Eq. (3)

$$\hat{\rho}[\rho] = \widehat{\mathcal{U}}_{f[\rho]}^+\hat{\rho}_g\widehat{\mathcal{U}}_{f[\rho]}\,. \tag{30}$$

It is straightforward to check that the corresponding 1- and 2-RDMs are related to the generating $D_g^{(1)}$ and $D_g^{(2)}$ by the same Eqs. (17)-(18) as in the zero-temperature case. The transformed 1-RDM, for example, is

$$\begin{aligned} D_\rho^{(1)}(\mathbf{r};\mathbf{r}') &= Tr\Big(\hat{\rho}[\rho]\psi_g^+(\mathbf{r})\psi_g(\mathbf{r}')\Big) \qquad (31)\\ &= \left[\frac{\rho(\mathbf{r})}{\rho_g(\mathbf{f}(\mathbf{r}))}\right]^{1/2}\left[\frac{\rho(\mathbf{r}')}{\rho_g(\mathbf{f}(\mathbf{r}'))}\right]^{1/2}\\ &\times \sum_\alpha n_\alpha\phi_{g,\alpha}^*(\mathbf{f}(\mathbf{r}))\phi_{g,\alpha}(\mathbf{f}(\mathbf{r}'))\\ &= \left[\frac{\rho(\mathbf{r})}{\rho_g(\mathbf{f}(\mathbf{r}))}\right]^{1/2}\left[\frac{\rho(\mathbf{r}')}{\rho_g(\mathbf{f}(\mathbf{r}'))}\right]^{1/2} D_g^{(1)}(\mathbf{f}(\mathbf{r});\mathbf{f}(\mathbf{r}'))\,. \end{aligned}$$

The local density distribution corresponding to transformed density matrix operator $\hat{\rho}[\rho]$ is

$$\begin{aligned} D_\rho^{(1)}(\mathbf{r};\mathbf{r}) &= \left[\frac{\rho(\mathbf{r})}{\rho_g(\mathbf{f}(\mathbf{r}))}\right]^{1/2}\left[\frac{\rho(\mathbf{r})}{\rho_g(\mathbf{f}(\mathbf{r}))}\right]^{1/2} D_g^{(1)}(\mathbf{f}(\mathbf{r});\mathbf{f}(\mathbf{r}))\\ &= \rho(\mathbf{r})\,. \end{aligned} \tag{32}$$

C. The Gibbs free-energy functional

Let a given generating density matrix operator $\hat{\rho}_g$ be fixed. Consider a "class" of transformed density matrix operators $\hat{\rho}[\rho] = \widehat{\mathcal{U}}_{f[\rho]}^+\hat{\rho}_g\widehat{\mathcal{U}}_{f[\rho]}$ as trial density operators. The Gibbs free energy Eq. (24) evaluated with trial density operator $\hat{\rho}[\rho]$ transforms into a functional of the local density distribution ρ

$$\mathcal{G}[\rho] \equiv \mathcal{G}[\hat{\rho}[\rho]] = Tr\Big(\hat{\rho}[\rho][\widehat{H} - \mu\widehat{N} + \beta^{-1}\ln\hat{\rho}[\rho]]\Big)\,. \tag{33}$$

At fixed $\hat{\rho}_g$, minimization w.r.t. trial density operator $\hat{\rho}[\rho]$ in Eq. (33) is equivalent to minimization w.r.t. local desnity distribution ρ. To get a global minima, the minimization w.r.t. $\hat{\rho}_g$ also should be performed

$$\begin{aligned} \mathcal{G}_0 &= \min_{\hat{\rho}_g}\Big(\min_{\hat{\rho}}\mathcal{G}[\hat{\rho}[\rho]]\Big)\\ &= \min_{\hat{\rho}_g}\Big(\min_{\rho}\mathcal{G}[\rho]\Big)\,. \end{aligned} \tag{34}$$

The Gibbs free energy in Eq. (33) is a functional of the local density distribution ρ with following components

$$\mathcal{G}[\rho] = \mathcal{F}[\rho] - \mu N[\rho] = \mathcal{E}[\rho] - {\rm T}\mathcal{S}[\rho] - \mu N[\rho]\,, \tag{35}$$

where

$$\mathcal{S}[\rho] = -Tr\Big(\hat{\rho}[\rho]\ln\hat{\rho}[\rho]]\Big)\,, \tag{36}$$

is the entropy density functional and

$$N[\rho] = -Tr\Big(\hat{\rho}[\rho]\widehat{N}\Big) = \int \rho(\mathbf{r})d\mathbf{r}\,, \tag{37}$$

is the number of electrons. $\mathcal{F}[\rho] = \mathcal{E}[\rho] - \mathrm{T}\mathcal{S}[\rho]$ is the Helmholtz free energy, where

$$\begin{aligned} \mathcal{E}[\rho] &= \mathcal{T}[\rho] + \mathcal{U}[\rho] \\ \mathcal{T}[\rho] &= Tr\Big(\hat{\rho}[\rho]\widehat{T}\Big) \\ \mathcal{U}[\rho] &= Tr\Big(\hat{\rho}[\rho]\widehat{U}\Big)\,, \end{aligned} \tag{38}$$

are density functionals of the internal energy and its components. All these functionals are evaluated using transformed 1- and 2-RDM Eqs. (31)-(32).

Consider again 1-RDM and an example of the kinetic energy functional. Transformed 1-RDM Eq. (31) may be presented as

$$\begin{aligned} D^{(1)}_{\rho}(\mathbf{r};\mathbf{r}') &= Tr\Big(\hat{\rho}[\rho]\psi^{+}_{g}(\mathbf{r})\psi_{g}(\mathbf{r}')\Big) \qquad (39) \\ &= \left[\frac{\rho(\mathbf{r})}{\rho_g\big(\mathbf{f}(\mathbf{r})\big)}\right]^{1/2}\left[\frac{\rho(\mathbf{r}')}{\rho_g\big(\mathbf{f}(\mathbf{r}')\big)}\right]^{1/2} \\ &\times \sum_{\alpha} n_\alpha \phi^{*}_{g,\alpha}(\mathbf{f}(\mathbf{r}))\phi_{g,\alpha}(\mathbf{f}(\mathbf{r}')) \\ &\equiv \sum_{\alpha} n_\alpha \phi^{*}_{\rho,\alpha}(\mathbf{f}(\mathbf{r}))\phi_{\rho,\alpha}(\mathbf{f}(\mathbf{r}'))\,, \qquad (40) \end{aligned}$$

where transformed density-dependent single particle wave functions were defined as

$$\phi_{\rho,\alpha}(\mathbf{r}) \equiv \phi_\alpha([\rho];\mathbf{r}) = \left[\frac{\rho(\mathbf{r})}{\rho_g\big(\mathbf{f}(\mathbf{r})\big)}\right]^{1/2}\phi_{g,\alpha}(\mathbf{f}(\mathbf{r}))\,, \tag{41}$$

and the kinetic energy functional Eq. (38) become

$$\begin{aligned} \mathcal{T}[\rho] &= Tr\Big(\hat{\rho}[\rho]\widehat{T}\Big) \\ &= \int \frac{1}{2}\nabla_{\mathbf{r}}\nabla_{\mathbf{r}'}D^{(1)}_{\rho}(\mathbf{r};\mathbf{r}')\Big|_{\mathbf{r}'=\mathbf{r}}d\mathbf{r} \\ &= \sum_{\alpha} n_\alpha \int \frac{1}{2}|\nabla\phi_{\rho,\alpha}(\mathbf{r})|^2 d\mathbf{r} \\ &= \int \Big(\tau_{\mathrm{W}}([\rho];\mathbf{r}) + \tau_\gamma([\rho];\mathbf{r})\Big)d\mathbf{r}\,, \end{aligned} \tag{42}$$

where

$$\tau_{\mathrm{W}}([\rho];\mathbf{r}) = \frac{1}{8}\frac{|\nabla\rho(\mathbf{r})|^2}{\rho(\mathbf{r})}\,, \tag{43}$$

and

$$\tau_\gamma([\rho];\mathbf{r}) = \rho(\mathbf{r})\Big[\nabla_{\mathbf{r}}\nabla_{\mathbf{r}'}\gamma_g(f([\rho];\mathbf{r}); f([\rho];\mathbf{r}'))\Big]_{\mathbf{r}'=\mathbf{r}}\,. \tag{44}$$

In last equation

$$\gamma_g(\mathbf{r};\mathbf{r}') = \frac{D^{(1)}_{g}(\mathbf{r};\mathbf{r}')}{\sqrt{\rho_g(\mathbf{r})\rho_g(\mathbf{r}'))}}\,, \tag{45}$$

is the reduced density matrix introduced by Gazquer and Ludeña[5] which satisfy the condition $\gamma_g(\mathbf{r};\mathbf{r}) = 1$ for arbitrary $\mathbf{r} \in \mathcal{R}^3$.

III. CONCLUSIONS

- LST approximate functionals are N-representable by construction with all good consequences of that fact.
- LST functional of total energy (T=0 case) or free energy (finite-T case) is in principle orbital-free.
- Evaluation of functional derivative of the LST functional w.r.t. density is not trivial procedure because the energy components are not explicit functionals of the density.
- Practical result may be obtained for cases of simple forms of generating orbitals $\phi_{g,k}$ when LST Eq. (7) can be solved analytically.
- LST functionals might be useful for study of formal properties (constraints) on exact functionals. In particular, I guess they might be used to study bounds tighten bounds for KE energy for example.
- LST method was applied for study correlation (when the generating wave function Ψ_g was explicitly correlated N-electron wave function. Extension to finite-T case does not seems to be trivial.

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