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Journées Équations aux dérivées partielles (1997), p. 1-8

http://www.numdam.org/item?id=JEDP_1997____A9_0

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HEAVY MOLECULES IN THE STRONG MAGNETIC FIELD. †

VICTOR IVRII

ABSTRACT. We consider heavy atoms and molecules in the constant magnetic field under condition $B \ll N^3$ where B is the intensity of the magnetic field and N is the number of electrons and discuss asymptotics of the ground state and ionization energies and estimates of the negative excessive charges for atoms and molecules and estimates of positive excessive charges for molecules.

0. Preface. Multiparticle quantum theory is one of the main topics of modern mathematical physics, and one of the central questions in this theory is the problem of the high-density limit. There are different versions of this problem including the analysis of a heavy atom, and the analysis of a molecule consisting of heavy atoms. These two versions are the most popular and I am dealing with them.

The first step in the analysis is usually the Thomas-Fermi approximation, which leads to a non-linear partial differential system describing density and effective potential. This part of the theory is basically done.

However, justification of this approximation, error estimates and the obtaining of additional correction terms (Scott and Dirac-Schwinger) is a much more difficult matter requiring quite different techniques. Until last years the main tool has been variational methods of mathematical physics. After no less than 20 years of intensive investigations there remain major open problems, and even recently essential progress was obtained.

In some steps of the analysis there arise problems lying within the theory of semiclassical spectral asymptotics. This is a highly developed theory with the very strong machinery. However, problems specific for the multiparticle quantum theory have never been treated, and these problems have essential differences from standard problems of this theory. As a result these particular problems were treated either by variational methods as well (which led to non-accurate error estimate and the impossibility of recovering correction terms) or by separation of variables and investigation of ordinary differential equation by the WKB method

† Work was partially supported by NSERC grant OGP0138277.

(this approach has provided very precise error estimate but works only in the very special cases).

Few years ago M.Sigal and I applied semiclassical spectral asymptotics methods to the multiparticle quantum theory problems and justified the Scott correction term for the ground state energy for large molecules. Automatically this provided some progress in other problems as well. After I managed to recover Dirac-Schwinger corrections as well.

Now I am studying the same problem with magnetic field and this case seems to be really challenging. I would like to present my recent results. I was stimulated by papers [LSY1,2] where the principal term was recovered and it was shown that the cases $B \ll N^3$ and $B \gg N^3$ are really different (and the transition zone is the most difficult!). I have been considering the first and this is my final report. I hope to advance to the second case shortly.

1. Quantum mechanics model. Let us consider the following operator (quantum Hamiltonian)

$$(1) \quad H = \sum_{1 \leq j \leq N} \left((i\nabla_{x_j} - \mathbf{A}(x_j))^2 - B - V(x_j) \right) + \sum_{1 \leq j < k \leq N} |x_j - x_k|^{-1}$$

describing N electrons in the external electro-magnetic field with the potentials \mathbf{A}, V and repulsing one another according Coulomb law. Here $x_j \in \mathbb{R}^3$ and $(x_1, \dots, x_N) \in \mathbb{R}^{3N}$, functions $\mathbf{A}, V(x)$ are assumed to be real-valued; B is the scalar intensity of the magnetic field and we assume that

$$(2) \quad \mathbf{B} = \nabla \times A = B\mathbf{k}, \quad B = \text{const.}$$

Mass is equal to $\frac{1}{2}$ and Plank constant and a charge are equal to 1 here. The crucial question is the quantum statistics. We assume that the particles (electrons) are fermions. That means that the Hamiltonian should be considered on the Fock space $\mathcal{H} = \bigwedge_{1 \leq j \leq N} L^2(\mathbb{R}^3)$ of the functions antisymmetric with respect to all variables x_1, \dots, x_N while for bosons one should consider a space symmetric functions. I neglect the fact that one particle is described by the wave function $\phi \in L^2(\mathbb{R}^3, \mathbb{C}^q)$ rather than by the wave function $\phi \in L^2(\mathbb{R}^3, \mathbb{C})$ and that the more correct form of the one particle free Hamiltonian is a bit different because for $q \geq 1$ no essential modifications of arguments is required and results are the same with other numerical coefficients. We consider a large molecule with

$$(3) \quad V(x) = \sum_{1 \leq k \leq M} \frac{Z_k}{|x - x_k|}$$

where $Z_k > 0$ and x_k are charges and locations of nuclei, the number of nuclei M is fixed. It is known that operator H is self-adjoint and semi-bounded from below. This model, rather inconsistent from the physical point of view, is nevertheless very popular among mathematical physicists.

First, I am interested in the *ground state energy* $E = E(N)$ of our system i.e. in the lowest eigenvalue of the operator H on \mathcal{H} . The first approximation is the Hartree-Fock (or Thomas-Fermi) theory. Namely, let us introduce the space density of the particle with the state $\psi \in \mathcal{H}$:

$$(4) \quad \rho(x) = \rho_\psi(x) = N \int |\psi(x, x_2, \dots, x_N)|^2 dx_2 \cdots dx_N.$$

Then Hamiltonian, describing the corresponding "quantum liquid" is

$$(5) \quad \mathcal{E}^{\text{TF}}(\rho) = \int (j_B(\rho) - V\rho) dx + \frac{1}{2} D(\rho, \rho),$$

$$D(\rho, \rho) \stackrel{\text{def}}{=} \iint |x - y|^{-1} \rho(x) \rho(y) dx dy$$

where $j_B(\rho)$ is the Legendre transformation of

$$(6) \quad P_B(W) = 2\kappa \sum_{n \geq 0} (W - 2nB)_+^{\frac{3}{2}} B;$$

here and below κ is reserved for well-known numerical constant. The right-hand expression is the Riemann sum; replacing it by an integral as $B \rightarrow 0$ one obtains $P(W) = \frac{2}{5} \kappa W_+^{\frac{5}{2}}$ and $j(\rho) = \frac{3}{5} \kappa^{-\frac{2}{3}} \rho^{\frac{5}{3}}$ which are well-known in non-magnetic case. The classical sense of the second and the third terms is clear and the first term is the kinetic energy in the semiclassical approximation; $j_B(\rho)$ is its spatial density. So, the problem is to minimize this the functional under restrictions:

$$(7) \quad \mathcal{E}^{\text{TF}}(\rho) \mapsto \inf \quad \text{for } \rho \geq 0, \quad \int \rho dx \leq N.$$

There exists a unique solution ρ^{TF} with $\int \rho^{\text{TF}} dx = \min(N, Z)$. Some properties of this solution are known as well; in particular, is known that the main contributions to $E^{\text{TF}} = \mathcal{E}^{\text{TF}}(\rho^{\text{TF}})$ and $\int \rho^{\text{TF}} dx$ are delivered by zone $\{\ell(x) \asymp r_T\}$ with $r_T = N^{-\frac{1}{3}}$ and $r_T = B^{-\frac{2}{5}} N^{\frac{1}{5}}$ for $B \leq N^{\frac{4}{5}}$ respectively; $\ell(x) = \min_k |x - x_k|$. Further, ρ^{TF} is supported in $\{\ell(x) \leq cr_S\}$ with $r_S = \min((Z - N)^{-\frac{1}{3}}, B^{-\frac{1}{4}})$ (for $N \leq Z$) and it is not finite supported for $Z = N, B = 0$ only. Finally, Thomas-Fermi potential $W^{\text{TF}} = V - |x|^{-1} * \rho^{\text{TF}}$ satisfies

$$(8) \quad P'_B(W^{\text{TF}} + \nu) = -\frac{1}{4\pi} \Delta(V - W) = \rho^{\text{TF}}$$

where the chemical potential ν is the Lagrange multiplier in the variational problem (7) (and the value of $-W^{\text{TF}}$ on the boundary of $\text{supp } \rho^{\text{TF}}$) and $\nu \leq 0$ for $N \leq Z$.

2. Asymptotics of the ground state energy. I managed to prove the following

THEOREM 1. [Ivr3] Let $c^{-1}N \leq Z_j \leq cN$, $B \leq N^3$ and

$$(9) \quad a = \min_{j < k} |x_j - x_k| \geq c^{-1} \min(N^{-\frac{1}{3}}, B^{-\frac{2}{3}} N^{\frac{1}{3}}).$$

(i) Then

$$(10) \quad |E - \mathcal{E}^{\text{TF}} - \text{Scott}| \leq C(\mathcal{R} + \mathcal{R}_1 + \mathcal{R}' + \mathcal{R}'')$$

with the Scott correction term $\text{Scott} = \kappa_{\text{Scott}} \sum_k Z_k^2$, $\kappa_{\text{Scott}} = q^{-1}$ and remainder estimates

$$(11) \quad \begin{aligned} \mathcal{R} &= N^{\frac{5}{3}} + N^{\frac{3}{5}} B^{\frac{4}{5}}, & \mathcal{R}_1 &= B^{\frac{1}{3}} N^{\frac{4}{3}} \\ \mathcal{R}' &= B^{\frac{4}{5}} N^{\frac{3}{5}} L^4, & L &= 1 + \log N^3 (B+1)^{-1} \end{aligned}$$

and

$$(12) \quad \mathcal{R}'' = \begin{cases} \min((Z-N)^{\frac{3}{4}} N^{\frac{9}{20}} B^{\frac{3}{5}}, (Z-N)^{\frac{7}{15}} N^{\frac{8}{15}} B^{\frac{2}{3}}) L^4 & \text{for } N^{\frac{4}{3}} \leq B \leq N^3, \\ \min((Z-N)^{\frac{3}{4}} B^{\frac{15}{16}}, (Z-N)^{\frac{7}{15}} B^{\frac{16}{15}}) L^4 & \text{for } (Z-N)^{\frac{4}{3}} \leq B \leq N^{\frac{4}{3}}, \\ (Z-N)^{-\frac{7}{9}} B^2 L^4 & \text{for } B \leq (Z-N)^{\frac{4}{3}}; \end{cases}$$

here and below C depends only on M and c .

(ii) Further, for $M = 1$ these estimates hold with $\mathcal{R}' = \mathcal{R}'' = 0$ [Ivr2].

REMARK 2. (i) It is known that

$$(13) \quad E^{\text{TF}} \asymp \begin{cases} N^{\frac{5}{3}} & \text{for } B \leq N^{\frac{4}{3}}, \\ B^{\frac{2}{5}} N^{\frac{9}{5}} & \text{for } N^{\frac{4}{3}} \leq B \leq N^3; \end{cases}$$

(ii) One can easily improve a bit logarithmic factor L^4 but who cares?

(iii) For $B \ll N$, $a \gg N^{-\frac{1}{3}}$ one can improve these asymptotics and recover Dirac-Schwinger corrections (see [Ivr2,3]). \diamond

The proof of theorem 1 leads us to

THEOREM 3. [Ivr5] In frames of theorem 1 the following estimate holds for the ground state density ρ_ψ :

$$(14) \quad D(\rho_\psi - \rho^{\text{TF}}, \rho_\psi - \rho^{\text{TF}}) \leq C(\mathcal{R} + \mathcal{R}' + \mathcal{R}'').$$

Further, for $N = 1$ these estimates hold with $\mathcal{R}' = \mathcal{R}'' = 0$.

3. Maximal negative charge and ionization energy. One can prove easily (adapting classical arguments of G.Zhislin [Zh]) that

$$I(N, Z) \stackrel{\text{def}}{=} E(N-1, Z) - E(N, Z) > 0$$

for $N \leq Z$ which means that our system can bind at least Z electrons. Let us estimate now the maximal N satisfying this inequality (so, $N - Z$ is the excessive negative charge). It's known that the answer is $N = Z$ in frames of Thomas-Fermi theory (in which negative ions are impossible). Repeating with modifications arguments of [SSS] and [Sol1] and using heavily ground state energy asymptotics one can prove

THEOREM 4. [Ivr5] *In frames of theorem 1 for $N \geq Z$ satisfying with $I(N, Z) > 0$ the following estimates hold:*

$$(15) \quad |N - Z| \leq C \begin{cases} N^{\frac{5}{7}} & \text{for } B \leq N^{\frac{20}{21}} \\ N^{\frac{5}{6}} B^{-\frac{1}{8}} + B^{\frac{1}{2}} L^2 & \text{for } N^{\frac{20}{21}} \leq B \leq N^{\frac{4}{3}} L^2 \\ B^{\frac{1}{5}} N^{\frac{2}{5}} & \text{for } N^{\frac{4}{3}} \leq B \leq N^3 \end{cases}$$

$$(16) \quad I(N, Z) \leq C \begin{cases} N^{\frac{20}{21}} & \text{for } B \leq N^{\frac{20}{21}} \\ N^{\frac{20}{21}} + B^{\frac{5}{7}} L^3 & \text{for } N^{\frac{20}{21}} \leq B \leq N^{\frac{4}{3}} \\ B^{\frac{22}{35}} N^{\frac{4}{35}} L^2 & \text{for } N^{\frac{4}{3}} \leq B \leq N^3 \end{cases}$$

Moreover, for $M = 1$ these estimates hold with $L = 1$.

REMARK 5. (i) One can see the new "breaking point": $B \asymp N^{\frac{20}{21}}$. The origin is rather simple: it follows from theorem 3 that ρ^{TF} is good approximation for ρ_ψ as $\ell(x) \ll N^{-\frac{5}{21}}$, $B \leq N^{\frac{20}{21}}$ and as $\ell(x) \ll r_S$, $B \geq N^{\frac{20}{21}}$. However, all these exponents seem to be technical rather than "physical".

(ii) This result was motivated by [LSY1,2], [SSS], [Sol1] and I used big chunks of their arguments completed by theorem 3.

(iii) Further, for $B \leq N$ one can improve estimates (15),(16) adding the same factor $\left((aN)^{-1} + BN^{-1} + N^{-1}\right)^\delta$ into their right-hand expressions. This remains true for estimates (17), (18) and (19) below. \diamond

Now I would like to estimate from above and below the ionization energy. I will do it for $M = 1$ or $B = 0$ (or small enough) only (estimates are rather tedious otherwise).

THEOREM 6. [Ivr5] *Let either $M = 1$ or $B = 0$. Then the following estimate holds:*

$$(17) \quad I_N + \nu \leq C \begin{cases} \min\left((Z - N)^{\frac{17}{18}} N^{\frac{5}{18}}, (Z - N)^{\frac{17}{12}} N^{\frac{5}{12}} B^{-\frac{1}{2}}\right) & \text{for } (Z - N) \geq B^{\frac{3}{4}} + N^{\frac{5}{7}} \\ N^{\frac{20}{21}} + (Z - N)^{\frac{3}{4}} N^{\frac{5}{12}} & \text{for } (Z - N)^{\frac{4}{3}} \leq B \leq N^{\frac{4}{3}} \\ B^{\frac{22}{35}} N^{\frac{4}{35}} + (Z - N)^{\frac{3}{4}} B^{\frac{1}{2}} N^{-\frac{1}{4}} & \text{for } B \geq N^{\frac{4}{3}} \end{cases}$$

THEOREM 7. [Ivr5] *Let either $M = 1$ or $B = 0$ and let one of the following conditions be fulfilled:*

$$(18)_1 \quad (Z - N) \geq N^{\frac{21}{25}}, \quad (Z - N)^{\frac{4}{3}} \geq B;$$

$$(18)_2 \quad (Z - N) \geq N^{\frac{7}{5}} B^{-\frac{1}{2}}, \quad (Z - N)^{\frac{4}{3}} \leq B \leq N^{\frac{4}{3}}, \quad B \geq N^{\frac{42}{55}};$$

$$(18)_3 \quad (Z - N) \geq N^{\frac{13}{25}} B^{\frac{4}{25}}, \quad N^{\frac{4}{3}} \leq B \leq N^3.$$

Then

$$(19) \quad I_N + \nu \geq -C(\mathcal{S}_1 + \mathcal{S}_2)$$

with

$$(20) \quad \mathcal{S}_1 = \begin{cases} N^{\frac{1}{2}}(Z - N)^{\frac{31}{42}} & \text{in case (18)}_1 \\ & \text{for } B \leq N^{\frac{1}{2}}(Z - N)^{\frac{31}{42}}; \\ N^{\frac{7}{10}}(Z - N)^{\frac{31}{30}} B^{-\frac{2}{5}} & \text{in case (18)}_1 \\ & \text{for } (Z - N)^{\frac{4}{3}} \geq B \geq N^{\frac{1}{2}}(Z - N)^{\frac{31}{42}}; \\ N^{\frac{7}{10}}(Z - N)^{\frac{1}{2}} & \text{in case (18)}_2; \\ N^{\frac{3}{50}} B^{\frac{12}{25}}(Z - N)^{\frac{1}{2}} & \text{in case (18)}_3. \end{cases}$$

and

$$(21) \quad \mathcal{S}_2 = \begin{cases} N^{\frac{4}{11}}(Z - N)^{\frac{34}{33}} B^{-\frac{2}{11}} & \text{in case (3.16)}_1 \\ & \text{for } (Z - N)^{\frac{4}{3}} \geq B \geq N^{\frac{1}{2}}(Z - N)^{\frac{31}{42}}; \\ N^{\frac{14}{33}}(Z - N)^{\frac{6}{11}} B^{\frac{2}{11}} & \text{in case (3.16)}_2; \\ N^{\frac{2}{15}} B^{\frac{2}{5}}(Z - N)^{\frac{6}{11}} & \text{in case (3.16)}_3. \end{cases}$$

In particular, for $B \leq \mathcal{S} = N^{\frac{1}{2}}(Z - N)^{\frac{31}{42}}$ we get estimate $I_N + \nu \geq -C\mathcal{S}$.

6. Maximal positive charge. The last question I want to address is "What can be maximal positive excessive charge for stable molecule ($M \geq 2$ is very important here) where now positions x_1, \dots, x_M are assumed to be optimal as well". Sure, one needs to count energy of interaction between nuclei. It is known, that there no stable molecules in frames of Thomas-Fermi theory. Further, I want to find the estimate from below for the distance between nuclei.

THEOREM 8. [Ivr5] *Let in frames of theorem 1 with $M \geq 2$*

$$(22) \quad E(N, \underline{Z}, \underline{x}) + \sum_{j < k} Z_j Z_k |x_j - x_k|^{-1} <$$

$$\min_{N_1, \dots, N_n: N_1 + \dots + N_n = N} \sum_{1 \leq j \leq n} E_j(N_j, Z_j)$$

(where E_j are atomic ground state energies). Then

(i) For all $i \neq j$

$$(23) \quad |x_i - x_j| \geq \begin{cases} \epsilon N^{-\frac{5}{21}} & \text{for } B \leq N^{\frac{20}{21}} \\ \bar{r}_i + \bar{r}_j - CB^{-\frac{5}{16}} & \text{for } N^{\frac{20}{21}} \leq B \leq N^{\frac{4}{3}} \\ \bar{r}_i + \bar{r}_j - CB^{-\frac{7}{20}} N^{\frac{1}{20}} & \text{for } N^{\frac{4}{3}} \leq B \leq N^3 \end{cases}$$

where \bar{r}_i are radii of supports of ρ_i^{TF} of separate atoms with $N_i = Z_i$; I recall that $\bar{r}_i \asymp B^{-\frac{1}{4}}$ for $B \leq N^{\frac{4}{3}}$ and $\bar{r}_i \asymp B^{-\frac{2}{5}} N^{\frac{1}{5}}$ for $N^{\frac{4}{3}} \leq B \leq N^3$.

(ii) Further, for $N < Z$ estimate (15) holds with $L = 1$.

REMARK 9. (i) For estimate (15) remark 5(iii) holds and for $B \leq N$ estimate $|x_i - x_j| \geq \min(N^{-\frac{5}{21} + \delta}, \bar{r}_i + \bar{r}_j - CB^{-\frac{5}{16}} B^\delta N^{-\delta})$ holds.

(ii) All the estimates of theorems 6, 7 remain true if $M \geq 2$ and $\mathcal{R}' + \mathcal{R}'' \leq \mathcal{R}$ for given $Z, Z - N$ and B . The estimates in the opposite case are left to the reader.

(iii) Unfortunately, we don't prove that molecules exist, i.e. that (22) really holds for appropriate N . I am not aware of any rigorous result of this type in frames of our models. \diamond

6. Case $B \geq N^3$. Let me explain informally why condition $B \ll N^3$ is so important. First of all, for $B = 0$ the pressure $P(\lambda)$ is the Weyl expression for the density of energy of the non-interacting particles, occupying subsequent energy levels. The same is true for $B \neq 0$, but now one needs first to separate variables (x_1, x_2) : electron is described now by one-dimensional Schrödinger operator. The main contribution for $B \geq N^{\frac{4}{3}}$ is given by the zone $\{|x - x_i| \asymp r_S\}$ (or their union). In this zone the length of the electron is $\asymp r_S$ and the uncertainty in x_3 -momentum is $\sqrt{W} = \sqrt{N r_S^{-1}}$. To have Weyl formula valid one needs to keep their product $\sqrt{N r_S}$ well above 1 (uncertainty principle) and since $r_S = B^{-\frac{2}{5}} N^{\frac{1}{5}}$ as $B \geq N^{\frac{4}{3}}$, this gives us exactly condition $B \ll N^3$.

The next case $B \geq CN^3$ I am going to treat in the nearest future. Right now I am just referring to [LSY1]. One thing is clear: the answer involves an auxiliary one-dimensional Schrödinger operator on magnetic line depending on (x_2, x_3) (I recall that $\mathbf{B} = B\mathbf{k}$) and that this answer is simpler for $B \gg N^3$ and it is really complicated in the transition zone.

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