Расчеты физических и химических характеристик элементов 7-го периода и их гомологов

Илья И. Тупицын

Члены атомной группы (СПбГУ)

Михаил Кайгородов, Алексей Малышев, Юрий Кожедуб, Дмитрий Глазов и Владимир Шабаев

Нам помогали и консультировали

Ephraim Eliav	Tel Aviv University, Israel
Леонид Скрипников	СПбГУ-ПИЯФ
Александр Олейниченко	МГУ-ПИЯФ

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Outline of the Talk

- Introduction
- Electron Localization Function
- Shannon Entropy
- Numerical results
- Summary and Outlook

Introduction

Physical and chemical characteristics of atoms that we considered in this work

- One-particle density – electron density distribution
- Electron Localization Function (ELF)

  The electron localization function is a measure of the probability of finding an electron in the neighborhood of another electron with the same spin.
- Shannon entropy

  Shannon entropy of a discrete random variable X is a measure of the amount of uncertainty associated with the value of X.
- Root-mean-square radii and standard deviations (STD) of atomic valence shells
- Ionization potentials and electron affinities of atoms

  The ionization potentials and affinity energies of the elements of the 7th period were calculated earlier. See, for example, papers [1-3].

[1] *E. Eliav et. al., Nucl. Phys. A 944, p. 518 (2015).*

[2] *E. Eliav et al., PRA 53, 3926 (1996).*

[3] *B. G. C. Lackenby et al., Phys. Rev. A 98, 042512 (2018).*

In our work, we used two independent theoretical calculation methods

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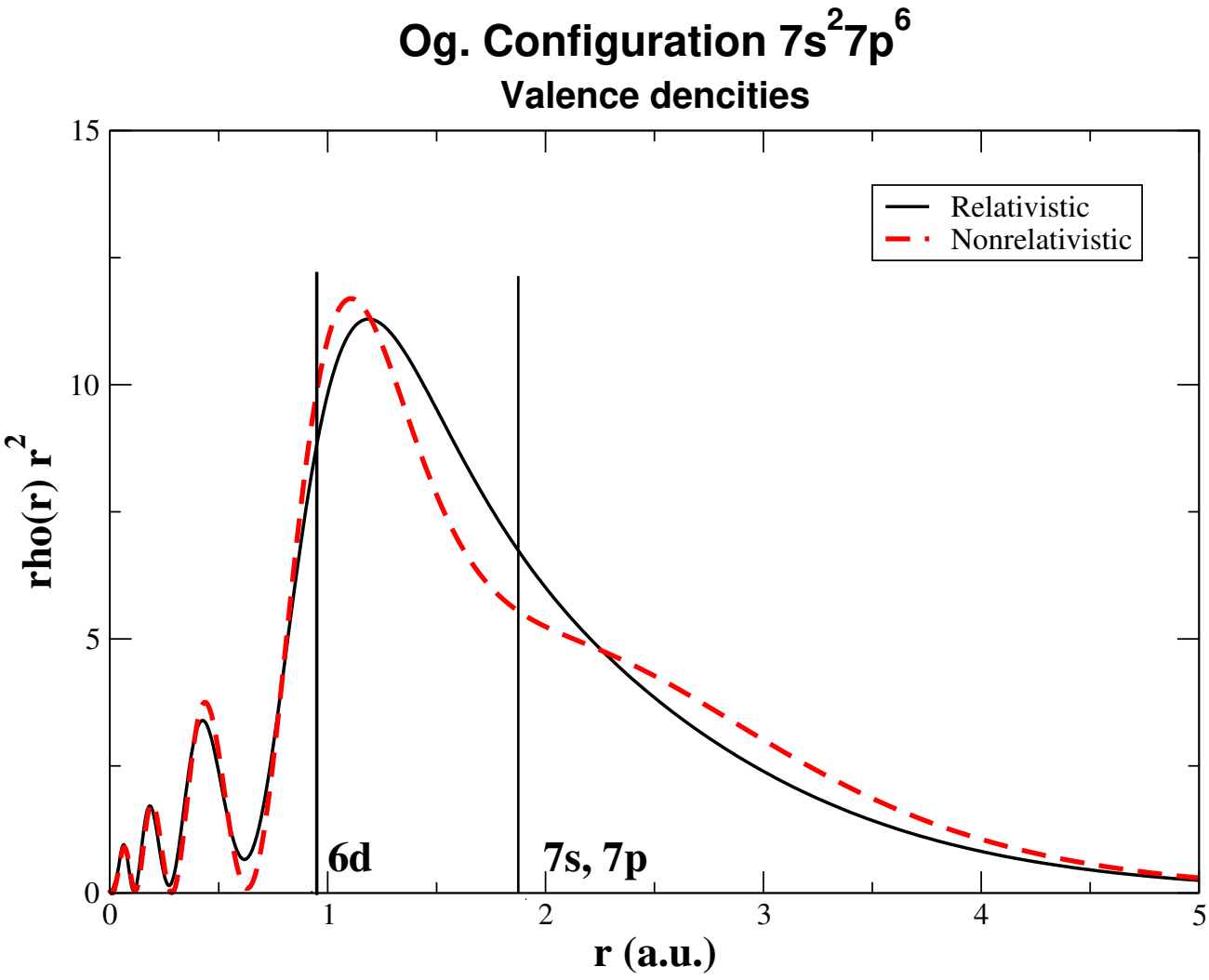
- Configuration Ineraction Dirac-Fock-Sturm method (CI-DFS)

  At the first step, to obtain the one-electron wave functions for the occupied atomic shells, we use the Dirac-Fock method. Then the DFS orbitals are obtained by solving the DFS equations for the vacant shells. At the last step, the relativistic CI+MBPT method is used to obtain the many-electron wave functions and the total energies.

- Fock Space Coupled-Cluster method (FSCC)

  DIRAC, a relativistic ab initio electronic structure program, Release DIRAC21 (2021), http://www.diracprogram.org

  FSCC method, in contrast to the one-configuration coupled-cluster method is capable of providing not only the ground-state energy of an N-electron system, but also an important fraction of system's excitation spectrum, including ionization potentials, electron affinities, etc.


Og. Configuration 7s2 7p6
Valence dencities
Relativistic
Nonrelativistic
rho(r) r2
r (a.u.)
6d
7s, 7p
0
5
10
15
0
1
2
3
4
5

Electron Localization Function (ELF)

The Electronic localization function is defined by [1]

$$\mathrm{ELF}(\boldsymbol{r}) = \left(1 + \left[\frac{D(\boldsymbol{r})}{D_0(\boldsymbol{r})}\right]^2\right)^{-1}, \tag{1}$$

where

$$D(\boldsymbol{r}) = \frac{1}{2}\left[\tau - \frac{1}{4}\frac{|\boldsymbol{\nabla}\rho(\boldsymbol{r})|^2}{\rho(\boldsymbol{r})}\right], \qquad \text{and} \qquad \tau = \sum_{i,\sigma} |\boldsymbol{\nabla}\varphi_{i\sigma}(\boldsymbol{r})|^2, \tag{2}$$

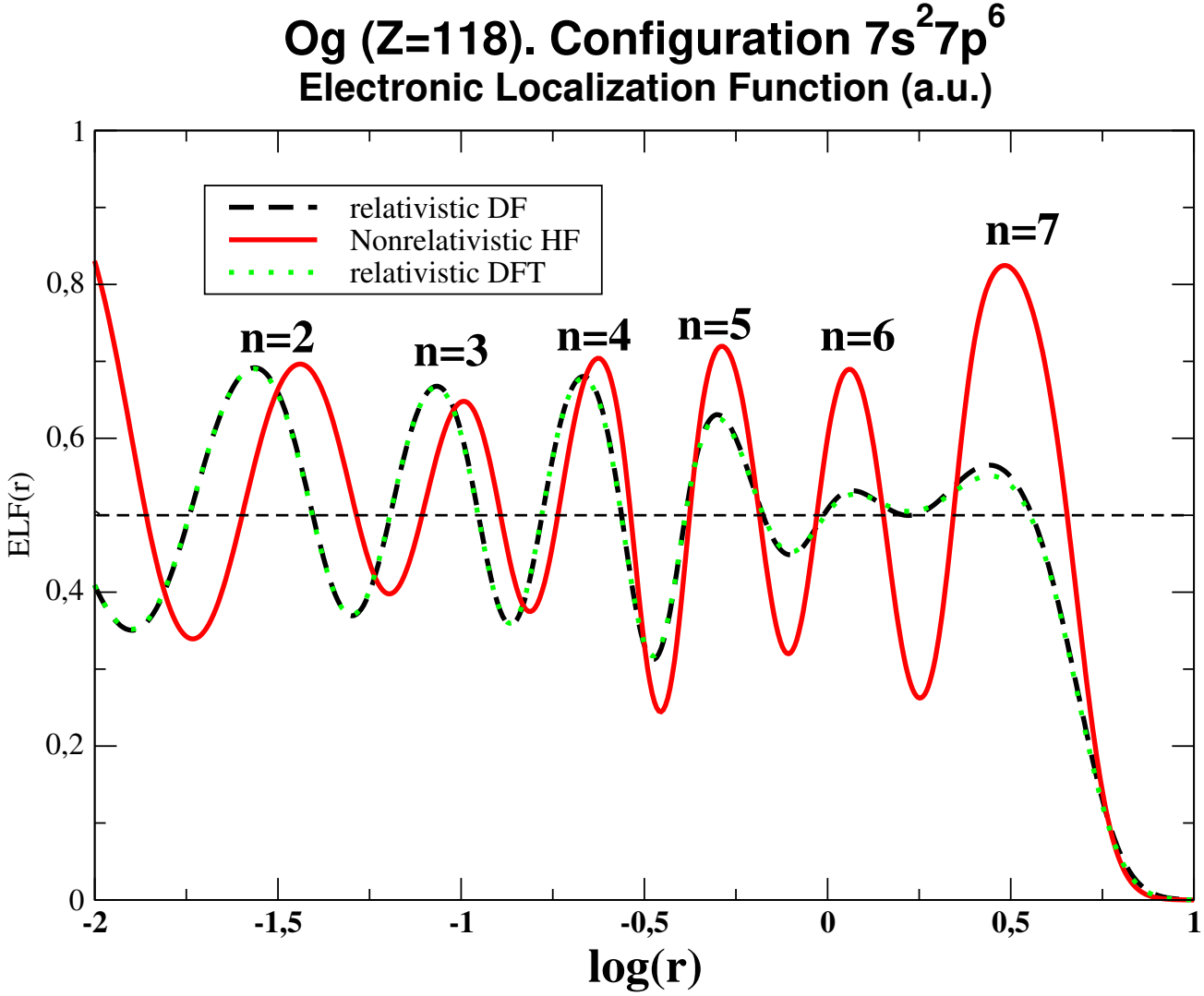
Here ρ is total density and τ the kinetic energy density.

$D_0(\boldsymbol{r})$ corresponds to a uniform electron gas (Thomas-Fermi) kinetic energy density

$$D_0(\boldsymbol{r}) = \frac{3}{10}(3\,\pi^2)^{2/3}\rho^{5/3}(\boldsymbol{r})\,. \tag{3}$$

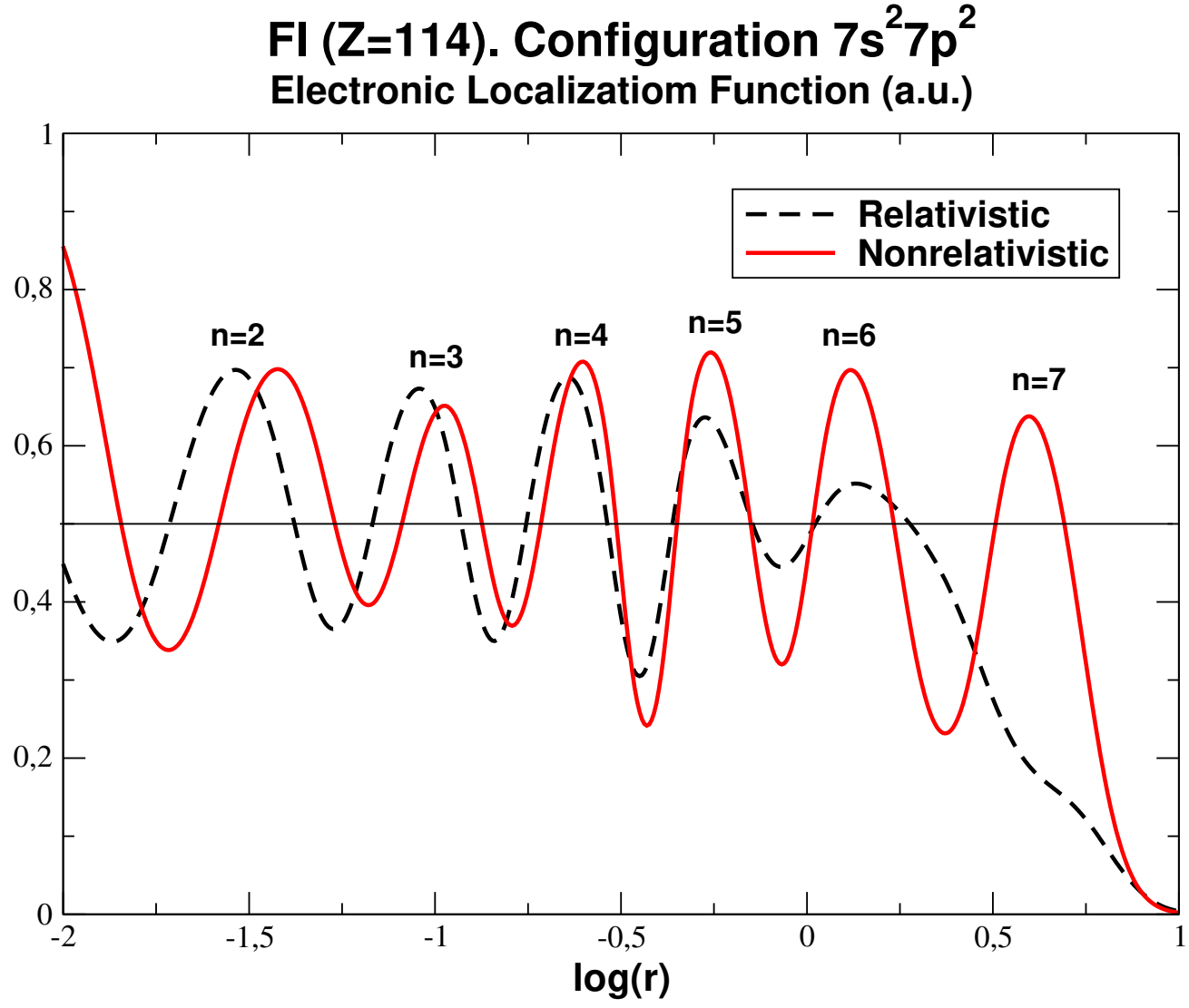
The ELF values lie by definition between zero and one. Small values are typical for the region between two electron shells. In a homogeneous electron gas, ELF = 0.5.

[1] *A. D. Becke and K. E. Edgecombe, J. Chem. Phys. v.92, 5397 (1990).*



"Spin-orbit splitting in the 7p electronic shell becomes so large (∼10 eV) that Og is expected to show uniform-gas-like behavior in the valence region" [1].

[1] *Jerabek et al., PRL 120, 053001 (2018).*


Fl (Z=114). Configuration 7s2 7p2
Electronic Localizatiom Function (a.u.)
Relativistic
Nonrelativistic
n=2
n=3
n=4
n=5
n=6
n=7
1
0,8
0,6
0,4
0,2
0
-2
-1,5
-1
-0,5
0
0,5
1
log(r)

Shannon entropy

According to the formula of K. Shannon [1], the amount of information is defined as:

$$S = -\sum_{i=1}^{N} p_i \, \ln p_i \tag{4}$$

where N is the number of random events, p_i is the probability of the i-th event and

$$\sum_{i=1}^{N} p_i = 1 \, . \tag{5}$$

The minimum value of S is reached for the deterministic event when one of the probabilities of p_i is 1, and the rest are zero. In this case, $S = S_{\min} = 0$.

The maximum value of S is reached for an equally probable distribution $p_i = 1/N$. Then, $S_{\max} = \ln(N)$

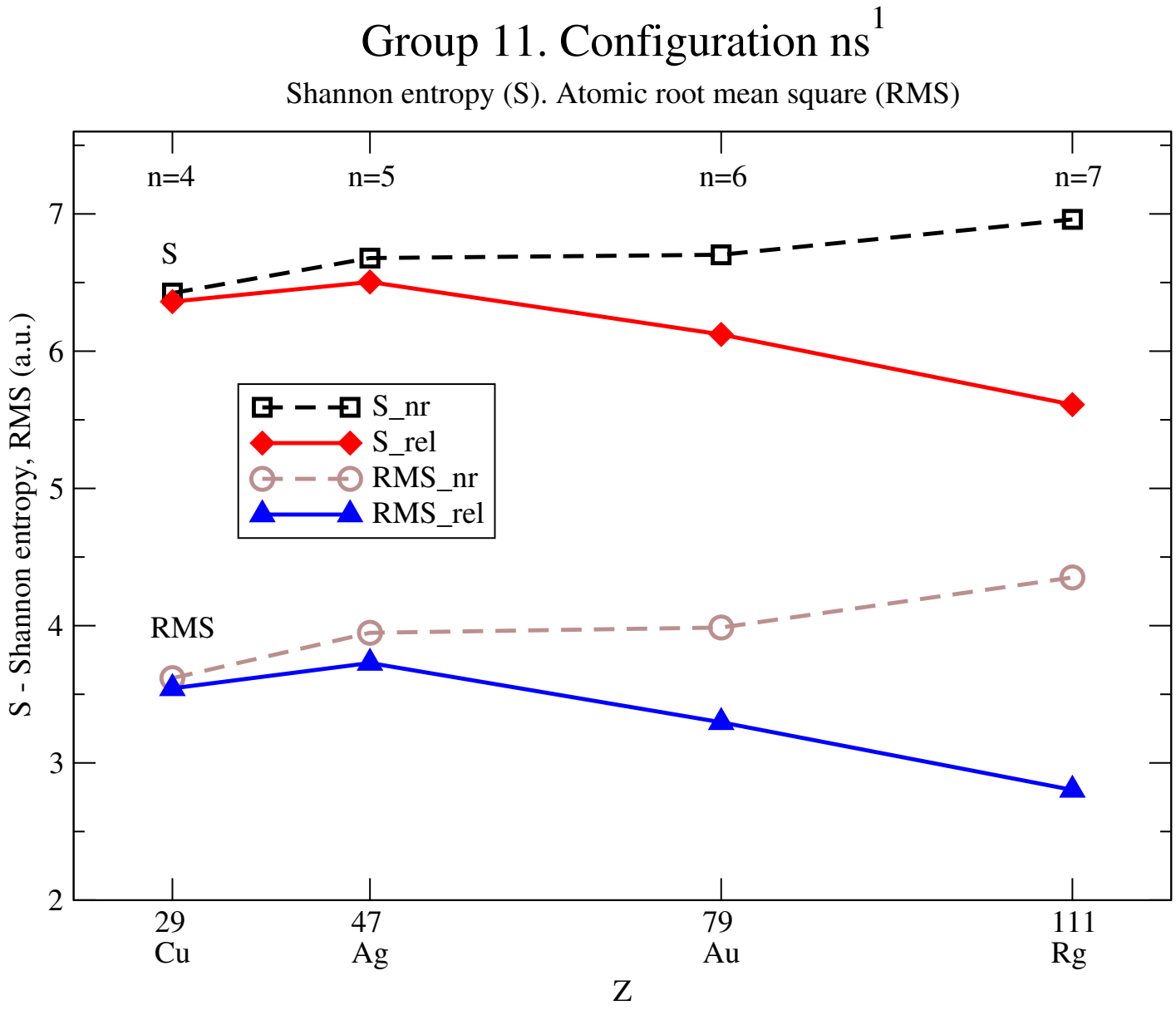
$$0 \leq S \leq \ln(N) \tag{6}$$

For continuous distribution

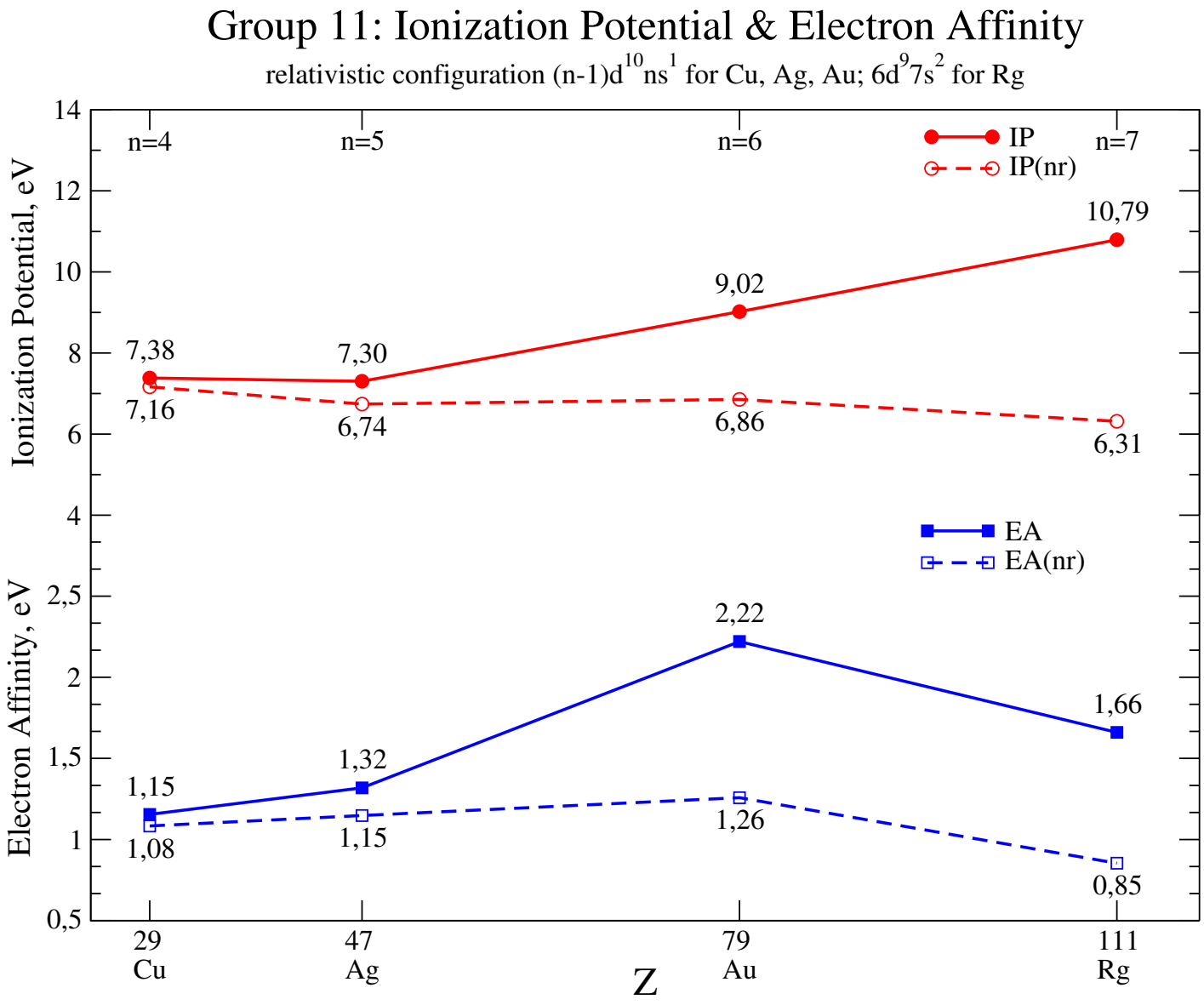
$$S = -\int d\boldsymbol{r} \, \ln(\rho(\boldsymbol{r})) \, \rho(\boldsymbol{r}) \, , \qquad \int d\boldsymbol{r} \, \rho(\boldsymbol{r}) = 1 \, . \tag{7}$$

[1] *C.E. Shannon, Bell Syst. Tech. J. 27, 379; 623 (1948).*

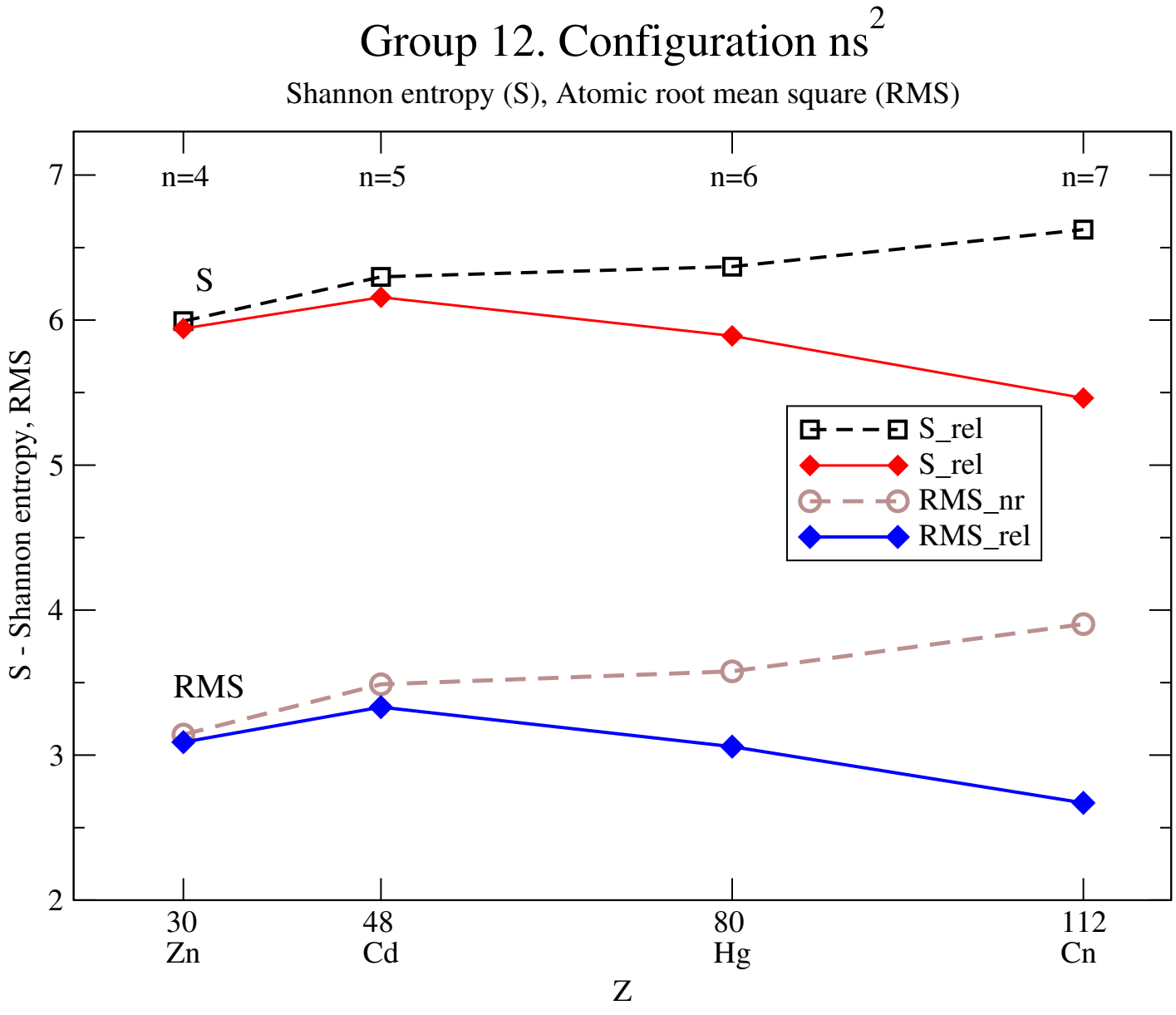
11th group of elements



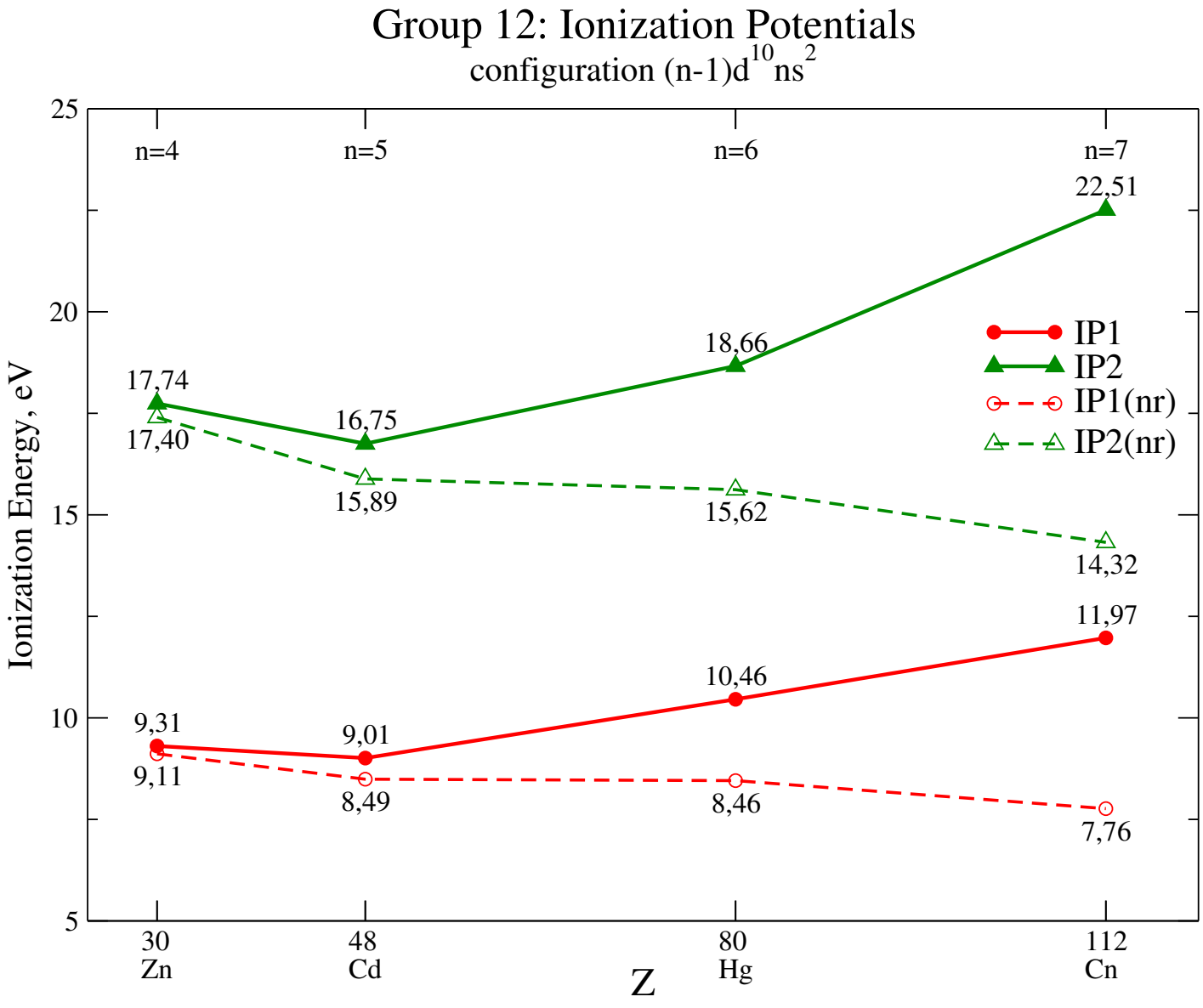
11th group of elements



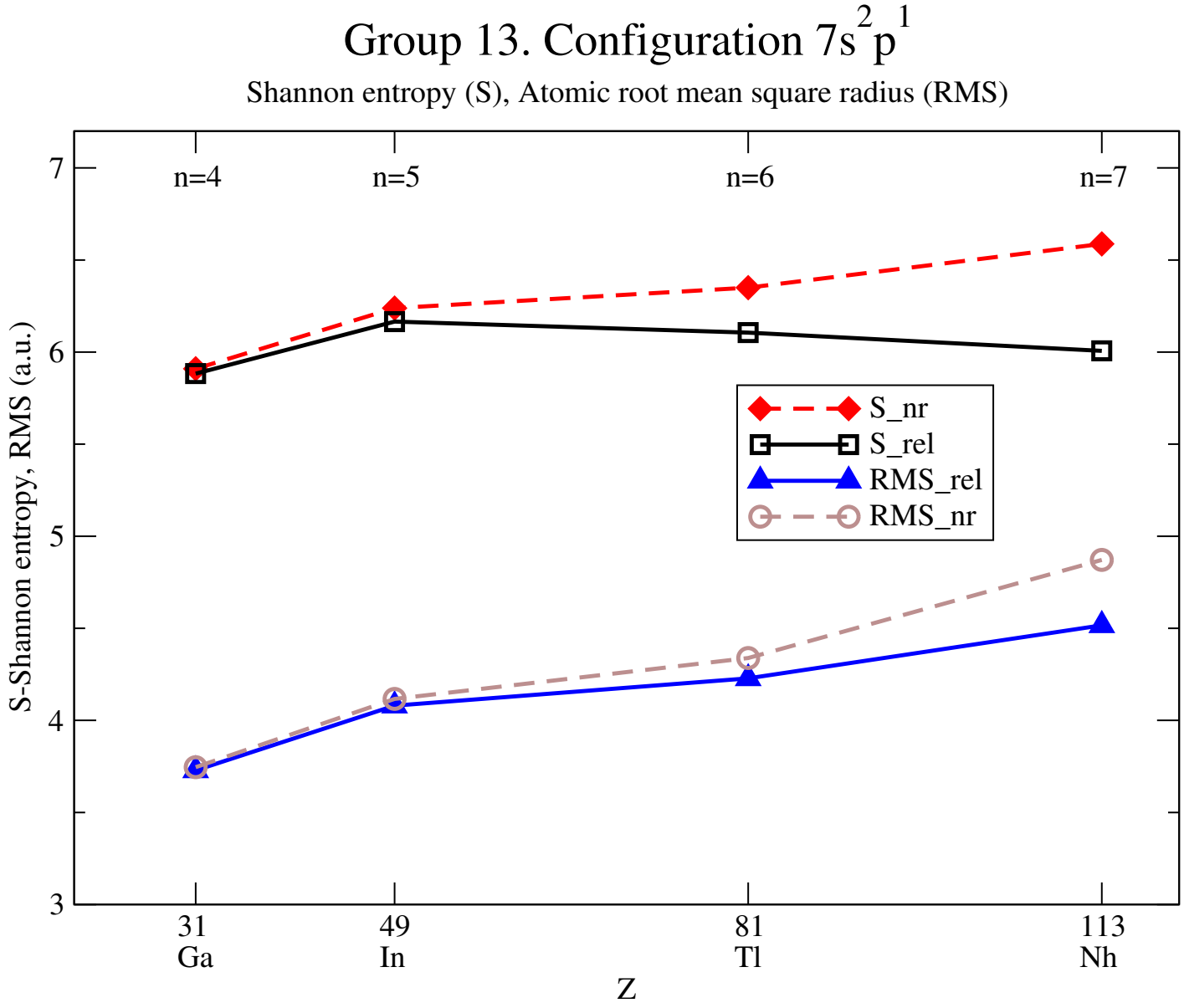
12th group of elements



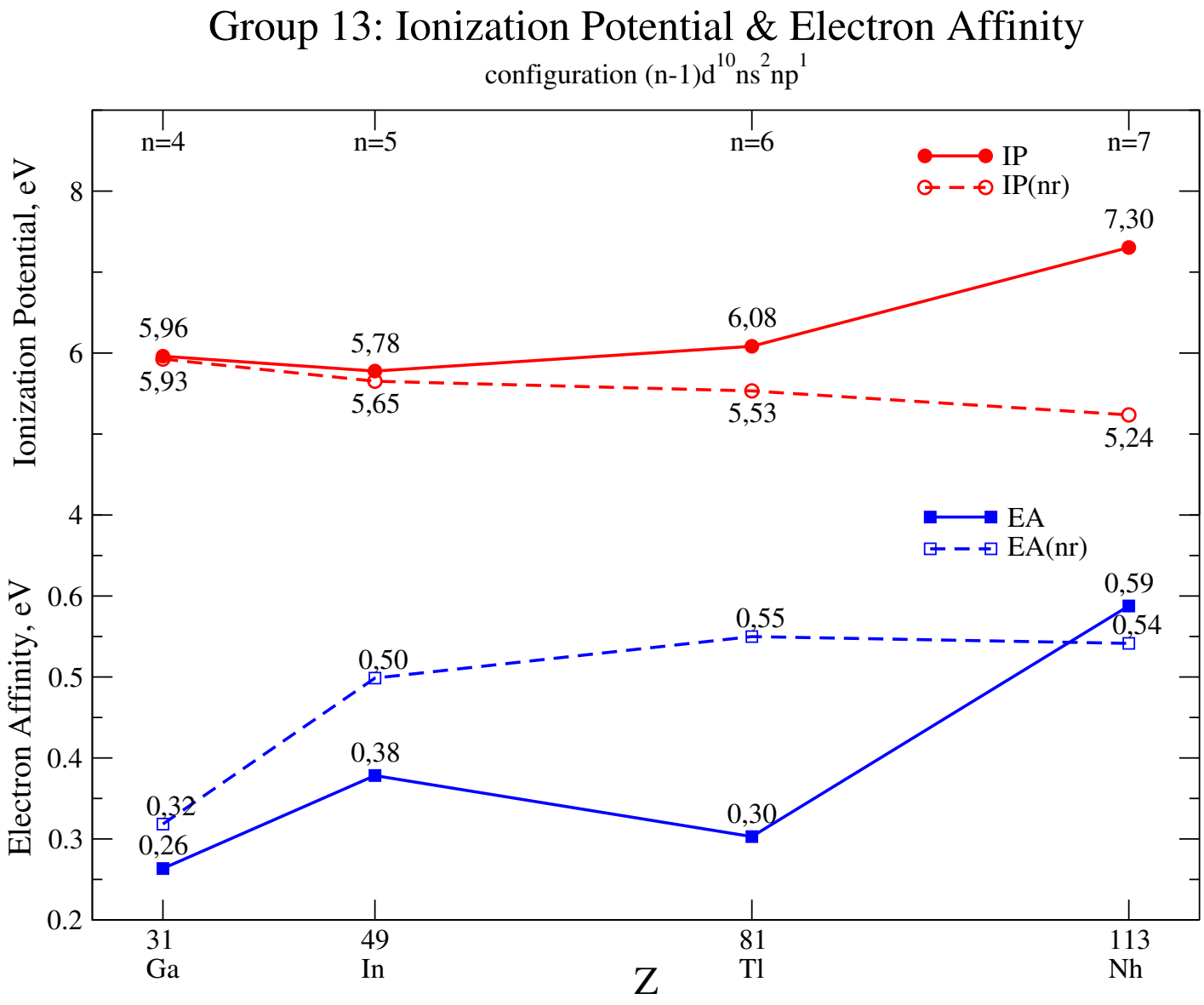
12th group of elements



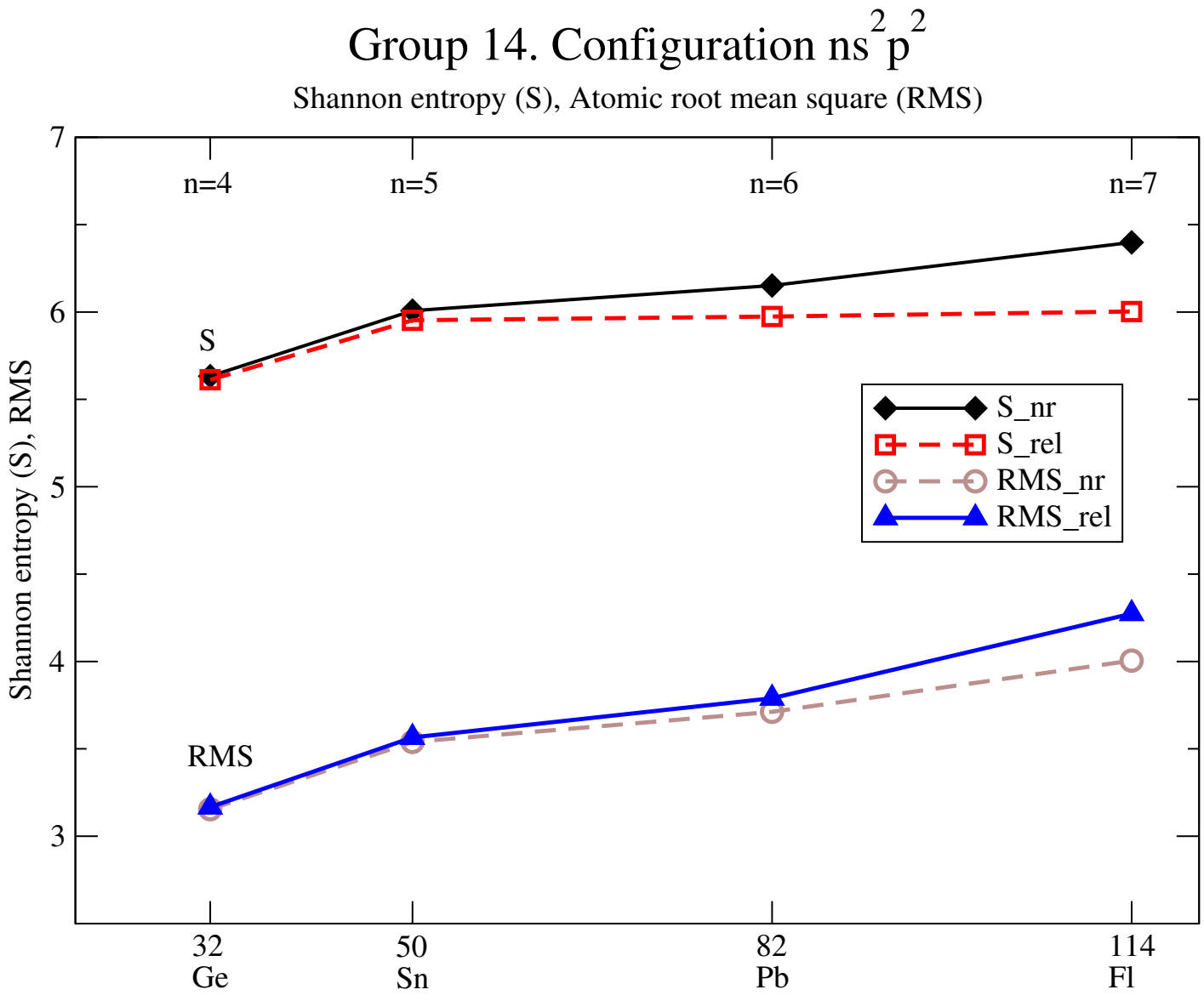
13th group of elements



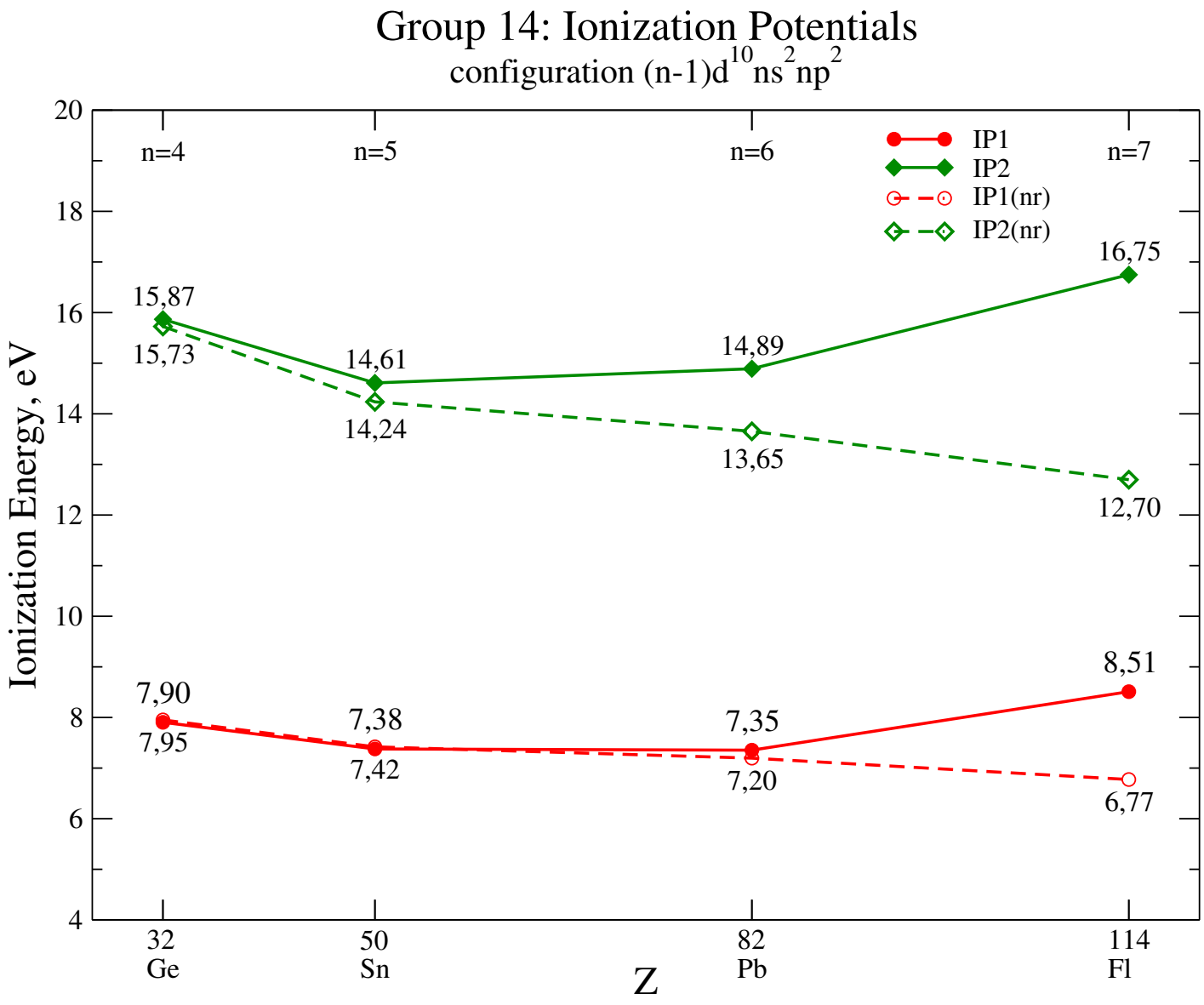
13th group of elements



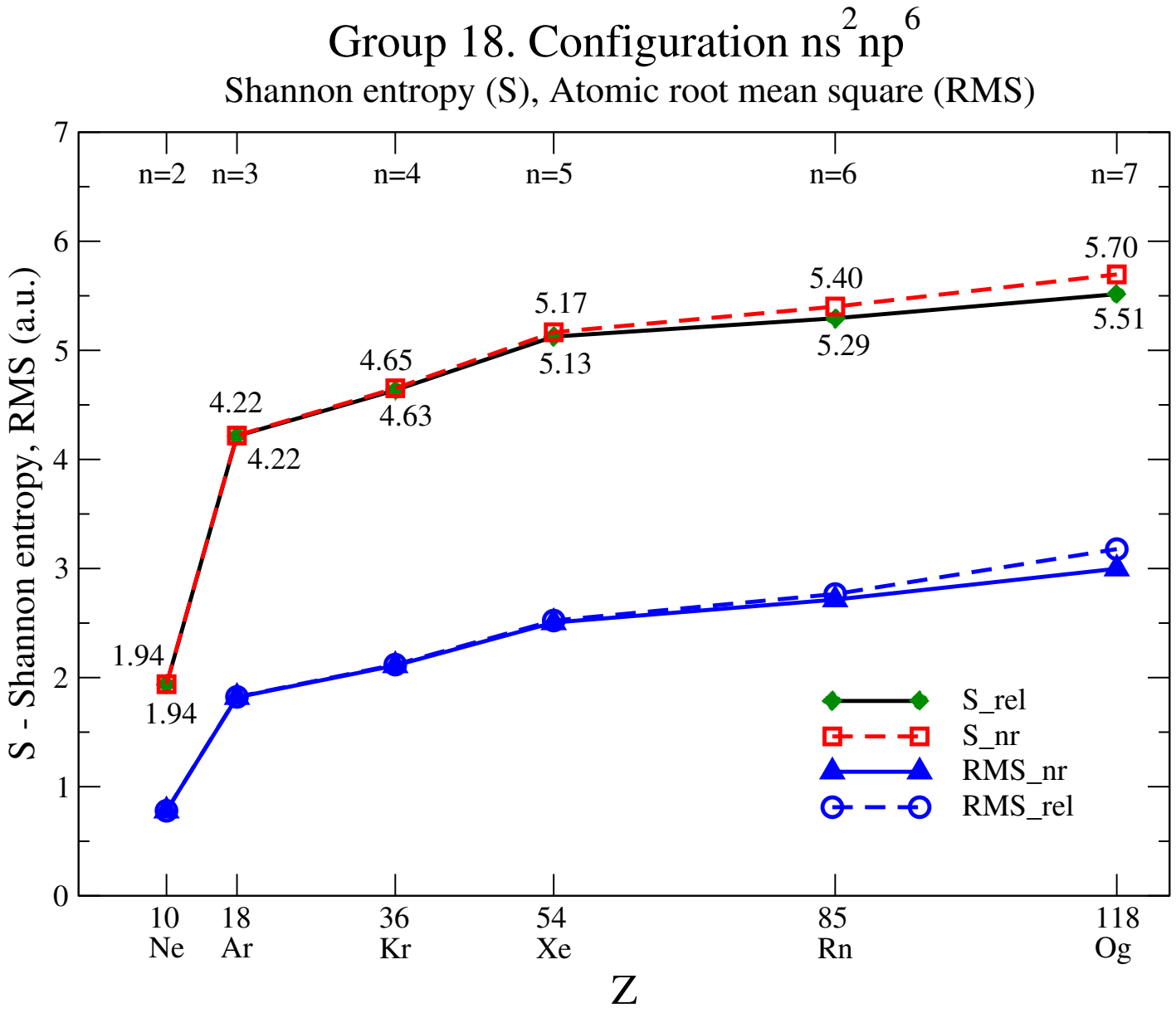
14th group of elements



14th group of elements



18th group of elements



18th group of elements

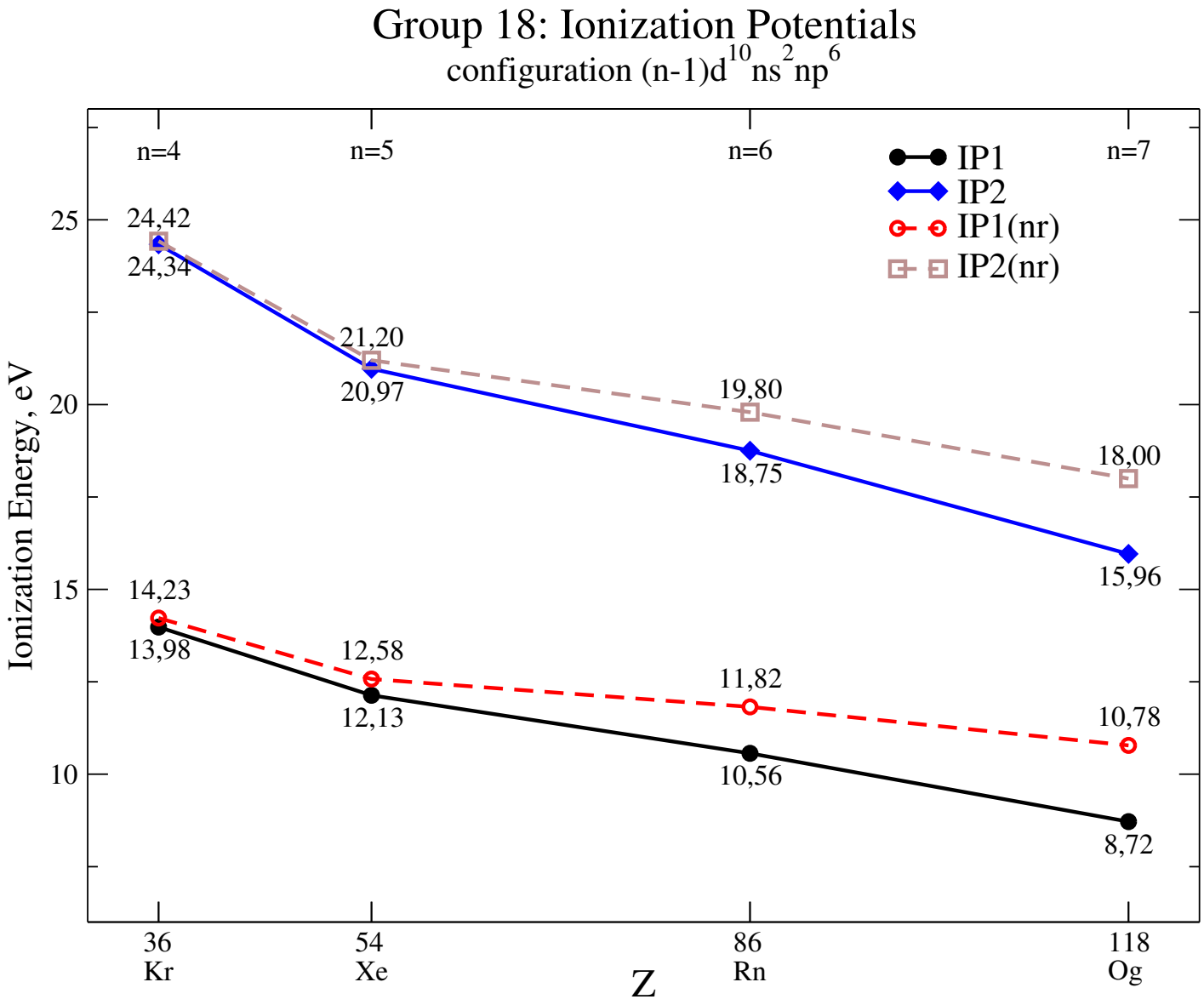


Таблица: Comparison of electron affinity (EA) for Og and the QED contribution to EA calculated with the FSCC and CI-DFS methods with results of other authors

Reference	Method	EA	$\delta\mathrm{EA}_{\mathrm{QED}}$	$\mathrm{EA}_{\mathrm{Total}}$
Present Work	FSCC	0.078(4)	-0.002(1)	0.076(4)
Present Work	CI-DFS	0.070(10)	-0.002(1)	0.068(10)
Eliav *et al.* [1]	FSCC	0.056(10)		
Goidenko *et al.* [2]	FSCC	0.064(2)	-0.0059(5)	0.058(3)
Lackenby *et al.* [3]	CI+MBPT			0.096

1. *E. Eliav, U. Kaldor, Y. Ishikawa, and P. Pyykkö, PRL 77, 5350 (1996).*
2. *I. Goidenko, L. Labzowsky, E. Eliav, U. Kaldor, and P. Pyykkö, PRA 67, 020102 (2003).*
3. *B. G. C. Lackenby, V. A. Dzuba, and V. V. Flambaum, PRA 98, 042512 (2018).*

Thank You for Attention.

Hydrogen-like (Coulomb) Sturmians