Similarities and differences between atomic and nuclear systems "Are atoms spherical?"

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科研費
KAKENHI

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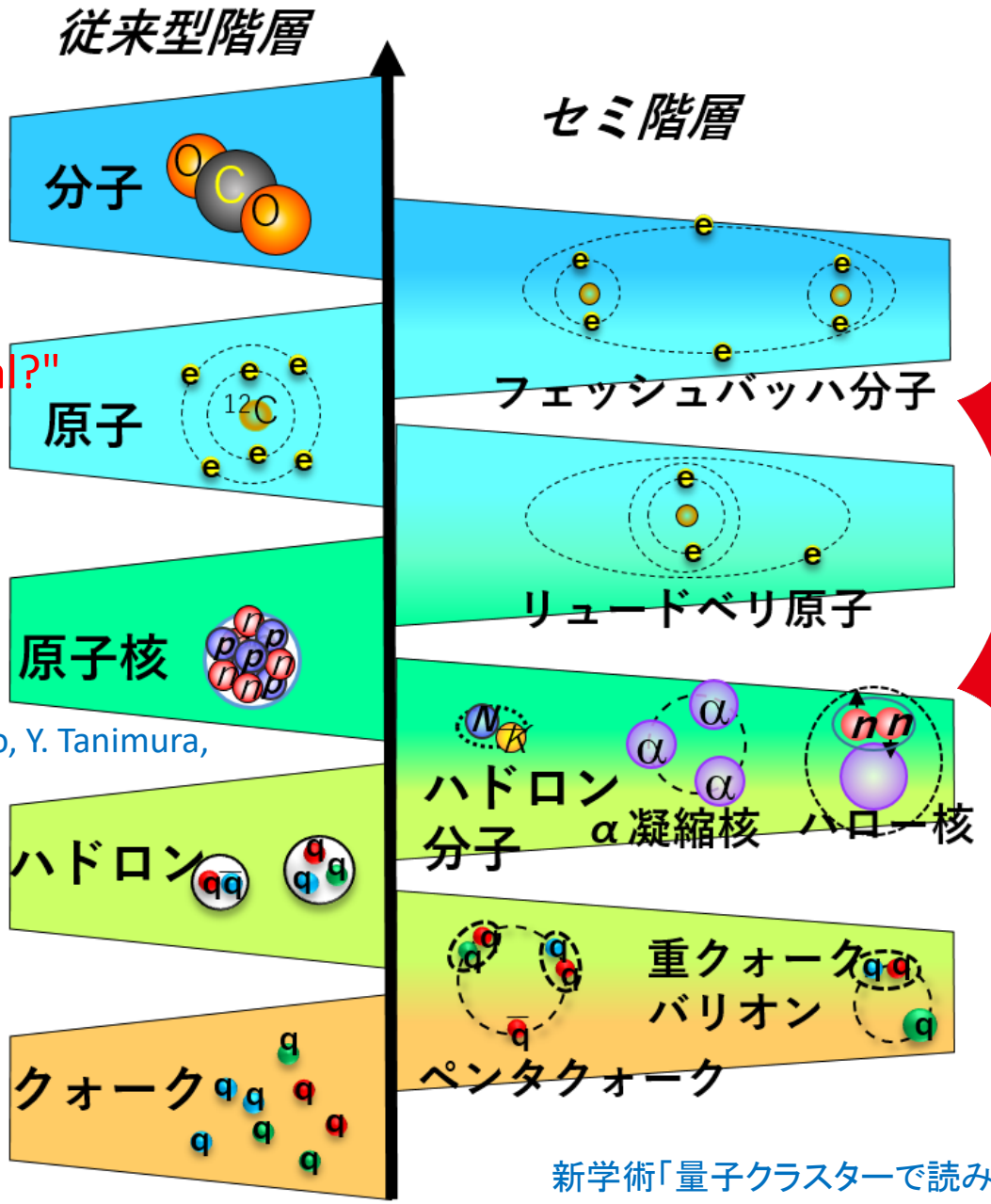
Japan Society for the Promotion of Science

Outline



This talk:
"Are atoms spherical?"

T. Naito, S. Endo, K. Hagino, Y. Tanimura,
arXiv:2009.05955

My Other Research
: cold atoms
Efimov states,
unitary Fermi gas

新学術「量子クラスターで読み解く物質の階層構造」HP

Are atoms spherical?

arXiv:2009.05955 (2020)

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Kouichi Hagino (萩野浩一),[5,‡] and Yusuke Tanimura (谷村雄介)[3,6,§]

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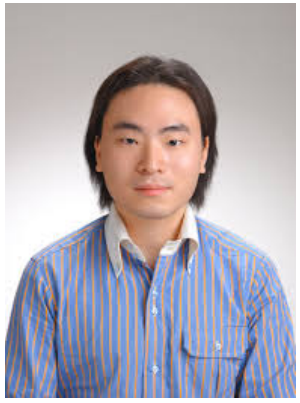
(Dated: September 13, 2020)

Atomic nuclei can be spontaneously deformed into non-spherical shapes as many-nucleon systems. We discuss to what extent a similar deformation takes place in many-electron systems. To this end, we employ several many-body methods, such as the unrestricted Hartree-Fock method, post-Hartree-Fock methods, and the density functional theory, to compute the electron distribution in atoms. We show that the electron distribution of open-shell atoms is deformed due solely to the single-particle valence orbitals, while the core part remains spherical. This is in contrast to atomic nuclei, which can be deformed collectively. We qualitatively discuss the origin for this apparent difference between atoms and nuclei by estimating the energy change due to deformation.



Tomoya Naito
(Univ. Tokyo & RIKEN)
Density Functional Theory,
numerical quantum many-body

Shimpei Endo(Tohoku Univ.)
Cold atoms, Universality of
few- and many-body system

Koichi Hagino (Kyoto Univ.)
Nuclear theory

Yusuke Tanimura(Tohoku Univ.)
Nuclear theory,
Hartree-Fock theory

Nuclei and atoms

- Interacting quantum many-body system of spin 1/2 fermions

Atomic Nuclei

- Many-body system of nucleons (protons & neutrons)
- Self-bound via nuclear force
- Inter-particle interaction essentially attractive

Atom

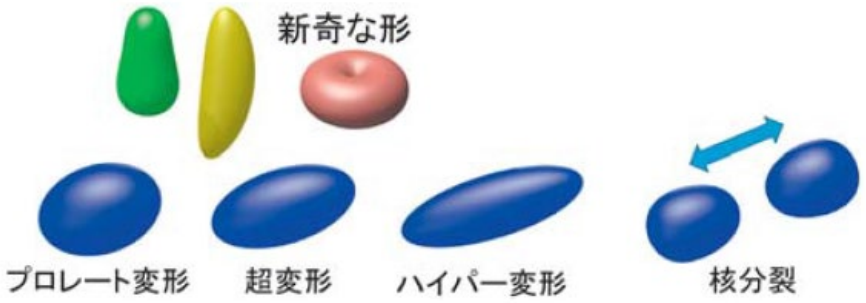
- Many-body system of electrons (+ central nucleus)
- Self-bound by central Coulomb force of the nucleus
- Inter-particle interaction repulsive Coulomb

Shape of Nuclei

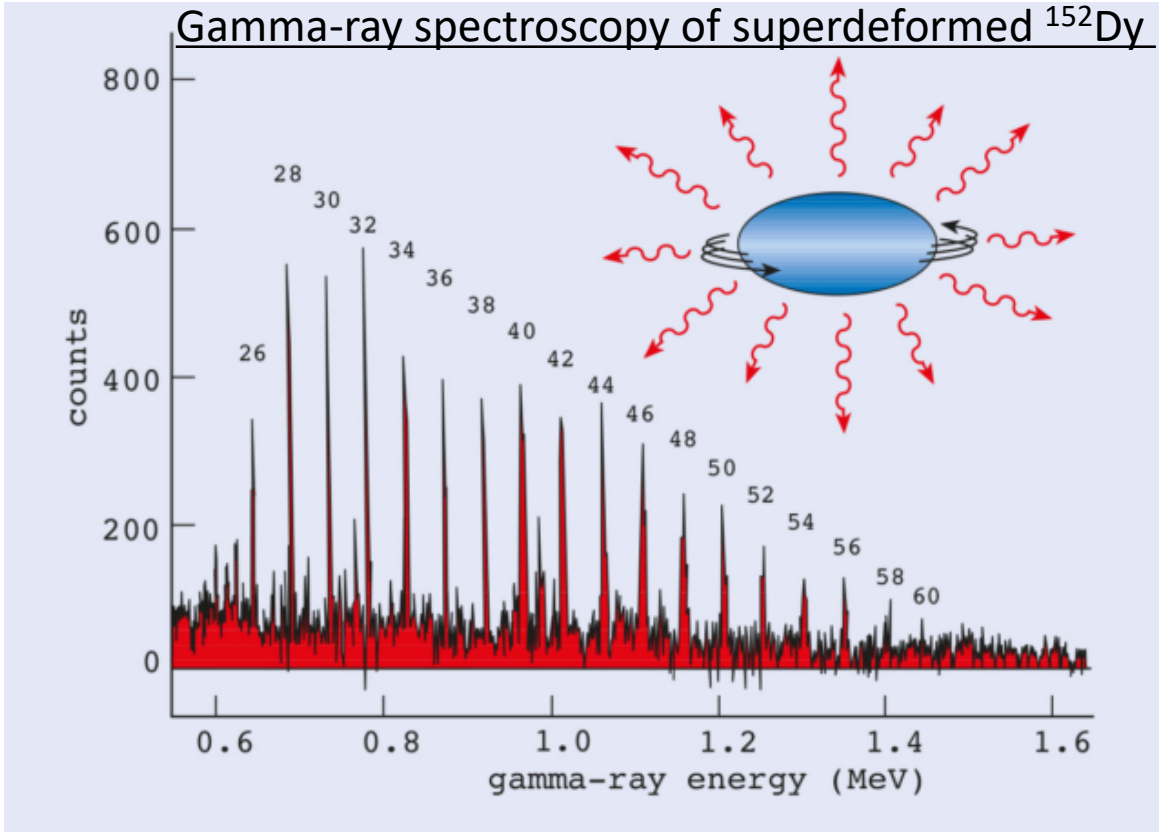
- Spontaneous breaking of rotational symmetry

Rotational spectrum

$$E = \frac{L(L+1)}{2I} \qquad L = 0, 1, 2, ...$$



Gamma-ray spectroscopy of superdeformed ^{152}Dy



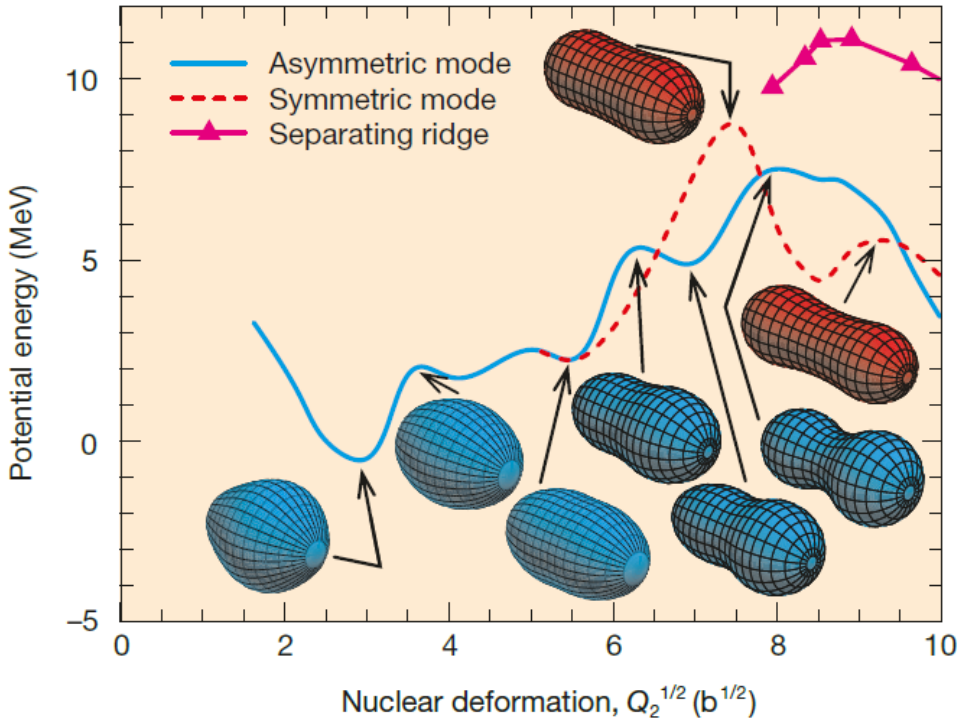
日本物理学会雑誌「物理学70の不思議」

Twin, et al., Phys. Rev. Lett. (1998)
Physics World, "A new spin on nuclei" (1998)

Collective deformations in nuclear systems

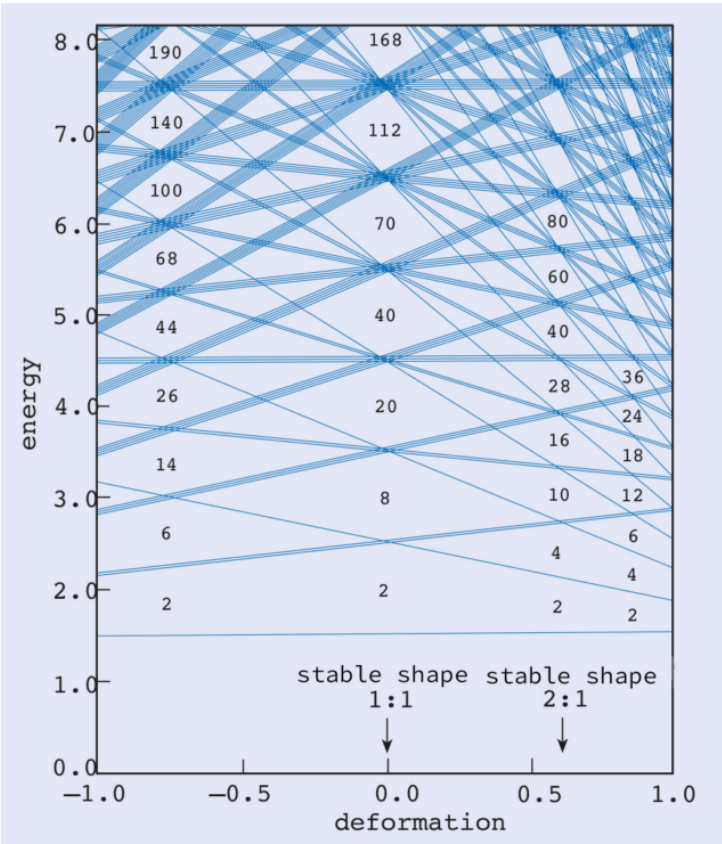
- Deformation parameter β , Multipole moment Q_i
  - Change of magic number in deformed nuclei
  - Nuclear fission
  - .....

Potential energy of nuclear fission of ^{228}Ra



Moller, Madland, Sierk, Iwamoto, Nature (2001)

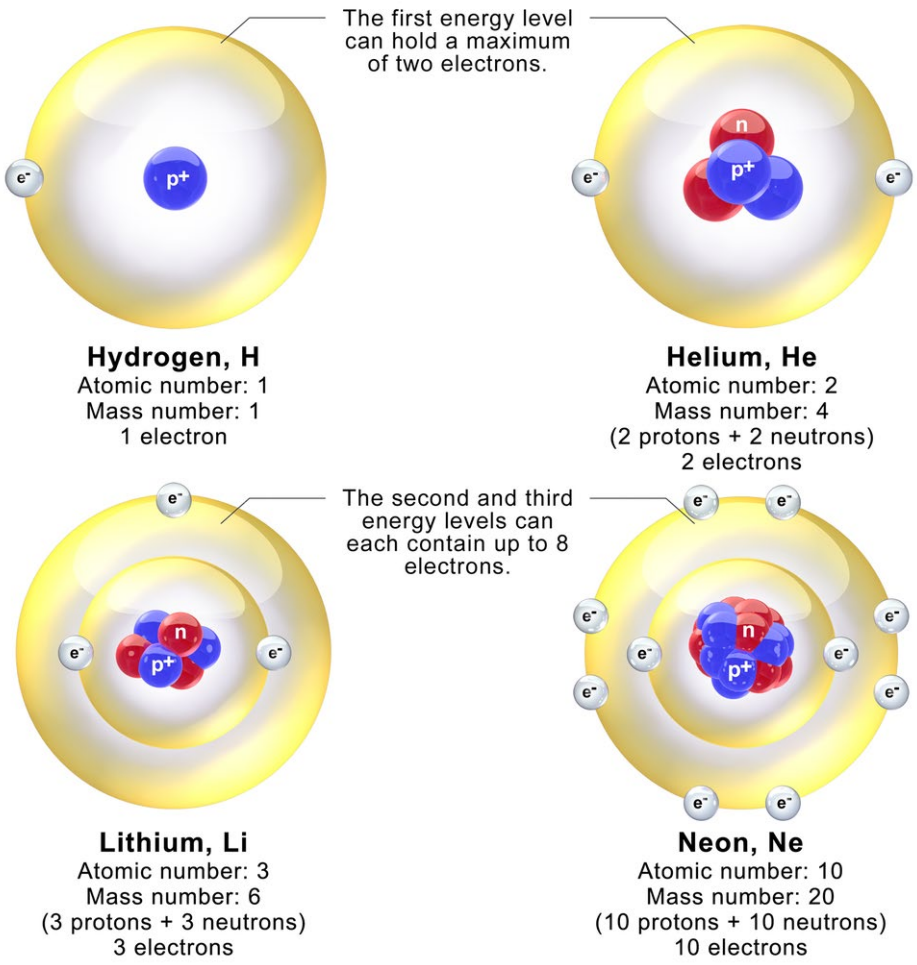
Nilson diagram



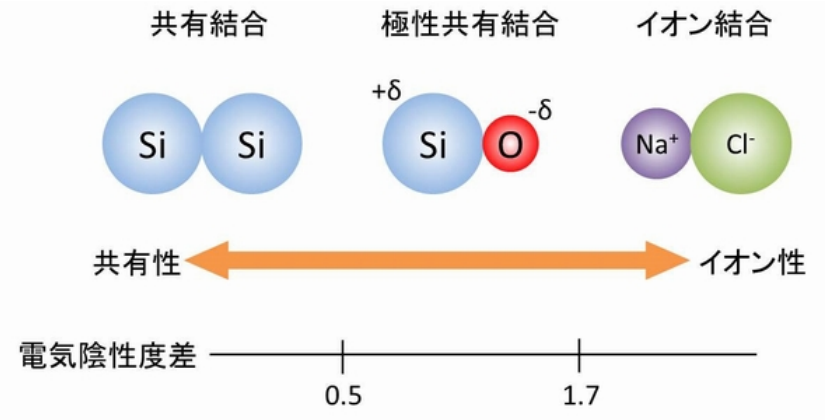
Twin, et al., Phys. Rev. Lett. (1986)
Physics World, "A new spin on nuclei" (1998)

What is the shape of atoms?

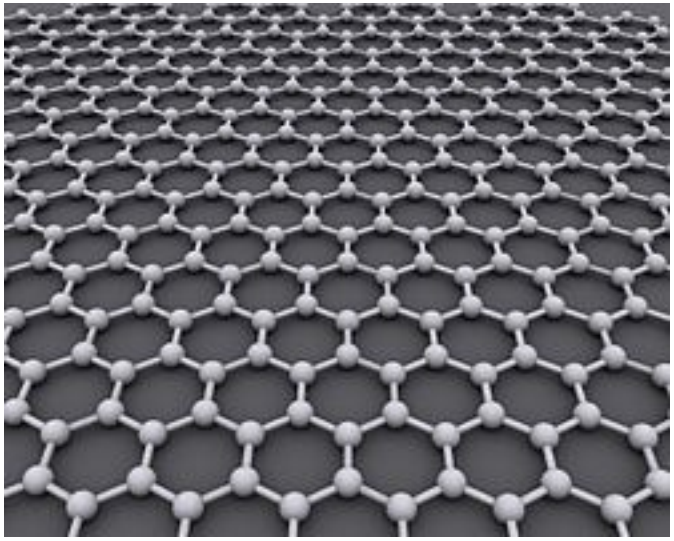
- Usually illustrated spherically



Bohr model of atoms: Wikipedia



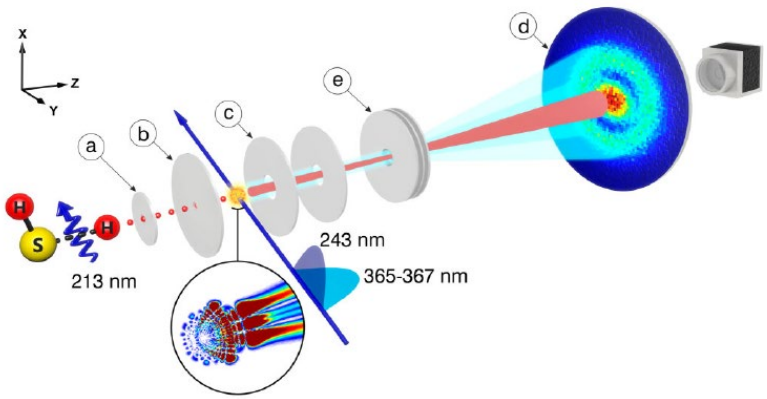
Univ. Tokyo, Press release



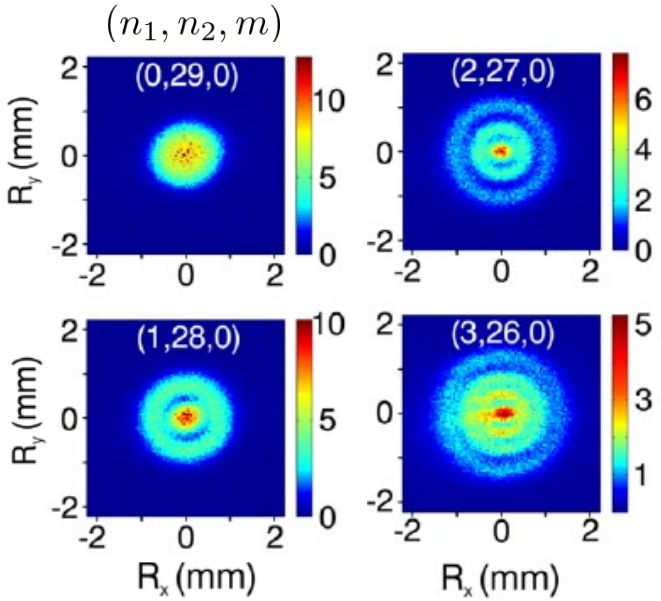
Graphene, Wikipedia

Observation of electron wave function in atoms & molecules

Electronic wave function of H atom
(measured by photoionization microscopy)

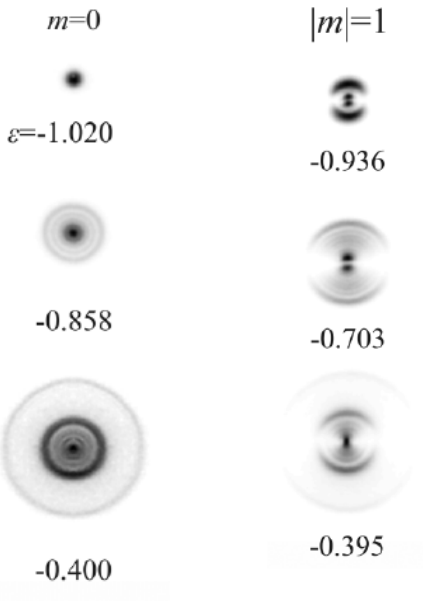


E-field along z(outplane)-axis

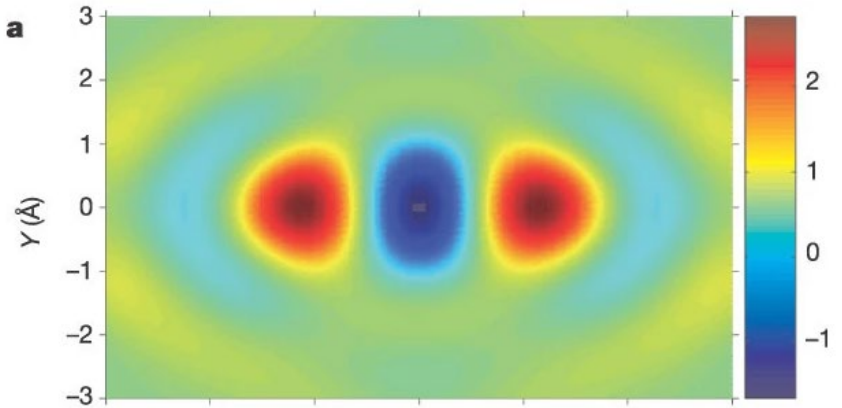


Stodolna, et al., Phys. Rev. Lett. (2013)

Electronic wave function of Li atom
(photoionization microscopy)

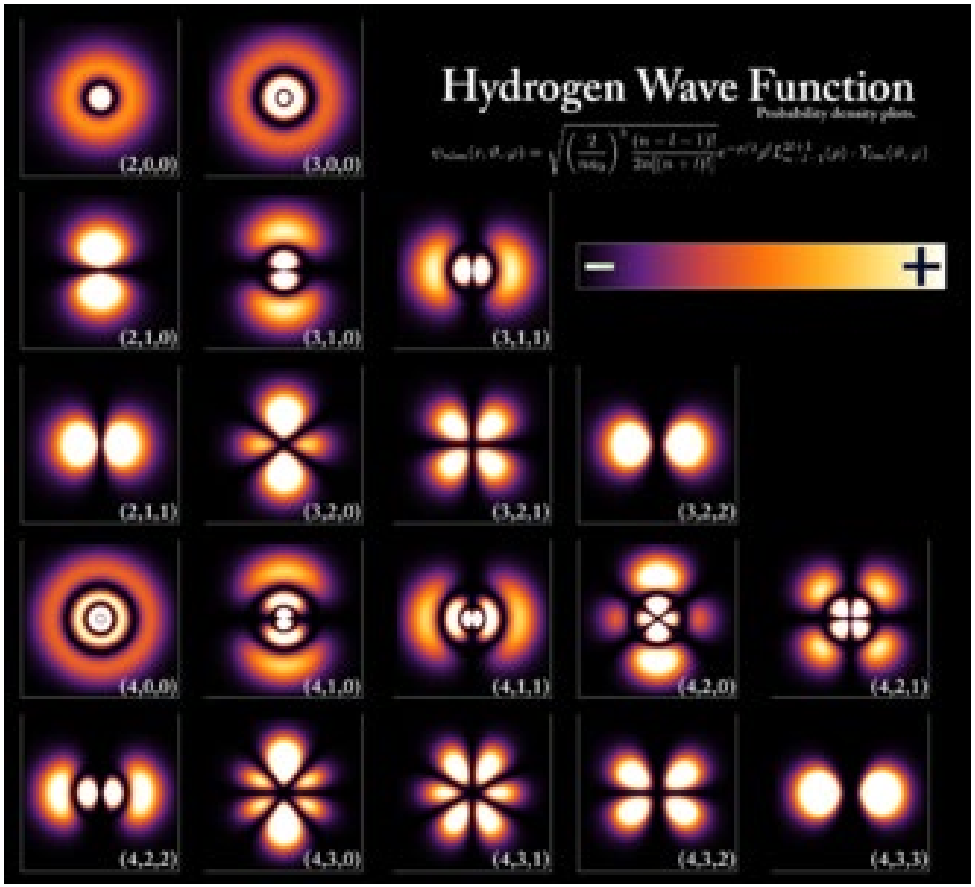


Tomographic imaging of HOMO orbital in N_2



Itatani, et al., Nature (2004)

Deformed single-particle orbital in atoms



Single-particle orbital in H (theory): Wikipedia

$\beta = 0, \quad Q_{ij} = 0$ for s-orbital

$\beta \neq 0, \quad Q_{ij} \neq 0$ for p-orbital, d-orbital,....

Single-particle deformation ≠ Collective deformation

Brief summary our work

Motivation

- Can atoms be deformed?
- What is the difference between atoms and nuclei?

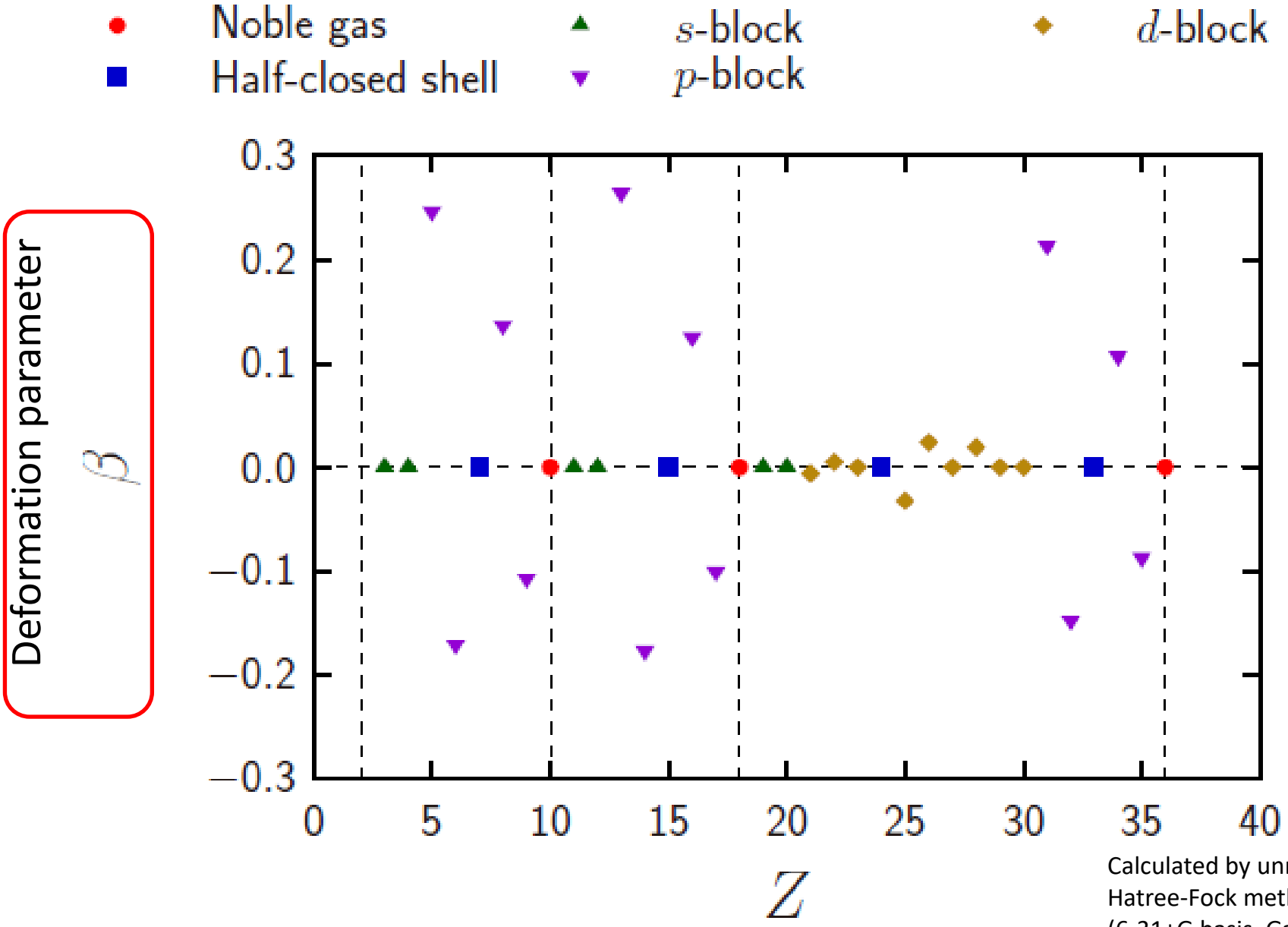
Method

- 3D electron many-body calculations for various atoms
- Analytical model

Main results

- Electron w.f. in atoms do not deform collectively.
- Atoms may be non-spherical only in the surface region via single-particle effect of the valence electron
- Many-body effects suppress deformations (screening)
- Repulsive Coulomb interaction ⇔ Attractive nuclear force

Mains result: Deformation parameters of atoms



Calculated by unrestricted Hatree-Fock method
(6-31+G basis, Gaussian code)
No spin-orbit.

Definition of the deformation parameter βi

※ Intrinsic frame ≠ Laboratory frame

Quadrupole moment

$$Q_{ij} = \int (3r_i r_j - \delta_{ij} r^2)\, \rho(r)\, dr$$

diagonalize ⇒ $Q_k \quad (k = x, y, z)$

Q-moment in the intrinsic frame

Deformation parameter (quadrupole)

$$\beta_k = \sqrt{\frac{\pi}{5}} \frac{Q_k}{N_{\rm tot} \langle r^2 \rangle} \qquad (k = x,\ y,\ z)$$

Relation to angle-dependent radius R(θ,ϕ)

$$R(\theta, \phi) = R_0 \left[1 + \sum_{l=2}^{\infty} \sum_{m=-l}^{l} a_{lm} Y_{lm}^*(\theta, \phi) \right]$$

For axially symmetric quadrupole deformation

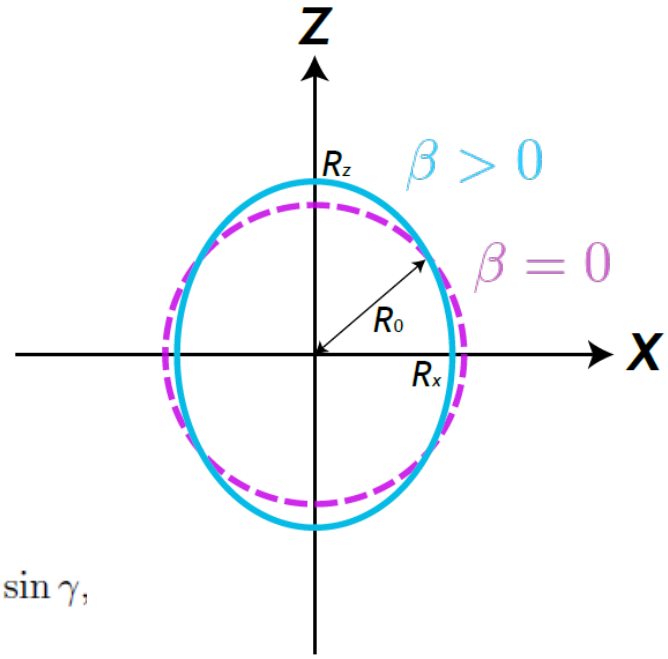
$$R_x = R_y = R(\pi/2, \phi) = R_0 \left(1 - \sqrt{\frac{5}{16\pi}} \beta \right)$$

$$R_z = R(0, \phi) = R_0 \left(1 + 2\sqrt{\frac{5}{16\pi}} \beta \right),$$

$$\beta = \beta_z \qquad \beta_x = \beta_y = -\beta_z/2$$

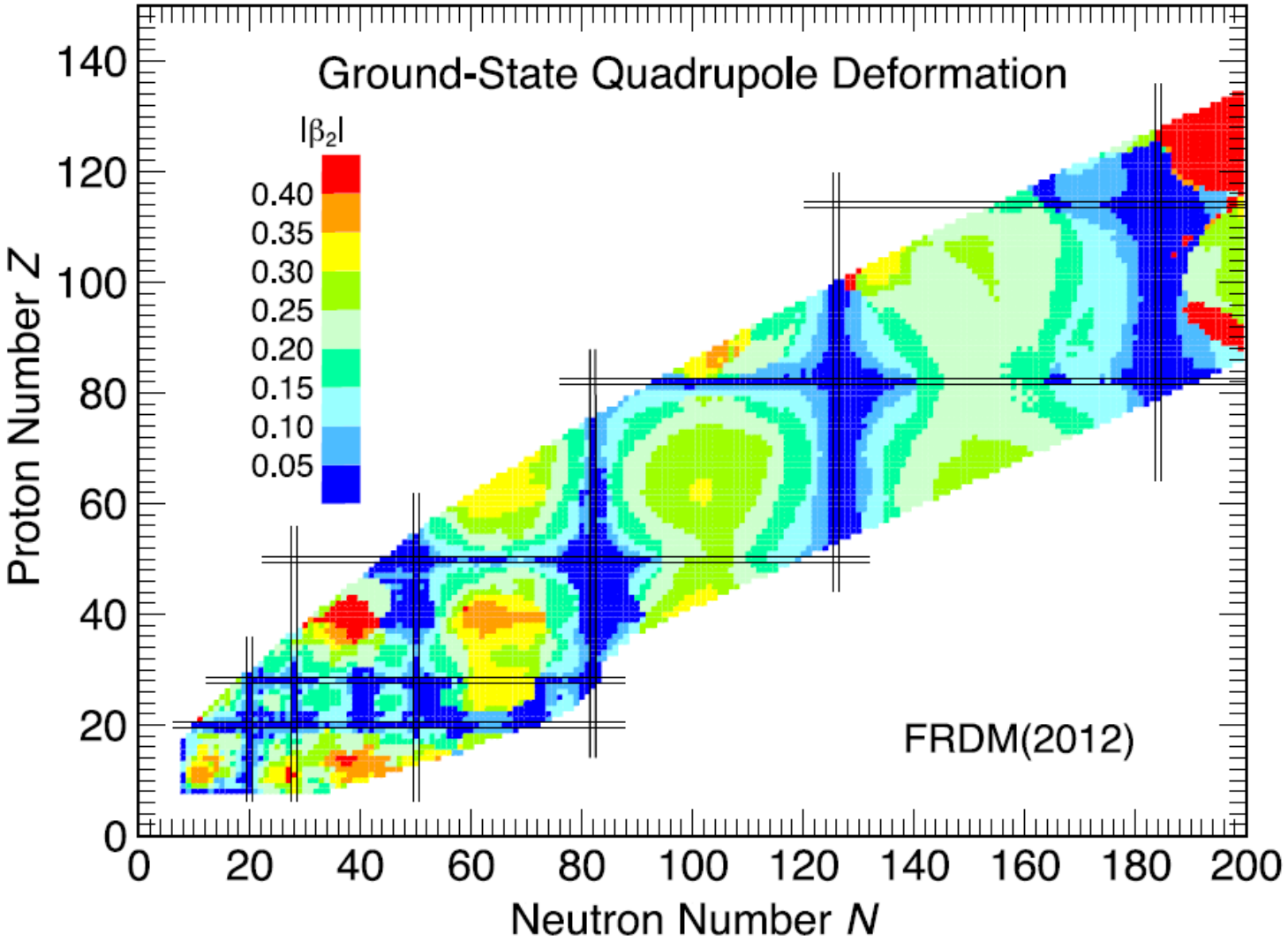
$$\gamma = 0$$

$$a'_{20} = \beta \cos\gamma,$$
$$a'_{22} = a'_{2-2} = \tfrac{1}{\sqrt{2}} \beta \sin\gamma,$$
$$a'_{21} = a'_{2-1} = 0.$$



Collective deformations in nuclear systems

- Deformed nuclei: $|\beta| \gtrsim 0.2 - 0.3$



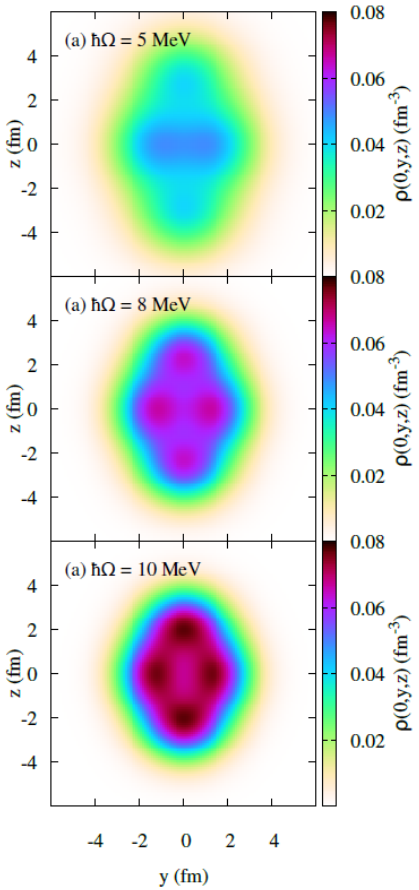
Moller, Sierk, Ichikawa, Sagawa, Atomic Data and Nuclear Data Table(2016): FRDM(2012)

Examples of deformation parameter β in nuclei

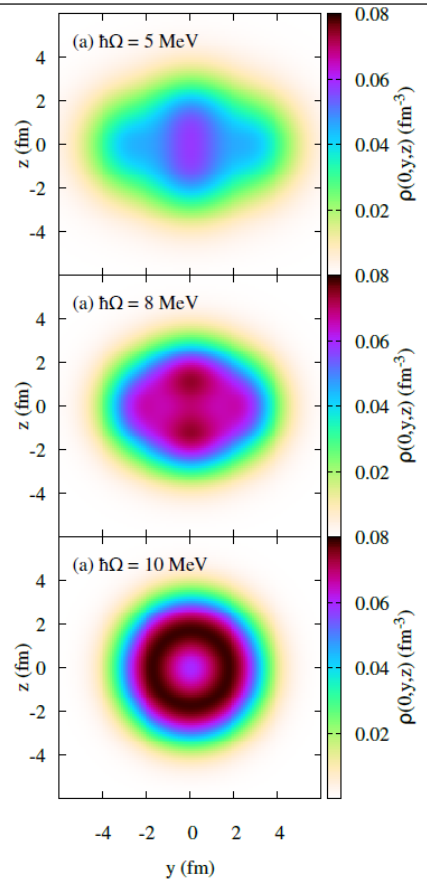
Neutron drop model (neutrons in a harmonic potential $m_n\Omega^2 r^2/2$)

- Deformations $|\beta| \gtrsim 0.2 - 0.3$

System	$\hbar\Omega$ (MeV)	$\langle r^2 \rangle^{1/2}$ (fm)	β_z
^{10}n	5	3.67	0.34
	8	3.14	0.28
	10	2.91	0.23

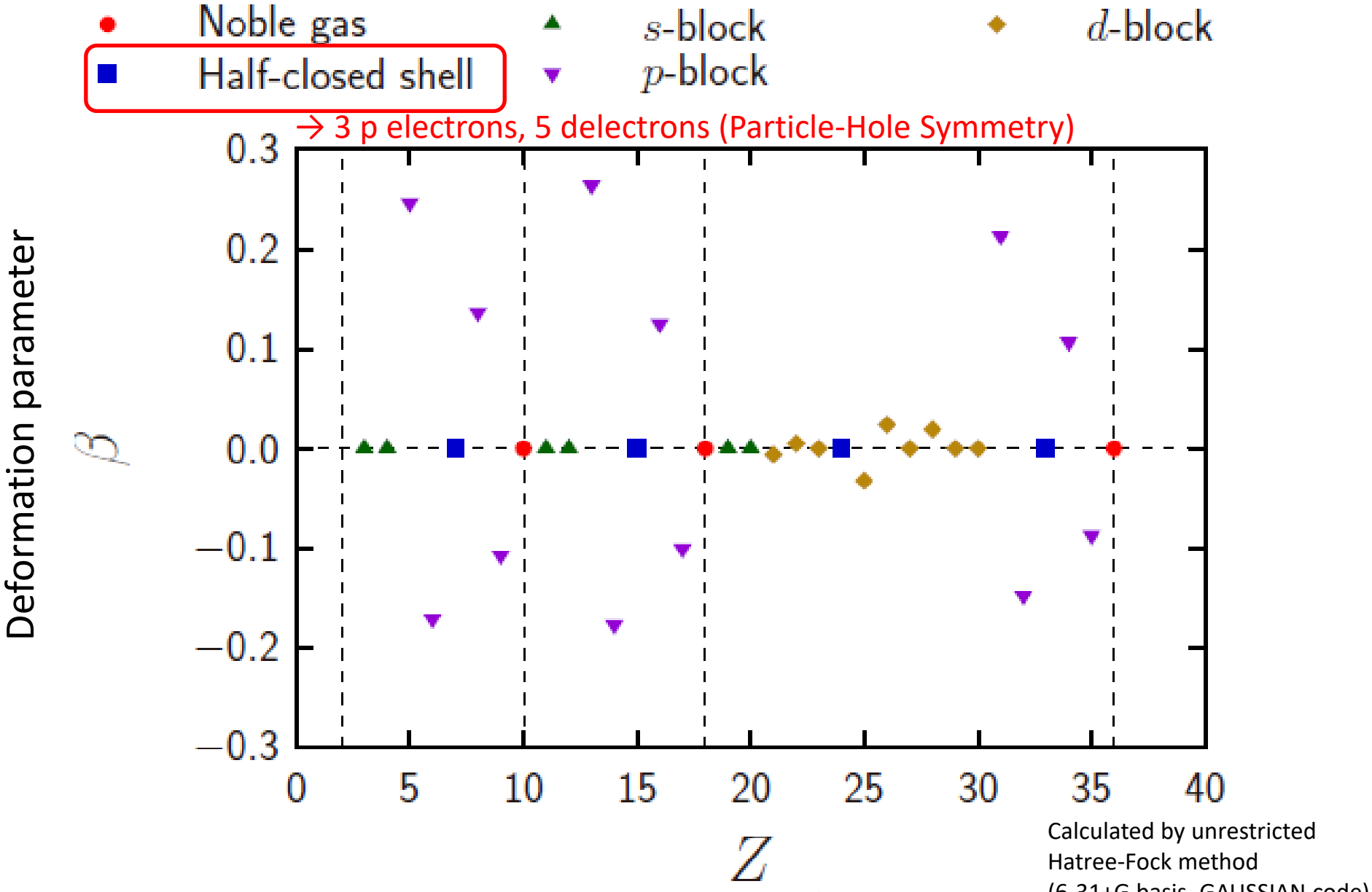


System	$\hbar\Omega$ (MeV)	$\langle r^2 \rangle^{1/2}$ (fm)	β_z
^{14}n	5	3.91	−0.31
	8	3.32	−0.20
	10	3.08	0.00



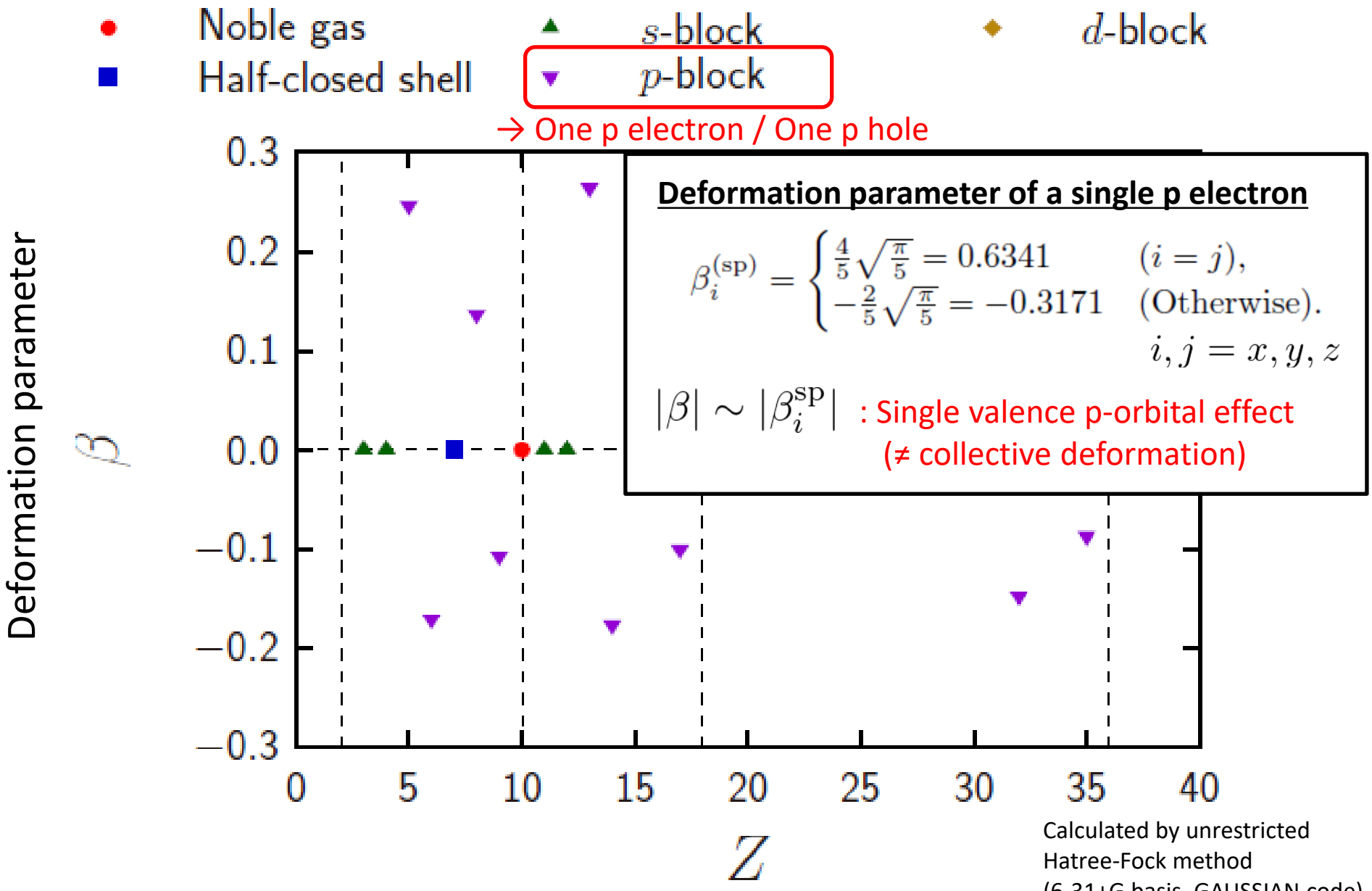
Calculated with 3D
relativistic Hatree-Fock method
(PK1 parameter for nucleon-meson)

Mains result: Deformation parameters of atoms



Calculated by unrestricted Hatree-Fock method
(6-31+G basis, GAUSSIAN code)
No spin-orbit.

Mains result: Deformation parameters of atoms



Deformation parameter of a single p electron

$$\beta_i^{(\mathrm{sp})} = \begin{cases} \frac{4}{5}\sqrt{\frac{\pi}{5}} = 0.6341 & (i = j), \\ -\frac{2}{5}\sqrt{\frac{\pi}{5}} = -0.3171 & (\text{Otherwise}). \end{cases}$$

$$i, j = x, y, z$$

$|\beta| \sim |\beta_i^{\mathrm{sp}}|$: Single valence p-orbital effect
(≠ collective deformation)

Calculated by unrestricted Hatree-Fock method
(6-31+G basis, GAUSSIAN code)
No spin-orbit.

Mains result: Deformation parameters of atoms

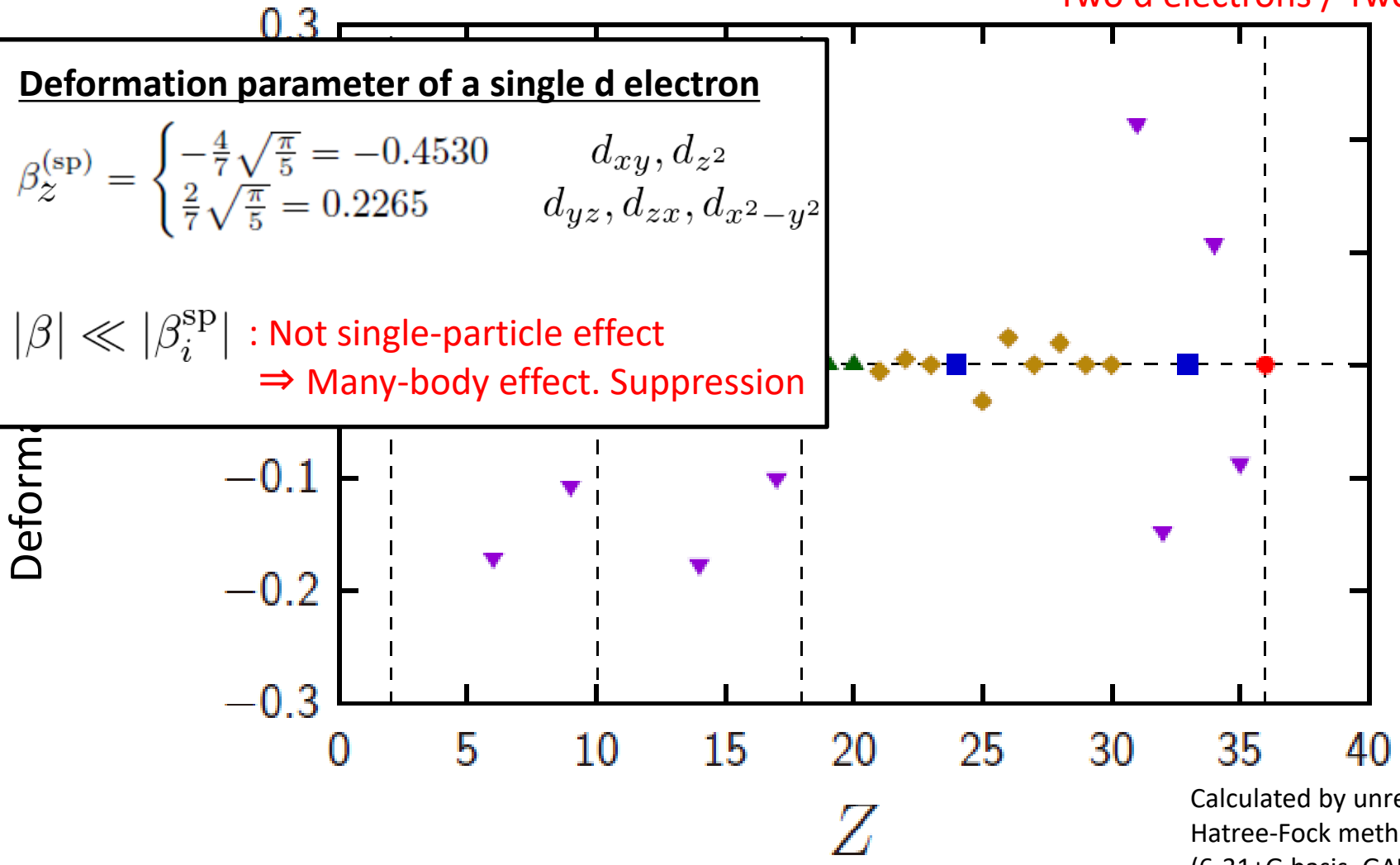
Noble gas
Half-closed shell
s-block
p-block
d-block

→ One d electron / One d hole
Two d electrons / Two d holes

Deformation parameter of a single d electron

$$\beta_z^{(\mathrm{sp})} = \begin{cases} -\frac{4}{7}\sqrt{\frac{\pi}{5}} = -0.4530 & d_{xy}, d_{z^2} \\ \frac{2}{7}\sqrt{\frac{\pi}{5}} = 0.2265 & d_{yz}, d_{zx}, d_{x^2-y^2} \end{cases}$$

$|\beta| \ll |\beta_i^{\mathrm{sp}}|$: Not single-particle effect
⇒ Many-body effect. Suppression



Calculated by unrestricted Hatree-Fock method
(6-31+G basis, GAUSSIAN code)
No spin-orbit.

Sc atom (Z=21): Closed shell + $(3d_{xy})^1$ +$(4s)^2$

Spin	NO	ε_j	$\langle r^2 \rangle_{nl}$	$q_z^{(\mathrm{sp})}$	$\beta_z^{(\mathrm{sp})}$
α	$1s$	-165.8952	0.0073	0.0000	0.0000
α	$2s$	-19.0956	0.1393	0.0000	0.0000
α	$2p_z$	-15.6861	0.1162	0.0930	0.6341
α	$2p_x$	-15.6830	0.1164	-0.0466	-0.3171
α	$2p_y$	-15.6830	0.1164	-0.0466	-0.3171
α	$3s$	-2.5988	1.3396	-0.0031	-0.0018
α	$3p_x$	-1.6232	1.6265	-0.6506	-0.3171
α	$3p_y$	-1.6232	1.6265	-0.6506	-0.3171
α	$3p_z$	-1.5893	1.6107	1.2886	0.6341
α	$3d_{xy}$	-0.3375	3.7873	-2.1642	-0.4530
α	$4s$	-0.2171	17.4198	-0.2152	-0.0098
β	$1s$	-165.8951	0.0073		
β	$2s$	-19.0832	0.1391		
β	$2p_z$	-15.6837	0.1162		
β	$2p_x$	-15.6657	0.1161		
β	$2p_y$	-15.6657	0.1161		
β	$3s$	-2.5451	1.3370	0.0387	0.0230
β	$3p_z$	-1.5798	1.6156	1.2925	0.6341
β	$3p_x$	-1.5273	1.6196	-0.6478	-0.3171
β	$3p_y$	-1.5273	1.6196	-0.6478	-0.3171
β	$4s$	-0.2051	18.5388	1.9406	0.0830
Total			53.1312	-0.4189	-0.0062

← Large deformation of $3d_{xy}$ electron

Deformation parameter of a single d electron

$$\beta_i^{(\mathrm{sp})} = \begin{cases} -\frac{4}{7}\sqrt{\frac{\pi}{5}} = -0.4530 & d_{xy}, d_{z^2} \\ \frac{2}{7}\sqrt{\frac{\pi}{5}} = 0.2265 & d_{yz}, d_{zx}, d_{x^2-y^2} \end{cases}$$

Large deformation of 4s orbital
← cancelling $3d_{xy}$ deformation!

← Very small total deformation

Sc atom (Z=21): Closed shell + $(3d_{xy})^1$ +$(4s)^2$

Spin	NO	ε_j	$\langle r^2 \rangle_{nl}$	$q_z^{(\mathrm{sp})}$	$\beta_z^{(\mathrm{sp})}$
α	$1s$	-165.8952	0.0073	0.0000	0.0000
α	$2s$	-19.0956	0.1393	0.0000	0.0000
α	$2p_z$	-15.6861	0.1162	0.0930	0.6341
α	$2p_x$	-15.6830	0.1164	-0.0466	-0.3171
α	$2p_y$	-15.6830	0.1164	-0.0466	-0.3171
α	$3s$	-2.5988	1.3396	-0.0031	-0.0018
α	$3p_x$	-1.6232	1.6265	-0.6506	-0.3171
α	$3p_y$	-1.6232	1.6265	-0.6506	-0.3171
α	$3p_z$	-1.5893	1.6107	1.2886	0.6341
α	$3d_{xy}$	-0.3375	3.7873	-2.1642	-0.4530
α	$4s$	-0.2171	17.4198	-0.2152	-0.0098
β	$1s$	-165.8951	0.0073	0.0000	0.0000
β	$2s$	-19.0832	0.1391	0.0001	0.0007
β	$2p_z$	-15.6837	0.1162	0.0929	0.6341
β	$2p_x$	-15.6657	0.1161	-0.0464	-0.3171
β	$2p_y$	-15.6657	0.1161	-0.0464	-0.3171
β	$3s$	-2.5451	1.3370	0.0387	0.0230
β	$3p_z$	-1.5798	1.6156	1.2925	0.6341
β	$3p_x$	-1.5273	1.6196	-0.6478	-0.3171
β	$3p_y$	-1.5273	1.6196	-0.6478	-0.3171
β	$4s$	-0.2051	18.5388	1.9406	0.0830
Total			53.1312	-0.4189	-0.0062

← Small deformation (Pauli principle)

Screening effect of e-e Coulomb interaction (repulsive Coulomb force)

$\langle r^2 \rangle_{4s} > \langle r^2 \rangle_{3dxy}$

Large deformation of 4s orbital

← cancelling $3d_{xy}$ deformation!

Ga atom (Z=31) : Closed shell + $(4p_z)^1$ +$(4s)^2$

Spin	NO	ε_j	$\langle r^2 \rangle_{nl}$	$q_x^{(\mathrm{sp})}$	$q_y^{(\mathrm{sp})}$	$q_z^{(\mathrm{sp})}$
α	$1s$	−378.6802	0.0033	0.0000	0.0000	0.0000
α	$2s$	−47.5198	0.0623	0.0000	0.0000	0.0000
α	$2p_z$	−42.4832	0.0452	−0.0181	−0.0181	+0.0362
α	$2p_x$	−42.4826	0.0452	+0.0362	−0.0181	−0.0181
α	$2p_y$	−42.4826	0.0452	−0.0181	+0.0362	−0.0181
α	$3s$	−6.2558	0.5350	+0.0004	+0.0004	−0.0008
α	$3p_x$	−4.4690	0.5485	+0.4388	−0.2194	−0.2194
α	$3p_y$	−4.4690	0.5485	−0.2194		
α	$3p_z$	−4.4678	0.5474	−0.2189		
α	$3d_{x^2-y^2}$	−1.1813	0.7753	+0.2215		
α	$3d_{xy}$	−1.1813	0.7753	+0.2215		
α	$3d_{yz}$	−1.1761	0.7753	−0.4430		
α	$3d_{zx}$	−1.1761	0.7753	+0.2215		
α	$3d_{z2}$	−1.1743	0.7757	−0.2239		
α	$4s$	−0.4471	7.5754	+0.0036	+0.0036	−0.0072
α	$4p_z$	−0.2086	13.9574	−5.5830	−5.5830	+11.1659
β	$1s$	−378.6792	0.0033	0.0000	0.0000	0.0000
β	$2s$	−47.5192	0.0623	0.0000	0.0000	0.0000
β	$2p_x$	−42.4823	0.0452	+0.0362	−0.0181	−0.0181
β	$2p_y$	−42.4823	0.0452	−0.0181	+0.0362	−0.0181
β	$2p_z$	−42.4796	0.0452	−0.0181	−0.0181	+0.0362
β	$3s$	−6.2535	0.5349	+0.0007	+0.0007	−0.0013
β	$3p_x$	−4.4682	0.5483	+0.4386	−0.2193	−0.2193
β	$3p_y$	−4.4682	0.5483	−0.2193	+0.4386	−0.2193
β	$3p_z$	−4.4585	0.5477	−0.2191	−0.2191	+0.4382
β	$3d_{x^2-y^2}$	−1.1808	0.7753	+0.2215	+0.2215	−0.4430
β	$3d_{xy}$	−1.1808	0.7753	+0.2215	+0.2215	−0.4430
β	$3d_{yz}$	−1.1720	0.7753	−0.4430	+0.2215	+0.2215
β	$3d_{zx}$	−1.1720	0.7753	+0.2215	−0.4430	+0.2215
β	$3d_{z2}$	−1.1691	0.7756	−0.2133	−0.2133	+0.4267
β	$4s$	−0.3877	7.3770	−0.0070	−0.0070	+0.0139
Total			41.4245	−5.5788	−5.5788	+11.1576

Deformation parameter of a single p electron

$$\beta_i^{(\mathrm{sp})} = \begin{cases} \frac{4}{5}\sqrt{\frac{\pi}{5}} = 0.6341 & (i=j), \\ -\frac{2}{5}\sqrt{\frac{\pi}{5}} = -0.3171 & (\text{Otherwise}). \end{cases} \quad i,j = x,y,z$$

←Large deformation of $4p_z$ electron ($\beta z = 0.6341...$)

Screening effect impossible

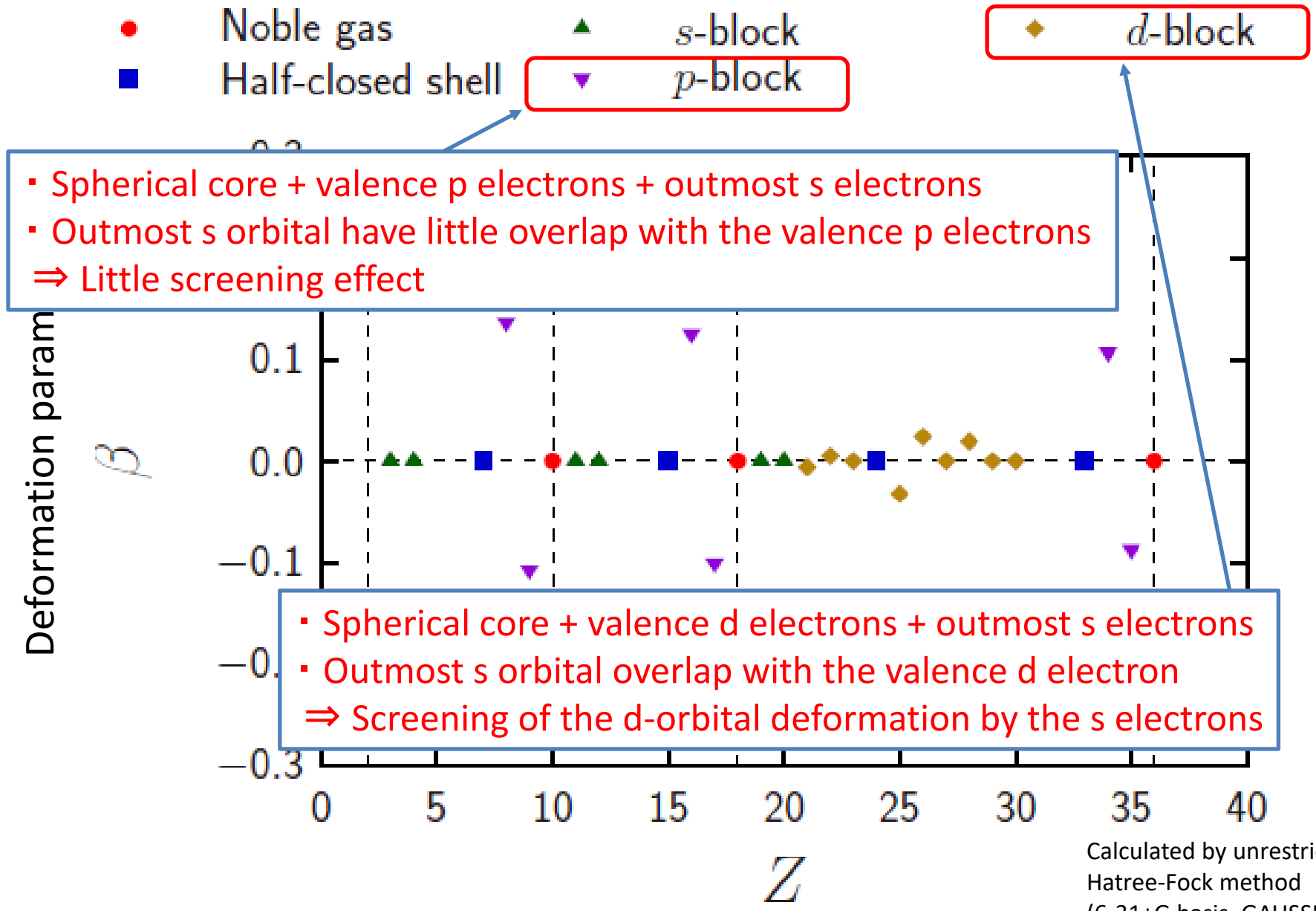
$\langle r^2 \rangle_{4s} \ll \langle r^2 \rangle_{4pz}$

Little wavefunction overlap

← Small deformation of 4s orbital!

← Very large total deformation

Mains result: Deformation parameters of atoms



- Spherical core + valence p electrons + outmost s electrons
- Outmost s orbital have little overlap with the valence p electrons

⇒ Little screening effect

- Spherical core + valence d electrons + outmost s electrons
- Outmost s orbital overlap with the valence d electron

⇒ Screening of the d-orbital deformation by the s electrons

Calculated by unrestricted Hatree-Fock method
(6-31+G basis, GAUSSIAN code)
No spin-orbit.

Why atoms and nuclei differ?: Qualitative discussion

Deformed many-body wavefunction

Bohigas, Lane, Martorell, Phys. Rep. (1979).
Bertsch, Feldmeier, Phys. Rev. C (1997)

$$|\Psi_\beta\rangle = e^{\beta[H,Q]}\,|\Psi_0\rangle \qquad Q = m\sum_{i=1}^{N}\left[z_i^2 - \frac{1}{2}\left(x_i^2+y_i^2\right)\right]$$

$$\Psi_\beta\left(x_1,y_1,z_1,x_2,y_2,z_2,\ldots\right) = \Psi_0\left(e^{\beta}x_1, e^{\beta}y_1, e^{-2\beta}z_1, e^{\beta}x_2, e^{\beta}y_2, e^{-2\beta}z_2,\ldots\right)$$

Variation of energy with the infinitesimal deformation |β|<<1

Kinetic part

$$\langle\Psi_\beta|T|\Psi_\beta\rangle = \langle\Psi_0|T|\Psi_0\rangle + 4\beta^2\langle\Psi_0|T|\Psi_0\rangle + O(\beta^4)$$

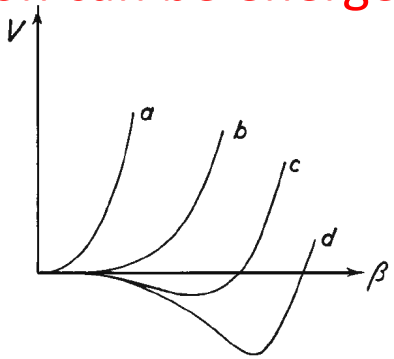
≧0 : Kinetic part disfavor deformation

Interaction part (direct interaction energy)

$$\langle\Psi_\beta|V_{\rm int}|\Psi_\beta\rangle = \langle\Psi_0|V_{\rm int}|\Psi_0\rangle + \frac{16\pi\beta^2}{5}\int r^3dr\int r'^3dr'V_2(r,r')\frac{d\rho_0(r)}{dr}\frac{d\rho_0(r')}{dr'} + O(\beta^4)$$

If < 0, deformation can be energetically favored

$$V_{\rm int}(\boldsymbol{r}-\boldsymbol{r'}) = \sum_{\ell,m} V_\ell(r,r')Y_{\ell,m}(\hat{\boldsymbol{r}})Y^*_{\ell,m}(\hat{\boldsymbol{r'}})$$



Mottelson, Nobel Lecture (1975)

Why atoms and nuclei differ?: Qualitative discussion

$$\frac{16\pi\beta^2}{5}\int r^3 dr \int r'^3 dr' V_2(r,r')\frac{d\rho_0(r)}{dr}\frac{d\rho_0(r')}{dr'}$$

$$V_{\rm int}(\boldsymbol{r}-\boldsymbol{r'}) = \sum_{\ell,m} V_\ell(r,r') Y_{\ell,m}(\hat{\boldsymbol{r}}) Y^*_{\ell,m}(\hat{\boldsymbol{r'}})$$

Atoms and nuclei

$\frac{d\rho(r)}{dr} \le 0$ for most region (except for complicated surface)

⇒ Sign of V_2 determines deformability

Nuclei

$$V_{\rm int}(\boldsymbol{r}) = -g\delta^{(3)}(\boldsymbol{r}) \le 0 \Rightarrow V_2 = -g\frac{\delta(r-r')}{rr'} < 0$$

Deformation may be energetically favored (c,d)

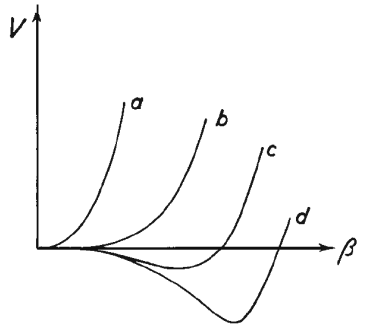
Atoms

$$V_{\rm int}(\boldsymbol{r}) = \frac{e^2}{r} \Rightarrow V_2(r,r') = \frac{4\pi}{5}\frac{r_<^2}{r_>^3} > 0$$

Deformation energetically disfavored (a,b)

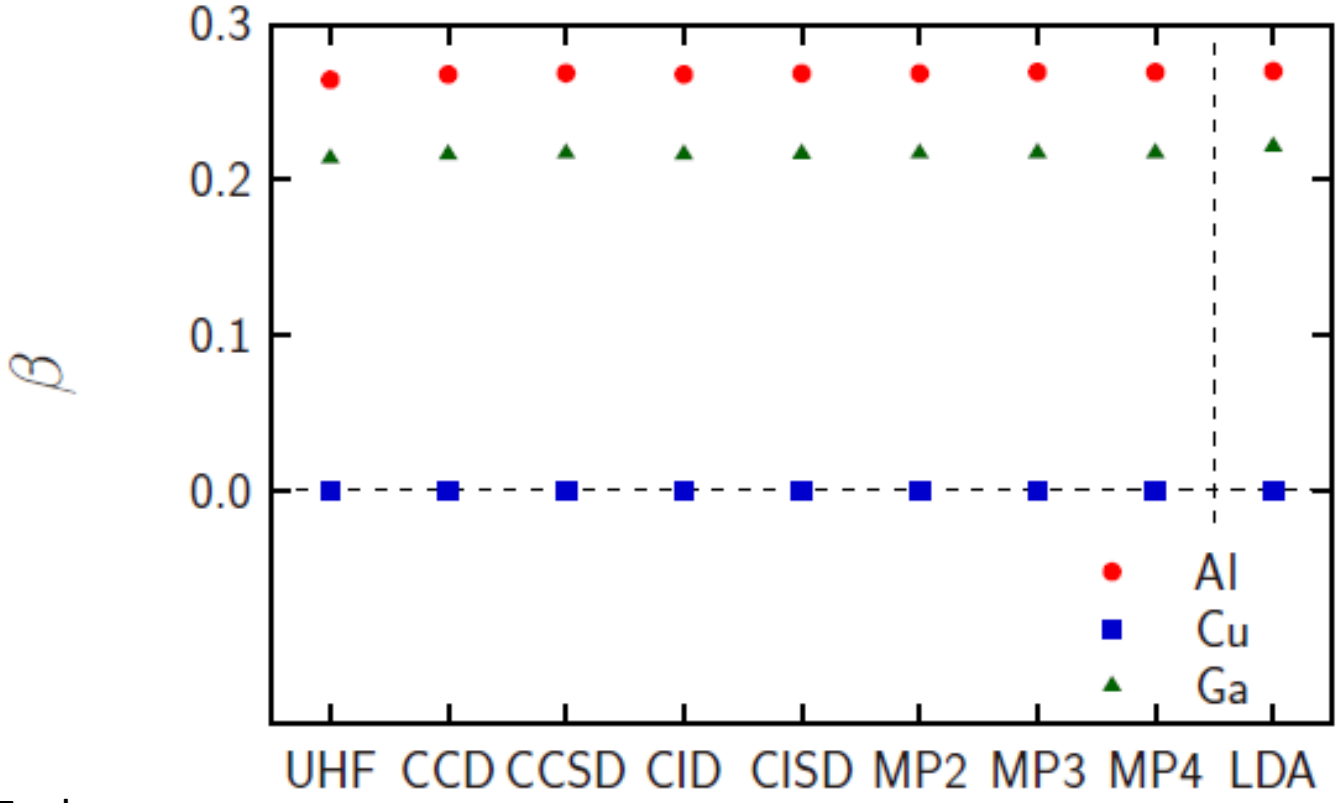
※ Surface (valence) region may show deformation because

1. Exchange-correlation part of int. energy is relevant.
2. $\frac{d\rho(r)}{dr} \le 0$ does not hold.



Mottelson, Nobel Lecture (1975)

Comparison between various different methods



6-31+G basis (dAug-CC-pV5Z basis and STO-3G basis essentially give the same results)

Hartree-Fock
- UHF: unrestricted Hatree-Fock

Coupled-cluster method
- CCD: with double excitations
- CCSD: with single-double excitations

Configuration Interaction
- CID: with double excitations
- CISD: with single-double excitations

Møller-Plesset perturbation theory
- MP2: second order
- MP3: third order
- MP4: fourth order (restricted to single, double, quadruple)

Density functional theory
- LDA: local density approximation PZ81

GAUSSIAN code for all the calculations

Conclusion

Motivation

- Can atoms be deformed?
- What is the difference between atoms and nuclei?

Main results (back-to-home message)

- Atoms can show large deformation $|\beta| \gtrsim 0.2 - 0.3$
- But they do not deform collectively.
- Atoms can be non-spherical only in the surface region via single-particle effect of the valence electron
- Many-body effects suppress deformations (screening)
- Repulsive Coulomb interaction ⇔ Attractive nuclear force

T. Naito, S. Endo, K. Hagino, Y. Tanimura,
arXiv:2009.05955

Repulsive long-range Coulomb force between electrons

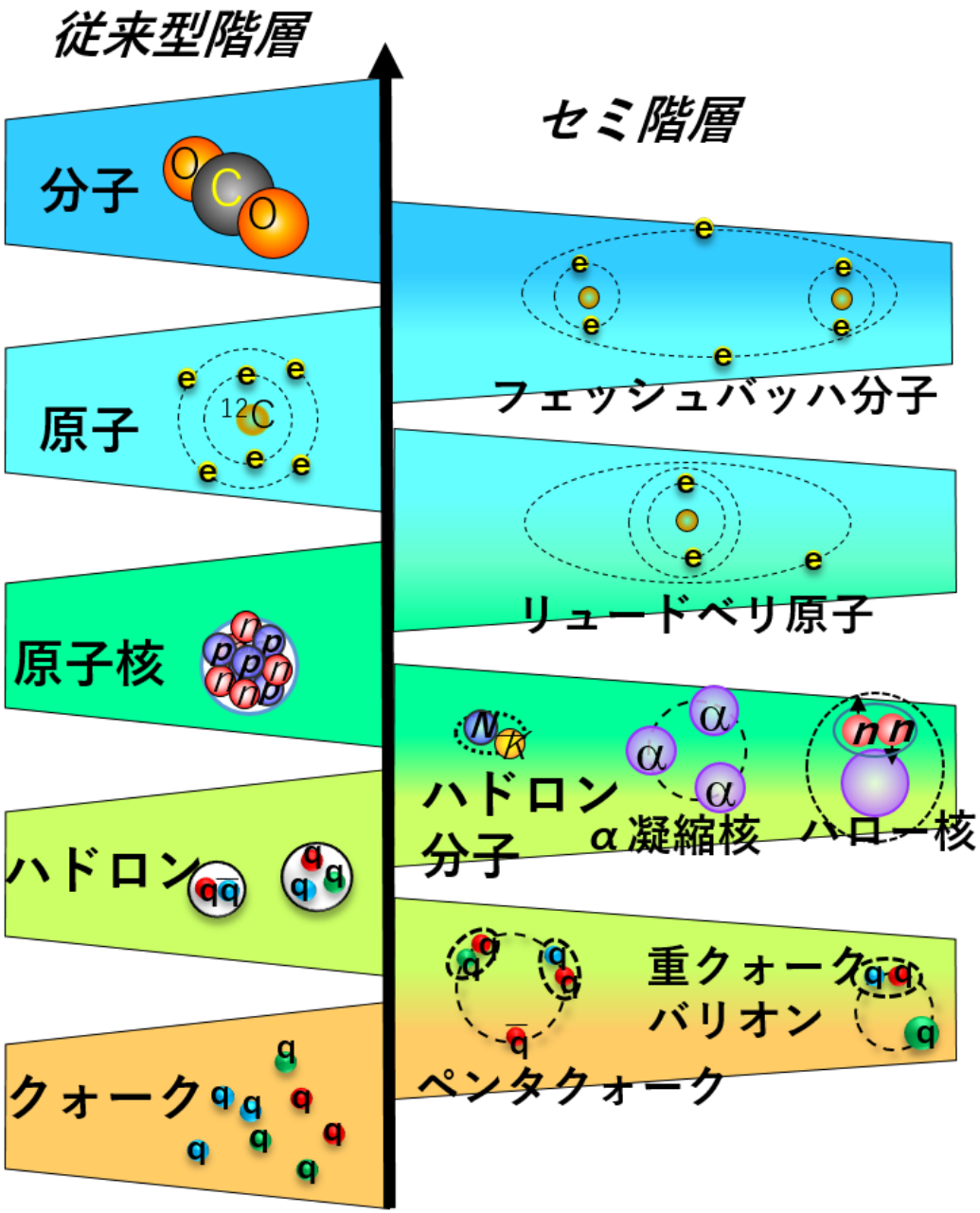
⇒ Deformation disfavored (screening effect)

⇒ No rotational spectra, No scissors mode,...

Attractive short-range nuclear force between nucleons

⇒ Deformation favored

⇒ Rotational spectra, scissors mode,...



Non-spherical effective-potential in atoms: H. A. Fertig, W. Kohn, PRA 62, 052511 (2000)

Relevant role of attractive p-n force in nuclear deformation: Dobaczewski, Nazarewicz, Skalski, Werner PRL. 60, 2254 (1988)