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PURE-STATE N-REPRESENTABILITY IN CURRENT-SPIN-DENSITY FUNCTIONAL THEORY*

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Abstract. This paper is concerned with the pure-state N-representability problem for systems under a magnetic field. Necessary and sufficient conditions are given for a spin-density 2×2 matrix R to be representable by a Slater determinant. We also provide sufficient conditions on the paramagnetic current $\mathbf{j}$ for the pair $(R,\mathbf{j})$ to be Slater-representable in the case where the number of electrons N is greater than 12. The case $N<12$ is left open.

Key words. Quantum theory, density functional theory, paramagnetic current, representability.

AMS subject classifications. 81Q05, 81V55.

1. Introduction

The density functional theory (DFT), first developed by Hohenberg and Kohn [5] then further developed and formalized mathematically by Levy [6], Valone [14], and Lieb [7], states that the ground state energy and density of a non-magnetic electronic system can be obtained by minimizing some functional of the density alone over the set of all admissible densities. Characterizing this set is called the *N-representability problem*. More precisely, as the so-called constrained search method leading to DFT can be performed either with N-electron wave functions [6, 7] or with N-body density matrices [7, 14], the N-representability problems can be recast in the pure-state setting resp. in the mixed-state setting as follows: *What is the set of electronic densities that come from an admissible N-electron wave function*, resp. *an admissible N-body density matrix?* This question was answered by Gilbert [1], Harriman [3], and Lieb [7] (see also Remark 3.3).

For a system subjected to a magnetic field, the energy of the ground state can be obtained by a minimization over the set of admissible pairs $(R,\mathbf{j})$, where R denotes the spin-density 2×2 matrix [2] (from which we recover the standard electronic density ρ and the spin angular momentum density $\mathbf{m}$) and $\mathbf{j}$ the paramagnetic current [16]. This has lead to several density-based theories that come from several different approximations. In spin-density functional theory (SDFT), one is only interested in spin effects. Hence the paramagnetic term is neglected. The SDFT energy functional of the system therefore only depends on the spin-density 2×2 matrix R. The N-representability problems in SDFT are therefore: *What is the set of spin-densities that come from an admissible N-electron wave function*, resp. *an admissible N-body density matrix?* (pure-state resp. mixed-state representability). These questions were left open in the pioneering work by von Barth and Hedin [17]. The mixed-state problem was answered recently in the mixed-case setting [2]. In parallel, in current-density functional theory (CDFT), one is only interested in magnetic orbital effects, and spin effects are neglected [15]. In this case, the CDFT energy functional of the system only depends on ρ and $\mathbf{j}$, and we need a characterization of the set of pure-state and mixed-state N-representable pairs $(\rho,\mathbf{j})$. Such a characterization was given recently by Hellgren, Kvaal, and Helgaker in the mixed-state setting [13] and by Lieb and Schrader in the pure-state setting when the number of electrons is greater than 4 [9]. In the latter article, the authors rely on

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the so-called Lazarev–Lieb orthogonalization process [8] (see also Lemma 4.2) in order to orthogonalize the Slater orbitals.

The purpose of this article is to give some answers to the N-representability problems in the current-spin-density functional theory (CSDFT): *What is the set of pairs* $(R,\mathbf{j})$ *that come from an admissible N-electron wave-function*, resp. *an admissible N-body density matrix?* (pure-state resp. mixed-state representability). We will answer the question in the mixed-state setting for all $N\in\mathbb{N}^*$ and in the pure-state setting when $N\geq 12$ by combining the results in [2] and in [9]. In the process, we will answer the N-representability problem for SDFT for all $N\in\mathbb{N}^*$ in the pure-state setting. The proof relies on the Lazarev–Lieb orthogonalization process. In particular, our method does not give an upper-bound for the kinetic energy of the wave-function in terms of the previous quantities (we refer to [8,11] for more details). We leave open the case $N<12$ for pure-state CSDFT representability.

The article is organized as follows. In Section 2, we recall briefly what the sets of interest are. We present our main results in Section 3, the proofs of which are given in Section 4.

2. The different Slater-state, pure-state and mixed-state sets

We recall in this section the definition of Slater-states, pure-states and mixed-states. We denote by $L^p(\mathbb{R}^3)$, $H^1(\mathbb{R}^3)$, $C^\infty(\mathbb{R}^3)$, ... the spaces of *real-valued* L^p, H^1, C^∞, ... functions on $\mathbb{R}^3$ and by $L^p(\mathbb{R}^3,\mathbb{C}^d)$, $H^1(\mathbb{R}^3,\mathbb{C}^d)$, $C^\infty(\mathbb{R}^3,\mathbb{C}^d)$, ... the spaces of $\mathbb{C}^d$-*valued* L^p, H^1, C^∞ functions on $\mathbb{R}^3$. We also make the identification $L^p(\mathbb{R}^3,\mathbb{C}^d)\equiv(L^p(\mathbb{R}^3,\mathbb{C}))^d$ (and similarly for $H^1(\mathbb{R}^3,\mathbb{C}^d)$, ...). The one-electron state space is

$$L^2(\mathbb{R}^3,\mathbb{C}^2)\equiv\left\{\Phi=(\phi^\uparrow,\phi^\downarrow)^T,\ \|\Phi\|_{L^2}^2:=\int_{\mathbb{R}^3}|\phi^\uparrow|^2+|\phi^\downarrow|^2<\infty\right\},$$

endowed with the natural scalar product $\langle\Phi_1|\Phi_2\rangle:=\int_{\mathbb{R}^3}\left(\overline{\phi_1^\uparrow}\phi_2^\uparrow+\overline{\phi_1^\downarrow}\phi_2^\downarrow\right)$. The Hilbert space for N-electrons is the fermionic space $\bigwedge_{i=1}^N L^2(\mathbb{R}^3,\mathbb{C}^2)$, which is the set of wave-functions $\Psi\in L^2((\mathbb{R}^3,\mathbb{C}^2)^N)$ satisfying the Pauli-principle: for all permutations p of $\{1,\dots,N\}$,

$$\Psi(\mathbf{r}_{p(1)},s_{p(1)},\dots,\mathbf{r}_{p(N)},s_{p(N)})=\varepsilon(p)\Psi(\mathbf{r}_1,s_1,\dots,\mathbf{r}_N,s_N),$$

where $\varepsilon(p)$ denotes the parity of the permutation p, $\mathbf{r}_k\in\mathbb{R}^3$ the position of the k-th electron, and $s_k\in\{\uparrow,\downarrow\}$ its spin. The set of admissible wave-functions, also called the set of pure-states, is the set of normalized wave-functions with finite kinetic energy

$$\mathcal{W}_N^{\text{pure}}:=\left\{\Psi\in\bigwedge_{i=1}^N L^2(\mathbb{R}^3,\mathbb{C}^2),\|\nabla\Psi\|_{L^2}^2<\infty,\|\Psi\|_{L^2(\mathbb{R}^{3N})}^2=1\right\},$$

where ∇ is the gradient with respect to the $3N$ position variables. A special case of wave-functions is given by Slater determinants: letting $\Phi_1,\Phi_2,\dots,\Phi_N$ be a set of orthonormal functions in $L^2(\mathbb{R}^3,\mathbb{C}^2)$, the Slater determinant generated by $(\Phi_1,\dots,\Phi_N)$ is (we denote by $\mathbf{x}_k:=(\mathbf{r}_k,s_k)$ the k-th spatial-spin component)

$$\mathscr{S}[\Phi_1,\dots,\Phi_N](\mathbf{x}_1,\dots,\mathbf{x}_N):=\frac{1}{\sqrt{N!}}\det(\Phi_i(\mathbf{x}_j))_{1\leq i,j\leq N}.$$

The subset of $\mathcal{W}_N^{\text{pure}}$ consisting of all finite energy Slater determinants is noted $\mathcal{W}_N^{\text{Slater}}$. It holds that $\mathcal{W}_1^{\text{Slater}}=\mathcal{W}_1^{\text{pure}}$ and $\mathcal{W}_N^{\text{Slater}}\subsetneq\mathcal{W}_N^{\text{pure}}$ for $N\geq 2$.

For a wave-function $\Psi \in \mathcal{W}_N^{\rm pure}$, we define the corresponding N-body density matrix $\Gamma_\Psi := |\Psi\rangle\langle\Psi|$, which corresponds to the projection on $\{\mathbb{C}\Psi\}$ in $\bigwedge_{i=1}^N L^2(\mathbb{R}^3,\mathbb{C}^2)$. The set of pure-state (resp. Slater-state) N-body density matrices is

$$G_N^{\rm pure} := \{\Gamma_\Psi, \Psi \in \mathcal{W}_N^{\rm pure}\} \text{ resp. } G_N^{\rm Slater} := \left\{\Gamma_\Psi, \Psi \in \mathcal{W}_N^{\rm Slater}\right\}.$$

It holds that $G_1^{\rm Slater} = G_1^{\rm pure}$ and that $G_N^{\rm Slater} \subsetneq G_N^{\rm pure}$ for $N \geq 2$. The set of mixed-state N-body density matrices $G_N^{\rm mixed}$ is defined as the convex hull of $G_N^{\rm pure}$:

$$G_N^{\rm mixed} = \left\{ \sum_{k=1}^{\infty} n_k |\Psi_k\rangle\langle\Psi_k|, 0 \leq n_k \leq 1, \sum_{k=1}^{\infty} n_k = 1, \Psi_k \in \mathcal{W}_N^{\rm pure} \right\}.$$

It is also the convex hull of $G_N^{\rm Slater}$. The kernel of an operator $\Gamma \in G_N^{\rm mixed}$ will be denoted by

$$\Gamma(\mathbf{r}_1, s_1, \ldots, \mathbf{r}_N, s_N; \mathbf{r}_1', s_1', \ldots, \mathbf{r}_N', s_N').$$

The quantities of interest in DFT are the spin-density 2×2 matrix and the paramagnetic-current. For $\Gamma \in G_N^{\rm mixed}$, the associated spin-density 2×2 matrix is the 2×2 hermitian function-valued matrix

$$R_\Gamma(\mathbf{r}) := \begin{pmatrix} \rho_\Gamma^{\uparrow\uparrow} & \rho_\Gamma^{\uparrow\downarrow} \\ \rho_\Gamma^{\downarrow\uparrow} & \rho_\Gamma^{\downarrow\downarrow} \end{pmatrix}(\mathbf{r}),$$

where, for $\alpha, \beta \in \{\uparrow,\downarrow\}^2$,

$$\rho_\Gamma^{\alpha\beta}(\mathbf{r}) := N \sum_{\vec{s} \in \{\uparrow,\downarrow\}^{(N-1)}} \int_{\mathbb{R}^{3(N-1)}} \Gamma(\mathbf{r}, \alpha, \vec{\mathbf{z}}, \vec{s}; \mathbf{r}, \beta, \vec{\mathbf{z}}, \vec{s}) \, {\rm d}\vec{\mathbf{z}}.$$

In the case where Γ comes from a Slater determinant $\mathscr{S}[\Phi_1, \ldots, \Phi_N]$, we get

$$R_\Gamma(\mathbf{r}) = \sum_{k=1}^N \begin{pmatrix} |\phi_k^\uparrow|^2 & \phi_k^\uparrow \overline{\phi_k^\downarrow} \\ \overline{\phi_k^\uparrow} \phi_k^\downarrow & |\phi_k^\downarrow|^2 \end{pmatrix}. \tag{2.1}$$

In order to lighten the notation, we will denote by $\rho_\Gamma^\uparrow := \rho_\Gamma^{\uparrow\uparrow}$, $\rho_\Gamma^\downarrow := \rho_\Gamma^{\downarrow\downarrow}$, and $\sigma_\Gamma := \rho_\Gamma^{\uparrow\downarrow}$ the elements of such a matrix R so that

$$R_\Gamma = \begin{pmatrix} \rho_\Gamma^\uparrow & \sigma_\Gamma \\ \overline{\sigma_\Gamma} & \rho_\Gamma^\downarrow \end{pmatrix}.$$

The total electronic density is $\rho_\Gamma = \rho_\Gamma^\uparrow + \rho_\Gamma^\downarrow$, and the spin angular momentum density is $\mathbf{m}_\Gamma = {\rm tr}_{\mathbb{C}^2}[\sigma R_\Gamma]$, where

$$\sigma := (\sigma_x, \sigma_y, \sigma_z) := \left(\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \begin{pmatrix} 0 & -{\rm i} \\ {\rm i} & 0 \end{pmatrix}, \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \right)$$

contains the Pauli matrices. As ρ_Γ only depends on Γ through R_Γ, we will sometimes use the notation $\rho_{R_\Gamma} := \rho_\Gamma$ or simply ρ_R when no confusion is possible. Note that the pair $(\rho_\Gamma, \mathbf{m}_\Gamma)$ contains the same information as R_Γ. Hence, the N-representability problem for the matrix R is the same as the one for the pair $(\rho, \mathbf{m})$. However, as noticed in [2], it is more natural mathematicaly speaking to work with R_Γ. The Slater-state, pure-state and mixed-state sets of spin-density 2×2 matrices are respectively defined by

$$\mathcal{J}_N^{\text{Slater}} := \{R_\Gamma,\ \Gamma\in G_N^{\text{Slater}}\},$$
$$\mathcal{J}_N^{\text{pure}} := \{R_\Gamma,\ \Gamma\in G_N^{\text{pure}}\},$$
$$\mathcal{J}_N^{\text{mixed}} := \{R_\Gamma,\ \Gamma\in G_N^{\text{mixed}}\}.$$

Since the map $\Gamma\mapsto R_\Gamma$ is linear, it holds that $\mathcal{J}_N^{\text{Slater}}\subset\mathcal{J}_N^{\text{pure}}\subset\mathcal{J}_N^{\text{mixed}}$ and that $\mathcal{J}_N^{\text{mixed}}$ is convex and is the convex hull of both $\mathcal{J}_N^{\text{Slater}}$ and $\mathcal{J}_N^{\text{pure}}$.

For an N-body density matrix $\Gamma\in G_N^{\text{mixed}}$, we define the associated paramagnetic current $\mathbf{j}_\Gamma=\mathbf{j}_\Gamma^\uparrow+\mathbf{j}_\Gamma^\downarrow$ where

$$\mathbf{j}_\Gamma^\alpha=\operatorname{Im}\left(N\sum_{\vec{s}\in\{\uparrow,\downarrow\}^{N-1}}\int_{\mathbb{R}^{3(N-1)}}\nabla_{\mathbf{r}'}\Gamma(\mathbf{r},\alpha,\vec{\mathbf{z}},\vec{s};\mathbf{r}',\alpha,\vec{\mathbf{z}},\vec{s})\Big|_{\mathbf{r}'=\mathbf{r}}\mathrm{d}\vec{\mathbf{z}}\right).$$

In the case where Γ comes from a Slater determinant $\mathcal{S}[\Phi_1,\dots,\Phi_N]$, we get

$$\mathbf{j}_\Gamma=\sum_{k=1}^N\operatorname{Im}\left(\overline{\phi_k^\uparrow}\nabla\phi_k^\uparrow+\overline{\phi_k^\downarrow}\nabla\phi_k^\downarrow\right). \tag{2.2}$$

Note that, while only the total paramagnetic current $\mathbf{j}$ appears in C(S)DFT, the pair $(\mathbf{j}^\uparrow,\mathbf{j}^\downarrow)$ is sometimes used to design accurate current-density functionals (see [16] for instance). In this article, however, we will only focus on the representability of $\mathbf{j}$.

3. Main results

3.1. Representability in SDFT. Our first result concerns the characterization of $\mathcal{J}_N^{\text{Slater}}$, $\mathcal{J}_N^{\text{pure}}$, and $\mathcal{J}_N^{\text{mixed}}$. For this purpose, we introduce

$$\begin{aligned}\mathcal{C}_N:=\Big\{&R\in\mathcal{M}_{2\times2}(L^1(\mathbb{R}^3,\mathbb{C})),\ R^*=R,\quad R\geq 0,\\ &\int_{\mathbb{R}^3}\operatorname{tr}_{\mathbb{C}^2}[R]=N,\quad \sqrt{R}\in\mathcal{M}_{2\times2}(H^1(\mathbb{R}^3,\mathbb{C}))\Big\}\end{aligned} \tag{3.1}$$

and $\mathcal{C}_N^0:=\{R\in\mathcal{C}_N,\ \det R\equiv 0\}$, where $\mathcal{M}_{2\times2}(E)$ denotes the set of 2×2 matrices with elements in the Banach space E.

The following characterization of $\mathcal{C}_N$ was proved in [2].

LEMMA 3.1. *A function-valued matrix* $R=\begin{pmatrix}\rho^\uparrow & \sigma\\ \overline{\sigma} & \rho^\downarrow\end{pmatrix}$ *is in* $\mathcal{C}_N$ *if and only if its coefficients satisfy*

$$\begin{cases}\rho^{\uparrow/\downarrow}\geq 0,\quad \rho^\uparrow\rho^\downarrow-|\sigma|^2\geq 0,\quad \displaystyle\int_{\mathbb{R}^3}\rho^\uparrow+\int_{\mathbb{R}^3}\rho^\downarrow=N,\\ \sqrt{\rho^{\uparrow/\downarrow}}\in H^1(\mathbb{R}^3),\quad \sigma,\sqrt{\det(R)}\in W^{1,3/2}(\mathbb{R}^3),\\ |\nabla\sigma|^2\rho^{-1}\in L^1(\mathbb{R}^3),\\ \left|\nabla\sqrt{\det(R)}\right|^2\rho^{-1}\in L^1(\mathbb{R}^3).\end{cases}$$

The complete answer for N-representability in SDFT is given by the following theorem, whose proof is given in Section 4.1.

THEOREM 3.2.

Case $N=1$***:*** *It holds that*

$$\mathcal{J}_1^{\mathrm{Slater}} = \mathcal{J}_1^{\mathrm{pure}} = \mathcal{C}_1^0 \quad \textit{and} \quad \mathcal{J}_1^{\mathrm{mixed}} = \mathcal{C}_1.$$

Case $N \geq 2$***:*** *For all* $N \geq 2$*, it holds that*

$$\mathcal{J}_N^{\mathrm{Slater}} = \mathcal{J}_N^{\mathrm{pure}} = \mathcal{J}_N^{\mathrm{mixed}} = \mathcal{C}_N.$$

The equality $\mathcal{J}_N^{\mathrm{mixed}} = \mathcal{C}_N$ for all $N \in \mathbb{N}^*$ was already proven in [2].

REMARK 3.3. Gilbert [1], Harriman [3], and Lieb [7] proved that the N-representability set for the total electronic density ρ is the same for Slater-states, pure-states, and mixed-states and is characterized by

$$\mathcal{I}_N := \left\{ \rho \in L^1(\mathbb{R}^3),\ \rho \geq 0,\ \int_{\mathbb{R}^3} \rho = N,\ \sqrt{\rho} \in H^1(\mathbb{R}^3) \right\}. \tag{3.2}$$

Comparing (3.2) and (3.1), we see that our theorem is a natural extension of the previous result.

3.2. Representability in CSDFT. We first recall some classical necessary conditions for a pair $(R,\mathbf{j})$ to be N-representable (we refer to [9, 13] for the proofs).

LEMMA 3.4. *If a pair* $(R,\mathbf{j})$ *is representable by a mixed-state* N*-body density matrix, then*

$$\begin{cases} R \in \mathcal{C}_N \\ |\mathbf{j}|^2/\rho \in L^1(\mathbb{R}^3). \end{cases} \tag{3.3}$$

From the second condition of (3.3), it must hold that the support of $\mathbf{j}$ is contained in the support of ρ. The vector $\mathbf{v} := \rho^{-1}\mathbf{j}$ is called the velocity field, and $\mathbf{w} := \mathbf{curl}(\mathbf{v})$ is the vorticity.

Let us first consider the pure-state setting. Recall that, in the spin-less setting, in the case $N=1$, a pair $(\rho,\mathbf{j})$ representable by a single orbital ϕ generally satisfies the curl-free condition $\mathbf{curl}(\rho^{-1}\mathbf{j}) = \mathbf{0}$ (this is the case for instance when ϕ is of the form $\phi = |\phi| \mathrm{e}^{-\mathrm{i}u}$, where the phase u is in $C^1(\mathbb{R}^3)$; see [9,13]). This is no longer the case when spin is considered, as is shown in the following lemma (see Section 4.2 for the proof).

LEMMA 3.5 (CSDFT, case $N=1$). *Let* $\Phi = (\phi^\uparrow, \phi^\downarrow)^T \in \mathcal{W}_1^{\mathrm{Slater}}$ *be such that both* $\phi^\uparrow$ *and* $\phi^\downarrow$ *have phases in* $C^1(\mathbb{R}^3)$*. Then the associated pair* $(R,\mathbf{j})$ *satisfies* $R \in \mathcal{C}_1^0$*,* $|\mathbf{j}|^2/\rho \in L^1(\mathbb{R}^3)$*, and the two curl-free conditions*

$$\mathbf{curl}\left(\frac{\mathbf{j}}{\rho} - \frac{\mathrm{Im}\,(\overline{\sigma}\nabla\sigma)}{\rho\rho^\downarrow} \right) = \mathbf{0},\ \mathbf{curl}\left(\frac{\mathbf{j}}{\rho} + \frac{\mathrm{Im}\,(\overline{\sigma}\nabla\sigma)}{\rho\rho^\uparrow} \right) = \mathbf{0}. \tag{3.4}$$

REMARK 3.6. If we write $\sigma = |\sigma| \mathrm{e}^{\mathrm{i}\tau}$, then $|\sigma|^2 = \rho^\uparrow \rho^\downarrow$ and

$$\mathrm{Im}\,(\overline{\sigma}\nabla\sigma) = |\sigma|^2 \nabla\tau = \rho^\uparrow\rho^\downarrow\nabla\tau. \tag{3.5}$$

In particular, it holds that

$$\boldsymbol{curl}\left(\frac{\mathrm{Im}(\overline{\sigma}\nabla\sigma)}{\rho\rho^{\downarrow}}+\frac{\mathrm{Im}(\overline{\sigma}\nabla\sigma)}{\rho\rho^{\uparrow}}\right)=\boldsymbol{curl}\,(\nabla\tau)=\mathbf{0},$$

so that one of the equalities in (3.4) implies the other one.

REMARK 3.7. We recover the traditional result in the collinear-spin setting, where $\sigma\equiv 0$.

In the case $N>1$, things are very different. In [9], the authors gave a rigorous proof for the representability of a pair $(\rho,\mathbf{j})$ by a Slater determinant whenever $N\geq 4$ under a mild condition (see equation (3.6) below). By adapting their proof to our case, we are able to ensure representability of a pair $(R,\mathbf{j})$ by a Slater determinant for $N\geq 12$ under the same mild condition (see Section 4.3 for the proof).

THEOREM 3.8 (CSDFT, case $N\geq 12$). *A sufficient set of conditions for a pair $(R,\mathbf{j})$ to be representable by a Slater determinant is:*

- $R\in\mathcal{C}_N$ *with* $N\geq 12$ *and* $\mathbf{j}$ *satisfies* $|\mathbf{j}|^2/\rho\in L^1(\mathbb{R}^3)$.
- *There exists* $\delta>0$ *such that*

$$\sup_{\mathbf{r}\in\mathbb{R}^3} f(\mathbf{r})^{(1+\delta)/2}\big(|\mathbf{w}(\mathbf{r})|+|\nabla\mathbf{w}(\mathbf{r})|\big)<\infty, \tag{3.6}$$

where $\mathbf{w}:=\mathbf{curl}\,(\rho^{-1}\mathbf{j})$ *is the vorticity, and*

$$f(\mathbf{r}):=(1+(r_1)^2)(1+(r_2)^2)(1+(r_3)^2).$$

REMARK 3.9. The condition (3.6) is the one found in [9]. The authors conjectured that this condition “can be considerably loosened.”

Let us finally turn to the mixed-state case. We notice that, if $(R,\mathbf{j})$ is representable by a Slater determinant $\mathscr{S}[\Phi_1,\ldots,\Phi_N]$, then, for all $k\in\mathbb{N}^*$, the pair $(k/N)(R,\mathbf{j})$ is mixed-state representable, where N is the number of orbitals (simply take the uniform convex combination of the pairs represented by $\mathscr{S}[\Phi_1]$, $\mathscr{S}[\Phi_2]$, etc.). In particular, from Theorem 3.8, we deduce the following corollary.

COROLLARY 3.10 (CSDFT, case mixed-state). *A sufficient set of conditions for a pair $(R,\mathbf{j})$ to be mixed-state representable is that $R\in\mathcal{C}_N$ for some $N\in\mathbb{N}^*$, $\mathbf{j}$ satisfies $|\mathbf{j}|^2/\rho\in L^1(\mathbb{R}^3)$, and (3.6) holds for some $\delta>0$.*

In [13], the authors provide different sufficient conditions than (3.6) for a pair $(\rho,\mathbf{j})$ to be mixed-state representable, where ρ is the electronic density. They proved that, if

$$(1+|\cdot|^2)\rho\left|\nabla(\rho^{-1}\mathbf{j})\right|^2\in L^1(\mathbb{R}^3),$$

then the pair $(\rho,\mathbf{j})$ is mixed-state representable. Their proof can be straightforwardly adapted for the representability of the pair $(R,\mathbf{j})$, so that similar results hold. The details are omitted here for the sake of brevity.

4. Proofs

4.1. Proof of Theorem 3.2. The mixed-state case was already proved in [2]. We focus on the pure-state representability.

Case $N=1$**.** The fact that $\mathcal{J}_1^{\rm Slater}=\mathcal{J}_1^{\rm pure}$ simply comes from the fact that $G_1^{\rm Slater}=G_1^{\rm pure}$. To prove $\mathcal{J}_1^{\rm Slater}\subset\mathcal{C}_1^0$, we let $R\in\mathcal{J}_1^{\rm Slater}$ be represented by $\Phi=(\phi^\uparrow,\phi^\downarrow)^T\in H^1(\mathbb{R}^3,\mathbb{C}^2)$, so that

$$R=\begin{pmatrix}|\phi^\uparrow|^2 & \phi^\uparrow\overline{\phi^\downarrow}\\ \phi^\downarrow\overline{\phi^\uparrow} & |\phi^\downarrow|^2\end{pmatrix}.$$

Since $R\in\mathcal{J}_1^{\rm Slater}\subset\mathcal{J}_1^{\rm mixed}=\mathcal{C}_1$ and $\det R\equiv 0$, we deduce $R\in\mathcal{C}_1^0$.

We now prove that $\mathcal{C}_1^0\subset\mathcal{J}_1^{\rm Slater}$. Let $R=\begin{pmatrix}\rho^\uparrow & \sigma\\ \overline{\sigma} & \rho^\downarrow\end{pmatrix}\in\mathcal{C}_1^0$. From $\det R\equiv 0$ and Lemma 3.1, we get

$$\begin{cases}\rho^{\uparrow/\downarrow}\geq 0,\quad \rho^\uparrow\rho^\downarrow=|\sigma|^2,\quad \displaystyle\int_{\mathbb{R}^3}\rho^\uparrow+\int_{\mathbb{R}^3}\rho^\downarrow=1,\\ \sqrt{\rho^{\uparrow/\downarrow}}\in H^1(\mathbb{R}^3),\quad \sigma\in W^{1,3/2}(\mathbb{R}^3),\\ |\nabla\sigma|^2/\rho\in L^1(\mathbb{R}^3).\end{cases}\tag{4.1}$$

There are two natural choices that we would like to make for a representing orbital, namely

$$\Phi_1=\left(\sqrt{\rho^\uparrow},\,\frac{\overline{\sigma}}{\sqrt{\rho^\uparrow}}\right)^T\quad\text{and}\quad\Phi_2=\left(\frac{\sigma}{\sqrt{\rho^\downarrow}},\,\sqrt{\rho^\downarrow}\right)^T.\tag{4.2}$$

Unfortunately, it is not guaranteed that these orbitals are indeed in $H^1(\mathbb{R}^3,\mathbb{C}^2)$. It is the case only if $|\nabla\sigma|^2/\rho^\downarrow$ is in $L^1(\mathbb{R}^3)$ for Φ_1 and if $|\nabla\sigma|^2/\rho^\uparrow$ is in $L^1(\mathbb{R}^3)$ for Φ_2. Due to (4.1), we only know that $|\nabla\sigma|^2/\rho\in L^1(\mathbb{R}^3)$. The idea is therefore to interpolate between these two orbitals, taking Φ_1 in regions where $\rho^\uparrow\gg\rho^\downarrow$ and Φ_2 in regions where $\rho^\downarrow\gg\rho^\uparrow$. This is done via the following process.

Let $\chi\in C^\infty(\mathbb{R})$ be a non-decreasing function such that $0\leq\chi\leq 1$, $\chi(x)=0$ if $x\leq 1/2$, and $\chi(x)=1$ if $x\geq 1$. We write $\sigma=\alpha+\mathrm{i}\beta$, where α is the real-part of σ and β is its imaginary part. We introduce

$$\begin{aligned}\lambda_1&:=\frac{\sqrt{\alpha^2+\chi^2(\rho^\uparrow/\rho^\downarrow)\beta^2}}{\sqrt{\rho^\downarrow}}, & \mu_1&:=\sqrt{1-\chi^2(\rho^\uparrow/\rho^\downarrow)}\frac{\beta}{\sqrt{\rho^\downarrow}},\\ \lambda_2&:=\frac{\alpha\lambda_1+\beta\mu_1}{\rho^\uparrow}, & \mu_2&:=\frac{\beta\lambda_1-\alpha\mu_1}{\rho^\uparrow},\end{aligned}$$

and we set

$$\phi^\uparrow:=\lambda_1+\mathrm{i}\mu_1\quad\text{and}\quad\phi^\downarrow:=\lambda_2+\mathrm{i}\mu_2.$$

Let us prove that $\Phi:=(\phi^\uparrow,\phi^\downarrow)$ represents R and that $\Phi\in\mathcal{W}_1^{\rm Slater}$. First, an easy calculation shows that

$$\begin{aligned}|\phi^\uparrow|^2&=\lambda_1^2+\mu_1^2=\frac{\alpha^2+\chi^2\beta^2+(1-\chi^2)\beta^2}{\rho^\downarrow}=\frac{|\sigma|^2}{\rho^\downarrow}=\rho^\uparrow,\\ |\phi^\downarrow|^2&=\frac{(\alpha^2+\beta^2)(\lambda_1^2+\mu_1^2)}{(\rho^\uparrow)^2}=\frac{|\sigma|^2}{\rho^\uparrow}=\rho^\downarrow,\end{aligned}$$

$$\operatorname{Re}\left(\phi^\uparrow\overline{\phi^\downarrow}\right)=\lambda_1\lambda_2-\mu_1\mu_2=\frac{\alpha(\lambda_1^2+\mu_1^2)}{\rho^\uparrow}=\alpha,$$
$$\operatorname{Im}\left(\phi^\uparrow\overline{\phi^\downarrow}\right)=\lambda_1\mu_2+\lambda_2\mu_1=\frac{\beta(\lambda_1^2+\mu_1^2)}{\sqrt{\rho^\uparrow}}=\beta,$$

so that $\Phi\in L^2(\mathbb{R}^3,\mathbb{C}^2)$ with $\|\Phi\|=1$, and Φ represents R. To prove that $\Phi\in\mathcal{W}_1^{\text{Slater}}$, we need to check that $\lambda_1,\lambda_2,\mu_1$, and μ_2 are in $H^1(\mathbb{R}^3)$. For λ_1, we choose another non-increasing function $\xi\in C^\infty(\mathbb{R})$ such that $0\le\xi\le 1$, $\xi(x)=0$ for $x\le 1$, and $\xi(x)=1$ for $x\ge 2$. Note that $(1-\chi)\xi\equiv 0$. It holds that

$$\nabla\lambda_1=(1-\xi^2(\rho^\uparrow/\rho^\downarrow))\nabla\lambda_1+\xi^2(\rho^\uparrow/\rho^\downarrow)\nabla\lambda_1. \tag{4.3}$$

The second term in the right-hand side of (4.3) is non-null only if $\rho^\uparrow\ge\rho^\downarrow$, so that $\chi(\rho^\uparrow/\rho^\downarrow)=1$ on this part. In particular, from the equality $\rho^\uparrow\rho^\downarrow=|\sigma|^2$, we get

$$\xi^2(\rho^\uparrow/\rho^\downarrow)\lambda_1=\xi^2(\rho^\uparrow/\rho^\downarrow)\frac{|\sigma|}{\sqrt{\rho^\downarrow}}=\xi^2(\rho^\uparrow/\rho^\downarrow)\sqrt{\rho^\uparrow},$$

and similarly,

$$\xi^2(\rho^\uparrow/\rho^\downarrow)\nabla\lambda_1=\xi^2(\rho^\uparrow/\rho^\downarrow)\nabla\sqrt{\rho^\uparrow},$$

which is in $L^2(\mathbb{R}^3)$ according to (4.1). On the other hand, the first term in the right-hand side of (4.3) is non-null only if $\rho^\uparrow\le 2\rho^\downarrow$, so that $(1/3)\rho\le\rho^\downarrow$ on this part. In particular, from the following pointwise estimate

$$|\nabla\sqrt{f+g}|\le|\nabla\sqrt{f}|+|\nabla\sqrt{g}|,$$

which is valid almost everywhere whenever $f,g\ge 0$, the inequality $(a+b)^2\le 2(a^2+b^2)$, and the fact that $\alpha^2+\chi^2\beta^2\le|\sigma|^2$, we get (we write χ for $\chi(\rho^\uparrow/\rho^\downarrow)$)

$$\begin{aligned}|\nabla\lambda_1|^2&=\left|\frac{\sqrt{\rho^\downarrow}\nabla\sqrt{\alpha^2+\chi^2\beta^2}-\sqrt{\alpha^2+\chi^2\beta^2}\nabla\sqrt{\rho^\downarrow}}{\rho^\downarrow}\right|^2\\&\le 2\left(\frac{|\nabla\sqrt{\alpha^2+\chi^2\beta^2}|^2}{\rho^\downarrow}+\frac{(\alpha^2+\chi^2\beta^2)}{(\rho^\downarrow)^2}|\nabla\sqrt{\rho^\downarrow}|^2\right)\\&\le 2\left(\frac{|\nabla\alpha|^2}{\rho^\downarrow}+\frac{2\left|\nabla\chi\frac{\rho^\downarrow\nabla\rho^\uparrow-\rho^\uparrow\nabla\rho^\downarrow}{(\rho^\downarrow)^2}\right|^2\beta^2}{\rho^\downarrow}+\frac{2\chi^2|\nabla\beta|^2}{\rho^\downarrow}+\frac{2|\sigma|^2}{(\rho^\downarrow)^2}|\nabla\sqrt{\rho^\downarrow}|^2\right).\end{aligned}$$

Finally, we use the inequality $(\rho^\downarrow)^{-1}\le(3/\rho)$, and the inequality $|\sigma|^2/(\rho^\downarrow)^2=\rho^\uparrow/\rho^\downarrow\le 2$ and get

$$|\nabla\lambda_1|^2\le C\left(\frac{|\nabla\alpha|^2}{\rho}+\|\nabla\chi\|_{L^\infty}^2\left(\frac{|\nabla\rho^\uparrow|^2}{\rho^\uparrow}+\frac{|\nabla\rho^\downarrow|^2}{\rho^\downarrow}\right)+\frac{|\nabla\beta|^2}{\rho}+|\nabla\sqrt{\rho^\downarrow}|^2\right).$$

The right-hand side is in $L^1(\mathbb{R}^3)$ according to (4.1). Hence, $(1-\xi^2(\rho^\uparrow/\rho^\downarrow))|\nabla\lambda_1|\in L^2(\mathbb{R}^3)$, and finally $\lambda_1\in H^1(\mathbb{R}^3)$. The other cases are treated similarly, observing the following:

- Whenever $\rho^{\uparrow} \geq \rho^{\downarrow}$, then $\chi = 1$, and $\Phi = \Phi_1$, where Φ_1 was defined in (4.2). We then control $(\rho^{\uparrow})^{-1}$ with the inequality $(\rho^{\uparrow})^{-1} \leq 2\rho^{-1}$.
- Whenever $\rho^{\uparrow} \leq \rho^{\downarrow}/2$, then $\chi = 0$, $\Phi = \Phi_2$. We control $(\rho^{\downarrow})^{-1}$ with the inequality $(\rho^{\downarrow})^{-1} \leq \frac{3}{2}\rho^{-1}$.
- Whenever $\rho^{\downarrow}/2 \leq \rho^{\uparrow} \leq \rho^{\downarrow}$, then both $(\rho^{\uparrow})^{-1}$ and $(\rho^{\downarrow})^{-1}$ are controlled via $(\rho^{\uparrow})^{-1} \leq 3\rho^{-1}$ and $(\rho^{\downarrow})^{-1} \leq 2\rho^{-1}$.

Case $N \geq 2$**.** Since $\mathcal{J}_N^{\mathrm{Slater}} \subset \mathcal{J}_N^{\mathrm{pure}} \subset \mathcal{J}_N^{\mathrm{mixed}} = \mathcal{C}_N$, it is enough to prove that $\mathcal{C}_N \subset \mathcal{J}_N^{\mathrm{Slater}}$. We start with a key lemma.

Lemma 4.1. *For all* $M, N \in \mathbb{N}$*, it holds that* $\mathcal{J}_{N+M}^{\mathrm{Slater}} = \mathcal{J}_N^{\mathrm{Slater}} + \mathcal{J}_M^{\mathrm{Slater}}$.

Proof. (Proof of Lemma 4.1.) The case $\mathcal{J}_{N+M}^{\mathrm{Slater}} \subset \mathcal{J}_N^{\mathrm{Slater}} + \mathcal{J}_M^{\mathrm{Slater}}$ is trivial: if $R \in \mathcal{J}_{N+M}^{\mathrm{Slater}}$ is represented by the Slater determinant $\mathscr{S}[\Phi_1, \ldots \Phi_{N+M}]$, then, by denoting by R_1 (resp. R_2) the spin-density 2×2 matrix associated to the Slater determinant $\mathscr{S}[\Phi_1, \ldots, \Phi_N]$ (resp. $\mathscr{S}[\Phi_{N+1}, \ldots, \Phi_{N+M}]$), it holds that $R = R_1 + R_2$ (see Equation (2.1) for instance), with $R_1 \in \mathcal{J}_N^{\mathrm{Slater}}$ and $R_2 \in \mathcal{J}_M^{\mathrm{Slater}}$.

The converse is more involving, and it requires an orthogonalization step. Let $R_1 \in \mathcal{J}_N^{\mathrm{Slater}}$ be represented by the Slater determinant $\mathscr{S}[\Phi_1, \ldots, \Phi_N]$ and $R_2 \in \mathcal{J}_M^{\mathrm{Slater}}$ be represented by the Slater determinant $\mathscr{S}[\widetilde{\Phi}_1, \ldots, \widetilde{\Phi}_M]$. We cannot directly consider the Slater determinant $\mathscr{S}[\Phi_1, \ldots, \Phi_N, \widetilde{\Phi}_1, \ldots, \widetilde{\Phi}_M]$, for $(\Phi_1, \ldots, \Phi_N)$ is not orthogonal to $(\widetilde{\Phi}_1, \ldots, \widetilde{\Phi}_M)$.

We use the following lemma, which is a smooth version of the Hobby–Rice theorem in [4] (see also [10]) and was proven by Lazarev and Lieb in [8] (see also [9]).

Lemma 4.2. *For all* $N \in \mathbb{N}^*$ *and for all* $(f_1, \ldots, f_N) \in L^1(\mathbb{R}^3, \mathbb{C})$*, there exists a function* $u \in C^\infty(\mathbb{R}^3)$*, with bounded derivatives, such that*

$$\forall 1 \leq k \leq N, \quad \int_{\mathbb{R}^3} f_k \mathrm{e}^{\mathrm{i}u} = 0.$$

Moreover, u *can be chosen to vary in the* r_1 *direction only.*

We now modify the phases of $\widetilde{\Phi_1}, \ldots, \widetilde{\Phi_M}$ as follows. First, we choose $\widetilde{u_1}$ as in Lemma 4.2 such that

$$\forall 1 \leq k \leq N, \quad \int_{\mathbb{R}^3} \left(\overline{\phi_k^{\uparrow}} \widetilde{\phi_1^{\uparrow}} + \overline{\phi_k^{\downarrow}} \widetilde{\phi_1^{\downarrow}} \right) \mathrm{e}^{\mathrm{i}\widetilde{u_1}} = 0,$$

and we set $\Phi_{N+1} = \widetilde{\Phi_1} \mathrm{e}^{\mathrm{i}\widetilde{u_1}}$. Note that, by construction, Φ_{N+1} is normalized, in $H^1(\mathbb{R}^3, \mathbb{C}^2)$, and orthogonal to $(\Phi_1, \ldots, \Phi_N)$. We then construct $\widetilde{u_2}$ as in Lemma 4.2 such that

$$\forall 1 \leq k \leq N+1, \quad \int_{\mathbb{R}^3} \left(\overline{\phi_k^{\uparrow}} \widetilde{\phi_2^{\uparrow}} + \overline{\phi_k^{\downarrow}} \widetilde{\phi_2^{\downarrow}} \right) \mathrm{e}^{\mathrm{i}\widetilde{u_2}} = 0,$$

and we set $\Phi_{N+2} = \widetilde{\Phi_2} \mathrm{e}^{\mathrm{i}\widetilde{u_2}}$. We continue this process for $3 \leq k \leq M$ and construct $\Phi_{N+k} = \widetilde{\Phi_k} \mathrm{e}^{\mathrm{i}\widetilde{u_k}}$. We thus obtain an orthonormal family $(\Phi_1, \ldots, \Phi_{N+M})$. By noticing that the spin-density 2×2 matrix of the Slater determinant $\mathscr{S}[\widetilde{\Phi_1}, \ldots, \widetilde{\Phi_M}]$ is the same as the one of $\mathscr{S}[\Phi_{N+1}, \ldots, \Phi_{N+M}]$ (the phases cancel out), we obtain that $R = R_1 + R_2$, where R is the spin-density 2×2 matrix represented by $\mathscr{S}[\Phi_1, \ldots, \Phi_{N+M}]$. The result follows. ∎

We now prove that $\mathcal{C}_N \subset \mathcal{J}_N^{\mathrm{Slater}}$. We start with the case $N=2$.

Case $N=2$. Let $R=\begin{pmatrix}\rho^\uparrow & \sigma\\ \overline{\sigma} & \rho^\downarrow\end{pmatrix}\in\mathcal{C}_2$. We write $\sqrt{R}=\begin{pmatrix}r^\uparrow & s\\ \overline{s} & r^\downarrow\end{pmatrix}$, with $r^\uparrow,r^\downarrow\in\left(H^1(\mathbb{R}^3)\right)^2$ and s in $H^1(\mathbb{R}^3,\mathbb{C})$. Let

$$R^\uparrow := \begin{pmatrix}|r^\uparrow|^2 & sr^\uparrow\\ \overline{s}r^\uparrow & |s|^2\end{pmatrix} \quad\text{and}\quad R^\downarrow := \begin{pmatrix}|s|^2 & sr^\downarrow\\ \overline{s}r^\downarrow & |r^\downarrow|^2\end{pmatrix}. \tag{4.4}$$

It is easy to check that $R=R^\uparrow+R^\downarrow$, that $R^{\uparrow/\downarrow}$ are hermitian, of null determinant, and $\sqrt{R^{\uparrow/\downarrow}}\in\mathcal{M}_{2\times2}\left(H^1(\mathbb{R}^3,\mathbb{C})\right)$. However, it may hold that $\int_{\mathbb{R}^3}\mathrm{tr}_{\mathbb{C}^2}[R^\uparrow]\notin\mathbb{N}^*$, so that $R^\uparrow$ is not in $\mathcal{C}_M^0$ for some $M\in\mathbb{N}^*$.

The cases $R^\uparrow=0$ and $R^\downarrow=0$ are trivial. Let us suppose that, for $\alpha\in\{\uparrow,\downarrow\}$, $m^\alpha:=\int_{\mathbb{R}^3}\rho_{R^\alpha}\neq0$. In this case, the matrices $\widetilde{R^\alpha}=(m^\alpha)^{-1}R^\alpha$ are in $\mathcal{C}_1^0$ and hence are representable by a single orbital, due to the first statement of Theorem 3.2. Let $\widetilde{\Phi}=(\widetilde{\phi_1^\uparrow},\widetilde{\phi_1^\downarrow})^T\in H^1(\mathbb{R}^3,\mathbb{C}^2)$ and $\widetilde{\Phi}_2=(\widetilde{\phi_2^\uparrow},\widetilde{\phi_2^\downarrow})^T\in H^1(\mathbb{R}^3,\mathbb{C}^2)$ be normalized orbitals that represent respectively $\widetilde{R^\uparrow}$ and $\widetilde{R^\downarrow}$. It holds that

$$\widetilde{\Phi}_1\widetilde{\Phi}_1^*=\widetilde{R^\uparrow}=(m^\uparrow)^{-1}R^\uparrow \quad\text{and}\quad \widetilde{\Phi}_2\widetilde{\Phi}_2^*=\widetilde{R^\downarrow}=(m^\downarrow)^{-1}R^\downarrow.$$

From the Lazarev–Lieb orthogonalization process (see Lemma 4.2), there exists a function $u\in C^\infty(\mathbb{R})$ with bounded derivatives such that

$$\langle\widetilde{\Phi}_1|\widetilde{\Phi}_2\mathrm{e}^{\mathrm{i}u}\rangle=\int_{\mathbb{R}^3}\left(\overline{\widetilde{\phi_1^\uparrow}}\widetilde{\phi_2^\uparrow}+\overline{\widetilde{\phi_1^\downarrow}}\widetilde{\phi_2^\downarrow}\right)\mathrm{e}^{iu}=0. \tag{4.5}$$

Once this function is chosen, there exists a function $v\in C^\infty(\mathbb{R})$ with bounded derivatives such that

$$\langle\widetilde{\Phi}_1|\widetilde{\Phi}_1\mathrm{e}^{\mathrm{i}v}\rangle=\langle\widetilde{\Phi}_1|\widetilde{\Phi}_2\mathrm{e}^{\mathrm{i}(u+v)}\rangle=\langle\widetilde{\Phi}_2\mathrm{e}^{\mathrm{i}u}|\widetilde{\Phi}_1\mathrm{e}^{\mathrm{i}v}\rangle=\langle\widetilde{\Phi}_2|\widetilde{\Phi}_2\mathrm{e}^{\mathrm{i}v}\rangle=0. \tag{4.6}$$

We finally set

$$\Phi_1:=\frac{1}{\sqrt{2}}\left(\sqrt{m^\uparrow}\widetilde{\Phi}_1+\sqrt{m^\downarrow}\widetilde{\Phi}_2\mathrm{e}^{\mathrm{i}u}\right)$$

and

$$\Phi_2:=\frac{1}{\sqrt{2}}\left(\sqrt{m^\uparrow}\widetilde{\Phi}_1-\sqrt{m^\downarrow}\widetilde{\Phi}_2\mathrm{e}^{\mathrm{i}u}\right)\mathrm{e}^{\mathrm{i}v}.$$

From (4.5), we deduce $\|\Phi_1\|^2=\|\Phi_2\|^2=1$, so that both Φ_1 and Φ_2 are normalized. Also, from (4.6), we get $\langle\Phi_1|\Phi_2\rangle=0$. Hence, $\{\Phi_1,\Phi_2\}$ is orthonormal. As $\widetilde{\Phi}_1$ and $\widetilde{\Phi}_2$ are in $H^1(\mathbb{R}^3,\mathbb{C}^2)$, and u and v have bounded derivatives, Φ_1 and Φ_2 are in $H^1(\mathbb{R}^3,\mathbb{C}^2)$. Finally, it holds that

$$\begin{aligned}\Phi_1\Phi_1^*+\Phi_2\Phi_2^*=&\frac{1}{2}\Big(m^\uparrow\widetilde{\Phi}_1\widetilde{\Phi}_1^*+m^\downarrow\widetilde{\Phi}_2\widetilde{\Phi}_2^*+2\sqrt{m^\uparrow m^\downarrow}\mathrm{Re}\left(\widetilde{\Phi}_1\widetilde{\Phi}_2^*\mathrm{e}^{-\mathrm{i}u}\right)\\ &+m^\uparrow\widetilde{\Phi}_1\widetilde{\Phi}_1^*+m^\downarrow\widetilde{\Phi}_2\widetilde{\Phi}_2^*-2\sqrt{m^\uparrow m^\downarrow}\mathrm{Re}\left(\widetilde{\Phi}_1\widetilde{\Phi}_2^*\mathrm{e}^{-\mathrm{i}u}\right)\Big)\\ =&m^\uparrow\widetilde{\Phi}_1\widetilde{\Phi}_1^*+m^\downarrow\widetilde{\Phi}_2\widetilde{\Phi}_2^*=R.\end{aligned}$$

We deduce that the Slater determinant $\mathscr{S}[\Phi_1,\Phi_2]$ represents R, so that $R\in\mathcal{J}_2^{\mathrm{Slater}}$. Altogether, $\mathcal{C}_2\subset\mathcal{J}_2^{\mathrm{Slater}}$, and therefore $\mathcal{C}_2=\mathcal{J}_2^{\mathrm{Slater}}$.

Case $N>2$. We proceed by induction. Let $R\in\mathcal{C}_{N+1}$ with $N\geq 2$, and suppose $\mathcal{C}_N=\mathcal{J}_N^{\rm Slater}$. We use the decomposition (4.4) and write $R=R^\uparrow+R^\downarrow$, where $R^{\uparrow/\downarrow}$ are two null-determinant hermitian matrices. For $\alpha\in\{\uparrow,\downarrow\}$, we note $m^\alpha:=\int_{\mathbb{R}^3}\rho_{R^\alpha}$. Since $m^\uparrow+m^\downarrow=N+1\geq 3$, at least $m^\uparrow$ or $m^\downarrow$ is greater than 1. Let us suppose without loss of generality that $m^\uparrow\geq 1$. We write $R=R_1+R_2$ with

$$R_1:=(m^\uparrow)^{-1}R^\uparrow \quad \text{and} \quad R_2:=\left(\left(1-(m^\uparrow)^{-1}\right)R^\uparrow+m^\downarrow R^\downarrow\right).$$

It holds that $R_1\in\mathcal{C}_1^0=\mathcal{J}_1^{\rm Slater}$ and $R_2\in\mathcal{C}_N=\mathcal{J}_N^{\rm Slater}$ (by induction). Together with Lemma 4.1, we deduce that $R\in\mathcal{J}_{N+1}^{\rm Slater}$. The result follows.

4.2. Proof of Lemma 3.5. Let $\Phi=(\phi^\uparrow,\phi^\downarrow)\in H^1(\mathbb{R}^3,\mathbb{C}^2)$. For $\alpha\in\{\uparrow,\downarrow\}$, we let τ^α be the phase of ϕ^α, so that $\phi^\alpha=\sqrt{\rho^\alpha}{\rm e}^{{\rm i}\tau^\alpha}$, and we suppose that $\tau^\uparrow$ and $\tau^\downarrow$ are in $C^1(\mathbb{R}^3)$. Let $(R,\mathbf{j})$ be the associated spin-density 2×2 matrix and paramagnetic current. It holds

$$R=\begin{pmatrix}\rho^\uparrow & \sigma\\ \overline{\sigma} & \rho^\downarrow\end{pmatrix}=\begin{pmatrix}|\phi^\uparrow|^2 & \phi^\uparrow\overline{\phi^\downarrow}\\ \phi^\downarrow\overline{\phi^\uparrow} & |\phi^\downarrow|^2\end{pmatrix}.$$

Setting $\tau=\tau^\uparrow-\tau^\downarrow$, we obtain $\sigma=|\sigma|{\rm e}^{{\rm i}\tau}=\sqrt{\rho^\uparrow\rho^\downarrow}{\rm e}^{{\rm i}\tau}$. The paramagnetic current is

$$\mathbf{j}=\rho^\uparrow\nabla\tau^\uparrow+\rho^\downarrow\nabla\tau^\downarrow=\rho\nabla\tau^\downarrow+\rho^\uparrow\nabla\tau=\rho\nabla\tau^\uparrow-\rho^\downarrow\nabla\tau.$$

In particular, using (3.5),

$$\frac{\mathbf{j}}{\rho}-\frac{{\rm Im}(\overline{\sigma}\nabla\sigma)}{\rho\rho^\downarrow}=\frac{\mathbf{j}-\rho^\uparrow\nabla\tau}{\rho}=\nabla\tau^\downarrow \quad \text{and} \quad \frac{\mathbf{j}}{\rho}+\frac{{\rm Im}(\overline{\sigma}\nabla\sigma)}{\rho\rho^\uparrow}=\nabla\tau^\uparrow$$

are curl-free.

4.3. Proof of Theorem 3.8. We break the proof in several steps.

Step 1: Any $R\in\mathcal{C}_N$ can be written as $R=R_1+R_2+R_3$ with $R_k\in\mathcal{C}_{N_k}^0$, $N_k\geq 4$. Let $R=\begin{pmatrix}\rho^\uparrow & \sigma\\ \overline{\sigma} & \rho^\downarrow\end{pmatrix}\in\mathcal{C}_N$, with $N\geq 12$. We write $\sqrt{R}=\begin{pmatrix}r^\uparrow & s\\ \overline{s} & r^\downarrow\end{pmatrix}$, with $r^\uparrow,r^\downarrow\in\left(H^1(\mathbb{R}^3)\right)^2$ and s in $H^1(\mathbb{R}^3,\mathbb{C})$. We write $R=R^\uparrow+R^\downarrow$, where $R^{\uparrow/\downarrow}$ were defined in (4.4). As in the proof of Theorem 3.2 for the case $N=2$, $R^{\uparrow/\downarrow}$ are hermitian and of null determinant, and $\sqrt{R^{\uparrow/\downarrow}}\in\mathcal{M}_{2\times 2}\left(H^1(\mathbb{R}^3,\mathbb{C})\right)$. However, it may hold that $\int{\rm tr}_{\mathbb{C}^2}[R^\uparrow]\notin\mathbb{N}^*$, so that $R^\uparrow$ is not in $\mathcal{C}_M^0$ for some $M\in\mathbb{N}^*$. In order to handle this difficulty, we will distribute the mass of $R^\uparrow$ and $R^\downarrow$ into three spin-density 2×2 matrices.
More specifically, let us suppose without loss of generality that $\int{\rm tr}_{\mathbb{C}^2}[R^\uparrow]\geq\int{\rm tr}_{\mathbb{C}^2}[R^\downarrow]$. We set

$$\begin{aligned}R_1&=(1-\xi_1)R^\uparrow+\xi_2R^\downarrow,\\ R_2&=\xi_1(1-\xi_3)R^\uparrow,\\ R_3&=(1-\xi_2)R^\downarrow+\xi_3R^\uparrow,\end{aligned}\tag{4.7}$$

where ξ_1,ξ_2,ξ_3 are suitable non-decreasing functions in $C^\infty(\mathbb{R}^3)$ that depend only on (say) r_1, such that, for $1\leq k\leq 3$, $0\leq\xi_k\leq 1$. We will choose them of the form $\xi_k(\mathbf{r})=0$ for $r_1<\alpha_k$ and $\xi_k(\mathbf{r})=1$ for $r_1\geq\beta_k>\alpha_k$, and such that

$$(1-\xi_1)\xi_2=(1-\xi_2)\xi_3=(1-\xi_1)\xi_3=0.\tag{4.8}$$

Finally, these functions are tuned so that $\int_{\mathbb{R}^3} \mathrm{tr}_{\mathbb{C}^2}(R_k) \in \mathbb{N}^*$ and $\int_{\mathbb{R}^3} \mathrm{tr}_{\mathbb{C}^2}(R_k) \geq 4$ for all $1 \leq k \leq 3$. We represent in Figure 4.1 a canonical example of such a triplet (ξ_1, ξ_2, ξ_3). In this figure, we clearly see how the non-overlapping condition (4.8) guarantees the null-determinant condition everywhere. Note that such a spatial decomposition could not have been performed with only two spin-density 2×2 matrices. Although it is not difficult to convince oneself that such functions ξ_k exist, we provide a full proof of this fact in the Appendix.

From (4.8), it holds, for all $1 \leq k \leq 3$, that $R_k \in \mathcal{C}^0_{N_K}$ and $R_1 + R_2 + R_3 = R^\uparrow + R^\downarrow = R$.

(a)

(b)

(c)

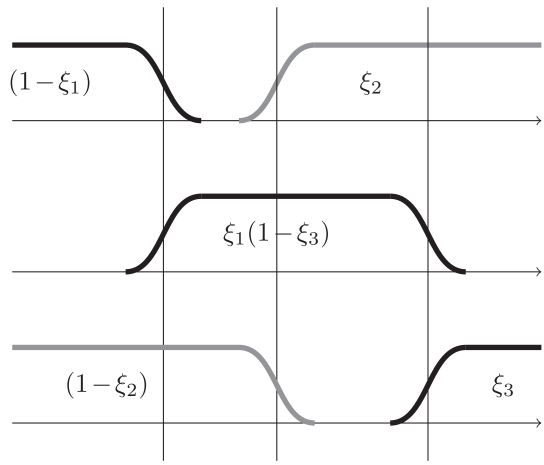

FIG. 4.1. *Weights of the matrices $R^\uparrow$ (black) and $R^\downarrow$ (gray) in (a) $R_1 = (1-\xi_1)R^\uparrow + \xi_2 R^\downarrow$, (b) $R_2 = \xi_1(1-\xi_3)R^\uparrow$, and (c) $R_3 = (1-\xi_2)R^\uparrow + \xi_3 R^\downarrow$.*

In the sequel, we decompose the current $\mathbf{j}$ in a similar way to (4.7). In order to simplify the notation, we introduce the total densities of $R^\uparrow$ and $R^\downarrow$:

$$f^\uparrow := |r^\uparrow|^2 + |s|^2 \quad \text{and} \quad f^\downarrow := |r^\downarrow|^2 + |s|^2.$$

Recall that $\rho = f^\uparrow + f^\downarrow$. We write $\mathbf{j} = \mathbf{j}_1 + \mathbf{j}_2 + \mathbf{j}_3$ with

$$\begin{aligned}
\mathbf{j}_1 &:= (1-\xi_1)\left(\frac{f^\uparrow}{\rho}\mathbf{j} - \mathrm{Im}(\overline{s}\nabla s)\right) + \xi_2\left(\frac{f^\downarrow}{\rho}\mathbf{j} + \mathrm{Im}(\overline{s}\nabla s)\right),\\
\mathbf{j}_2 &:= \xi_1(1-\xi_3)\left(\frac{f^\uparrow}{\rho}\mathbf{j} - \mathrm{Im}(\overline{s}\nabla s)\right),\\
\mathbf{j}_3 &:= (1-\xi_2)\left(\frac{f^\downarrow}{\rho}\mathbf{j} + \mathrm{Im}(\overline{s}\nabla s)\right) + \xi_3\left(\frac{f^\uparrow}{\rho}\mathbf{j} - \mathrm{Im}(\overline{s}\nabla s)\right).
\end{aligned} \tag{4.9}$$

Step 2: The pair $(R_1, \mathbf{j}_1)$ is representable by a Slater determinant. Following [9], we introduce

$$\xi(x) = \frac{1}{m}\int_{-\infty}^{x} \frac{1}{(1+y^2)^{(1+\delta)/2}}\mathrm{d}y,$$

where δ is the one in (3.6) and m is a constant chosen such that $\xi(\infty) = 1$. We then introduce

$$\begin{aligned}
\eta_{1,1}(\mathbf{r}) &= \frac{2}{N}\xi(\mathbf{r}+\alpha),\\
\eta_{1,2}(\mathbf{r}) &= \frac{2}{N-1}\xi(x_1+\beta)(1-\eta_1(\mathbf{r})),\\
\eta_{1,3}(\mathbf{r}) &= \frac{2}{N-2}\xi(x_2+\gamma)(1-\eta_1(\mathbf{r})-\eta_2(\mathbf{r})),\\
\eta_{1,k}(\mathbf{r}) &= \frac{1}{N-3}(1-\eta_1(\mathbf{r})-\eta_2(\mathbf{r})-\eta_3(\mathbf{r})) \quad \text{for} \quad 4\le k\le N,
\end{aligned} \tag{4.10}$$

where α,β,γ are tuned so that, if $\rho_1 := \mathrm{tr}_{\mathbb{C}^2} R_1$ denotes the total density of R_1, then

$$\forall 1\le k\le N_k, \quad \int_{\mathbb{R}^3}\eta_{1,k}\rho_1 = 1. \tag{4.11}$$

It can be checked (see [9]) that $\eta_{1,k}\ge 0$ and $\sum_{k=1}^N \eta_{1,k}=1$. We seek orbitals of the form

$$\Phi_{1,k} := \sqrt{\eta_{1,k}}\left(\sqrt{(1-\xi_1)}\begin{pmatrix} r^\uparrow \\ \overline{s}\end{pmatrix} + \sqrt{\xi_2}\begin{pmatrix} s \\ r^\downarrow\end{pmatrix}\right)\mathrm{e}^{\mathrm{i}u_{1,k}}, \ 1\le k\le N_1,$$

and where the phases $u_{1,k}$ will be chosen carefully later. From (4.8), we recall that $(1-\xi_1)\xi_2=0$, so that, by construction, $\Phi_{1,k}$ is normalized and

$$\Phi_{1,k}\Phi_{1,k}^* = \eta_{1,k}R_1.$$

Let us suppose for now that the phases $u_{1,k}$ are chosen so that the orbitals are orthogonal. This will indeed be achieved, thanks to the Lazarev–Lieb orthogonalization process (see Lemma 4.2). Then, $\Psi_1 := \mathcal{S}[\Phi_{1,1},\ldots,\Phi_{1,N}]$ represents the spin-density 2×2 matrix R_1. According to (2.2), the paramagnetic current of Ψ is (we recall that $r^\uparrow$ and $r^\downarrow$ are real-valued, and we write $s=|s|\mathrm{e}^{\mathrm{i}\tau}$ for simplicity)

$$\begin{aligned}
\mathbf{j}_\Psi &= \sum_{k=1}^{N_1}\eta_{1,k}(1-\xi_1)\left(|r^\uparrow|^2\nabla u_{1,k}+|s|^2\nabla(-\tau+u_{1,k})\right)\\
&\quad+\sum_{k=1}^{N_1}\eta_{1,k}\xi_2\left(|s|^2\nabla(\tau+u_{1,k})+|r^\downarrow|^2\nabla u_{1,k}\right)\\
&= \left((1-\xi_1)f^\uparrow+\xi_2 f^\downarrow\right)\left(\sum_{k=1}^{N_1}\eta_{1,k}\nabla u_{1,k}\right)+(\xi_2-(1-\xi_1))|s|^2\nabla\tau.
\end{aligned}$$

Since $|s|^2\nabla\tau = \mathrm{Im}(\overline{s}\nabla s)$, this current is equal to the target current $\mathbf{j}_1$ defined in (4.9) if and only if

$$\rho_1\frac{\mathbf{j}}{\rho} = \rho_1\sum_{k=1}^{N_1}\eta_k\nabla u_{1,k}.$$

In [9], Lieb and Schrader provided an explicit solution of this system when $N_1\ge 4$ [1] (their proof uses Lemma 4.2 and in particular the fact that the phase may be chosen

[1] In the same article, the authors recall (see [12] for instance) that there exist pairs $(\rho,\mathbf{j})$ for which no smooth solution exists when $N_1=2$. The case $N_1=3$ is still open. Of course, should someone find an explicit solution for $N_1=3$, the condition $N\ge 12$ in Theorem 3.8 could be replaced by the weaker condition $N\ge 9$.

to vary in one direction only). We do not repeat the proof, but we emphasize the fact that, because condition (3.6) holds true, the phases $u_{1,k}$ can be chosen to have bounded derivatives, so that the functions $\Phi_{1,k}$ are in $H^1(\mathbb{R}^3,\mathbb{C}^2)$. Also, as their proof relies on the Lazarev–Lieb orthogonalization process, it is possible to choose the phases $u_{1,k}$ so that the functions $\Phi_{1,k}$ are orthogonal, and orthogonal to a finite-dimensional subspace of $L^2(\mathbb{R}^3,\mathbb{C}^2)$.

Altogether, we proved that the pair $(R_1,\mathbf{j}_1)$ is representable by the Slater determinant $\mathscr{S}[\Phi_{1,1},\ldots,\Phi_{1,N_1}]$.

Step 3: Representability of $(R_2,\mathbf{j}_2)$ and $(R_3,\mathbf{j}_3)$, and finally of $(R,\mathbf{j})$. In order to represent the pair $(R_2,\mathbf{j}_2)$, we first construct the functions $\eta_{2,k}$ for $1\le k\le N_2$ of the form (4.10) so that (4.11) holds for $\rho_2:=\mathrm{tr}_{\mathbb{C}^2}R_2$. We then seek orbitals of the form

$$\Phi_{2,k}:=\sqrt{\eta_{2,k}\xi_1(1-\xi_3)}\begin{pmatrix} r^{\uparrow}\\ \overline{s}\end{pmatrix}\mathrm{e}^{\mathrm{i}u_{2,k}},\quad \text{for}\quad 1\le k\le N_2.$$

Reasoning as above, the Slater determinant of these orbitals represents the pair $(R_2,\mathbf{j}_2)$ if and only if

$$\rho_2\frac{\mathbf{j}_2}{\rho}=\rho_2\sum_{k=1}^{N_2}\eta_{2,k}\nabla u_{2,k}.$$

Again, due to the fact that $N_2\ge 4$, this equation admits a solution. Moreover, it is possible to choose the phases $u_{2,k}$ so that the functions $\Phi_{2,k}$ are orthogonal to the previously constructed $\Phi_{1,k}$.

We repeat again this argument for the pair $(R_3,\mathbf{j}_3)$. Once the new set of functions $\eta_{3,k}$ is constructed, we seek orbitals of the form

$$\Phi_{3,k}:=\sqrt{\eta_{3,k}}\left(\sqrt{(1-\xi_2)}\begin{pmatrix} s\\ r^{\downarrow}\end{pmatrix}+\sqrt{\xi_3}\begin{pmatrix} r^{\uparrow}\\ \overline{s}\end{pmatrix}\right)\mathrm{e}^{\mathrm{i}u_{3,k}}$$

and construct the phases so that the functions $\Phi_{3,k}$ are orthogonal to the functions $\Phi_{1,k}$ and $\Phi_{2,k}$.

Altogether, the pair $(R,\mathbf{j})$ is represented by the (finite energy) Slater determinant $\mathscr{S}[\Phi_{1,1},\ldots,\Phi_{1,N_1},\Phi_{2,1},\ldots,\Phi_{2,N_2},\Phi_{3,1},\ldots,\Phi_{3,N_3}]$, which concludes the proof.

Appendix A. We explain in this section how to construct three functions $\xi_1,\xi_2,\xi_3\in(C^\infty(\mathbb{R}))^3$ like in Figure 4.1. In order to simplify the notation, we introduce

$$f(r):=\iint_{\mathbb{R}\times\mathbb{R}}\mathrm{tr}_{\mathbb{C}^2}(R^{\downarrow})(r,r_2,r_3)\;\mathrm{d}r_2\mathrm{d}r_3,$$
$$g(r):=\iint_{\mathbb{R}\times\mathbb{R}}\mathrm{tr}_{\mathbb{C}^2}(R^{\uparrow})(r,y,z)\;\mathrm{d}r_2\mathrm{d}r_3,$$

where $R^{\uparrow}$ and $R^{\downarrow}$ were defined in (4.4). We denote

$$F(\alpha)=\int_{-\infty}^{\alpha}f(x)\mathrm{d}x\quad\text{and}\quad G(\alpha)=\int_{-\infty}^{\alpha}g(x)\mathrm{d}x,$$

and finally $\mathcal{F}=F(\infty)=\int_{\mathbb{R}}f$ and $\mathcal{G}=G(\infty)=\int_{\mathbb{R}}g$. Note that F and G are continuous non-decreasing functions going from 0 to $\mathcal{F}$ (resp. $\mathcal{G}$) and that it holds $\mathcal{F}+\mathcal{G}=N$. Let us suppose without loss of generality that $\mathcal{F}\le\mathcal{G}$, so that $0\le\mathcal{F}\le N/2\le\mathcal{G}\le N$. If $\mathcal{F}=0$, then $R^{\downarrow}=0$ and we can choose $R_1=R_2=(4/N)R^{\uparrow}\in\mathcal{C}_4^0$ and $R_3=(N-8)/NR^{\uparrow}\in\mathcal{C}_{N-8}^0$. Since $N\geq 12$, it holds that $N-8\geq 4$, so that this is the desired decomposition. We now consider the case $\mathcal{F}\neq 0$.

In order to keep the notation simple, we will only study the case $\mathcal{F}<8$ (the case $\mathcal{F}>8$ is similar by replacing the integer 4 by a greater integer M such that $\mathcal{F}<2M<N-4$ in the sequel). We seek for α such that

$$\begin{cases} \int_{-\infty}^{\alpha} f(x)\mathrm{d}x<4 \quad \text{and} \quad \int_{-\infty}^{\alpha} f(x)+\int_{\alpha}^{\infty} g(x)>4, \\ \int_{\alpha}^{\infty} f(x)\mathrm{d}x<4 \quad \text{and} \quad \int_{-\infty}^{\alpha} g(x)\mathrm{d}x+\int_{\alpha}^{\infty} f(x)\mathrm{d}x>4, \end{cases}$$

or equivalently

$$\mathcal{F}-4<F(\alpha)<4 \quad \text{and} \quad F(\alpha)+4-\mathcal{F}<G(\alpha)<F(\alpha)+\mathcal{G}-4. \tag{A.1}$$

Let $\alpha_{(\mathcal{F}-4)}$ be such that $F(\alpha_{(\mathcal{F}-4)})=\mathcal{F}-4$ (with $\alpha_{(\mathcal{F}-4)}=-\infty$ if $\mathcal{F}\leq 4$) and $\alpha_{(4)}$ be such that $F(\alpha_{(4)})=4$ (with $\alpha_{(4)}=+\infty$ if $\mathcal{F}\leq 4$). As F is continuous and non-decreasing, the first equation of (A.1) is satisfied whenever $\alpha_{(\mathcal{F}-4)}<\alpha<\alpha_{(4)}$. The function $[\alpha_{(\mathcal{F}-4)},\alpha_4]\ni\alpha\mapsto m(\alpha):=F(\alpha)+4-\mathcal{F}$ goes continuously and non-decreasingly from 0 to $8-\mathcal{F}$, and the function $[\alpha_{(\mathcal{F}-4)},\alpha_4]\ni\alpha\mapsto M(\alpha):=F(\alpha)+\mathcal{G}-4$ goes continuously and non-decreasingly from $N-8$ to $\mathcal{G}$ between $\alpha_{(\mathcal{F}-4)}$ and $\alpha_{(4)}$. In particular, since $G(\alpha)$ goes continuously and non-decreasingly from 0 to $\mathcal{G}$, only three cases may happen:

- There exists $\alpha_0\in(\alpha_{(\mathcal{F}-4)},\alpha_{(4)})$ such that $m(\alpha_0)<G(\alpha_0)<M(\alpha_0)$. In this case, (A.1) holds for $\alpha=\alpha_0$. By continuity, there exists $\varepsilon>0$ such that

$$\begin{cases} F(\alpha+\varepsilon)<4, \\ F(\alpha)+\mathcal{G}-G(\alpha+\varepsilon)>4, \\ G(\alpha)+\mathcal{F}-F(\alpha+\varepsilon)>4. \end{cases}$$

Let $\xi_2\in C^\infty(\mathbb{R})$ be a non-decreasing function such that $\xi_2(x)=0$ for $x<\alpha$ and $\xi_2(x)=1$ for $x>\alpha+\varepsilon$. Then, as $0\leq\xi_2\leq 1$, it holds that

$$\int_{\mathbb{R}}(1-\xi_2)f\leq F(\alpha+\varepsilon)<4$$

and

$$\int_{\mathbb{R}}(1-\xi_2)f+\int_{\alpha+\varepsilon}^{\infty} g\geq F(\alpha)+\mathcal{G}-G(\alpha+\varepsilon)>4.$$

We deduce that there exists an non-decreasing function $\xi_3\in C^\infty(\mathbb{R})$ such that $\xi_3(x)=0$ for $x<\alpha+\varepsilon$ and such that

$$\int_{\mathbb{R}}(1-\xi_2)f+\xi_3 g=4.$$

Note that $(1-\xi_2)\xi_3=0$. On the other hand, from

$$\begin{cases} \int_{\mathbb{R}}\xi_2 f\leq\mathcal{F}-F(\alpha)<4 \\ \int_{\mathbb{R}}\xi_2 f+\int_{-\infty}^{\alpha} g\geq\mathcal{F}-F(\alpha+\varepsilon)+G(\alpha)>4, \end{cases}$$

we deduce that there exists a non-decreasing function $\xi_1 \in C^\infty(\mathbb{R})$ such that $\xi_1(x) = 1$ for $x > \alpha$:

$$\int_{\mathbb{R}} (1-\xi_1) g + \xi_2 f = 4.$$

and $(1-\xi_1)\xi_2 = (1-\xi_1)\xi_3 = 0$. Finally, we set

$$\begin{aligned} R_1 &= (1-\xi_1) R^\uparrow + \xi_2 R^\downarrow, \\ R_2 &= \xi_1 (1-\xi_3) R^\uparrow, \\ R_3 &= (1-\xi_2) R^\downarrow + \xi_3 R^\uparrow. \end{aligned}$$

By construction, $R = R^\uparrow + R^\downarrow = R_1 + R_2 + R_3$, $R_1 \in \mathcal{C}_4^0$, and $R_3 \in \mathcal{C}_4^0$. We deduce that $R_4 \in \mathcal{C}_{N-8}^0$, where $N-8 \geq 4$. This leads to the desire decomposition.

- For all $\alpha \in (\alpha_{(\mathcal{F}-4)}, \alpha_{(4)})$, it holds that $G(\alpha) < m(\alpha)$. Note that this may only happen if $m(\alpha_{(4)}) > 0$ or $\mathcal{F} < 4$, so that $\mathcal{G} > N - 4 \geq 8$. It holds that $G(\alpha_{(\mathcal{F}-4)}) = 0$, so that $g(r)$ is null for $r < \alpha_{(\mathcal{F}-4)}$. Let α_0 be such that $\alpha_{(\mathcal{F}-4)} < \alpha_0 < \alpha_{(4)}$. As

$$\int_{\mathbb{R}} f = \mathcal{F} > 4 \quad \text{and} \quad \int_{\alpha_0}^{\infty} f = \mathcal{F} - F(\alpha_0) < 4,$$

there exists a non-decreasing function $\xi_1 \in C^\infty(\mathbb{R})$ satisfying $\xi_1(x) = 1$ for $x \geq \alpha_0$ and such that

$$\int_{\mathbb{R}} \xi_1 f = 4.$$

Now, since $G(\alpha_{(4)}) < m(\alpha_{(4)}) = 8 - \mathcal{F}$, it holds that

$$\begin{cases} \int_{\mathbb{R}} (1-\xi_1) f \leq F(\alpha_{(4)}) = 4 \\ \int_{R} (1-\xi_1) f + \int_{\alpha_0}^{\infty} g \geq F(\alpha_{(\mathcal{F}-4)}) + \mathcal{G} - G(\alpha_{(4)}) > 4. \end{cases}$$

There exists a non-decreasing function $\xi_2 \in C^\infty(\mathbb{R})$ satisfying $\xi_2(x) = 0$ for $x \leq \alpha_0$ and such that

$$\int_{\mathbb{R}} (1-\xi_1) f + \xi_2 g = 4.$$

Note that $(1-\xi_1)\xi_2 = 0$. Finally, we set

$$\begin{aligned} R_1 &= \xi_1 R^\downarrow, \\ R_2 &= (1-\xi_2) R^\uparrow, \\ R_3 &= \xi_2 R^\uparrow + (1-\xi_1) R^\downarrow. \end{aligned}$$

By construction, it holds that $R = R_1 + R_2 + R_3$, $R_1 \in \mathcal{C}_4^0$, and $R_3 \in \mathcal{C}_4^0$. We deduce $R_2 \in \mathcal{C}_{N-8}^0$, and the result follows.

- For all $\alpha \in (\alpha_{(\mathcal{F}-4)}, \alpha_{(4)})$, it holds that $\mathcal{G}(\alpha) > M(\alpha)$. This case is similar to the previous one.

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