

Naval Research Laboratory

Washington, DC 20375-5000

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AD-A243 718



NRL Memorandum Report 6927

**Evidence for MeV Particle Emission From Ti
Charged with Low Energy Deuterium Ions**

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December 18, 1991

91-19130



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91 1227 006

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE 1991 December 18	3. REPORT TYPE AND DATES COVERED Final	
4. TITLE AND SUBTITLE Evidence for MeV Particle Emission From Ti Charged with Low Energy Deuterium Ions			5. FUNDING NUMBERS 46-3765-01 ONR 63218C	
6. AUTHOR(S) George P. Chambers, Graham K. Hubler and Kenneth S. Grabowski				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Naval Research Laboratory 4555 Overlook Avenue Washington, DC 20375-5000			8. PERFORMING ORGANIZATION REPORT NUMBER NRL Memorandum Report 6927	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) ONR Alexandria, VA			10. SPONSORING / MONITORING AGENCY REPORT NUMBER	
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) Thin titanium films have been bombarded with low energy (350 eV) deuterium ions at high current density (0.2-0.4 mA.cm ⁻²) to investigate the reported occurrence of nuclear reactions at ambient temperatures in deuterium charged metals. A silicon charged particle detector was used to search for charged particles produced by such reactions. Evidence is reported for the detection of hydrogen isotopes with 5 MeV energy at a rate of 10 ⁻¹⁶ events/deuteron pair/s.				
14. SUBJECT TERMS Deuterium, Titanium, Ion Bombardment, nuclear reactions, charged particles			15. NUMBER OF PAGES 34	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL	

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EVIDENCE FOR MEV PARTICLE EMISSION FROM TI CHARGED WITH LOW ENERGY DEUTERIUM IONS

ABSTRACT: Thin titanium films have been bombarded with low energy (350 eV) deuterium ions at high current density (0.2-0.4 mA/cm²) to investigate the reported occurrence of nuclear reactions at ambient temperatures in deuterium charged metals. A silicon charged particle detector was used to search for charged particles produced by such reactions. Evidence is reported for the detection of hydrogen isotopes with ~5 MeV energy at a rate of 10⁻¹⁶ events/deuteron pair/s.

I. Introduction

In pioneering work by Jones et al. [1], a titanium electrode was charged with deuterium ions in an electrochemical cell by means of an electric current passing through an electrolyte containing various Li salts and D₂O. The experiment was monitored for neutron production which was found to occur at a rate of 1x10⁻²³ fusions/deuteron pair/sec. Jones et al. further observed short bursts of n emission during thermal cycling of Ti alloys charged with deuterium - hydrogen gas mixtures [2]. Our investigations were begun because we were intrigued by the plausibility of arguments by Jones et al. that low levels of fusion reactions could explain certain anomalies in geological and planetary data [1], and because we calculated that our deuterium ion beam charging apparatus could achieve a sensitivity of ~ 10⁻²³ fusions/deuteron-pair/s for charged particle detection, and would

therefore be able to detect extremely low levels of radiation. In a search for these reactions, long bombardment times and very low count rates were anticipated.

We have used the approach of employing low energy ion beams to charge metal lattices with deuterium in vacuum [3, 4], so that we can monitor the emission of energetic charged particle reaction products, such as helium or isotopes of hydrogen, that have extremely short ranges in air or water. Because ion beams can implant deuterium below the surface of a metal, the charging efficiency is roughly 75-80% after (reflection loss as computed by TRIM [5]) compared to the electrochemical charging efficiency at saturation of <30% [6]. Moreover, energetic ion implantation can create metastable, high nonequilibrium concentrations of deuterium within the metal lattice, thus allowing access to a larger range of deuterium -solid interaction parameters. Thin films have been used in this experiment as opposed to bulk metals because thin films can be saturated quickly and thoroughly by the ion beam and energetic charged particles can be detected because their range is much larger than the thickness of the films.

In this paper, evidence is presented for the generation of sustained high energy nuclear reactions produced as a result of low energy ion beam charging of titanium with deuterium at a surprising rate of $\sim 10^{-16}$ events/deuteron pair/s. This rate corresponds to a cross-section at least 26 orders of magnitude higher than the cross sections for conventional nuclear fusion [7] at the incident ion energies (350 eV) used in this experiment and higher than that postulated by Jones et al. for planetary fusion reactions by 7 orders-of-magnitude.

II. Experimental Description

The experimental arrangement is shown in Fig. 1. Low energy deuterium (350 eV) ions produced by an ECR microwave source impinge normally on a thin metal film in

vacuum, while a Si particle detector placed directly behind the film detects particle emission. The advantages of this method are rapid and efficient deuterium charging of any material (including insulators), high particle detection efficiency and sensitivity (low background), and the ability to measure the particle energy and determine the particle type.

The sample and particle detector were mounted in an ice-water-cooled Cu block as shown in Fig. 1. Titanium was chosen as the target because previous work by Jones had shown neutron emission and because Ti retains more hydrogen near room temperature than does Pd [8]. A thin teflon sleeve (0.64 mm wall thickness) electrically insulated the particle detector to eliminate ground loop noise, but was thin enough to provide detector cooling from the Cu block. Thin target foils were mounted on a 1-mm-thick Al washer with Duco cement. This was required because the deposited films contained high compressive stress which caused unsupported foils to curl up. The foils were electrically grounded and cooled through conduction from the contact to the Cu block around the perimeter of the foils. The hole in the front of the block was 9.5 mm in diameter and was beveled at a 30 degree angle to minimize sputtering of Cu onto the sample.

The deuterium ions were produced by a Microscience, Inc. electron cyclotron resonance (ECR) microwave (2.45 GHz) plasma source, typically operating at an input power of ~500 watts. The source was equipped with dual graphite extraction grids to allow acceleration of the ions up to 1000 Volts, although 350 V was most commonly used. These operating conditions produced a beam estimated to be 80% D⁺ and 20% D₂⁺ [9] and a flux of $\sim 5 \times 10^{15}$ deuterium atoms/cm²/sec at the sample. Ion beam current was monitored during experimental runs by an electrostatically suppressed Faraday cup attached to the Cu block, 2.5 cm below the sample position. The sample stage could be moved vertically in the chamber to measure the beam current at the actual sample position where the beam current was consistently 20% larger than in the monitoring position for typical source

operating conditions. A 1000 l/s turbo-molecular pump provided a base pressure of 5×10^{-8} Torr in the all metal seal vacuum chamber and the operating pressure was $1 - 2 \times 10^{-4}$ Torr of 99.5% purity Matheson D_2 gas as measured by an ionization gauge in the target chamber. Actual H pressure was a factor of 2.4 greater taking into account the ionization gauge factor between nitrogen and hydrogen. At these pressures, 40-60% of the beam was charge-exchange neutralized while traversing the 25 cm drift distance between the ion source extraction grids and the Faraday cup. These corrections for Faraday cup position, charge-exchange neutralized fraction, reflected beam and molecular species in the beam have been applied to the beam currents quoted in Table I and the text. The projected range (straggling) of 175 and 350 eV deuterium in Ti are 47 Å (26 Å) and 78 Å (39 Å), respectively, from TRIM calculations.

The detector was a Ruggedizedtm Ortec silicon charged particle detector [10], with a depletion depth of 500 μm and an active area of 50 mm², the surface of which was mounted four mm from the back side of the sample. The detector was calibrated using 5.486 MeV alpha particles from an ²⁴¹Am radioactive source in conjunction with a precision pulse generator. The intrinsic efficiency of particle detection for this detector was 100%; the geometric efficiency of the particle detection system was $10 \pm 2\%$. Pulse shapes were continuously monitored with a storage oscilloscope during ion bombardment. Due to the operation of the ECR source, axial magnetic fields on the order of 100 gauss, microwave radiation on the order of 10mW/cm², and uv radiation of unknown but substantial intensity were present at the sample during bombardment.

Considerable effort was expended in minimizing noise in the system and characterizing those sources that could not readily be eliminated. The particle detector ground was electrically insulated from vacuum chamber at the detector cable vacuum feed-through and the preamplifier case was isolated from ground. An isolation transformer and

surge protector powered the detection system electronics. The coaxial signal cable was shielded where possible with aluminum foil. Without the ion source turned on, there was a background signal from cosmic ray secondary particles which yielded 25.3 ± 0.26 counts/h < 1 MeV, 1.13 ± 0.05 counts/h between 1 - 3 MeV and 0.29 ± 0.03 counts/h > 3 MeV.

With the ion source operating there were two noise sources; one of which generated white noise in the low energy region of the spectrum (< 1.5 MeV) due to reflected ions impinging on unprotected regions of the detector cable and to gradual detector warming during the course of the experiment. This noise produced broadening of all radiation peaks acquired during operation of the source. The second noise source was identified as arcing, associated with voltage breakdown of the ion accelerating grids [11]. It increased in frequency with increasing voltage and was found to be capable of producing isolated pulses anywhere in the spectrum. This noise, however, appeared at a low, well-defined rate and did not produce the well resolved spectral peaks typical of charged particle events. The signal pulses produced by both of these sources, as monitored on the storage oscilloscope, were observed to be highly anomalous in shape and therefore easily recognizable; in the case of beam induced white noise the pulses occurred randomly and were oscillatory with positive/negative excursions around zero and of fluctuating height, while pulses due to arcing exhibited a pronounced negative excursion. By contrast, pulses produced by the passage of energetic charged particles through the detector were Gaussian in shape.

Thin films were prepared by electron beam evaporation in a separate chamber. Typical base pressure was 5×10^{-7} Torr which improved during evaporation as a result of Ti getter pumping. High purity Ti films were deposited onto $3.8 \mu\text{m}$ -thick nickel foils at an average rate of about $60 \text{ \AA}/\text{s}$. After deposition, foils were mounted *tautly* onto an aluminum washer and were then moved immediately to the ECR bombardment chamber.

Table I describes the samples used in these experiments. Some films had 0.5 μm Au deposited prior to deposition of Ti to provide a barrier to deuterium out-diffusion on the back side of the sample. Room temperature diffusion coefficients for D in Ti ($\sim 10^{-13}$ cm^2/s) [12] indicate that there is sufficient mobility at 0 degrees C for deuterium implanted into the surface to diffuse through the 1 μm thick films within 300 seconds.

III. Experimental Results

Three experiments which yielded positive results are described in detail below. Figures 2a and 2b show spectra from two experiments in which sustained particle emission was observed. These spectra were produced during low energy (350 eV) deuterium ion bombardment of samples 1 and 2, respectively, in Table I. The spectrum in Fig. 2a was obtained after bombardment at a current density of 0.27 mA/cm^2 for 25 minutes followed by bombardment at 0.38 mA/cm^2 for fifteen minutes. At this time, counts appeared in multiple bursts over a two to three minute period. No additional counts were observed even though bombardment was continued for two additional hours. A total of 1171 counts were acquired in a peak centered at an energy of 4.99 MeV. (X-ray diffraction to search for TiD on this sample was inconclusive due to the large interference from Au.)

The spectrum shown in Fig 2b was obtained after bombardment of sample 2 for slightly over five minutes at a current density of 0.38 mA/cm^2 at which time counts began to appear in a sustained continuous manner lasting approximately five minutes. During acquisition of this spectrum, which consists of 8035 counts, the detector bias was purposely turned to zero, held there for several minutes, and then turned back to -200 volts. Zeroing the detector bias has the effect of reducing the active depletion region from 500 μm to ~ 20 μm , in effect simulating a particle telescope. When the bias voltage was at -200 volts, counts were observed to appear at the higher energy peak, centered at 5.08

MeV; when the bias was at zero voltage, counts were observed to appear at the lower energy peak, centered at 3.5 MeV. This response to the reduction of detector bias voltage is characteristic of the that expected for the impingement of energetic charged particles on the detector when their range is greater than the depletion depth at the reduced bias voltage. In other words, incomplete charge collection occurred. Therefore, the behavior described above clearly establishes that the signal is due to a charge collection process occurring within the detector. Continued bombardment for a period of one hour produced no further evidence of reactions occurring in the film. X-ray diffraction analysis performed subsequent to bombardment showed that the film was predominantly titanium.

In order to provide additional information on the particle emissions, a 11.3-mg/cm² thick Ni foil, which partially covered the open area of the Al washer, was cemented to the backside of the washer (1 mm behind the Ni foil). This additional foil left an open region accounting for ~1/5 of the planar area of the 0.94 mm ID washer. Fig. 3 shows a ~60 count spectrum for sample 7 which was collected twenty minutes after initiation of bombardment with a 0.4 mA/cm² deuterium ion beam. The twin peaks exhibited in this spectrum appeared concurrently during a several second long burst. The ratio of the counts in the lower energy peak (~2.5 MeV) to the counts in the higher energy peak (~5.7 MeV) is approximately 2/1, consistent with particles originating from a volume in the sample directly over the area unshielded by the back Ni foil, given the angles and geometry involved. Particles passing through the open region showed no shift, while particles passing through the 11.3 mg/cm² foil were shifted to lower energy. This behavior clearly establishes that the counts are produced by MeV- energy charged particles which originate external to the detector.

In one other significant result (sample 12), a peak centered at 3.8 MeV appeared during a 1 minute period after 25 minutes charging with a beam current of 400 μ A/cm².

The peak had 25 total counts. An additional 11.3 mg/cm² Ni foil covered all the area behind the sample so the total thickness of Ni traversed was ~16.5 μm. Therefore, the minimum starting energy required to deposit an energy of 3.8 MeV in the detector from p, d, t, or ³He particles would be ~ 4.6 MeV, 5.0 MeV, 5.3 MeV, and 8.7 MeV, respectively, assuming normal incidence on the detector. Tritons produced at ~>5.3 MeV are consistent with results in figures 2 and 3.

Control experiments were performed using H⁺ bombardment of metallic films under nearly identical experimental conditions. No events of a similar nature were observed during over ten hours of operation. As an example, Fig. 4 exhibits a spectrum acquired during a four hour period by bombarding a 7-μm thick TiD₂ film, prepared by ion beam assisted deposition, mounted on a 3.8 μm thick nickel foil, using hydrogen ions at 500 eV. No spectral peaks can be seen in this spectrum. Pulses in the lower energy region of the spectrum are due primarily to white noise while higher energy pulses are due to arcing which was substantial at the higher grid voltage of 500 volts. Arcing was much less frequent at the lower grid voltage of 350 volts, which was used during the course of the deuterium runs.

As is shown in Table I, of the 13 samples run during the course of the experiment, four samples yielded positive results, four of the films delaminated at an unknown time during the run and were destroyed, while five remained intact and produced no result. No films which delaminated during bombardment produced a positive result. If all the implanted D remained in the samples, the amount of D loading at the time of reaction onset was ~ 55%, 8%, 30% and 45% of that required to convert all of the samples to TiD₂ for samples 1, 2, 7 and 12, respectively.

IV. Analysis and Particle Identification

Both the bias shift experiment and the partial foil experiment provide information on the particle identity. For analysis of the bias shift data, 5.1-MeV ^3He , ^4He and 5.0-MeV protons and deuterons produced in a tandem Van DeGraaff accelerator were scattered by a foil at a 2 degree forward angle and into the same detector at normal incidence and at 30 and 60 degree angles to the detector normal. This provided a calibration of the shifts expected for different particles with energies close to the unknown particle energy. Table II summarizes the data. The deposited energy shifts registered with the bias on and off for both helium isotopes were in each case smaller than the observed shift which eliminates helium as the particle. The energy deposited in the detector for protons travelling at various angles with respect to the detector normal are well fitted by an inverse $\cos \theta$ distribution, as is expected since the path length through the active region scales accordingly. Based on this relationship, the energy shift measured for 5.0-MeV deuterons indicate that deuterons travelling anisotropically at an angle of 30° to the detector normal could have produced the shift observed in figure 2b. While not measured directly, comparison of particle ranges in silicon suggest that tritons at or near normal incidence can account for the observed energy deposited, suggesting the triton as the most likely candidate to explain this result. This is illustrated by Fig. 5, which plots the energy deposited in the unbiased detector by normally incident particles against the average particle stopping power in silicon. A least-squares fit of the four data points to an exponential function shows that the stopping power of a triton in Si intersects along this curve with the energy deposited by the unknown particle in our experiment.

The energy shift observed in the partial foil experiment is also consistent with a hydrogen isotope. Based on the stopping powers given by Ziegler et al. [13], the energy loss through a foil of this thickness is too small to be accounted for by He^3 , but too large to be accounted for by a proton or deuteron. A triton, however, travelling at an angle ~ 60 degrees with respect to the surface normal could account for this result. As the ratio of the peaks suggests, the particles likely originated directly over the open region of the foil. This is not inconsistent with an energy shift for particles travelling at a high angle while particles at more normal angles would miss the extra foil and not experience an energy loss. Of course, the particles could have been produced in multiple active regions within the sample.

In Figs. 2 and 3, the spectrum is cut-off at energies ≤ 1 MeV by the multichannel analyzer (MCA) discriminator to eliminate white noise signals. The width of the particle peaks is considerably less than this white noise width. This occurred because the MCA was on during the entire experiment, and the microwave source would sometimes drift out of resonance alignment which produced a maximum noise signal represented by the cut-off. During appearance of the particle signals, the source was in relatively stable conditions so that the noise width was less than the maximum. This prevented examination of the spectrum at energies < 1 MeV for low energy particles. Information about the particle type cannot be extracted from the peak broadening due to energy loss straggling contributions to the width of the peaks because of the additional broadening caused by noise which is variable and much greater than the predicted straggling width for proton or helium isotopes. However, the width of the spectral peak exhibited in Fig. 2 is small enough to be accounted for by an isotropically produced MeV energy hydrogen isotope, taking into account the stopping powers of isotopic hydrogen ions through titanium, gold and nickel.

It is significant that the spectra presented above are shaped correctly in both the full and zero bias voltage cases. From Fig. 2a, it can be seen that a distinct "tail" (extra counts on the low energy side of the peak) is exhibited in the spectrum, as is expected due to energy straggling of energetic particles traversing the foil. In the zero bias case, however, an extra density of counts was observed on the high energy side, consistent with the effects expected when the range of the particle exceeds the active depletion depth. This situation is described on page 10 of the EG&G - ORTEC manual for silicon charge particle detectors [14] where it states that when "the range of the ionizing particles exceeds the sensitive depth of the detector...will produce an extra density of counts per channel on the high energy side of the spectrum." Examination of Fig. 2b reveals that the spectrum acquired at zero bias voltage is distorted in precisely this way.

This point is illustrated in figure 6, which shows spectra taken from an ^{241}Am source passing through a 1.9- μm Ni foil taken at intrinsic bias (power supply off) and full bias. This spectral peak at 4.7 MeV, taken at full bias, exhibits a distinct "tail" region, as is similarly exhibited by the spectrum in Fig. 2a. However, the spectrum acquired at intrinsic bias, peaked at an energy of 2.3 MeV, is distorted so that the leading edge is sharp, while the high energy side has an extra density of counts, similar in shape to the spectral peak exhibited in Fig. 2b.

Further, pulses that generated the spectral peaks described above appeared on our storage oscilloscope to be identical in shape to pulses produced by the ^{241}Am source. This is a clear and direct indication of normal charge collection processes occurring in the Si detector due to the stopping of energetic particles.

Assuming isotropic particle emission and uniform particle production over the entire area of the film exposed to the ion beam, the reaction rate required to produce the counts observed in Fig. 2b, averaged over the time in which the reaction was active, was $\sim 5 \times 10^6$

16 events/deuteron pair/s. A lower limit on the cross-section for this reaction is similarly found to be $\sim 5 \times 10^{-5}$ barns, a value 26 orders of magnitude higher than the cross-section for conventional $d-d$ fusion reactions at the incident ion energies used in this experiment (13). However, if instead the particles are originating from small active volumes within the film, as is suggested by the rapid reaction onset and cessation, then the reaction cross-section could be higher by many orders of magnitude.

V. Investigation of Experimental Artifacts

After the result for sample 1 was obtained, a concerted effort was made to devise additional artifact recognition procedures during the experiment. Accordingly, during the accumulation of counts from sample 2, the following tests were performed. In addition to changing the bias on the detector for the purpose of particle identification, other parameters were changed while the bias was off. The grid voltage of the ECR source was turned from 350V to zero volts, left at zero for several seconds, and returned to 350V. This action reduced the beam current to zero while the voltage was off. There was no observable effect on the rate of the particle pulses or pulse shape and amplitude as monitored on the oscilloscope. This suggests that the acceleration voltage and electronics were not causing spurious signals, and that the detected particles were not directly driven by the ion beam, since particles continued to be counted even when the acceleration voltage was turned off for a few seconds. The microwave power level was then turned from 500W to 150W, held there for several seconds, and returned to 500W. There also was no observable effect on the rate, height or shape of the incident particle pulses. This suggests that electromagnetic interference from the microwave flux did not causing the signal.

Other potential artifacts were investigated by independent tests on the apparatus.

With the ion source on, $\leq 2\text{mW/cm}^2$ of microwave power exits from the sample's vacuum chamber through a 5 inch diameter viewport at right angles to the ion beam. At the sample position, the microwave flux could be considerably higher. Therefore, it was imperative to further examine possible artifacts associated with the microwave radiation on the detector and electronics. From the front, the sample prevented the microwave radiation from penetrating to the detector, while the diameter of the detector housing aperture (1.8 cm) was too small to allow penetration of 2.45 GHz microwaves to the rear of detector which was recessed 3 cm into the hole. Moreover, extensive tests where the detector, cable, preamp and electronics were directly irradiated with 10 mW/cm^2 microwave power produced by a commercial microwave generator at 2.45 GHz produced no observable signal on our oscilloscope or MCA. This included direct irradiation of the detector face, and a portion of the center conductor of the coaxial cable where the shield was deliberately stripped to expose it. Attempts to generate the observed spectral peaks artificially by adjustment of the ECR gun parameters while running deuterium onto uncoated Ni foils over a ten hour period produced no counts over the background signals.

Pulses caused by electronic system noise are unaffected by a change in detector bias voltage while white noise originating within the detector is known to increase, not decrease, in amplitude as a result of decreasing the bias voltage. Deliberate generation of ground loop noise sources did produce resolved peaks, albeit with anomalous pulse shapes, but these peaks were always observed to shift to higher, not lower, energy when the bias voltage was reduced to zero.

Many other potential artifacts were investigated. Noise sources such as microplasma breakdown in the detector or in the gas in the volume between the foil and the detector would be expected to disappear when the detector voltage was removed. Attempts to induce microplasma breakdown by deliberately over-biasing a detector produced

anomalous square shaped pulses at high voltage, easily distinguishable from real particle pulses. Microplasma breakdown was deliberately induced in a different but similar specification detector by backfilling the vacuum chamber with D₂ to mTorr pressure. The signals produced had square shaped pulses and appeared in the MCA at energies <1 MeV. Charge sensitive preamplifiers experience oscillation if the impedance match to the detector is incorrect. Attempts were made to cause the preamplifier to oscillate by connecting very long cables between it and the detector. The preamp remained stable with up to 50 feet of cable, and the only effect was to increase the noise width as the cable length increased. In addition, the overload protection network in the preamplifier was in place during these experiments, which ensures more stable operation of the preamplifier.

The ion beam typically delivers a power of less than a quarter Watt to our samples. To ensure that the ion beam did not impinge directly onto the detector through ruptures in the foils during the experiment, samples were checked both prior to, and after, bombardment for pin-holes by holding them up to the light, as well as by examination in a back-lit optical microscope. No pin-holes or ruptures of any kind were found. Moreover, if our signals had been due to the beam impinging on the detector through a foil rupture, they would be expected to disappear when the beam was turned off. Lastly, deliberate impingement of an ion beam on a detector through a pin hole aperture produced noise pulses in the low energy region of the spectra *only*, inconsistent with our signals which were observed at much higher energy.

Microphonic noise from preamplifier or detector vibration can produce well shaped pulses, but not at a well defined energy. The chamber was also very vibration-free. Noise generated on the system ground by other equipment in the laboratory was checked both with and without the ECR source running and none was found. Contamination from a radioactive source is unlikely because of the sporadic nature of the count rate, the fact that

high count rate signals have been observed four times, and as described above, the particle is not helium, thus ruling out ^{241}Am contamination. Noise from photons is ruled out because the detector is coated with Al and can be operated in bright light and in air. Electrons and gamma-rays cannot deposit enough energy in the sensitive volume of the detector to give the high energy peaks observed. Cosmic rays are known to have temporally periodic and sporadic fluctuations in intensity. Records of cosmic ray activity and solar flare activity were examined for the dates and times of our positive results, and no correlations were found - rates corresponding to those in Fig 2 would be extremely unlikely.

Lastly, the detector behaved normally both before and after each of the runs described above. The detector leakage current was typically $0.3\ \mu\text{A}$ but occasionally rose to $\sim 1.5\ \mu\text{A}$ during a long (several hours) run and recovered after sitting in vacuum or air after several hours. This variation can affect the depletion depth (50% reduction at $1\ \mu\text{A}$ leakage) but the leakage was always low at the early time into runs when particles were observed.

VI. Discussion

Given that the pulses producing the spectral peaks describe above were well shaped, that the spectral peak shifted to lower energy in response to a decrease of the active depletion depth, that a distinct energy shift was observed in the partial-foil experiment, and that the spectral peaks are shaped correctly in both the full and zero bias voltage cases, we are led to conclude that we have detected MeV-energy charged particles emanating from our films as a result of low-energy deuterium bombardment.

It is important to point out that these energetic particles are not the result of direct collisions of 350 eV D^+ ions with D atoms at the surface of the target. If this were the case, normal nuclear products from d-d fusion would be observed at the appropriate energies, and the rate would scale with the impinging ion current density. The ion beam merely serves as a convenient and controlled means to obtain supersaturated solid solutions of deuterium within the metal lattice and to induce diffusion by means of high concentration gradients near the surface. Rather, the particles appear to be a consequence of the fact that high non-equilibrium concentrations of D exist within the solid environment. Also, nearly all measurements of the D-D cross-section have been performed using gas targets. The energetic charged particles observed here would not be expected in those measurements if the solid state environment is in fact involved in the reaction mechanism.

To account for the production of >5 MeV tritons by conventional d-d fusion, kinematic calculations indicate that a deuteron would have to be accelerated to >4 MeV. There is also a small probability that the particles are >5 MeV deuterons. In either case, the particles are clearly produced externally to the particle detector, originating either in the ion source, downstream from the ion source, or in the target or target holder. At the target and target holder, the deuterium charging will cause swelling and cracking of the irradiated material. Cracking or localized delamination of the target may cause ionization of the deuterium and acceleration [15], but it is difficult to justify acceleration voltages of MV by this mechanism. Jones et al. has postulated that piezo-fusion could occur in mineral rich brines or rocks under high temperature and pressure in the earth's mantle. A piezoelectric effect could conceivably produce greater voltages than cracking in a metal, but megavolt potentials are still unlikely.

We therefore conclude that the process appears to be due to a nuclear reaction other than conventional *d-d* fusion. Analysis of the spectra presented in figures 2 and 3 provides some information on the identity of this reaction. The small energy width of the spectral lines suggest that the reaction which produced them generates only two products in the exit channel, since three or more reaction products would be expected to produce a broader energy spectrum. Further, the bias shift and foil shift result imply that one of these particles is likely a triton. TRIM calculations indicate that a 5.38 MeV triton originating at the Ti/Au interface and travelling at normal incidence to the detector could account for the energy of the spectral peak (4.99 MeV) in Fig 2a. Alternatively, a 6.0 MeV triton originating at the front surface of the Ti film and travelling to the detector at the maximum geometrically possible angle of 65 degrees, could also account for this peak energy. Thus, the initial triton kinetic energy should fall between these two limits, depending on the exact region of origin and angle of passage through the foil to the detector.

It is conceivable that the spectra presented above could be accounted for by a deuterium pick-up reaction with a thermal neutron ($Q=6.25$ MeV) producing an energetic triton, provided lattice atoms or impurities either singly or collectively were somehow available to carry away recoil momentum. This reaction has the advantage that there is no Coulomb barrier to overcome. However, the source of neutrons for such a reaction is not apparent. It is of interest to note that low rates of unidentified 5- MeV charged particles have also been observed by Cecil et al. [4] in a deuterated palladium system, while low rates of MeV-energy particles have been observed by Taniguchi et al. [16] in an electrochemically charged Pd foil.

It is possible that these reactions may be originating from a small active region, perhaps a single grain or grain boundary. In this case, when the active region is sufficiently

charged by the beam, reactions are initiated. Reactions would cease when the active region is sputtered away by the beam (the surface of the films is continually eroded by the beam during the ion bombardment process) or when an irreversible change in the microstructure occurs due to hydrogen loading or stress relief. Such a scenario could account for the onset and cessation of the reactions, and for the observation that once the reaction terminates it cannot be reinitiated. Low reproducibility could be caused by subtle differences in deposition conditions of the films, variations in the state of stress during deposition or mounting, or sample temperature excursions during bombardment. Thermal flow calculations indicate that the rise in sample temperature should be less than 15 C for the experimental conditions used. However, at the time these experiments were performed, the contact between the sample and the Cu-block was maintained only by a press fit. It is possible that the contact was not maintained in some of the runs in which case the sample temperature could have been higher from heating caused by the ion, microwave, and uv fluxes on the sample.

The high particle production rate observed in these experiments was obtained using an ECR microwave ion source. Similar experiments have been performed using a Kaufmann ion source in place of the ECR ion source. High particle production rates similar to those shown in Figs. 2 and 3 have so far not been observed. This suggests that an as yet unidentified condition or set of conditions present in the environment created by the ECR ion source may be important for the high rate of particle production. In addition, except for the multichannel analyzer employed, the same detector electronics and detector was used in the follow-up experiments, which also implies that the high rate results are not artifacts associated with the detector or electronics.

VII. Summary and Conclusions

Strong evidence that nuclear reactions can occur at high rates, 10^{-16} events/deuteron pair/sec, within deuterium-charged Ti thin films was presented. The method used was low energy (350 eV), high current density (0.2-0.4 mA/cm²) deuterium ion beam charging of evaporated titanium thin film targets while monitoring with a silicon detector to measure charged particle production. These particles have been characterized as ~5 MeV isotopic hydrogen in at least two experiments, with the triton indicated as the most likely candidate. It was concluded that 1) conventional d-d fusion is not responsible for the particle production, 2) the reactions are not produced in collisions of 350 eV deuterium with the target ions, 3) the reactions occur as a consequence of high concentrations of deuterium in the Ti films, and 4) the rate of reactions is 7 orders-of-magnitude larger than that measured by Jones et al. and which are needed to explain anomalies in geophysical data.

According to the laws of conventional nuclear physics, nuclear reactions cannot occur at such large rates for sub-keV energy deuteron bombardment; yet we are unable to uncover a plausible artifact that could have produced the observed signals. As such, we believe that the data is very provocative and that the effect warrants investigation by other researchers.

Moreover, the experimental technique of low energy bombardment of thin samples in a transmission geometry is demonstrated to be a simple and sensitive method to search for low level fusion reactions that could explain anomalies in geophysical data. It has the advantage that any material, including insulators, may be charged with deuterium to high concentrations. A sensitivity of $\sim 10^{-23}$ events/deuteron pair/s can be achieved for 25- μ m-thick samples in charging periods of 1 to 10 hours, depending on the ion current. By simple changes in the detection system to separate the detector from the target holder, the

sample could be mounted on a temperature controlled holder with little loss of detection efficiency.

We would like to acknowledge the people who assisted in this work, including Al Knudson, Jim Sprague, Bill Coleman and Tom Hargraves. Discussions with Steve Jones, Mike Haftel, Vince Folen, and Jim Eridon were very helpful. We also thank Ed Cecil, Gary Herling, Jim Mayer, Rich Petrasso and Dan Mumick for critical reading of the manuscript.

REFERENCES

1. S. E. Jones, *et al.*, *Nature* 338, 737-40 (1989).
2. S. E. Jones, F. Scarramuzzi, and D. H. Worledge, *Proceedings of Anomalous Nuclear Effects in Deuterium/Solid Systems Workshop 22-24 Oct 1990, Provo UT* (American Institute of Physics, NY, no. 228)
3. G. P. Chambers, J. Eridon, K. S. Grabowski, *Phys. Rev. B* 41, (1990).
4. F. Cecil, *et al.*, in *Workshop on Cold Fusion Phenomena* R. A. Krakowski, Eds. (*Journal of Fusion Energy*, Sante Fe, New Mexico, 1989),
5. J. Ziegler, J. Biersack, G. Como, "The Stopping and Range of Ions in Matter," TRIM-89
6. B.Dandapani, M. Fleischmann, *J. Electroanal. Chem.* 39, 323 (1972).
7. R. E. Brown, N. Jarmie, *Phys. Rev. C* 41, 1391 (1990).
8. W. Moller, F. Besenbacher, and J. Bottinger, *Appl. Phys. A* 27, 19 (1982)
9. O. Popov, Microscience, Inc. *private communication* (1990).

10. This detector has a Schottky barrier junction on the rear surface protected by a thick organic sealant, and an 1850 Å-thick Al coating on the front surface. Standard Schottky barrier detectors short out within ~1 hour in the hydrogen pressure used in these experiments. A non-Schottky barrier detector of proper thickness for use as a particle telescope is not yet commercially available.
11. Our early version of this ion source had a defect in the accelerator grid design.
12. G.R. Caskey, in *Hydrogen in Metals* , I.M. Bernstein, ed. American Society for Metals, (1974) p. 465
13. J. Ziegler, J. Biersack, and U. Littmark, *The Stopping and Range of Ions in Matter* (Pergamon Press, New York).
14. EG&G Ortec, *Silicon Charged Particle Radiation Detectors Instruction Manual* , p. 10
15. J. T. Dickenson, L. C. Jensen, S. C. Langford, R. R. Ryan, E. Garcia, *J. Mater. Res.* 5, 109 (1990).
16. R. Taniguchi, T. Yamamoto, S. Irie, *Jap. Jour. Appl. Phys.* 28, 198 (1989).

TABLE I Sample compositions, ion beam conditions, dates of runs, and results are tabulated for deuterium runs with the ECR source. NR indicates that no positive result was obtained. Beam energies are in eV while current densities are in mA/cm².

Run #	Date	Sample	E, I	Result	Final Sample Condition
1	12/14/89	Ti/Au/Ni	350, 380	Peak at 5 MeV after 40 min 1171 cnts	Intact
2	12/17/89	Ti/Au/Ni	350, 300	Peak at 5 MeV after five min Bias zeroed Peak at 3.5 MeV 8035 cnts	Intact local blistering on surface
3	12/19/89	Ti/Ni	350, 400	NR	Intact
4	12/21/89	Ti/Au/Ni	350, 320	NR	Delaminated
5	12/23/89	Ti/Au/Ni	350, 400	NR	Delaminated
6	12/24/89	Ti/Au/Ni	350, 400	NR	Delaminated
7	1/16/90	Ti/Ni Partial Ni Foil behind sample	350, 300	5.7MeV, 2.5 MeV after 20 min 60 Total cnts	Intact
8	1/28/90	Ti/Ni	350, 400	NR	Delaminated
9	2/24/90	Ti/Ni	350, 400	NR	Intact
10	2/25/90	Ti/Ni	350, 300	NR	Intact
11	2/26/90	Ti/Ni	350, 400	NR	Intact
12	2/27/90	Ti/Ni Additional Ni foil behind sample	350, 300	Peak at 3.8 MeV after 25 min 25 total counts	Intact
13	3/2/90	Ti/Ni	350, 400	NR	Intact

Table II - 5.1-MeV ^3He , ^4He and 5.0-MeV protons and deuterons produced in a tandem Van DeGraaff-type accelerator were scattered by a foil at a 2 degree forward angle and into our detector. Energies deposited in the detector for the bias voltage on and off for both helium isotopes, deuterons, and protons at several incident angles are compared to the observed energy registered at full and zero bias by the particles detected in our experiment. Energies are presented in units of MeV.

Particle	Energy (MeV)	Incident Angle (degrees)	Energy Deposited at Zero Bias (MeV)
Unknown	5.08	Unknown	3.5
^4He	5.1	0	4.30
^3He	5.1	0	4.10
Deuteron	5.0	0	2.95
Proton	5.0	0	1.84
Proton	5.0	45	2.53
Proton	5.0	60	3.23

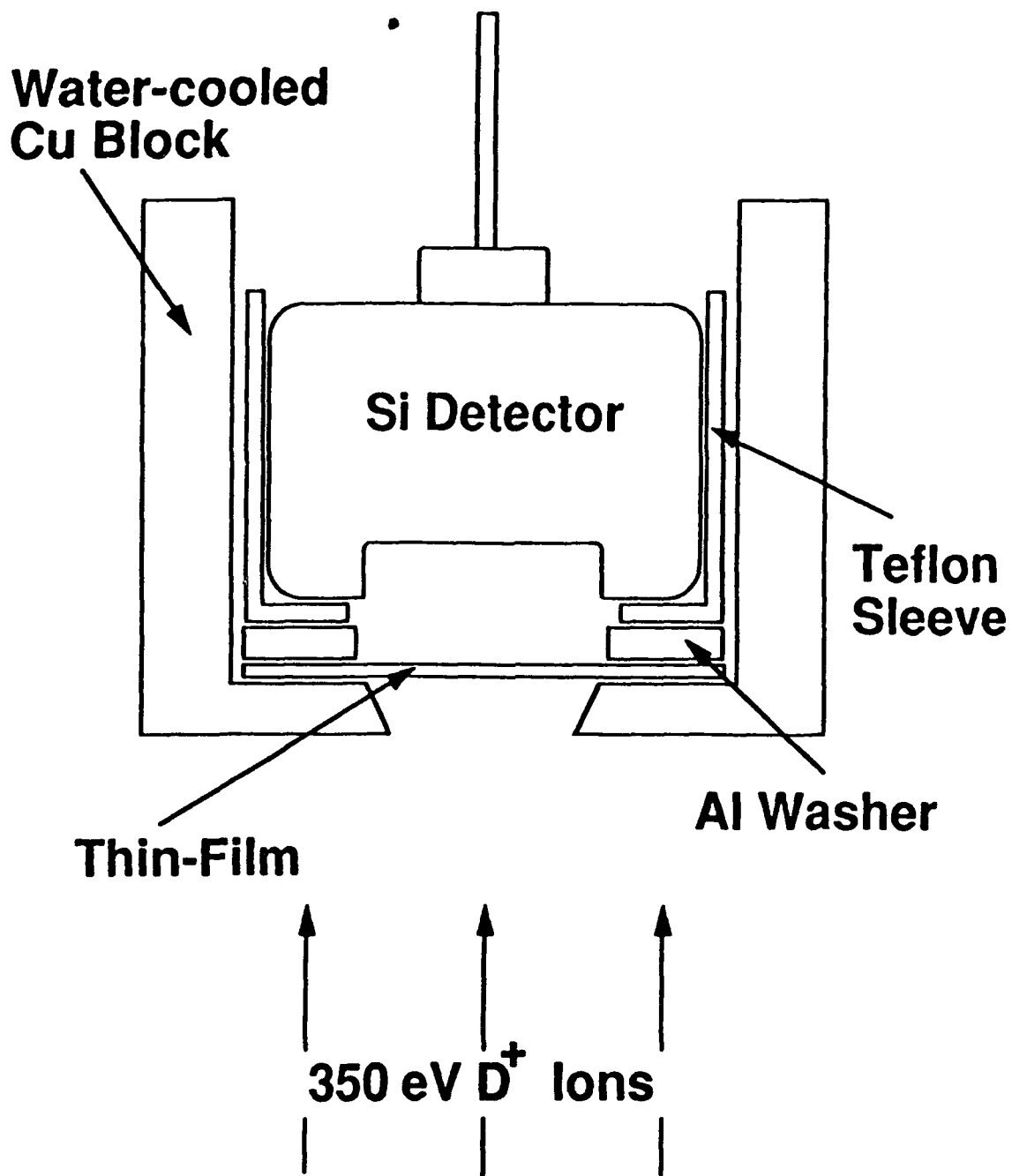


Figure 1. Schematic of transmission geometry used in this experiment. Samples were mounted in a water cooled copper block with an aperture to allow bombardment by the ion beam generated by an ECR plasma source. A silicon detector was mounted ~ 4 mm behind the sample to detect energetic charged particles.

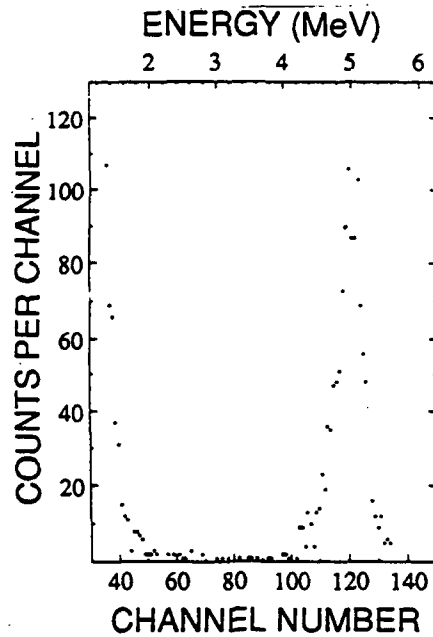


Figure 2(a). Spectra acquired after 40 minutes of 350 eV deuterium bombardment of a 1- μm thick titanium film evaporated onto 500 nm gold on a 3.8 μm thick nickel foil. The spectral peak, centered at 5 MeV, occurred during two approximately minute long bursts and consists of over 1100 counts.

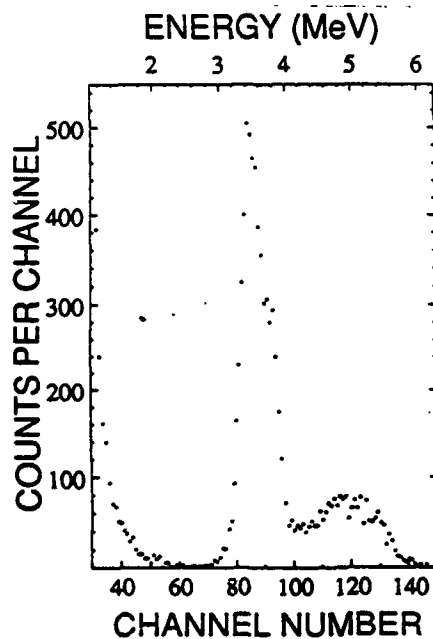


Figure 2(b). Spectra acquired after 5 minutes of 350 eV deuterium bombardment of a 1- μm thick titanium film evaporated onto a 3.8 μm thick nickel foil. These spectral peaks were acquired during a five minute long burst. The double peaks are due to the changing of detector bias voltage from -200 volts to zero and back to -200 volts again during the time the signal was present. There are 8035 counts in this spectrum.

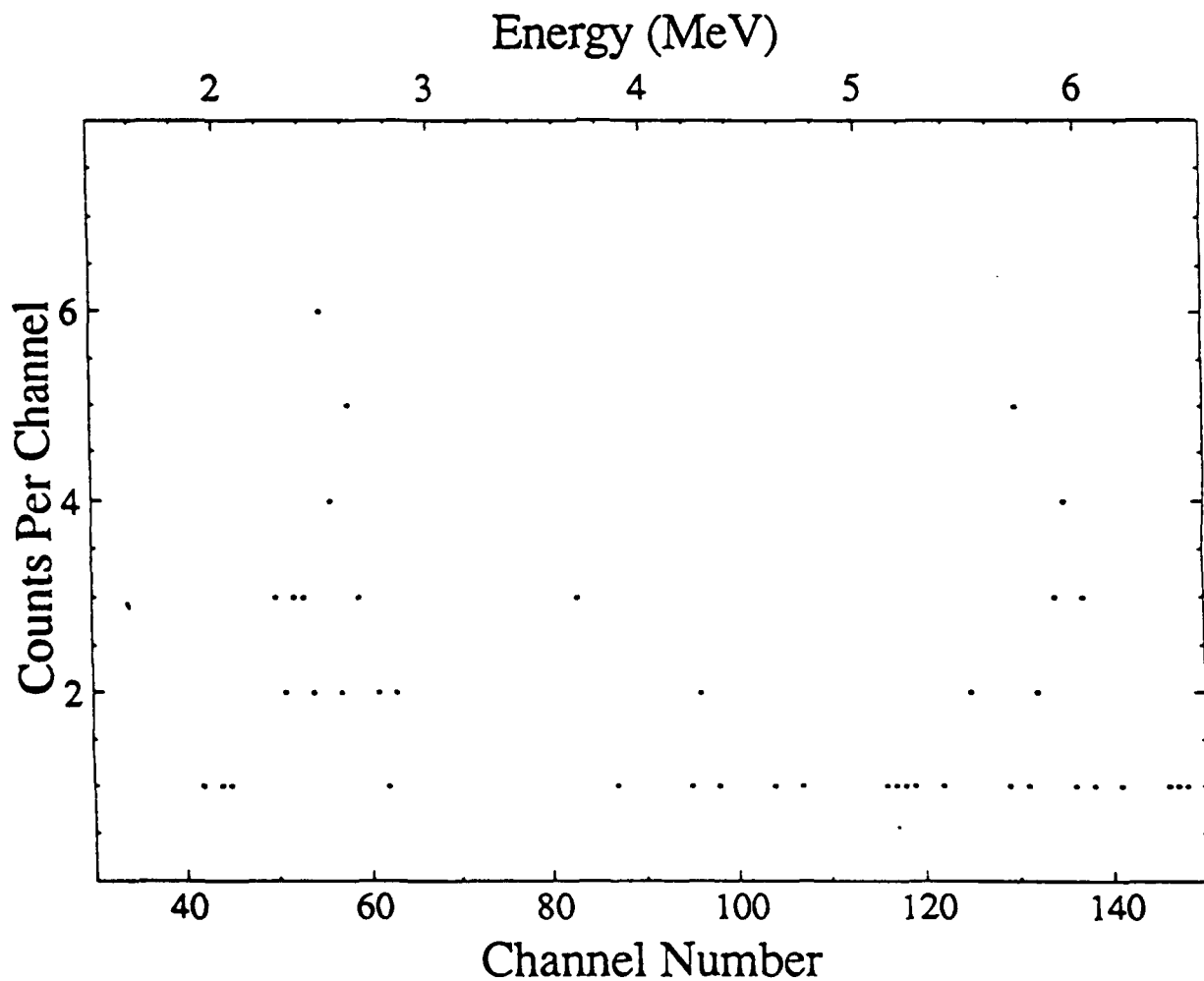


Figure 3. A spectrum acquired over a five minute period during 350 eV deuterium bombardment of a 1- μm thick titanium film evaporated onto a 3.8 μm thick nickel foil backed with an 11.26 mg/cm^2 Ni foil which partially blocked the particle path from the film to the detector. Two peaks appeared simultaneously during a three to five second burst , at 5.7 MeV and 2.5 MeV , respectively. This shift could be accounted for by energetic tritons travelling from the film to the detector.

Hydrogen Bombardment of TiD₂

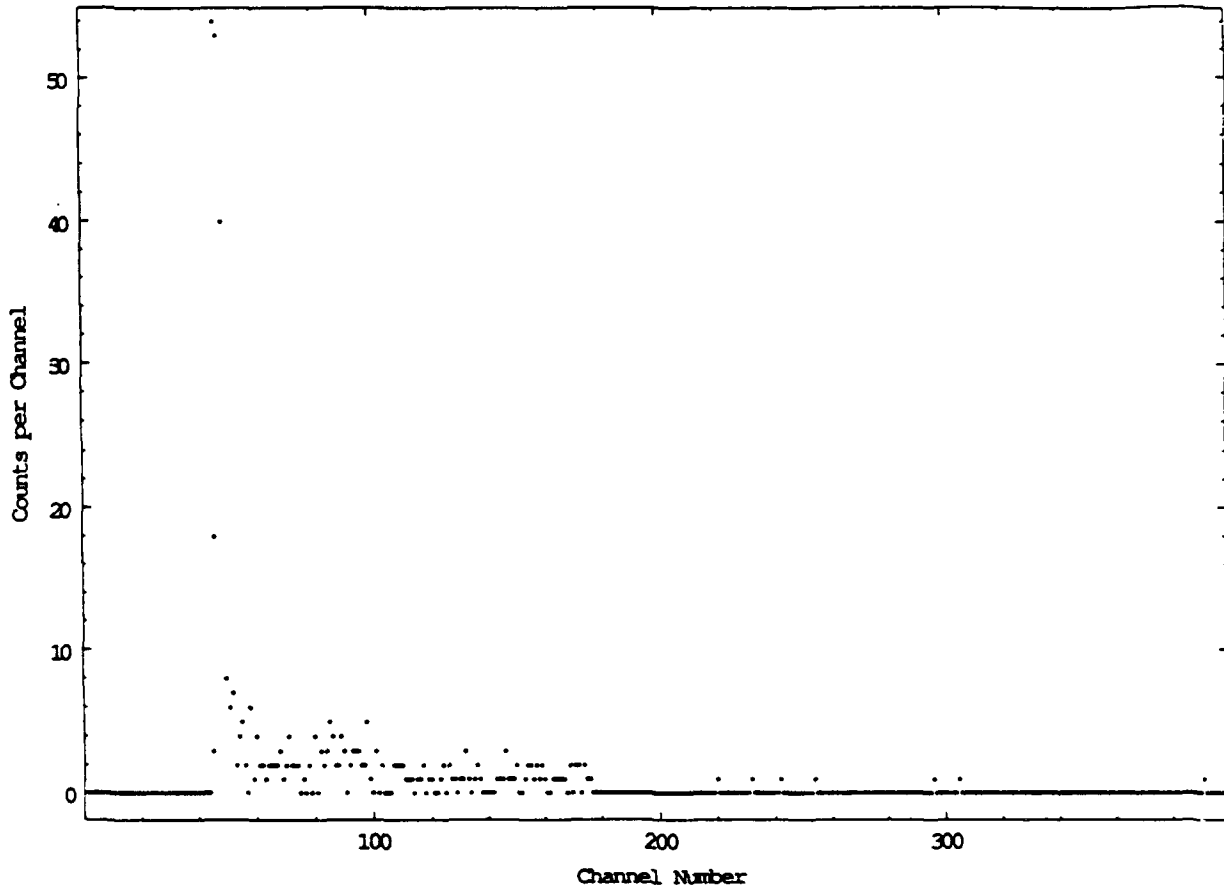


Figure 4. Spectrum acquired for a four hours during a control experiment where hydrogen ions were accelerated an energy of 500 eV at a current density of $200 \mu\text{a}/\text{cm}^2$ onto a TiD₂ thin-film sample. Counts above 1 MeV are caused by arcing (voltage breakdown of the accelerator grids, see text) as observed on a storage oscilloscope. There are no peaks in the spectrum.

Energy Deposited in Unbiased Detector

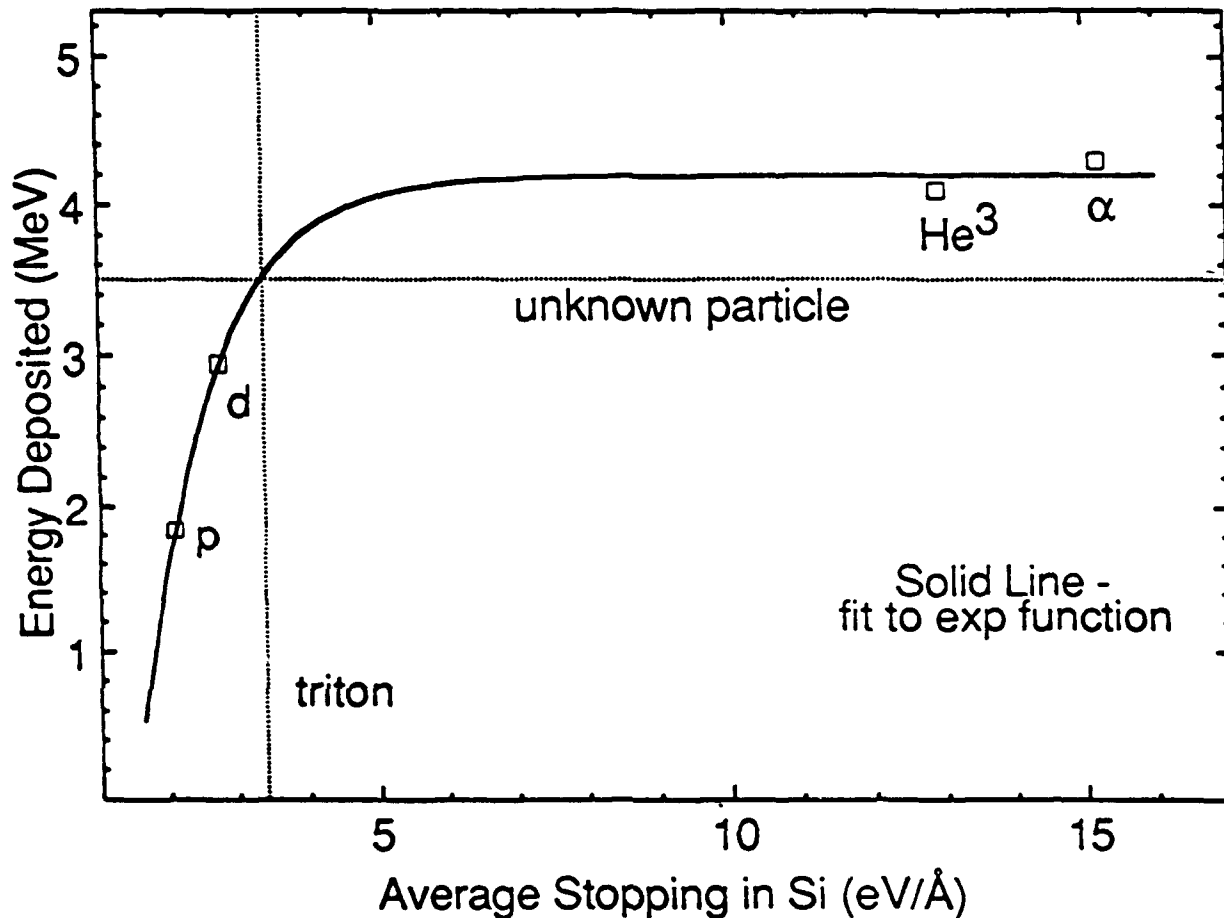


Figure 5. Energies deposited in the unbiased detector for normally incident 5-MeV ^3He , ^4He , protons and deuterons produced in a tandem Van DeGraaff accelerator are plotted against particle stopping powers in silicon averaged over the particle range. The solid line represents a least-squares fit of the data to an exponential function. The observed energy registered at zero bias by the particles detected in our experiment is shown as a horizontal dotted line. The intersection of this line with a vertical dotted line representing the average stopping power of tritium in Si and with the fitted curve suggests that a triton is the most likely candidate for the detected particles.

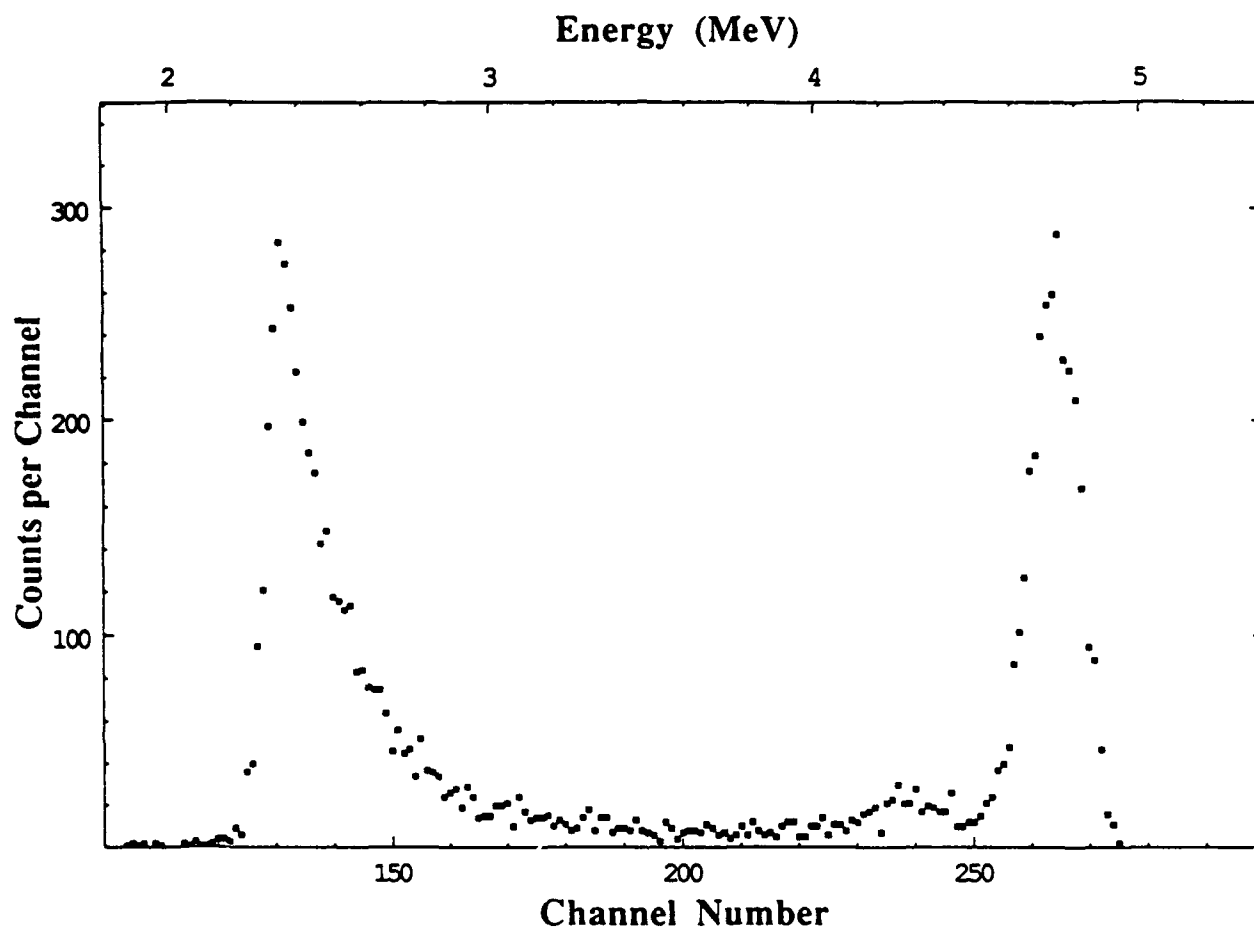


Figure 6. Spectrum of 5.49 MeV alpha particles from a ^{241}Am source taken in vacuum passing through a $1.9\ \mu\text{m}$ Ni foil at two different voltage bias conditions. The peaks at 4.7 MeV and 2.3 MeV were taken at full bias and intrinsic bias (power off), respectively (see text). The 2.3 MeV peak for intrinsic bias exhibits a spectral distortion (an extra density of counts on the high energy side of the peak) caused by the particle range exceeding the depletion depth of the detector. This distortion is similar to that of the peak in Fig. 2b.