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Vincent Tang, Glenn Meyer, Greg Schmid, Chris Spadaccini, Phil Kerr, Brian Rusnak, Steve Sampayan, Brian Naranjo, Seith Putterman

August 10, 2007

Neutron Production from Feedback Controlled Thermal Cycling of a Pyroelectric Crystal

V. Tang, G. Meyer, J. Morse, G. Schmid, C. Spadaccini, P. Kerr, B. Rusnak, and S. Sampayan, Lawrence Livermore National Laboratory, Livermore, Ca

B. Naranjo and S. Putterman, University of California at Los Angeles, Los Angeles, Ca

Abstract

The LLNL Crystal Driven Neutron Source is operational and has produced record ion currents of ~10 nA and neutron output of 1.9 (±0.3) x 10^5 per thermal cycle using a crystal heating rate of 0.2 ^{o}C/s from 10 ^{o}C to 110 ^{o}C. A 3 cm diameter by 1 cm thick $LiTaO_3$ crystal with a socket secured field emitter tip is thermally cycled with feedback control for ionization and acceleration of deuterons onto a deuterated target to produce D-D fusion neutrons. The entire crystal and temperature system is mounted on a bellows which allows movement of the crystal along the beam axis and is completely contained on a single small vacuum flange. The modular crystal assembly permitted experimental flexibility. Operationally, flashover breakdowns along the side of the crystal and poor emitter tip characteristics can limit the neutron source. The experimental neutron results extend earlier published work by increasing the ion current and pulse length significantly to achieve a factor-of-two higher neutron output per thermal cycle. These findings are reviewed along with details of the instrument.

I. Introduction

Recent work [1-3] has shown that electrostatic fields from the pyroelectric effect in specific crystals may be utilized to produce neutrons. Specifically, a $LiTaO_3$ crystal with a nominal pyroelectric coefficient of K=190 μC/m^2-^{o}C [4] can be thermally cycled to produce charges and corresponding electrostatic fields which ionize and accelerate deuterons to ~100's of keV. A tungsten tip field emitter on top of the crystal is typically used to enhance the electric field for ionization. The deuterium beam bombards a deuterated target and ~2.5 MeV neutron emission occurs through the D-D fusion reaction. Figure 1 graphically illustrates the concept along with a typical Faraday cup setup for current measurement and neutron

production. The crystals after heating or cooling, and assuming no charge losses (i.e. ion emission, parasitic electrons, etc.), can reach a potential very roughly given by the 1-D estimate $V = KA\Delta T/(C_{cr}+C_g)$ [5], where C_{cr} is the capacitance of the crystal and C_g is the stray capacitance in the gap between the front of the crystal and the target. A is the front surface area of the crystal, and ΔT is the crystal temperature change. Practically, "pyrofusion" neutron sources could potentially be made very small with little high voltage equipment, and could for example, potentially be used for active interrogation of materials in the field.

In this work, we present first results form the LLNL Crystal Driven Neutron Source (CDNS) experiment based on the pyroelectric effect. The goal of the CDNS project is to extend the work done previously and ultimately raise the D-D neutron output from $\sim 10^3$ n/s to $\sim 10^6$ n/s by, for example, increasing the beam energy and current using different crystal configurations. Pulsed neutron emission is also desired. The beginning project objectives are to extend and reproduce the previous results from ref [1] using a more experimentally flexible crystal assembly and start to verify pyrofusion system models. To these objectives, the CDNS is operational and has produced record ion currents of ~ 10 nA, neutron flat-tops ~ 170 s long at 924 ± 141 n/s, and total neutron output of $1.9\ (\pm 0.3)\ \text{x}\ 10^5$ per thermal cycle. This is compared with neutron outputs of $8.5\ \text{x}\ 10^4$ per cycle and $5.9\ \text{x}\ 10^4$ per cycle from ref [1] and [3] with peak currents of ~ 4 nA in ref [1]. The heightened neutron output and ion current is achieved by extending the thermal cycling range of the crystal from ~ 40 ^{o}C to ~ 100 ^{o}C in a controlled manner using a novel modular crystal assembly with feedback temperature control and configured for rapid changing of field emitters. Additionally, basic pyroelectric charge measurements were also performed and agreed with predictions. Lastly, it is found that the neutron emission and run reproducibility can be susceptible to HV breakdowns and field emitter tip condition.

II. Experiment Setup

The present experimental setup primarily consists of a Kimball Physics 8.4" diameter spherical chamber with an array of modular beam and nuclear diagnostics, and a novel crystal assembly with a feedback controlled thermoelectric heater/cooler (TEC). Figure 2 illustrates the general setup with some of the diagnostics and provides a detailed view of the crystal assembly.

The modular crystal assembly consists of a crystal clamped onto the feedback controlled 39 W TEC which allows the crystal to be cycled from -20 ^{o}C to 120 ^{o}C in a proscribed manner. The ability to proscribe temperature cycles in detail permit numerous possible crystal operating schemes. The entire crystal assembly is mounted onto a single 2 ¾ vacuum flange with a bellows which allows the crystal to traverse along the beam axis. Relevant experimental parameters, such as target distance, can be changed easily in-situ. The crystal setup used for the neutron results in this work employ a 3 cm diameter by 1 cm thick LiTaO$_3$ crystal with optical finish. The front and rear faces of the crystal is coated with a 1000 Å of platinum in lieu of the large copper plate used in ref [1] and ensures an even charge distribution. A smaller ~1 cm diameter copper plate is mounted on the center of the crystal and has a single pin socket from a standard dual in-line package (DIP) mount for holding the 0.02" diameter shank of a tungsten tip field emitter with a 1000 Å radius apex. The tip apex is ~1.1 cm above the face of the crystal but can be changed for different experiments. This socket approach allows rapid changing of field emitter tips for different experiments, and when tips are damaged. The base of the crystal is electrically and physically connected to a thin copper backplate via a low temperature indium-silver solder. There is a coat of Torrseal epoxy around the base of the crystal to ease the electric field stresses at the triple point, similar to the setup in ref [1]. The crystal with backplate is clamped to a copper heatsink with the TEC sandwiched in-between them using a Kel-F ring. Indium foil is compressed between the interfaces to ensure good thermal contact. The use of indium foil and mounting clamps instead of epoxies permit changing the setup quickly. The back of the crystal is grounded using thin indium strips which connects the crystal backplate to the heatsink and system ground electrically. The copper heatsink is cooled with circulating cooling water. A Labview program interfaces with an Alpha Omega 800 TEC feedback controller based on thermocouple readings from the center of the backplate and executes proscribed temperature ramps or flat-tops. Based on 1-D thermal modeling, the temperature gradient between the front and back of the crystal should be less than 10 ^{o}C for thermal ramps with rates of 0.1-0.2 ^{o}C/s. The crystal is aligned so that positive potentials are created when it is heated.

The experiment has several primary diagnostics. The first consists of a Faraday cup for ion beam

measurement and has a fully deuterated erbium (ErD_2) target mounted in it, similar to ref [1] and shown

schematically in Figure 1. The target has a 0.5 μm thick layer of erbium with an estimated deuterium

loading between 1.95 and 2.05 based on analysis of other targets produced using the same setup and

procedures [6]. Using an erbium density of 9 gm/cc and a deuterium loading of 2, the deuterium number

density of the deuterated erbium film is 6.5 x 10^{22}/cc. The cup or target plate is biased at 54V to capture

secondary electrons [7] and is 0.8 cm behind a grounded mesh with ~85% transparency. Almost all of the

secondary electrons are expected to have energies noticeably lower than the bias. The current from the

target is recorded using a Keithley 6514 electrometer. A 2" x 2" Ortec NaI detector mounted on a 2 ¾"

window provides an indirect voltage measurement by detecting the end point of the bremsstrahlung

spectrum from backstreaming or stray electrons. Neutron emission is recorded through a 14 17" long ^{3}He

detector tube array covered with 1.5" thick polyethylene moderator. The array is essentially insensitive to

gammas and has a calibrated 2.5 MeV neutron detection efficiency of 3 (±0.45) x 10^{-3} for the experiment.

The calibration was done with both a known D-D neutron tube and californium-252 source, and accounts

for the experiment geometry along with the slight anisotropy of the D-D neutron emission. A set of

firewire CCD cameras are used to observe breakdowns and for in-situ examination of the field emitter tip.

III. Operating Results

The first set of experiments simply involved a direct charge measurement in air (1 atm) using the Keithley

6514 electrometer. The Coulomb probe was placed in direct contact with the front of a crystal which had a

2.8 cm diameter copper plate mounted to its face using vacuum silver epoxy instead of the more complex

platinum coating described in Section II. The backplate in this preliminary setup was also mounted with

the same epoxy. The TEC was ramped as quickly as possible from room temperature to a specific set

point, and ramped back down after thermal equilibrium was reached. The detected charge was >90% of the

expected value, based on 190 μC/m^2-^{0}C. Figure 3 plots the measurement. The finite rise time of the curves

is due to the thermal diffusion time of the crystal, estimated as ~100 s.

Experiments to produce beams and neutrons followed the basic charge experiments. Figure 4 plots the

relevant parameters for an experiment using the assembly described in Section II. The crystal temperature

was heated from 10 ^{o}C to 110 ^{o}C at 0.2 ^{o}C/s and held at 110 ^{o}C for 600 s, then cooled back down to 10 ^{o}C.

The data after t=600 s is not shown in the figure since no ion current was detected afterwards.   The

proscribed feedback controlled temperatures and measured rear crystal temperatures essentially matched

each other with little overshoot, undershoot, or lag.   A static fill of ~3 mTorr D_2 gas was used. The

emitter tip apex was ~2.4 cm away from the grounded mesh of the Faraday cup, and ~3.2 cm away from

the biased target or current collector. The height of the tip apex above the crystal was again ~1.1 cm. As

the beam current rose, the bremsstralung spectrum from the NaI detector indicated beam energies of ~80

keV, resulting in a noticeable neutron emission flat-top from t=280 to 450 s at an average rate of 924 $\pm$ 141

n/s after background subtraction and accounting for the detector efficiency of 3 ($\pm$0.45) x 10^{-3}. A

background value of 1.1 n/s was used. This background is theorized to stem from cosmic-ray interactions.

The total number of measured neutrons from t=0 to t=600 s, after background subtraction, was 578 $\pm$ 43.

This gives a total source neutron output per thermal cycle of 1.9 ($\pm$0.3) x 10^5 neutrons. Assuming a mesh

transparency of ~85%, a peak current of ~9.5 nA was recorded for this run. These results extend the beam

current and neutron output of previous work by a factor-of-two.

For comparison, Figure 5 plots another ~3 mTorr D_2 run using the preliminary crystal setup described in

the charge measurement experiment. The field emitter tip apex in this case was only ~1.4 cm from the

grounded mesh. The tip was not mounted using a socket and had an apex ~0.4 cm above the crystal. A

slower ramp rate and shorter temperature swing was also used. This run resulted in greater measured

current, ~10 nA, but lower beam energies at ~35-40 keV. Very few neutrons above background were

detected for this run due to the low beam energy.

The enhanced electric field at the tip apex for both of the above runs should have been greater than ~100

MV/cm, based on field enhancement factors extrapolated and estimated from ref [8]. The measured

neutron emission rate and beam energies for the first run indicate that the beam was dominantly made of

D^+, and not D_2^+; this concurs with beam-target simulations using stopping powers from SRIM [9] and

fusion cross-sections from ENDL99 evaluations which indicates a neutron output of 850 n/s for an 8 nA 80

keV D^+ beam impinging on the fully deuterated erbium target. The slight increase in recorded current for

the second run could be due to higher collection efficiency, since the beams are expected to be divergent.

The lower potential or beam energy could be partly due to an increase in stray capacitance, or C_g, as the

crystal is closer to the ground mesh, and also charge neutralization due to any electrons pulled from the

mesh or target. During the cooling portion of the thermal cycle for the numerous runs performed, electron

emission was seen as expected. A systems modeling effort is underway to simulate and understand these

experimental data.

Reproducibility of runs is complicated by changing field emitter tip condition and high voltage

breakdowns. Each time a breakdown occurs, the beam collapses and the effective available charge on the

crystal is changed. Breakdowns occur easily as the electric fields between the front and back of the crystal

are on the order of ~100 kV/cm, easily exceeding the typical DC surface breakdown fields of ~20 kV/cm

[10]. Figure 6 shows a breakdown captured by an Imaging Source color CCD firewire camera, and also

another run where an arc occurred at the tip apex. The breakdowns typically occur as a flashover on the

side of the crystal, as shown by Figure 6a. Emitter tip condition affects the ion current noticeably; runs

with damaged tips like the one in Figure 6b resulted in significantly less or zero ion current. Typically,

depending on experimental conditions, maximum ion currents in the ~0.5 to ~10 nA range were achieved.

IV. Conclusion

We have shown a factor-of-two increase in ion current and neutron output in the LLNL CDNS experiment

over previous pyrofusion work. This was achieved by increasing the temperature swing of the pyroelectric

crystal in a controlled feedback manner using a novel crystal assembly. The incorporation of feedback

temperature control, socket mounts for rapid changing of field emitters, and the ability of the assembly to

traverse the beam axis permits maximum experimental flexibility. In general, flashover breakdowns and

poor emitter tip conditions were found to affect the reliability of the source. We are now working on

applying high voltage engineering methods such as corona rings, and accelerator techniques to reliably

achieve greater voltage and beam currents in order to increase the D-D neutron emission from $\sim 10^3$ n/s to $\sim 10^6$ n/s.

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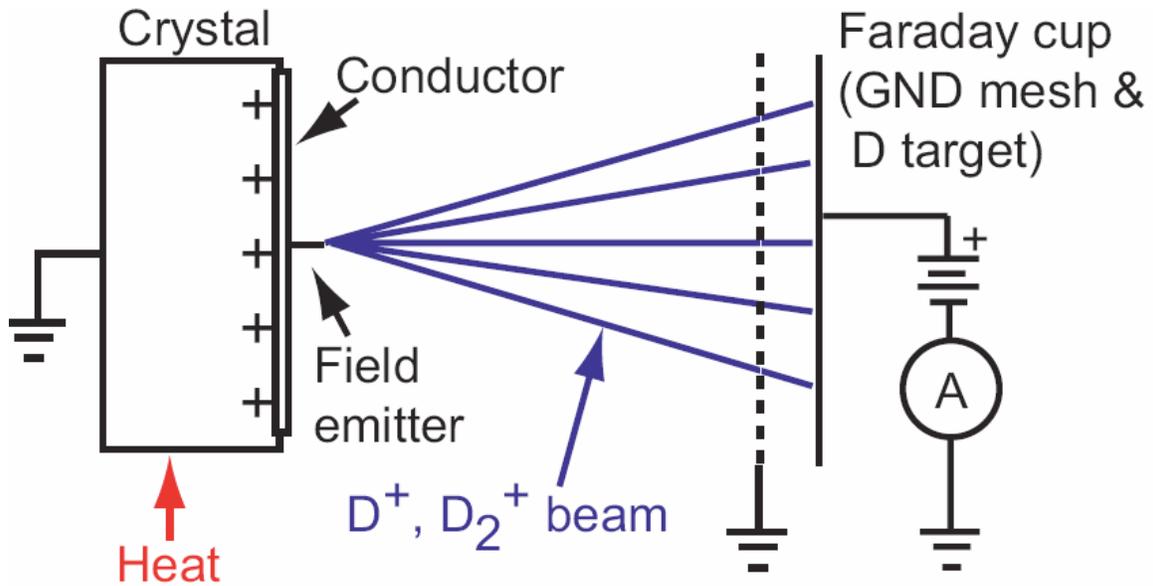[10] S. Humphries, *Principles of Charged Particle Acceleration* (Wiley and Sons, 1986).

Figure 1) Schematic of the pyrofusion concept and a typical Faraday cup-target setup for ion current measurement [1]. The current collector or target plate behind the grounded mesh is loaded with deuterium for D-D fusion and connected to an ammeter for current measurement. The collector is operated with a positive bias to prevent the escape of secondary electrons [7].
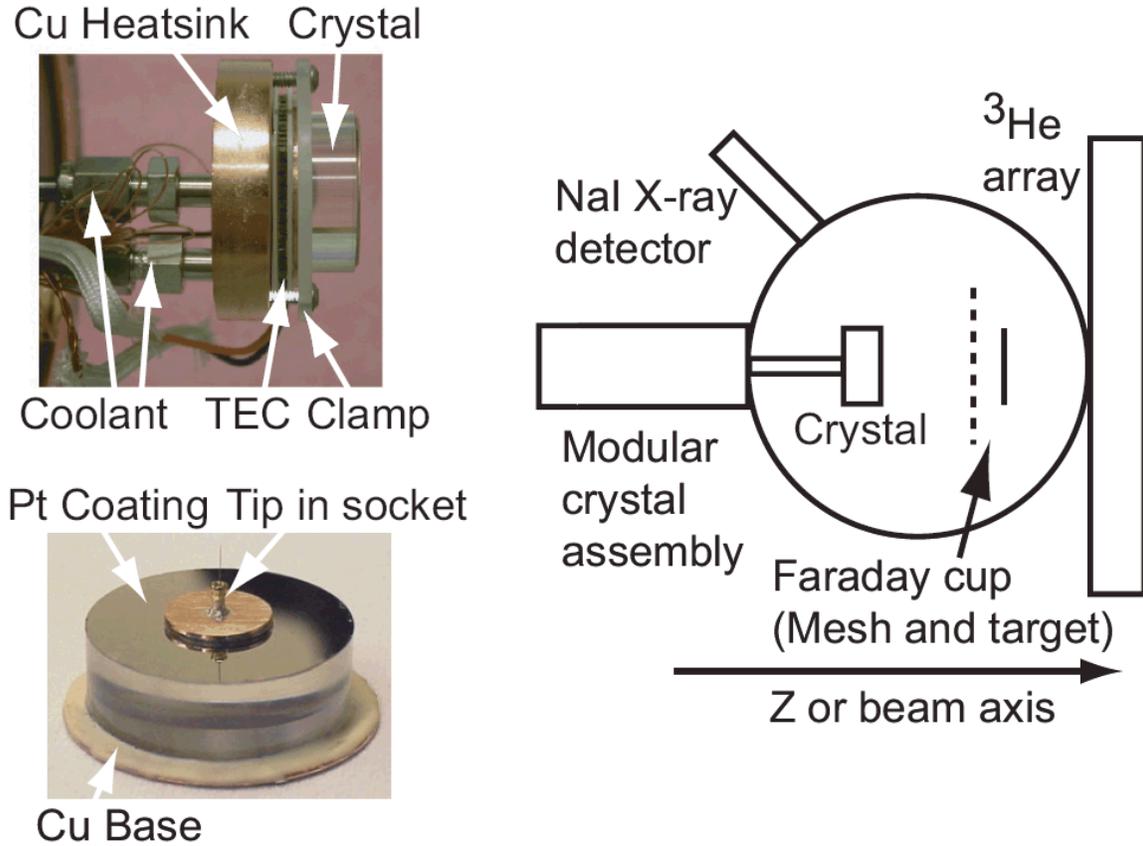
Figure 2) Schematic of the CDNS with close-up views of the modular crystal assembly and one of the crystals. The crystal can be moved back and forward along the beam axis.   The thermocouple for feedback control is sandwiched between the TEC and the copper backplate on the rear of the crystal. A groove cut into the backplate allows the thermocouple to be sandwiched snuggly without causing a gap between the TEC and the backplate.
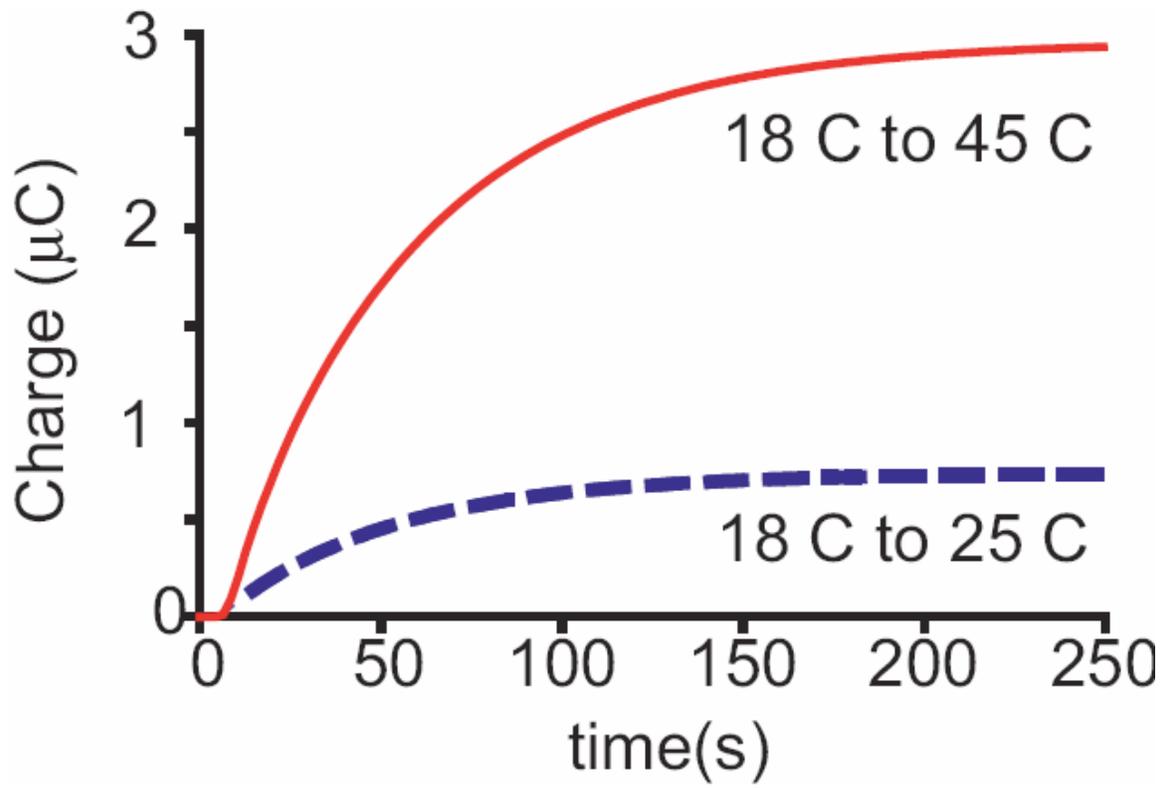
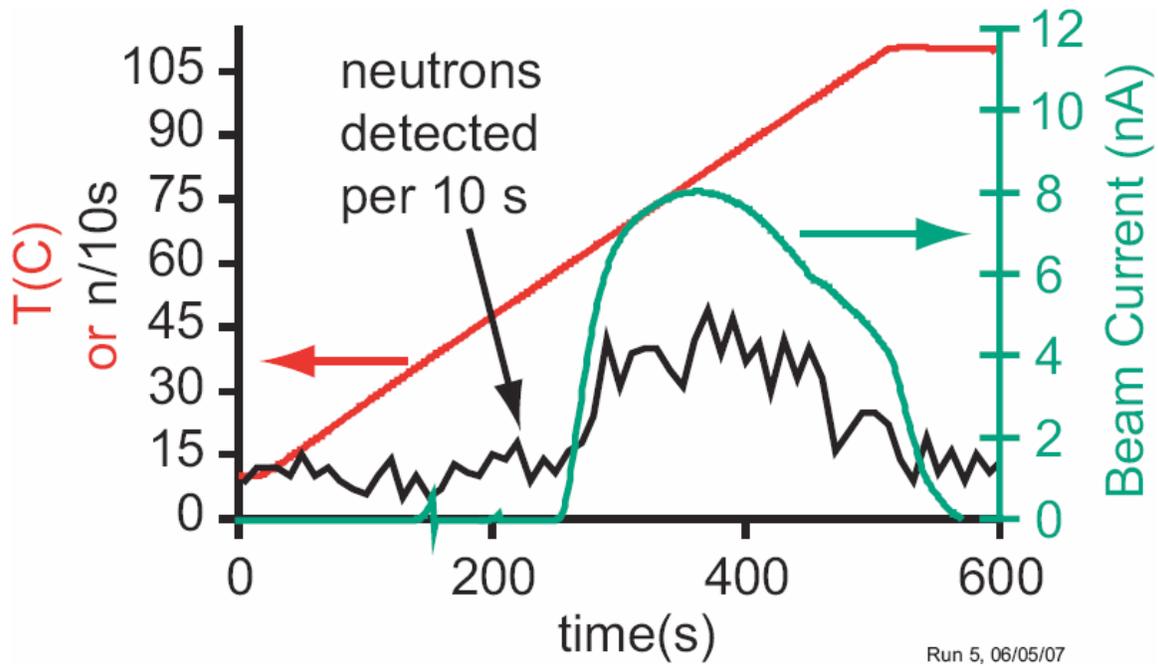Figure 3) Direct charge measurements in air (1 atm) for different temperature ramps.

Figure 4) Measured beam current (right), detected neutrons per 10 s (left), and rear crystal temperature

(left). The prosribed temperatures essentially match the measurements and hence are not shown for clarity.

The neutron flat-top between t=280-450 s has an average source emission rate of 924 ± 141 n/s based on a

detector efficiency of 0.3%. A total of 1.9 (±0.3) x 10^5 neutrons was produced by the source from t=0 to

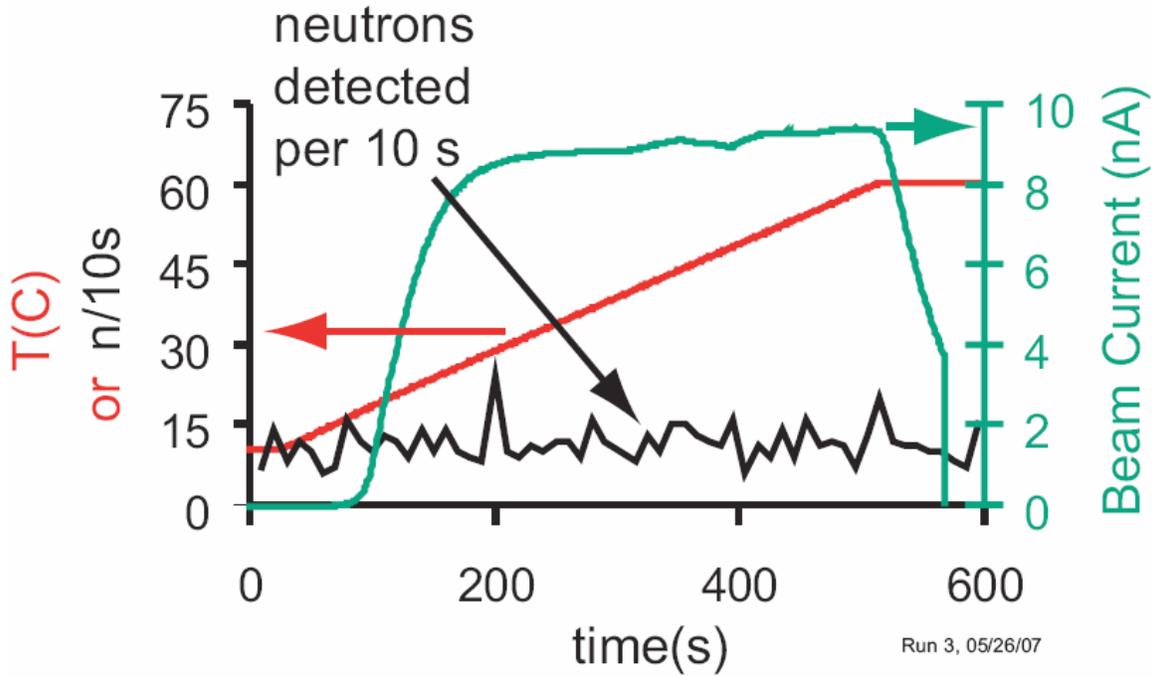t=600 s. A background neutron rate of 1.1 n/s was used for background subtraction.

Figure 5) Measured beam current (right), detected neutrons per 10 s (left), and rear crystal temperature

(left) for highest measured current run. The beam energy was only ~35-40 keV. The beam collapsed at

~575 s due to breakdown. Very few neutrons above background were detected for this run due to the low
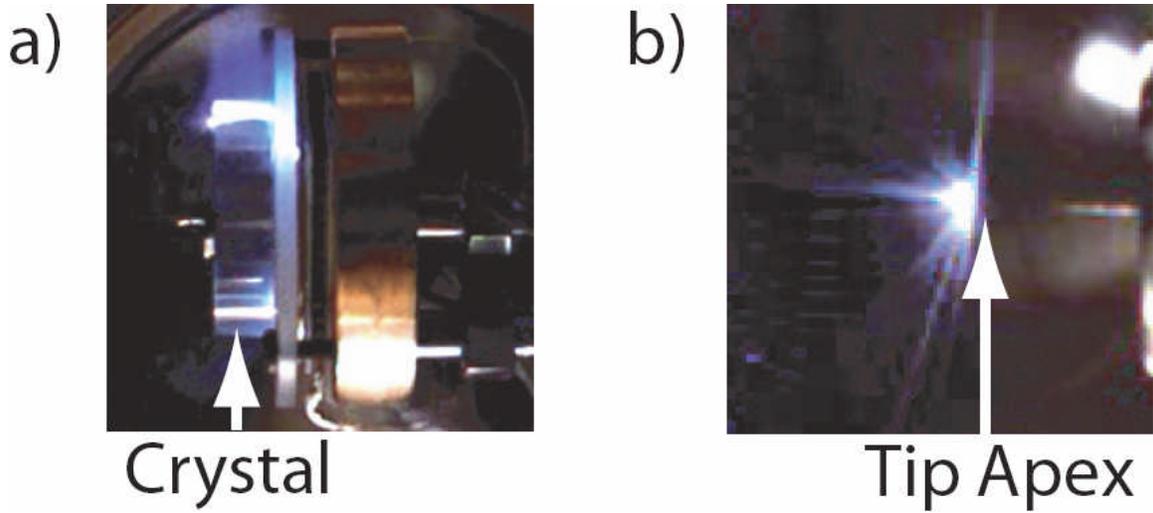
beam energy.

Figure 6a) Typical surface flashover breakdown on crystal side. 6b) Arcing to field emitter tip; the 0.02"
diameter shank is visible but the tungsten tip and its apex is not due to its small radius. The socket mount
design shown here is slightly different than the one described in Section II.