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Low Energy D+D Reactions in Metal

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D+D reactions in various metals were investigated for the deuteron bombardment with bombarding energies at around 150 keV and below 15 keV. Energetic protons and α-particles which can never be attained in the D+D reaction were observed in bombardments with higher energy deuterons. In order to explain the spectra, reaction processes in which three deuterons are involved are considered; sequential reaction and simultaneous three-body reaction. The sequential reaction can well explain the observed bump structure, and the three-body reaction can reproduce the continuum spectral shape of protons and α-particles, although an anomalously large enhancement factor is required. For the lower energy bombardment, thick target yields for the D+D reactions in Ti were measured down to 4.7 keV. They were well explained with the astrophysical S-factors deduced from gas target measurements. This indicates that the effect of the environment is not so much different for the deuterons in Ti and in gas phase.

1. Introduction

The effect of the electron environment on the nuclear phenomena has been investigated for many years.[1] For the nuclear decay processes accompanying the change of bound electrons, such as electron captures and internal conversions, it is shown that any change in the configuration of the outer electronic shells modifies the electron density near the nucleus, and hence causes the change in the transition rate, although the observed change is very small. So-called cold fusion[2] has roused attention more generally on the influence of the environment where the nuclear processes take place. Although most of the experiments reported that time were known to be difficult to reproduce and hence to be under suspicion, the influence of the environment in various nuclear processes is one of the interesting subjects to be studied more, because of its interdisciplinary nature involving nuclear physics, condensed matter physics, material science, and so on. In addition, one can develop its applications in various fields if the environment really does affect the nuclear processes very strongly.

Nuclear reactions at very low energies are naturally considered to be affected by the environment, since surrounding electrons contribute to the effective Coulomb interaction between the projectile and target nuclei. Actually, recently reported experiments showed the innegligible effect caused by the bound electrons in low energy reactions with solid or gas targets.[3] One might expect much stronger effects than caused by bound electrons when the nucleus is embedded in different materials.[4] Ichimaru et al.[5] calculated the effective static potential for hydrogen in Ti and Pd, and showed the stronger screening effect together with the density effect. However, low energy reactions in such conditions have not been studied, so far.

We have started to perform two series of experiments on bombardments of low energy deuterons on various metals, in order to see how the low energy deuterons react with implanted deuterons in metal. One series employed relatively high energy deuterons (around 150 keV), and aimed to search for reactions other than the D+D reaction. The other aimed to measure the reaction rates of the D+D reaction in metal at very low energies. In this paper, we report on the results of these experiments obtained up to the present and describe our future plans.

2. 150-keV deuteron bombardment on various metals

The experiments with 150-keV deuterons were carried out by using a Cockcroft-Walton accelerator at the Department of Chemistry at Tohoku University. As the experiments with deuterated Ti were reported in ref. 6 and ref. 7 in detail, only results are shown here.

2-1 High energy charged particle spectrum

Very energetic particles were observed in the bombardment of 150 keV deuterons on TiD_x (x=1.2). In Fig. 1(a) is shown such a spectrum measured at 135° with a 15-μm thick Al absorber. A huge peak appearing at about 2.45 MeV is attributed to protons emitted in the D(d,p)T reaction. Events due to double and triple pileups of the protons are distributed up to about 4.9 and 7.4 MeV, respectively, where sharp edges are clearly seen in the spectrum. In addition to these normal events, events up to 17 MeV are also seen. They are neither pileups nor events produced by the protons in the detector as proved by Fig. 1(b) which shows the spectrum measured with a 200-μm thick Al foil stopping the 2.75-MeV protons. As seen, the huge proton peak and the pileups disappear, but the high-energy structure remains, and the amount of its reduced energy clearly indicates that these high energy particles are protons emitted in the target.

The characteristics of the high energy proton structure are these: a broad bump ranging from 12.5 to 16.5 MeV, a sharp peak at 14.1 MeV, and a continuum up to ~ 17 MeV where the bump and the peak are superimposed.

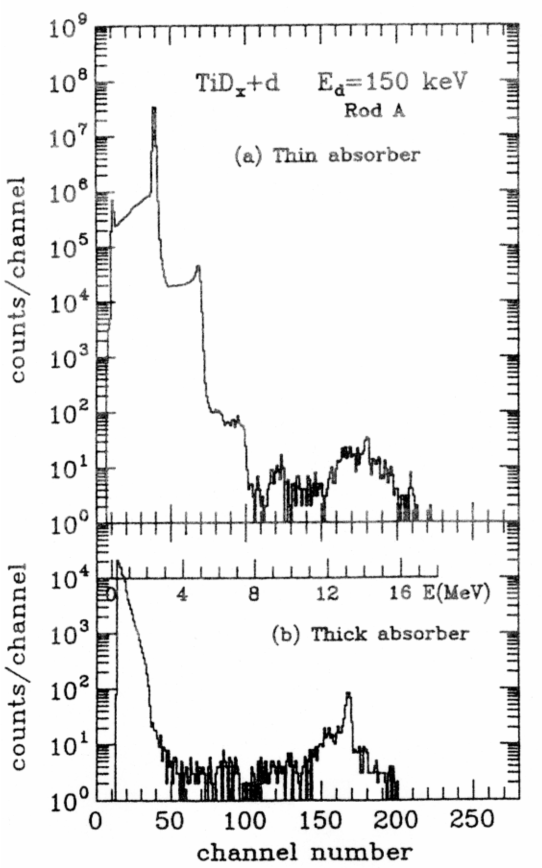

Fig. 1. Charged particle spectra obtained in the 150-keV deuteron bombardment on TiD$_x$ at 135°; (a) measured with a 15-μm thick Al absorber and (b) with a 200-μm thick Al absorber. The energy scale represents energy of charged particles after passing through the absorber.

2-2 Anomalous concentration of ^{3}He

The sharp peak at 14.1 MeV in Fig. 1 was interpreted as protons emitted in the ^{3}He(d,p)^{4}He (Q = 18.35 MeV) reaction. The observed peak just follows the kinematical prediction as shown in Fig. 2, where open circles show measured energies of the peak detected at angles of 90°, 110° and 135°, and a solid line shows the kinematical calculation for the ^{3}He(d,p)^{4}He reaction with E_d=150 keV. This interpretation requires an anomalous concentration of ^{3}He in the target before the bombardment began as discussed in ref. 6. Unfortunately, the reproducibility of appearance of the peak is quite low so that we have had only four cases which show the peak clearly, out of more than 100 bombardments on various TiD$_x$. Thus, a systematic study of the peak is very difficult. Nevertheless, during experiments in July 1993, we had a chance to see the sharp peak, again, and to investigate the region where ^{3}He atoms are concentrated. Fig. 3 shows the quantity of the ^{3}He atoms, which is proportional to the yield of the peak measured at various positions of the TiD$_x$. As seen, the ^{3}He atoms are heavily concentrated in a very narrow region; each position in Fig. 3 differs only 3 mm. Moreover, they seem to be concentrated on the surface area, because the yields measured after scraping off the surface of the TiD$_x$ are much smaller than those obtained for surface positions; open circles are data for the surface positions and open squares are those after scraping off.

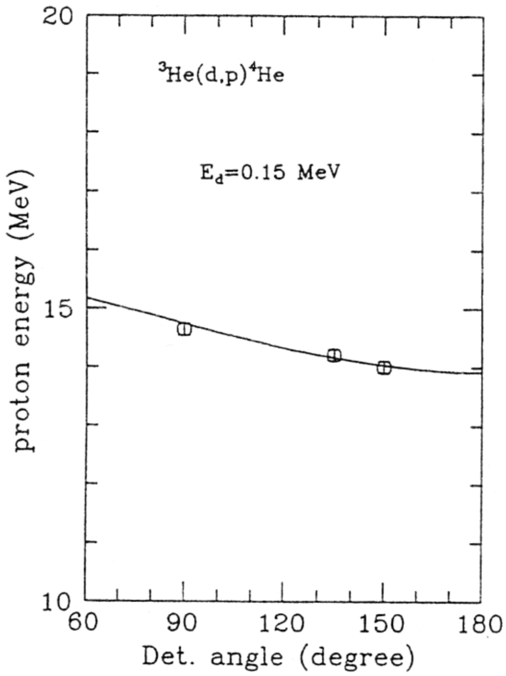

Fig. 2. Energy of the sharp peak observed in the 150-keV deuteron bombardment on TiD_x versus detected angle. The solid line is the kinematical calculation for the ^{3}He(d,p)^{4}He reaction at E_d = 150 keV.

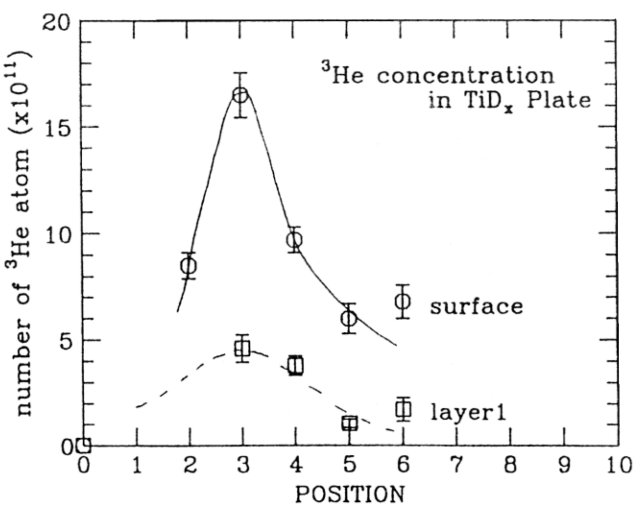

Fig. 3. Position dependence of the anomalous concentration of ^{3}He in TiD_x. The number of ^{3}He atoms is deduced from the yield of the peak from the ^{3}He(d,p)^{4}He reaction by changing the position of the bombardment on TiD_x. Open circles are data for the surface of TiD_x and open squares are those after scraping off.

2-3 Continuum protons and α-particles

The bump and the continuum in Fig. 1 always appear in any measurements on TiD_x as long as x > 1.2. In Fig. 4, high energy part of the proton spectra, measured at 110°, 135° and 155° are shown. Since the spectra in Fig. 4 were obtained with different targets from the one in Fig. 1, they do not show the sharp peak at 14.1 MeV, but do show the bump and continuum; its line shape and the maximum energy depends; on the detection angle.

In the subsequent measurements with a counter telescope, continuum energy α-particles were also observed. Fig. 5 shows a scatter plot of ΔE vs. E measured at 135° in the bombardment on TiD_x. Protons from the D(d,p)T reaction as well as their pileups are seen as a heavy locus, although the low energy part of ΔE is cut. In addition, a broad locus consisting of several tens of events are observed at larger ΔE, and which was actually assigned to be α-particles based on the observed energy loss. Energy spectra of the α-particles at 135° and 155° were deduced by setting a banana gate on the two-dimensional spectra. These spectra are shown with open circles in Fig. 6. As shown, α-particles are observed up to ~ 6.5 MeV (those with energy lower than 4.5 MeV were stopped in the ΔE detector).

In order to check the possibility that the α-particles are emitted in the secondary reactions induced by any materials in the target with the products of the primary D+D reactions, we have irradiated the target directly with neutrons, protons and ^{3}He particles and measured the charged particles. For neutron radiation, we used the same setup as the α-particle measurement. Another 2-mm thick TiDx plate was placed between the primary target and the detector. In this way, neutrons from the D+D reaction can irradiate the other target. No α-particles were observed for the irradiation with neutron dose of 10 times more than the experimental condition. The TiDx target were directly bombarded with 3.3 MeV proton and 1.5 MeV ^{3}He beam obtained from a Van de Graaff accelerator at Tokyo Institute of Technology. The irradiated dose was more than 100 times of that of the products of the D+D reaction. Again, no α-particles were observed. Thus, the α-particle production rate due to the secondary reactions induced by neutrons, protons and ^{3}He-particles is less than 10^{-7} of the reaction rate of the D+D reaction, i.e., at least one order of magnitude smaller than observed.

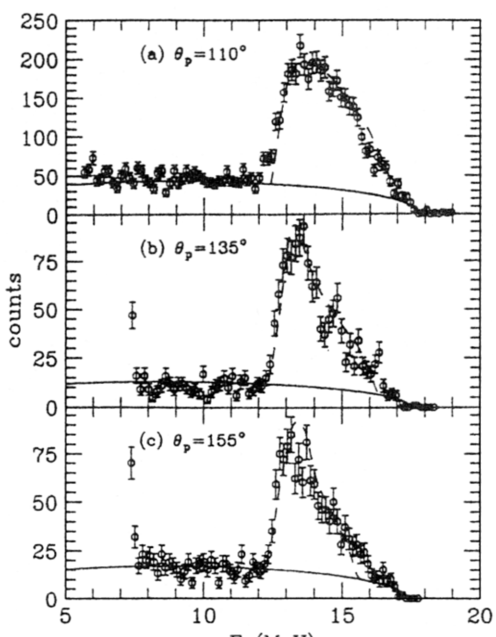

Fig. 4. High energy proton spectra measured at (a) 110°, (b) 135° and (c) 155°. Solid lines represent available phase spaces of protons emitted in the three-body reaction. Dashed lines are the calculated spectral shapes of protons emitted in the sequential reaction and are superimposed on the solid lines.

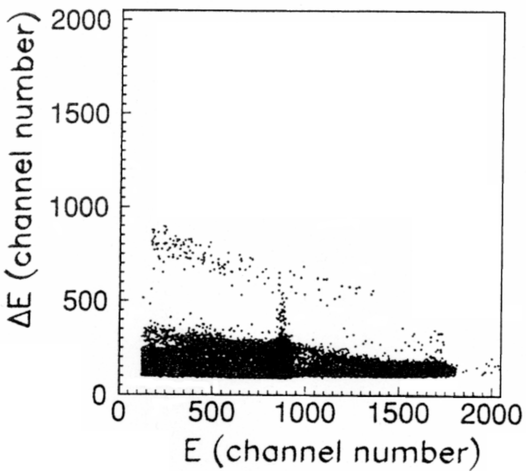

Fig. 5. Scatter plot of E vs ΔE, measured in the 150-keV deuteron bombardment on TiD_x at 135°. A broad locus at higher ΔE region is due to α-particles.

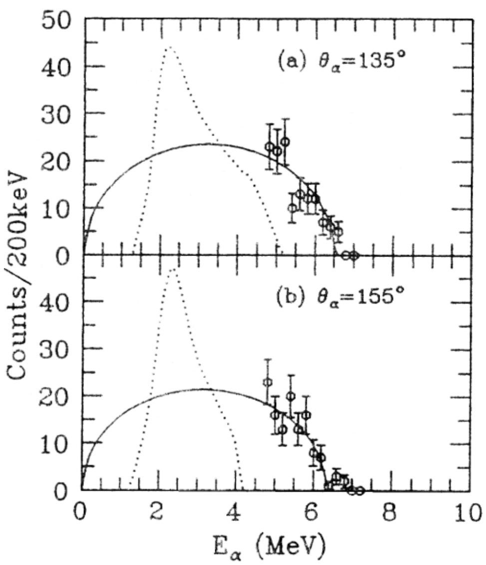

Fig. 6. Alpha-particle spectra measured at (a) 135° and (b) 155°. Solid lines represent available phase spaces of α-particles emitted in the three-body reaction. Dotted lines are the calculated spectral shapes of α-particles emitted in the sequential reaction.

2-4 Sequential reaction involving three deuterons

For the broad bump observed between 12 and 16 MeV in Fig. 4, protons are interpreted to be emitted in the $D(^3He,p)^4He$ reaction (Q = 18.35 MeV) which sequentially occurs following the primary $D(d,^3He)n$ reaction. In this case the ^3He particle ejected in the primary reaction reacts with deuterons at rest in the target. However, the protons emitted in the secondary reaction cannot form a sharp peak, because of the spread of energy and direction of the ^3He. In order to verify this situation quantitatively, the spectral shape of the protons has been calculated for the sequential reaction. An excitation function of the cross section and angular distributions of the primary $D(d,^3He)n$ reaction for Ed < 150 keV were taken from ref. 8. Angular distributions of the secondary $D(^3He,p)^4He$ reaction were assumed to be isotropic in the CM frame for $E3_{He}$ < 1-33 MeV (the maximum energy for the secondary reaction), and cross sections were estimated from the differential cross sections[9] and S-factors[8]. Values of deuteron and ^{3}He energy loss in Ti were taken from ref. 10.

In Fig. 4, spectra measured at 110°, 135° and 155° are compared with the calculations. Dashed lines in the figure are the calculated spectral shapes superimposed on the assumed continuum (discussed later) drawn with solid lines. As seen, the calculation reproduces the experimental shapes very well for each angle. Since the production rate of ^{3}He is proportional to the density of deuterons which are also the targets of the secondary reaction, the reaction rate of the sequential reaction is proportional to the square of the deuteron density in Ti. In order to reproduce the ratio of the yield of the bump to that of the peak of the D(d,p)T reaction (order of 10^{-5}), the required D/Ti ratio at the beam spot is more than 1.2. This explains why the bump was not easily seen for the targets with small D/Ti ratio (< 0.6).

2-5 Anomalous protons and a-particles

The fact that protons are observed with energies up to ~ 17 MeV, as clearly seen in Fig. 4, is really anomalous. As already mentioned, none of the secondary reactions with neutron, triton or ^{3}He can produce such high energy protons. The yield of the continuum part shown with solid lines in Fig. 4 is about 10^{-6} of that of the protons from the D(d,p)t reaction.

Alpha-particle emission in the bombardment is quite anomalous, as well. As shown in Fig. 6, the yield of the α-particles is quite small, again, about 10^{-6} of that of the protons produced in the D(d,p)T reaction. The spectral shape of emitted α-particles in the sequential reactions in which the D(^{3}He,α)p and the D(t,α)n reaction follows the D(d,^{3}He)n and the D(d,t)p reaction was calculated using the same program used to calculate the proton spectrum. The result also gave a bump in the α-particle spectrum. In this case, however, the expected bump lies in a lower energy region than observed, as shown with dotted lines in Fig. 6. Thus, the observed α-particles are not explained as the products of the sequential reactions involving another deuteron in the target.

The anomalous protons and α-particles were also observed in the deuteron bombardments on other metals. We have obtained such spectra for the bombardements on Au, Pd and Zr, as well as Ti. In Fig. 7 and 8 shown are the proton spectrum and ΔE-E plot measured in the Zr bombardment, respectively. In both spectra, the same structures are clearly seen as shown in Fig. 1 and Fig. 5. Therefore, the emissions of continuous protons up to 17 MeV and α-particles up to 6.5 MeV, that cannot be explained by the conceivable reactions, are common features for the low energy deuteron bombardments on metals.

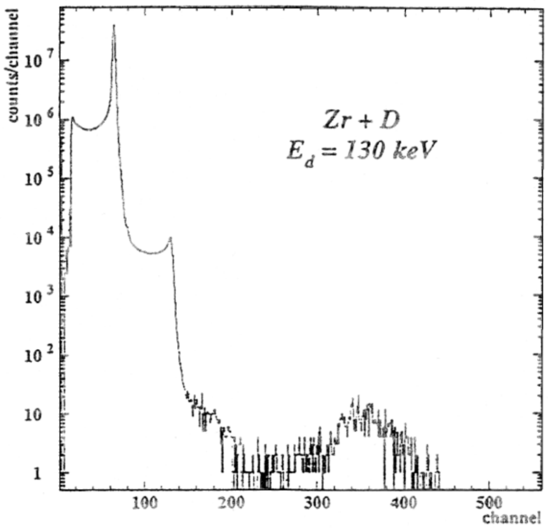

Fig. 7. Charged particle spectra obtained in the 130-keV deuteron bombardment on Zr at 135°.

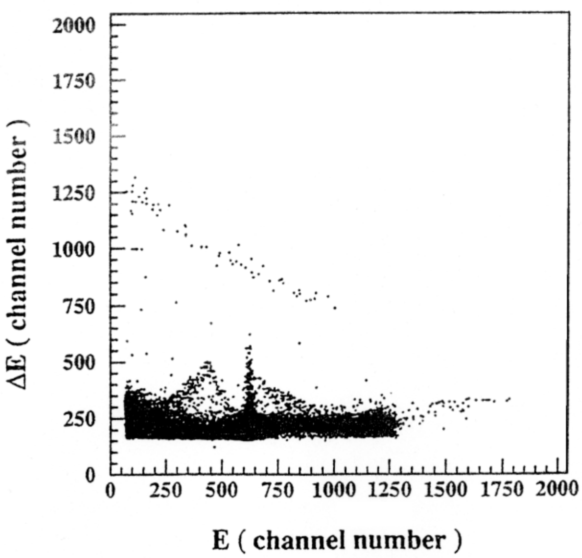

Fig. 8. Scatter plot of E vs ΔE, measured in the 130-keV deuteron bombardment on Zr at 135°.

2-6 Possibility of the simultaneous three-body D+D+D reaction

We have inferred, that the anomalous energetic protons and α-particles are attributed to a common origin. Of particular interest is the fact that the observed maximum energies up to which the anomalous particles are emitted are always larger than possible from the sequential reaction. Thus, we have speculated that the protons and α-particles are emitted in the reaction where the bombarded deuteron reacts with two deuterons in the target, without forming a real intermediate state. The incident deuteron first interacts with a deuteron to form a virtual intermediate state, which subsequently interacts with another deuteron and produces neutron, proton and α-particle. In this case, the available energy is shared with the three particles in the final state; the maximum energy of each particle can be larger than that of the sequential reaction.

Based on this very naive picture, we have calculated the phase spaces of protons and α-particles being available in the D+D+D→p+n+ α reaction (Q = 21 62 MeV). in the calculation, we assumed that the 150-keV deuteron reacts with two deuterons whose center of mass is at rest. The results of the calculation are drawn with solid lines in Fig. 4 and Fig. 6. Since the calculation depends only on the kinematical factor, yields are normalized with the data. As seen, the calculation explains both spectra very well; i.e., the maximum energies and their spectral shapes. Thus, the observed spectra indicate the possibility that the incident deuteron interacts with two other deuterons to produce n, p and α without forming any real nuclear state in an intermediate stage.

The integrated yields of the anomalous protons and α particles are about 10^{-5} times of that of the protons of the D(d,p)T reaction. The reaction rate of the hypothetical three-body reaction can be factorized as $R_{ddd} = A \times P(r_0)$, where $P(r_0)$ is the probability that the two deuterons in the target are within a distance of r_0. If the reaction proceeds only through the nuclear interaction, then r_0 should be on the order of a nuclear radius, about 10 fm. In order to obtain a rough value of R_{ddd}, we assume that A is equal to the reaction rate of the usual D+D reaction; the bombarded deuteron interacts with one of the deuterons of the pair and the virtual intermediate state immediately interacts with the other deuteron, resulting in neutron, proton and α-particle in the final state. The probability of finding the two deuterons at r_0 is basically determined by the Coulomb penetration factor. Ichimaru et al. calculated the probability in Pd and Ti, including an enhancement due to the many-particle processes of the screened deuterons.[5] The ratio of the three-body reaction rate to the two-body one is then estimated to be order of $\sim 10^{-17}$, which is far below the experimental ratio of 10^{-5}. Thus, various factors which are not included in the model calculation should be considered. For example, high concentration of deuteron realized by the continuous bombardment on highly deuterated metal would cause a flow of deuterons; the situation cannot be a static one. In a very low energy region, the penetration factor of deuterons might be much less than the expected one from the adiabatic approximation as shown for fusion reactions with light nuclei.[9] A possibility that the second step interaction is the electromagnetic one increases the value of r_0 and hence $P(r_0)$ can be much larger.

Recently, Kim et al.[11] developed the formulation to treat the simultaneous three-body reactions at low energies, and calculated the cross sections for the D+D+D reactions. The experimental ratio of the three- to two-body reaction, about 10^{-6}, can be explained by the calculation.

3. Reaction rates of the D+D reaction in Ti

The experiments were performed with low energy deuteron beams obtained from an RF ion source. The experimental details are reported in ref. 12. Thus, we will describe only the results here.

3-1 Thick target yield for the D+D reactions down to 4.8 keV

Due to the implanted beams, the deuterons were accumulated in TiD_x metal. The stopped deuterons do not stay at the same place, but diffuse to the surface so that the number of target deuterons increases during the measurements. Therefore, the thick target yields of p, t and ^{3}He measured at lower incident energies were normalized to the averaged yield at Ed = 15.4 keV which were measured before, during and after the run at each bombarding energy.

In Fig. 9, the relative thick target yields obtained for the D(d,p)T and D(d,n)^{3}He reactions are plotted against the bombarding energy. Errors associated with the data in the figure include possible sources of uncertainty; (a) statistics of the peaks measured for each bombarding energy, (b) background subtraction, (c) statistics of the peaks measured at 15.4 keV for the normalization, and (d) uncertainty of the target current measurement. Relative uncertainties of (c) and (d) are typically 3 ~ 5%.

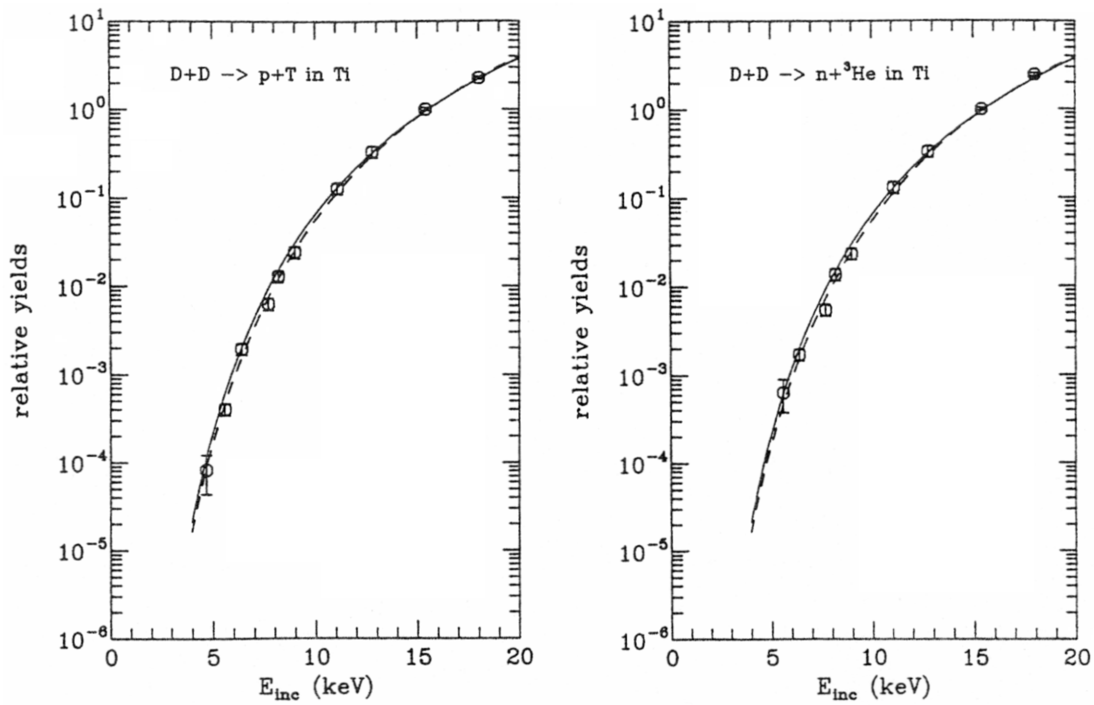

Fig. 9. Relative yields of protons and tritons (left) and ^{3}He emitted in the D+D→p+t and D+D→n+^{3}He reaction occurring in a thick; Ti plate against the bombarding energy. The data are normalized to the one measured at the bombarding energy of 15.4 keV. Thick target yields calculated with S-factors deduced from gas target measurements are plotted with a solid curve for uniform distribution of the deuterons in Ti and with a dashed curve for the frozen distribution.

3-2 Comparison of the reaction rate with gas target measurement

The D+D reaction at low incident energies have been, studied, so far, with gas target down to 6 keV by Krauss et al.[8] They deduced the astrophysical S-factors by dividing the reaction cross sections by the Gamow factor and the incident energy. The S-factors for the (d,p) and (d,n) reactions were then parameterized as a function of the incident energy. They are S(E) = 52.9 + 0.019E + 0.00192E^2 (keV·barn), for the D(d,p)T reaction, and S(E) = 49.7 + 0.170E + 0.00212E^2 (keV·barn), for the D(d,n)^{3}He reaction. We have compared the measured thick target yields with those calculated with the above S-factors; i.e., with the cross sections measured with gas target.

In the present experiment, the bombarded deuterons are slowed down in the Ti metal and the reactions can be generated at energies below the bombarding energy. The energy loss of deuterons were calculated with the dE/dx reported in ref. 10 for every 1-nm layer and the reaction yields were calculated with the S-factors for the corresponding energy. Total reaction yields are the sum of the yields of all layers. In the calculation, target deuterons are assumed to be uniformly distributed along the beam path. This assumption seems to be reasonable, because the range of the implanted deuterons are very small and the deuterons diffuse to the surface. However, in order to see the effect of the deuteron density distribution in Ti, we performed another calculation with an extreme assumption, i.e., a frozen distribution in which a target deuteron is assumed to stay at its stopped position. In this case, the ratio of the thick target yield at a given bombarding energy to that at E_d = 15.4 keV is calculated with the distribution predicted with the deuteron range and its straggling. The effect of the distribution becomes larger as the bombarding energy decreases, as shown in Fig. 8 and 9 in which the results with the uniform and the frozen distributions are plotted with solid and dotted lines, respectively. The frozen distribution gives about 20% lower relative yield at 5 keV. The experimental data agree very well with both calculations within error bars down to the lowest bombarding energy.

In a naive picture for the atomic target, the effect of the electron screening is approximately estimated with an enhancement factor

$$F(E) = \exp(k/E^{1/2})/\exp(k/(E+U_e)^{1/2}),$$

where $k = e^2/hc\cdot(mc^2/2)^{1/2}$, and U_e is the potential energy of the electrons at the screening distance of the target; i.e., at 0.053 nm (U_e = 27 eV) for hydrogen. In the D+D reaction with gas target, the effect should be observed below E_d = 20 keV. The S-factors used for the comparisons were deduced from the data including at E_d = 5.96 and 13.26 keV for the (d,p) and (d,n) reactions, respectively. Thus, the S-factors, especially the one for the (d,p) reaction, include the effect of screening by the atomic electrons. The present observation that the S-factors from gas target measurements can well explain the D+D reaction yields in Ti metal indicates that the screening effect is not different so much for deuterons in metal and in gas. Ichimaru[5] proposed the short-range screening distance to be 0.028 nm in TiD_2, which gives U_e = 51 eV instead of 27 eV of the atomic target. Thus the reaction rate is expected to be enhanced by about 20% at the bombarding energy of 6 keV when the comparison is made with bare nuclei cross sections. Thus, in the present case, where the comparisons are made with the S-factors from gas target measurements, the expected enhancement of the reaction rate is smaller than 10%, which cannot be distinguished within the experimental accuracy. In order to see the difference, data for much lower incident energies are required.

4. Summary and future plans

In the 150-keV deuteron bombardment on highly deuterated Ti, we have observed energetic protons and α-particles which cannot be emitted in the D+D reaction. They are the characteristics of the high energy proton structure, which include a sharp peak at 14.1 MeV, a broad bump between 12.5 and 16.5 MeV and a continuum up to 17 MeV, and continuous α-particles up to 6.5 MeV.

The sharp proton peak at 14.1 MeV was found out to be emitted in the ^{3}He(d,p)^{4}He reaction. The systematic study of the occurrence of this reaction was almost impossible because the reaction seldom takes place. Nevertheless, we have found that the concentration of ^{3}He was limited in a very narrow region on the surface of TiD_x.

A bump structure at around 14 MeV seen in the proton spectrum can be well explained as emitted in the sequential reaction involving three deuterons. The first step of the reaction is the D(d, ^{3}He)n reaction and the second one is the D(^{3}He,p)^{4}He reaction in which ^{3}He produced in the first step reacts with another deuteron in metal. It was found that the yield of the bump depends strongly on the deuteron density, since two deuterons in the target are involved in the reaction.

The continua of protons and α-particles up to ~17 and ~6.5 MeV, respectively, cannot be explained as the products of the conceivable two-body nuclear reactions. The possibility of attributing the protons and the α-particles to the three-body reaction, D+D+D→p+n+α, is suggested, since it can explain the maximum energies and spectral shapes. However, this possibility requires an anomalously large enhancement factor which is not understood at present. Thus, further experimental studies are highly desirable as well as theoretical studies, although the formulation proposed by Kim et al.[11] can explain the experimental yields.

Thick target yields of the D(d,p)T reaction were obtained down to 4.7 keV and those of the D(d,n)^{3}He reaction to 5.4 keV. They were compared with the thick target yields calculated with the S-factors which were deduced from gas target measurements. The comparisons show that the S-factors can well explain the present data down to the lowest bombarding energy. This indicates that the screening effects of deuterons in Ti metal and in gas: are not so much different. In order to see the difference of the screening effects, it is highly desirable to measure the reaction rate at lower incident energies.

For further experimental studies, especially for lower energy bombardments, we have constructed a low-energy high-intensity ion beam generator, which consists of a duoplasmatron ion source, a beam analyzer magnet, electrostatic lenses, decelerator (accelerator) electrode and a filter magnet for neutral beams. It was designed to produce deuteron beams from 1 keV to 100 keV with 1 mA. The machine was just installed in our laboratory, and a beam test has begun. We will measure the reaction rate of D+D reactions in various metals down to at least 2 keV in near future, in order to see whether the reactions in metal is really enhanced or not. Furthermore, p-α coincidence measurements are being planned in order to clarify that the high energy protons and a-particles are really originated from the simultaneous three-body D+D+D reaction in metal. These investigations may reveal interesting and important effects of the environment on the low energy nuclear reactions in metal and may open up a new interdisciplinary field which involves nuclear physics and condensed matter physics.

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