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ON THE NUMERICAL ANALYSIS OF DECAY RATE ENHANCEMENT IN METALLIC ENVIRONMENT

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Motivated on the very recent experiments to determine the acceleration of the alpha decay of meta-stable radionuclides in metallic environment some work has been done to strengthten the importance in the process of electrons screening in metals. Thus, by combining the Gamow decay theory with electrostatic screening in Debye–Hückel approximation (jellium model) a formula for "the shift" in screening energy which enters in the decay enhancement factor expression that copes well with these experiments has been derived. It was established that to simulate the poly-atoms system containing decaying isotopes in QM&MD codes calculations, and to include "the screening energy shift" of protons, decay alpha, beta$^+$ particles due to all surrounding interacting effects, it is sufficiently only to substitute the code ruly pseudo-potential input for hydrogen-like atoms (including alpha) by a screened Coulomb potential as from the well-known Gamow alpha decay theory. For demonstration is used the QM&MD code package which usually performs density-functional theory (DFT) total-energy calculations for materials ranging from insulators to transition metals. This package employs first-principles pseudo-potentials and a plane-wave basis-set, and it was used to do a special calculus for some metal environments (Pd) where protons–deuterons are implanted or when it is alloyed with a radionuclide-like isotopes ($^{174}\mathrm{Hf}_{72}$), the results compare well with the existing experiments on the decay enhancement. These works give further arguments for a cheap solution to remove the transuranic waste (involving all alpha-decay) of used-up rods of fission reactors in a time period of a few years.

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1. Introduction

In a recent publication, Kettner *et al.* [1] have suggested that one could speed up alpha decay of transuranic nuclear waste material by embedding it in metals at low temperature. This would have a huge impact on waste disposal management and quite likely initiate a completely renewed public debate on the subject of nuclear energy. The proposed idea is that the electron screening provided by the metal will lower the alpha-decay lifetime.

The screening enhancement effect in laboratory nuclear reactions at astrophysical energies has attracted a lot attention recently, especially after the recent accomplishments of the LUNA collaboration at Gran Sasso [2]. Other low-energy experiments of the proton–proton chain [1–3] (past, current or planned) still need a theoretical model that could account for the observed enhancement. The use of insulators and metallic alloys in LUNA [2] experiments indicate that only when the metallic character remained essentially unchanged by a small content (a few %) of alloy meta-stable element (alpha, beta$^+$ decay) the screening effect is remarkable, mainly due to the free electrons in metallic environment.

If we have alpha and beta particles both with a chance of escaping from nucleus, it might be thought that every radioactive element should disintegrate partly with expulsion of alpha particles and partly with beta particles. But we would argue that the escape rate is extremely sensitive to the height to which the potential energy curve rises above the energy-level in question; if the size of this potential barrier is increased by a small factor the probability of escape might decrease more than million-fold.

In the literature, the different analytical approaches [4, 5] are formulated in order to describe these important experimental findings, but most of them fail due to a "forced" application to solids — some comments are presented in Section 2 of this paper.

In the present paper, the new ideas are advanced, mainly to calculate the shift of the screening energy of the decay alpha particle following interactions in order to explain all results of Ketner *et al.* [1]. Thus, firstly a new formula is derived and, secondly a numerical analysis based on the density functional theory (DFT) is proposed. It is known that this theory permits to account for all interaction effects of the "environment" of a system of atoms which is only possible if more sophisticated and reliable tools in quantum mechanics (QM) are used. Therefore, it will be shown that in order to obtain "a screening energy shift" of the decay alpha particle due to atom system with crystalline symmetry, it is sufficiently to substitute the ruly pseudo-potential (QM-code input) for hydrogen-like atoms (including alpha) with the screened Coulomb potential as deduced from the well-known Gamow alpha decay theory.

In the present paper we have performed dedicated calculations for some metal environments like (Pd) where either protons–deuterons are implanted, or when they are alloyed with radionuclide-like isotopes ($174\,\mathrm{Hf}_{72}$). We have used an available tool, namely FHI98MD computer code [6] that employs quantum mechanical effects (QM) and molecular dynamics (MD).

Before the discussion of the results obtained by means of the above computer code, firstly, in Section 2 the screened Coulomb potentials for hydrogen-like atoms is derived in a detailed fashion, which turns out to be very useful in Section 3, where the binding energies calculated by the code are used for the calculation of the decay enhancement factor and half-live shortening.

2. The derivation of new formula for decay enhancement

2.1. Gamow factor for decay

It is well known from textbooks that the alpha particle can be regarded as trapped by a potential barrier. In order to escape into the environment, the alpha must tunnel through the barrier. This description of alpha decay, which also explains the wide range in lifetimes, was given by Gamow as well as Gurney, Condon [7] and was one of the first successes of the new quantum theory (which introduced such counter-intuitive ideas as tunneling).

Suppose we have a wave packet representing an alpha particle mass μ_{a} and kinetic energy E impinging on a square potential barrier of height $V-E$ and width Δr. Then the transmission coefficient T is obtained from 2nd year quantum mechanics as:

$$T \approx e^{-2k\Delta r}\,, \tag{1}$$

where

$$k = \sqrt{2m_\alpha \left|V-E\right|/\hbar}\,, \qquad \hbar = \frac{h}{2\pi}\,, \tag{2}$$

and h is a Planck constant. This can be extended to any barrier shape in the form of the WKB approximation:

$$T \approx \exp\left(-2\int\limits_R^b k(r)dr\right). \tag{3}$$

Here, R and b are the classical turning points of the motion inside and outside the barrier. We may take the barrier to be the sum of a square well nuclear potential of radius R and a Coulomb potential arising from a charge within R

$$V(r) = \begin{cases} 0\,, & \text{for} \quad r < R\ , \\ \frac{1}{[4\pi\varepsilon_0]}\frac{Z_\alpha Z_{\mathrm{D}} e^2}{r}\,, & \text{for} \quad r \geq R\ . \end{cases} \tag{4}$$

We can equate (approximately) the energy release Q in the alpha decay to the kinetic energy E of the alpha particle and to the potential at the outer classical turning point

$$Q \approx E = \frac{Z_\alpha Z_{\mathrm{D}} e^2}{[4\pi\varepsilon_0]\,b}\,, \tag{5}$$

and hence determine b

$$b = \frac{Z_\alpha Z_{\mathrm{D}} e^2}{[4\pi\varepsilon_0]\, Q}\,. \tag{6}$$

Therefore the integral over $k(r)$ becomes

$$G = \frac{2}{\hbar}\sqrt{2m_\alpha Q}\int_R^b \left[\frac{b}{r} - 1\right]^{1/2} dr\,, \tag{7}$$

where $Q = \frac{1}{2}m_\alpha v^2$ and the above expression for b has been used.

For thick barriers ($R/b \ll 1$ or $V(R) \gg Q$) and $R \ll b$ we have

$$G \approx \frac{4Z_\alpha Z_{\mathrm{D}} e^2}{[4\pi\varepsilon_0]\, \hbar v}\left(\frac{\pi}{2} - 2\sqrt{\frac{R}{b}}\right) \cong 2\pi\frac{Z_\alpha Z_{\mathrm{D}} e^2}{\hbar v}\,. \tag{8}$$

The decay constant for alpha decay is thus

$$\lambda = \lambda_0 e^{-G}\,, \tag{9}$$

where λ_0 is the frequency factor or the number of assaults on the barrier per unit of time, and is usually estimated as

$$\lambda_0 \; = \; \frac{v}{2R} \approx \frac{e}{2R}\sqrt{\left(\frac{2Q_\alpha}{m_\alpha e^2}\right)}\,, \tag{10}$$

$$\eta \; \equiv \; \frac{Z_1 Z_2 e^2}{\hbar v}\,, \tag{11}$$

where v is the relative velocity. As a rule, the known Sommerfeld parameter is given by $2\pi\,\eta = 31.29\ Z_1 Z_2 (\mu/E)^{1/2}$ (see [1]). The quantities Z_1 and Z_2 are the nuclear charges of the interacting particles in the entrance channel, μ is the reduced mass (in units of amu), and $E \equiv Q$ is the centre of mass energy (in units of keV).

Thus, the transmission coefficient is given as $T = \exp(-2\pi\eta)$. Otherwise, from [8] the cross section for nuclear reactions between charges Z_1 and Z_2 at low energies E (*i.e.*, below the Coulomb barrier) has the form $\sigma(E) = (S(E)/E)e^{-2\pi\eta}$, where $S(E)$ is generally referred to as the astrophysical "constant".

A shift on Q with U_{D} as calculated below gives the screening enhancement factor defined [8] as $f_{\mathrm{lab}}(E) = \sigma_{\mathrm{s}}(E)/\sigma_{\mathrm{b}}(E)$, where $\sigma_{\mathrm{s}}(E)$ is the higher cross section due to screening and $\sigma_{\mathrm{b}}(E)$ that for bare nuclei, and $f_{\mathrm{lab}}(E) \cong \exp(\pi\eta U_{\mathrm{D}}/E)$.

2.2. Models based on astrophysical stellar plasma

At astrophysical energies the potential energy is found to be shifted by a constant screening energy U_e. In Refs. [4,5] the screening energy for the collision of a light bare nucleus (Z_2e) with a neutral multielectron atom (Z_1e) has been calculated in the framework of the Thomas–Fermi model for a special screened Coulomb potential

$$U_{\mathrm{TF}}^{\mathrm{SL}} = -1.21\, Z_1^{4/3} Z_2 \frac{e^2}{a_0}\,. \tag{12}$$

Obviously, the screening effect reduces the half-life of the decaying nucleus. This is of course expected, since the screening cloud reduces the Coulomb barrier thus easing the way of the α particle out of the parent nucleus. In Fig. 1, we calculated the value of $U_{\mathrm{TF}}^{\mathrm{SL}}$ for

$$174\,\mathrm{Hf}_{72} \xrightarrow{\alpha} 170\,\mathrm{Yb}\,, \tag{13}$$

with half-life 2.0×10^{15} years, mode of decay: alpha to Yb-170, decay energy 2.495 MeV. The screened α decay according to Liolios formula (13) being $U_{\mathrm{TF}}^{\mathrm{SL}} = 500\,\mathrm{a.u.} \times 27.2 = 13.6\,\mathrm{keV}$.

We know that the abundance follows the usual law of exponential decay, that is

$$\lambda \;=\; \frac{\ln 2}{T_{1/2}}\,, \tag{14}$$

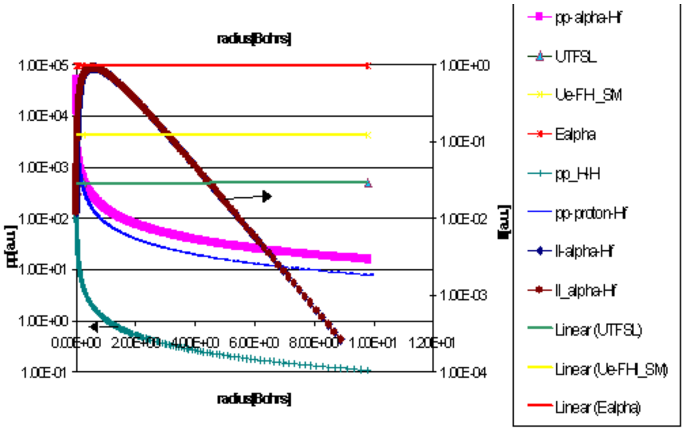

Fig. 1. The evolution of pseudo-potential (*pp*) *versus* atom radius for alpha particle and proton according to Eg. (37); on the right axes the local component of the potential (II) is shown; Ealpha-energy of alpha particle.

$$N(t) \;=\; N(0)\exp\left(-\frac{\ln 2}{T_{1/2}}t\right) . \qquad (15)$$

A similar result based on the astrophysical approach has been obtained in Ref. [1], where the cross section of a charge-particle-induced nuclear reaction is enhanced at subcoulomb energies by the electron clouds surrounding the interacting nuclides, with an enhancement factor [1,3]

$$f_{\mathrm{lab}}(E) = E(E+U_e)^{-1}\exp\left(\frac{\pi\eta U_e}{E}\right) , \qquad (16)$$

where E is the centre-of-mass energy, η is the Sommerfeld parameter and U_e is the screening energy. The electron screening in $d\ (d,p)\ t$ has been studied for deuterated metals, insulators and semiconductors, *i.e.* 58 samples in total [3]. As compared to the measurements performed with a gaseous D_2 target ($U_e = 25\,\mathrm{eV}$), a large screening was observed in all metals (of order $U_e = 300\,\mathrm{eV}$), while a small (gaseous) screening was found for the insulators and semiconductors. In [1], an explanation of the large screening was suggested by the Debye plasma model applied to the quasi-free metallic electrons. In this approach, they combined the Drude model of metals (with a kinetic energy $0.5\,kT$ for the quasi-free valence electrons) with the Debye model of plasma: the Drude–Debye model, in short Debye model.

The electron Debye radius around the deuterons in the lattice is given by

$$R_{\mathrm{D}} = \left(\frac{\varepsilon_0 kT}{e^2 n_{\mathrm{eff}}\rho_a}\right)^{1/2} = 69\left(\frac{T}{n_{\mathrm{eff}}\rho_a}\right)^{1/2}\ [\mathrm{m}] ,$$

with the temperature T of the quasi-free electrons in units K, n_{eff} the number of thesis electrons per metallic atom and the atomic density ρ_a in units of atoms m^{-3}. For $T = 293^\circ\,\mathrm{K}$, $\rho_a = 6\times 10^{28}\,\mathrm{m}^{-3}$, and $n_{\mathrm{eff}} = 1$ obtained radius R_{D} is about a factor 10 smaller than the Bohr radius of a hydrogen atom. With the Coulomb energy of the Debye electron cloud and a deuteron projectile at R_{D} set equal to $U_e \equiv U_{\mathrm{D}}$, one obtains $U_{\mathrm{D}} = 2.09\times 10^{-11}\,(n_{\mathrm{eff}}\rho_a/T)^{1/2}\,\mathrm{eV}$. With the Coulomb energy between two deuterons at R_{D} set equal to U_e, one obtains $U_e = (4\pi\varepsilon_0)^{-1}e^2/R_{\mathrm{D}} = 300\,\mathrm{eV}$ the order of magnitude of the observed U_e values.

An important deduction made in [1] is it that the Debye energy U_{D} should scale with the nuclear charge Z_t of the target atoms

$$U_{\mathrm{D}} \;=\; 2.09\times 10^{-11}\,(Z_{\mathrm{t}}\,(Z_{\mathrm{t}}+1))^{1/2}\left(\frac{n_{\mathrm{eff}}\rho_a}{T}\right)^{1/2}[\mathrm{eV}] , \qquad (17)$$

$$U_{\mathrm{D}} \;=\; Z_\alpha Z_{\mathrm{t}} U_e(d-d)\left(\frac{290}{4}\right)^{1/2} . \qquad (18)$$

Thus, for Pd–Hf (alpha decay), $U_{\mathrm{D}} = 2 \times 300 \times 72 \times 8.5 = 355\,\mathrm{keV}$.

The proposed parametric model is mainly based on the known approach of Debye–Hückel and well fits the experiment. Note that this model is in fact based on the Maxwell–Boltzmann statistics when the temperature T is high (which is not the case). At low temperature when T is approximately equal to the kinetic energy E_{F} — the Fermi energy obeys Thomas–Fermi statistics as for free electrons gas (of non-Coulomb interacting electrons). For strongly interacting plasma at finite temperatures, as in the case of metals, other methods based on, say density functional theory, are needed. This idea is pursued in the present paper. The perturbation theory applied in both approximations Debye–Hückel or Thomas–Fermi are introduced into the first Maxwell equation, giving

$$\left[\nabla^2 - k_0^2\right] \phi(r) = -\frac{Q}{\varepsilon_0}\delta(r)\,, \tag{19}$$

which is known as the screened Poison equation. The solution is

$$\phi(r) = \frac{Q}{4\pi\varepsilon_0 r}\, e^{-k_0 r}\,, \tag{20}$$

which is called screened Coulomb potential. It is a Coulomb potential multiplied by an exponential damping term, with the strength of damping factor given by the magnitude of k_0, the Debye or Fermi–Thomas wave vector, where for the first case

$$k_0 \;\equiv\; \sqrt{\frac{\rho e^2}{\varepsilon_0 k_{\mathrm{B}} T}}\,, \qquad \text{(Debye)} \tag{21}$$

and

$$k_0 \;\equiv\; \sqrt{\frac{3e^2\rho}{\varepsilon_0 E_{\mathrm{F}}}}\,, \qquad \text{(Fermi–Thomas).} \tag{22}$$

The associated Debye length $\lambda_{\mathrm{D}} = 1/k_0$ is the fundamental length scale of classical plasma. These results are obtained in perturbation theory, which fails when the Wigner–Seitz radius $r_{\mathrm{s}} = (3/(4\pi n))^{1/3} > 1$, where n is the number of particles per unit of volume: r_{s} is also called the coupling constant $r_{\mathrm{s}} =$ (potential energy)/(kinetic energy). In the case of metals $2 < r_{\mathrm{s}} < 6$. At high density of electrons when $r_{\mathrm{s}} < 1$ the effect of Coulomb interaction is weak and the free electron gas is a good approximation.

3. The application of the Density Functional Theory codes to calculate the decay enhancement in poly-atomic structures

The package `fhi96md` is an efficient code to perform density-functional theory total-energy calculations for materials ranging from insulators to transition metals. The package employs first-principles pseudopotentials and a plane-wave basis-set. For exchange and correlation both the local density and generalized gradient approximations are implemented.

In poly-atomic systems as for example molecules, crystals, defects in crystals, surfaces, it is highly desirable to perform accurate electronic structure calculations, without introducing uncontrollable approximations.

The package `fhi96md` used in this paper is especially designed to investigate the material properties of large systems. It is based on an iterative approach to obtain the electron ground state. Norm-conserving pseudopotentials [9] in the fully separable form of Kleinman and Bylander are used to describe the potential of the nuclei and core electrons acting on the valence electrons. Exchange and correlation are described by either the local-density approximation [6] or generalized gradient approximation. The equation of motion of the nuclei (EOM) is integrated using standard schemes in molecular dynamics.

The key variable in DFT is the electron density $n(\boldsymbol{r})$. As stated by the fundamental theorem of Hohenberg and Kohn the ground state energy $E_0(\{\boldsymbol{R}_J\})$ of the system for given positions of the nuclei $\{\boldsymbol{R}_J\}$ is the minimum of the Kohn–Sham total energy functional with respect to the electron density n. The total energy functional $E[n]$ is:

$$E\left[n\right] = T^{\mathrm{S}}\left[n\right] + E^{\mathrm{H}}\left[n\right] + E^{\mathrm{e-nuc}}\left[n\right] + E^{\mathrm{XC}}\left[n\right] + E^{\mathrm{nuc-nuc}}\,, \qquad (23)$$

where T^{S} is the kinetic energy of non-interacting electrons, E^{H} is Hartree energy, and E^{XC} is the exchange-correlation energy. The expressions for the energy are briefly discussed in the following. The explicit expression for each of the contributions to the total-energy, potentials and forces are given in a form implemented in the program.

The energy of the electron–nuclei and nuclei–nuclei interaction $\mathrm{E}^{\mathrm{e-nuc}}$ and $\mathrm{E}^{\mathrm{nuc-nuc}}$ are

$$E^{\mathrm{e-nuc}}[n] = \int d^3r\, V^{\mathrm{e-nuc}}(r)n(r)\,, \qquad E^{\mathrm{nuc-nuc}} = \frac{1}{2}\sum_{IJ,I\neq J}\frac{Z_1 Z_2}{|R_I - R_J|}\,, \quad (24)$$

.where Z_1 and Z_J are the charges of the corresponding nuclei. Throughout the paper we employ atomic units (energy in hartree) unless otherwise noted. As approximations to the exchange-correlation energy functional $E^{\mathrm{XC}}[n]$ the local-density approximation (LDA) is employed in a form obtained for the homogeneous electron gas by Ceplerley and Alder in the parameterization of Perdew and Zunger, and the generalized gradient approximations of Becke and Perdew and of Perdew *et al.* are used.

The effect of the core electrons and the Coulomb potentials of the nuclei is replaced by soft pseudo-potentials which enables the efficient use of a plane-wave basis.

We employ the norm conserving pseudopotentials (GNCPP) constructed *e.g.* following the schemes of Hamann [9], shown bellow. These have been proven to yield transferable potentials for a broad class of nuclei ranging from first row elements to transition metals. The pseudopotentials are represented in the fully separable form as proposed by Kleinman and Bylander.

In this form the pseudopotential splits into a local part $\vec{V}_{I_s}^{\text{ps,local}}$ and a non-local but separable part $\vec{V}_{I_s}^{\text{ps,nl}}$. Correspondingly the potential $\vec{V}^{\,\text{e}-\text{nuc}}$ and the $E^{\,\text{e}-\text{nuc}}$ are expressed as:

$$\vec{V}^{\,\text{e}-\text{nuc}} \;=\; \vec{V}^{\,\text{ps,local}} + \vec{V}^{\,\text{ps,nl}}\,,$$

and

$$E^{\,\text{e}-\text{nuc}} \;=\; E^{\,\text{ps,local}} + E^{\,\text{ps,nl}}\,. \tag{25}$$

The energy expressions Eq. (24) evaluated in `vofrho` subroutine becomes:

$$E = E^{\text{kin}} + E^{\text{H}} + E^{\text{sr}} - E^{\text{self}} + E^{\text{ps,local}} + E^{\text{ps,nl}} + E^{\text{XC}} = E^{\text{dipole}}\,, \tag{26}$$

where, for example, E^{sr} is the energy of screened ions-variable.

In the Kohn–Sham scheme [6] the electron density is expressed by a set of orthogonal, normalized Kohn–Sham orbitals $\phi_\alpha(\boldsymbol{r})$

$$n(r) = \sum_\alpha f_\alpha \left|\phi_\alpha(r)\right|^2 \,. \tag{27}$$

The occupation numbers f_α vary between 0 and 2 as the electron spin is not included explicitly and the sum over all occupation numbers is the total number of electrons N_{el} per super cell. The ground state electron density is calculated by solving the Kohn–Sham equation self-consistently for these orbitals

$$\underbrace{\left(\tfrac{1}{2}\nabla^2 + V^{\,\text{e}-\text{nuc}} + V^{\text{H}}[n] + V^{\text{XC}}[n]\right)}_{\vec{H}^{\text{KS}}}\phi_\alpha(r) = e_\alpha\,\phi_\alpha(r)\,. \tag{28}$$

The Kohn–Sham orbitals are represented by a plane-wave basis-set

$$\phi_{i,k}(r) = \sum_{G,\frac{1}{2}|G+K|^2 \le E_{\text{cut}}} c_{iG+K}\, e^{i(G+K)\,r} \tag{29}$$

truncated and a energy cut-off E_{cut}. The Brillouin zone integral over the k-points is replaced by a sum over a set of special k-points with the corresponding weights $w_{\boldsymbol{k}}$.

Once the ground state of the electrons is calculated the atomic EOMs are integrated with standard MD techniques. Two schemes are implemented — namely the Verlet algorithm and a predictor-corrector algorithm.

3.1. Construction of GNCPP'S

In electronic-structure calculations the desired functions are bare-ions pseudo-potentials based on local-density-functional theory (LDF) which depend on angular momentum l. The following functional from that enforces a r_{cl} — "core radius" or lower bound radius for unbound states

$$f\left(\frac{r}{r_{cl}}\right) = \exp\left[-\left(\frac{r}{r_{cl}}\right)^{\lambda}\right], \tag{30}$$

with $\lambda = 3.5$, has proven effective. Following the standard procedure, an intermediate pseudopotential is introduced

$$V_{1l}(r) = \left[1 - f\left(\frac{r}{r_{cl}}\right)\right] V(r) + c_l\, f\left(\frac{r}{r_{cl}}\right) \tag{31}$$

which converges to the full potential $V(r)$ for $r > r_{cl}$. The final pseudo-potential, found from analytically inverting the Schrödinger equation, is then [9]

$$\begin{aligned} V_{2l}(r) \; &= \; V_{1l}(r) + \gamma_l \delta_l r^{l+1} f\left(\frac{r}{r_{cl}}\right) / 2w_{2l}(r) \\ &\quad \times \left[\frac{\lambda^2}{r^2}\left[\frac{r}{r_{cl}}\right]^{2\lambda} - \frac{2\pi l + \lambda(\lambda+1)}{r^2}\left[\frac{r}{r_{cl}}\right]^{\lambda} + 2\varepsilon_l - 2V_{1l}(r)\right], \end{aligned} \tag{32}$$

where γ_l-scale factor for wave functions u_l, w_l reads $\gamma_l = u_l(R_l)/w_{1l}(R_l)$, $c_l^{(n)}$ are trial functions used in Eq. (31) which correspond to "eigenvalue" $\varepsilon_l^{(n)}$.

In contrast to the case of bound-state-only pseudopotentials found for several different atomic configurations, a single valence-electron electrostatic and exchange-correlation potential can be subtracted from V_{2l} to find the full set of ionic pseudopotentials,

$$V_l^{\text{ion}}(r) = V_{2l}(r) - \frac{4\pi}{r}\int_0^r \rho(r')r'^2 dr' - 4\pi \int_r^{\infty} \rho(r')r' dr' - \frac{\delta E_{\text{exc}}\left[\rho(r)\right]}{\delta\rho(r)}, \tag{33}$$

where $w_{1l}(r)$ is an intermediate wave function; $\lambda = 3.5$, $\rho(r) = \sum_l n_l \left(\frac{w_{2l}(r)}{r}\right)^2$ and n_l is the occupancy of bound valence state l.

In the case of rare gases, when all the electrons in the ground configuration are being treated as a core, there is no unscreening, and $V_l^{\text{ion}}(r) = V_{2l}(l)$. For computational convenience Kleinman and Bylander [6] proposed a fully separable form as mentioned above.

In the following it will be shown that this form could be reduced to the one more "understandable", especially for low charge number atoms like protons, alpha. Thus, in [10] as a canonical first step these algorithms generate angular momentum-dependent screened pseudopotentials $V_l^{\mathrm{eff}}\,[n_0]$ from a particular reference configuration, *e.g.*, the ground state of the neutral atom, assuming spherical screening. These act as effective potentials on the atomic pseudo valence states via the radial Schrödinger equations

$$\left(-\frac{1}{2}\frac{d^2}{dr^2}+\frac{l(l+l)}{2r^2}+V_l^{\mathrm{eff}}\,[n_0;r]-\varepsilon_l\right)rR_l(r)=0\,. \tag{34}$$

$V_l^{\mathrm{eff}}\,[n_0]$ contain a common spherical screening potential which is self-consistent with the total atomic charge density $n_0(r)$, comprised of the (pseudo) valence density n_0^v and the core charge density n_0^c obtained from the all-electron core states. The effective potentials can be decomposed rigorously into the Hartree potential V_{H} and the XC potential due to valence and core electrons and an angular-momentum-dependent bare potential V_l^{bare} which conveys the nuclear attraction and the Pauli repulsion due to the core states. For an arbitrary valence configuration one has

$$V_l^{\mathrm{eff}}\,[n;r]=V_l^{\mathrm{bare}}(r)+V_{\mathrm{H}}\,[n_0^c;r]+V_{\mathrm{H}}\,[n^v;r]+V_{\mathrm{XC}}\,[n^v+n_0^c;r]\;, \tag{35}$$

which, in the reference configuration ($n=n_0^v+n_0^c$), reduces of course, to the screened pseudopotentials.

The approximate procedure for dealing with exchange and correlation proposed by Kohn and Sham [11] has been adopted with V_{eff} given by

$$V_{\mathrm{eff}}(\vec{r})=-\frac{e^2}{r}+e^2\int d^3\,r'\,\frac{n(\vec{r'})}{|\vec{r}-\vec{r'}|}+\mu_{\mathrm{XC}}(n(\vec{r}))\,. \tag{36}$$

The first two terms of Eq. (3) constitute the electrostatic Hartree potential. The last term accounts for exchange and correlation by approximating the electron gas locally by a uniform gas of mean density $n(\vec{r})$, where $\mu_{\mathrm{XC}}(n_0)$ is the exchange-correlation part of the chemical potential for the uniform interacting electron gas of mean density n_0. This approximation, valid for slowly varying densities, will be poor near the proton. However, in this region V_{eff} will be dominated by the Hartree potential $\sim -e^2/r$. Therefore we suggest to use for hydrogen-like atoms (for other atoms the original formulation is kept) the pseudo-potential form as in Eq. (4) neglecting the non-local contributions at the level of the atom itself

$$pp=U_{\mathrm{D}}=\frac{1}{[4\pi\varepsilon_0]}\frac{Z_\alpha Z_{\mathrm{D}}e^2}{r}\qquad\text{for}\quad r\geq R\,. \tag{37}$$

Consequently, the complementary tool from the package FHIPP was modified (the package being of open source type), and the results of this modification for pseudo-potentials of alpha particles and protons are shown in Fig. 1. The start utility (FHIstart) of the code includes an automatic search for point group symmetries and the symmetry center in the system.

The input file `start.inp` used in FHIstart describes the geometry of the super cell, the configuration of the nuclei and parameters relevant for the MD simulation or the structure optimization, and the calculation of the electron ground state.

For each of the *nsp* atomic species one declares the properties of the pseudopotential ($zv, l_{\max}$ and $l_{\rm loc}$) and the radius of the screening charge $n_{i_{\rm s}}^{{\rm Gau}\beta}(r)$ (*rgauss*). The positions of the nuclei τ_0 and operationally also the velocities vau_0 follow the declaration (*c.f. npos*). Pseudopotential data need to be provided in the files `fort.11, fort.12`,... (see Table I) for the *nsp* atomic species as tabulated on a logarithmic mesh (parameter *tpsmesh*).

Output generated during the calculation is written to several files. The chief output file is `fort.6`. It contains a complete protocol of the initialization, the molecular dynamic simulation or the structure optimization on each step of the energy minimization. Partial results are presented on Table I.

4. Discussion of the results and conclusions

An analysis of the existing models and experiments on strong enhancement of alpha decay in metallic environments show that the Fermi–Thomas free gas electrons based models [4, 5] fail at high atom charge numbers (Z). By comparison a parametric Debye plasma model [1] which gives reliable results is theoretically controversial (low temperature and high electron density). Therefore, in the present paper we derive a screening potential from the Gamow decay theory in order to substitute the Hatree pseudo-potential in case of the hydrogen-like atoms (including alpha particle), that is subsequently used in density functional theory (DFT) calculations. The results of DFT code package FHI98MD for the system Pd lattice (bcc)+ $174\,{\rm Hf}_{72}$ +He (alpha) are given in Table I. From there on can remark the following:

In Fig. 1, if we look to position of atoms as given in Table I, it results that the distance between alpha particle and Hf nuclei (0.04 a.u.) is just the turning point when the kinetic alpha energy plus the "screening energy shift" equals the pseudo-potential (*pp*), thus confirming the following rule: the out of tunneling [4] is at about $a_0/Z_{\rm Hf}$, where the Hf atom radius $a_0 = 3.1$ a.u., but more that it could be considered at $\cong 0.01\,a_0$.

With Somerfeld factor as given by Eq. (11) equal 184 and U_e = 4215 a.u. × 27.2 = 116 keV (from code results, see Table I) and with enhancement factor as given by Eq. (16) $f_{\rm lab} = 95$, the half-life is shortened

TABLE I

Input file: Fort.11 & output file: Fort.6 (fragments)

Pd–He–Hf				
Lattice vectors				
a1		10.470000	0.000000	0.000000
a2		0.000000	10.470000	0.000000
a3		0.000000	0.000000	10.470000
Reciprocal lattice vectors				
b1		1.000000	0.000000	0.000000
b2		0.000000	1.000000	0.000000
b3		0.000000	0.000000	1.000000
Atomic positions τ_0*:*				
Species	No.	x	y	z
Palladium	1	−2.6175	−2.6175	0.0000
Palladium	2	2.6175	2.6175	0.0000
Palladium	3	2.6175	−2.6175	5.2350
Palladium	4	−2.6175	2.6175	5.2350
Helium	1	0.0000	0.0000	2.6175
Hafnium	1	0.0000	0.0000	2.6594

Number of k-points: $n_{\mathrm{kpt}} = 1$
internal energy at zero temperature = −4215.495899 a.u.
Kohn–Sham orbital energy = −4654.635696 a.u.

Pd–proton–proton				
Atomic positions τ_0:				
Species	No.	x	y	z
Palladium	1	−2.6175	−2.6175	0.0000
Palladium	2	2.6175	2.6175	0.0000
Palladium	3	2.6175	−2.6175	5.2350
Palladium	4	−2.6175	2.6175	5.2350
Hydrogen	1	0.0000	0.0000	2.6175
Hydrogen	2	0.0000	0.0000	2.6384

internal energy at zero temperature = −21.848761 a.u.

case Pd–Hf–proton				
Atomic positions τ_0:				
Species	No.	x	y	z
Palladium	1	−2.6175	−2.6175	0.0000
Palladium	2	2.6175	2.6175	0.0000
Palladium	3	2.6175	−2.6175	5.2350
Palladium	4	−2.6175	2.6175	5.2350
Hydrogen	1	0.0000	0.0000	2.6175
Hafnium	1	0.0000	0.0000	2.6594

internal energy at zero temperature = −2041.224846 a.u.

to 2.0×10^{15} years/95 = 2.0×10^{13} years, unfortunately this is an unseizeable effect at this scale for the chosen isotope, so other reactions need to be considered in order to be proven by experiment, the present case could be viewed as an exercise. If we apply formula from Eq. (18), we obtain

$$U_{\rm D} = 2 \times 72 \times 300\,{\rm eV} \times (290/4)^{1/2} = 2 \times 72 \times 0.3 \times 8.5 = 355 \text{ KeV}$$

or 42 keV without temperature correction. Quite different values are explained due to temperature correction in formula but not considered in the code.

In the case of the isotopes sensitive to beta$^+$ decay, when a proton from inside the nuclei + energy converts into a neutron with a positron release (e^+), or the same beta$^+$ decay plus an electron capture from inner shell neutrons when neutrinos are released, like in a reaction $176\,{\rm Lu}_{71} \to 176\,{\rm Hf}_{70}$, 1.316 MeV, 3.666 h. It follows from [1] that in an experiment realized by proton bombardment in the range: 0.8 MeV÷1.4 MeV of ${\rm PdLu}_{10\%}$ alloy, the measured neutrons yield, implies an enhancement factor of 2.5 around $E_{\rm pr} = 0.81$ MeV that may be attributed to Debye electrons screening of $U_{\rm D} = 33\,{\rm keV}$. In the case of code application with proton inside nuclei the value of screening energy shift is equal $2041. \times 27.2 = 54\,{\rm keV}$, and with Somerfeld factor of 39, $f_{\rm lab} = 1.29$, and the half-life is shortened to 8.09/1.29 h, so the values are comparable with the experiment.

In case of fusion reaction $d + d$ after deuterons implantation in Pd, the code gives $20 \times 27.2 = 544\,{\rm eV}$ which is in the range of experimental values (800 eV) [3] for Pd.

Now, it becomes clear that the Debye formula for data parametrization as advanced by Kettner *et al.* [1] agrees well with experiments, the question is why this works so well. One of the major motivations of the present work is to find an answer to this question. If we look at the formula for $U_{\rm D}$, we can observe that it could be split in two parts: the part due to the screening at an "early" turning point of Debye length Eq. (21) $\lambda_{\rm D} = 1/k_0$ in place of b, so $b = \lambda_{\rm D} \equiv 1/\sqrt{\frac{\rho e^2}{\varepsilon_0 k_{\rm B} T}}$, or at such $1/b$ which accounts for overall "interaction" as coming from the "plasma sea" of free electrons of the metallic environment as described by a density $\rho_a\, n_{\rm eff}$; and the second part which accounts for the alpha particle already screened till the reduced Debye length by a type of interaction suggested by Coulomb potential introduced by Gamow Eq. (5).

By repeating the calculus of Gamow factor (Eq. (2)–(8)) with WKB integration limit $b = \lambda_{\rm D}$ we obtain:

$$V = E + U_{\rm D} = Z_1 Z_2 e^2 4\pi\varepsilon_0 \lambda_{\rm D}\,,$$

with

$$V(r) = \frac{\lambda_{\rm D}(E + U_{\rm D})}{r}\,, \qquad E + U_{\rm D} = \frac{1}{2}\, m_{\rm red} v_{\rm relativ}^2 v_{\rm relativ}\,,$$

where v_{relativ} is the relative velocity of fragments, m_{red} is the reduced mass of disintegrating system. Screening is then governed by:

$$\begin{aligned}
G_{\mathrm{scr}} &= \frac{2}{\hbar}\sqrt{2m_{\mathrm{red}}U_{\mathrm{D}}}\int\limits_{\cong 0}^{\lambda_{\mathrm{D}}}\left[\frac{\lambda_{\mathrm{D}}}{r}-1\right]^{1/2}dr\,,\\
G_{\mathrm{scr}} &\approx \frac{4Z_\alpha Z_{\mathrm{D}}e^2}{[4\pi\varepsilon_0]\,\hbar v_{\mathrm{relativ}}}\left(\frac{\pi}{2}-2\sqrt{\frac{\cong 0}{\lambda_{\mathrm{D}}}}\right)\cong 2\pi\frac{Z_\alpha Z_{\mathrm{D}}e^2}{\hbar v_{\mathrm{relativ}}}\,,\\
2\pi\eta &= 31.29\,Z_\alpha Z_{\mathrm{D}}\frac{m_{\mathrm{red}}}{(E+U_{\mathrm{D}})^{1/2}}\,.
\end{aligned}$$

So, the enhancement of the cross section $\sigma(E)=\frac{S(E)}{E}\,e^{-2\pi\eta(E)}$ is then simply like in [12].

$$\begin{aligned}
f_{\mathrm{lab}} &= \frac{\sigma(E+U_d)}{\sigma(E)}=\frac{S(E+U_{\mathrm{D}})}{S(E)}\,\frac{E}{E+U_{\mathrm{D}}}\,\frac{\exp[-2\pi\eta(E+U_{\mathrm{D}})]}{\exp[-2\pi\eta(E)]}\\
&\approx \exp\left\{\pi\eta(E)\frac{U_{\mathrm{D}}}{E}\right\}\,,
\end{aligned}$$

The penetrability factor through external barrier is $T_{\mathrm{scr}}=\exp(-2\pi\eta)$.

Finally, the energy shift is

$$U_{\mathrm{D}}=\frac{Z_1Z_2e^2}{4\pi\varepsilon_0\lambda_{\mathrm{D}}}\cong 2.09\times 10^{-11}Z_1Z_2\left(\frac{\rho}{T}\right)^{1/2}[\mathrm{eV}]\,, \tag{38}$$

where $e^2=1.4399652\,\mathrm{MeV\,fm}$, for example for metals $\rho\cong 6\times 10^{28}m^{-3}$ and $T=293\,\mathrm{K}$, for two deuterons: $U_{\mathrm{D}}=2.09\times 10^{-11}\times 1\times 1\times 2.4\times 10^{14}/17\cong 370\,\mathrm{eV}$ and for Hf-alpha decay and $T=1\,\mathrm{K}$, and $U_{\mathrm{D}}\equiv 2.09\times 10^{-11}\times 2\times 74\times 2.4\times 10^{14}/1\cong 148\,\mathrm{keV}$. The values calculated by the DFT code are comparable with the ones above if we consider the temperature correction.

Therefore, the present DFT code application by using a modified pseudopotential in accord with the decay theory because it describes much better the overall neighboard interactions, that permits to furnish the correct results, and finally it could be applied for other different atoms systems, except the atoms containing f electron shells. Due to the present simple formula, it has been constructed on the basic physical assumptions and can be used to approximate quickly the screening energy "shift" which enters in the final expression of the enhancement decay factor of alpha, beta$^+$ decay type isotopes in metallic environment.

If these kind of predictions continue to be verified by appropriate experiments, one may have a cheap solution to remove the transuranic waste (involving all alpha-decay) of used-up nuclear fuel rods of fission reactors in a time period of a few years.

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