Li+D Reaction in Pd and Au for 30<E_d<75 keV(I. Nuclear Physics)

Li+D Reaction in Pd and Au for $30 < E_d < 75$ keV

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Thick target yields of α particles emitted in the ^{6,7}Li $(d, \alpha)^{4,5}$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.

Recently, the screening energy of the D+D reaction in various materials has been measured by several authors[1-7]. 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.* [6] have discussed several possibilities to interpret such large screening in materials, no satisfactory explanation could be given. Yuki *et al.* [3] and Kasagi *et al.* [7] 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 $(d, \alpha)^{4,5}$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; $U_s = 250\sim310$ eV in Refs. [10, 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 $U_s = 20\sim70$ eV in Refs. [10, 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 experiments were performed using a low-energy ion beam generator [1] at the Laboratory of Nuclear Science of Tohoku University, designed to produce deuteron beams with several 100 μA from 2 to 100 keV. The target used was a foil of Pd-Li alloy, which was prepared by arc melting Pd and Li as described in Ref. [8]. The atomic ratio Li/Pd obtained was $5\sim7\%$ in a foil of $13\times13\times0.3$ 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 20×20×1 mm³. In addition, a 2-mm thick LiF foil was bombarded to deduce the excitation function of the ⁷Li (d, α)⁵He 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 ℃ 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 ⁶,⁷Li (d, α)⁴,⁵He reactions, a ΔE-E counter telescope consisting of 30- and 100-μ m thick Si surface barrier detectors was used. The front face of the ΔE detector was covered with a 2 μ m 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.

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 ⁷Li(d, α)⁵He reaction and events B are from the ⁶Li(d, α)⁴He reaction. Events with ΔE < 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. In Fig.1(b), the projected energy spectrum of α-particles is shown, in which two peaks are clearly seen; one for the ⁶Li(d, α)⁴He reaction and the other for ⁷Li(d, α)⁵He. Although the lower energy peak of the ⁷Li(d, α)⁵He reaction does not possess a symmetric shape, we simply fixed the low energy side of the gate by using the same channel number of

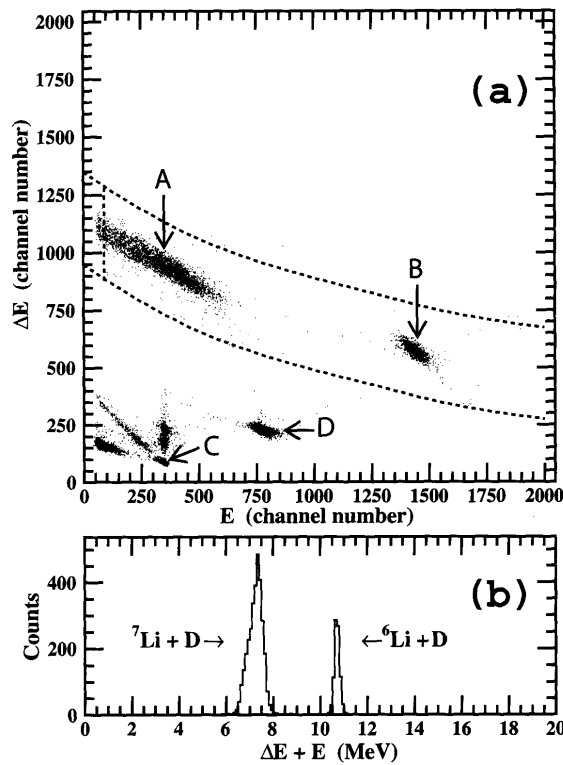Fig.1. Measured spectra for the deuteron bombardment on AuLi$_x$; (a) two-dimensional scatter plot of ΔE vs E with α-particle events indicated between the dashed lines and (b) energy spectrum of α particles emitted in the ⁶,⁷Li(d, α)⁴,⁵He reaction in Au.

the E spectra for all the measurements, indicated by a vertical line in Fig.1(a).

The target Li in the metal host was present in form of an alloy, i.e., $PdLi_x$ or $AuLi_x$ with $x = 0.05 \sim$ 0.10, and the number of Li atoms was found to decrease during the measurements. Thus, in the present work, we employed a method to obtain the relative yields; 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, $^6Li + d$ and $^7Li + d$) measured at $E_d = 75$ keV for the $PdLi_x$
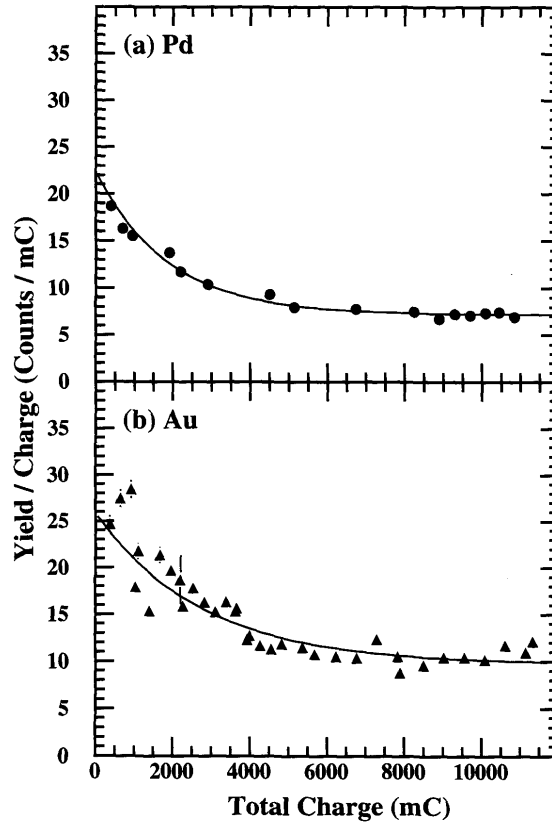

Fig.2. Summed yields of α particles emitted in the $^6Li + d$ and $^7Li + d$ reaction, measured at $E_d = 75$ keV for (a) $PdLi_x$ and (b) $AuLi_x$ as a function of the dose of the deuteron beam.

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.

No measurement has been reported for the $^7Li(d, \alpha)^5He$ reaction, so far. Thus, in the present work, the thick target yields of both the $^6Li(d, \alpha)^4He$ and the $^7Li(d, \alpha)^5He$ reactions were measured using LiF target. The purpose of this measurement was not to deduce the S-factor of the $^7Li(d, \alpha)^5He$ reaction, but to obtain the ratio of the α-particle yield in the gate employed in the $^7Li(d, \alpha)^5He$ reaction to the yield in the $^6Li(d, \alpha)^4He$ 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 $^6Li(d, \alpha)^4He$ reaction and (b) for the $^7Li(d, \alpha)^5He$ reaction.

First, we analyze the excitation function of the $^6Li(d, \alpha)^4He$ reaction. Since incident deuterons slow down in the target and the reaction can occur until the deuterons stop, the observed α-particle yield Y
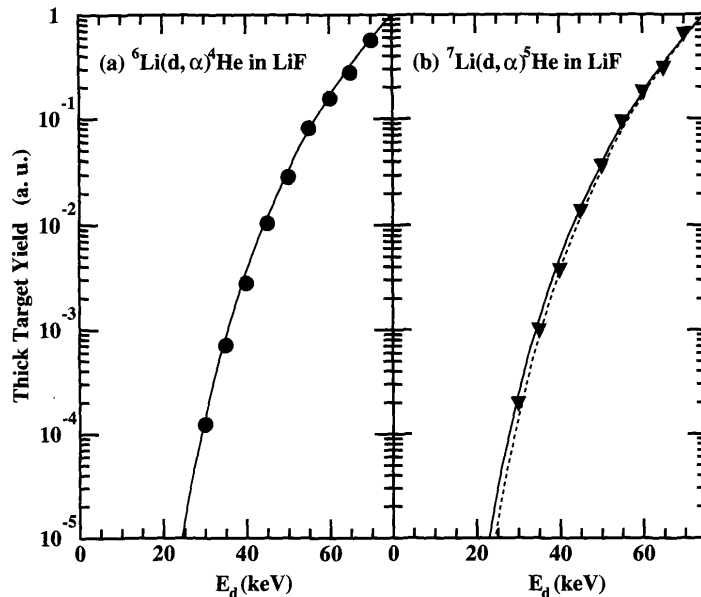
Fig.3. Relative yield of α particles emitted in the ^{6,7}Li(d, α)^{4,5}He reaction in LiF as a function of the bombarding energy of deuterons.

(E_d) at the bombarding energy E_d is given by

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

Here, N_{Li} is the number of target Li, $d\Omega_{cm}/d\Omega_{lab}$ 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. Since the detector is placed at $125°$ with respect to the beam direction, $d\sigma_{^6Li} (E_{cm})/d\Omega_{cm}$ can be replaced as $\sigma(E_{cm})/4\pi$. For the calculation of the relative yield $Y(E_d)$ / $Y(75$ keV), N_{Li} cancels out. The parameterization by Anderson and Ziegler [9] is employed for the stopping power of deuterons and the S-factor in Ref. [10] 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.

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. 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 was deduced as the energy dependent yield function, $G_7(E) = \sigma_{^6Li}(E)\times(1.576-0.00712E)$, and have re-calculated the thick target yield for the ^{7}Li(d, α)^{5}He reaction by replacing $\sigma_{^6Li}(E)$ with $G_7(E)$ in Eq.(1). The result of the calculation is shown in Fig.3(b) by 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 $E_d = 75$ keV. The standard calculations are carried out with Eq.(2), by using the stopping

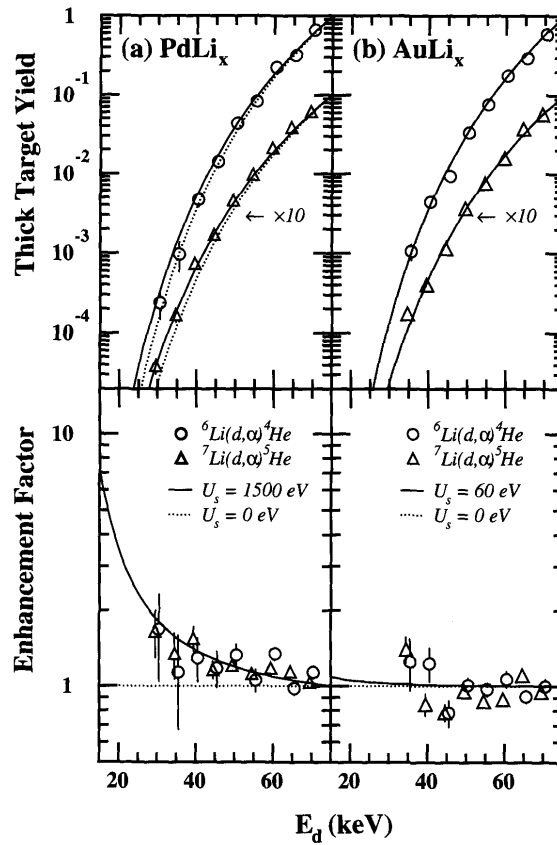Fig.4. Relative yield of α particles emitted in the ^{6,7}Li$(d, \alpha)^{4,5}$He reaction as a function of the bombarding energy of deuterons; (a) for PdLi$_x$ and (b) for AuLi$_x$. In the upper part, the data normalized to the yield at 75 keV are plotted. In the lower part, the experimental yields 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.

power parameterized in Ref. [9] for Pd and Au together with $\sigma_{^6\text{Li}}(E)$ 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$_x$ 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 Ref. [1]. The value of U_s for the ^{6}Li$(d, \alpha)^4$He and the ^{7}Li$(d, \alpha)^5$He reactions is searched for by fitting the experimental relative yields in Fig.4. The calculations with screening energy U_s are shown by the solid lines in the upper and lower parts of Fig.4. The results obtained are $U_s = 1500 \pm 310$ and 60 ± 150 eV for the Li+d reactions in Pd and Au, respectively. The errors shown are statistical errors only. The systematic errors are considered to originate mainly from the uncertainty in the bombarding energy (± 25 eV) and from fluctuations in the Li density (± 100 eV) in the target.

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 reported in Ref. [10] for the gas target and LiF target. 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 [11] as $\phi_s(r) = Ze/r \cdot \exp(-k_e r)$; $k_e = (6\pi e^2 n_e/E_F)^{1/2}$, n_e is the number density of electrons and E_F is the Fermi energy of the electrons. The corresponding screening energy is approximated as $U_{ce} = Ze^2 k_e$. For the Li+d reaction in Pd metal, $E_F = 2.66$ eV and $n_e = 1.97 \times 10^{22}$ cm^{-3} [12], thus $U_{ce} = 61$ 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. [10] is used for the bound electrons, the summed value is 410~480 eV. Therefore, the large screening energy of ~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 ($U_s = 250~310$ eV [3,7] and 800 eV [6]), 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 [3,7]. 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 $n_d = 3 \times 10^{21}$/cm^3 [3,7] and $T = 200°$ K, $k_d = 56.5$ nm^{-1} is deduced. This corresponds to the screening energy of ~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. 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 $Z^{0.58~1.43}$.

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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