Current Densities in Density Functional Theory

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and

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The Old and The New Problems

Density functionalists, and others, like to work with Hartree-Fock theory, which means working with determinantal functions (ignoring spin here for simplicity)

$$\Psi(\vec{x}_1, \vec{x}_2, \dots, \vec{x}_N) = \det\{\phi_i(\vec{x}_j)\}_{i,j=1}^N \quad \phi_i \text{ orthonormal in } L^2(\mathbb{R}^3).$$

This Ψ has a kinetic energy T, density $\rho(x)$, and current $\vec{j}(\vec{x})$

$$T = \sum_{i=1}^N \int |\nabla\phi_i(\vec{x})|^2 \mathrm{d}\vec{x}, \;\; \rho(\vec{x}) = \sum_{i=1}^N |\phi_i(\vec{x})|^2, \;\; \vec{j}(\vec{x}) = \tfrac{1}{2i} \sum_{i=1}^N \phi_i^*(\vec{x})\nabla\phi_i(\vec{x}) - \phi_i(\vec{x})\nabla\phi_i^*(\vec{x})$$

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Old Question: Given $\rho(\vec{x}) \geq 0$ with $\int \rho = N$ does there exist a finite energy Ψ?
The answer is YES, but the upper bound is lousy. (Harriman 1981, Lieb 1982) The idea goes back to March & Young 1958 who showed that in 1D there is always a Ψ such that

$$T \leq \int |\nabla\sqrt{\rho(\vec{x})}|^2 \mathrm{d}\vec{x} + C^{\text{semiclassic}} \int \rho(\vec{x})^{1+2/D} \mathrm{d}\vec{x}$$

and conjectured the same for $D > 1$. This conjectured bound is still an open problem!!

New Question : Given *both* $\rho(\vec{x})$ *and* $\vec{j}(\vec{x})$ does there exist a finite energy Ψ? (in 3D, of course.) Our answer is:

- 1. Define the velocity field, $\vec{v}(\vec{x}) = \vec{j}(\vec{x})/\rho(\vec{x})$.
- 2. If $\vec{v}$ is *curl-free* then the answer is YES. The proof of this is closely related to the proof of the 'old question' and the estimate for T is just as bad.
- 3. If curl $\vec{v} \neq 0$ the answer is YES if $N \geq 4$, (and with some mild conditions on the decay of $\vec{j}(\vec{x})$ and its derivatives). *We have no bound for T, however, only that it is finite!* This took us 6 years because we needed one small lemma to finish the construction, namely the smooth generalization of the Hobby-Rice theorem. This was proved by L & Lazarev and will be described later.
- 4. If curl $\vec{v} \neq 0$ and $N = 2$ the answer is MAYBE. We give examples where there is a Ψ and where there assuredly is none.
- 5. If curl $\vec{v} \neq 0$ and $N = 3$ the question is open.

Solution When v is Curl-Free, including $\vec{v} = 0$

Recall $\vec{v}(\vec{x}) = \vec{j}(\vec{x})/\rho(\vec{x})$. In this case we can write $\vec{v} = \nabla\tau$. Let us write

$$\phi_i(\vec{x}) = \psi_i(\vec{x}) \exp[i\tau(\vec{x})/N], \qquad \text{with } \psi_i \text{ orthonormal.}$$

Then the ϕ_i are O.N. as well. The condition on the current is satisfied if the current of $\det\psi_i$ is 0. All that remains is to make the density of the ψ_i equal to ρ. I.e., the problem reduces to the $\vec{v} = 0$ question solved 3 decades ago. This is accomplished as follows: Let $\vec{x} = (x^1,\, x^2,\, x^3)$.

$$f(x^3) = \tfrac{2\pi}{N} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{x^3} \rho(s,t,u)\, \mathrm{d}s\, \mathrm{d}t\, \mathrm{d}u$$

and $\psi_k(\vec{x}) = \sqrt{\rho(\vec{x})/N}\ \exp\{i\,k\,f(x^3)\}$ with $k = -N/2, \dots N/2$. These are O.N. because (after changing variables) $\int \exp\{imf(x^3)\}\rho(\vec{x})/N\,\mathrm{d}\vec{x} = \int_0^1 \exp\{i2\pi my\}\,\mathrm{d}y = 0$ if $m \neq 0$.

It is not too hard to prove that T is bounded by a constant ($\sim N$) times the March-Young conjecture plus the expected quantity $\int |\vec{j}(\vec{x})|^2/\rho(\vec{x})\ \mathrm{d}\vec{x}$.

When $\vec{v} = \vec{j}/\rho$ is Not Curl-Free and $N \geq 4$

Full disclosure: We will need that $\nabla \times \vec{v}$ and its derivatives vanish faster than $|x|^{-3}$ at ∞.

Step 1. Fixing $\rho_j,\ j = 1, \ldots, N$: Let $\xi \vec{x}) = \frac{1}{m} \int_{-\infty}^{x} \frac{1}{(1+y^2)^{(1+\varepsilon)/2}} \mathrm{d}y / \int_{-\infty}^{\infty} \frac{1}{(1+y^2)^{(1+\varepsilon)/2}} \mathrm{d}y$

$$\rho_1(\vec{x}) = \tfrac{1}{N} \xi(x^1 + \alpha)\rho(\vec{x}) \tag{1}$$

$$\rho_2(\vec{x}) = \tfrac{1}{N-1} \xi(x^1 + \beta)(\rho(\vec{x}) - \rho_1(\vec{x}))$$

$$\rho_3(\vec{x}) = \tfrac{1}{N-2} \xi(x^2 + \gamma)(\rho(\vec{x}) - \rho_1(\vec{x}) - \rho_2(\vec{x}))$$

$$\rho_i(\vec{x}) = \tfrac{1}{N-3} \widehat{\rho}(\vec{x}) = \tfrac{1}{N-3}(\rho(\vec{x}) - \rho_1(\vec{x}) - \rho_2(\vec{x}) - \rho_3(\vec{x}))), \quad 4 \leq i \leq N.$$

and we can easily fix $\alpha,\ \beta,\ \gamma$ uniquely so that all $\rho_1 \cdots \rho_N$ are normalized.

We write $\phi_i(\vec{x}) = \sqrt{\rho_i(\vec{x})}\exp\{i\chi_i(\vec{x})\}$. The conditions on χ_i are

$$\textstyle\sum_{i=1}^{3} \rho_i \nabla \chi_i + \rho_4 \nabla (\sum_{i=4}^{N} \chi_i) = \vec{j} = \ \rho \vec{v}. \qquad (*)$$

Step 2: We can solve equation (*) for $\chi_i,\ \chi_2,\ \chi_3,\ \text{and}\ \sum_4^N \chi_i$ by integrating the equations and noting the important facts that ρ_1 and ρ_2 depend only on x^1 while ρ_3 depends only on $x^1,\ x^2$. There is some freedom in the solution, which will be exploited later. This freedom is parametrized by 3 functions $h_i(x^1),\ i = 1, 2, 3$ of x^1 alone.

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Step 3: We now want to choose the $\chi_i,\ i = 4, ..., N$ so that $\sum_4^N \chi_i$ equals the required value and so that the $\phi_i,\ i = 4, ..., N$ are orthonormal. But this is *deja vu all over again,* namely it is essentially the old problem when the density is $(N-3)\rho_4$.

What is left is, somehow, to make $\phi_1,\ \phi_2,\ \phi_3$ orthogonal to each other and to the $\phi_i,\ i = 4, ..., N$. This is the next step.

Orthogonality and the Hobby-Rice Theorem

Step 4: It turns out that for $4 \leq i \leq N$ the inner product (after integration $\mathrm{d}x^2\mathrm{d}x^3$ and lots of calculations) has the form:

$$\langle \phi_3, \phi_i \rangle = \int_{\mathbb{R}} \exp\{ih_3(x^1)\} F_i(x^1)\mathrm{d}x^1$$

The function h_3 is arbitrary. Wouldn't it be nice if, given any (N-3) (complex) L^1 functions F_i one could always find a real, C_c^∞ function h_3 that makes this integral vanish for all i ?

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The function h_3 is arbitrary. Wouldn't it be nice if, given any (N-3) (complex) L^1 functions F_i one could always find a real, C_c^∞ function h_3 that makes this integral vanish for all i ? Yes, indeed, it is true! This is the smoothed Hobby-Rice Theorem. (L and Oleg Lazarev)

The H-R theorem says: *Given n real functions $f_1, ..., f_n$ in $L^1(\mathbb{R})$ there is a function g that takes values ± 1 (in at most n intervals) such that*

$$\int_{\mathbb{R}} g(x^1)^* f_i(x^1)\mathrm{d}x^1 = 0$$

In other words, n different loaves of bread can be *simultaneouly* cut into 2 equal weight pieces by at most n cuts! The proof by Pinkus cleverly uses the Borsuk Antipodality Theorem.

Denouement!

The smooth H-R theorem says that the same thing can be accomplished by a *smooth function* $g(x) = e^{ih(x)}$ that takes values on the unit circle in $\mathbb{C}$. Moreover, we can let the f_j be complex-valued. The H-R $g(x) = \pm 1$ also takes values on the unit circle in $\mathbb{C}$, but it is not smooth.

We now see that by proper choice of $h_3(x)$, $\langle \phi_3,\ \phi_1 \rangle = 0$ for $4 \leq i \leq N$. We need h_3 to be smooth so that $\int |\nabla \phi_3|^2 < \infty$, but we have no way to estimate this integral.
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The next step is to repeat the argument for the N-2 inner products $\langle \phi_2, \phi_i \rangle$ with $3 \leq i \leq N$, by using H-R to find an $h_2(x)$. Finally, we use H-R again to make $\langle \phi_1, \phi_i \rangle = 0$ for $2 \leq i \leq N$ by choosing $h_1(x)$.
This finishes the job. The ϕ_i we have constructed are O.N. and give the desired density and current.

Useful(?) Corollary

Given n functions on $\mathbb{R}^3$, $f_1(\vec{x}), \ldots, f_n(\vec{x})$, we can orthogonalize them with n smooth phase functions of x^1, $\theta_1(x^1), \ldots \theta_n(x^1)$. That is, there are smooth θ's so that the n functions

$$g_j(\vec{x}) = f_j(\vec{x}) e^{i\theta_j(x^1)}$$

are mutually orthogonal.

To prove this we use the smooth HR to find θ_2 so that $\int f_1^*(\vec{x}) f_2(\vec{x}) e^{i\theta_2(x^1)} d\vec{x} = 0$. That is, $g_1 = f_1$ and $g_2 = f_2 e^{1\theta_2(x^1)}$.

Next, we can find θ_3 so that $\int g_k^*(\vec{x}) f_3(\vec{x}) e^{i\theta_3(x^1)} d\vec{x} = 0$ for $k = 1, 2$.

Proceeding in this way we succesively find $\theta_4, \ldots, \theta_n$.

THANKS FOR LISTENING!