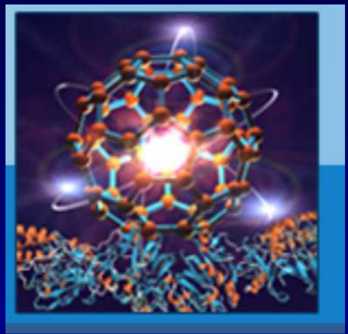
CHARGED METALLIC CLUSTERS



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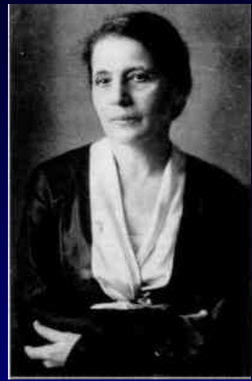
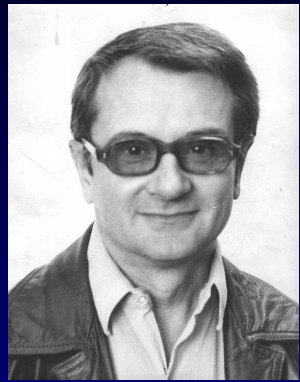
OUTLINE

Up to now we studied neutral clusters.

- Metallic behaviour
- Evaporation of neutral cluster and fission
- Macroscopic-microscopic approach. LDM of a charged cluster
- Coulomb energy
  - Nonmetals
  - Metals
- Fissility. Stability and metastability
- Dissociation energy and Fission barrier
- Shape isomer as a precursor
- Spheroidal shapes
- Hemispheroidal and cylindrical shapes
- Comparison of valley on PES: nuclei (superheavy, cluster decay, alpha decay) and atomic clusters (trimer decay)
- Conclusions

Liquid Drop Model + corrections



John William Strutt (Lord Rayleigh) (1842–1919), Phil. Mag. **14** (1878) 184: On the equilibrium of liquid conducting masses charged with electricity.

Niels Bohr, Nature **137** (1936) 344: LDM applied to atomic nuclei

Explained the induced nuclear fission:

- Lise Meitner and O. Frisch, Nature **143** (1939) 239
- N. Bohr and J. Wheeler, Phys. Rev. **56** (1939) 426

V.M. Strutinsky Nucl. Phys. **A 95** (1967) 420: shell+pairing corrections. Since 1967, the Macroscopic-Microscopic method successfully used in Nuclear Physics.

Adapted to Atomic Cluster Physics in the 80s and 90s.

Electrostatics – Coulomb & Poisson

Charles-Augustin de Coulomb (1736–1806)

Coulomb's law (vector form)

$$\mathbf{F} = \frac{1}{4\pi\varepsilon_0}\frac{q_1 q_2(\mathbf{r_1} - \mathbf{r_2})}{|\mathbf{r_1} - \mathbf{r_2}|^3} \qquad q_1 q_2 - \text{electric charges}$$

Coulomb's constant $1/4\pi\varepsilon_0 = 8.987 \times 10^9$ N·m^2·C^{-2} $\gg$ universal gravitational constant $G = 6.674 \times 10^{-11}$m^3·kg^{-1}·s^{-2}

"In conducting objects, the electric fluid expands on the surface of the body and does not penetrate into the interior (1786)"

Siméon-Denis Poisson (1781–1840)

Brilliant student and Professor (succeeding Fourier) at the École Polytechnique, Paris. Professors: Legendre; Lagrange; Laplace.

Poisson equation (of potential theory):

$$\nabla^2\phi = \Delta\phi = -4\pi\rho_e \qquad \rho_e - \text{charge density}$$

The Laplacean operator $\Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}$

Electrostatics – Gauß

Johann Carl Friedrich Gauß (1777–1855)

"Prince of Mathematicians": "Mathematics is the queen of sciences." Born in Braunschweig as the only son of a mason. The Duke of Braunschweig awarded him a fellowship to the Collegium Carolinum (now Technische Universität Braunschweig). Among his students in Götingen: Richard Dedekind, Bernhard Riemann, and Friedrich Bessel.

Contributed to: number theory; statistics; analysis; differential geometry; geodesy; electrostatics; astronomy; optics, etc.

Gauss's flux theorem

$$\oint_S \mathcal{E} \cdot d\mathbf{A} = 4\pi \int_V \rho_e(\mathbf{r}) d^3\mathbf{r}$$

The electric flux through a closed surface is proportional to the enclosed electric charge.
Differential form

$$\nabla \mathcal{E} = 4\pi \rho_e$$

where the electric field intensity $\mathcal{E} = -\nabla\phi$.

See: Walter Greiner, *Klassiche Elektrodynamik* (Harri Deutsch, Frankfurt am Main, 2008).

Few references

C. Bréchignac et al., Phys. Rev. Lett.: Photoionization of mass-selected K_n^+ ions: a test for the ionization scalling law, 63 (1989) 1368; Asymmetric fission of Na_n^{++} around the critical size of stability, 64 (1990) 2893; Dissipation Effects in Cluster Fission, 92 (2004) 083401

R. N. Barnett et al., Patterns and barriers for fission of charged small metal clusters, Phys. Rev. Lett., 67 (1991) 3058

C. Yannouleas, U. Landman: Shell-correction method for calculating the binding energy of metal clusters, Phys. Rev. B, 48 (1993) 8376; Electronic Entropy, Shell Structure, and Size-Evolutionary Patterns of Metal Clusters, Phys. Rev. Lett., 78 (1997) 1424

A.G. Lyalin, A.V. Solov'yov, W. Greiner, Comparative study of metal-cluster fission in Hartree-Fock and local-density approximations, Phys. Rev. A, 65 (2002) 043202; A.G. Lyalin, O.I. Obolensky, A.V. Solov'yov, W. Greiner, Dissociation and fission of small sodium and strontium clusters, Europ. Phys. J. D, 34 (2005) 93.

K. Seeger and R. E. Palmer, Fabrication of silicon cones and pillars using rough metal films as plasma etching masks, Appl. Phys. Lett. 74 (1999) 1627.

Charged atomic clusters

An ion is an atom or molecule which has lost or gained one or more electrons. The definition is extended to an atomic cluster with N atoms.

- Positively charged M_N^{z+} *cation*
- Negatively charged M_N^{z-} *anion*

From Greek: $\kappa\alpha\tau\alpha =$ down, $\alpha\nu\alpha =$ up.

The number of delocalized electrons left after ionization or electron attachment are

$$n_e = N - z \text{ for a cation}$$

$$n_e = N + z \text{ for an anion}$$

z is called the excess charge and N the size of the cluster.

In the most frequently studied fission (“Coulomb explosion”) process

$$\mathbf{M_N^{z+} \rightarrow M_{N_1}^{z_1+} + M_{N_2}^{z_2+} \; ; \quad n_e = n_{e1} + n_{e2} \; ; \quad z = z_1 + z_2}$$

the parent is doubly charged ($z = +2$) so that the fragments are single ionized: $z_1 = z_2 = 1$. The numbers of electrons are conserved:

$$N = N_1 + N_2 \; ; \quad z = z_1 + z_2 \; ; \quad n_e = N - z = n_{e1} + n_{e2} \; ; \quad n_{ei} = N_i - z_i$$

Charged clusters are produced by photoionization with laser beams, or by collision. The ionization energy of metals is generally much lower than the ionization energy of nonmetals hence metals will generally lose electrons to form cations while nonmetals will generally gain electrons to form anions.

Metallic behaviour

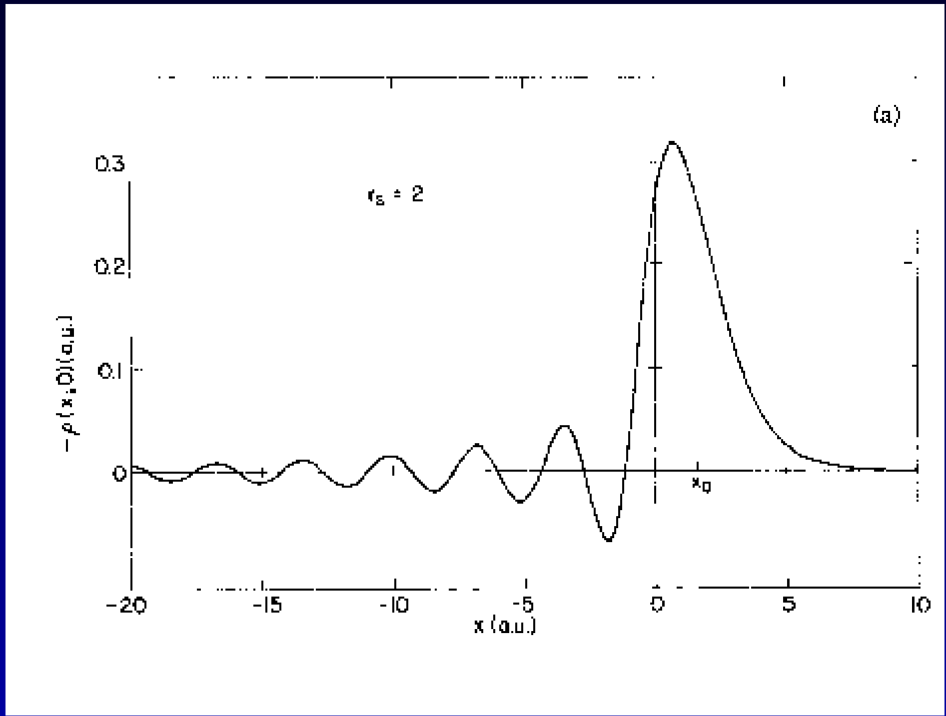


Fig 2(a) from N.D. Lang, W. Kohn, *Phys. Rev.* B **7** (1973) 3541. DFT result: profile of induced surface charge density $\rho(x;0)$ at a planar metal by a small static external charge.

The electronic screening charge distribution shows that the external field has been largely (about 90 %) screened out.

SCREENING OF EXTERNAL FIELD IS METALLIKE. The electronic polarizability of small metallic particles, investigated within the LDA applied to the spherical gellium model are leading to a similar result for the induced polarization density (see Fig 2 of W. Ekardt, *Phys. Rev. Lett.* **52** (1984) 1925).

This is a microscopic justification for the classical solving the electrostatic problems of metallic clusters within LDM by assuming a surface charge distribution instead of the homogeneously distributed one in the bulk.

Macroscopic-microscopic meth.

Successfully used in Nuclear Physics is suitable since delocalized conduction (valence) electrons of a metallic cluster form a Fermi liquid like the nucleons in an atomic nucleus.

- Macroscopic Liquid Drop Model: E_{LD}
- Single-particle shell model (SPSM): energy levels vs. deformation. E.g. two center shel model time consuming computations
- Shell + pairing correction method: $\delta E = \delta U + \delta P$
- Total deformation energy: $E_{def} = E_{LD} + \delta E$

for a given parametrization of the drop surface $\rho = \rho(z)$. The potential part of SPSM Hamiltonian should admit $\rho = \rho(z)$ as an equipotential surface.

D.N. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner,
Europhys. Lett. **79** (2007) 63001.
D.N. Poenaru, R.A. Gherghescu, I.H. Plonski, A.V. Solov'yov, W. Greiner,
Europ. Phys. J. D **47** (2008) 379-393. HIGHLIGHT PAPER.
D.N. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner,
Phys. Lett. A **372** (2008) 5448-5451.
R.A. Gherghescu, D.N. Poenaru, A.V. Solov'yov, W. Greiner,
Int. J. Mod. Phys. B **22** (2008) 4917-4935.

Liquid drop model of a charged cluster

M_N^{z+} will have $n_e = N - z$ delocalized electrons. Deformation energy

$$E_{LDM} = E - E^0 = (E_s - E_s^0) + (E_C - E_C^0)$$

$$= E_s^0(B_s - 1) + E_C^0(B_C - 1)$$

Spherical shapes: $E_s^0 = 4\pi R_0^2 \sigma = a_s n_e^{2/3} = 4\pi r_s^2 n_e^{2/3}$;
$E^0_{C-metal} = z^2e^2/(2R_0) = z^2e^2/(2r_s n_e^{1/3})$ for a surface distrib. of charge. The ratio to $E_C^0 = 3z^2e^2/(5R_0)$ for bulk homog. charge distr. is 5/6, i.e. 17 % smaller.
$\sigma -$ surface tension. $r_s -$ Wigner-Seitz radius.

Fissility

$$X = \frac{E_c^0}{2E_s^0} = \frac{e^2}{16\pi r_s^3 \sigma} \frac{z^2}{n_e} < 1 \; ; \quad n_e > n_c = \frac{e^2 z^2}{16\pi r_s^2 \sigma}$$

Within LDM VERY LIGHT CHARGED ATOMIC CLUSTERS ARE UNSTABLE.
For nuclear fission $X = E_c^0/(2E_s^0) \simeq [3e^2/(10r_0 a_s)](Z^2/A) < 1$ leading to $Z^2/A < (Z^2/A)_c \simeq 10 r_0 a_s/(3e^2)$ SUPERHEAVY NUCLEI ARE UNSTABLE.
Tables of material properties (r_s, σ or a_s, a_v, etc):

J.P. Perdew, Y. Wang, E. Engel, *Phys. Rev. Lett.* **66** (1991) 508.
U. Näher, S. Bjørnholm, S. Frauendorf, F. Garcias, C. Guet, *Phys. Rep.* **285** (1997) 245.

Coulomb energy - nonmetals

The electrostatic energy of a charge distribution with a density ρ_e in the volume V_n

$$E_C = \frac{1}{8\pi}\int_{V_\infty} \mathcal{E}^2(\mathbf{r})d^3r = \frac{1}{2}\int_{V_n} \rho_e(\mathbf{r})V(\mathbf{r})d^3r$$

where $\mathcal{E}$ is the electric field produced by this distribution of charges. The potential V is a solution of the Poisson equation $\Delta V(\mathbf{r}) = -4\pi\rho_e(\mathbf{r})$.

$$E_C = \frac{1}{2}\int_{V_n}\int \frac{\rho_e(\mathbf{r})\rho_e(\mathbf{r}_1)d^3rd^3r_1}{|\mathbf{r}-\mathbf{r}_1|} = -\frac{\rho_e^2}{12}\oint\oint \frac{(d\mathbf{S}_1\mathbf{r}_{12})(d\mathbf{S}_2\mathbf{r}_{12})}{r_{12}}$$

The double-volume integrals are transformed into double-surface integrals. By reducing the number of integrations from six to four, the computer running time becomes substantially shorter.

By using Gauss's theorem twice (with respect to $\mathbf{r}_1$ and $\mathbf{r}_2$), after expressing r_{12}^{-1} in terms of a double divergence, K.T.R. Davies, A.J. Sierk, *J. Comput. Phys.* **18** (1975) 311 have obtained the last relationship invariant under the interchange of $\mathbf{r}_1$ and $\mathbf{r}_2$.

Generalized for different charge densities: D.N. Poenaru et al., Comp. Phys. Comm. **16** (1978) 85, **19** (1980) 205.

Shape dependent B_s and B_C

B_s is proportional with surface area. For axially simmetric shapes

$$B_s = \frac{d^2}{2} \int_{-1}^{+1} \left[y^2 + \frac{1}{4} \left(\frac{dy^2}{dx} \right)^2 \right]^{1/2} dx$$

In cylindrical coordinates with -1, +1 intercepts on the symmetry axis $y = y(x)$ or $y_1 = y(x')$ is the surface equation. $d = (z'' - z')/2R_0$ – seminuclear length in units of R_0.

Assume uniform charge density in the bulk $\rho_{0e} = \rho_{1e} = \rho_{2e}$.

$$B_c = \frac{5d^5}{8\pi} \int_{-1}^{+1} dx \int_{-1}^{+1} dx' F(x, x')$$

$$\begin{aligned} F(x, x') \quad = \quad & \{ y y_1 [(K - 2D)/3] \cdot \\ & \left[2(y^2 + y_1^2) - (x - x')^2 + \frac{3}{2}(x - x') \left(\frac{dy_1^2}{dx'} - \frac{dy^2}{dx} \right) \right] + \\ & K \left\{ y^2 y_1^2/3 + \left[y^2 - \frac{x - x'}{2} \frac{dy^2}{dx} \right] \left[y_1^2 - \frac{x - x'}{2} \frac{dy_1^2}{dx'} \right] \right\} \} a_\rho^{-1} \end{aligned}$$

Complete elliptic integrals of the 1st and 2nd kind:

$K(k) = \int_0^{\pi/2} (1 - k^2 \sin^2 t)^{-1/2} dt$; $K'(k) = \int_0^{\pi/2} (1 - k^2 \sin^2 t)^{1/2} dt$.

$D = (K - K')/k^2$. Gauss-Legendre numerical quadratures.

Numerical quadratures

For Gauss-Legendre numerical quadratures of a single and double integral

$$\mathcal{I}_1 = \int_a^b dz f(z) \simeq \sum_{i=1}^{n} v_i f(z_i)$$

$$\mathcal{I}_2 = \int_a^b dz \int_a^b dz^{'} g(z, z^{'}) \simeq \sum_{i=1}^{n} \sum_{j=1}^{n} v_i v_j g(z_i, z_j)$$

we need the abscisas and weights (v_i, z_i) $i = 1, 2, ...n$ for the domain (a, b), which may be obtained from the tabulated (w_i, x_i) in the interval $(-1, +1)$ by using the relationships

$$v_j = \frac{b-a}{2} w_j \;\; ; \;\; z_j = \frac{b-a}{2} x_j + \frac{b+a}{2}$$

Coulomb energy - metals

The electrostatic energy of a charge distribution with a surface density σ_e

$$E_C[\sigma_e] = \frac{1}{2}\int\int \frac{\sigma_e(\mathbf{r})\sigma_e(\mathbf{r}_1)d^2\mathbf{S}d^2\mathbf{S_1}}{|\mathbf{r}-\mathbf{r}_1|}$$

The distribution σ_e on the surface is obtained by minimizing the energy under the constraint

$$Q = ze = \int \sigma_e(\mathbf{r})d^2\mathbf{S}$$

i.e. the functional derivative

$$\frac{\delta(E-\lambda Q)}{\delta\sigma_e} = \frac{1}{2}\int \frac{\sigma_e(\mathbf{r}_1)d^2\mathbf{S_1}}{|\mathbf{r}-\mathbf{r}_1|} - \lambda = 0$$

H.J. Krappe, *Z. Phys. D* **23** (1992) 269

H. Koizumi, S. Sugano, Y. Ishii, *Z. Phys. D* **28** (1993) 223

Stability of a slightly distorted sphere I

For *axially symmetric* shapes, the deformations parameters $\{\alpha_n\}$ may be defined by expanding the radius in a series of Legendre polynomials, leading to

$$B_C \simeq 1 - 5 \sum_n \frac{(n-1)}{(2n+1)^2} \alpha_n^2$$

$$B_s \simeq 1 + \frac{1}{2} \sum_n \frac{(n-1)(n+2)}{2n+1} \alpha_n^2$$

The stability of nuclear shape relative to small quadrupolar deformations, α_2, can be studied by developing the relative deformation energy $\xi = (E - E^0)/E_s^0$ in terms of α_2 around $\alpha_2 = 0$. To a very good approximation, one has

$$\xi = B_s - 1 + 2X(B_C - 1) \simeq (2/5)\alpha_2^2(1 - X)$$

Stability of slightly distorted sphere II

When $X < 1$ the deformation energy increases with the deformation parameter α_2 and there is a driving force toward the potential minimum at $\alpha_2 = 0$ - the equilibrium shape.

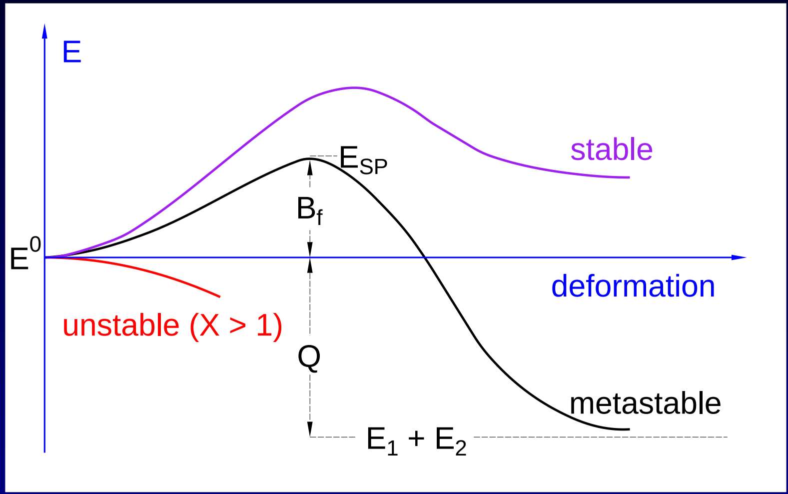
Within LDM any nucleus or charged atomic cluster with $X > X_{cr} = 1$, has no chance to survive because its energy is decreasing continuously with increasing deformation parameter.

For metallic cluster, apart the constant factor (1/2 instead of 3/5) an almost identical relationship was obtained:

$$E_C = \frac{z^2 e^2}{2R_0}\left(1 - \frac{\alpha_2^2}{5} - \frac{4\alpha_2^3}{105} + \frac{53\alpha_2^4}{245}\right)$$

W. A. Saunders, Phys. Rev. A **46** (1992) 7028.

Stable and metastable states



$X < 1$; $Q < 0$; $B_f > 0$ stable
$X < 1$; $Q > 0$; $B_f > 0$ metastable
$X \geq 1$; $B_f \leq 0$ unstable

The released (dissociation) energy $Q = E^0 - (E_1 + E_2)$

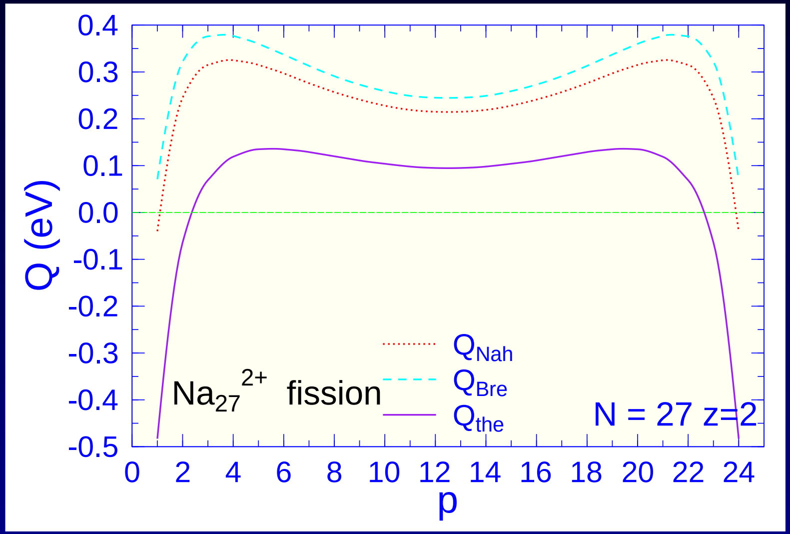
When $Q > 0$ (exothermic reaction) the spontaneous fission is possible.
For $Q < 0$ (endothermic reaction) – induced fission: one has to excite the charged cluster to surpass the barrier.

Fission barrier $B_f = E_{SP} - Q$; SP – saddle point

Fussion barrier approx. $B_C = B_f + Q$

In a metastable state the two fragments are temporarily held together by the potential barrier. There is a finite probability for the penetration of the barrier by the quantum-mechanical tunneling effect.

LDM Dissociation energy (Q-value)



$p = n_{e1}$

Surface component

$Q_s = a_s[n_e^{2/3} - n_{e1}^{2/3} - n_{e2}^{2/3}]$

Coulomb component

$$Q_C = \frac{e^2}{r_s}\left[\frac{\alpha}{n_e^{1/3}} - \frac{\beta}{n_{e1}^{1/3}} - \frac{\gamma}{n_{e2}^{1/3}}\right]$$

where $\alpha = \frac{z^2}{2} - \frac{z}{8}$, $\beta = \frac{z_1^2}{2} - \frac{z_1}{8}$

$\gamma = \frac{z_2^2}{2} - \frac{z_2}{8}$; $Q_{Bre} = Q_s + Q_C$

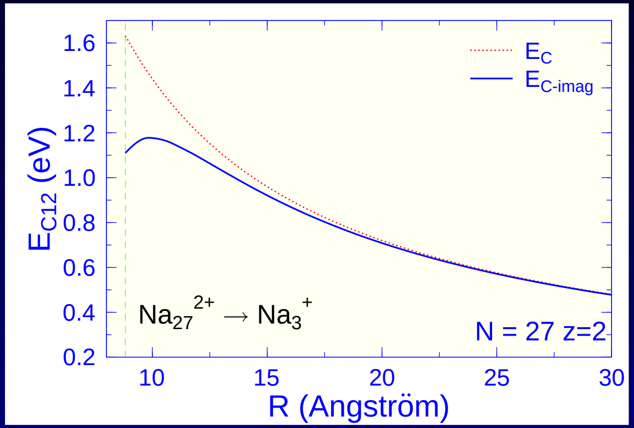
C. Bréchignac et al. *Phys. Rev. B* **44** (1991) 11386.

$$Q_{Nah} = E(n_e, z) - [E(n_{e1}, z_1) + E(n_{e2}, z_2)] \, ; \quad E(n_e, z) = E_s(n_e) + z(z+2k-1)e^2/(2R_0)$$

because the total energy needed to remove z electrons from a neutral cluster is $zW_b + z(k - 1/2 + z/2)e^2/R_0$. Since the volume and the charge are conserved, the work function W_b and the cohesive energy a_v are not contributing to Q. The classical value of $k = 0.5$ but the experiment confirmed by DFT gives $k = 0.4$.

$$Q_{the} = E^0 - (E_1 + E_2) \, ; \quad E_i = E_{si} + E_{Ci}$$

Coulomb Barrier - image charge



$$E_C = \frac{z_1 z_2 e^2}{R}$$

$$E_{C-Nah} = \frac{z_1 z_2 e^2}{R} - \frac{z_2 e^2 R_1^3}{2R^2(R^2 - R_1^2)}$$

$$R_1 > R_2$$

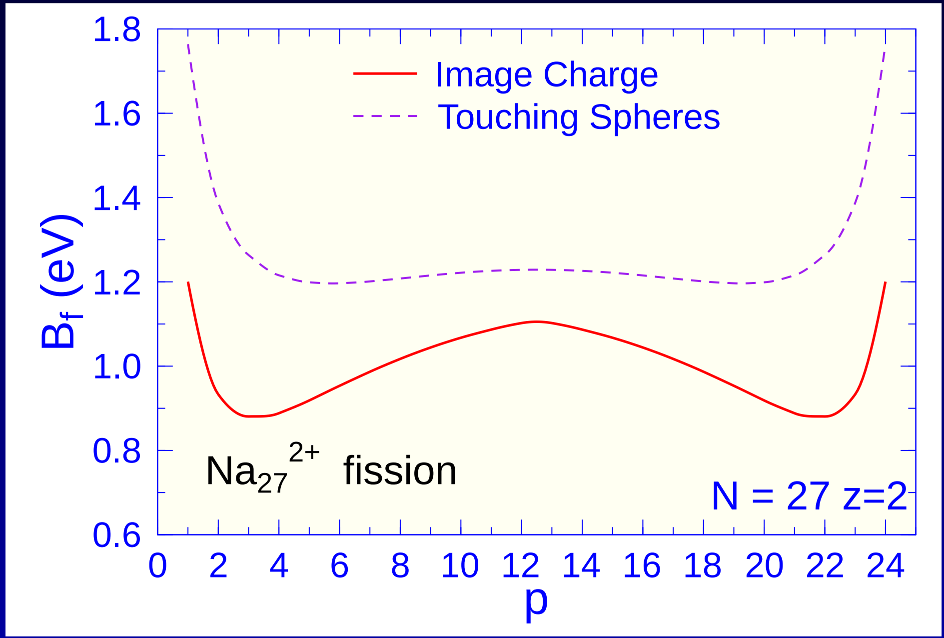
Interaction energy for separated fragments. $Na_{27}^{2+} \rightarrow Na_3^+ + Na_{24}^+$ fission.

Classical image charge model. A point charge q_2 at the distance R from the center of the conducting sphere with a radius R_1 and the charge q_1 will produce two image charges: one at the center of the sphere ($q_2 R_1/R$) and the other ($-q_2 R_1/R$) at the distance R_1^2/R. The total force between the point charge and the charged conducting sphere is $F(R) = \frac{q_1 q_2}{R^2} + \frac{q_2^2 R_1}{R^2} - \frac{q_2^2 R_1}{R(R-R_1^2/R)^2}$ i.e. a sum of monopole-monopole and monopole-dipole interactions.

The total energy $E_{C-Nah}(R) = \int_\infty^R F(R)dR = \frac{q_1 q_2}{R} - \frac{q_2^2 R_1^3}{2R^2(R^2 - R_1^2)}$

The maximum of the interaction energy is placed at the distance $R = R_m = R_t + 2$ Å. When there is no solution for $R_m > R_t$, one takes $R_m = R_t$.

LDM fission barrier



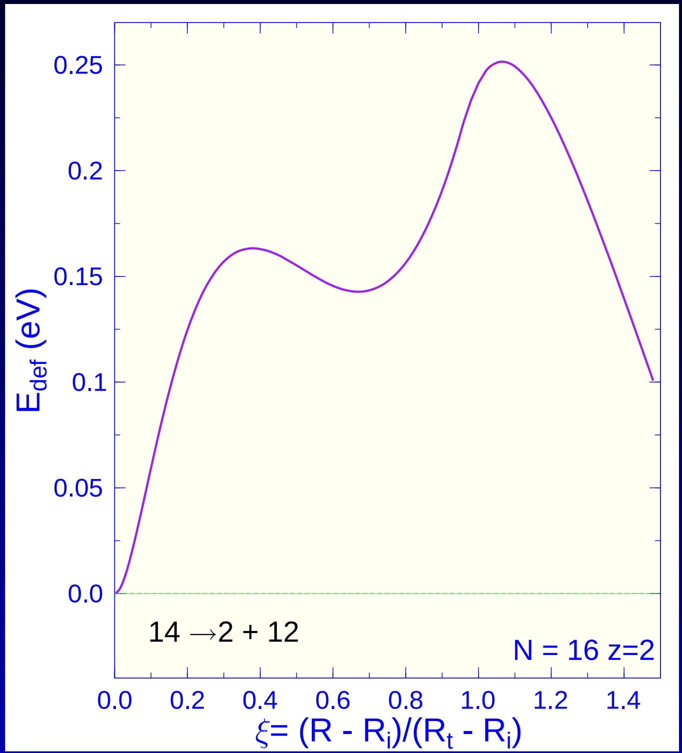
Low barrier means an increased yield.

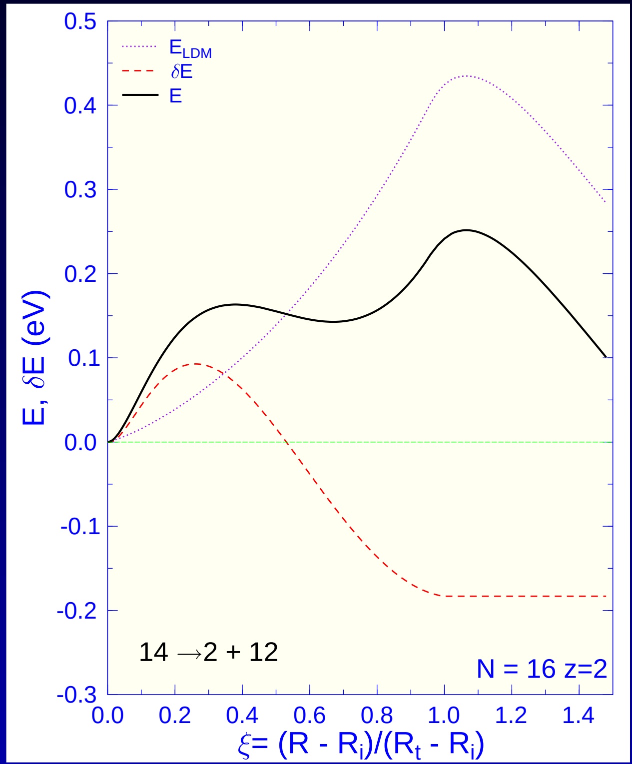
$p = n_{e1}$

Na_{27}^{2+} fission.

The low n_{e1} channels (in particular the trimer Na_3^+ fragment) are promoted by the LDM!

Preformation of Na_3^+





LDM + shell effects. Here we have $Na_{16}^{2+} \rightarrow Na_3^+ + Na_{13}^+$ A two-humped barrier was also obtained for $Na_{10}^{2+} \rightarrow Na_3^+ + Na_7^+$ by

R.N. Barnett, U. Landman, G. Rajagopal, *Phys. Rev. Lett.* **67** (1991) 3058

by A. Vieira, C. Fiolhais, *Phys. Rev. B* **57** (1998) 7352.

and by P. Blaise et al., *Phys. Rev. Lett* **87** (2001) 063401.

The shape isomeric state was interpreted as a precursor (kind of preformed trimer Na_3^+) prior eventual separation.

CHARGED SPHEROIDAL Na CLUSTER

Spherical shape. Material properties

Energies in eV for Na (monovalent) charged spherical cluster Na_N^{z+} with $N - z = n_e$ delocalized electrons

$$E_v^0 = -2.252 n_e$$

Volume energy is proportional to the volume.

$$E_s^0 = 0.541 n_e^{2/3}$$

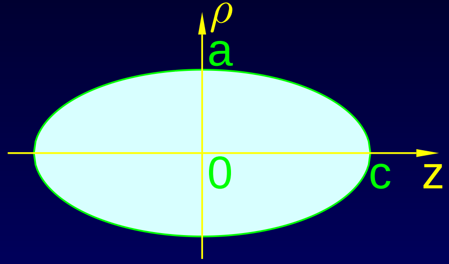
Surface energy is proportional to the surface area and to the surface tension σ: $E_s^0 = 4\pi R_0^2 \sigma = 4\pi r_s^2 \sigma n_e^{2/3}$, $4\pi r_s^2 \sigma = 0.541$ eV

Coulomb (electrostatic energy) of a metallic cluster

$$E_C^0 = \frac{z^2 e^2}{2R_0} = \frac{z^2 e^2}{2 r_s n_e^{1/3}}$$

$e^2/2 = 7.1998259$ eV·Å.

Spheroidal deformation



Spheroidal deformation $\delta < 0$ oblate

$\delta > 0$ prolate

(K.L. Clemenger, PhD Thesis, Univ. of California, Berkeley, 1985)

Dimensionless semiaxes (units of $R_0 = r_s n_e^{1/3}$)

$$a = \left(\frac{2-\delta}{2+\delta}\right)^{1/3} \quad ; \quad c = \left(\frac{2+\delta}{2-\delta}\right)^{2/3}$$

$$\frac{a}{c} = \frac{2-\delta}{2+\delta} = a^3$$

Volume conservation: $a^2 c = 1$.

Energies of spheroidal shapes

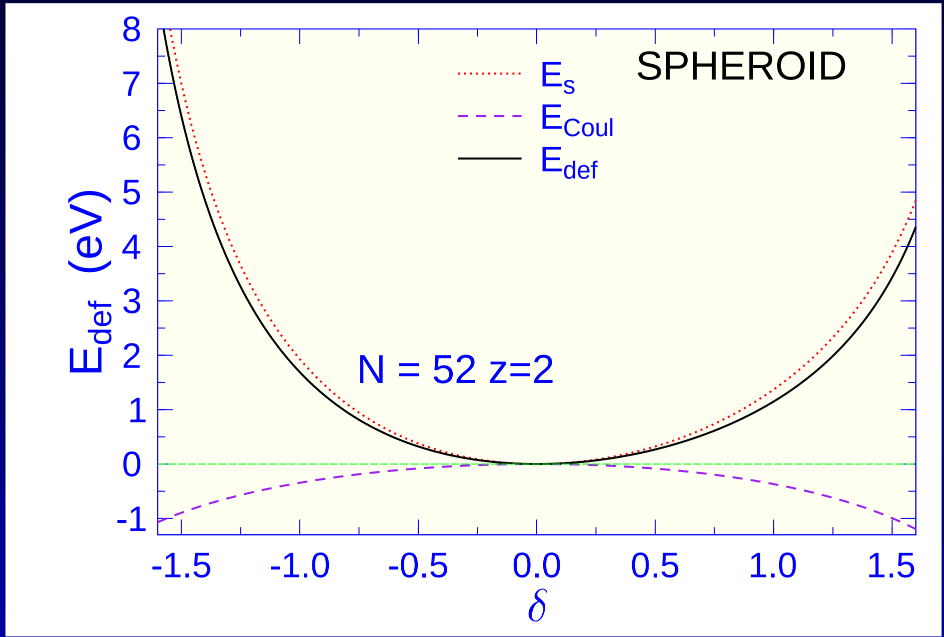
Oblate ($a > c$, eccentricity $\epsilon = \sqrt{a^2/c^2 - 1}$):

$$B_s = \frac{a}{2}\left(a + \frac{c}{2\epsilon}\ln\frac{a + c\epsilon}{a - c\epsilon}\right) \quad ; \quad B_C = \frac{1}{c\epsilon}\arctan\epsilon$$

Prolate ($a < c$, eccentricity $\epsilon = \sqrt{1 - a^2/c^2}$):

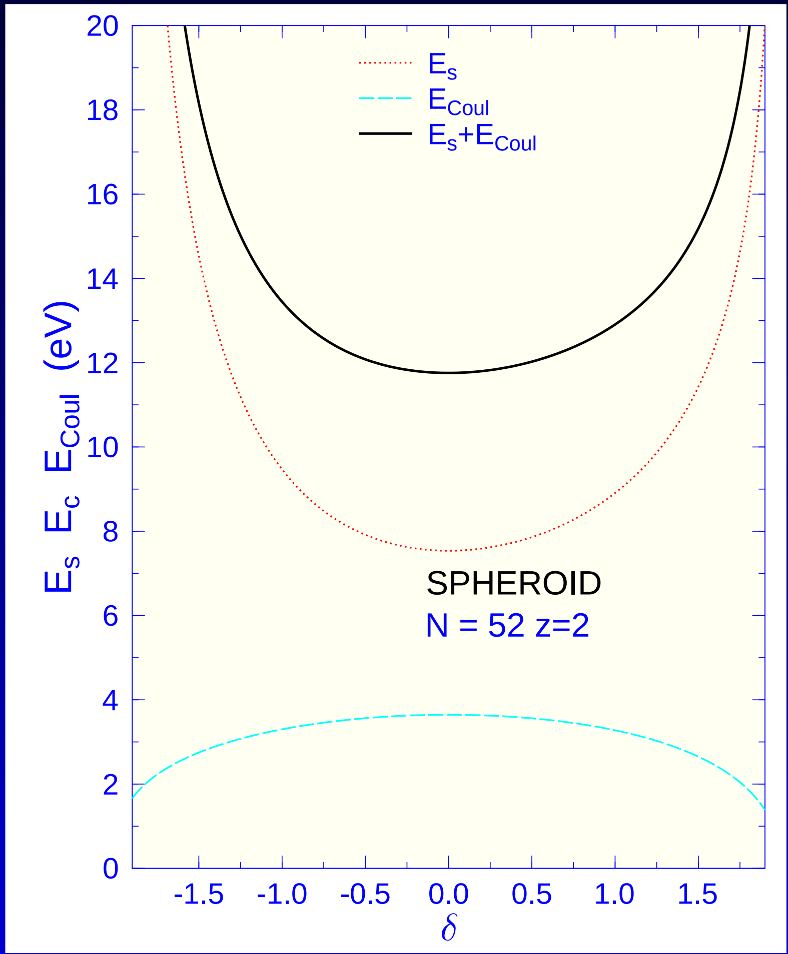
$$B_s = \frac{a}{2}\left(a + \frac{c}{\epsilon}\arcsin\epsilon\right) \quad ; \quad B_C = \frac{1}{2c\epsilon}\ln\frac{1 + \epsilon}{1 - \epsilon}$$

LDM def. energies of Na_{54}^{2+}



Doubly charged spheroidal Na cluster with 54 atoms and 52 delocalized electrons.

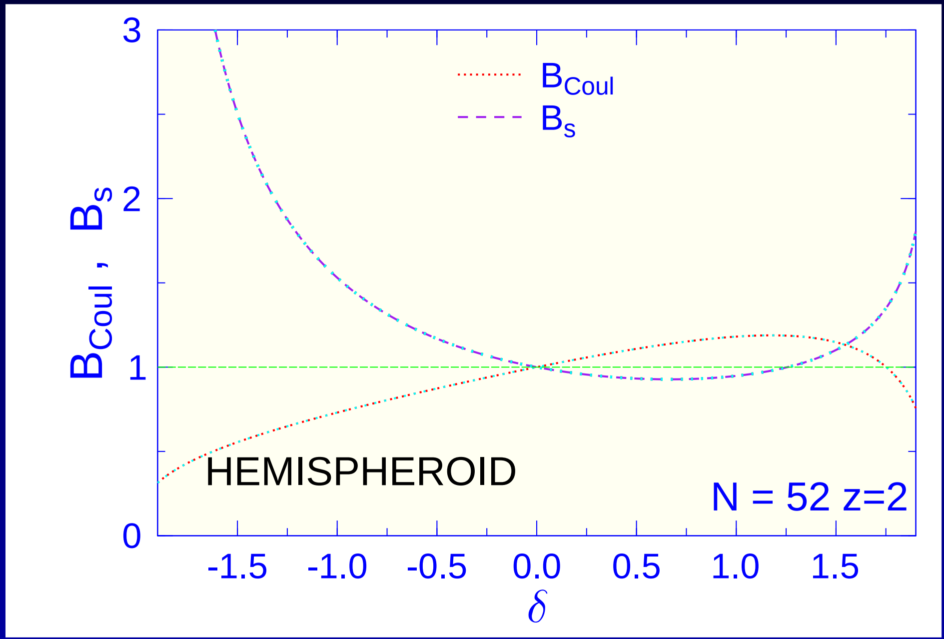
LDM energies of Na_{54}^{2+}



Doubly charged spheroidal Na cluster with 54 atoms and 52 delocalized electrons.

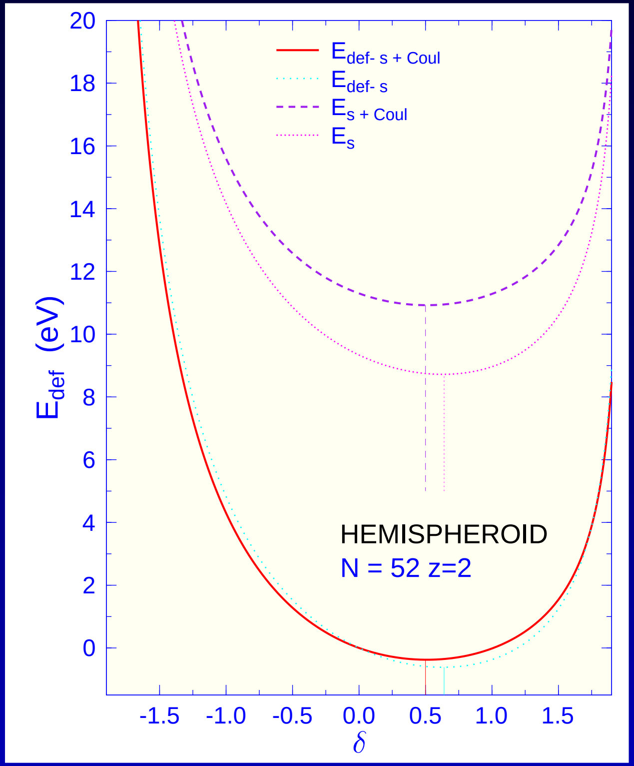
CHARGED HEMISPHEROIDAL and CYLINDRICAL Na CLUSTER

LDM def. en. of hemispheroidal Na_{54}^{2+}



Doubly charged hemi-spheroidal Na cluster with 54 atoms and 52 delocalized electrons.

LDM energies of hemisperoidal Na_{54}^{2+}



Doubly charged hemispheroidal Na cluster with 54 atoms and 52 delocalized electrons.

With Coulomb energy the minimum moved to smaller deformation ($\delta = 0.50$ instead of $\delta = 0.64$. Also the stability decreases (the minimum of the absolute value of energy is increased.

Cylindrical cluster

The body taken as a reference corresponds to $a_0 = 1$ and $c_0 = 2$, for which

$$V_0 = \frac{4\pi R_0^3}{3} = \pi R_{c0}^3 a_0^2 c_0 = 2\pi R_{c0}^3$$

$R_{c0} = \left(\frac{2}{3}\right)^{1/3} R_0$ and the volume conservation ($\pi R_{c0}^3 a^2 c = V_0$) leads to $a^2 c = 2$

Deformation parameter: $\xi = c/a$ so that $\xi_0 = 2$. For a given ξ we have

$a = \left(\frac{2}{\xi}\right)^{1/3} \quad ; \quad c = a\xi$

The surface area $S = 2\pi R_{c0}^2 a^2 + 2\pi R_{c0}^2 ac = 2\pi R_{c0}^2 a(a+c)$ so that

$E_{sc} = S\sigma = \frac{1}{2}\left(\frac{2}{3}\right)^{2/3} a_s N^{2/3} = a(c+a)E_{sc}^0/3 = \frac{2^{2/3} E_{sc}^0}{3} \frac{1+\xi}{\xi^{2/3}}$ When $a = 1$ and $c = 2$

$E_{sc}^0 = \frac{3}{2}\left(\frac{2}{3}\right)^{2/3} a_s N^{2/3} = \left(\frac{3}{2}\right)^{1/3} E_s^0$

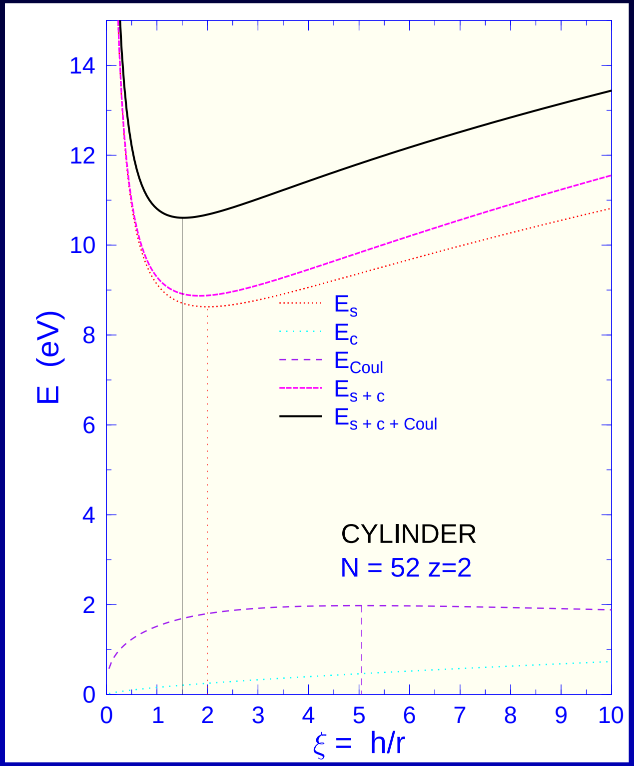
Since $\rho = a$, $\rho' = \rho'' = 0$, the integrated curvature $\mathcal{K} = \pi R_{c0} c$ and the curvature energy $E_{cc} = \mathcal{K}\gamma_c = \frac{1}{4}\left(\frac{2}{3}\right)^{1/3} a_c N^{1/3} c = cE_{cc}^0/2 = \frac{E_{cc}^0}{2^{2/3}}\xi^{2/3}$

$E_{cc}^0 = \frac{1}{2}\left(\frac{2}{3}\right)^{1/3} a_c N^{1/3} = \frac{E_c^0}{12^{1/3}}$

The Coulomb energy $E_{Coul-c} = \frac{q^2}{6\pi}\frac{Z^2}{R_{c0}} \int_0^c dz \int_0^c dz' F(z, z')$

From numerical quadrature we obtain $E_{Coul-c}^0 = 0.494095855522 E_C^0$ i. e. the cylinder with $h = 2a$ has an energy smaller than the sphere by about 51 %.

LDM energies of a cylindrical cluster



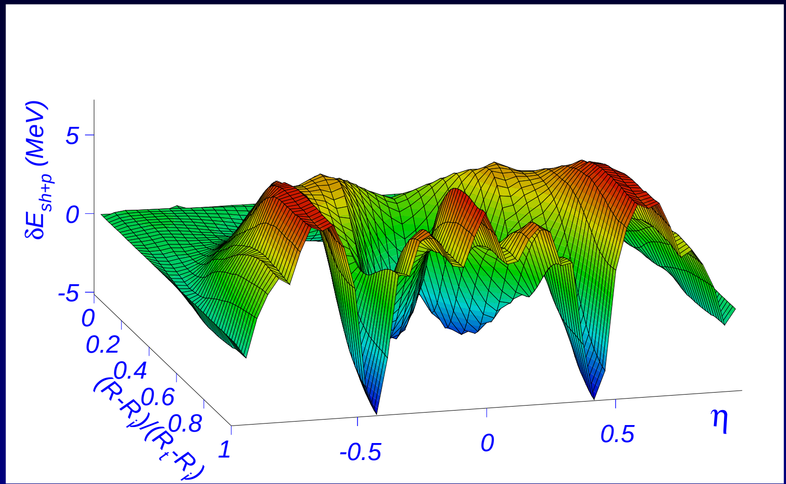
Doubly charged cylindrical Na cluster with 54 atoms and 52 delocalized electrons.

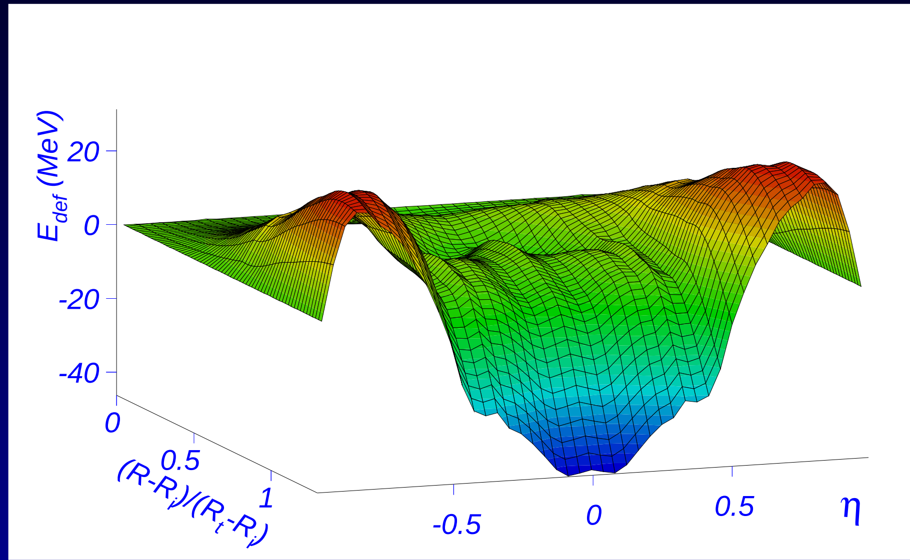
With Coulomb energy (max. at $\xi = 5.05$) the minimum moved to smaller deformation ($\xi = 1.50$ instead of $\xi = 2.00$. Also the stability decreases (the minimum of the absolute value of energy is increased).

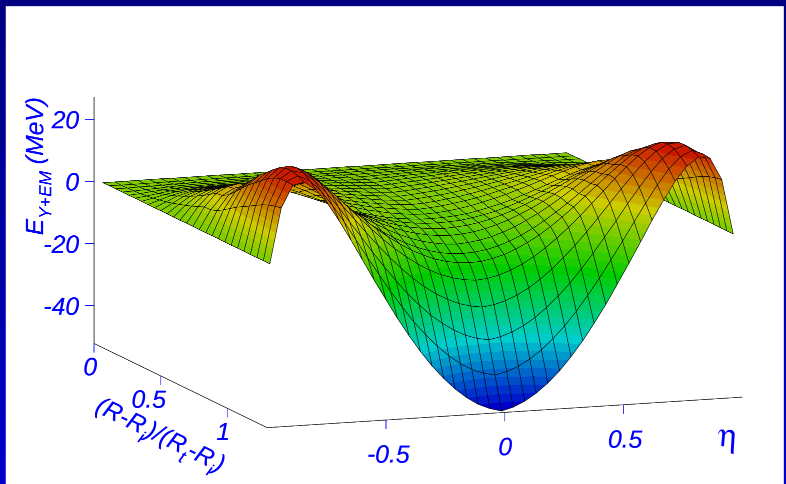
PES FOR FISSION AND FUSION OF NUCLEI AND METALLIC CLUSTERS:

- SUPERHEAVY NUCLEI
- CLUSTER RADIOACTIVITY
- α-DECAY
- CLUSTER FISSION

^{294}118: E_{Y+EM}, $\delta E_{shell+pair}$, E_{def}







normalized separation distance

$(R - R_i)/(R_t - R_i)$

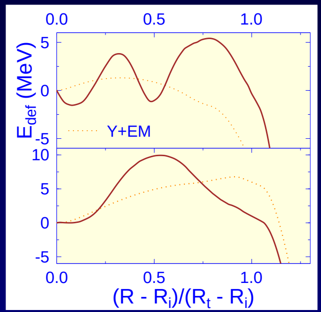
mass asymmetry

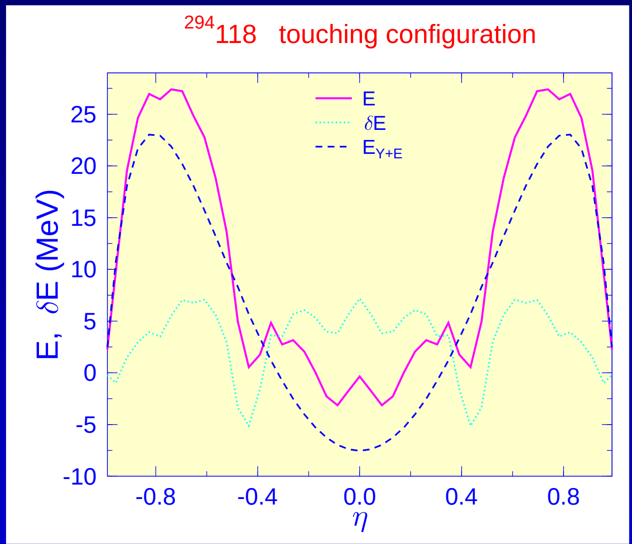
$\eta = (A_1 - A_2)/(A_1 + A_2)$

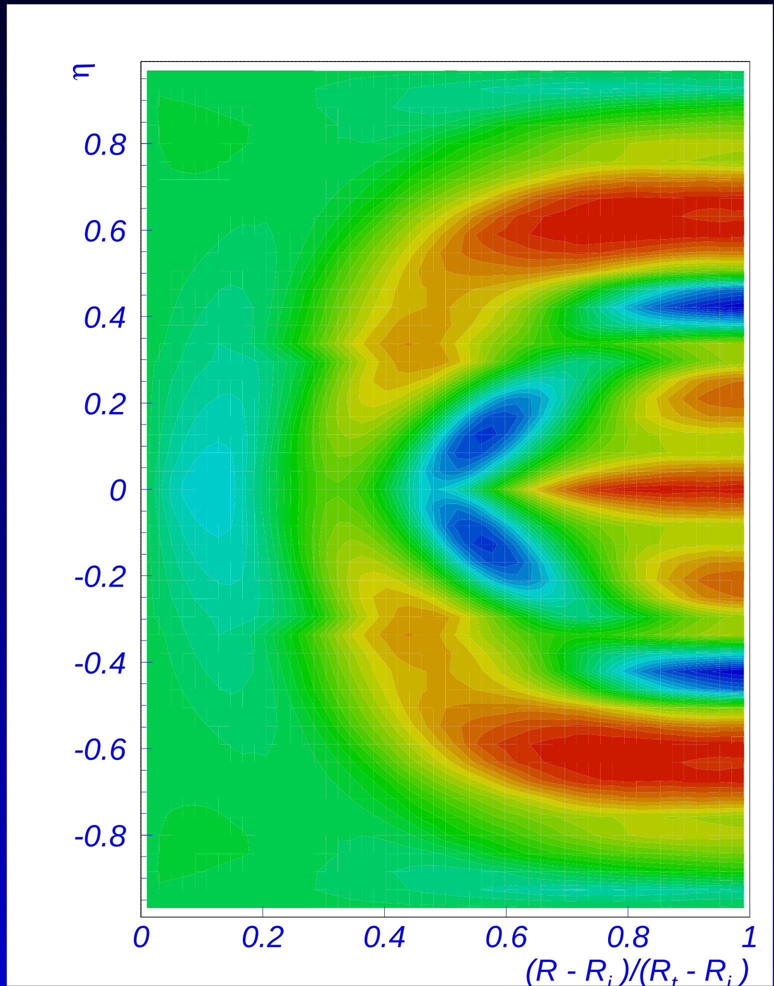
Fusion valley for production : ^{86}Kr$_{50}$ + ^{208}Pb at $\eta = 0.41$

D.N. Poenaru, I.H. Plonski, R.A. Gherghescu, W. Greiner, *J. Phys. G: Nucl. Part. Phys.* **32** (2006) 1223.

^{294}118 barr., touching, contour







$\delta E_{shell+pairing}$ *contour plot*

in the plane $(R - R_i)/(R_t - R_i), \eta$

Exp SH nuclei — Cold Valleys

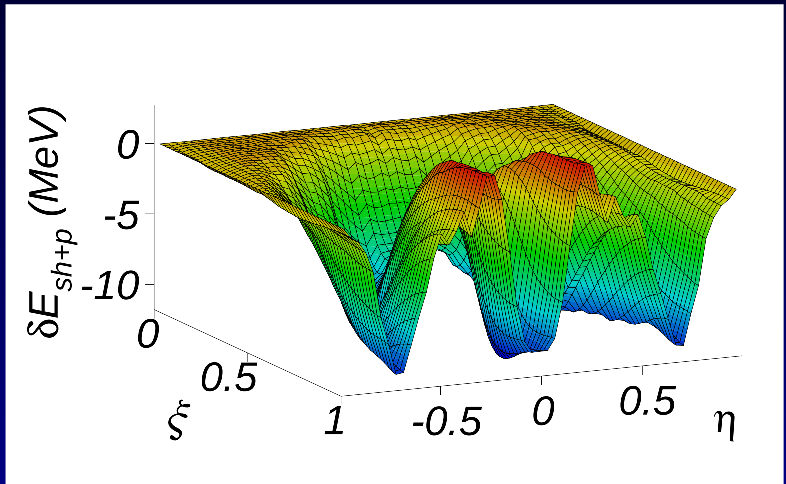
Element			Projectile			Target		
Z	Symbol	Name		N_t	Z_t		N_p	Z_p
107	Bh	Bohrium	^{54}Cr	30	24	^{209}Bi	126	83
108	Hs	Hassium	^{58}Fe	32	26	^{208}Pb	126	82
109	Mt	Meitnerium	^{58}Fe	32	26	^{209}Bi	126	83
110	Ds	Darmstadtium	^{62}Ni	34	28	^{208}Pb	126	82
111	Rg	Roentgenium	^{64}Ni	36	28	^{209}Bi	126	83
112	Cn	Copernicum?	^{70}Zn	40	30	^{208}Pb	126	82
113			^{70}Zn	40	30	^{209}Bi	126	83

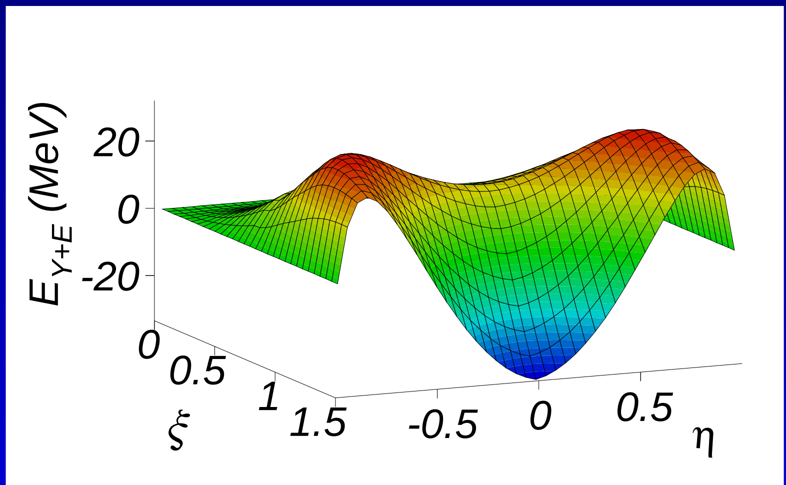
GSI: Gottfried Münzenberg, Sigurd Hofmann et al. 1981, 1984, 1994, 1996.

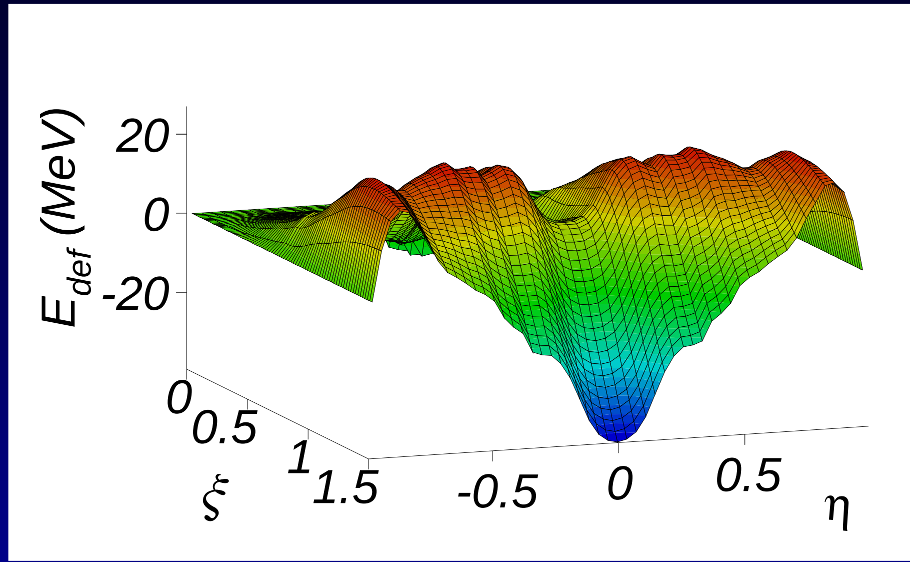
RIKEN (Z=113): Kosuke Morita et al. 2004.

Magic numbers of neutrons: 2, 8, 20, 28, 50, 82, 126

^{242}**Cm:** E_{Y+EM}, $\delta E_{shell+pair}$, E_{def}







separation distance

$\xi = (R - R_i)/(R_t - R_i)$

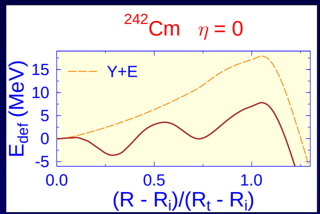
mass asymmetry

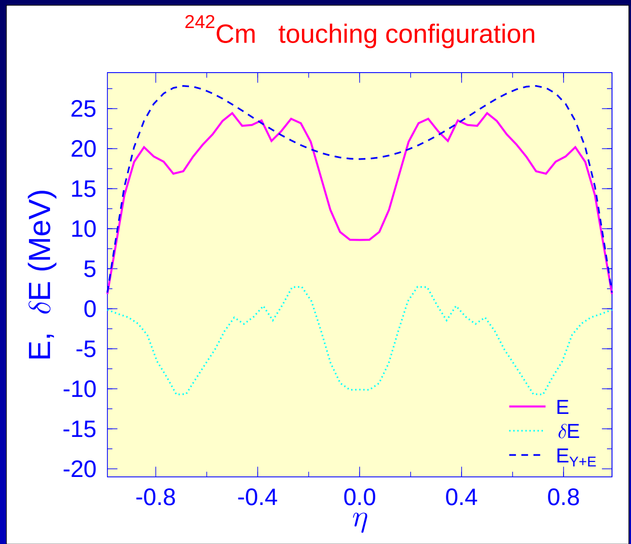
$\eta = (A_1 - A_2)/(A_1 + A_2)$

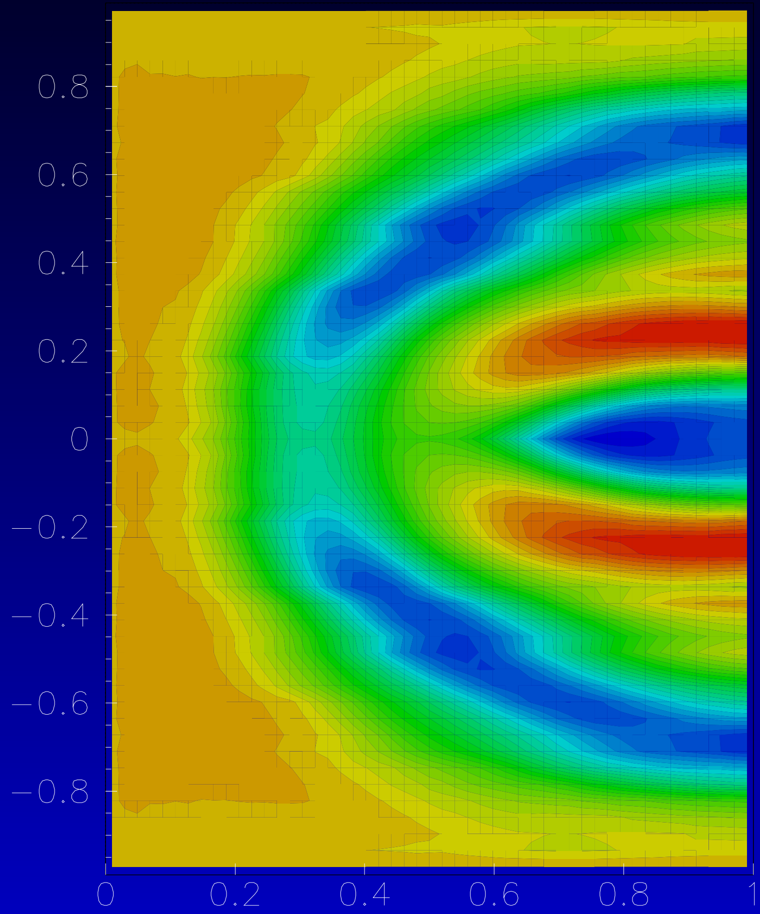
^{34}Si radioactivity + ^{208}Pb daughter valley at $\eta = 0.72$

D.N. Poenaru, R.A. Gherghescu, W. Greiner, *Phys. Rev.* **C 73** (2006) 014608.

^{242}Cm barr., touching, contour







$\delta E_{shell+pairing}$ *contour plot*

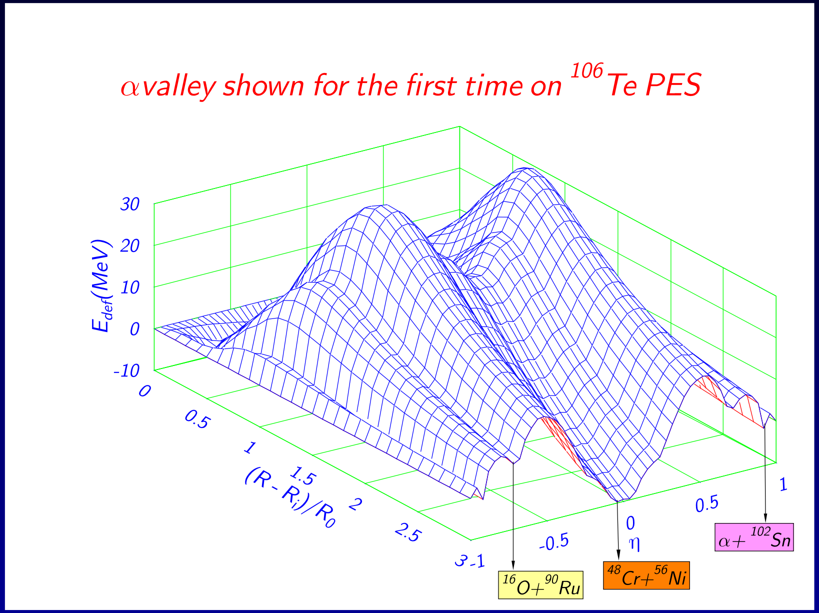
in the plane $\xi = (R - R_i)/(R_t - R_i), \eta$

CLUSTER RADIOACTIVITIES - confirmed

Cluster	Z_e	N_e	Parent - Daughter	Z_d	N_d	Cluster	Z_e	N_e	Parent - Daughter	Z_d	N_d
^{14}C	6	8	^{221}Fr	81	126	^{14}C	6	8	^{221}Ra	82	125
			^{222}Ra	82	126				^{223}Ra	82	127
			^{224}Ra	82	128				^{226}Ra	82	130
			^{223}Ac	83	126				^{225}Ac	83	128
^{20}O	8	12	^{228}Th	82	126	^{23}F	9	14	^{231}Pa	82	126
^{22}Ne	10	12	^{230}U	82	126	^{24}Ne	10	14	^{231}Pa	81	126
^{24}Ne	10	14	^{232}U	82	126				^{233}U	82	127
			^{234}U	82	128				^{235}U	82	129
^{25}Ne	10	15	^{233}U	82	128	^{25}Ne	10	15	^{235}U	82	128
^{26}Ne	10	16	^{234}U	82	126	^{28}Mg	12	16	^{234}U	80	126
^{28}Mg	12	16	^{236}U	80	128				^{236}Pu	82	126
			^{238}Pu	82	128	^{30}Mg	12	18	^{236}U	80	126
^{30}Mg	12	18	^{238}Pu	82	126	^{32}Si	14	18	^{238}Pu	80	126
^{34}Si	14	20	^{242}Cm	82	126						

Since 1984 experiments performed in: Oxford, Moscow, Orsay, Argonne, Berkeley, Dubna, Livermore, Geneva, Milano, Vienna, Beijing.

Alpha valley of ^{106}Te

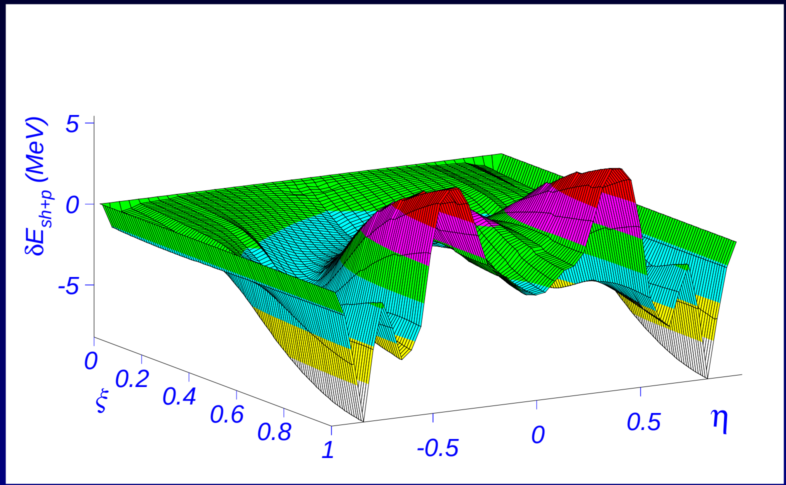


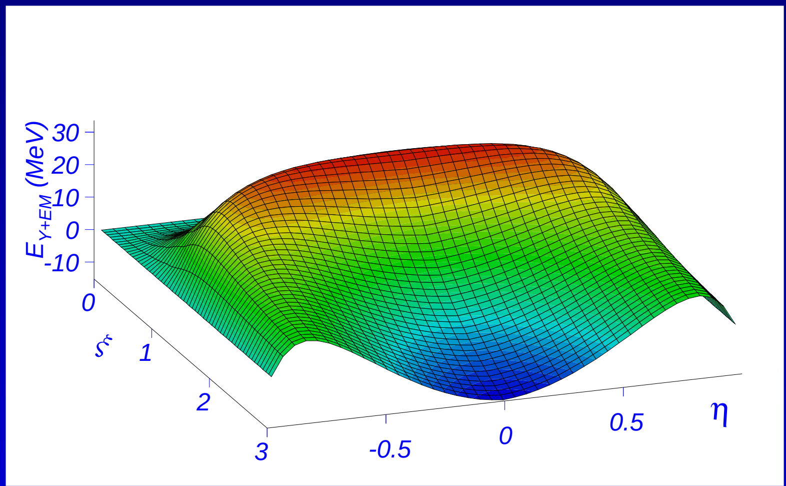
mass asymmetry $\eta = (A_1 - A_2)/(A_1 + A_2)$

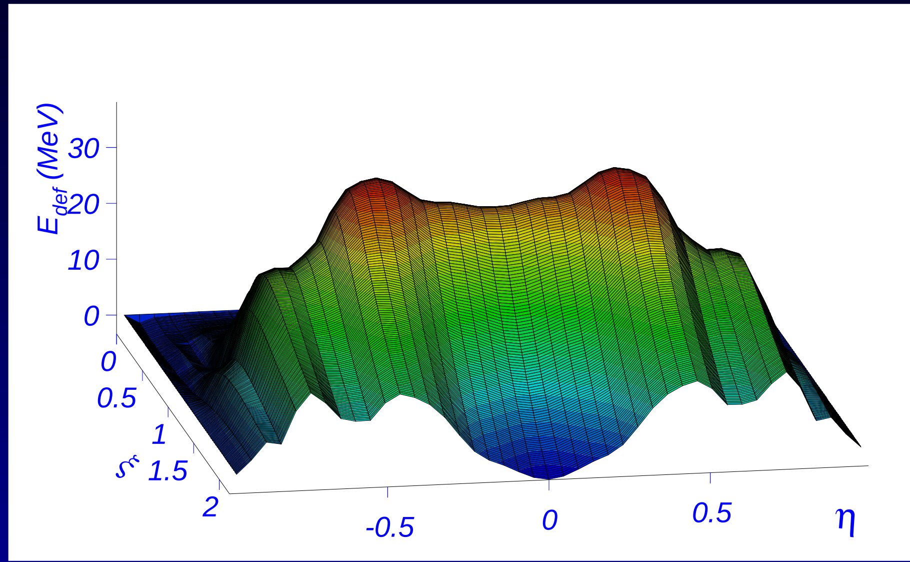
α-decay + ^{102}Sn daughter valley at $\eta = 0.92$

D.N. Poenaru, R.A. Gherghescu, W. Greiner, *Il Nuovo Cimento* **111 A** (1998) 887.

^{106}Te: E_{Y+EM}, $\delta E_{shell+pair}$, E_{def}







separation distance

$\xi = (R - R_i)/(R_t - R_i)$

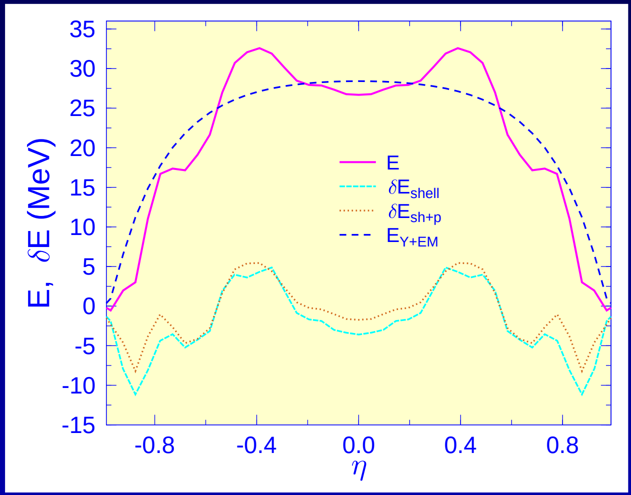
mass asymmetry

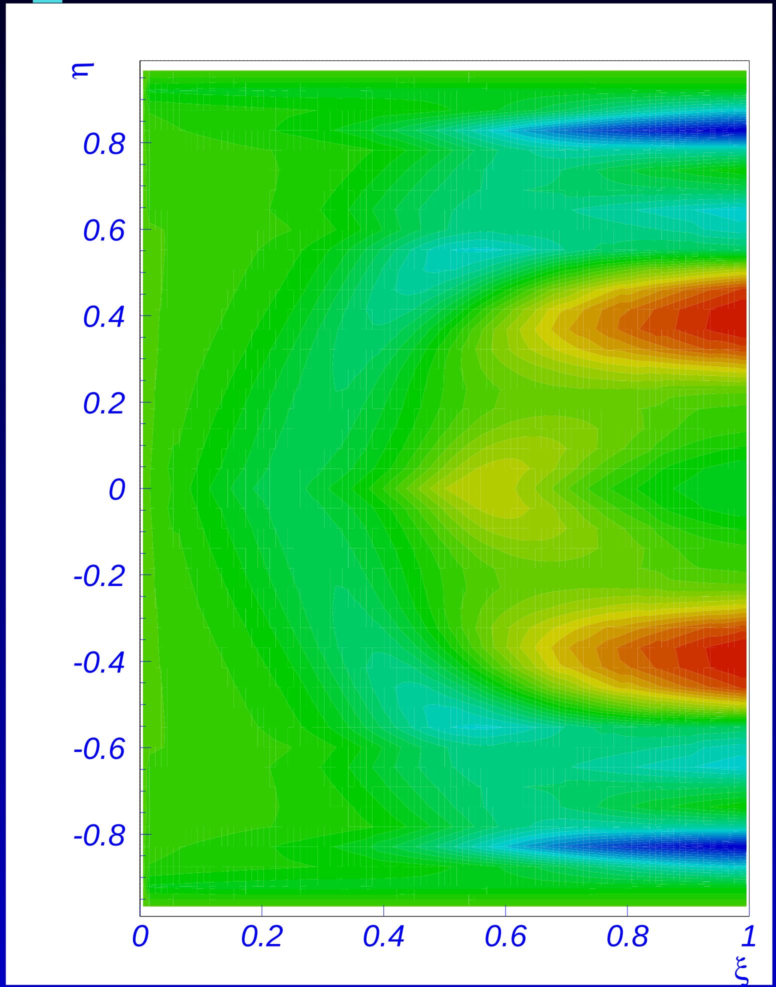
$\eta = (A_1 - A_2)/(A_1 + A_2)$

α-decay + ^{102}Sn daughter valley at $\eta = 0.92$

D.N. Poenaru, I.H. Plonski, R.A. Gherghescu, W. Greiner, *J. Phys. G: Nucl. Part. Phys.* **32** (2006) 1223.

^{106}Te touching point, contour

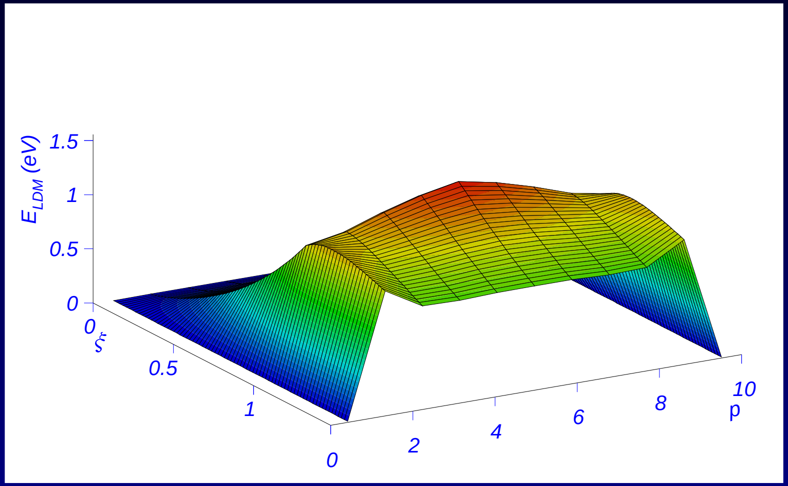


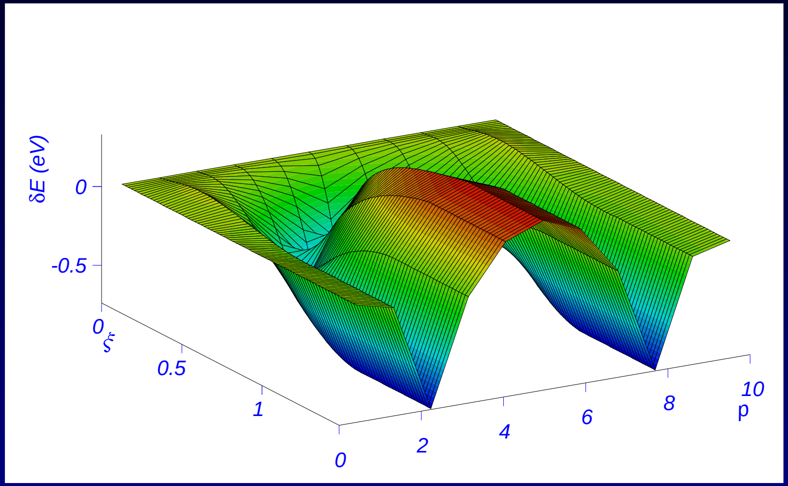


$\delta E_{shell+pairing}$ contour plot

in the plane $\xi = (R - R_i)/(R_t - R_i), \eta$

$\mathbf{Ag_{12}^{2+}}$ fission. $10 \rightarrow p + (10 - p)$





separation distance

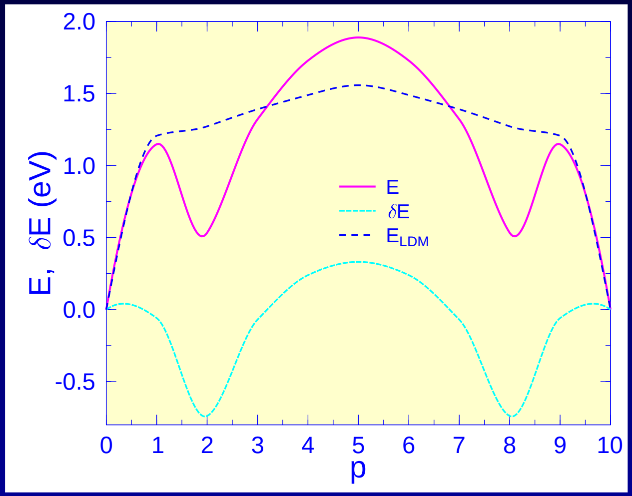
$\xi = (R - R_i)/(R_t - R_i)$

mass asymmetry

$\eta = (10 - 2p)/10$

$Ag_3^+ + Ag_9^+$ valley at $\eta = 0.6$

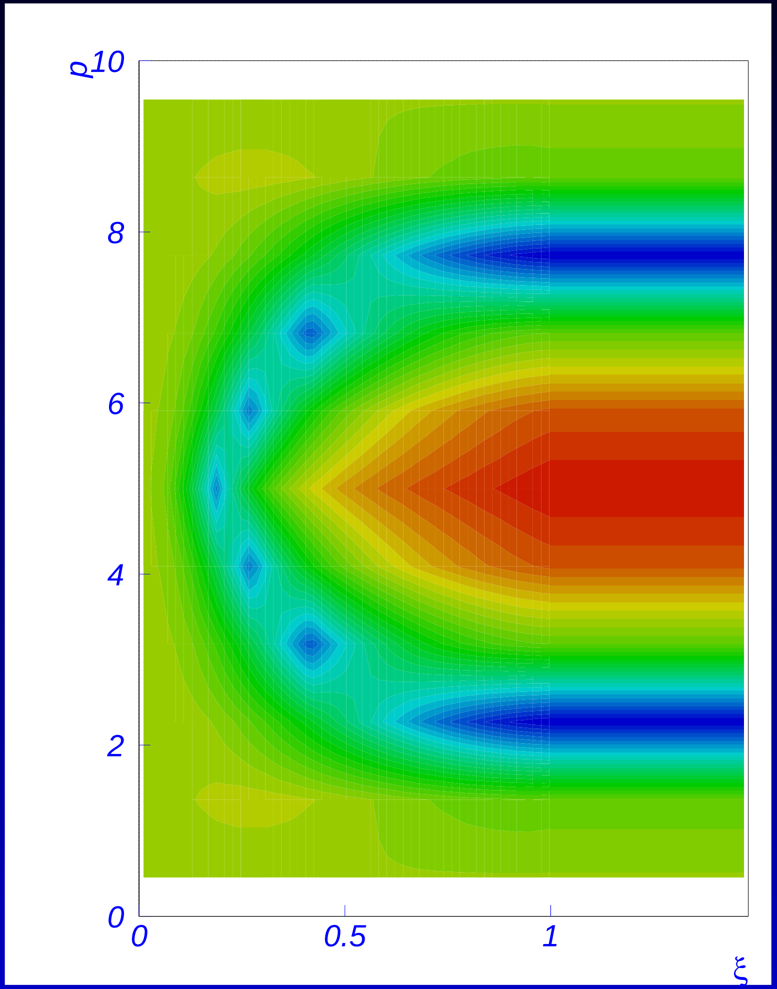
Ag_{12}^{2+} fiss.: touching p., contour



Smoothed lines.

Two magic numbers:

2 and 8



δE contour plot

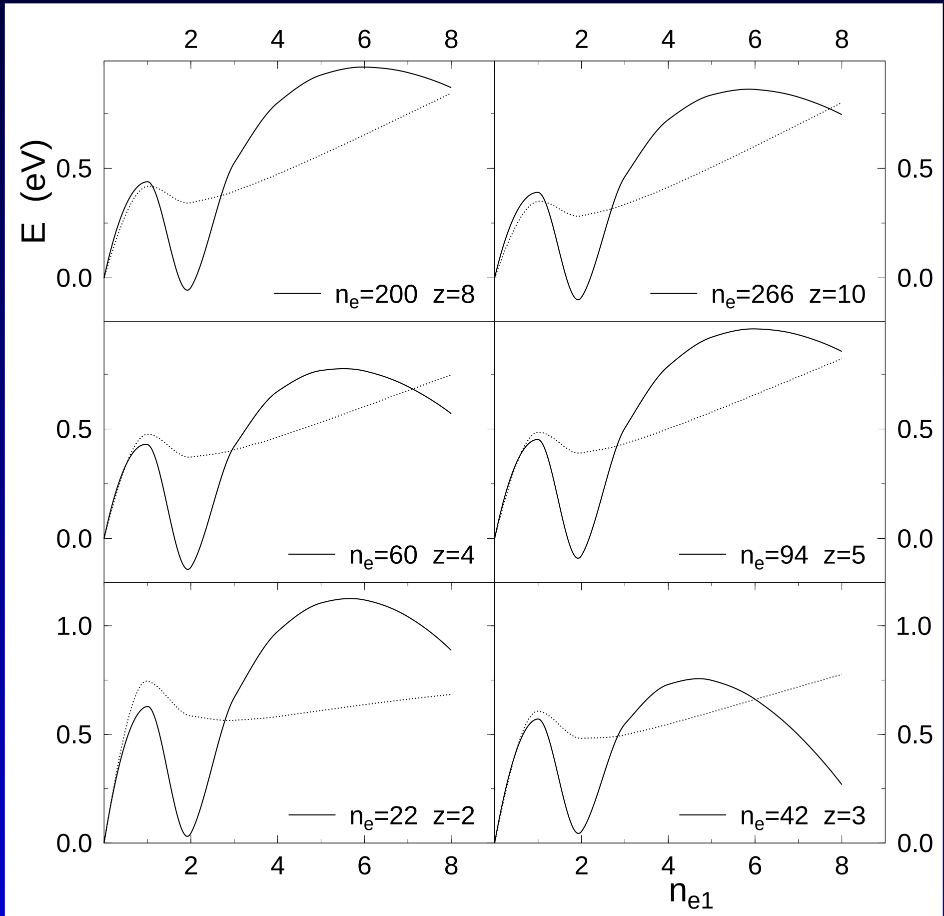
in the plane $\xi = (R - R_i)/(R_t - R_i), p$

IDEAL TRIMER EMITTERS

For Nuclear Decay Modes usually (except $^{264}Fm \rightarrow 2\ ^{132}Sn$ cold fission) the asymmetry where shell correction is minimum is different from that corresponding to LDM minimum.

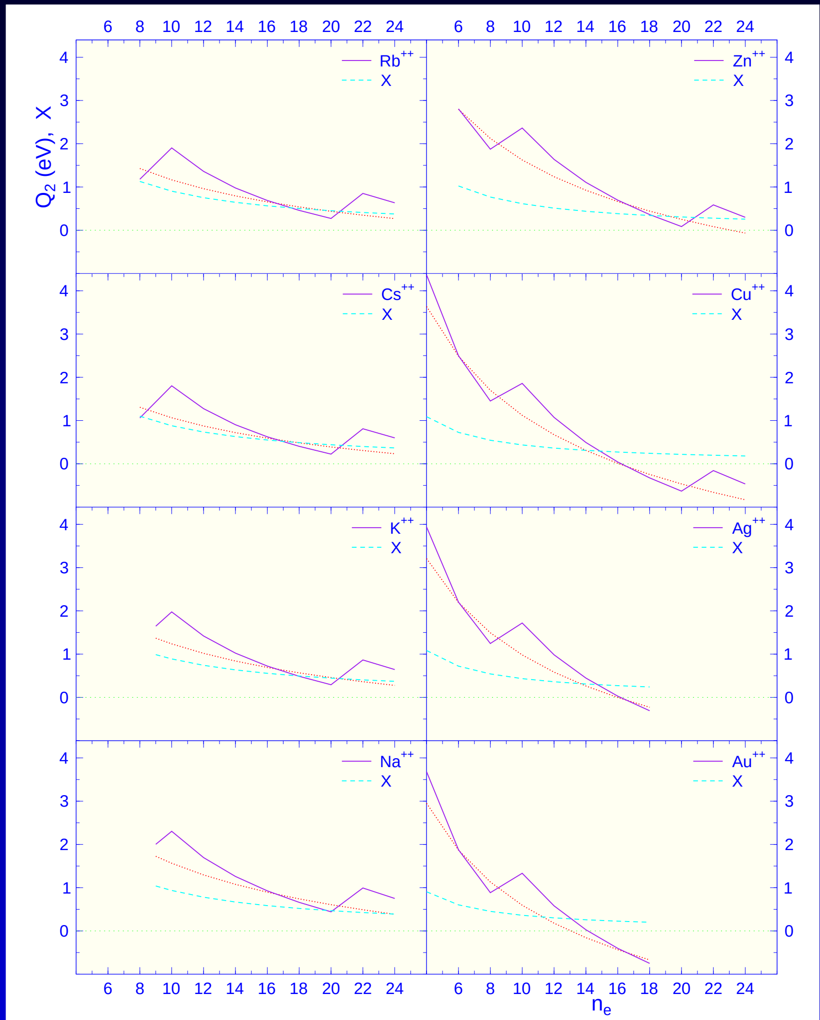
For alkali metal clusters the value of these two asymmetries are almost the same.

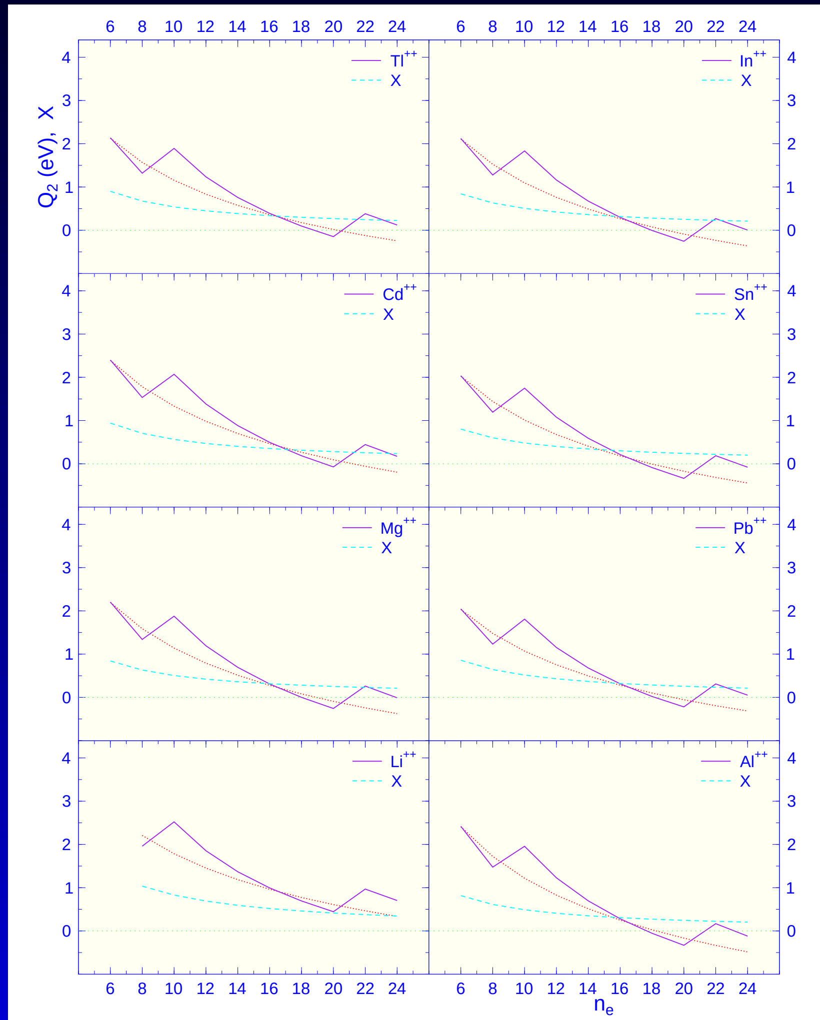
Cs as one of the Ideal Emitters



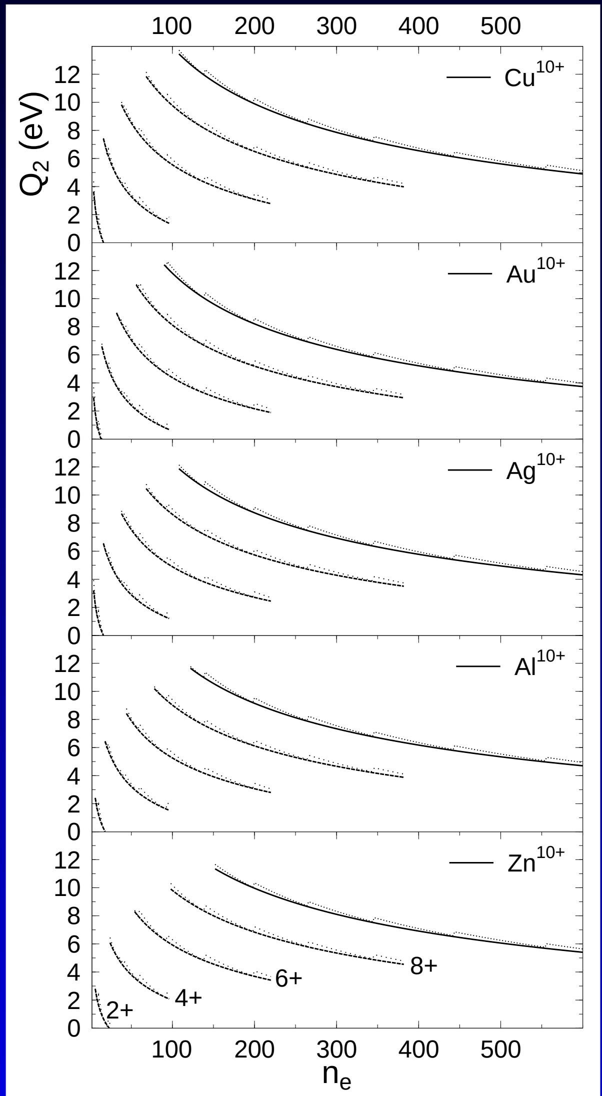
Details of the large asymmetry part of scission point deformation energies E_{LDM} (dotted line) and $E_{LDM} + \delta E$ (full line) for different values of z and n_e.

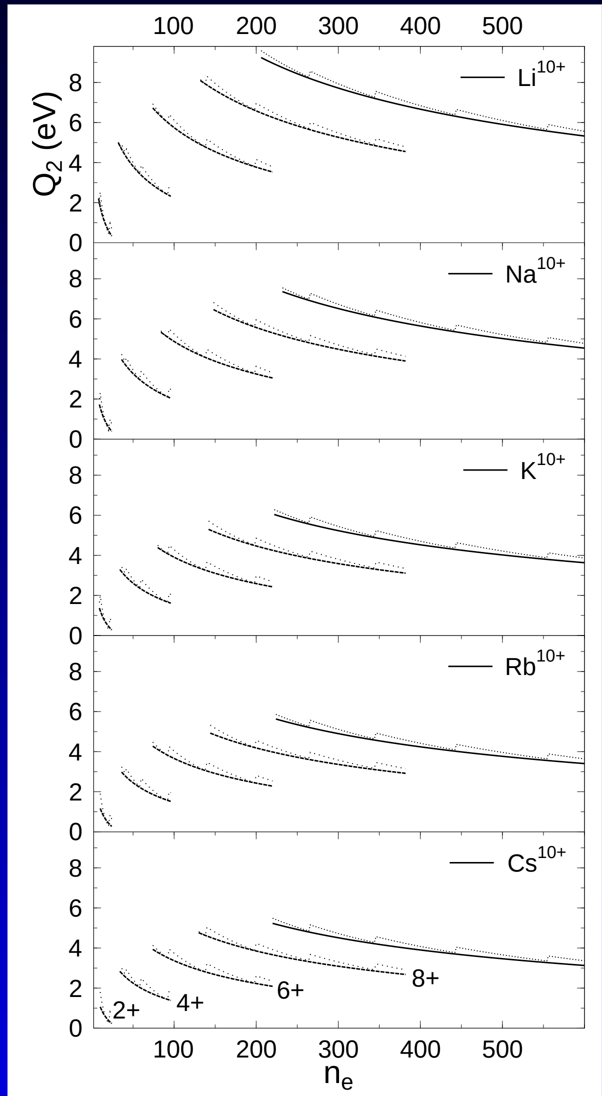
Metalic clusters. Sphere





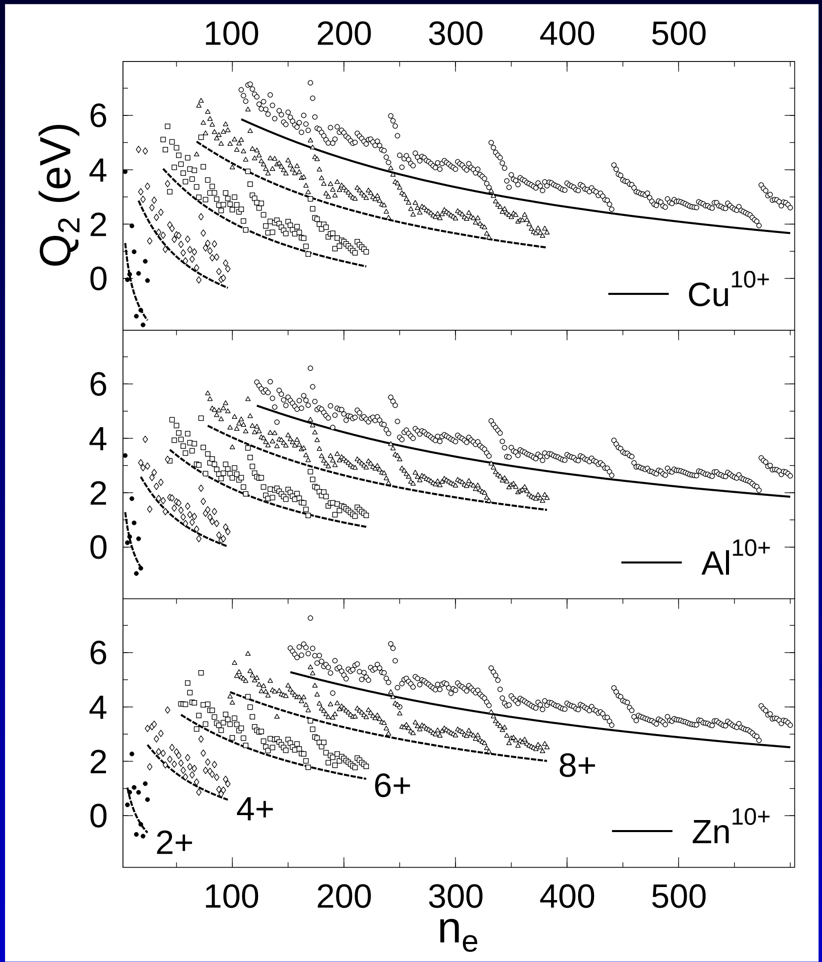
Transitions and alkali metals. Sphere

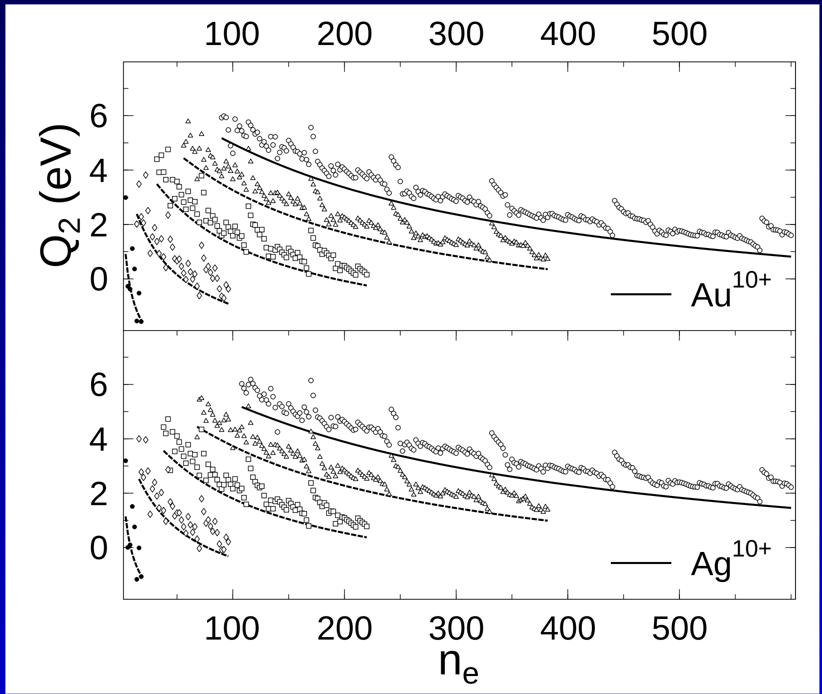




Large Q_{LDM} when both Q_s and Q_C are large, i.e. the ratio a_s/r_s is large: max. for transition metals and min. for alkali metals.

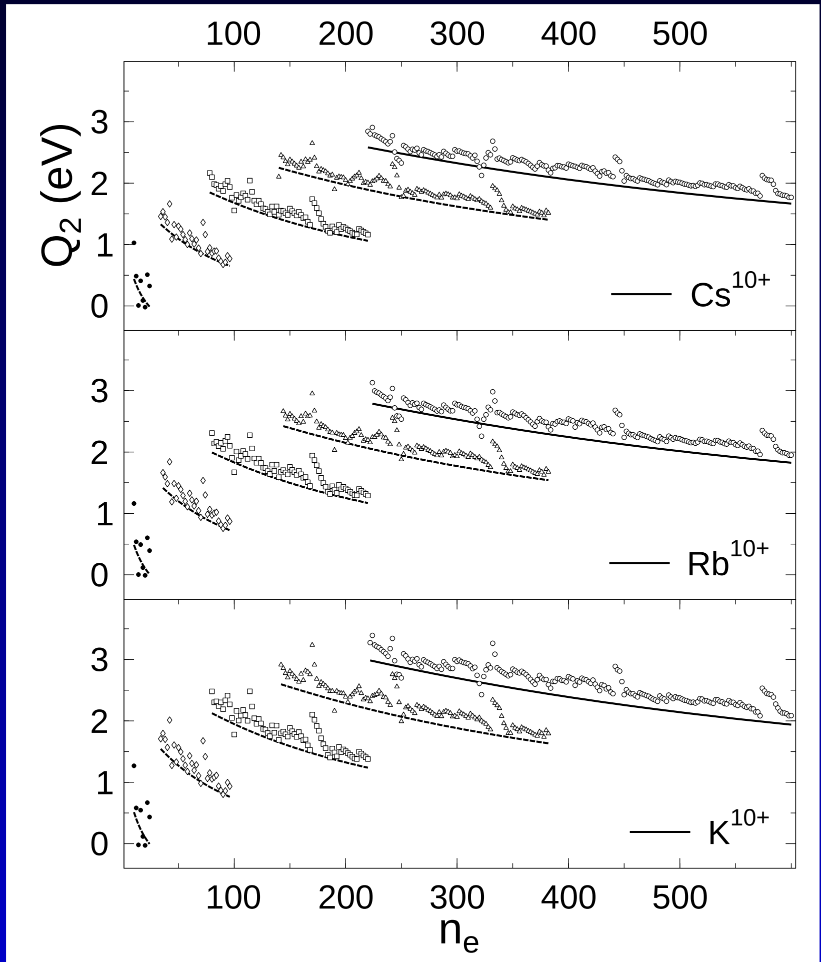
Transitions metals. Hemispheroid

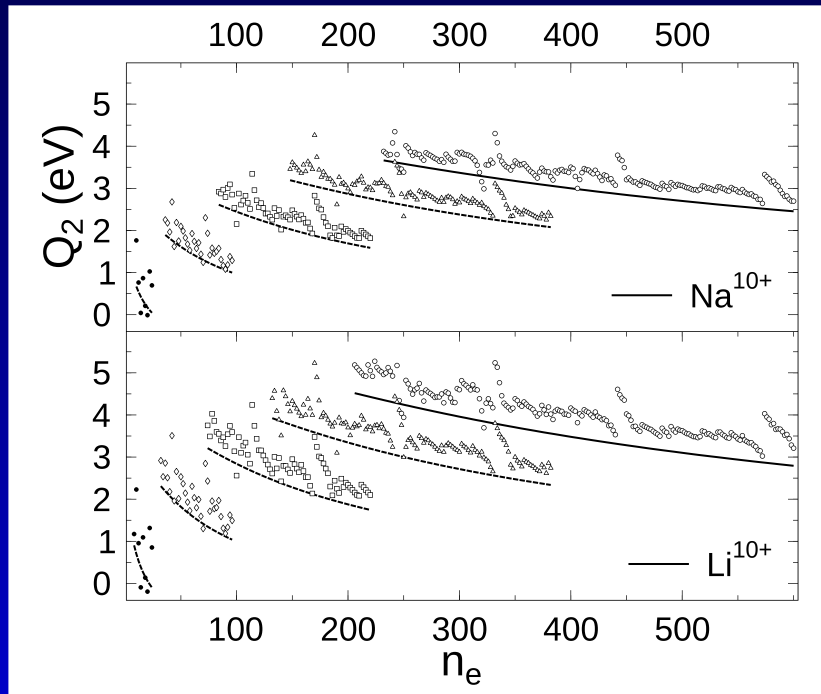




Q obtained after minimization of E_{LDM} and $E_{def} = E_{LDM} + \delta E$ of the parent, daughter and emitted cluster.

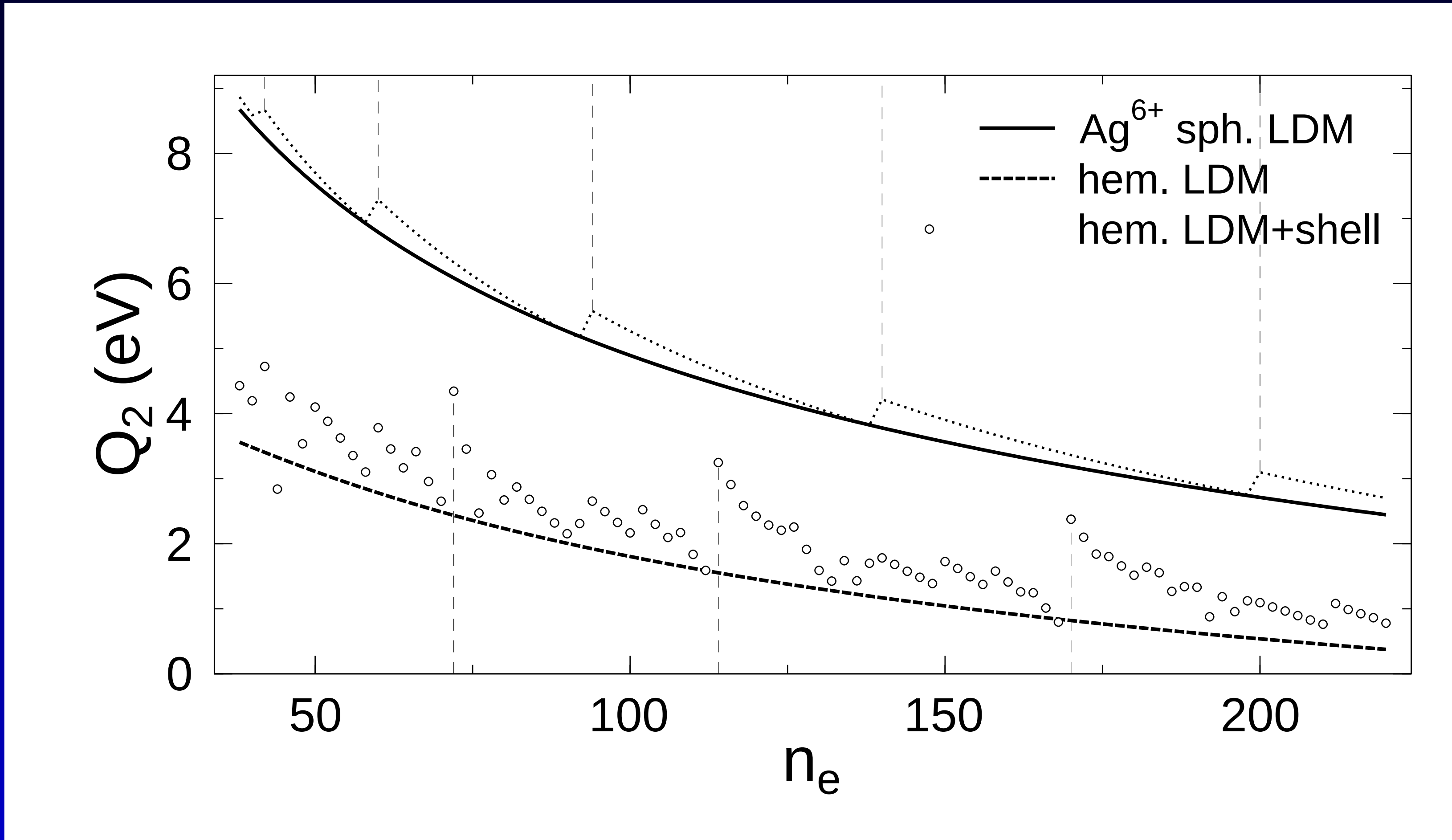
Alkali metals. Hemispheroid





Q obtained after minimization of E_{LDM} and $E_{def} = E_{LDM} + \delta E$ of the parent, daughter and emitted cluster.

$Ag^{6+}_{n_e+6}$ Spheres and Hemispheroids

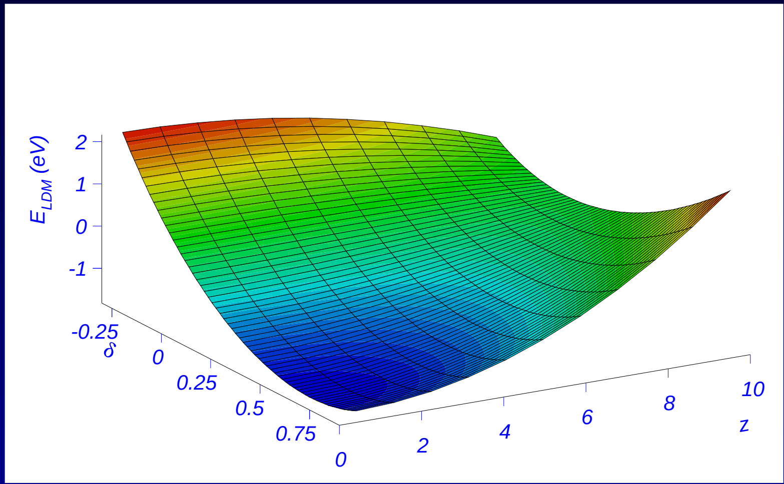


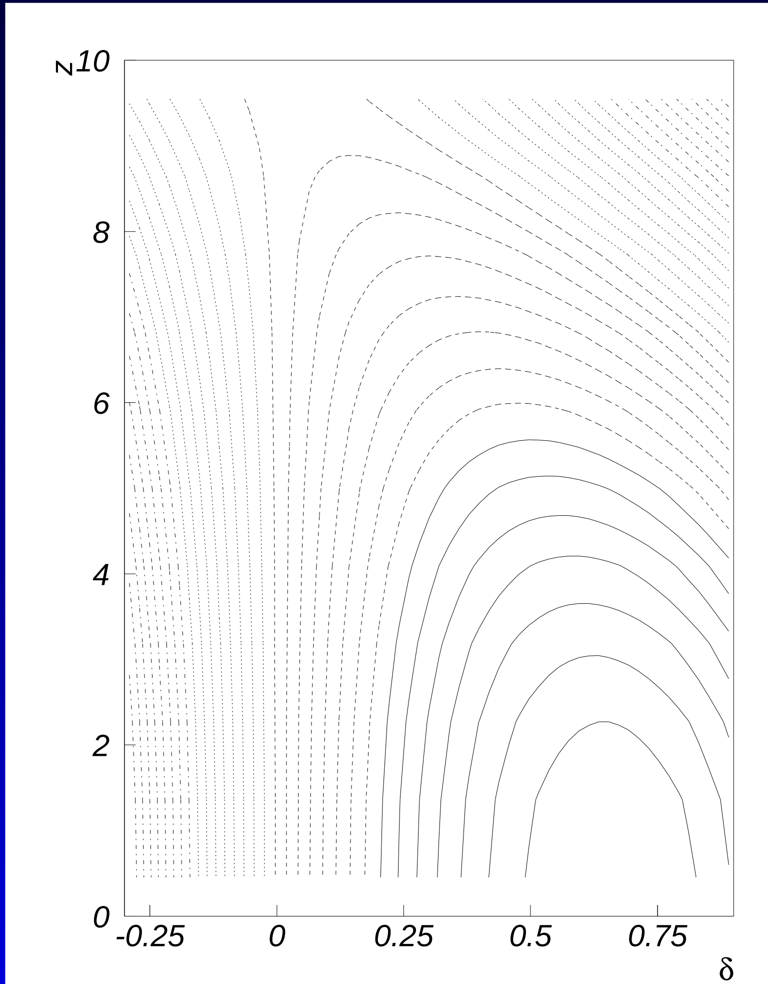
Max. Q-value for two fragments with magic numbers of electrons $n_e = 2 + n_{d-magic}$.

For spherical shape $n_{d-magic} = 40, 58, 92, 136, 198$.

For a superdeformed hemispheroid $n_{d-magic} = 70, 112, 168$.

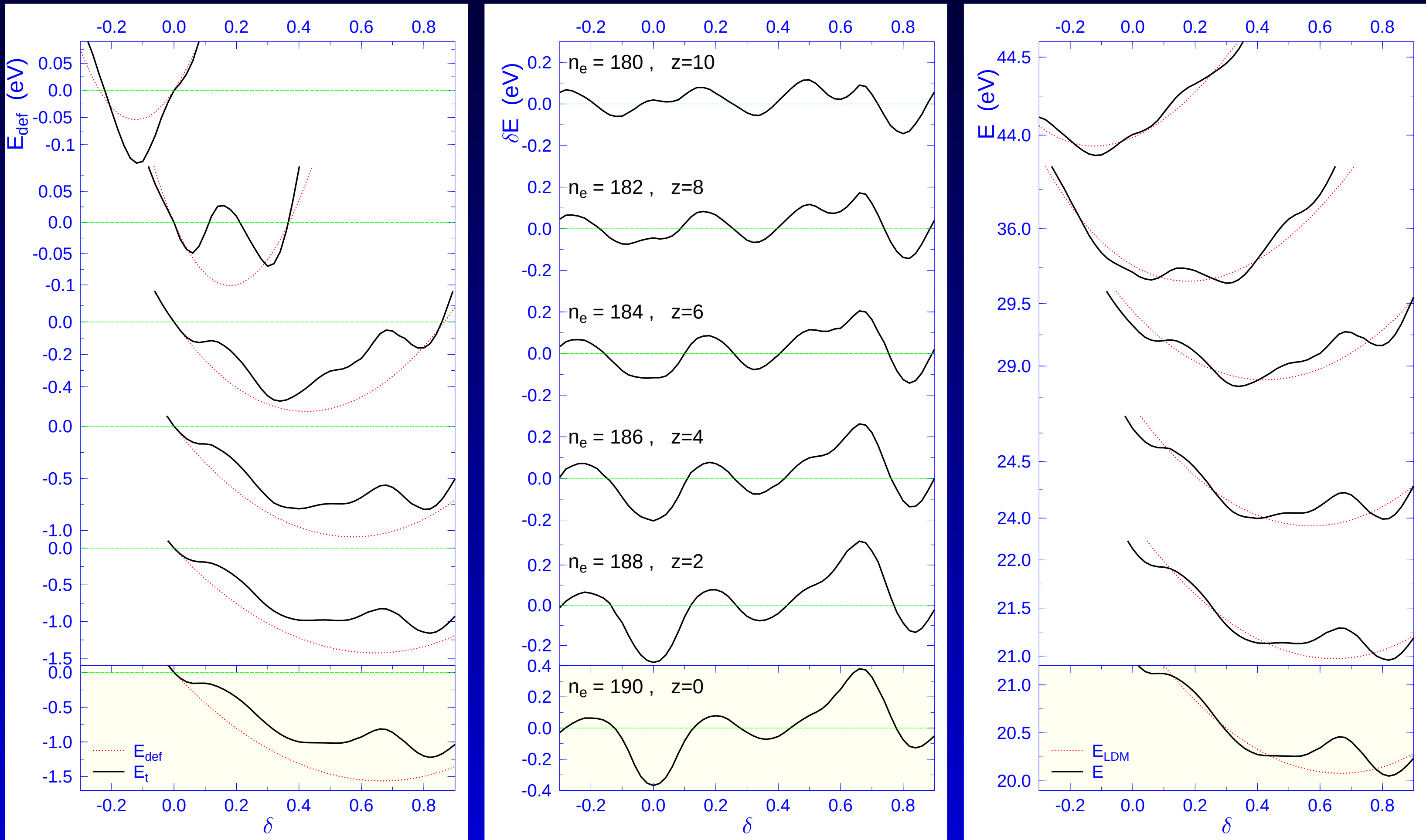
Cs_{240}^{z+} Hemispheroids





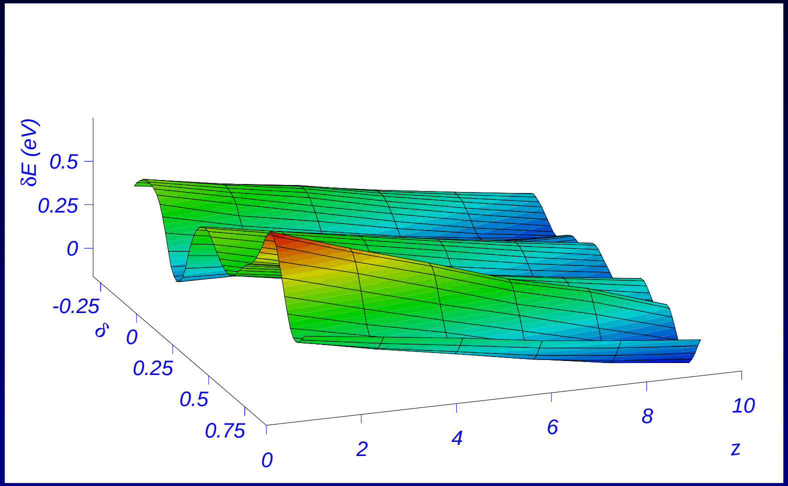
LDM equilibrium deformation decreases when z increases. For applications in which the substrate should be entirely covered with deposited cluster, the efficiency increases when the clusters are charged: with 60 % when δ_{eq} decreases from 0.66 to 0.

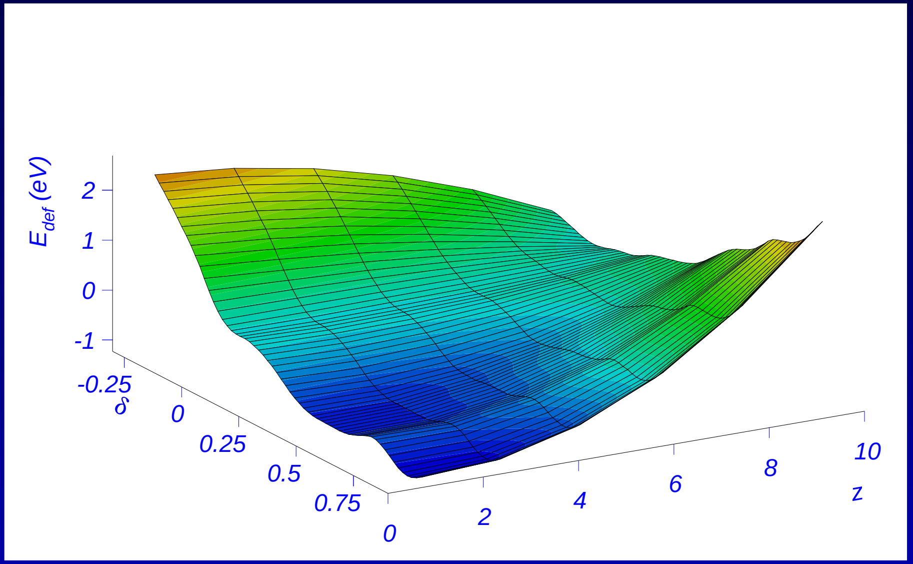
Cs_{190}^{z+} Hemispheroids

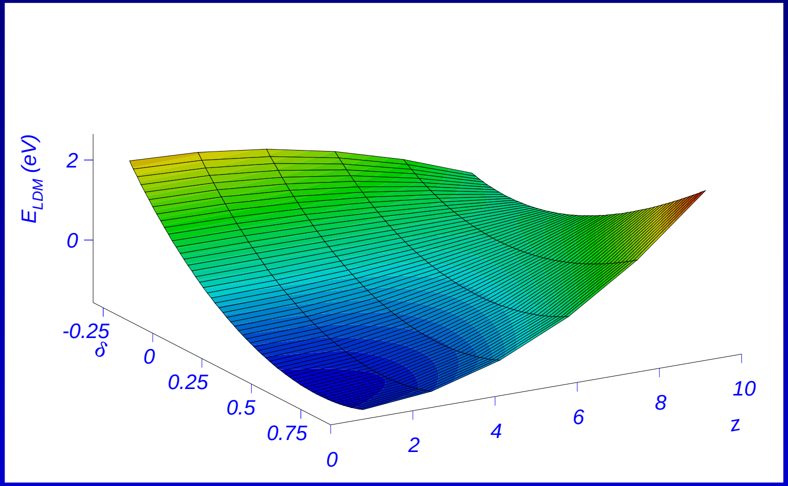


Equilibrium deformation decreases and the minimum energy increases when z increases.

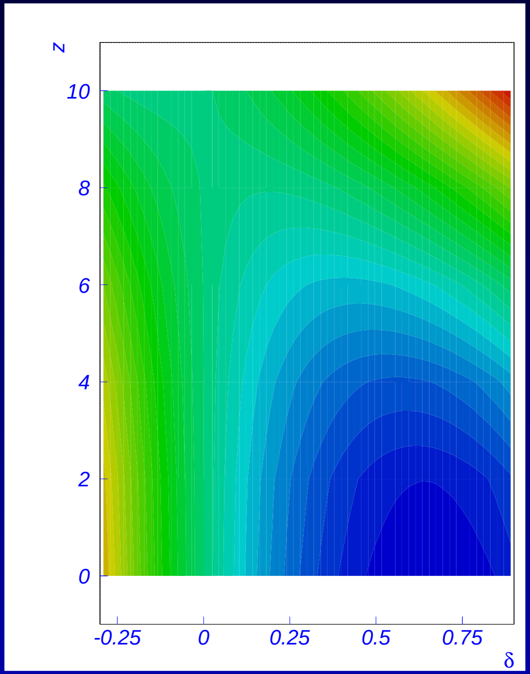
Cs_{190}^{z+} Hemispheroids. PES



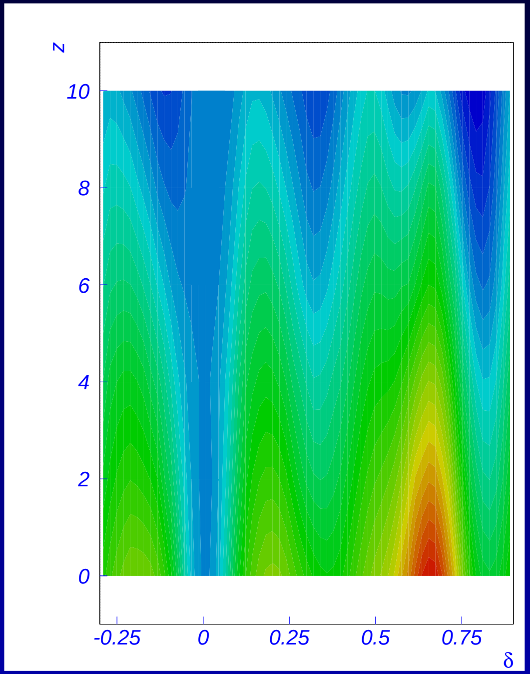




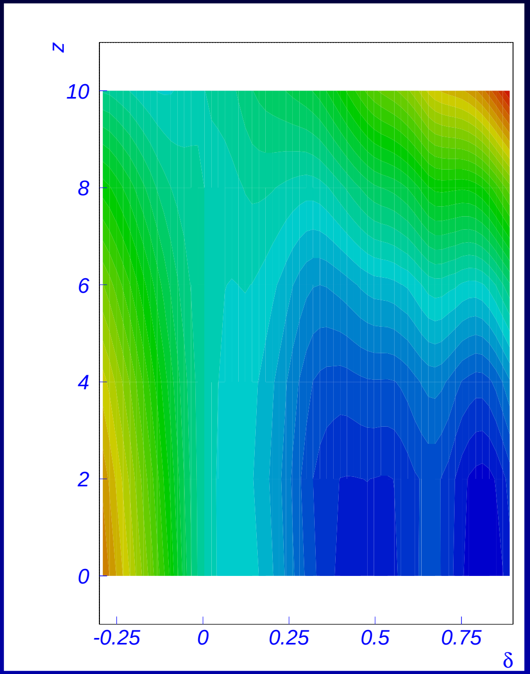
Cs_{190}^{z+} Hemispheroids. Contour plot



E_{LDM}



δE



E_{def}

CONCLUSIONS I

- The electric (excess) charge of a metallic cluster is distributed at the surface unlike in a nucleus
- For a spherical shape $E_C^0 = (ze)^2/(2R_0)$ - metal, $E_C^0 = 3(ze)^2/(5R_0)$ - nucleus
- For some shapes (e.g. spheroid and ellipsoid) the analytical relationships of B_C obtained for nuclei are the same for metals
- Interaction energy of separated cluster fission fragments should take into account the induced charges
- Small size charged clusters, $n < n_c$, are unstable against fission

CONCLUSIONS II

- A shape isomeric state obtained in some fission reactions (e.g. $Na_{16}^{2+} \rightarrow Na_3^+ + Na_{13}^+$) may be viewed as a precursor
- B_C is maximum at $\delta = 0$ for a spheroid and at a hyperdeformation $\delta = 1.17$ for a doubly charged Na_{54}^{2+} hemispheroid or cylinder ($c/a = 5.05$)
- The valleys produced by shell effects on PES of superheavy nuclei, cluster radioactive heavy nuclei, and α-emitters are usually shallower because of the LDM steep variation (e.g. Businaro-Gallone mountains).
- The fission channel with a light trimer M_3^+, frequently met in experiment, is promoted not only due to shell effects but also due to LDM low fission barrier. Charged clusters are ideally “alpha” emitters!

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