

Pair Interaction Energy of Hydrogen Isotopes in Metallic Lattices. Estimate of Fusion Rates.

F. MARCHESONI, C. PRESILLA and F. SACCHETTI

Dipartimento di Fisica, Università di Perugia - I-06100 Perugia, Italy

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Abstract. – The fusion rates for the reactions pd , dd and pp in a solid-state environment are determined under the assumption that the fusion process is mediated by bound states of the reactant pair. An effective binding potential which accounts for lattice, electronic and pair interactions is envisaged to model plausible equilibrium and nonequilibrium conditions. Our estimates for the upper-bound fusion rates in condensed matter lie well below the actual experimental limits.

Recent claims for surprisingly intense neutron production in electrolytic cells operated with heavy water at room temperature during the process of deposition of deuterium on palladium [1, 2] and titanium [2] cathodes revived the interest for the study of nuclear fusion reactions in condensed matter [3]. The consensus on the validity of the experimental data made available is still elusive. Deep concern has been expressed due to the lack of i) γ emission related with the neutron-capture process in the spectrometer medium [4]; ii) energetic charged particle production which would characterize most fusion reactions involving deuterium (d) and hydrogen nuclei (p) [5]; iii) reproducible experimental conditions (namely, the cathode preparation) and quantitative estimates of other direct or indirect neutron sources [6].

In the absence of further experimental evidence most theoretical work assumes the conservative value of 10^{-23} s^{-1} for the dd fusion rate in Pd [2]. The collisional mechanisms that have been advocated [7, 8] to overcome the Coulomb barrier, possibly screened by many-body effects [9], would require too a high activation energy of the reactants (three or four orders of magnitude larger than the relevant thermal energy) to be plausible. A different approach is based on the assumption that the reactant pair forms a bound state in the solid-state environment, the size and energy of which crucially depend on both the static and dynamical properties of the lattice. No appreciable fusion rates have been predicted for deuterium in bulk Pd at equilibrium [10, 11], whereas rigorous upper bounds to the fusion rates obtainable under diverse lattice configurations may be set following the discussion of ref. [12].

In the present letter we model the effective pair potential for couples of interacting pointlike nuclei with positive charge $+e$ in the presence of the electronic cloud and the screened ions of the metallic lattice. On imposing spherical symmetry we determine a lower bound to the actual pair potential, which can be solved numerically to calculate the upper

bound to the fusion rate of the reactions [9]:



The electronic charge density has been varied to assess the efficacy of a number of non-equilibrium mechanisms which might occur during the process of deposition of deuterium on the metallic cathode.

For the sake of simplicity we specialize our model to the study of nuclear fusion in Pd. Metallic Pd presents a f.c.c. lattice with spacing $l_0 \approx 4 \text{ \AA}$. Hydrogen nuclei, d and p, tend to occupy the octahedral sites of the f.c.c. cell. Introducing a pointlike nucleus with charge $+e$ in any point \mathbf{x}_1 of the lattice determines a local polarization of the unperturbed electronic charge density $n_0(\mathbf{x})$. Confining ourselves, for simplicity, to the linear response approximation and using the Thomas-Fermi expression for the dielectric function [13], the induced variation of $n_0(\mathbf{x})$ is given by

$$\delta n_1(|\mathbf{x} - \mathbf{x}_1|) = \frac{k_{\text{TF}}(\mathbf{x}_1)^2 \exp[-k_{\text{TF}}(\mathbf{x}_1)|\mathbf{x} - \mathbf{x}_1|]}{4\pi |\mathbf{x} - \mathbf{x}_1|}, \quad (4)$$

where $k_{\text{TF}}(\mathbf{x})^2 = 12\pi(3\pi^2)^{-2/3} n_0(\mathbf{x})^{1/3}/a_0$, $a_0 = 0.529 \text{ \AA}$ being the Bohr radius. $k_{\text{TF}}(\mathbf{x})$ has been taken constant over the local length scale $n_0(\mathbf{x})^{-1/3}$ and δn_1 has been normalized to one, the metallic lattice being treated as an infinite reservoir of electronic charge.

Let us consider, now, the case of two such charged nuclei located in the arbitrary points \mathbf{x}_1 and $\mathbf{x}_2 = \mathbf{x}_1 - \mathbf{R}$ of the lattice. Within the adiabatic approximation the total energy E_{tot} of the system formed by the bare nuclei and the polarized electronic charges δn_1 and δn_2 can be separated in six distinct contributions:

$$E_{\text{tot}} = E_{12} + E_1^s + E_2^s + E_{\text{ne}} + E_1^l + E_2^l, \quad (5)$$

E_{12} , E_1^s , E_2^s , E_1^l and E_2^l are electrostatic terms which denote the pair interaction, the self-energies and the lattice interaction of the reactants, respectively. E_{ne} represents the sum of the kinetic and interaction energies of the polarization clouds. E_{tot} depends on $n_0(\mathbf{x}_1)$ and $n_0(\mathbf{x}_2)$ and on the relative coordinate \mathbf{R} . The interaction between two diffusing nuclei is favourite in the directions connecting adjacent octahedral sites. We simplify our problem by determining the interaction energy along such directions, only.

The electrostatic energy related to the interaction of the reactants with the electronic cloud can be calculated explicitly:

$$E_{12} + E_1^s + E_2^s = \frac{1}{2} \iint d\mathbf{x} d\mathbf{y} \frac{\rho(\mathbf{x})\rho(\mathbf{y})}{|\mathbf{x} - \mathbf{y}|}, \quad (6)$$

with

$$\rho(\mathbf{x}) = e\delta(\mathbf{x} - \mathbf{x}_1) + e\delta(\mathbf{x} - \mathbf{x}_2) - e\delta n_1(|\mathbf{x} - \mathbf{x}_1|) - e\delta n_2(|\mathbf{x} - \mathbf{x}_2|). \quad (7)$$

On introducing the expression (4) for δn_1 and δn_2 into eq. (6) we obtain the electrostatic pair-interaction potential in an analytical form

$$E_{12} = -\frac{3}{4} e^2 (k_{\text{TF1}} + k_{\text{TF2}}) + \frac{e^2}{R} \frac{k_{\text{TF2}}^2 \exp[-k_{\text{TF2}}R] - k_{\text{TF1}}^2 \exp[-k_{\text{TF1}}R]}{k_{\text{TF2}}^2 - k_{\text{TF1}}^2}, \quad (8)$$

where k_{TF1} and k_{TF2} denote $k_{TF}(\mathbf{x}_1)$ and $k_{TF}(\mathbf{x}_2)$, respectively. Finally, the energy contribution due to the interaction of nuclei with the screened Pd atoms of the lattice follows immediately the Gauss theorem:

$$E_k^l = \sum_i \int_{R_i}^{\infty} dz \frac{e^2}{z^2} \left[Z_{Pd} - \int_0^z dy 4\pi y^2 n_{Pd}(y) \right], \quad (9)$$

where R_i is the distance between the point \mathbf{x}_k ($k = 1, 2$) and the i -th lattice site, $Z_{Pd} = 46$ is the atomic number and n_{Pd} the spherically symmetric electronic charge density of a free Pd atom [14].

The nonelectrostatic term E_{ne} has been studied extensively in the literature [15]. E_{ne} can be computed as the energy difference

$$E_{ne} = \int d\mathbf{x} \{ \varepsilon[n(\mathbf{x})] n(\mathbf{x}) - \varepsilon[n_0(\mathbf{x})] n_0(\mathbf{x}) \} \quad (10)$$

between the perturbed and unperturbed electronic cloud with densities n and n_0 obeying the identity

$$n(\mathbf{x}) = n_0(\mathbf{x}) + \delta n_1(|\mathbf{x} - \mathbf{x}_1|) + \delta n_2(|\mathbf{x} - \mathbf{x}_2|). \quad (11)$$

On assuming that n_0 has cylindric symmetry parallel to the directions joining two adjacent octahedral sites, the integral in (11) reduces to a double two-centre integral which must be computed numerically. The functional $\varepsilon[n]$ is the energy per particle of an interacting electron gas whose density is n . $\varepsilon[n]$ is usually written as the sum of various terms [16]:

$$\varepsilon[n] = \frac{3}{10} e^2 a_0 (3\pi^2 n)^{2/3} - \frac{3}{4\pi} e^2 (3\pi^2 n)^{1/3} + \varepsilon_c[n]. \quad (12)$$

The effective interaction potential $V(R)$ is defined as

$$V(R) = E_{12} + E_{ne} + E_{1/2}^l, \quad (13)$$

where the particle 1 sits in an octahedral site and the other, at distance R , is free to move along a direction connecting the first particle to an adjacent octahedral site. In fig. 1 we display the equilibrium electronic charge density n_0 in bulk Pd, $n_0(\mathbf{x}) \sum_i n_{Pd}(|\mathbf{x}_i - \mathbf{x}|)$ (\mathbf{x}_i being the i -th lattice site coordinates). The resulting effective potential, plotted in fig. 2, admits of a minimum at $R_0 = 5.34a_0$ with binding energy 1.23 eV.

The actual pair potential in the Pd lattice is bounded below by the spherically symmetric potential $V(R)$ [12]. This allows to determine upper bounds to the rates Γ of the fusion reactions (1)-(3). Γ can be calculated according to the well-known formula $\Gamma = \sigma v \rho$, where σ , v and ρ denote, respectively, the cross-section of the relevant process, the relative velocity and the density of the reactants [9]. In the case when the reaction studied involves low activation energies (cold fusion), only the bound states of $V(R)$ contribute (binding energy $0 \leq E \leq 1.23$ eV). In this case we interpret σ as the intrinsic cross-section of the nuclear fusion process, σ_N , and ρ as the probability for the annihilating pair to come to zero relative separation, *i.e.* $\rho = |\psi_E(0)|^2$, where $\psi_E(R)$ is the s -wave eigenfunction of the spherically symmetric potential $V(R)$ with energy E . In our calculations $\psi_E(R)$ have been computed numerically and the product $\sigma_N v$ approximated to its zero velocity limit $A(0)$ [9]. In the case of high-activation energies $E > 1.23$ eV (microscopically hot fusion), instead, we choose σ to define the cross-section for the total process including the penetration of the repulsive

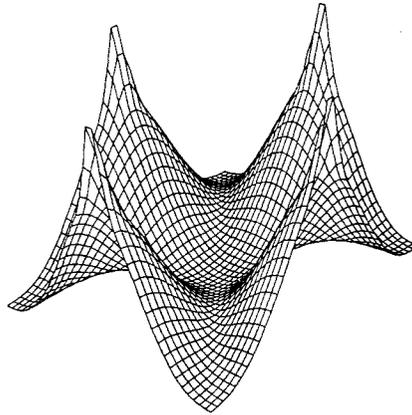


Fig. 1. - Electronic charge density $n_0(\mathbf{x})$ with $\mathbf{x} \equiv (x, y, l_0/2)$ and $0 \leq x, y \leq l_0$ for bulk Pd (logarithmic vertical scale). The five minima are the octahedral sites.

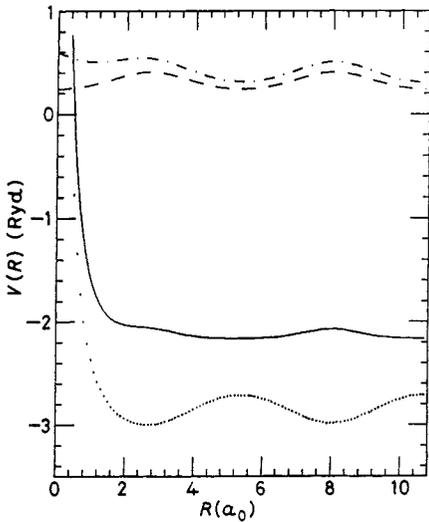


Fig. 2.

Fig. 2. - Potential of eq. (13) calculated along the direction joining two octahedral sites of fig. 1. $V(R)$ (solid line) is the sum of E_{12} (dots), E_{ne} (dot-dashed line) and $E_{\frac{1}{2}}$ (dashed line).

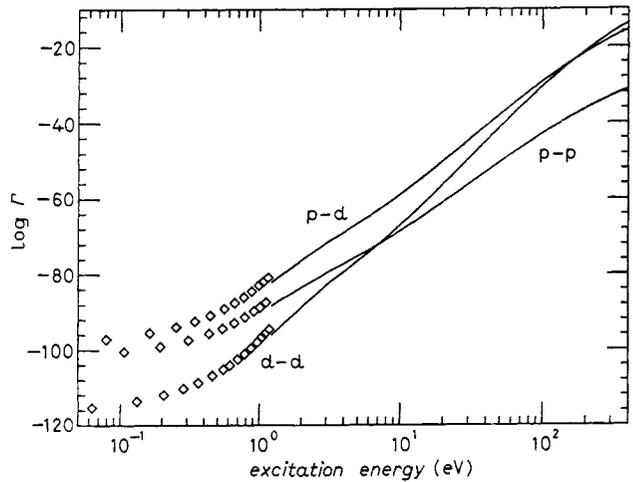


Fig. 3.

Fig. 3. - Fusion rate upper bounds for the processes (1)-(3) at different pair energy E .

Coulomb barrier, σ_T , and, consequently, ρ represents the macroscopic density of the reactants deposited in the Pd lattice. On calculating the penetration factor explicitly, $\Gamma = S(0)/EG(E)v\rho$, where $S(0)$ is the zero velocity limit of $\sigma_T v$ ($S(0) = A(0)\pi\mu e^2/\hbar$, μ being the reduced mass of the reactant pair), $v = \sqrt{2E/\mu}$ and the Gamow factor $G(E)$ depends on the shape of the pair potential $V(R)$:

$$G(E) = \exp \left[-\frac{2}{\hbar} \int_{R_c}^0 dR \sqrt{2\mu(V(R) - E)} \right], \tag{14}$$

with $V(R_c) = E$.

Our results for the fusion reactions (1)-(3) are plotted in fig. 3. Most notably, i) nuclear fusion rates in bulk Pd at equilibrium lie well below the values estimated from the experimental data of ref. [2]. In particular comparable values for Γ can only be obtained by advocating unplausible collisional mechanisms that would involve pair energies as large as 160 eV [7, 8]. ii) An interesting signature of cold nuclear fusion would be an abundant production of energetic positrons due to the fusion of pp pairs, eq. (1). Such an effect has not been observed, so far [5].

The very low fusion rates in fig. 3 are a consequence of the large internuclear separation, $R_0 \approx 5.34a_0$, of the reactant pairs in bulk Pd at equilibrium [10]. However, we observed that the effective potential $V(R)$ strongly depends on the choice of the electronic charge density. Large spatial variations of the density on a short length scale supersede the binding action of the lattice and may bring the reactant nuclei to close distances. In fig. 4 we plotted the

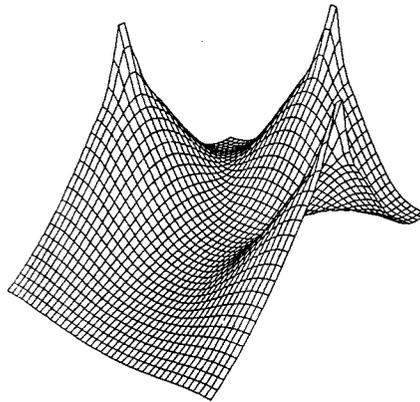


Fig. 4. - As in fig. 1 but assuming the plane $x = l_0/2$ as an ideal surface.

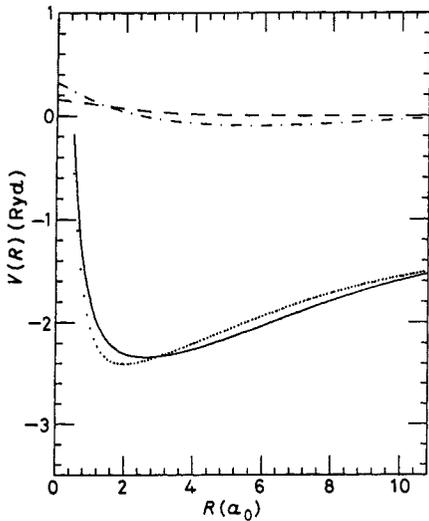


Fig. 5.

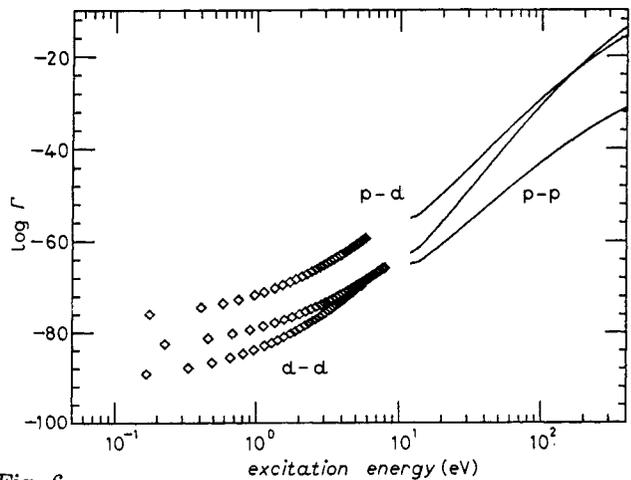


Fig. 6.

Fig. 5. - Potential (13) for $x \geq l_0/2, y = l_0/2, z = l_0/2$. Notation is as in fig. 2.

Fig. 6. - Fusion rate upper bounds for the potential of fig. 5.

electronic density $n_0(\mathbf{x})$ at the surface of an ideal Pd lattice. That to model the largest spatial variation of $n_0(\mathbf{x})$ which is expected to occur in a *stationary* lattice configuration (including dislocations, cavities, defects, etc.). The natural length scale of the electronic density variations associated with such phenomena is the lattice spacing l_0 . The corresponding pair potential $V(R)$ and the fusion rates for processes (1)-(3) are displayed in fig. 5 and 6, respectively. The potential is calculated as in the bulk case with the particle 1 fixed in the outermost octahedral site and the particle 2 at distance R in the vacuum. The minimum of the potential $V(R)$ is now located at a shorter relative distance, $R_0 \approx 2.5a_0$, and exhibits a larger binding energy, 12 eV. Accordingly, the cold fusion rates, which involve the bound states of the reactants pairs, are enhanced by up to 23 orders of magnitude, whereas no significant variation can be expected for the «in flight» processes (*i.e.* in the continuum of the energy spectrum, $E > 12$ eV). However, *no stationary* lattice mechanism seemingly suffices to make cold fusion processes detectable by the apparatuses employed in ref. [1, 2, 5, 6].

Finally, we observe that the occurrence of cold nuclear fusion in the Pd lattice would require large fluctuations of n_0 on spatial scales of fractions of l_0 and relatively long-time scales to allow for the formation of the closer bound states of the reactant pairs. We could speculate about various nonequilibrium situations allowing for the above condition but, at present, none appear to be favourite because of the lack of clear experimental evidence.

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