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Measurements of the D + D Reaction in Ti Metal with Incident Energies between 4.7 and 18 keV

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The D+D reactions in Ti metal were investigated for the deuteron incident energies between 4.7 and 18 keV. Observed were protons, tritons and ^{3}He particles emitted in the deuteron bombardment on TiD_x. Thick target yields for the D(d, p)T and D(d, n)^{3}He reactions were measured at bombarding energies down to 4.7 and 5.4 keV, respectively, for the first time. They were well explained with the reported astrophysical S-factors which were deduced from gas target measurements at $E_d > 6$ keV for the D(d, p)T reaction and $E_d > 13.3$ keV for the D(d, n)^{3}He reaction. The cross section ratio σ(d, p)/σ(d, n) was obtained down to 6.4 keV, and was found to be constant at around 1.0 for $E_d < 20$ keV.

KEYWORDS: D+D reaction in Ti, D(d, p)T and D(d, n)^{3}He reactions, thick target yields, astrophysical S-factor, cross section ratios

§1. Introduction

At energies far below the Coulomb barrier, cross sections of nuclear reactions are essentially dominated by the Coulomb penetration factor. An astrophysical S-factor, which contains information on pure nuclear part, is deduced by dividing the reaction cross section by the Coulomb penetration factor of two nuclei. Recently, in several reactions with light nuclei, such as the ^{3}He + d, ^{6,7}Li + d and ^{6,7}Li + p reactions, the deduced astrophysical S-factors for very low energies were found to be larger than those extrapolated from higher energy data.[1,2] The deviation seems to occur below the incident energy of about 20 keV for the ^{3}He + d reaction and about 80 keV for the Li+p,d reactions; corresponding classical closest distances at theses energies are 140 and 70 fm, respectively. Therefore, nuclei which sit each other in the distance farther than about 100 fm might experience a weaker repulsive potential than the Coulomb potential between the two bare nuclei. This behavior is associated with an effect of the screening caused by surrounding electrons, since the target nuclei are in the form of neutral atoms or molecules in the laboratory. However, the observed enhancement of the S-factors is not fully understood as the electron screening effect.[3] Thus, measurements of reaction cross sections at very low energies under various circumstances are highly interested.

Reaction cross sections of the D + D reaction at low energies have been measured by several authors,[4-7] and the S-factors for the D(d, p) and D(d, n) reactions were parameterized down to several keV.[7] However, the measurements were performed only with gas target, up to the present. It has been suggested that hydrogen nuclei in metals are strongly screened,[8] since the electrons both in metallic d-band and in hydrogen-induced s-band can contribute to the screening effect. The proposed screening distance between two hydrogen nuclei in Pd or Ti is much shorter than that of the atomic hydrogen. Thus, it is very interesting to measure the reaction cross sections for the different target conditions to see the difference of the screening effects, if it exists. In the present work, we have studied the D + D reaction in a special condition in which target deuterons sit in metal. The target used was a thick Ti plate in which D_2 gas was absorbed before the bombardment made. Detected were protons, tritons and ^{3}He particles emitted in the deuteron bombardment on a thick TiD_x plate at several incident energies down to 4.7 keV, which is the lowest bombarding energy for the D + D reaction so far studied.

The characteristics of the low energy experiment with a thick target are as follows. Nuclei sitting only on the surface region of the target can contribute to the nuclear reactions, because the reaction yield rapidly decreases as the incident beam penetrates into the target. The effective thickness of the target, thus, depends on the incident energy; those estimated for TiD_x are about 50 nm for 15 keV, and 15 nm for 5 keV. Furthermore, in the present experiment, deuterons which can become targets are being implanted into a thick plate during the measurement. Therefore, special cares are necessary to discuss the reaction rate.

§2. Experimental Procedure

The experiments were performed with low energy deuteron beams obtained from an RF ion source. The extracted beam was bent by 90 degree by a dipole magnet in order to select the atomic or molecular beam. Atomic beams were used for the measurements at the incident energies of 7.7, 11.1, 12.8, 15.4 and 18.0 keV and molecular beams were used at those of 4.7, 5.4, 6.4, 8.2 and 9.0 keV. The extracted and accelerated voltage were measured directly by using a voltage meter. In general, the extracted energy of the beam from an RF ion source is larger than the applied voltage for the extraction due to the excess voltage given by the RF field. Thus the bombarding energy was corrected for the excess voltage estimated according to ref. 9. The systematic error of the bombarding energy of the deuteron beam was estimated to be ± 100 eV.

The deuterated titanium plate (TiD_x, x~0.4), in which D_2 gas was absorbed by the method described in ref. 10, were set at the center of a scattering chamber. In order to avoid carbon build-up on the target, liquid nitrogen traps were set at the beam line and at the chamber. Since no focusing elements were placed in the beam line, the size of the beam was large, about 20 mm diameter, at the entrance of the scattering chamber. In order to reduce the beam size, a collimator of 3 mm in diameter was set at the entrance of the chamber. Thus, the beam was expected to be uniformly distributed within the 3-mm diameter beam spot. The electronic current from the target was being integrated during the run, because the beam stopped in the target. The measured current, thus, included the effect of the secondary electron emission from the target. In order to estimate the total dose of the deuteron beams, the electric current were directly measured with a Faraday cup several times during the run and compared with the current from the target. In average, the current measured from the target is about 20% larger than the current from the Faraday cup for the atomic beams and about 30% larger for the molecular beams. The current on the target was about 0.7 to 2 μA.

Protons, tritons and ^{3}He particles emitted in the D(d, p)t and D(d, n)^{3}He reactions occurring in TiDx plate were detected with two silicon detectors (thickness-area: 200mm-150mm^2 and 500 mm-200 mm^2) which were placed at about 2.5 cm from the target and at ± 135° in respect to the beam direction. In order to prevent δ-electrons and scattered deuterons from hitting the detectors directly, a 50 μg/cm^2-thick carbon foil was placed in front of each detector. Energy resolution of the detectors was checked with an ^{241}Am source and was about 30 keV for 5.48 MeV.

§3. Experimental Results

Figure 1 shows charged particle spectra obtained at E_d =15.4 and 6.4 keV. Three peaks observed at about 90, 120 and 460 ch correspond to ^{3}He-particles, tritons and protons, respectively, emitted in the D + D reaction. As shown in the figure, the ^{3}He peak is clearly seen at E_d =15.4keV, while at E_d = 6.4keV large backgrounds caused by electric noise compete with the peak, because the reaction cross section drops about three orders of magnitude in this energy range. Yields of ^{3}He and t were obtained for each bombarding energy by subtracting the background yields at the peak regions. The ^{3}He yields could not be obtained at the incident energies below E_d = 5.4 keV.

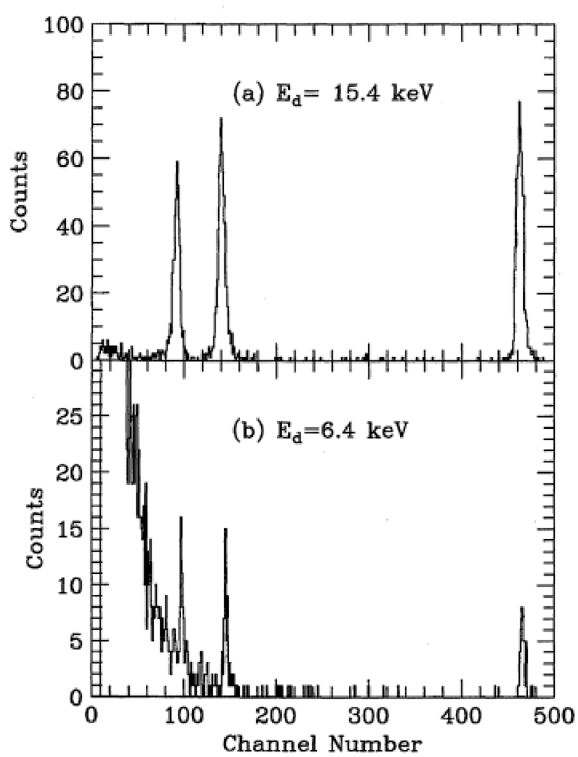

Fig. 1. Charged particle spectra obtained in the bombardment of the deuteron on thick deuterated Ti plate; (a) at E_d =15.4 keV and (b) 6.4 keV.

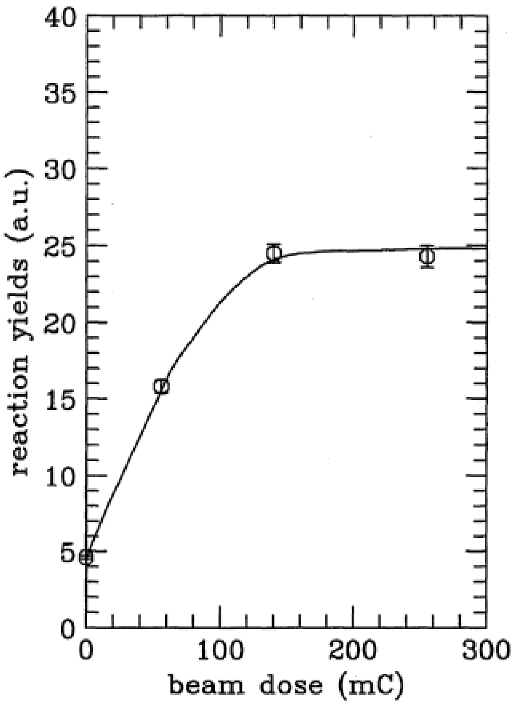

Fig. 2. Charged particle yields measured at E_d =15.4 keV against accumulated deuteron dose during the measurement at E_d = 6.4 keV.

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. This causes a serious problem to determine the number of the target deuterons, especially at lower bombarding energies. Since the reaction rate becomes smaller for the lower bombarding energies, the required total dose of the deuteron beams during a run becomes larger and, hence, the uncertainty of the number of the target deuterons becomes larger. For the measurements at lower bombarding energies, therefore, we measured the reaction yields at E_d = 15.4 keV during the run, because the reaction cross section is relatively large at E_d = 15.4keV so that it can be obtained without serious change of the amount of the target deuterons. The reaction yield at E_d = 15.4 keV, then, reflects the number of the target deuterons at that time. In Fig. 2 we show such a plot where the yields obtained at E_d = 15.4 keV during the 6.4-keV run are plotted against total amount of the deuteron beams of 6.4 keV. As shown, the number of the target deuterons increases as the total beam dose increases, and, then, it seems to saturate. The thick target yields of p, t and ^{3}He measured at lower incident energies were then normalized to the averaged yield at E_d = 15.4 keV which were measured before, during and after the run at each bombarding energy.

In order to deduce the angle integrated yields, the ratio of the detector solid angle in the center of mass system to that in the laboratory system was corrected for each bombarding energy and angular distributions are assumed to be isotropic in the center of mass system; this is well justified at these low energies.[3)] In Figs. 3 and 4, obtained relative thick target yields for the D(d, p)T and D(d, n)^{3}He reactions, which are normalized to the 15.4-keV data, are plotted against the bombarding energy, respectively. 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%. Errors associated with the ratio σ(d, p)/σ(d, n), which will be discussed below, include only (a) and (b).

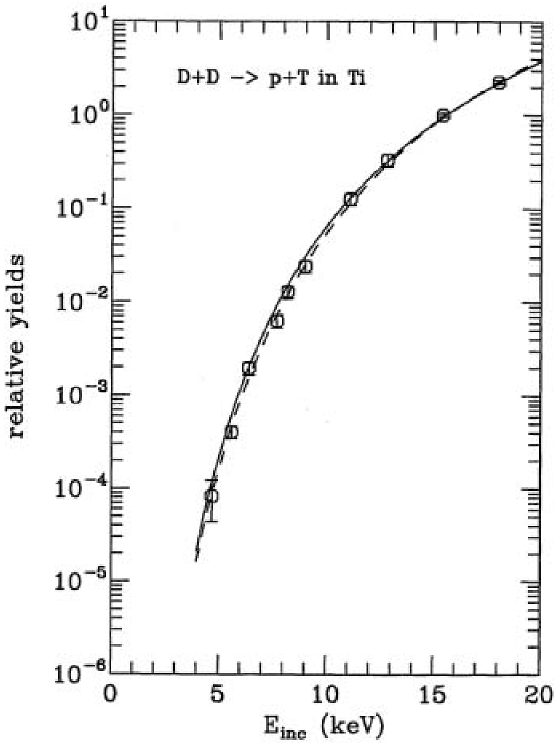

Fig. 3. Relative yields of protons and tritons emitted in the D + D → p + t reaction occurring in a thick Ti plate against the bombarding energy. The data are normalized to the one measured at the bombarding energy of 15.4 keV. Thick target yields calculated with S-factors deduced from gas target measurements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

The normalization method employed in the present analysis assumes that the intensity distribution of the beam for each bombarding energy is same as that for 15.4 keV. Although the uniformity of the beam intensity is expected to be very good within beam spot as mentioned in §2, we estimated the effect of nonuniformity of the beam intensity distribution. For an extreme case, the intensity distribution ($I(r)$) of the beam is assumed as $I(r) = I_{max} \times \exp(-r/R)$, with R = 10mm; the value is deduced from the beam size at the entrance of the collimator. It is natural to assume that the beam after the collimator keeps I_{max} within the beam spot (3 mm diameter), because the beam was always adjusted so as to obtain the maximum current during the measurement. Thus, comparisons were made for putting I_{max} on various positions within a circle of 3-mm diameter. The effect of the difference of I_{max} position does not exceed 4% to the normalized relative yield.

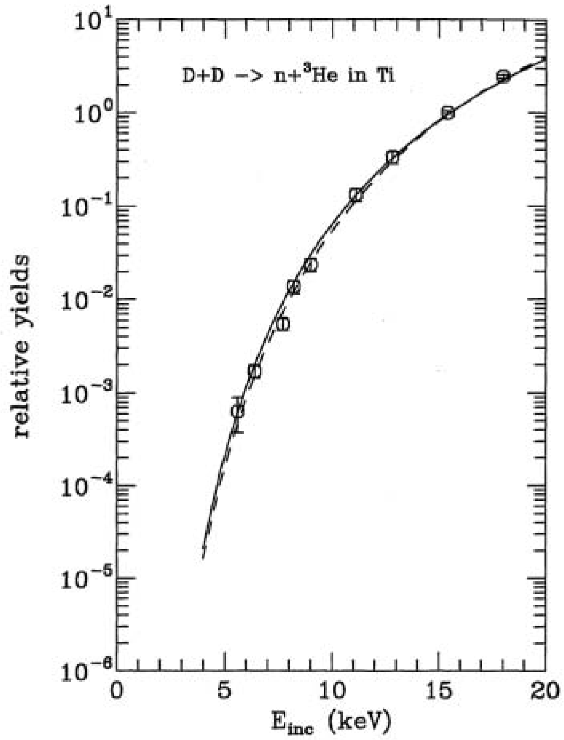

Fig. 4. Same as Fig. 3 except that relative yields of ^3He emitted in the D + D-»n + ^3He reaction are plotted.

§4. Discussion

In the present work, we have measured the relative reaction rates of the D + D reaction in Ti metal at bombarding energies down to 4.8 keV, for the first time. The D + D reaction at low incident energies have been studied, so far, with gas target down to 6 keV by Krauss *et al.*[7] 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 was calculated with the dE/dx reported in ref. 11 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 is 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=l5.4 keV is calculated with the distribution predicted with the deuteron range and its straggling.[11] The effect of the distribution becomes larger as the bombarding energy decreases, as shown in Figs. 3 and 4 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 $k=e^2/\hbar c\cdot(mc^2/2)^{1/2}$, 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 = 27eV) in the case of 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[8] 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.

In Fig. 5, the ratio of the yield of the D(d, p)T to that of the D(d, n)^{3}He reaction is plotted against incident energies (with open circles), together with the data (with squares and diamonds) by Krauss *et al.*[7] The previous data show the tendency that the proton emission overcomes the neutron emission as decreasing the incident energy. The present data at around 15 keV are consistent with the previous ones, and the data below 15 keV down to 6.4 keV show that the ratio is almost 1.0. Thus, it can be concluded that the ratio stays constant at about 1.0 for E_d < 20 keV and starts to decrease above 20 keV. A solid line in the figure is the ratio calculated from the S-factors mentioned above. As a matter of course, it explains the ratio very well for E_d > 20 keV, since the S-factor of the (d, n) reaction was parameterized above 13 keV. However, it overestimates the ratio below 20 keV. In order to obtain better fit to the data, we slightly modified the S-factor of the (d, n) reaction as $S(E)=51.0+0.16E+0.00212E^2$. The ratio calculated with the modified S-factor is plotted with a dashed and dotted line in the figure. The S-factor of the (d, n) reaction has stronger energy dependence than that of the (d, p) reaction as described with a larger coefficient of the linear term; this produces the energy dependence of the yield ratio σ(d, p)/σ(d, n). The recent theoretical calculation by Koonin and Mukerjee[12] including the Oppenheimer-Phillips effect[13] cannot reproduce this tendency; the calculated ratio stays almost constant at 0.83 for these incident energies as shown with a dashed line in Fig. 5.

A similar trend was reported for the reaction ratio for the ^6Li + d reactions at very low energies;[14)] the experimental ratio cannot be explained either.

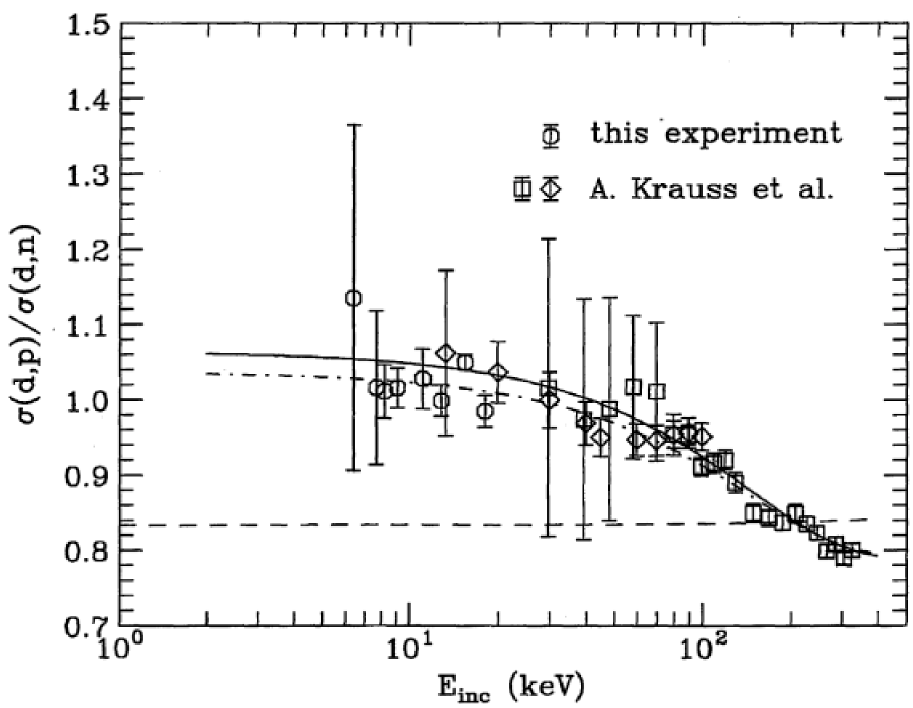

Fig. 5. Ratios of the cross section of the D(d, p)t reaction 10 that of the D(d, n)^{3}He reaction versus incident energy. Open circles are those obtained In this experiment (below 18 keV), while squares and diamonds are with the solid curve, while a slight modification of the S-factor for the D(d, n)^{3}He reaction gives the dotted-dashed curve. A dashed curve shows a theoretical model calculation from ref. 12.

§5. Conclusion

In the present work, the D + D reactions in Ti metal were investigated for the incident deuteron energy between 4.7 and 18.0 keV. 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 5.4keV. 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 different so much. In order to see the difference of the screening effects, more accurate experimental data are required as well as data at lower incident energies. The ratios of the yield of the D(d, p)T reaction to that of the D(d, n) ^{3} He reaction were obtained down to E_d = 6.4 keV in the present work. It was found that the ratios stay constant at about 1.0 up to E_d = 20 keV, and then decreases above 20 keV. We proposed the S-factor of the (d, n) reaction to be slightly modified in order to obtain better fit to the data. The fact that the ratios show rather strong incident energy dependence can never be explained by the calculation including Oppenheimer-Phillips effect. Theoretical considerations for the very low energy region are highly desirable.

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Note added in proof—Greife *et al.* recently reported the measurement of the d + d fusion reactions down to E_{cm} = 1.6keV.[15)] The obtained cross sections at E_{cm}<10keV show clear evidence for electron screening effect.