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Measurement of the D(d,p)T Reaction in Ti for 2.5 < E_d < 6.5 keV and Electron Screening in Metal

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In order to study the electron screening effect on low-energy nuclear reactions in metals, the D+D reaction in Ti was investigated. Measured were thick target yields of protons emitted in the D(d, p)T reaction from the bombardment of Ti metal with deuteron energies between 2.5 and 6.5 keV. The obtained yields were compared with those predicted by using the parameterization of cross sections at higher energies. It was found that the reaction rates in Ti are slightly enhanced over those of the bare D+D reaction for E_d < 4.3 keV, and the enhancement can be interpreted as caused by the electron screening. The electron screening potential in Ti is deduced for the first time to be 19 ± 12 eV.

KEYWORDS: nuclear reactions in metal, D(d,p)T reaction, thick target yield, electron screening

§1. Introduction

The effect of the electronic environment on nuclear phenomena has been investigated for many years.[1)] For nuclear decay processes accompanied with a 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 a change in the transition rate, although the observed change is very small. The so-called cold fusion[2)] has roused attention more generally on the influence of the environment where nuclear processes take place. Although most of the experiments reported at that time were known to have a difficulty in the reproducibility and, hence, to be under suspicion, the influence of the environment in various nuclear processes is one of the interesting subjects which need more study, 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 electron environment really affects the nuclear processes very strongly.

Nuclear reactions at very low energies are considered naturally 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 non-negligible 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] suggested that hydrogen nuclei in metals are strongly screened, since the electrons both in metallic *d*-band and hydrogen-induced *s*-band can contribute to the screening effect. They calculated the effective static potential for hydrogen in Ti and Pd, and proposed that the screening distance between two hydrogen ions in metal is much shorter than that of atomic hydrogen.

The D+D reactions have been investigated with gas targets by many authors.[6-11] Krauss *et al.*[9] parametarized the *S*-factors for the D(d,p) and D(d,n) reactions by fitting their data with a quadratic polynomial for $5 < -E_{\mathrm{c.m.}} < 120$ keV. Bosch and Hale[10] parameterized the reaction cross sections by using the *R*-matrix parameters of the D+D reaction which were determined with all types of experimental data including integrated cross sections, differential cross sections and polarizations. Greife *et al.*[11] recently reported the measurement at center-of-mass energies down to 1.6 keV. The deduced astrophysical *S*-factors below 10 keV are clearly larger than predicted from the parameterization of Bosch and Hale.[10] They interpreted the observed enhancement as the screening effect of the bound electron, and obtained a screening potential of 25 eV. The D+D reaction in Ti was studied first by Roth *et al.*[12] They reported that no enhancement of the cross section of the D+D reaction is observed down to 3 keV within their statistical error of ±50%. Kasagi *et al.*[13] measured the reaction rate of the D+D reactions in Ti for bombarding energies between 4.8 and 18 keV. The obtained thick target yields are well explained with the *S*-factors deduced from the gas target experiment. Thus, for the D+D reactions in metal, experiments with much lower energies and with good statistics are highly desirable to observe the screening effects of metallic electrons.

Recently, a low-energy high-current ion beam generator was introduced to our laboratory to study the nuclear reactions with much lower energies. The present work is a natural extension of the previous work in ref. 13, by using the newly installed machine. Lower energy data were obtained for the D+D reaction in Ti metal down to 2.5 keV, for the first time, in this experiment.

§2. Experimental Procedure

The low-energy high-current beam generator was designed to produce deuteron beams with several hundreds of μA. from 1 keV to 100 keV. As shown in Fig. 1. it consists of a duoplasmatron ion source, an ion beam extraction system, a 30-degree bending magnet, focusing lenses, an acceleration/deceleration electrode and a neutral beam filter magnet. The duoplasmatron ion source can provide high current beam (~1 mA) with low energy spread (≤25eV). The beam is extracted from the ion source with ~25 KV. After passing through the magnet and focusing lenses, the beam is accelerated or decelerated by changing the connection of the main power supply providing stable voltage up to 80 KV (stability and voltage ripple is less than 0.01%). In the acceleration mode, the beam is transported straight to the target position. In the deceleration mode, however, the beam is bent by 45° with a dipole magnet placed in the scattering chamber, in order to remove neutral beams which cannot be decelerated. The beam energy is determined by the electrical potential between the ion source and the ground, which is measured by a register chain with a digital meter.

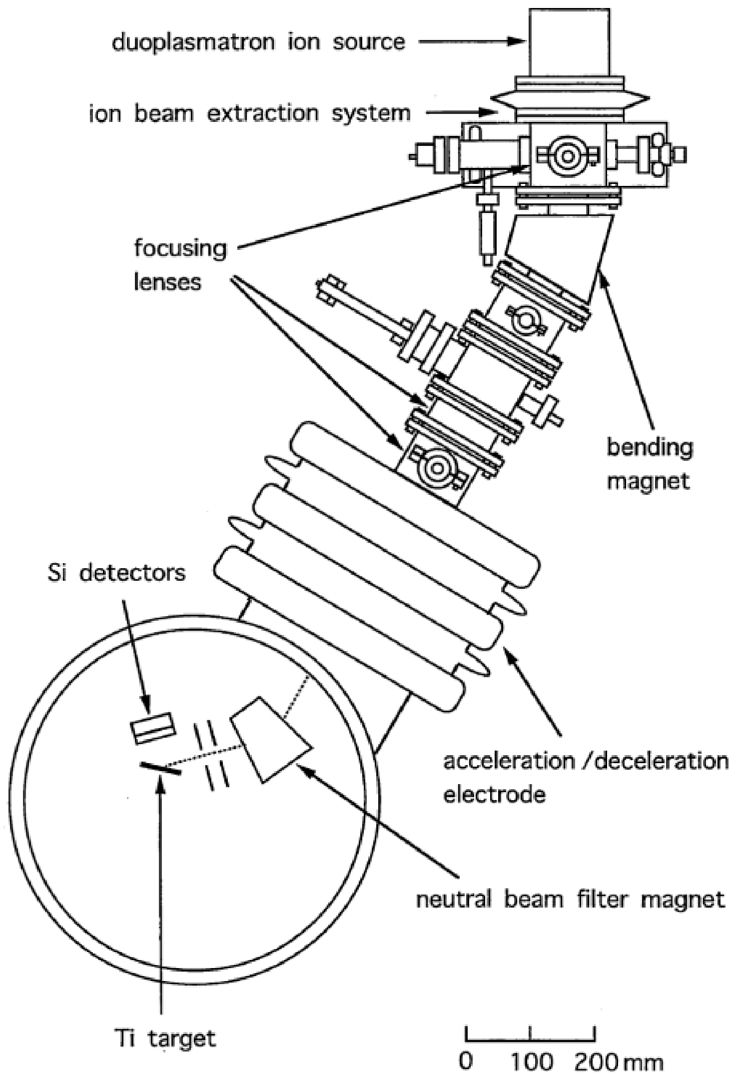

Fig. 1. Schematic diagram of low-energy beam generator and experimental setup.

A thick plate of Ti (10mm × 30mm × 2mm), in which deuterium gas was absorbed, was placed at the target position. The method of loading gas into the Ti plate is described in ref. 14. In order to fix the beam spot on the target, a beam collimator was set between the filter magnet and the target. The beam spot on the target was about 4mm in diameter. The target current, as well as the collimator current, was monitored during the run.

Since a spectrum measured with a single Si detector was found out to contain much electrical noise at low bombarding energies, a ΔE - E counter telescope consisting of 50-μm and 200-μm thick Si surface barrier detectors was employed. A requirement of a coincidence between two Si detectors almost completely reduced the electrical noise. The front face of the ΔE detector was covered with a 15-μm thick Al foil to prevent δ-rays and scattered deuterons from hitting the detector. The telescope was placed at 2 cm from the target and at 90° with respect to the beam direction. The target was tilted by 58° and the solid angle subtended with the telescope was about 3.5% of 4π sr. Signals from the detectors were fed to preamplifiers which generate fast outputs for time information as well as slow outputs for pulse height information. The fast outputs were fed into timing filter amplifiers and time signals were picked up from constant fraction discriminators. CAMAC ADCs and TDCs were used, respectively, to measure pulse height spectra for each detector and time spectra between the two detectors.

As will be mentioned below, the present measurements only give relative values of the reaction rate. The total dose of the deuteron beam for each run was deduced from the electric current from the target, which might depend on the bombarding energy since an amount of secondary electron emission from the target may depend on the energy. Thus, the electric current was also measured with a Faraday cup and was compared to the target current. Ratios of the current from the target to that from the Faraday cup were found to be quite constant for $2.45 < E_d < 7.95$ keV; they fall between 1.02 and 1.06. Thus, no correction was made for the total dose deduced from the target current for each bombarding energy. The target current was about 500μA at $E_d = 6.5$ keV and 100μA at $E_d = 2.5$ keV.

As was described in ref. 13, the difficulty is to determine the number of target deuterons in the D+D reaction. Since the deuterons are accumulated in TiD_x during the deuteron bombardment, the number of the target deuterons changes. We applied the same method as described in ref. 13 in order to measure the relative values of the reaction rates. In the present work, the yields of protons at $E_d = 6.45$ keV were frequently measured during the run; for example, the yields at $E_d = 6.45$ keV were measured every 2 mC of the beam charge accumulation for 200 mC of the total bombardment at $E_d = 2.55$ keV. The reaction rate at $E_d = 6.45$ keV is much larger than those at lower bombarding energies so that it reflects the number of the target deuterons at that time. The measured thick target yields at lower incident energies were normalized to the yield at $E_d = 6.45$ keV.

§3. Experimental Result

As mentioned, protons emitted in the D+D reaction were measured with a counter telescope in order to obtain good signal to noise ratio. Fig. 2 shows such spectra obtained at $E_d = 4.5$ keV; Fig. 2(a) is a TDC spectrum where a window employed to discriminate true events from those due to electrical noise is also shown, and two dimensional spectra of ΔE versus E are shown in Figs. 2(b) and 2(c); they are obtained, respectively, without and with setting a gate on the window. As expected, events due to electrical noise that are distributed mainly along both ΔE and E axes in Fig. 2(b) are completely eliminated in Fig. 2(c). A band seen as a line of a constant value of $\Delta E+E$ corresponds to the protons emitted in the D+D reaction, since heavier charge particles cannot punch through the ΔE detector. The solid angle and the area of both detectors are quite large and hence the energies deposited in the ΔE detector are rather widely spread.

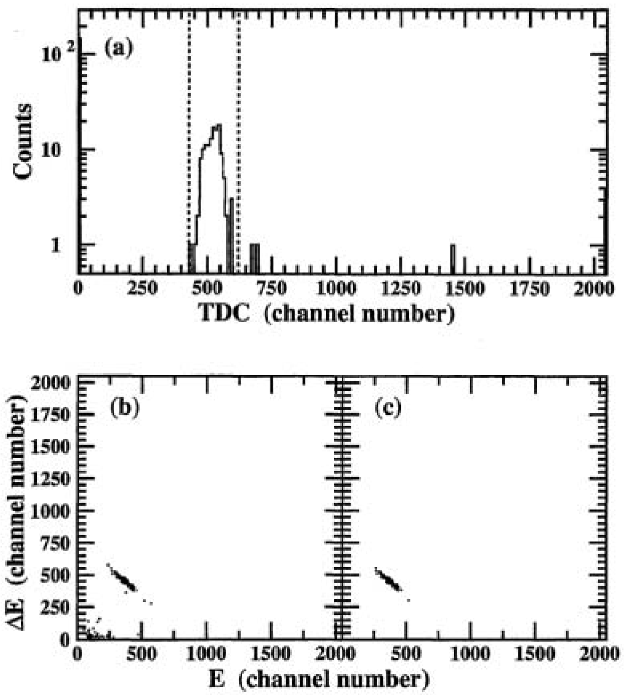

Fig. 2. Spectra measured with a *ΔE - E* Si counter telescope in the deuteron bombardment of TiD_x; (a) TDC spectrum with a gate position indicated by dotted lines, (b) two-dimensional spectrum of *ΔE* vs. *E* without any selection, and (c) same as m (b) but selected by setting the TDC gate.

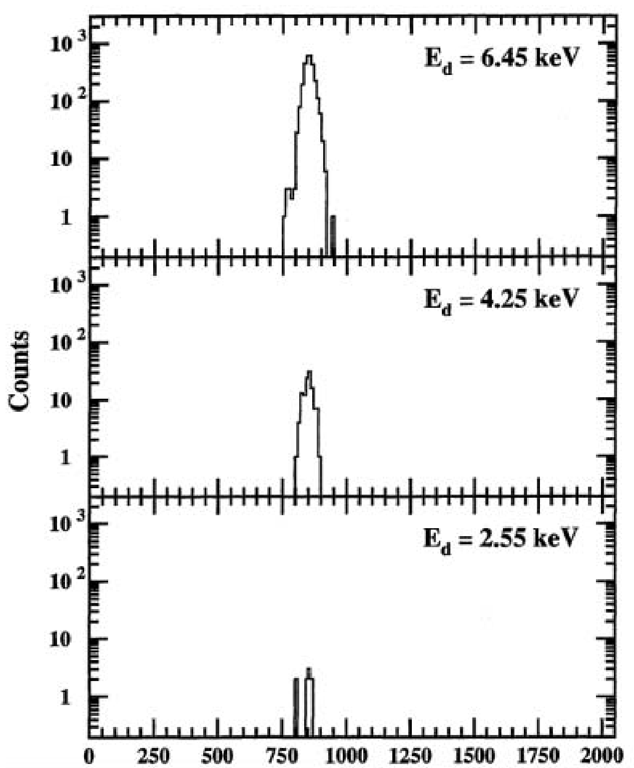

Fig. 3. Charged particle spectra measured in the D(d,p)T reaction in Ti plotted against *ΔE+E.*

In Fig. 3, yields of the true events are plotted against $\Delta E + E$ for various bombarding energies. As shown, the band observed in the two dimensional spectrum is seen as a sharp peak located at the same position, and no other events are observed than the peak. After the correction of the energy loss in Al absorber, the energy of the peak is deduced to be 3.0 MeV, corresponding to the proton emitted in the D+D → p+T reaction. The yield of the peak decreases steeply as the bombarding energy decreases.

In order to deduce the angle integrated yields, the correction only for the ratio of the detector solid angle $(\Delta\Omega)_{c.m.}/(\Delta\Omega)_{lab}$ was made, since the angular distributions are isotropic in the center of mass system at such low bombarding energies.[9)] In Fig. 4. the deduced thick target yields of the D(d,p)T reaction in Ti are plotted against the bombardment energy. The data are normalized at 6.45 keV as mentioned in the previous section, and errors associated with the data in the figure include only statistical ones; i.e., statistics of the yield at each bombarding energy and those at 6.45 keV for the normalization runs. As shown in the figure, the yield decreases very rapidly as the bombarding energy decreases.

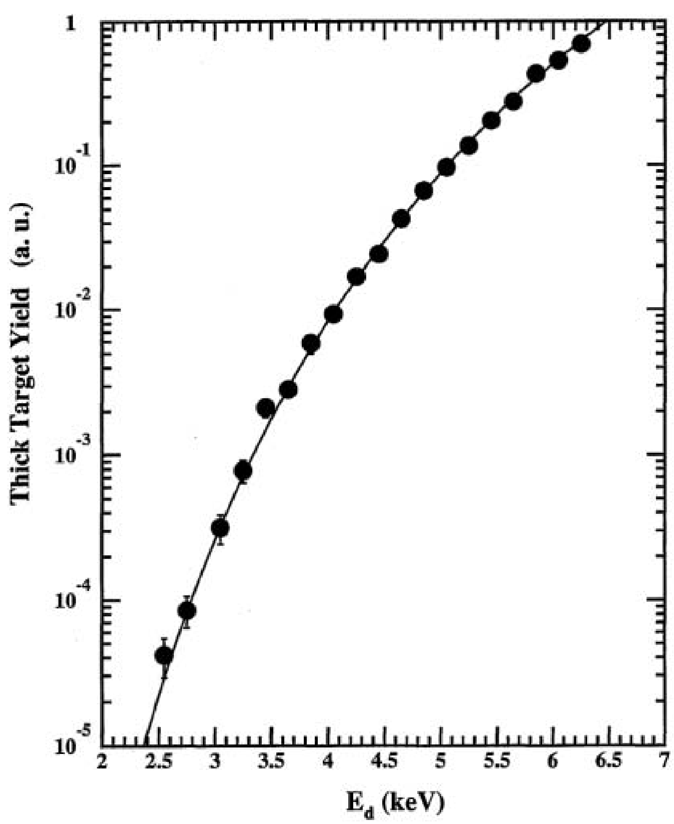

Fig. 4. Yield of the D(d,p)T reaction occurring in thick Ti metal target as a function of the deuteron bombarding energy. Data are normalized to the yield at 6.45 keV. A solid curve shows a thick target yield calculated with the bare cross section.

§4. Comparison of the Thick Target Yield with the Bare Cross Section

Since the bombarded deuterons are slowed down in Ti metal and the reactions can occur until the deuteron stops, the thick target yield Y_t at the bombarding energy of E_b is given as follows.

$$\begin{aligned} Y_t(E_b) &= A \cdot \int N_D(x) \cdot \sigma(E) dx \\ &= A \cdot \int N_D(x) \cdot \sigma(E) \cdot (dE/dx)^{-1} dE, \end{aligned} \tag{1}$$

where $N_D(x)$, $\sigma(E)$ and dE/dx are the number of the target deuterons per unit area, the reaction cross section and the stopping power of the deuteron, respectively. Thus, the obtained thick target yields should be compared with those calculated with the reaction cross section of the bare D+D reaction, in order to see whether the reaction rate in Ti is larger than that of the reaction in vacuum or not.

The parameterization of Bosch and Hale[10)] has been used for the cross section. The parameterization is based on the R-matrix theory which describes different reactions (T(p,p)T, D(d,p)T and D(d,n)^{3}He reactions) simultaneously with a single set of R-matrix parameters, and was made with the high-energy data ($E_d > 15$ keV), where no electron screening effects occur. Therefore, use of the parameterization is a better determination of the cross section of the D(d,p)T reaction, and the extrapolation of the reaction cross section to lower energies represents the case of the bare D+D reaction.

The stopping power of the deuteron is also necessary to calculate the thick target yield. Although an accurate value of the stopping power is important, no experimental information on the stopping power of the deuteron has been available in the region of the present bombarding energies. Thus we followed the recipe of Anderson and Ziegler[15)] (we will call this the stopping cross section of AZ), in which the electronic stopping power is assumed to be proportional to the velocity of the projectile (normalized to the experimental data at higher energies) at very low energies in accordance with the Thomas-Fermi model of the atom and the LSS[16)] nuclear stopping power is employed. The electronic stopping power is considered only for the Ti atom, whereas the nuclear stopping power of the D atom is included as well as the Ti atom. The atomic ratio of D/Ti is assumed to be 2.0 in the analysis. The density distribution of the target deuterons along the incident deuteron path is also important for estimating the reaction rate. The projected range of the 6.5-keV deuteron is only about 40 nm in Ti, and the target deuterons are assumed to be uniformly distributed.

The thick target yield calculated with eq. (1) is normalized to that at 6.45 keV as the experimental ones and is plotted with the solid line in Fig. 4. Since it is difficult to compare the experimental data with the calculations quantitatively in a log-scale graph, the ratios of the experimental yields to the calculated ones are plotted in Fig. 5 as a function of the bombarding energy. The ratios for all the experimental data are plotted in Fig. 5(a), in which the experimental reaction yields are slightly enhanced over the calculated ones at the lower bombarding energies, although large statistical errors and fluctuations obscure a definite conclusion. However, the enhancement can be seen more clearly for $E_d < 4$ keV in Fig. 5(b), where the weighted average values of the ratios of three points are plotted. Since a fine structure of the excitation function of the thick target yield cannot be expected, the averaging process is an effective method for reducing the statistical errors.

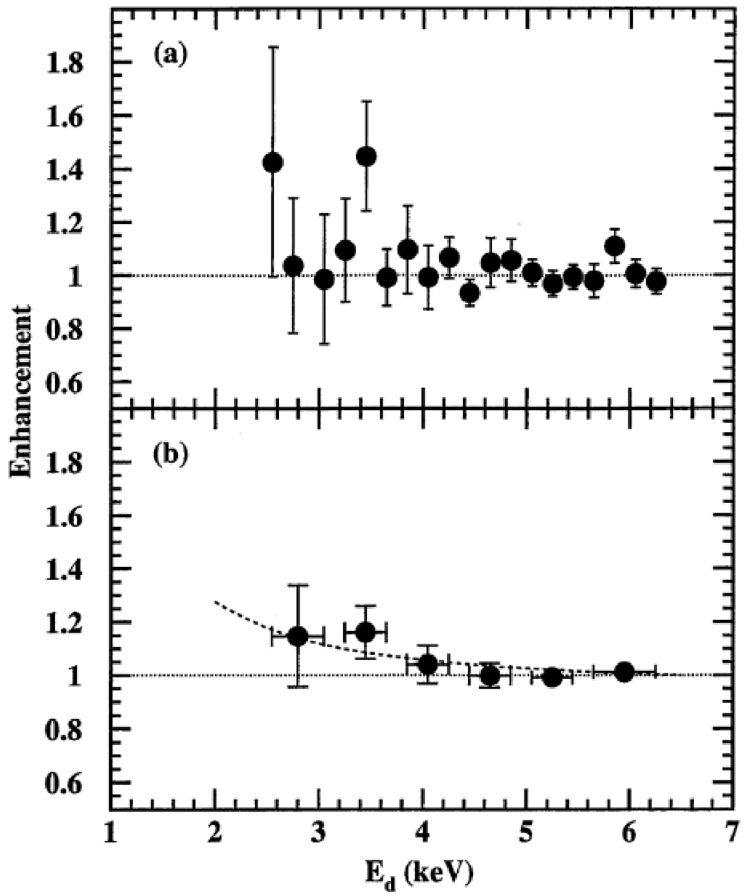

Fig. 5. Ratios of the thick target yield of the D(d, p)T reaction to the calculated one. The calculation was made with the bare cross section so that the ratio shows enhancement of the reaction rate. The ratios for each bombarding energy are shown in (a), and the weighted averages of the ratios of three points are shown in (b). A dashed curve in (b) shows the calculation with the electron screening potential of 19 eV.

§5. Electron Screening Potential

As seen in Fig. 5, the observed enhancement of the reaction rate seems to increase as the incident energy decreases. Thus it can be naively interpreted as the reduction of the effective Coulomb barrier due to the electron screening potential in metal. An enhancement factor of the reaction cross section is given as $f(E) \approx \exp(\pi\eta U_e/E)$, where U_e is the electron screening potential and η is the Sommerfeld parameter. We calculated thick target yields for various values of U_e and compared them with the experimental ones. Chi-squared values were calculated for $0 < U_e <$ 50 eV and the result is plotted in Fig. 6. As shown in the figure, the χ^2-plot has a minimum at 19 eV. A quadratic curvature around the minimum gives one standard deviation of 12 eV. Therefore, we can conclude that the electron screening potential affecting the D+D reaction is obtained for the first time in Ti metal as $U_e = 19 \pm 12$ eV. The dashed line in Fig. 5(b) is the calculated enhancement with $U_e = 19$ eV, and explains the data very well.

The effect of the stopping cross section on the deduced value of U_e should be discussed here. The first question is concerned with the velocity proportionality of the electronic stopping power. Recent measurements on electronic stopping cross sections of low-energy protons in helium gas show that they are not proportional to the projectile velocity but strongly deviate from proportionality below 20 keV.[17] A main cause of this large discrepancy from the commonly used assumption is due to the fact that the threshold energy, below which the projectile cannot transfer its kinetic energy to the He atom, is quite high because of its large ionization energy. In the present experiment, however, deuterons slow down in Ti metal and can transfer their kinetic energy to conduction electrons. Therefore, the velocity proportionality of the electronic stopping cross section at very low energies, which is the basic assumption of the stopping cross section of AZ, is considered to be valid, because the threshold energy does not exist in this case.

The second question is concerned with the nuclear stopping cross section. The stopping cross section of AZ (dE/dx_{AZ}) for deuterons in Ti was determined by the normalization of data at the deuteron energies between 30 and 100 keV.[18] At lower energies, it is expressed as

$$dE/dx_{AZ} = f_e \cdot (dE/dx_{LSS})_e + f_n \cdot (dE/dx_{LSS})_n ,$$

where (dE/dx_{LSS}) is the stopping cross section of LSS[16] and subscripts e and n represent electronic and nuclear, respectively. For the electronic stopping cross section, the normalization $f_e = 2.16$, and it is used in the present analysis, While $f_n = 1.0$ is simply assumed, since no experimental information on the nuclear stopping cross section has been available. This uncertainty may affect the deduced value of U_e, since the velocity dependence of the nuclear stopping cross section is very different from that of the electronic one. We calculated the thick target yields with various values of f_e and f_n, and discussed the effect on the final result. First, we consider the two extreme cases; one with only the electronic stopping cross section ($f_e = 2.16, f_n = 0.0$) and the other with only the nuclear stopping cross section ($f_e = 0.0, f_n = 1.0$). In the former case the stopping cross section decreases faster than that of AZ as the deuteron velocity decreases. While in the latter case, the stopping cross section increases as the deuteron slows down. The calculated thick target yields with these stopping cross sections give the values of $U_e = 6.6$ and 132 eV for the former and the latter, respectively. These values set the maximum uncertainties associated with the stopping cross section. However, such extreme cases never happen, since both the electronic and the nuclear stopping must contribute to the slowing down process. In fact, $f_e = 1.3$ and $f_n = 1.0$ was suggested for heavy atoms in Ti from Doppler-shift attenuation measurements;[19] $U_e = 27$ eV is deduced in this case. Therefore, we cannot determine the appropriate error associated with the uncertainty of the stopping cross sections because of the complete lack of experimental information on the nuclear stopping cross section. Instead, we present the values of $U_e = 12$ and 28 eV, which correspond to $f_n = 0.5$ and $f_n = 2.0$, respectively ($f_e = 2.16$ for both cases), as criteria. Measurements of the stopping cross sections of deuterons in Ti at very low energies are highly desirable.

The uncertainty of the number of the deuterons in Ti metal during the bombardment should be discussed also. Morimoto *et al.*[20] measured the depth profile of the deuteron in the 1.6-μm foil of Ti at a bombarding energy of 5 keV. According to them, deuterons are accumulated rather uniformly inside the foil in the beginning of the implantation. However, the higher the deuteron fluency, the worse the uniformity of the deuteron density becomes. The final profile shows a broad peak at about 200 nm from the surface. The density of the deuteron almost linearly increases with the depth from the surface, and differs about 25% between the surface and at 40 nm from the surface for the worst case. The reaction rate was calculated by using this distribution. The result gives $U_e = 23$ eV; the difference from the result with the uniform distribution is very small (+4 eV), since the reaction occurs effectively at the surface region.

Although the present result clearly shows that the D+D reaction rate is enhanced in Ti for $E_d < 4$ keV, the deduced electron screening potential $U_e = 19\pm12$ eV is smaller than the one predicted by Ichimaru.[5] They calculated the effective D-D potential in TiD_2 and obtained the short-range screening distance to be 0.028 nm, which corresponds to $U_e = 51$ eV. However, the comparison of the present result with their prediction may not be pertinent, since only the thermal motion of the deuteron in the Ti lattice is considered in their calculation, whereas deuterons are impinging with several keV and diffuse in the Ti in the experiment. A theoretical treatment for the low energy nuclear reaction including the effect of the electronic environment is highly desirable. It should be noticed that the present result is in good agreement with the one *(*$U_e = 25\pm5$ eV) obtained in the gas target measurement by Greife *et al*[11] They measured the reaction cross sections of the D(d, p)T reaction with a gas target down to $E_d = 3.2$ keV, and compared those with the same cross section as employed in the present analysis. Although their result is larger than the naive prediction *(*$U_e = 14$ eV) for the molecular D_2 gas case, the agreement of the two results indicates that electrons in the metallic d-band in Ti do not affect strongly the D+D reaction at a few keV.

§6. Summary

We measured the reaction rate of the D+D reaction in Ti metal at low bombarding energies down to $E_d = 2.55$ keV by using a deuteron beam from the low-energy high-current beam generator, installed recently in our laboratory. Protons emitted in the D(d, p)T reaction were detected without any electrical noise by employing a ΔE - E counter telescope. The measured thick target yield at each bombarding energy was normalized to that at 6.45 keV. The thick target yields were compared with the bare D+D reaction cross section; the parameterization of Bosch and Hale[10] for the cross section and the stopping power of Anderson and Ziegler[15] were used for the comparison. The enhancement of the reaction rate is clearly seen for $E_d < 4$ keV in the comparison, and can be interpreted as the reduction of the effective Coulomb barrier due to the electron screening in metal. The screening potential affecting the D+D reaction in Ti metal is obtained for the first time as $U_e = 19 \pm 12$ eV. Although we cannot determine the appropriate error associated with the uncertainty of the stopping cross section due to the lack of the experimental data at very low energy, we discuss that the most probable value of U_e might fall between 12 and 28 eV. The effect of uncertainty of the number of deuteron in Ti metal was found out to be very small. The deduced value $U_e = 19 \pm 12$ eV is smaller than the prediction of Ichmaru,[5] but is in good agreement with the one obtained in the gas target measurement by Greife *et al*.[11] This indicates that the electrons in the metallic d-band in Ti do not affect strongly the D+D reaction at a few keV. A theoretical treatment for the low energy nuclear reaction including the effect of the electronic environment is highly desirable.

Acknowledgments

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