Letter

Deuteron-deuteron nuclear reactions at extremely low energies

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The theoretical analysis of the ^{2}H$(d, p)^3$H reaction taking place in both metallic as well as gaseous targets at deuteron energies of several keV presented here indicates a strong contribution of the single-particle 0^+ threshold resonance. Additional arguments based on the weak coupling between the 2 + 2 and 3 + 1 clustering states of the compound nucleus ^{4}He support this resonance and suggest its large partial width for the internal electron-positron pair formation that overestimates the proton width. A detailed study of the interplay between the resonance transition and the electron screening effect enables estimation of the nuclear reaction rate of the deuteron-deuteron fusion reactions at room temperature.

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Introduction. Observation of an anomalously large amount of energy in the electrolysis of heavy water reported by Fleischman and Pons over thirty years ago was interpreted as a result of deuteron-deuteron (DD) fusion reactions in the Palladium electrode [1,2]. However, the lack of experimental reproducibility and expected nuclear reaction products observed in accelerator experiments at higher deuteron energies caused strong skepticism about the data. It was based mainly on two theoretical arguments. First of all, the penetration through the Coulomb barrier of a height of about 350 keV in the deuteron-deuteron system was supposed to be more than 40 orders magnitude too low to explain the energy production reported [3,4]. Additionally, the branching ratio of the DD reactions known from accelerator experiments [5,6] down to few keV should favorize the mirror neutron and proton channels being almost of the same strength. The ^{4}He channel resulting from the $E2$ deuteron radiative capture is seven orders of magnitude weaker than the nucleon channels [7]. In room temperature experiments, the branching ratios seem to be opposite: the measured heat excess is directly correlated to production of ^{4}He in the absence of any gamma radiation [8], and the neutron channel should be much weaker [2]. These observations, also confirmed in the last experiments [9], could not yet be theoretically explained in a consistent manner [10].

In particular, many attempts have been made to overcome the Coulomb barrier problem and enhance the tunneling probability. These include catalyzing the fusion by muons or antiprotons [11], driving cusps [12], spreading wave packets [13], coherent correlated states [14], Bose-Einstein condensation [15], or a scalar field contribution [16]. However, none of the above effects could either be confirmed experimentally or potentially strong enough to explain the results obtained for various metallic systems. Therefore, the electron screening effect observed in the DD reactions taking place in different metallic targets [17,18] seems to have great advantages.

This phenomenon was already anticipated for classical plasmas [19] in the 1950s and observed 30 years later in nuclear reactions [20] preceding on gaseous targets. The shielding of nuclear charges by surrounding electrons leads to a reduction of the Coulomb barrier and consequently, to an exponential-like increase of its penetrability for lowering projectile energies, which was found especially strong in metallic targets [17]. Mathematically, the screened nuclear reaction cross section can be determined as follows:

$$\sigma_{\rm scr}(E) = \frac{1}{\sqrt{E(E+U_e)}} S(E) \exp\left(-\sqrt{\frac{E_G}{E+U_e}}\right) = \frac{1}{\sqrt{EE_G}} P(E+U_e) S(E), \quad (1)$$

where $S(E)$ is the astrophysical S factor, and the s-wave penetration factor through the Coulomb barrier $P(E)$ is given by

$$P(E) = \sqrt{\frac{E_G}{E}} \exp\left(-\sqrt{\frac{E_G}{E}}\right). \quad (2)$$

Here, E and E_G stay for the center-mass energy and the Gamow energy equal to $E_G = \frac{2\pi^2}{137^2}(Z_1Z_2)^2\,\mu$, respectively. Z_1 and Z_2 are atomic numbers of colliding nuclei, and μ is the reduced mass.

To describe the screening effect, the screening energy U_e corresponding to reduction of the Coulomb barrier height can be simply added to the energy in the expression for the penetration factor [18]. The experimental value of U_e can be then determined by fitting the enhancement of the screened cross section measured at low projectile energies for reactions taking place in the target medium compared to the bare nuclear

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case. The electron screening plays a very important role in dense astrophysical plasmas of white dwarfs or giant planets, where nuclear reaction rates can be even increased by many orders of magnitude [21]. To study this effect under terrestrial conditions, the DD reactions preceding on metallic targets seemed to be an ideal tool because of a low Coulomb barrier and the relatively large S factor. The first measurements performed for metallic targets [17] showed that the experimental screening energies for heavy metals of about 300 eV exceeded at least three times the theoretical expectations and were larger by an order of magnitude than the value $U_e = 20 \pm 5$ eV obtained previously for the gaseous target [5]. These results were generally confirmed by other authors [22–24], although a strong variation of screening energies determined under different experimental setups could be observed [25]. The main reason for that was contamination of the target surface by oxygen and carbon [26]. Thus, application of the ultrahigh vacuum technique was necessary and led to the experimental screening energy $U_e = 120 \pm 7$ eV for the Zr target [27] which is now very close to the theoretical value of 110 eV predicted within the self-consistent dielectric function theory [18,26]. The discrepancy with the results obtained previously under worse vacuum conditions could be explained by the small target contamination which induces target lattice defects and, consequently, raises the effective electron mass and the screening energies to the values of 300–400 eV [27].

The last precise measurements on the Zr target also revealed another effect—the energy dependence of the measured enhancement factor could not be described only by the electron screening effect. An additional contribution has been postulated due to the threshold 0^+ resonance in the compound nucleus ^{4}He at the excitation energy of about 23.85 MeV. Despite its DD single-particle structure, the total resonance width should be very small (on the order of 1 eV) because of very low probability for tunneling through the Coulomb barrier, making it very difficult for direct experimental observation. On the other hand, this resonance may change the branching ratios of the DD reaction in the vicinity of the reaction threshold in favor of those observed at thermal energies and thus contribute to solving the cold fusion puzzle.

In the present Letter, additional experimental and theoretical arguments supporting the existence of the threshold resonance in ^{4}He will be given, and consequences for the DD reactions at extremely low energies down to room temperature will be discussed.

Observation of the DD threshold resonance. The DD reaction branching ratios at very low deuteron energies could be changed by this threshold resonance significantly due to its internal structure, expressed generally by partial resonance widths:

$$\Gamma_{\rm tot}(E) = \Gamma_d(E) + \Gamma_p + \Gamma_n + \Gamma_{\rm em}. \tag{3}$$

The total resonance width $\Gamma_{\rm tot}(E)$ is a sum of a deuteron, proton, neutron, and electromagnetic partial widths, and is strongly energy dependent because of the penetration factor in the deuteron channel [28] (other partial widths can be considered constant for high energies of emitted particles):

$$\Gamma_d(E) = 2k\, a\, P(E)\frac{\hbar^2}{\mu a^2}|\theta_d|^2, \tag{4}$$

where k denotes the deuteron wave number, and a and μ stand for the channel radius and the reduced mass, respectively. θ_d is the reduced resonance width of the deuteron channel and takes on a maximum value equal to unity since the single-particle resonance structure is assumed. Whereas the deuteron width dominates the total resonance width at keV energies, it is much smaller compared to other partial widths of Eq. (3) at eV energies. The resonance cross section for the ^{2}H $(d, p)^3$H reaction can be simply expressed by the Breit-Wigner formula:

$$\sigma_{\rm res} = \frac{\pi}{k^2}\,\frac{\Gamma_d\Gamma_p}{({\rm E}-{\rm E_{res}})^2 + \frac{1}{4}\Gamma_{\rm tot}^2}. \tag{5}$$

As discussed previously [27], this resonance should have spin and parity assignment $J^\pi = 0^+$ and can interfere in the total cross section with other 0^+ resonances of ^{4}He which are generally very broad and result in a structureless (flat) excitation function $\sigma_{\rm flat}$. Thus, the coherent 0^+ contribution to the total cross section can be expressed as follows:

$$\sigma_{\rm tot}^{0+} = \sigma_{\rm flat}^{0+} + \sigma_{\rm res} + 2\,\sigma_{\rm flat}^{0+}\,\sigma_{\rm res}\cos\left(\varphi_{\rm flat}^{0+} - \varphi_{\rm res}\right), \tag{6}$$

where $\varphi_{\rm res}$ is the resonance phase shift given by

$$tg\varphi_{\rm res} = \frac{\Gamma_{\rm tot}}{2({\rm E}-{\rm E_{res}})} \approx \frac{\Gamma_{\rm tot}}{2{\rm E}}. \tag{7}$$

The last approximation in the formula above can be adopted for keV deuterons used in accelerator experiments if the resonance energy is of eV. $\varphi_{\rm flat}^{0+}$ represents the nuclear phase shift of the $\alpha_0 = \langle ^1S_0|0^+|^1S_0\rangle$ transition matrix element [27]. In the past, the total (flat) cross sections, vector, and tensor analyzing powers for the ^{2}H $(d, n)^3$He and ^{2}H $(d, p)^3$H reactions could be parametrized [29] using 16 independent transition matrix elements and their nuclear phase shifts for the deuteron energies up to 500 keV. According to that, the α_0 transition matrix is responsible for about 1/3 of the total cross section at the deuteron energies below 50 keV.

The experimental data at very low energies are usually presented by means of enhancement factors that are defined either as a ratio of the experimentally determined cross section $\sigma_{\rm scr}$ (or S factor), undergoing the screening effect and the theoretical value expected for bare nuclei $\sigma_{\rm bare}$ or as a ratio of the corresponding thick-target yields:

$$f(E) = \frac{\sigma_{\rm scr}(E)}{\sigma_{\rm bare}(E)} = \frac{S_{\rm scr}(E)}{S_{\rm bare}(E)},$$
$$F(E) = \frac{Y_{\rm scr}(E)}{Y_{\rm bare}(E)} = \frac{\int_E^0 \sigma_{\rm scr}(E)\left(\frac{dE}{dx}\right)^{-1}dE}{\int_E^0 \sigma_{\rm bare}(E)\left(\frac{dE}{dx}\right)^{-1}dE}. \tag{8}$$

The fitting procedure of the thick-target enhancement factor F for the ^{2}H $(d, p)^3$H reaction taking place in the Zr target is described in detail in Ref. [27]. Here, for simplicity, a linear parametrization of the astrophysical S factor [30] was used, which agrees very well with the transition matrix approach [29] in the studied energy range. The results are presented in

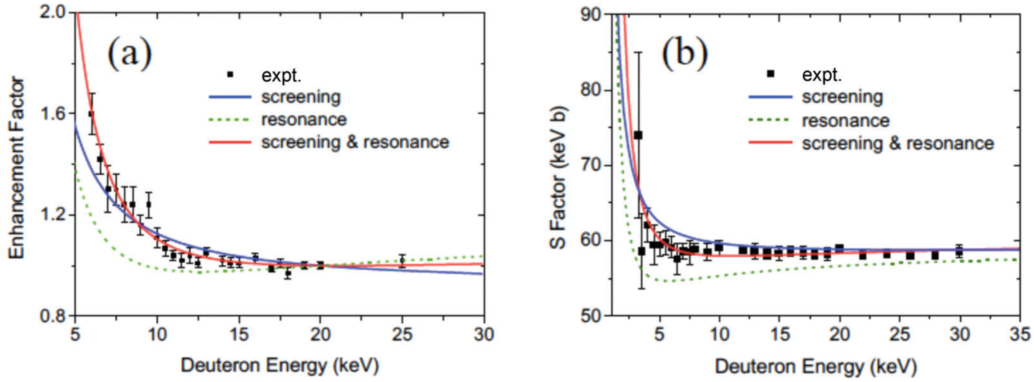

FIG. 1. The DD 0^+ threshold resonance contribution observed in the ^{2}H $(d, p)^3$H reaction. (a) Thick-target enhancement factor obtained for the Zr target. (b) S factor obtained for the gaseous target.

Fig. 1(a) and are very close to those obtained previously. The blue curve represents only the electron screening effect fitted by the screening energy U_e, whereas the red one takes into account also the resonance contribution described additionally by parameters Γ_p and $\varphi_{\text{flat}}^{0+}$ according to Eq. (6). The three-parameter fit gives the following values: $\Gamma_p = 40 \pm 15$ meV, $\varphi_{\text{flat}}^{0+} = 116 \pm 40^\circ$, and $U_e = 109 \pm 30$ eV. The phase shift determined is very close to the value 110° given by Paetz gen. Schieck [29]. If the phase shift is fixed to the latter value, the other two parameters are $\Gamma_p = 40 \pm 10$ meV and $U_e = 115 \pm 15$ eV. The experimentally estimated screening energy is now in very good agreement with the theoretical prediction of 112 eV [18], and the proton partial width is very small supporting the assumption made here about the DD single-particle structure of the 0^+ threshold resonance.

From the theoretical point of view, the resonance contribution should be also visible in the experimental data obtained many years ago by Greife *et al.* [5] studying the DD reactions on the gas target. In this case, the theoretical screening energy should be about 20 eV [31]. The experimental S factors together with theoretical fits are depicted in Fig. 1(b). Similar to the metallic Zr target, one can observe again that the pure screening curve cannot describe the experimental data correctly. If the screening energy is set to 20 eV and the destructive interference of the threshold resonance can be included, one gets $\Gamma_p = 10 \pm 2$ meV and $\varphi_{\text{flat}}^{0+} = 105 \pm 5^\circ$. This new result confirms that the characteristic shape of the Zr enhancement curve is not a crystal lattice effect, but rather relates to individual atoms.

Whereas the phase shifts in both cases agree very well, the proton resonance width estimated for Zr is clearly larger than that for a gaseous target. Here, the thermal broadening of the resonance of such a small width can play an important role. The room temperature broadening can just correspond to the width obtained for the gas target equal to 10 meV. Then, the higher value of the Zr target might be understood as a result of an additional broadening process taking place usually in the solid state such as for instance exciton excitation or local changes of the screening energy. Thus, one can conclude that both experimental results agree with each other, and the real proton partial width of the threshold 0^+ resonance (without broadening) can be smaller than 1 meV.

Theoretical arguments. Observation of a new resonance in the ^{4}He nucleus is rather surprising in view of the conviction that its level scheme has been well known for many years. Recent *ab initio* structure calculations of the four-nucleon system applying realistic nucleon-nucleon interactions and the microscopic cluster approach [32–34] confirm the level structure of ^{4}He earlier proposed by the multichannel R-matrix theory [35]. Theoretically, the DD stripping reactions have been successfully described within this theory taking into account a sequence of broad overlapping 1 + 3 cluster states. At higher excitation energy of about 28 MeV, series of pure 2 + 2 cluster states are known for which partial neutron and proton widths are very small.

Generally, the 1 + 3 cluster resonances in ^{4}He can be understood as predominantly single-particle states of different relative angular momenta, mixed by the internal isospin mixing with the states of the isospin equal to 1. A very good example for that is twin isospin mixed P-wave resonances which explain an anisotropic angular distribution of the DD reactions even at the lowest projectile energies. All of them have very small deuteron partial widths. Similarly, the negative parity 2 + 2 states 1^-, 2^-, and 0^- located around the excitation energy $E_x = 28$ MeV show very small nucleon widths, which was already postulated many years ago [36], suggesting their very weak coupling to the 1 + 3 cluster levels of the same spin and parity. Therefore, these states can be also interpreted as single-particle resonances of the 2 + 2 structure with the angular momentum of 1. Within this picture, the corresponding s-wave single-particle resonance 0^+ should be close to the DD reaction threshold, which was also recognized in the last four-nucleon calculations [32,34]. The authors have assumed, however, a strong mixing of this state with the 1 + 3 configuration, leading to formation of the first excited state at $E_x = 20.21$ MeV. This is, nonetheless, in contradiction to the finding of papers [37,38] pointing to a large single-particle 1 + 3 strength of the second 0^+ state of ^{4}He and denying earlier supposition of its breathing vibration nature [39]. In order to explain the very weak mixing, I propose here, in analogy to shape coexistence effects known in heavier nuclei, a four-nucleon energy surface with two different minima for two different clustering states, depending on the radius mean square (rms) parameter (see Fig. 2). For a larger rms value of

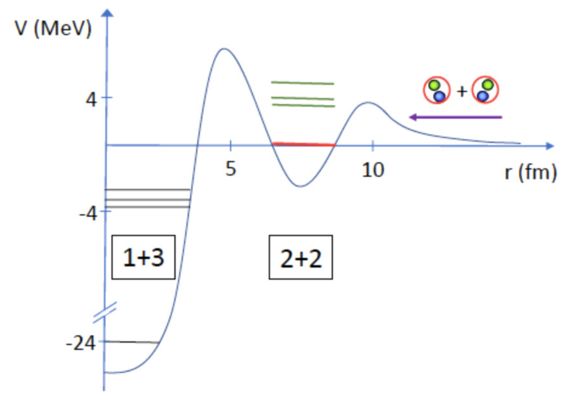

FIG. 2. Schematic four-nucleon energy surface separating 1 + 3 and 2 + 2 cluster states for the angular momentum $L = 1$. The red colored level represents the DD 0^+ threshold resonance.

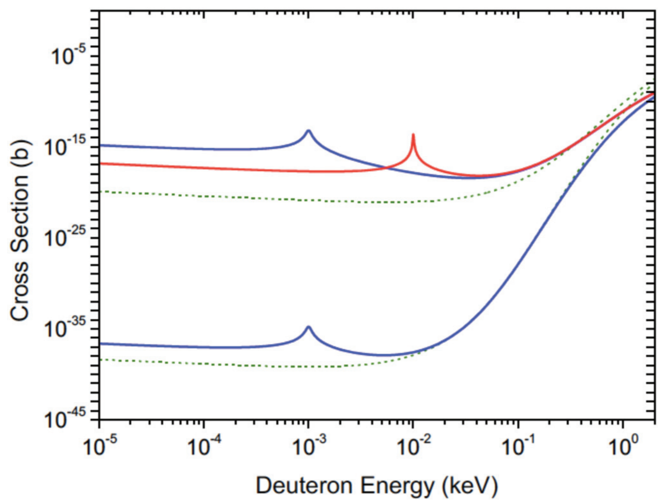

FIG. 3. Nonresonant (dotted lines) and resonant (solid lines) cross sections for $U_e = 400$ eV (upper part) and $U_e = 100$ eV (lower part). Blue lines correspond to the resonance energy of 1 eV and the red one to 10 eV. The resonance width is assumed to be 0.1 eV.

about 8 fm, the $d + d$ structure could be realized, whereas at values less than 5 fm (see Ref. [38]), the 1 + 3 structure takes place. Similarly large radius differences are already applied in the R-matrix parametrization of the four-nucleon system [35]. The potential barriers of the energy surface are composed of the Coulomb and centrifugal barriers. Therefore, the $l = 1$ states of the 2 + 2 structure, localized about 4 MeV above the DD threshold (Fig. 2), have to penetrate 4.1 MeV (centrifugal barrier) and 0.32 MeV (Coulomb barrier) to mix with the 3 + 1 configuration. For the $l = 0$ state, there is only the pure Coulomb barrier for which the transition probability is as low as 10^{-5}. An additional effect, contributing to the very low mixing, certainly results from a very low overlap between the different cluster wave functions.

Resonance decay width. If the 0^+ resonance is located only a few eV above the DD threshold, the deuteron partial width will be very small because of the strongly decreasing penetration factor [Eq. (2)]. Since nucleon partial widths are also very small and gamma emission for the $0^+ \to 0^+$ ground state transition is strictly forbidden, other electromagnetic decay channels as the electron conversion or internal pair creation can contribute to the total width. The electron conversion process is, however, much less probable for light nuclei and high excitation energies [40]. For a rough estimation of the total resonance width, one can therefore focus on a partial width related to the $E0$ transition of the internal pair production. The corresponding energy weighted sum rule (EWSR) can be written as [41]

$$\sum_{n\geqslant 2}^{\infty} (E_n - E_1)\, |\langle 0_n^+|\sum_p r_p^2|0_1^+\rangle|^2 = \frac{Z\hbar^2}{m_N}\langle r_p^2\rangle_{0_1^+}, \qquad (9)$$

where E_n and $|0_n^+\rangle$ are the energy and wave function of the nth state of the four-nucleon system, respectively; m_N is the nucleon mass, and $Z = 2$.

Four-nucleon calculations using a realistic NN force and phenomenological three-body force performed for both the ^{4}He ground and first excited 0^+ states ($E_x = 20.21$ MeV) gave the rms radii $\langle r_p^2\rangle^{1/2}$ equal to 1.66 and 5.3 fm, respectively [37], and the transition matrix element $\langle 0_2^+|\sum_p r_p^2|0_1^+\rangle =$ 1.38 fm^2, which exhausts only 17% of EWSR and provides the partial resonance width for pair production of 0.52 meV. Assuming the same value of the transition matrix element for the DD threshold resonance, the partial resonance width would be equal to 1.6 meV due to the strong excitation energy dependence of the internal pair creation [40]. On the other hand, taking into account the large rms radius of the proposed resonance of about 8 fm and assuming its maximum mixing with the ground state, the transition matrix element would take on its maximum possible value $\langle 0_3^+|\sum_p r_p^2|0_1^+\rangle = (\langle r_p^2\rangle_{0_3^+} - \langle r_p^2\rangle_{0_1^+})$ of 61.2 fm^2 and the partial width of 3.1 eV. For a more realistic estimation, one can suppose that the mixing with the ground state is the same as for the first excited state; then one gets 5.91 fm^2 and 29 meV, respectively. Therefore, the real total resonance width should be dominated by the internal pair creation, and its value lies somewhere between both limits determined above.

Extrapolation to room temperature. The ^{2}H $(d, p)^3$H reaction cross sections calculated for the threshold resonance and simply extrapolated using the known astrophysical S factor [30] which takes into account only the known broad ^{4}He resonances are presented in Fig. 3. The curves are evaluated for the screening energies of 100 and 400 eV, which correspond to the lower (theoretical) and upper (experimental) limit values. The reduction of the screening energy at extremely low deuteron energies to about 78% of its value observed in accelerator experiments has been taken into account according to the prescription presented previously [18,42]. Since the resonance position is not known exactly, the resonant cross section was also estimated for two different resonance energies. The total resonance width has been set to 100 meV, which roughly corresponds to the theoretical estimation for the internal pair production and includes the thermal broadening effects discussed above. Strong dependence of the cross sections on the screening energy value can be observed at very low energies. The threshold resonance cross section overestimates the values calculated for the known broad resonances by many orders of magnitude. Thus, any interference effects do not need to be included at thermal energies. The difference between both reaction amplitude contributions is much bigger for the larger screening energy. Since the partial resonance width of the electron-positron pair production should be two orders of magnitude bigger than that for the proton channel, the DD reaction at thermal energies will predominantly lead to the ^{4}He production.

Therefore, to determine the power density of a hypothetical energy source based on the ^{4}He production at room temperature, the nuclear reaction rate has to be estimated for deuterons implanted in a host metal:

$$R = N_A \langle \sigma_{\rm scr} v \rangle \cong N_A \sigma_{\rm scr}(E) \sqrt{\frac{2E}{\mu}} = N_A \sqrt{\frac{2}{\mu U_0}}\, S_0 \exp\left(-\sqrt{\frac{E_G}{U_0}}\right). \quad (10)$$

Here, the screened cross section has been assumed to be constant over the relative velocity v distribution range, and $N_{\rm A}$ is the Avogadro number. The screen energy U_0 is equal to 78% of the value U_e determined in the accelerator experiments, and S_0 represents the S factor at room temperature. Assuming conservatively that $\sigma_{\rm scr} = 10^{-15}$ b at $E = 25$ meV for the ^{4}He channel, one gets the reaction rate of 2.9×10^{-9} 1/s, which would lead to the power of 0.59 W per gram of Pd with the stoichiometric metal-to-deuteron ratio equal to unity. This power is estimated under the assumption that all deuterons can move freely in the Pd crystal lattice, which of course is not satisfied due to limited hydrogen diffusion in the metallic lattice. However, it is large enough to use it for commercial applications.

Conclusions. In conclusion, it has been shown that the deuteron fusion reactions at room temperature can take place with relatively large cross sections because of large electron screening energies realized in metallic deuterides. The reaction probability might be additionally increased by many orders of magnitude if the DD threshold resonance is taken into account. As accelerator experiments show, this 0^+ resonance and its single-particle nature are necessary to explain the enhanced reaction probability of the ^{2}H $(d, p)^3$H reaction for decreasing deuteron energies in experiments with both metallic and gaseous targets. The existence of the proposed resonance is also supported by theoretical arguments based on the weak coupling between different cluster structures and large nuclear radius differences. The latter implies a large partial resonance width for the internal pair creation resulting in the strongly increased branching ratio for synthesis of the ^{4}He nucleus. Instead of the strong suppression of the ^{4}He channel observed in accelerator experiments, its domination by a factor of 100 or more over the proton channel can be expected at room temperature. Finally, the estimated reaction rates suggest commercial applicability of the DD fusion reactions in metallic environments as a new effective nuclear energy source with a strongly reduced emission of neutrons—the largest part of the reaction Q value will lead to production of charge particles that can be absorbed in the active material. The effectivity of the new energy source will critically depend on the value of the screening energy of the applied material. As shown before [27], an increase of the effective electron mass arising from the crystal lattice defects can be here especially helpful. Because of the small resonance width, the electron screening and its local material dependence also decide about a resonance position and its width, being crucial for the experimental reaction rates. These effects explain why the cold fusion experiments are so difficult to reproduce. Here, new experimental studies focused on the internal pair production could be very helpful. The proposed threshold resonance might play a similar important role for future energy production utilizing the DD fusion reactions, as another single-particle 0^+ resonance, so-called Hoyle resonance [43], postulated in the past to explain helium burning and synthesis of ^{12}C in massive stars.

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