Low-Energy Nuclear Reactions in Metals

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In order to investigate the interplay between nuclei and their surroundings, we have studied deuteron induced fusion reactions in metals at very low energies. We summarize the results on the following measurements; reaction rates of the D(d,p)T reaction in various metals for bombarding energies between 2.5 and 10 keV and those of the ^{6,7}Li(d,α)^{4,5}He reactions in Pd and Au for bombarding energies between 30 and 75 keV. These measurements clearly showed that the low energy nuclear reactions are strongly affected by the metal environments surrounding the nuclei.

§1. Introduction

When two nuclei collide at energy far below the Coulomb barrier, the reaction rate is extraordinarily low, since the projectile should penetrate the huge potential barrier. At such low energies, the effect of the environment surrounding the nuclei may appear clearly. For instance, the screening effect is mainly due to bound electrons, which cancel the Coulomb potential of the bare nuclei and thereby increase the reaction cross section.

One might expect a more abundant manifestation of the effect of the surrounding matter when the nucleus is embedded in a metal lattice, where the host atoms are arranged periodically and there are numerous free conduction electrons.[1)] In order to explore such circumstances for the DD fusion reaction, we have developed an experimental technique in which deuterons are implanted in a metal and then serve as target nuclei for subsequent bombardment with deuterons.[2)–5)] By using this technique, the screening energy in metal was first deduced for the DD reaction in Ti and Yb.[3)] The results were comparable to those for DD reaction using a gas target. In subsequent measurements with Pd, PdO, however, much larger screening energy than expected with conduction electrons were deduced,[4)] for the first time. The measurements have been extended for various hosts.[5)]

The large screening energy for the DD reaction in metal was also reported, later, by Czerski et al.[6)] and Raiola et al.[7)] for Ta as the metal host. Recently, Raiola et al.[8)] have obtained the screening energies of the DD reaction for a wide range of materials, and showed that most of the metals enhance the DD reaction rate very much.

We have also studied other effects of the environments acting on the nuclear reactions; the Li + d reactions in metals[9)] and the branching ratio of the DD reaction in metals. In this paper, we summarize the results on the measurements of reaction rates of D + D and Li + D reactions in metals.

§2. Experimental procedure

The measurements were performed using the low-energy ion beam generator at the Laboratory of Nuclear Science of Tohoku University. The experimental procedure is almost same as described in Refs. 2)–5). A deuteron beam extracted from the low-energy ion beam generator, which consists of a duoplasmatron ion source, an extraction lens, a bending magnet, focusing lenses, a deceleration electrode and a neutral beam filter magnet, was used to bombard the target. The low-energy beam of several 100 μA intensity was collimated by passing it through two apertures so as to fix the beam position and spot size at each energy; the beam spot on the target was about 3 mm in diameter.

We have performed the following measurements in various materials; (1) yield of protons from the D+D reaction as a function of bombarding energy between 2.5 and 10 keV to deduce the reaction rates, and (2) yield of α particles from the Li + D reactions for $30 < E_d < 75$ keV.

For the measurements (1), protons emitted in the D(d,p)T reactions were detected with a ΔE-E counter telescope consisting of 50- and 200-μm thick Si surface barrier detectors. During the measurement at the specific bombarding energy, the yield at 10 keV was measured very frequently at intervals in order to verify the number of target deuterons. We always normalized the proton yield to that at 10 keV measured just before and after the run. Up to the present, Be, Ti, Fe, Cu, Ni, Pd, Re, Sm, Yb, Au and PdO were bombarded as a host material in which the DD fusion reaction occurs.

For the ^{6,7}Li(d,α)^{4,5}He reaction measurements, the targets were foils of Pd-Li (PdLi$_x$, $x \sim 0.07$) and AuLi$_x$ ($x \sim 0.10$). They were kept at low temperature, during the bombardment, between -80 and $-70°C$ to prevent the Li atom from escaping from the spot where the beam bombards. Alpha particles were detected by a ΔE-E counter telescope consisting of 20- and 100-μm thick Si detectors. Aluminum foil of 10 μm in thickness covered the front face of the ΔE detector in order to prevent secondary electrons and scattered deuterons from hitting the detector.

§3. Results and discussion

3.1. *DD fusions in metals*

In Fig. 1, we show the relative excitation functions of the D(d,p)T reaction in PdO, Pd, Fe, Au and Ti, to demonstrate that the reaction rates depend strongly on the host materials. As expected from the bare D+D reaction the yields decrease very rapidly as the bombarding energy decreases. We show the standard yield by dotted curve, which corresponds to the thick target yield calculated for the bare D + D reaction in materials by the following equation,

$$Y_P(E_d) = A \int_0^{E_D} N_D(x)\sigma(E)(dE/dx)^{-1}dE, \tag{3·1}$$

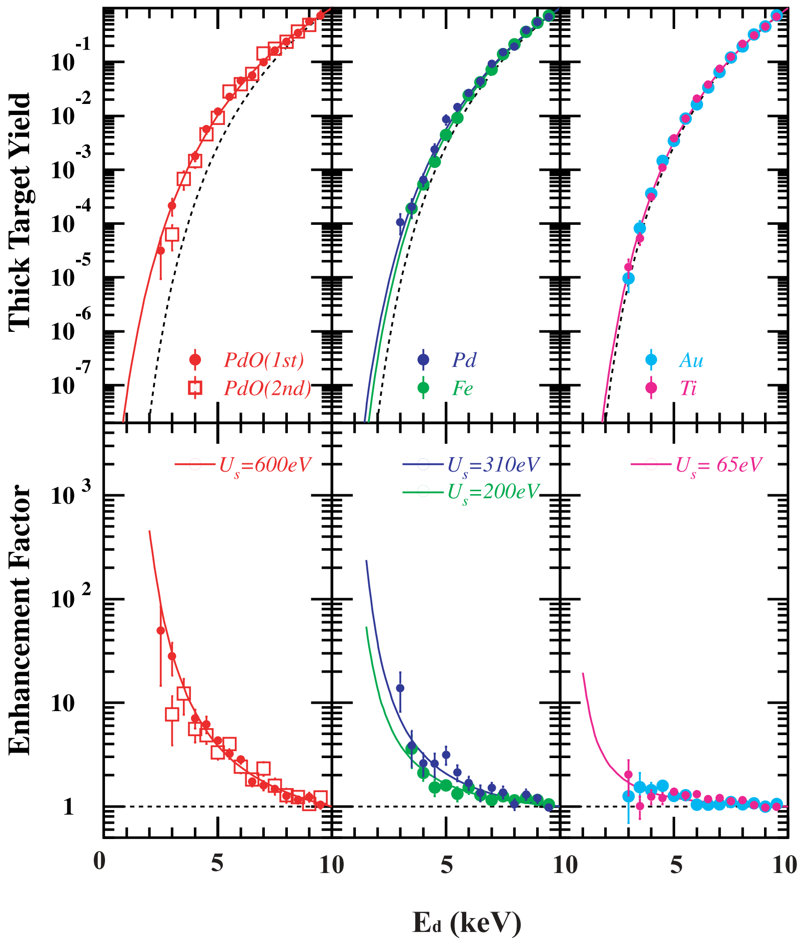

Fig. 1. Relative yield of protons emitted in the D(d,p)T reaction in the five hosts as a function of the bombarding energy; (a) two independent measurements for PdO, (b) for Pd and Fe, and (c) for Au and Ti. In the upper sections, the data normalized to the yield at 10 keV are plotted. In the lower sections, the experimental yields divided by the standard calculations are shown as enhancement factor.

where $\sigma(E)$ is the bare cross section parameterized by Bosch and Hale[10)] and (dE/dx) is the energy loss curve of deuteron parameterized by Anderson and Ziegler.[11)] It is clearly seen that the yields at lower energies depend on the host material very strongly. In the lower part of Fig. 1, we plot the ratio of the experimental yields to the standard calculation in order to make comparisons more clearly. As seen, the reaction rate in PdO is enhanced very much, about 50 times the standard at E_d = 2.5 keV. It is also enhanced in Pd and Fe. On the other hand, the deduced enhancement is very small for Au and Ti.

In a naive picture, the screening effect may be described quantitatively by a screening energy U_s and the enhanced cross section is described with the relative energy in center of mass system, as

$$\sigma(E) = \frac{S(E)}{E + U_s} \exp\left(-31.39 Z_1 Z_2 \sqrt{\frac{\mu}{E + S}}\right), \qquad (3\cdot2)$$

where $S(E)$ is the astrophysical S factor and μ is the reduced mass. The greater the value of U_s, the greater the enhancement and vice versa. The value of U_s is searched for so as to reproduce the data; those so far obtained in our measurements are listed in Table I.

Table I. Screening energy of the D+D reaction in metals obtained from our measurements.

Material	screening energy (eV)
PdO	600 ± 30
Pd	310 ± 30
Fe	200 ± 20
Re	200 ± 20
Cu	120 ± 20
Yb	80 ± 20
Ni	80 ± 20
Au	70 ± 20
Ti	65 ± 30

Conduction electrons in metal can be regarded as Fermi gas moving in a mean field produced by the nuclei and electrons. If a nucleus is placed in such an electron sea, the scalar potential produced by the nucleus is screened by the surrounding electrons. A simple estimation based on the Thomas-Fermi model gives $\phi_s(r) = Ze/r \exp(-k_e r)$, where k_e is given with the electron density n_e and Fermi energy E_F as $k_e = (6\pi e^2 n_e/E_F)^{-1/2}$. The potential can be approximated as $\phi_s(r) = Ze/r - Zek_e$ for $k_e r \ll 1$, and, thus the screening energy for deuterons in the conduction electrons is estimated as $U_s = e^2 k_e$.

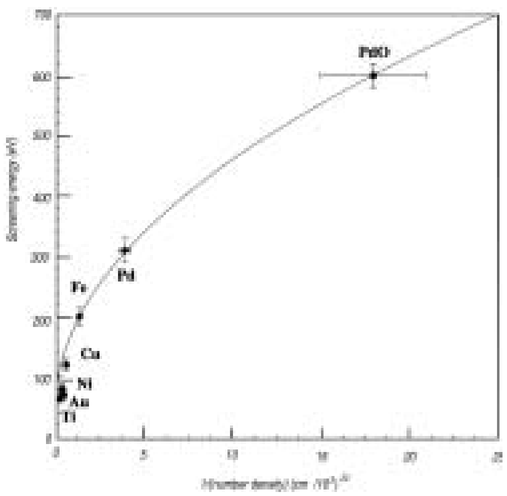

Fig. 2. Deduced screening energy as a function of inverse of the deuteron density.

The screening energy for a deuteron in the conduction electrons in Fe is estimated as $U_s = 24$ eV in this picture; the value is only 1/10 of the experimentally deduced one. For Pd, Ichimaru et al., including the effects of deuterons in the lattice, calculated the screening energy as $U_s = 75$ eV. The experimental value is about 4 times larger than the calculation. Thus, we conclude that the screening energy for the DD fusion reactions in Fe, Pd and PdO cannot be explained by the electron screening alone. It should be stressed that the value of 600 eV for PdO is extraordinary large.

A relevant correlation may be that between the screening energy and the deuteron density in the host during the bombardment, as shown in Fig. 2. We see there that a large screening energy goes with a small density (note that the abscissa is the inverse of the density). The density may be related to the diffusivity, or mobility of D^+ ions in the host; large mobility results in small density of the target deuterons and large screening energy. The fluid deuterons and conduction electrons might behave like a plasma in the host. In a plasma, both electrons and positive ions are fluid, and, hence, their electric charges can be distributed so as to satisfy simultaneously the Poisson equation and the statistical distribution. As a result, the Coulomb repulsion is reduced not only by electrons but also by positive ions. We suggest the possibility of such a dynamic screening mechanism during the deuteron bombardment and penetration into the host wherein the fluidity of deuterons must play a decisive role.

In Fig. 3, we plot, again, screening energy vs inverse of deuteron density, including the data of Rolfs group. The squares are our data and the circles are those in Ref. 8). Although the data are scattered largely, the correlation between the screening energy and the deuteron density may be seen. It should be noted that correlations are clearly seen for the data indicated by larger circles and squares connected by arrows, which are for the same host metal but different density.

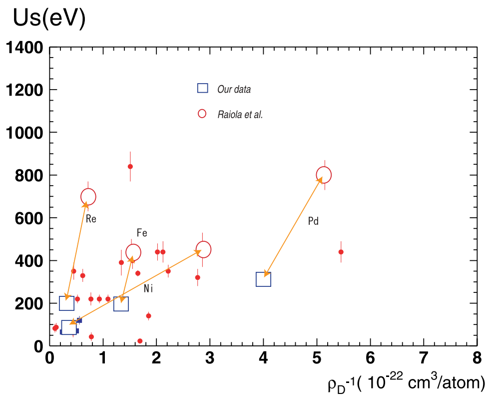

Fig. 3. Screening energy vs inverse of deuteron density. Our data are plotted with squares and data reported in Ref. 8) are plotted with circles. A pair of large circle and square connected with an arrow is for the same host metal.

3.2. *Screening energy for the Li+D reaction*

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 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.

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 results are $U_s = 1500 \pm 310$ 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 screening energy of the ^{6}Li + d and ^{6,7}Li + p reactions has been obtained in Ref. 12) 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 (∼170 eV), i.e., the difference of the binding

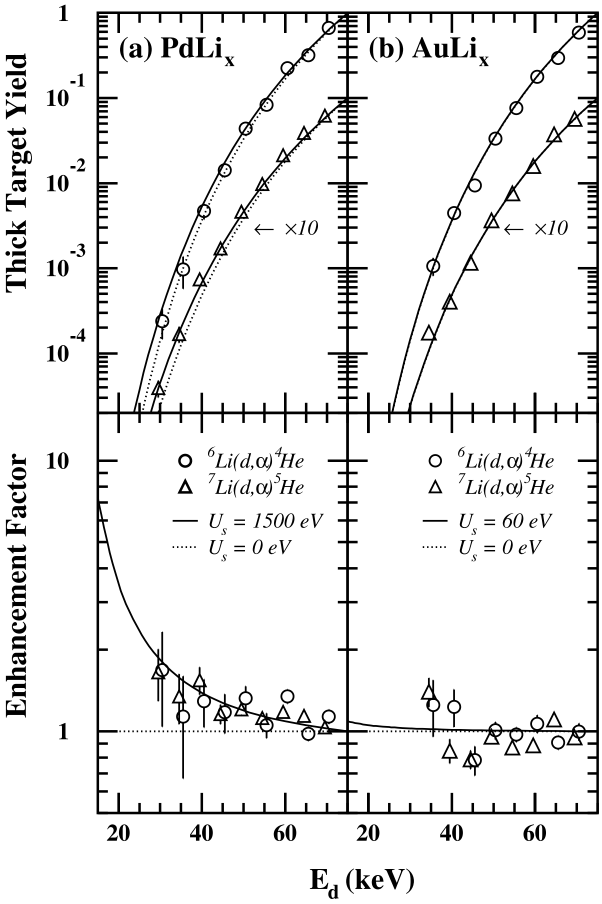

Fig. 4. Relative yield of α particles emitted in the ^{6,7}Li(d,α)^{4,5}He reaction as a function of the bombarding energy of deuterons; (a) for PdLi$_x$ and (b) for AuLi$_x$. In the upper part, the data normalized to the yield at 75 keV are plotted. In the lower part, the experimental yields divided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

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 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. Thus, the beam deuteron experiences a reduced Coulomb repulsion force, when it collides with the nucleus, and the corresponding screening energy is approximated as $U_{ce} = Z_e^2 k_e$. For the Li + d reaction in Pd metal, $E_F = 2.66$ eV and $n_e = 1.97 \times 10^{22}\text{cm}^{-3}$, 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. 12) is used for the bound electrons, the summed value is $410 \sim 480$ 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, 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. 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 $n_d = 3 \times 10^{21}/\text{cm}^3$ and $T = 200\text{K}$, $k_d = 56.5\text{nm}^{-1}$ 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. With the given values of n_d and T, however, the screening energy for the D + D reaction in Pd is only ~ 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 $1.9 \sim 4.8$ times larger than the one for implanted D, or a scaling form of $Z^{0.58\sim 1.43}$.

§4. Summary

We have measured yields of the D(d,p)T reactions in various metals for bombarding energies between 2.5 and 25 keV. The results of the measurements clearly showed that the D + D reaction is strongly affected by the environments surrounding the nuclei. Enhanced reaction rates were deduced for the DD fusions in PdO, Pd and Fe. These enhancements can be explained by introducing the screening energy as a parameter determined so as to reproduce the experimental data for each reaction and host. The large values of the screening energy are obtained for PdO, Pd, Fe and

Re. These large values cannot be explained by the effect of screening of electrons alone, since the conduction electrons can produce only several tens eV of screening energy for the D + D reaction.

Subsequently, we have also measured yields of the ^{6,7}Li(d,α)^{4,5}He reactions in Pd and Au for bombarding energies between 30 and 75 keV. Again, large enhancement was observed for the Li + D reactions in Pd. The screening energy deduced for the Li + D reaction in Pd amounts to 1500 ± 230 eV. This, again, is not due to the electron screening alone.

Therefore, we conclude that there should be another important mechanism for the fusion reactions to enhance the reaction rate in some particular metal like Pd. Based on the correlation between the screening energy and the deuteron density, we have suggested that the enhanced mechanism might be related with the fluidity of deuterons in metals.

The present work indicates clearly that the metal environment affects the fusion reactions of light nuclei at low energies. Thus, it is highly desirable to develop the theoretical studies in which that the low-energy nuclear reactions should be treated not as just isolated two-body system, but as the whole system including the environment.

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