Anomalous enhancement of *DD* reaction in Pd and Au/Pd/PdO heterostructure targets under low-energy deuteron bombardment

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Yields of protons emitted in the $D+D$ reaction in Pd, Au/Pd/PdO, Ti, and Au foils are measured by a $dE-E$ counter telescope for bombarding energies between 2.5 and 10 keV. The experimental yields are compared with those predicted from a parametrization of the cross section and stopping power at higher energies. It is found that for Ti and Au target the enhancement of the $D(d,p)T$ reaction is similar to that observed with a deuterium gas target (several tens of eV). The dependence of the yields on the bombarding energy corresponds well to the screening potential parameters $U_s=250\pm15$ eV for Pd and 601 ± 23 eV for Au/Pd/PdO. Possible models of the enhancement obtained are discussed. © *1998 American Institute of Physics.*
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Nuclear reactions at very low energies are naturally assumed to be affected by the environment, since the surrounding electrons contribute to the effective Coulomb interaction between the projectile and target nuclei. Indeed, recently reported experiments have revealed non-negligible effects caused by bound or free electrons in low-energy reactions with solid or gas targets.[1,2]

The $D+D$ reaction in gas targets has been investigated by many authors.[3–7] Krauss *et al.*[5] parametrized the S factor for $D(d,p)$ and $D(d,n)$ reactions with a quadratic polynomial for $5<E_{c.m.}<120$ keV. Bosch and Hale parametrized the reaction cross section using the R-matrix parameters of the *DD* reaction which were determined from all types of experimental data, including integrated cross section, differential cross section, and polarization.[6] Greife *et al.*[7] recently reported measurements in a deuterium gas target at center-of-mass energies down to 1.6 keV. The deduced astrophysical S factors below 10 keV are clearly larger than predicted in Ref. 6. They interpreted the observed enhancement as the screening effect of the bound electrons and obtained a screening potential

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$U_s = 25 \pm 5$ eV. The $D+D$ reaction in Ti at room temperature was studied in Ref. 8 down to 3 keV (lab) and more recently to as low as 2.5 keV (lab).[9] At very low deuteron energies $E_d < 4.0$ keV there was enhancement of the *DD* reaction in Ti, with a screening potential $U_s = 20 \pm 12$ eV. This value doesn't exceed the screening in deuterium gas[7] and indicates the absence of specific conditions in the Ti crystal lattice that could increase the electronic screening of deuterons.

However, for solids that interact with deuterium one might expect stronger crystal-lattice effects than were obtained in Refs. 7–9. Thus it was suggested in Ref. 10 that hydrogen nuclei in some metals are strongly screened, since the electrons both in the metallic *d* band and in the hydrogen-induced *s* band can contribute to the screening effect. Moreover, the diffusivity of deuterium and its mobility in Pd metal are very high, and they are accompanied by a quantum interaction between the deuterons and conduction electrons[11] that could create additional conditions for screening as compared to other metals with high deuterium concentration but low diffusivity.

Therefore, in this work we have carried out low-temperature studies of possible screening effects in Pd metal and Au/Pd/PdO heterostructure samples with high deuterium diffusivity and compared the results with the electronic screening in Ti and Au. It should be emphasized that our experiments used a deuteron beam with a minimum energy of 2.5 keV (lab). The use of such a low deuteron beam energy in a *DD* reaction study has not been reported previously.

The low-energy, high-current ion beam generator, operating within a 1–100 keV energy interval (using a duoplasmatron ion source with a low energy spread $\leqslant 25$ eV) and producing a several hundred μA deuteron beam, is described elsewhere.[9] The targets were Pd, Ti, and Au foils 100 μm thick that had been annealed in high vacuum at 800 °C for 5–10 h before the experiment. The Au/Pd/PdO foil, 60 μm thick (with a PdO layer of 40 nm and an Au coating of about 0.1 μm on the opposite side), was prepared in accordance with Ref. 12.

To eliminate electrical noise at low bombarding energies during the proton yield measurements, we used a $\Delta E - E$ counter telescope consisting of Si surface barrier detectors 50 μm and 150 μm thick. The telescope was placed 1.5 cm from the target and at 90° with respect to the beam direction. The target was tilted by 58°, and the solid angle of detection was about 5.0% of 4π sr.

The target bombardment was carried out at deuteron energies of 2.5–10 keV using a beam current that was varied within the range 240–60 μA in such a way that for each beam energy E_d the power applied to the sample would be the same. During the experimental procedure the beam energy was changed continuously: before, during, and after runs at different deuteron energies $E_d < 10.0$ keV. Proton yield measurements at $E_d = 10.0$ keV were made in order to monitor the deuteron concentration in the sample. It should be noted that for long times of continuous bombardment (typically about 10 days) at different energies the mean concentration corresponding to a given effective average deuteron stopping range in the sample could be considered nearly the same for any energy used. The cause of this effect at low deuteron energies is the overlapping of the ranges for 2.5–10 keV deuterons in all the materials used if allowance is made for their range stragglings, which are comparable to the mean ranges in the energy interval under consideration.[13] For our samples of Pd and Au/Pd/PdO implanted with 5.0–10.0 keV

deuterons this hypothesis was confirmed by deuterium depth profile measurements made by the elastic recoil technique,[14] which showed very broad, uniform deuterium distributions extending 0.0–100 nm for Pd and more than 250 nm in the Au/Pd/PdO case, while the effective ranges in Pd for energies in the interval 5.0–10 keV amount to only 20–40 nm. We can therefore consider the mean deuterium concentration in the sample under bombardment to be quite constant and independent of the projectile energy.

The thick-target yield Y_t of the $D(d,p)T$ reaction at bombarding energy E_d is:[9]

$$Y_t(E_d)=\epsilon\int N_D(x)\sigma_{lab}(E)dx=\epsilon\int N_D(x)\sigma_{lab}(E)(dE/dx)^{-1}dE, \tag{1}$$

where $x=x_d$, $N_D(x)$, $\sigma_{lab}(E)$, and dE/dx are the mean range of an incident deuteron, the density of target deuterons, the reaction cross section, and the stopping power, respectively, and ϵ is the proton detection efficiency in our case. The parametrization of Bosch and Halle[6] has been used to describe the cross section with an extrapolation to lower energies that gives good agreement with the gas target experiment.[7] The stopping power of deuterons in the target used is assumed to be proportional to the projectile velocity[13] at low deuteron energies. This assumption was recently confirmed experimentally down to deuteron (proton) energies as low as 1.0 keV.[15,16]

The thick-target yield calculated according to Eq. (1) for $E_d<10.0$ keV is normalized to the experimental yield at $E_d=10.0$ keV (where enhancement is negligible[7,9]) and is plotted by the solid line in Fig. 1. This plot reflects the behavior of the bare $D+D$ reaction cross section in vacuum as the deuteron energy is decreased. In view of the procedure of normalization by the value at 10 keV we did not use the actual values of N_D in our calculation of the bare cross section, assuming them to be equal for all deuteron energies. Thus the thick-target yields obtained for the bombarded samples should be compared with the reaction cross section of the bare $D+D$ reaction in order to see whether or not the reaction rate for them is larger than in vacuum.

At the same time, by using the experimental proton yield at $E_d=10$ keV (with negligible enhancement), it is possible to estimate the mean concentration of deuterium in the subsurface layer of the sample:

$$N_D=Y_t(10)/Q\times\epsilon\times I(E), \tag{2}$$

where Q is the deuteron charge transferred through the sample during 10.0 keV bombardment and $I(E)$ is the integral from 0 to 10 keV: $I(E)=\int\sigma(E)(dE/dx)^{-1}dE$.

The screening potential U_s was calculated from the enhancement data

$$f(E)=Y\exp(E)/Y_t(E_b)=\exp[\pi\eta(E)U_s/E],$$

where $Y\exp(E)$ is the experimental yield at deuteron energy E and $Y_t(E_b)$ is the bare yield at the same energy, and $2\pi\eta=31.29Z^2(\mu/E)^{1/2}$ is the Sommerfeld parameter (Z is the charge number of the deuteron in the case of D^+ projectile and target, μ is the reduced mass, and E is the center-of-mass energy).

The thick-target yields of Ti and Au foils are shown in Fig. 1. The measured yields in these metals fall almost on the standard bare yield, although slight enhancements were deduced from these data (Table I). The screening potential values derived for Ti and Au with the systematic errors taken into account do not exceed the value of U_s that was

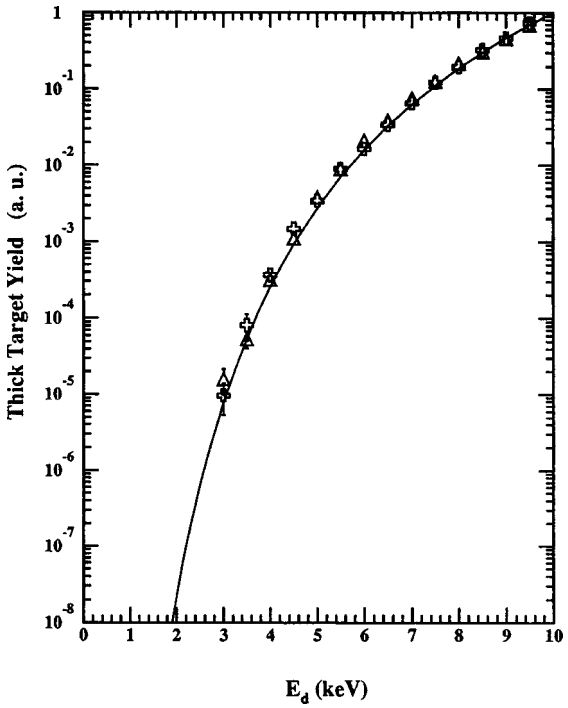

FIG. 1. Experimental yields of the $D(d,p)T$ reaction in Ti (crosses) and Au (triangles) targets under cooled conditions versus the bombarding energy. The solid curve is the bare yield calculated according to formula (1) without any enhancement.

obtained in Ref. 7 for a D_2 gas target. The U_s value for Ti (Table I) in low-temperature bombardment is also in close agreement with the screening potential value $U_s = 20 \pm 12$ eV that was deduced earlier in our experiments at room temperature.[9] The screening potential values for Ti and Au therefore indicate an absence of the strong electron screening that might be expected to occur in a metal crystal lattice loaded with a very high deuterium concentration.

In contrast to Ti and Au, the samples of Pd (during bombardment both under cooled and room temperature conditions) and Au/Pd/PdO exhibit a strong, perfectly reproducible

TABLE I. Mean temperature $\langle T \rangle$ under D^+ bombardment, average deuterium concentration corresponding to the ratio $\langle x \rangle = D/\mathrm{Me}$ determined from formula (2) at $E_d = 10$ keV, and screening potential U_s for the samples used

Sample	$\langle T \rangle$, K	$\langle x \rangle = D/\mathrm{Me}$	U_s, eV
Ti	185.5	3.76 ± 0.50	35.5 ± 10.5
Au	179.2	1.66 ± 0.32	22.8 ± 11.0
Pd (cool)	190.1	0.23 ± 0.10	250.1 ± 16.0
Pd (room)	313.0	0.38 ± 0.12	257.5 ± 21.6
Au/Pd/PdO	193.3	0.11 ± 0.07	601.7 ± 23.4

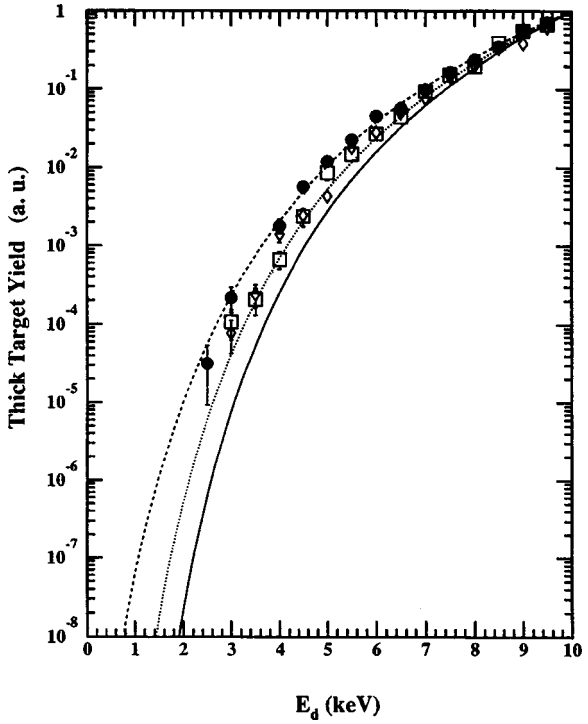

FIG. 2. Experimental yields of the $D(d,p)T$ reaction: in Pd under cooled conditions, $\langle T\rangle = 190.1$ K (open squares); in Pd at $\langle T\rangle = 313.0$ K (open diamonds); and in an Au/Pd/PdO heterostructure at $\langle T\rangle = 193.3$ K (solid circles). The solid curve is the calculated bare yield without enhancement. The dotted and dashed curves are parametrizations of the experimental yields with screening potentials $U_s = 250$ eV and $U_s = 600$ eV, respectively.

deviation to the positive side of the standard bare curve (Fig. 2) that increases as the deuteron energy decreases, so that at $E_d = 2.5$ keV for the Au/Pd/PdO heterostructure the experimental yield is about 50 times larger than the standard bare value. In Fig. 3 the ratio of the experimental yield to the standard yield (i.e., the enhancement) is plotted as function of bombarding energy. The enhancement values obtained are well parametrized by the screening potential values $U_s = 601 \pm 23$ eV for the Au/Pd/PdO heterostructure (Fig. 3, dashed line) and $U_s = 250 \pm 16$ eV for the Pd sample (Fig. 3, dotted line). The screening potentials deduced from the experimental yield for both the Pd and Au/Pd/PdO samples are surprisingly large and cannot be explained by electron screening in the metals, even with allowance for the possibility of sd hybridization of electrons in Pd deuteride.[10] At the same time, the concentration of deuterium in both the Pd and heterostructure samples is rather low (Table I), and their loading ratios (D/Pd) are too far from the maximum ratio ($x \sim 0.95$) that could be achieved in electrochemical loading.

This peculiarity of the yield in different materials under bombardment is obviously connected with deuterium diffusivity in metals. The diffusivity of D in Pd and in the Au/Pd/PdO heterostructure is much higher than in Ti and even more so than in Au.[11] The presence of high diffusivity may create conditions of deuterium ''fluidity'' in the subsurface layer of the crystal lattice of Au/Pd/PdO and Pd. In this case conditions may also be

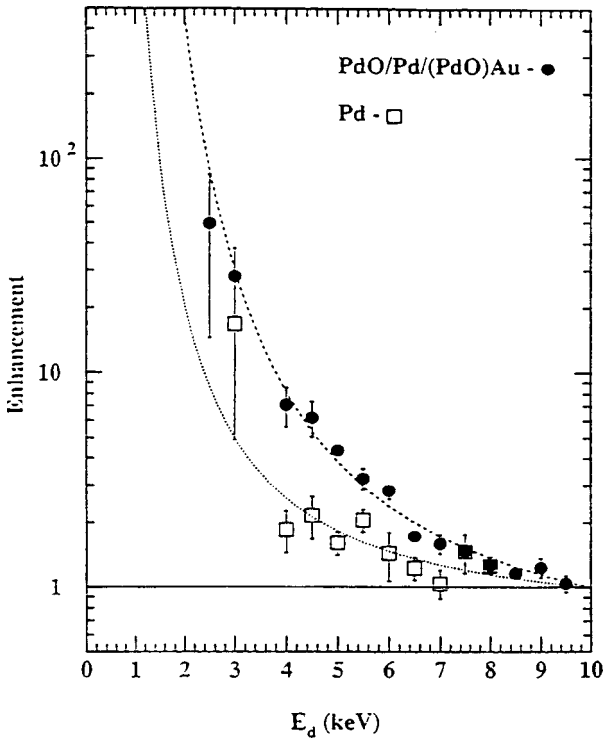

FIG. 3. Observed enhancement of thick-target yields of $D(d,p)T$ reactions in Pd (squares) and Au/Pd/PdO (circles) versus the bombarding energy under cooled conditions. The solid curve is the calculated bare curve without any enhancement. The dotted and dashed curves are for screening potential values $U_s = 250$ eV and 600 eV, respectively.

conducive to dynamic deuteron–deuteron screening[17–19] due to coherent motion of the deuterons. The total short-range screening radius of a deuteron in a Pd matrix for the case of both electronic and dynamic ion screening is given by[19]

$$r_s = k^{-1} = (k_e^2 + k_+^2)^{-1/2}, \tag{3}$$

where k_e^{-1} is the electronic screening radius of a deuteron in the metal and k_+^{-1} is the dynamic screening length due to coherent deuteron motion:

$$k_e^2 = 6\pi e^2 n_e / E_F = 3.94 n_e^{1/3} / a_0$$

(here n_e is the electron concentration in the metal, E_F is the Fermi energy, and a_0 is the Bohr radius), while within the linearized Poisson–Boltzmann equation dynamic ion model the radius r_+ will be represented in Debye-like form:

$$r_+ = (k_B T / 4\pi e^2 N_D)^{1/2} \text{Å} = 6.91 (T/N_D)^{1/2},$$

where T is the temperature and N_D is the mobile deuteron concentration in the sample.

The values of the screening potentials in Ti and Au are in good agreement with only the electronic screening part of (3), so that their radii $k_e^{-1} \sim 0.45$–0.50 Å. In this case the screening potential value must be 25–30 eV, which corresponds within error limits to our

experimental values (Table I) as well as to the deuterium gas target result.[7] On account of the low diffusion rate in these metals the concentration of mobile deuterium in them will be rather low and cannot contribute to the deuteron–deuteron screening.

For the case of Pd and the Au/Pd/PdO heterostructure, however, the dynamic ion screening term of Eq. (3) must be used, on account of their high U_s values. In the case under consideration the total concentration of diffusing deuterons is uncertain and cannot be obtained from Eq. (2), since the target yield provides information mainly on the concentration of trapped species. Nevertheless, direct estimates for the case of Pd bombarded under cooled conditions, using the actual temperature ($\langle T\rangle = 190.1$ K) and the concentration that is derived from Eq. (2) ($\langle N_D\rangle = 1.56\times 10^{22} D/\mathrm{cm}^3$), gives in accordance with Eq. (3) a screening radius $r_s = 0.075$ Å and a screening potential $U_s = 193$ eV. In the case of the Pd sample at room temperature (Table I) the values were $r_s = 0.07$ Å and $U_s = 196$ eV. The values screening potential obtained for the two Pd cases at different temperatures and D loadings without taking into consideration the total deuterium concentration are, as expected, 25% smaller than the experimental values, which, however, are also close to each another (Table I).

In the case of the Au/Pd/PdO heterostructure target the deuterium diffusion rate is much higher than for Pd, so that direct estimation of the screening potential as a function of deuterium concentration on the basis of only the yield measurements would not be correct. To permit estimation of the deuterium mobility in the samples, additional *in situ* experiments are now in progress.

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