Bound on the Ionization Energy of Large Atoms

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Abstract

We present a simple argument which gives a bound on the ionization energy of large atoms and at the same time proves the bound on the excess charge of Fefferman and Seco[1].

1 Introduction

An atom of nuclear charge Z with N electrons is described by the Schrödinger operator

$$H_{N,Z} = \sum_{i}^{N}(-\Delta_i - \frac{Z}{|x_i|}) + \sum_{1\leq i<j\leq N} \frac{1}{|x_i - x_j|} \tag{1}$$

acting on the antisymmetric space $\mathcal{H}_F = \wedge_{i=1}^{N}(L^2(I\!R^3) \otimes \mathbb{C}^2)$.

The **Energy** of the atom is

$$E(N,Z) = \inf\ \mathrm{spec}_{\mathcal{H}_F} H_{N,Z} \tag{2}$$

and the **Ionization Energy** is

$$I(N,Z) = E(N-1,Z) - E(N,Z). \tag{3}$$

It is well known that there is a critical number of electrons $Z \leq N_c(Z) < 2Z+1$ (see [9] and [4]) such that

$$\begin{aligned} I(N,Z) > 0 \quad &\text{if} \quad N \leq N_c \\ I(N,Z) = 0 \quad &\text{if} \quad N > N_c. \end{aligned}$$

The **Excess Charge** is

$$Q_c(Z) = N_c(Z) - Z. \tag{4}$$

For $N \leq N_c$ the operator $H_{N,Z}$ has a ground state $\psi_{N,Z} \in \mathcal{H}_F$.

We define the **Radius**$R(N,Z)$ of the atom by

$$\int_{|x| \leq R(N,Z)} \rho_{N,Z} dx = N - 1, \tag{5}$$

where $\rho_{N,Z}$ is the **Density**

$$\rho_{N,Z}(x) = N \sum_{\sigma=1,2} \int |\psi_{N,Z}(x,\sigma; x_2,\sigma_2; \ldots; x_N,\sigma_N)|^2 d(x_2,\sigma_2)\ldots d(x_N,\sigma_N),$$

x_i are the space variables and σ_i the spin variables, $\int d(x,\sigma) = \sum_\sigma \int dx$. (Throughout most of the paper explicit mentioning of the spin variables will be omitted). Outside $R(N,Z)$ there is an average of one electron.

It is expected that as $Z \to \infty$

$$Q_c(Z),\ I(Z,Z),\ R(Z,Z)\ = O(1), \tag{6}$$

this is part of a much stronger conjecture saying that as $Z \to \infty$ the atomic structure shows a universal behavior, which is to say that the quantities in (6) actually converge to non-zero values as $Z \to \infty$.

In Thomas-Fermi theory this universality has been known for some time (see [5]). In the present paper we will indeed compare with TF theory. In the Thomas-Fermi-von Weizsäcker theory universality was recently proved in [8].

It is not hard to prove that

$$Q_c(Z) \leq CZ, \ I(Z,Z) \leq CZ^{4/3} \text{ and } \ R(Z,Z) \geq CZ^{-1/3}. \tag{7}$$

In [6] it was proved that $Q_c(Z) = o(Z)$. This has recently been improved in [1] (an announcement was made in [2]) to $Q_c(Z) \leq CZ^{(1-\varepsilon)}$ with $\varepsilon = 9/56$.

Using the key estimate of [1], i.e. (13) below, we give a simple argument proving

Theorem 1 *For $Z \leq N \leq N_c$*

$$I(N,Z) \leq C_1 Z^{(4/3)(1-\varepsilon)} - C_2(N-Z)Z^{(1/3)(1-\varepsilon)}. \tag{8}$$

We get as an immediate consequence

Corollary 2

$$Q_c(Z) \leq CZ^{(1-\varepsilon)}$$

and for $N \geq Z$,

$$I(N,Z) \leq CZ^{(4/3)(1-\varepsilon)}.$$

As a very easy consequence of the proof of Theorem 1 we also find (see Lemma 7)

Theorem 3 *For $N \geq Z$*

$$R(N,Z) \geq CZ^{-(1/3)(1-\varepsilon)}. \tag{9}$$

We prove Theorem 1 by first proving a general estimate on $I(N, Z)$ which for an arbitrary radius R bounds I in terms of quantities we call the Screening Charge at radius R, the Excess Charge at radius R, and the 2-point correlation outside R. This general bound is given in Section 2. In Section 3 we show how to estimate the above quantities using the key estimate from [1].

Our method emphasizes the importance of controlling the 2-point correlation function

$$\rho^{(2)}(x,y) = \\ N(N-1) \sum_{\sigma_1,\sigma_2} \int |\psi(x,\sigma_1; y,\sigma_2; \ldots; x_N,\sigma_N)|^2 d(x_3,\sigma_3)\ldots d(x_N,\sigma_N),$$

(we will often omit the subscripts N, Z). In fact the key step is to estimate the truncated correlation function

$$\rho^{(2)}(x,y) - \rho(x)\rho(y),$$

this is done in Section 3 Lemma 5.

2 General Argument

Given δ, choose $\theta_0 \in C^\infty(I\!R_+)$ with $0 \le \theta_0 \le 1$, and $\theta_0(t) = 0$ if $t \le 1-\delta$, $\theta_0(t) = 1$ if $t \ge 1$.

For all R, let $\theta_R^\pm : I\!R^3 \mapsto I\!R^3$ be given by

$$\theta_R^+(x) = \theta_0(|x|/R) \quad \text{and} \quad \theta_R^-(x) = (1-\theta_R^+(x)^2)^{1/2}.$$

We define

The Excess Charge at Radius R,

$$Q_\delta(R) = \int \rho(x)\theta_R^+(x)^2 dx,$$

The Screening Charge at Radius R,

$$\nu_\delta(R) = Z - \frac{1}{Q_\delta(R)}\int \rho^{(2)}(x,y)\frac{|x|}{|x-y|}\theta_R^-(y)^2\theta_R^+(x)^2 dxdy,$$

The Normalized 2-point Correlation Outside R,

$$\mathcal{C}_\delta(R) = \frac{1}{Q_\delta(R)^2}\int \rho^{(2)}(x,y)\theta_R^+(x)^2\theta_R^+(y)^2 dxdy.$$

We will prove an upper bound to the ionization energy in terms of these quantities by using a very simple trick which in fact goes back to Benguria (see [5]) and was used in [4] to prove $N_c < 2Z+1$. The idea here is to use the trick on the outside problem ($|x| > R$). The same method was used in [8].

Theorem 4 *For all $\delta > 0$ and $R > 0$*

$$I \le \left[\nu_\delta(R) - \tfrac{1}{2}\mathcal{C}_\delta(R)Q_\delta(R)\right]R^{-1} + X_\delta, \tag{10}$$

where the error term is bounded by

$$X_\delta \le c_\delta R^{-2}\frac{Q_\delta(R(1-\delta))}{Q_\delta(R)}.$$

Proof. From the IMS formula we find

$$\begin{aligned}
&E_{N,Z}\int \rho(x)|x|\theta_R^+(x)^2 dx \\
&= \sum_i \left\langle \psi_{N,Z}\left|\theta_R^+(x_i)^2|x_i|H_{N,Z}\right|\psi_{N,Z}\right\rangle \\
&= \sum_i \left\langle \psi_{N,Z}\left|\theta_R^+(x_i)|x_i|^{1/2}H_{N,Z}\theta_R^+(x_i)|x_i|^{1/2}\right|\psi_{N,Z}\right\rangle \\
&\quad - \sum_i \left\langle \psi_{N,Z}\left||\nabla_i(\theta_R^+(x_i)|x_i|^{1/2})|^2\right|\psi_{N,Z}\right\rangle \\
&\ge E_{N-1,Z}\int \rho(x)|x|\theta_R^+(x)^2 dx \\
&\quad + \sum_i \left\langle \psi_{N,Z}\left|\theta_R^+(x_i)^2|x_i|\Big(-\frac{Z}{|x_i|} + \sum_{j,j\ne i}\frac{1}{|x_j-x_i|}\Big)\right|\psi_{N,Z}\right\rangle \\
&\quad + \sum_i \int \Big(\nabla_i(\theta_R^+(x_i)^2|x_i|^{1/2}\psi)\Big)^2 - \psi^2|\nabla_i(\theta_R^+(x_i)|x_i|^{1/2})|^2 dx.
\end{aligned}$$

Using $|\nabla(\theta_R^+(x)|x|^{1/2}| \leq cR^{-1}$ we rewrite this inequality as

$$\begin{aligned} -RIQ_\delta(R) &\geq -ZQ_\delta(R) + \int \rho^{(2)}(x,y)\frac{|x|}{|x-y|}\theta_R^+(x)^2 dxdy \\ &\quad - c_\delta Q_\delta(R(1-\delta))R^{-1}. \end{aligned}$$

In [4] the error term (the last term above) could be ignored by use of the uncertainty principle: $\int(\nabla u)^2 \geq (1/4)\int u^2/|x|^2$. Here the uncertainty principle can be used to improve c_δ, but this is not necessary.

The trick is now to symmetrize and use the triangle inequality

$$\begin{aligned} RIQ_\delta(R) &\leq \nu_\delta(R)Q_\delta(R) - \int \rho^{(2)}(x,y)\frac{|x|}{|x-y|}\theta_R^+(x)^2\theta_R^+(y)^2 dxdy \\ &\quad + c_\delta Q_\delta(R(1-\delta))R^{-1} \\ &= \nu_\delta(R)Q_\delta(R) - \tfrac{1}{2}\int \rho^{(2)}(x,y)\frac{|x|+|y|}{|x-y|}\theta_R^+(x)^2\theta_R^+(y)^2 dxdy \\ &\quad + c_\delta Q_\delta(R(1-\delta))R^{-1} \\ &\leq \nu_\delta(R)Q_\delta(R) - \tfrac{1}{2}\mathcal{C}_\delta(R)Q_\delta(R)^2 + c_\delta Q_\delta(R(1-\delta))R^{-1}. \end{aligned}$$

∎

3 Estimates

We proceed as in [1]. Choose $\varphi_1 \in C_0^\infty(I\!R^3)$ radially symmetric, positive and with $\int \varphi_1 = 1$. Let

$$\varphi(x) = \varphi_Z(x) = Z^2\varphi_1(Z^{2/3}x), \tag{11}$$

then $\varphi = 1$. With ρ_{TF} the Thomas-Fermi density for a neutral atom with nuclear charge Z(see [5] for a review of TF theory), we define a function $K_N : I\!R^N \to I\!R_+$ by

$$K_N(x_1,\ldots,x_N) = D\Big(\sum_{i=1}^N \varphi(\cdot - x_i) - \rho_{TF}\,,\, \sum_{i=1}^N \varphi(\cdot - x_i) - \rho_{TF}\Big), \tag{12}$$

where

$$D(f,g) = \tfrac{1}{2}\int f(x)|x-y|^{-1}g(x)dxdy.$$

It follows from the proof of the Scott correction [1] (see [3] and [7]) that for all N with $Z - \text{const} \leq N$ we have an operator inequality

$$H_{N,Z} \geq E(N,Z) + K_N(x_1,\ldots,x_N) - C_S Z^{7/3-b}, \tag{13}$$

for some b, $1/3 < b \leq 2/3$.

Our main estimate is

Lemma 5 *Given $\theta \in C^\infty(I\!R^3)$ with $0 \leq \theta \leq 1$, $\operatorname{supp}\theta \subset \{|x| \geq R\}$ and $|\nabla\theta| < c_1 R^{-1}$ and $\chi \in C^\infty(I\!R^3 \times I\!R^3)$ with $0 \leq \chi$ and $\chi_x = \chi(x,\cdot)$ compactly supported. Then for all N with $Z \leq N \leq N_c(Z)$*

$$\begin{aligned}
&\left|\int[\rho^{(2)}(x,y) - \rho_{TF}(y)\rho(x)]\theta(x)^2\chi(x,y)dxdy\right| \\
&\leq C_1 \sup_x \|\nabla_y\chi_x\|_{L^2(I\!R^3)} \left\{ (Z^{(7/3-b)} + ZR^{-1})\int \rho(x)\theta(x)^2dx + ZR^{-2} \right\}^{1/2} \\
&\qquad\qquad \times \left\{ \int \rho(x)\theta(x)^2 dx \right\}^{1/2} \\
&\quad + C_2 Z^{1/3}\|\nabla_y\chi\|_{L^\infty}\int\rho(x)\theta(x)^2dx,
\end{aligned} \tag{14}$$

where ρ and $\rho^{(2)}$ are the ground state density and correlation function. C_1, C_2 depend only on C_s and φ_1.

Remark. The reason for the rather peculiar cutoff in (14) will be clear in Lemma 8 below.

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[1] Without using the Scott correction, but only the fact (see [5]) that the TF-energy approximates the quantum energy, one can still prove Theorem 1, but the value for ε will be much smaller.

Proof. Define $N_x\colon \mathbb{R}^{N-1} \to \mathbb{R}_+$ and $N_x^{TF} \in \mathbb{R}_+$ by

$$
\begin{aligned}
N_x(x_2,\ldots,x_N) &= \sum_{i=2}^{N} \chi_x * \varphi(x_i) && (15)\\
N_x^{TF} &= \int \rho_{TF}(y)\chi_x(y)dy. && (16)
\end{aligned}
$$

Then

$$
\begin{aligned}
&|\int [\rho^{(2)}(x,y) - \rho_{TF}(y)\rho(x)]\theta(x)^2\chi(x,y)dxdy| \\
&\quad\le \int \rho^{(2)}(x,y)|\chi_x * \varphi(y) - \chi_x(y)|\theta(x)^2 dxdy \\
&\qquad + N\left|\int |\psi(x,x_2,\ldots,x_N)|^2\Big(N_x(x_2,\ldots,x_N) - N_x^{TF}\Big)dx_2\ldots dx_N\theta(x)^2dx\right| \\
&\quad\le C_2 Z^{1/3}\|\nabla_y\chi_x\|_{L^\infty}\int \rho(x)\theta(x)^2dx \\
&\qquad + \left\{ N|\int |\psi(x,x_2,\ldots,x_N)|^2|N_x - N_x^{TF}|^2\theta(x)^2 dxdx_2\ldots dx_N \right\}^{1/2} \\
&\qquad\qquad \times\left\{\int \rho(x)\theta(x)^2dx\right\}^{1/2}, \qquad (17)
\end{aligned}
$$

where we have used Cauchy-Schwarz inequality.

Since

$$
N_x(x_2,\ldots,x_N) - N_x^{TF} = \int \Big(\sum_{i=2}^{N}\varphi(y-x_i) - \rho_{TF}(y)\Big)\chi(x,y)dy,
$$

we get again from Cauchy-Schawarz

$$
\begin{aligned}
|N_x - N_x^{TF}|^2 &\le \int |\hat{\chi}_x(\xi)|^2|\xi|^2 d\xi \int \Big|\Big(\sum_{i=2}^{N}\varphi(y-x_i) - \rho_{TF}(y)\Big)\hat{}(\xi)\Big|^2|\xi|^{-2}d\xi \\
&\le C\|\nabla\chi_x\|^2_{L^2(\mathbb{R}^3)}K_{N-1}(x_2,\ldots,x_N), && (18)
\end{aligned}
$$

where $\hat{}$ denotes Fourier transform. From (13) we find using IMS,

$$
E_{N-1,Z}\int \rho(x)\theta(x)^2dx \ge E_{N,Z}\int \rho(x)\theta(x)^2dx
$$

$$\begin{aligned}
&\geq \sum_{i=1}^{N}\Big\{ <\psi \mid \theta(x_i) H_{N-1,Z}\theta(x_i) \mid \psi> - <\psi \mid (\nabla_i \theta(x_i))^2 \mid \psi> \\
&\quad + <\psi \mid -\theta(x_i)^2 \frac{Z}{|x_i|} + \sum_{j,j\neq i} \frac{\theta(x_i)^2}{|x_i - x_j|} \mid \psi> \Big\} \\
&\geq E_{N-1,Z} \int \rho(x)\theta(x)^2 dx + N \int |\psi|^2 K_{N-1}\theta(x)^2 dx dx_2 \ldots dx_N \\
&\quad + C_S Z^{(7/3-b)} \int \rho(x)\theta(x)^2 dx - cNR^{-2} - cR^{-1} \int \rho(x)\theta(x)^2 dx.
\end{aligned}$$

Thus

$$\begin{aligned}
&N \int |\psi|^2 \|\nabla \chi_x\|_{L^2}^2 K_{N-1}\theta(x)^2 dx dx_2 \ldots dx_N \\
&\leq C \sup_x \|\nabla \chi_x\|_{L^2}^2 \left((Z^{(7/3-b)} + R^{-1}Z) \int \rho\theta(x)^2 dx + ZR^{-2} \right). \qquad (19)
\end{aligned}$$

Putting together (17),(18) and (19) gives (14). ∎

A simplification of the above proof gives

Lemma 6 *With the notation of Section 2*

$$\left| \int (\rho(x) - \rho_{TF}(x))\theta_R^-(x)^2\, dx \right| \leq C(R^{1/2} Z^{(7/6-b/2)} + R^{-1} Z^{1/3}) \qquad (20)$$

Estimate on Q_δ

Using that $\rho_{TF}^{(Z)}(x) = Z^2 \rho_{TF}(Z^{1/3}x)$ and

$$|x| \geq 1 \quad \Rightarrow \quad C_-|x|^{-6} \leq \rho_{TF}^{(1)}(x) \leq C_+|x|^{-6},$$

gives

$$|x| \geq Z^{-1/3} \quad \Rightarrow \quad C_-|x|^{-6} \leq \rho_{TF}(x) \leq C_+|x|^{-6}. \qquad (21)$$

Furthermore $\int \rho_{TF}\, dx = Z$. Thus

$$\begin{aligned}
Q_\delta(R) &= N - \int \rho(x)\theta_R^-(x)^2 dx \\
&= N - \int \rho_{TF}(x)\theta_R^-(x)^2 dx - \int (\rho(x) - \rho_{TF}(x))\theta_R^-(x)^2 dx \\
&= N - Z + \int \rho_{TF}(x)\theta_R^+(x)^2 dx - \int (\rho(x) - \rho_{TF}(x))\theta_R^-(x)^2 dx.
\end{aligned}$$

Choosing

$$R = R_Z = \alpha Z^{-1/3+b/7} \tag{22}$$

gives

$$\alpha^{-3}\tilde{C}_- R^{-3} \le \int \rho_{TF}(x)^2 dx \le \alpha^{-3}\tilde{C}_+ R^{-3} \tag{23}$$

and from Lemma 6 we then find

$$(\tilde{C}_-\alpha^{-3} - C\alpha^{1/2} Z^{(1-3b/7)}) \le Q_\delta(R_Z) - (N-Z) \le (\tilde{C}_+\alpha^{-3} + C\alpha^{1/2} Z^{(1-3b/7)}).$$

Choosing α appropriately we have proved

Lemma 7 *With $\varepsilon = 3b/7$ there exists $\alpha > 0$ such that for $R_Z = \alpha Z^{-(1/3)(1-\varepsilon)}$*

$$N - Z + C_- Z^{(1-\varepsilon)} \le Q_\delta \le N - Z + C_+ Z^{(1-\varepsilon)}. \tag{24}$$

¿From the lower bound in (24) we get the result in Theorem 3 with $\varepsilon = 3b/7$.

Estimate on ν_δ

Lemma 8 *For ε and R_Z as in Lemma 7*

$$\nu_\delta(R_Z) \le C_\delta Z^{(1-\varepsilon)} \tag{25}$$

Proof. If in (14) we choose $\theta(x) = \theta_R^+(x)$ and

$$\chi(x,y) = \frac{|x|}{|x-y|}\theta^-_{R(1-2\delta)}(y)^2 \theta_R^+(x)^2$$

then, on supp χ we have $|x-y| \ge \delta R$, and

$$\chi(x,y)\theta(x)^2 = \frac{|x|}{|x-y|}\theta^-_{R(1-2\delta)}(y)^2 \theta_R^+(x)^2.$$

Furthermore it is easy to see that

$$\sup_x \|\nabla_y \chi_x\|_{L^2(I\!R^3)} \le c_\delta R^{1/2} \quad \text{and} \quad \|\nabla\chi\|_{L^\infty} \le c_\delta R^{-1}.$$

From (14) we obtain

$$\begin{aligned}&\left|\int \left[\rho^{(2)}(x,y) - \rho_{TF}(y)\rho(x)\right] \theta_R^+(x)^2 \frac{|x|}{|x-y|}\theta^-_{R(1-2\delta)}(y)^2 dxdy\right| \\ \leq\ & \tilde{C}_\delta \left(Q_\delta(R)Z^{(1-\varepsilon)}Q_\delta(R)^{1/2}Z^{(2/3-\varepsilon/6)} + Q_\delta(R)Z^{(2/3-\varepsilon/3)}\right) \\ \leq\ & \tilde{C}_\delta \left(Q_\delta(R)Z^{(1-\varepsilon)} + Q_\delta(R)^{1/2}Z^{(2/3-\varepsilon/6)}\right),\end{aligned}$$

where we have used that $1/3 < b \leq 2/3$ implies $1/7 \leq \varepsilon \leq 2/7$. From Lemma 7 we can now conclude

$$\begin{aligned}\nu_\delta(R_Z) &\leq Z - \frac{1}{Q_\delta(R)}\rho_{TF}(y)\rho(x)\theta_R^+(x)^2\frac{|x|}{|x-y|}\theta^-_{R(1-2\delta)}(y)^2 dxdy \\ &\quad + \tilde{C}_\delta(Z^{(1-\varepsilon)} \\ &\leq Z - \int \rho_{TF}(y)\theta^-_{R(1-2\delta)}(y)^2 dy + \tilde{C}_\delta Z^{(1-\varepsilon)},\end{aligned}$$

the last inequality follows since $|x-y|^{-1}$ is the harmonic potential, $\theta^-_{R(1-2\delta)}$ and ρ_{TF} are spherically symmetric and $\theta^-_{R(1-2\delta)}$ is supported disjointly from θ_R^+. Recalling (21) we get

$$\begin{aligned}\nu_\delta(R_Z) &\leq \int \rho_{TF}(y)\theta^+_{R(1-2\delta)}(y)^2 dy + \tilde{C}_\delta Z^{(1-\varepsilon)} \\ &\leq C_\delta Z^{1-\varepsilon}.\end{aligned}$$

∎

Estimate on $\mathcal{C}_\delta$

Lemma 9 *For R_Z as in Lemma 7*

$$\mathcal{C}_\delta(R_Z) \geq C_\delta > 0 \tag{26}$$

Proof. This can be done without the use of Lemma 5. Indeed notice that the inequality $\langle F^2\rangle - \langle F\rangle^2 \geq 0$ used on $F = \sum_{i=1}^N f(x_i)$ implies

$$\int \rho^{(2)}(x,y)f(x)f(y)dxdy \geq \left(\int \rho(x)f(x)dx\right)^2 - \int \rho(x)f(x)^2 dx.$$

Hence since $\theta_R^+ \leq 1$

$$\mathcal{C}_\delta \geq \frac{1}{Q_\delta(R)^2}\Big(Q_\delta(R)^2 - Q_\delta(R)\Big)$$

and the result follows from Lemma 7 ∎

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