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Strongly Enhanced Li + D Reaction in Pd Observed in Deuteron Bombardment on PdLi$_x$ with Energies between 30 and 75 keV

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Thick target yields of α particles emitted in the ^{6,7}Li(d,α)^{4,5}He reactions in PdLi$_x$ and AuLi$_x$ were measured as a function of the bombarding energy between 30 and 75 keV. It was found that the reaction rate in Pd at lower energies is enhanced strongly over the one predicted by the cross section for the reaction with bare nuclei, but no enhancement is observed in Au. A screening energy is introduced to reproduce the excitation function of the thick target yield for each metal. The deduced value for Pd amounts to 1500 ± 310 eV, whereas it is only 60 ± 150 eV for Au. The enhancement in the Pd case cannot be explained by electron screening alone but suggests the existence of an additional and important mechanism of screening in metal.

KEYWORDS: low-energy nuclear reaction, Li + D reaction in metal, reaction rate enhancement, screening energy
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1. Introduction

At energies far below the Coulomb barrier, cross sections of nuclear reactions are essentially dominated by the Coulomb penetration factor.[1,2)] However, significant cross section enhancements over the predictions with simple penetration have been reported.[3–7)] These enhancements are qualitatively understood as a screening effect due to bound electrons, since the prediction is based on the assumption that Coulomb potential of the target nucleus and projectile is resulting from bare nuclei. In a naive picture, the screening effect may be described quantitatively by a screening energy U_s, and the cross section at the center-of-mass energy E_{cm} is described as

$$\sigma(E_{cm}) = S(E_{cm} + U_s)/(E_{cm} + U_s)\exp\{-2\pi\eta(E_{cm} + U_s)\}, \tag{1}$$

where S(E) is the astrophysical S-factor, and $\eta(E) = Z_1 Z_2 \alpha(\mu c^2/2E)^{1/2}$ is the Sommerfeld parameter; Z_1 and Z_2 are atomic numbers of the target and projectile, α is the fine structure constant and μ is the reduced mass. The greater the value of U_s, the greater the enhancement and vice versa.

Recently, the screening energy of the D + D reaction in various materials has been measured by several authors.[8–14)] Surprisingly, some metals provide anomalously large size screening effects for the D + D reaction, while others exhibit normal electron screening enhancement. The enhancement of the reaction rate strongly depends on the host material, and deduced values of the screening energy vary from several tens of eV to 800 eV. Although Riola *et al.*[13)] have discussed several possibilities to interpret such large screening in materials, no satisfactory explanation could be given. Yuki *et al.*[10)] and Kasagi *et al.*[14)] have proposed that high fluidity of deuterons in the host may be responsible for the enhancement.

In order to explore the mechanism of enhanced screening, we have studied other nuclear reactions in metal hosts. In the present work, we have investigated the ^{6,7}Li(d,α)^{4,5}He reactions in metal, for the first time. Two host metals were selected in which the Li + d reactions occur, Pd and Au. The screening energy of the D + D reaction in Pd is confirmed to be very large, although two reported values are not in good agreement with each other; $U_s = 250$–310 eV in refs. 10 and 14 and ~800 eV in ref. 13. On the other hand, the normal value of screening obtained for the D + D reaction in Au is $U_s = 20$–70 eV in refs. 10 and 14 and ~60 eV in ref. 13. Thus, a naive and natural question is whether the Li + D reaction in Pd is also strongly enhanced compared to Au. The screening energies of the ^{6}Li(d,α)^{4}He reaction obtained with LiF target and the one of the D(^{6}Li,α)^{4}He reaction with gas target were reported to be 380 ± 250 and 330 ± 120 eV, respectively,[15)] slightly larger than expected from a simple prediction of the atomic model.

2. Experimental Procedure

The experiments were performed using a low-energy ion beam generator[8)] at the Laboratory of Nuclear Science of Tohoku University, designed to produce deuteron beams with several 100 μA from 2 to 100 keV. It consists of a duoplasmatron ion source, an extraction lens, a bending magnet, focusing lenses, and an acceleration/deceleration electrode. The duoplasmatron ion source delivers high beam currents (~1 mA) with low energy spread (< 25 eV). In the acceleration mode, the beam, extracted from the ion source with an extraction voltage of 25 keV, first passes through the bending magnet and focusing lenses and is then accelerated up to 100 keV. This beam is transported straight to the target position after passing through two apertures determining the beam position and size (~4 mm diameter).

The target used was a foil of Pd–Li alloy, which was prepared by arc melting Pd and Li as described in ref. 16. The atomic ratio Li/Pd obtained was 5–7% in a foil of $13 \times 13 \times 0.3$ mm^3. A foil of Au–Li alloy was obtained from Tanaka Metal Co. Its atomic ratio Li/Au was ~10% and the size of the foil was $20 \times 20 \times 1$ mm^3. In addition, a 2-mm thick LiF foil was bombarded to deduce the excitation function of the ^{7}Li(d,α)^{5}He reaction as reference of non-metallic targets, since no measurement was reported so far in this low energy region. During the bombardment, the target was kept at low temperature between −80 and −70°C to minimize the possible thermal diffusion of Li contained in the target alloy from the beam spot.

In order to detect α particles emitted in the ^{6,7}Li(d,α)^{4,5}He reactions, a ΔE-E counter telescope consisting of 30- and 100-µm thick Si surface barrier detectors was used. The front face of the ΔE detector was covered with a 2-µm thick Al foil to prevent electrons and scattered deuterons from hitting the detector. The counter telescope was placed at 125° to the beam direction and subtended a solid angle of 0.14 sr. Signals from the detectors were fed to preamplifiers which generated fast outputs for time information as well as slow outputs for pulse height information. The fast outputs were fed into timing filter amplifiers and timing signals were generated by constant fraction discriminators. A signal of coincidence between the ΔE and E detectors was used as an event gate for CAMAC data taking system; CAMAC ADCs and TDCs were used to measure pulse height spectra for each detector and a time spectrum between the two detectors in event-by-event mode.

Figure 1(a) shows a scatter plot of ΔE vs E measured during the bombardment on AuLi$_x$. Alpha particles are identified clearly as the events on a locus between the dashed lines; events A correspond to those from the ^{7}Li(d,α)^{5}He reaction and events B are from the ^{6}Li(d,α)^{4}He reaction. Events with $\Delta E < 400$ ch were assigned as originating from the D + D reaction, in which the incident deuterons interact with the ones implanted by the beam in the target. Tritons and ^{3}He particles emitted in the D + D reactions cannot punch through the ΔE detector. Although the threshold for the ΔE detector is nominally set above the energy loss of the proton, a small portion of the huge proton peak is inevitably observed in Fig. 1(a) as indicated by C, above which pile-up events in the ΔE detector are observed as a vertical line. Events indicated by D correspond to two-proton pile-up events. In Fig. 1(b), the projected energy spectrum of α-particles is shown, in which two peaks are clearly seen; one for the ^{6}Li(d,α)^{4}He reaction and the other for ^{7}Li(d,α)^{5}He. The lower energy peak of the ^{7}Li(d,α)^{5}He reaction does not possess a symmetric shape, because of the asymmetric nature of the ground state ^{5}He coupled to a contribution of the direct three-body channel, $\alpha + \alpha + n$. Since the low energy side of the spectrum is cut off by the ΔE detector and the contribution of the three-body channel cannot be estimated correctly, it is impossible to measure the exact yield of α particles corresponding to the ground state of ^{5}He. Instead, we simply fixed the low energy side of the gate by using the same channel number of the E spectra for all the measurements, indicated by a vertical line in Fig. 1(a).

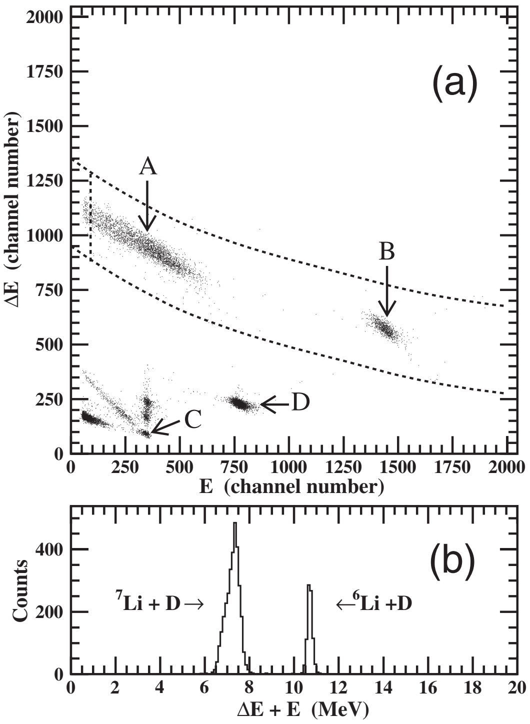

Fig. 1. Measured spectra for the deuteron bombardment on AuLi$_x$; (a) two-dimensional scatter plot of ΔE vs E with α-particle events indicated between the dashed lines and (b) energy spectrum of α particles emitted in the ^{6,7}Li(d,α)^{4,5}He reaction in Au.

The present measurements only give relative thick target yields of the ^{6,7}Li(d,α) reactions in metal. The total dose of the incident deuteron beam for each run was deduced from the electric current from the target with a small correction for secondary electron emission. For the correction factor, separate measurements were made to compare the electric current in the target (I_T) with the one measured directly by an electron-suppressed Faraday cup (I_F) placed behind the target. The measured ratio ($R = I_F/I_T$) of the direct current to the target one for $30 < E_d < 75$ keV is always smaller than 1.0 exhibiting a small dependence on the bombarding energy. Assuming a linear dependence on the bombarding energy, we have obtained the ratio as a function of the bombarding energy E_d, $R = 0.874 - 0.0008E_d$ for Pd and Au. For the LiF target, a metal mesh-screen was placed on the surface to make good contact and to prevent the target from being charged up by the beam. The correction factor $R = 0.531 - 0.0003E_d$ was obtained for LiF.

The target Li in the metal host was present in form of an alloy, i.e., PdLi$_x$ or AuLi$_x$ with $x = 0.05$–0.10, and the number of Li atoms was found to decrease during the measurements. Thus, in the present work, we employed a similar method to obtain the relative yields as was done in the measurements of the D + D reaction;[8–10,14] the α particle yield at 75 keV was repeatedly measured at frequent intervals to average out small fluctuations, and the yield at energy E_d was divided by the averaged yield at 75 keV measured just before and after each measurement at E_d. Figure 2 shows α-particle yields (sum yields of both channels, ^{6}Li + d and ^{7}Li + d) measured at $E_d = 75$ keV for the PdLi$_x$ and AuLi$_x$ target as a function of the accumulated dose of deuteron beam. As can be seen, the yield decreases initially and becomes stable later on. For the LiF target, the yield at $E_d = 75$ keV was also measured frequently, although this yield remained more or less constant.

3. Data Analysis and Results

In order to discuss the effect of the surroundings on the reaction rate, the reaction cross section with bare nuclei is indispensable for a standard. The S-factor of the ^{6}Li(d,α)^{4}He reaction, deduced by Engstler *et al.*[15] from the data at higher energies where the screening effect is negligibly small, can be used as a standard cross section unaffected by the surroundings. However, no measurement has been reported

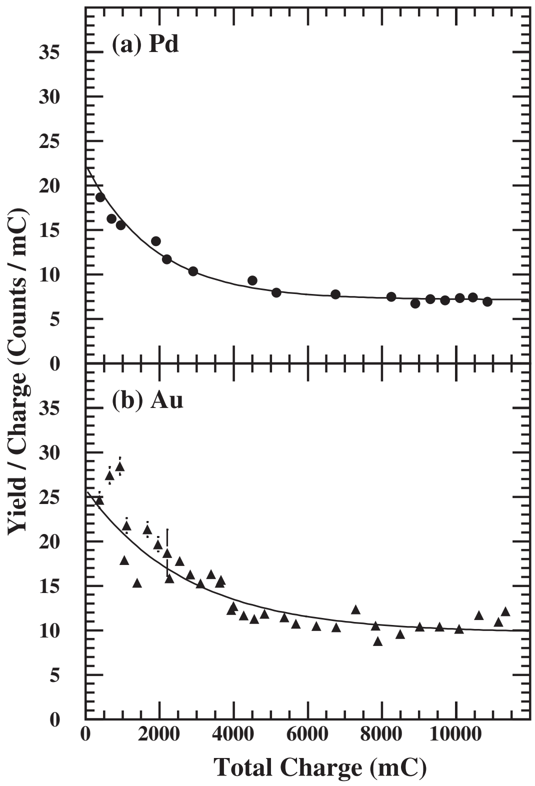

Fig. 2. Summed yields of α particles emitted in the ^{6}Li + d and ^{7}Li + dreaction, measured at $E_d = 75$ keV for (a) PdLi$_x$ and (b) AuLi$_x$ as a function of the dose of the deuteron beam.

for the ^{7}Li(d,α)^{5}He reaction. Thus, in the present work, the thick target yields of both the ^{6}Li(d,α)^{4}He and the ^{7}Li(d,α)^{5}He reactions were measured using LiF target. The purpose of this measurement was not to deduce the S-factor of the ^{7}Li(d,α)^{5}He reaction, but to obtain the ratio of the α-particle yield in the gate employed in the ^{7}Li(d,α)^{5}He reaction to the yield in the ^{6}Li(d,α)^{4}He reaction. Figure 3 shows results of the such measurement, i.e., the relative thick target yield for the LiF target as a function of the bombarding energy; (a) for the ^{6}Li(d,α)^{4}He reaction and (b) for the ^{7}Li(d,α)^{5}He reaction.

First, we analyze the excitation function of the ^{6}Li(d,α)^{4}He reaction. Since incident deuterons slow down in the target and the reaction can occur until the deuterons stop, the observed α-particle yield $Y(E_d)$ at the bombarding energy E_d is given by

$$Y(E_d) = (\text{constant}) \times N_{\text{Li}} \int_0^{E_d} d\sigma_{6\text{Li}}(E_{\text{cm}})/d\Omega_{\text{cm}} \times (d\Omega_{\text{cm}}/d\Omega_{\text{lab}})(dE/dx)^{-1} dE. \quad (2)$$

Here, N_{Li}, is the number of target Li, $d\Omega_{\text{cm}}/d\Omega_{\text{lab}}$ is the ratio of the solid angle in the center-of-mass to laboratory system, and dE/dx is the energy dependent stopping power for deuterons in LiF. The differential cross section, $d\sigma_{6\text{Li}}(E_{\text{cm}})/d\Omega_{\text{cm}}$, can be expressed with Legendre polynomials up to order 2, $P_2(\theta_{\text{cm}})$ as reported in ref. 5; i.e.,

$$d\sigma_{6\text{Li}}(E_{\text{cm}})/d\Omega_{\text{cm}} = \sigma(E_{\text{cm}}) \cdot (4\pi)^{-1} \cdot (1 + A_2(E_{\text{cm}})P_2(\theta_{\text{cm}})).$$

Since the detector is placed at 125° with respect to the beam direction and the value of A_2 is less than 0.1 at these energies,[5] the second term contributes little at most 0.001, and is therefore neglected in the present analysis. For the calculation of the relative yield $Y(E_d)/Y(75\,\text{keV})$, N_{Li} cancels out. The parameterization by Anderson and Ziegler[17] is employed for the stopping power of deuterons and the S-factor in ref. 15 is used for the cross section of the ^{6}Li(d,α)^{4}He reaction. The result of this calculation is given by the solid line in Fig. 3(a). It is seen that the calculation with the standard parameter set reproduces the experimental data reasonably well. This is at variance with the result of ref. 15 requiring a screening energy of about 300 eV. However, this discrepancy is not investigated thoroughly here, and may be caused by electric charge up of the LiF target due to imperfect contact of the mesh.

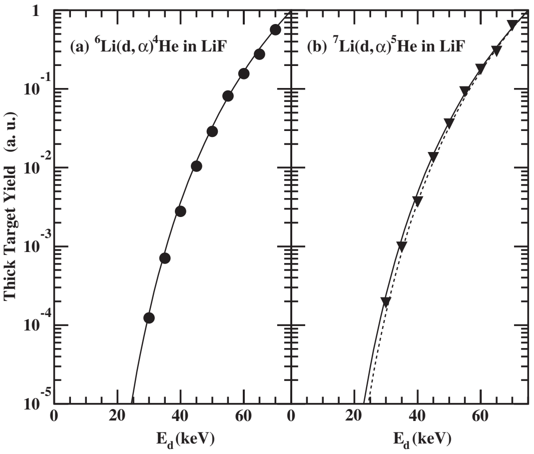

Fig. 3. Relative yield of α particles emitted in the ^{6,7}Li(d,α)^{4,5}He reaction in LiF as a function of the bombarding energy of deuterons.

The relative yield for the ^{7}Li(d,α)^{5}He reaction is then calculated in the same way using the cross section of the ^{6}Li + d reaction and is compared with the data. In this case, however, the calculation indicated by the dashed line in Fig. 3(b) deviates increasingly from the experimental data as the bombarding energy decreases. Two reasons are considered as follows. The one is due to kinematics: In the ^{7}Li(d,α)^{5}He reaction, the low energy gates of yield integration for α particles detected at 125° were used at identical energy in the laboratory system. This causes an increase in the integration at the relative energy of the residual ^{4}He–n system or the excitation energy of ^{5}He (this increase amounts to 40 keV for every 10 keV decrease in the bombarding energy). The other is the possibility that the S-factor differs from the one for the ^{6}Li(d,α)^{4}He reaction. In the present work, the yield of the ^{5}He ground state cannot be separated from the contribution of the three-body $\alpha + \alpha + n$ channel because of the missing low energy part in the α-particle spectrum.

We have measured the yield of the ^{7}Li(d,α)^{5}He reaction with a common gate setting for all the target. Thus, an effective excitation function corresponding to the present gate is needed in order to analyze the ^{7}Li and ^{6}Li data for the PdLi$_x$ and AuLi$_x$ targets. We have introduced the energy dependent yield function, $G_7(E) = \sigma_{6\text{Li}}(E) \times (1.576 - 0.00712E)$, and have re-calculated the thick target yield for the ^{7}Li(d,α)^{5}He reaction by replacing $\sigma_{6\text{Li}}(E)$ with $G_7(E)$ in eq. (2). The result of the calculation is shown in Fig. 3(b) by

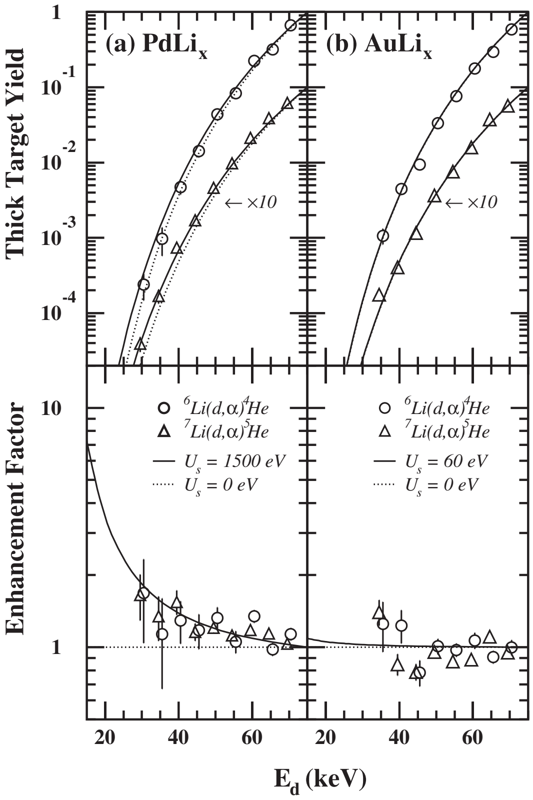

Fig. 4. Relative yield of α particles emitted in the ^{6,7}Li(d,α)^{4,5}He reaction as a function of the bombarding energy of deuterons; (a) for PdLi$_x$ and (b) for AuLi$_x$. In the upper part, the data normalized to the yield at 75 keV are plotted. In the lower part, the experimental yields divided by those presented with the dotted curve are shown. The dotted curves correspond to the relative yields calculated without screening. Solid curves correspond to calculations with the screening energy indicated in each section.

the solid line, which reproduces the data very well. Thus, we have obtained the standard excitation function, corresponding to the ^{7}Li(d,α)^{5}He reaction without the effect of the surroundings to be $G_7(E)$.

The results for the PdLi$_x$ and AuLi$_x$ targets are shown in Fig. 4; Fig. 4(a) for PdLi$_x$ and Fig. 4(b) for AuLi$_x$. The upper part of Fig. 4 shows the excitation functions of the ^{6,7}Li(d,α)^{4,5}He reactions relative to the yield at $E_\mathrm{d} = 75$ keV. The standard calculations are carried out with eq. (2), by using the stopping power parameterized in ref. 17 for Pd and Au together with $\sigma_{6\mathrm{Li}}(E)$ and $G_7(E)$. The results are plotted with dotted lines in Fig. 4 (the dotted line in Fig. 4(b) is covered by the solid line). Relative to the dotted line, the yield of the Li + d reaction in PdLi$_\mathrm{x}$ is larger at the lower energies.

In the lower part of Fig. 4, we plot the ratio of the experimental yields to the standard calculations in order to make the comparison easier. As seen, the reaction rate in Pd is systematically enhanced for both reactions. On the other hand, the deduced enhancement is negligibly small in Au and scatters around 1.0. In order to explain the observed enhancement, thick target yields have been calculated using the enhanced cross section with the screening energy U_s as described in eq. (1).

We deduced the values of U_s of the ^{6}Li(d,α)^{4}He and the ^{7}Li(d,α)^{5}He reaction, separately, for each data by fitting the experimental relative yields. The deduced values are $U_\mathrm{s} = 1400 \pm 480$ and 1580 ± 380 eV, respectively for the ^{6}Li(d,α)^{4}He and the ^{7}Li(d,α)^{5}He reaction in Pd, and $U_\mathrm{s} = 310 \pm 280$ and -180 ± 180 eV, respectively for the ^{6}Li(d,α)^{4}He and the ^{7}Li(d,α)^{5}He reaction in Au. The screening energy of both reactions in the common host metal should be equal, since the screening effect is considered to originate in electrical effects which cannot be different for ^{6}Li and ^{7}Li. Moreover, the deduced values of U_s almost overlap each other within statistical errors. Thus, we have considered that the difference of U_s for the ^{6}Li(d,α)^{4}He and the ^{7}Li(d,α)^{5}He reactions in each metal is due to fluctuations of experimental data including systematic errors. The final values of U_s in the present work are, then, obtained by averaging the above values. The results are $U_\mathrm{s} = 1500 \pm 310$ and 60 ± 150 eV for the Li + d reactions in Pd and Au, respectively. The calculations with screening energy U_s are shown by the solid lines in the upper and lower parts of Fig. 4. The errors shown are statistical errors only. The systematic errors are considered to originate mainly from the uncertainty in the bombarding energy ($\pm$25 eV) and from fluctuations in the Li density ($\pm$100 eV) in the target.

4. Discussion

The screening energy of the ^{6}Li + d and ^{6,7}Li + p reactions has been obtained in ref. 15 to be 420 ± 120 eV for the LiF target and 350 ± 80 eV for the hydrogen and deuterium gas target. Although the screening effect in this case is considered to originate from the bound electrons, the reported values are somewhat larger than the prediction of the naive atomic model ($\sim$170 eV), i.e., the difference of the binding energy of electrons between Be and Li, and this small enhancement is not fully understood, yet. The present work shows, for the first time, that, in the metallic environment, the size of the screening effect in the Li + d reaction depends strongly on the metal host. The obtained value of the screening energy in Pd is about 4 times larger than those mentioned above. Therefore, we conclude that another screening mechanism exists in Pd apart from the one due to bound electrons in the naive atomic picture.

In metals, the screening effect due to conduction electrons should also be considered. The screened electrostatic potential of the nucleus with atomic number Z existing in the sea of conduction electrons is given[18] as $\phi_\mathrm{s}(r) = Ze/r \cdot \exp(-k_\mathrm{e}r)$; $k_\mathrm{e} = (6\pi e^2 n_\mathrm{e}/E_\mathrm{F})^{1/2}$, n_e is the number density of electrons and E_F is the Fermi energy of the electrons. Thus, the beam deuteron experiences a reduced Coulomb repulsion force, when it collides with the nucleus, and the corresponding screening energy is approximated as $U_\mathrm{ce} = Ze^2k_\mathrm{e}$. For the Li + d reaction in Pd metal, $E_\mathrm{F} = 2.66$ eV and $n_\mathrm{e} = 1.97 \times 10^{22}\,\mathrm{cm}^{-3}$,[19] thus $U_\mathrm{ce} = 61$ eV is expected. The effect of the bound electron should be added, since the Li atom is considered to remain in metal in the form of Li$^+$. Summing up the values of the screening energy due to conduction electrons and bound electrons, we obtain a value of about 230 eV. Even if the experimental value in ref. 15 is used for the bound electrons, the summed value is 410–480 eV. Therefore, the large screening energy of $\sim$1500 eV obtained for the Li + D reaction in Pd cannot be due to electron screening alone.

Of particular interest is the fact that the Pd metal provides a large screening effect not only for the Li + d reaction but also for the D + D reaction ($U_s = 250$–$310\,\mathrm{eV}$[10,14] and $800\,\mathrm{eV}$[13]), whereas the Au metal host does not in both cases. Thus the mechanism of enhanced screening in metal might have the same origin in the D + D and Li + d reactions. Although the enhanced screening is not fully understood, we have previously discussed the possibility that the large screening effect might originate from fluid deuterons in Pd.[10,14] If the same argument is applied to both reactions, the electrostatic potential of the nucleus with atomic number Z is also screened by mobile D^+ ions and by conduction electrons. In this case, the screened potential due to D^+ is given as $\phi_s(r) = Ze/r \cdot \exp(-k_d r)$, where $k_d = (4\pi e^2 n_d / k_B T)^{1/2}$ and n_d is the deuteron density. When we use the experimental values $n_d = 3 \times 10^{21}/\mathrm{cm}^3$[10,14] and $T = 200\,\mathrm{K}$, $k_d = 56.5\,\mathrm{nm}^{-1}$ is deduced. This corresponds to the screening energy of $\sim$240 eV for the Li + d reaction, which is similar in size to the electron screening but is still not sufficient to explain the observed screening energy. With the given values of n_d and T, however, the screening energy for the D + D reaction in Pd is only $\sim$80 eV, which does not explain the observed values either.

The above discussion indicates that the screening energy of the Li + d reaction should be 3 times larger than the one of the D + D reaction as long as the screened Coulomb potential is expressed in the form $\phi_s(r) = Ze/r \cdot \exp(-kr)$, whatever the origin of the screening might be. Unfortunately, the two reported values for the D + D reaction do not agree with one another (which might be an indication that the enhancement phenomenon is not well controlled experimentally) and we cannot examine the scaling in detail. Thus, at present, we can only deduce that the enhanced screening observed in Pd depends on the atomic number Z of the implanted target nucleus; the value for implanted Li is 1.9–4.8 times larger than the one for implanted D, or a scaling form of $Z^{0.58-1.43}$.

5. Conclusions

In summary, we have studied the low-energy Li + d reactions in metal, and observed the strong effect of the environment. In the present work, thick target yields of α particles emitted in the Li + d fusion reaction in PdLi_x, AuLi_x and LiF were measured as a function of the bombarding energy for $30 < E_d < 75\,\mathrm{keV}$. It was found that the reaction rate at low energies normalized to the one at 75 keV, increases strongly in Pd but not in Au. We have introduced the screening energy (U_s), which reduces the Coulomb barrier between the nuclei, to parameterize the observed enhancement for each host metal. The values of the deduced screening energy are 1500 ± 310 for Pd and $60 \pm 150\,\mathrm{eV}$ for Au. Since the screening energy caused by electrons in metal is expected up to about 480 eV, the large screening energies deduced for Pd indicates the existence of a new mechanism enhancing the reaction rate. Of particular interest is the fact that the Pd metal provides a large screening energy not only for the Li + d reaction but also for the D + D reaction, whereas the Au metal host does not in both reactions. However, a quantitative comparison of the screening energies in the two reactions is difficult at present. Additional experimental efforts are highly desirable to reduce the systematic errors and to obtain more data for various host metals as well as for different reactions. In order to understand the mechanism of the enhanced screening, new theoretical treatments of the nuclear reaction including the surrounding environment are also highly desirable.

The present work reveals a non negligible effect of the environment surrounding the nuclei on the cross section. Thus, low-energy nuclear reactions at energies far below the Coulomb barrier should be explored under various conditions, experimentally as well as theoretically.

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