# Anomalous Energetic Proton Emission during 170 keV Deuteron Bombardment of TiD<sub>2</sub>

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TiH<sub>2</sub> and TiD<sub>2</sub> thick targets were bombarded with 170 keV protons and with 170 keV deuterons. Charged particles from nuclear reactions were observed by means of a surface barrier particle detector shielded by an energy absorbing foil. Deuteron irradiation of TiD<sub>2</sub> produced these observations: copious ~2.5 MeV neutrons and ~3 MeV protons from direct d–d reactions; gamma rays from p,  $\gamma$  and n,  $\gamma$  reactions; ~14 MeV protons from the secondary d(<sup>3</sup>He,p) $\alpha$  reaction; and an anomalous, broad energy distribution proton signal between ~5–11 MeV. Our observations confirm the anomalous proton emission at a rate ~4 × 10<sup>-5</sup> of the d–d fusion rate.

KEYWORDS: energetic proton emission, d-d nuclear reaction, deuteron irradiation, Ti deuteride, anomalous nuclear effects, cross section

# 1. Introduction

The anomalous particle measurements of Kasagi et al. from Tohoku University suggest that three deuterium atoms undergo fusion in metal deuteride targets when exposed to low energy (90–150 keV) deuterium bombardment.<sup>1)</sup> The emissions consisted of protons at  $\sim$ 6–17 MeV and <sup>4</sup>He at  $\sim$ 4.5–6.5 MeV, where the end-point energies of the proton and the alpha signals were consistent with that expected from a 3-body d-d-d-fusion reaction. The proton emission yields were between  $10^{-5}$  and  $10^{-6}$  that of the normal 2-d fusion cross section. Additional evidence was supplied by Takahaashi who presented emission of 4.5 MeV particles consistent with two-body breakup in d–d–d reactions.<sup>2)</sup> If the anomalous proton emission is due to d-d-d fusion, the result is anomalous since there is no currently accepted physical mechanism that would explain the phenomenon. The proposed source of the anomalous proton emission in refs. 1 and 2 raises serious concerns because is implies that two deuterons are close together in the TiD<sub>2</sub> lattice, and the energetic deuteron reacts with the deuteron pair. If this is true, the signal strength could be interpreted as a measure of the probability of finding two deuterons within a certain distance in the solid-a problem that is a theoretical challenge. This conjecture as to the origin of the anomalous proton signal in ref. 1, however remote the possibility of being true, warrants a second look with improved signal-tonoise experiments.

In this work, a series of experiments were undertaken to, 1) determine if the initial result of Kasagi *et al.* is caused by an experimental artifact, and 2) extend knowledge of the anomaly if it is reproduced at another laboratory. The experiment is straightforward but suffers from poor signal-to-noise due to the effective  $\sim 0.1 \,\mu$ barn effective cross section measured in ref. 1, and the fact that the signal is a continuous energy distribution of protons with no well defined proton energy peak. We hoped to improve upon the signal-to-noise of the measurements by employing  $\sim 20 \times$ 

higher beam current, and targets that contain  ${\sim}20\%$  greater concentration of deuterium.

### 1.1 Relevant nuclear reactions

Reaction products that are expected from deuterium bombardment of  $TiD_2$  at 170 keV energy include products from primary reactions (d+target d, d+impurity), and secondary reactions (d+d reaction products with target and impurities). Possible reactions with impurities are discussed in a later section.

### 1.1.1 Primary reactions (d+d)

A list of such reactions for incident deuterons include:

$$d + d \rightarrow t + p, \qquad Q = 4.03 \text{ MeV} (1.01 + 3.02) \quad (1a)$$
  

$$\rightarrow {}^{3}\text{He} + n, \qquad Q = 3.27 \text{ MeV} (0.82 + 2.45) \quad (1b)$$
  

$$\rightarrow {}^{4}\text{He} + \gamma, \qquad Q = 23.85 \text{ MeV}. \quad (1c)$$

The numbers in parentheses are the energies of the final state nuclei at 0 incident energy. The reaction (1c) is an electromagnetic channel so it is safely neglected.

### 1.1.2 Secondary reactions

For the above primary products  $[t(1.01 \text{ MeV}), p(3.02 \text{ MeV}), {}^{3}\text{He}(0.82 \text{ MeV}), n(2.45 \text{ MeV})]$  as projectiles and d as target, the only open channels are:

$$\begin{array}{rl} t+d \rightarrow {}^{4}\text{He}+n, & 17.59 \ \text{MeV} \ (3.52+14.07) & (2a) \\ \rightarrow {}^{5}\text{He}^{*}, & 16.70 \ \text{MeV} & (2b) \\ {}^{3}\text{He}+d \rightarrow {}^{4}\text{He}+p, & 18.35 \ \text{MeV} \ (3.67+14.68) & (2c) \\ \rightarrow {}^{5}\text{Li}^{*}, & 16.39 \ \text{MeV} & (2d) \\ n+d \rightarrow t+\gamma, & 6.26 \ \text{MeV} & (2e) \end{array}$$

$$p + d \rightarrow {}^{3}\text{He} + \gamma, \quad 5.49 \text{ MeV}.$$
 (2f)

For reactions (2b) and (2d) the mass 5 products are too short lived ( $\sim 10^{-21}$  s) to react with another nucleus before decay and need not be considered. Reactions (2e) and (2f) are electromagnetic and second generation and can be neglected for a charged particle detector. This leaves only reactions (2a) and (2c) to be accounted for.

If we take the Kasagi et al. conjecture at face value, then

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<sup>1.1.3</sup> Decay channels for d-d-d fusion

the decay channels to consider for d–d–d fusion with high Q values are:

$$d + d + d \rightarrow {}^{4}\text{He} + p + n$$

$$Q = 21.62 \text{ MeV} (3\text{-body}) \qquad (3a)$$

$$d + d + d \rightarrow {}^{3}\text{He} + t$$

$$Q = 9.53 \text{ MeV} (4.76 + 4.76)$$
 (3b)

$$d + d + d \rightarrow {}^{4}\text{He} + d$$

$$Q = 23.84 \text{ MeV} (8.0 + 13.9)$$
 (3c)

Kasagi *et al.* measured ~6–17 MeV protons and ~4.5–6.5 MeV alpha particles in independent experiments. The only reaction that would produce a continuous proton and alpha spectrum is (3a) since it is a 3-body decay channel. We did not study the alpha particle emission since there are numerous d,alpha and p,alpha reactions with target impurities that could conspire to produce a spectrum that mimics the alpha particle from a d–d–d reaction. The energetic proton emission, however, is difficult to attribute to any known d,p reactions with impurities and alpha particle detection in high background of the protons from the primary d–d reaction is problematic.<sup>2)</sup> Therefore, we sought only the proton signal.

### 2. Experimental Procedure

Targets were 1.27-cm-diameter, 1.6-mm-thick buttons with compositions of  $\text{TiH}_{1.93}$  and  $\text{TiD}_{1.97}$  (determined by precision x-ray diffraction) produced by gas charging at high temperature. Details of the target preparation were previously reported.<sup>3)</sup> Table I is the known impurity content of the targets reproduced from ref. 3.

A 50 mm<sup>2</sup>, 2-mm-depletion-depth Si surface-barrier particle detector was placed at scattering angles between 90° and 135°. Al, Ta or Pb absorber foils of various thickness served to protect the detector face from scattered primary beam, or provided ranging foil thickness for particle identification. For most of the runs, they prevented the ~3 MeV protons from reaching the detector. The detector solid angle subtended was 32 or 21 msr depending on the experiment. Detector calibration at high proton energy was accomplished by deuterium bombardment of a pure Ti sample that was implanted with <sup>3</sup>He (5 × 10<sup>16</sup>/cm<sup>2</sup>, 50 keV). For low energy calibration, radioactive <sup>241</sup>Am was employed. The total unscanned beam currents of 170 keV deuterons were 6–30  $\mu$ A in a beam spot 4 mm by 4 mm square. The projected range of the deuterons in TiD<sub>2</sub> is 1.1  $\mu$ m.

Ion beams were produced by a 200 kV commercial ionimplanter. A modified end station shown in Fig. 1 provided the scattering chamber that was isolated from ground for charge collection. The particle detector and signal cable were isolated from chamber ground. The samples were mounted on a Ta sheet with conducting silver paste, and the Ta sheet was fixed to a Mo plate that could be cooled. No cooling was found to be necessary as monitored by a thermocouple placed on the Mo plate that did not exceed 80 °C. Scattering chamber vacuum was typically  $< 2 \times 10^{-6}$  Torr.

### 3. Results: Basic Spectra

Several initial spectra with different foil thickness were accumulated to compare with the results of ref. 1. A  $TiD_2$  target was bombarded with 170 keV deuterium ions with the

**Table I.** Impurity concentrations in the Ti stock used to synthesize the titanium hydrides, and impurity concentrations in the hydrides.

Element	Concentration (at. %)	Notes
Н	0.002	For TiD <sub>2</sub> sample only
Li	0.00002	Measured <sup>3)</sup>
В	0.0003	Measured <sup>3)</sup>
С	0.01	From vendor assay
Ν	0.013	//
Ο	0.015	//
Ti	99.7	//
Fe	0.1	//



Fig. 1. (Color) Schematic diagram of the scattering chamber and detector configurations.

particle detector placed at a scattering angle of 90°. A 10um-thick Al absorber foil was placed in front of the particle detector. Figure 2 shows the multichannel analyzer, semilog plot of counts per mC collected charge versus channel number. The peak near channel  $\sim 125$  is the  $\sim 3$  MeV proton from the (t, p) channel of the d-d reaction [reaction (1a) above]. The first pile-up peak extends to channel  $\sim$ 250 and the second pile-up peak extends to channel  $\sim$ 375. There is also a low intensity, high-energy peak in channels  $\sim$ 550–670. This peak, identified as protons by ranging foil experiments, is the 14.7 MeV proton from the secondary (<sup>3</sup>He, d) reaction [(2c) above], broadened by energy-loss in the target and absorber foil and kinematic energy spread arising from the variation in combined angles of the two reaction events. Between channels  $\sim$ 375 and  $\sim$ 550 is a small yield of counts of unknown origin. Kasagi et al. also observed these counts.

Another particle spectrum is shown in Fig. 3 under identical conditions as in Fig. 2 except that the Al foil thickness was increased to  $130\,\mu\text{m}$  to absorb all of the ~3 MeV protons from the primary d-d reaction and all alpha particles emitted from primary and secondary reactions. There is a monotonically decreasing low energy yield extending to channel ~150; a peak due to the 14.7 MeV proton from the secondary reaction (2c) shifted down in energy compared to Fig. 1 to channels ~500–630 or 10.8–



Fig. 2. Energy spectrum of charged particles emitted at an angle of  $90^{\circ}$  from a TiD<sub>2</sub> target bombarded by 170 keV deuterium ions. A 10-µm-thick Al foil in front of the detector stops elastically scattered deuterium.



**Fig. 3.** Energy spectrum of charged particles emitted at an angle of  $90^{\circ}$  from a TiD<sub>2</sub> target bombarded by 170 keV deuterium ions. A 130-µm-thick Al foil in front of the detector stops protons with energy <4 MeV.

13.7 MeV (11.8–14.6 MeV initial) by the Al absorber foil; and an approximately uniform particle yield of unknown origin in channels  $\sim$ 150–500 corresponding to an energy range 3.1–10.8 MeV.

#### 3.1 Investigation of noise sources

Several possible backgrounds contributions were investigated. During d bombardment of TiD<sub>2</sub>, high fluxes of 3 MeV protons and 2.5 MeV neutrons are incident on the chamber walls, absorber foils, the target and target holder. Since neutrons are not stopped by the absorber foil, the detector sees high fluxes of neutrons. Furthermore, the protons and neutrons produce a very large gamma ray flux by generating (p,  $\gamma$ ) and (n,  $\gamma$ ) reactions.

### 3.1.1 Fast and thermal neutrons

To investigate the effect of fast neutrons and thermal neutrons on the detector, the Al absorber foil was replaced



**Fig. 4.** Energy spectrum of charged particles emitted at an angle of  $90^{\circ}$  from a TiD<sub>2</sub> target bombarded by 170 keV deuterium ions. A 550-µm-thick Ta foil placed in front of the detector stops protons with energy <20 MeV. Paraffin shielding was placed around the chamber.

by a 0.55 mm thick Ta absorber that stopped up to 20 MeV protons. This Ta foil would also stop many fast neutrons, but few thermal neutrons. Most gamma rays would also penetrate the Ta. The particle spectrum for this experiment is shown in Fig. 4. Compared to Fig. 3, the low energy monotonically decreasing yield is virtually unchanged, indicating that it is not caused by primary reaction charged particles. The low energy yield is absent with no beam on the target so it is not electronic noise. Furthermore, there remains a signal above channel  $\sim$ 150 up to channel  $\sim$ 580 of approximately the same magnitude as in Fig. 3. The counts below channel 150 in Figs. 3 and 4 must be caused by the large neutron flux from the (<sup>3</sup>He, n) channel of the primary d-d reaction (1b), or by gamma rays from  $(n, \gamma)$  or  $(p, \gamma)$ reactions, or both. A very high flux of thermal neutrons was present during these experiments due to a large amount of paraffin neutron moderators placed near the target chamber to serve as radiation shielding. There was concern about  $(n, \gamma)$  reactions producing gamma rays that would generate background, so steps were taken to reduce this contribution.

Figure 5 shows a spectrum taken under identical geometrical conditions as in Fig. 4 but with the paraffin removed, and a 1 mm Pb absorber in place of the Ta absorber due to its smaller fast neutron absorption cross section. The counts in channels 150 up to 580 are significantly reduced compared to Fig. 4. The counts remaining in these channels are attributed to the 2.5 MeV neutrons from the primary d-d reaction [reaction (1b) above] interacting directly with the detector or indirectly with detector packaging materials. These counts remain of unknown specific origin. However, the yield is lower than that expected from the anomalous proton signal strength measured by Kasagi, so yield of these counts is not a significant artifact in his measurement. The spectrum in Fig. 5 is very useful here since it provides a quantitative "neutron correction", when normalized for incident collected charge, to any subsequent spectra taken without the thick Pb absorber foil. This correction is independent of the aperture size on the detector since the neutrons penetrate the Pb aperture, and the correction scales with total charge collected.



Fig. 5. Energy spectrum of charged particles emitted at an angle of  $90^{\circ}$  from a TiD<sub>2</sub> target bombarded by 170 keV deuterium ions. A 1-mm-thick Pb foil placed in front of the detector stops protons with energy <20 MeV. Paraffin shielding was removed from around chamber.

### 3.1.2 Gamma rays

The low energy monotonically decreasing signal in Fig. 5 changed shape from a positive curvature in Fig. 4 to a negative curvature, and melds with the background at channel  $\sim 120$  as opposed to channel  $\sim 150$  in Fig. 4. The yield above channel 120 is significantly reduced without the paraffin around the chamber. Gamma ray spectra were taken for the bombardment conditions used in Figs. 2-5 using a  $3.8 \times 2.5 \,\mathrm{cm}^2$  NaI gamma ray detector placed a 90° that viewed the target through a quartz window. There was an enormous gamma ray flux extending up to  $\sim 10 \,\text{MeV}$  that decreased significantly when the paraffin was removed. With the paraffin, few peaks were visible. Without the paraffin, peaks were noted but the flux was still so high that any anomalous peaks from low probability events would be masked. The gamma detector was not used after this. The low energy monotonically decreasing signal in Figs. 3-5, that was characterized by Kasagi as electronic noise, is caused by gamma ray Compton scattering events in the 2 mm thick Si detector. Their maximum energy corresponds to the maximum energy that 10 MeV gammas could deposit in the detector ( $E_{\text{max}} \simeq 3.5 \text{ MeV}$ ) by Compton events. While there is a high flux of these events, they cut off at sufficiently low energy that they do not interfere with the measurement of protons at higher energy. The rate of the low energy Compton counts was found to decrease by about a factor of 2 when a 1-mm-thick Pb can was placed to shield the detector (see Fig. 1).

### 3.1.3 Detector efficiency effects

The <sup>3</sup>He-implanted Ti sample was used for two purposes. Bombardment with 170 keV deuterons produces 14.7 MeV protons from the inverse of reaction (2c) above that is convenient for a detector calibration point at high energy. It also provides a quantitative correction for detector artifacts.

The implanted target was bombarded without ( $\sim$ 32 msr) and with ( $\sim$ 21 msr) a detector defining aperture consisting of 1 mm Pb foil with a 6 mm diameter hole centered over the detector. The spectrum for the case with the defining aperture is shown in Fig. 6(a) that displays the sharp proton



**Fig. 6.** Energy spectrum of charged particles emitted at an angle of  $135^{\circ}$  from a <sup>3</sup>He ion-implanted Ti target bombarded by 170 keV deuterium ions. A 140 µm thick Al foil was placed in front of the detector: (a)  $10^4$  counts full scale; (b) 50 counts full scale.

peak from the  $d^{-3}$ He reaction. Figure 6(b) is the same spectrum on an expanded scale that shows background counts between the peak and the low energy cut-off (~channels 120-550). These counts interfere with a measurement of anomalous protons in these channels. The counts are caused by incomplete charge collection in the detector (no detector is perfect), by protons that escape out the sides of the detector before losing all their energy, by protons that scatter from the edges of the 1-mm-thick Pb washer or in the absorber foil, and possibly by reaction protons that undergo multiple scattering in the target on their way to the detector. These counts were greatly diminished by application of the aperture as compared to similar spectra without the aperture. In Figs. 2 and 3, the proton peak is considerably broadened by the fact that the primary d-d reaction is isotropic so that the <sup>3</sup>He can be moving toward or away from the detector when the reaction occurs.

By taking the ratio of the sum of the counts in channels  $\sim$ 120 to 500 to the sum of the counts in the <sup>3</sup>He-d proton peak (channels 501–700) in Fig. 6(b), a quantitative

correction can be applied to any spectra taken for deuteron bombardment of  $TiD_2$ . This is called the "detector correction" and is applied by summing the counts from the secondary <sup>3</sup>He-d reaction protons and applying the ratio to the counts in the channel of interest. This correction depends on the detector aperture, and on the specific detector employed. It is independent of collected charge.

### 3.1.4 Reactions with impurities

The TiH<sub>2</sub> samples were used to search for reactions with impurities as a possible cause of the anomalous protons measured by Kasagi. Our TiD<sub>2</sub> and TiH<sub>2</sub> samples were synthesized from the same Ti rod, so their impurity content should be the same. This was thoroughly checked and found to be a correct assumption as reported in ref. 3. In that work, the targets were found to contain  $\sim 0.2$  ppm Li and  $\sim 3$  ppm B that were not indicated by the material vendor as impurities in the material (see Table I). Due to energy loss of the incident low energy deuterium, any primary reaction with an impurity is confined to well within the first 0.5 µm of the target. Therefore, reaction product protons with MeV energy would lose very little energy exiting the target (e.g., a 5 MeV proton loses <10 keV). Since only high energy protons can penetrate the Al absorber foil, only (d, p) reactions need to be considered. The light elements are most likely to react with the low energy deuterium since the Coulomb barrier is prohibitive for Ti and other heavy impurities. Those possible (d, p) reactions with light elements and with Q > 3 MeV are (with Q in MeV):  ${}^{6}\text{Li}(5.03), {}^{9}\text{Be}(4.59), {}^{10}\text{B}(9.23), {}^{13}\text{C}(5.95), {}^{14}\text{N}(8.61),$ <sup>17</sup>O(5.82), <sup>19</sup>F(4.38).

TiH<sub>2</sub> targets were bombarded with 170 keV deuterium so the large d-d primary reaction products that mask low level reactions with trace impurities would be absent. The spectrum in Fig. 7 shows the result of this experiment.<sup>3)</sup> The sharp 14.7 MeV proton peak in channel  $\sim$ 630 was initially a surprise. It is caused by direct reaction of deuterium elastically scattered from the target with <sup>3</sup>He that is implanted into the Al absorber foil covering the detector. The implanted <sup>3</sup>He concentration builds up over time from the  $\sim 0.8 \,\text{MeV}^{-3}$ He produced in the d-d reactions in the target. Use of a fresh Al absorber foil eliminates these counts. The (d, p) reactions with impurities listed above would appear as a sharp peak in the spectrum, and no peaks are observed. The origin of these counts is unknown, but those below  $\sim 8 \text{ MeV}$  could be caused by primary (d, p) reactions with multiple impurities.

We consider only the primary (d, p) reactions with impurities likely. The yields for the reaction of impurities with the d–d reaction products (e.g., secondary <sup>3</sup>He, p, n, t in TiD<sub>2</sub> targets) are deemed too weak to be observed. An indication of this is the fact that proton signal from the secondary (<sup>3</sup>He, d) reaction is weak, even though the concentration of deuterium in the target is high. The light impurities are 4 to 6 orders-of-magnitude less concentrated than Ti or deuterium in the targets, so the signal from secondary reactions with impurities would be correspondingly less. Secondary reactions of 3 MeV protons with impurities is experimentally examined in a later section.

There are a small number of counts in channels  $\sim$ 120–550 that constitutes a quantifiable background correction for the

spectra taken with a deuteron beam. This signal, that we define as a "impurity correction", is sufficiently small that it does not prevent the measurement of the anomalous protons at the yield measured by Kasagi *et al.* This correction

Fig. 7. Energy spectrum of charged particles emitted at an angle of  $135^{\circ}$  from a TiH<sub>2</sub> target bombarded by 170 keV deuterium ions. A 140  $\mu$ m thick

## 3.1.5 Natural backgrounds

depends on collected charge.

Al foil was placed in front of the detector.

A particle spectrum was collected for  $3.5 \times 10^5$  s for the conditions of no ion beam, and with the aperture and absorber in place to determine the contribution from the sum of natural radioactivity background and cosmic ray background. This produced 76 counts in the region of interest. From this data, the contribution of natural background to the spectra collected in these experiments was found to vary between 0 and 4 counts. This correction was too small for consideration and was ignored in the data analysis.

### 3.2 Measurement of anomalous proton emission

Figure 8 is one of four spectra accumulated for 170 keV deuterons on TiD<sub>2</sub> targets. It is for a 135° detector angle, a 140  $\mu$ m Al absorber, and 242 mC collected charge accumulated over 17777 s. Between channels 500–780 there is a sharp peak riding on a broad hump. The sharp peak is the 14.7 MeV proton from the primary (d, <sup>3</sup>He) reaction. As the experiment progresses, <sup>3</sup>He accumulates in the target as a product of the primary d, d reaction [reaction (1b) above]. The rate of accumulated charge. Kasagi *et al.* definitively determined that the broad hump is protons from the secondary (<sup>3</sup>He, d) reaction. The counts between channels 150–500 (3–10 MeV) are mostly anomalous counts.

### 3.2.1 Signal and noise

A summary of four runs on two different  $TiD_2$  targets bombarded with 170 keV deuterons is shown in Fig. 9 in bar-graph format. The bar height is the total number of counts in the anomalous region at lower energies than the 14.7 MeV protons and above the counts from Compton





Fig. 8. Energy spectrum of charged particles emitted at an angle of  $135^{\circ}$  from a TiD<sub>2</sub> target bombarded by 170 keV deuterium ions. A  $140 \,\mu\text{m}$  thick Al foil was placed in front of the detector.

scattering of gammas. The experiments are ordered chronologically from left to right, and the last bar corresponds to the data in Fig. 8. The three corrections ("neutrons", "detector", "impurities", §3.1.1, 3.1.3, 3.1.4, respectively) applied to the data as described above are shown with error bars. The first bar is data taken at a scattering angle of  $110^{\circ}$ and with no aperture on the detector. The second bar is data with the aperture, and also at a 110° scattering angle. The "detector correction" is reduced nearly a factor-of-10 by the aperture. The particle detector was replaced between the experiments for bars two and three since the energy resolution had noticeably degraded. This detector had experienced large fluxes of thermal neutrons in the early stages of the work that probably contributed to the degradation. The new detector improved the signal-to-noise in bars three and four as it allowed a larger channel width to sum the anomalous counts. Runs three and four were for a detector angle of 135°. All four experiments produced anomalous counts. The reproducibility of the signal is 100% for the statistical sampling of four runs.

### 3.2.2 Relative yield of anomalous proton emission

The measured yield of anomalous counts versus the target D/Ti ratio is plotted in Fig. 10 expressed as the yield of anomalous counts divided by the yield of 3 MeV protons from the primary d-d reaction [reaction (1a) above]. The four data points from this work are all for the same Ti/D ratio and are displaced on the abscissa for clarity. Also included is the data point from the work of Kasagi et al. They bombarded 100 different targets and state that the anomalous counts did not appear for D/Ti < 1.2, and that the maximum target ratio investigated was  $\sim 1.6$ . The target ratio is plotted as 1.4 with error bars over the range of target composition for which they observed anomalous counts. Kasagi noticed no angular dependence for measurements between the angle of 110 and  $155^{\circ}$ .<sup>1)</sup> The yield measured in this work is  $\sim 4 \times 10^{-5}$  of the d–d reaction cross section, or  $\sim$ 4 times larger than the yield measured by Kasagi *et al.* The yield of anomalous protons per deuteron in the target may increase about a factor-of-2 to  $\sim 1 \times 10^{-4}$  due to the



Fig. 9. (Color) Bar graph of the total number of counts in the channels between the low energy cut-off and the protons from secondary reactions ( $\sim$ 3–10.5 MeV; as in Fig. 8) for four different bombardments of TiD<sub>2</sub> with 170 keV deuterons. The scattering angle is shown on the bottom. Each bar is composed "neutrons", "detector", and "impurities" corrections (see text) and the anomalous counts. For bars 2, 3, and 4, a 1-mm-thick Pb aperture, 6-mm in diameter was placed over the detector. For bars 3 and 4, a new particle detector was used.



Fig. 10. The ratio of yield of anomalous counts to the yield of 3 MeV protons from the (p, t) branch of the d–d reaction versus the D/Ti ratio of the target. The data point at D/Ti of 1.4 is from ref. 1. The four data points at D/Ti = 1.97 are from this work.

possibility of uncounted proton yield underneath the 14.7 MeV proton peak and those protons that are slowed down in the 140  $\mu$ m Al absorber that do not penetrate to the detector.

### 4. Possible Artifacts

The experiments described above confirm the existent of anomalous energetic proton emission when titanium deuteride is bombarded with low energy deuterons. In this section, a number of artifacts are examined and discarded as a source of the anomalous emissions.

### 4.1 Artifacts eliminated by Kasagi et al.

In the work of Kasagi *et al.*, a number of possible explanations were examined and discarded.<sup>1)</sup> These were; products of the primary d–d reactions (1a) and (1b) reacting with target deuterons yielding reactions (2a) and (2c); products of the primary d–d reactions (1a) and (1b) reacting with known target impurities including Li and the Al

absorber, and knock-on of 14 MeV protons with alphas or deuterons. They also bombarded  $TiD_x$  samples directly with 3.3 MeV protons, 1.5 MeV <sup>3</sup>He, and 2.5 MeV neutrons and observed no alpha particle emission from 4.5 to 6.5 MeV. Similar tests for the anomalous protons were not performed. They also found that the ratio of anomalous proton yield to anomalous alpha yield was roughly constant with deuteron energy, which suggests the two particles are connected to the same process.

### 4.2 3-MeV secondary protons

As stated previously, there are a number of possible d, alpha reactions with trace amounts of light impurities. It is possible that combinations of concentrations and depths of impurities in the sample could conspire to produce the anomalous alpha particles. This is why we concentrated on the anomalous protons. It is appropriate to check for the interaction of the secondary products with trace impurities for the proton signal. Therefore, we bombarded a TiD<sub>2</sub> target with 3.3 and 6.0 MeV protons using our 3 MV Pelletron facility. The same detector was used in conjunction with the aperture, 140  $\mu$ m Al absorber, and an angle of 135°. There were no proton counts observed for a proton dose 20× larger than the total proton dose from secondary d, p reactions during 170 keV deuterium bombardment.

### 4.3 Collision cascade

The collision cascade could, in very unlikely events, arrange to drive three deuterons together in the solid. As a test of this notion, TiD<sub>2</sub> was bombarded with 170 keV <sup>4</sup>He and 170 keV protons with same 2-mm-thick detector, the Pb aperture, 140  $\mu$ m Al absorber, and an angle of 135°. If the collision cascade is involved, the cascade caused by <sup>4</sup>He is more violent than for deuterons and less violent for the protons. Only 10 counts were observed between 3 and 10 MeV for 105 mC collected charge for either beam. Therefore, if collision cascades are involved, it must be required that the beam nucleus be a reaction participant, and/or, the beam nucleus have the same mass as the other nuclei involved in the reaction.

### 4.4 Cracks in the targets

The deuteride targets had numerous cracks in the surface caused by the large volume expansion when Ti is charged with deuterium at high temperature and then cooled.<sup>3)</sup> If this crack area were 20 to 50% of the surface area, it could perhaps account for the anomalous protons. A fraction of the deuterons could penetrate deep into the target, react with a target deuteron to produce 14.7 MeV protons through the secondary reaction (2c). Energy loss through various thickness of target before emerging to be detected would produce the anomalous proton spectrum. Accordingly, an experiment was run where 170 keV <sup>3</sup>He impinged upon the sample at normal incidence, 30°, and 45°. The detector angle was 135°, the Pb aperture and a 140 µm Al foil protected the detector.

The spectra obtained were very similar to Figs. 6(a) and 6(b). The presence of the cracks was observed to slightly broaden the sharp 14.7 MeV proton peak on the low energy side to varying degrees with scattering and detector angle. The ratio of the summed detector fault counts in the lower

channels to the sum of the counts in the peak was statistically identical to the ratio from previous numerous measurements from deuterium bombardment of <sup>3</sup>He-implanted Ti. Therefore, the anomalous counts are not an artifact caused by cracks in the sample.

#### 4.5 RF discharges

Silicon particle detectors are quite efficient RF antennae due to the relatively large area and capacitance. Radio frequency (RF) discharges will readily induce ~mV pulses that are converted by the amplifier and multichannel analyzer (MCA) system into counts in a particle spectrum. While it is difficult to produce a false peak in a spectrum, RF discharges typically produce a continuous background over the complete energy range, and it is common to observe amplifier saturation pulses at high energy, if the MCA system is set to collect them. These pulses were observed and identified as such, for example, during the experiment in §4.2 above where 3.3 and 6 MeV protons bombarded TiD<sub>2</sub>. It was observed that high-voltage breakdown in the Pelletron accelerator tube transmitted RF to the detector and produced false counts in the spectrum. The possibility of RF-induced artifacts must be considered for the measurements at 170 keV using the ion implanter.

Several experiments were performed to attempt to induce RF in the experimental system. With the particle analysis system in the collection mode, high voltage breakdown of the accelerator and ion source extraction was artificially induced, and all accelerator components were abruptly turned on and off. No spurious counts appeared in the MCA. RF problems can arise from high voltage breakdown of an electrically insulated component in the target chamber that charges up from bombardment of secondary electrons. Care was taken to ensure there were no insulated parts. Furthermore, the same conditions of beam current, beam energy and chamber geometry were present during the bombardment by protons and <sup>4</sup>He in §4.3 above, and only 10 counts were observed. While RF discharge cannot be completely ruled out as the source of anomalous counts, these experimental tests make RF discharges very unlikely.

### 5. Discussion and Conclusions

It is concluded from this study that the emission of high energy protons from deuterium bombarded TiD<sub>2</sub> is a robust anomaly. The extensive search for possible artifacts both here and in ref. 1 have not revealed a plausible conventional explanation. The anomalous proton emission reported in this work is the broad energy signal of 3 to 10.5 MeV protons at scattering angles of 110 and 135° at a rate of  $\sim 4 \times 10^{-5}$  of the d–d fusion rate when TiD<sub>2</sub> is bombarded with 170 keV deuterons. It is 100% reproducible based on four successes in four tries. Expressed in terms of a nuclear cross section, the anomalous proton emission has a value of about 1 µbarn. Our value of the proton emission rate is 4× larger than that measured by Kasagi, perhaps due to the ~20% higher concentration of deuterons in our targets.

Kasagi *et al.* offer the exotic suggestion that the proton emission is the result d-d-d fusion, primarily because they also measured the companion <sup>4</sup>He emission that would accompany the proton for the three-body branch of the d–d–d reaction [reaction (3a) above]. However, the measurements were independent and not in coincidence. We prefer at this point to leave open the question of its origin of this anomaly, since the attribution of d–d–d fusion only raises more questions such as; why is only the three-body branch seen, and is there any corresponding p–d–d fusion for proton bombarded TiD<sub>2</sub> targets? We leave discussion of the source of the anomalous proton emission as d–d–d fusion until after definitive evidence is obtained for the its existence, since the effect could still be an artifact as yet uncovered. Such evidence would be provided by the detection of time coincidence between the proton and alpha particle emissions.

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- J. Kasagi, T. Ohtsuki, K. Ishii, and M. Hiraga: J. Phys. Soc. Jpn. 64 (1995) 777.
- A. Takahashi, K. Maruta, K. Ochiai, and H. Miyamaru: Phys. Lett. A 255 (1999) 89.
- K. S. Grabowski, G. K. Hubler, D. L. Knies, R. A. Walker, S. B. Qadri, T. S. Wang, and D. J. Nagel: J. Phys. Soc. Jpn. **71** (2002) 757.