1-9-2004

Development of a Fourier Transform-Based Time-of-Flight Electron Spectrometer with Ultra-High Resolution

Brian Frederick
Principal Investigator; University of Maine, Orono, brian.frederick@umit.maine.edu

Peter H. Kleban
Co-Principal Investigator; University of Maine, Orono, kleban@maine.edu

Recommended Citation

Frederick, Brian and Kleban, Peter H., "Development of a Fourier Transform-Based Time-of-Flight Electron Spectrometer with Ultra-High Resolution" (2004). *University of Maine Office of Research and Sponsored Programs: Grant Reports*. 120.
https://digitalcommons.library.umaine.edu/orsp_reports/120

Final Report for Period: 09/1999 - 08/2003 **Submitted on:** 01/09/2004
Principal Investigator: Frederick, B. G. **Award ID:** 9977800
Organization: University of Maine
Title:
Development of a Fourier Transform-Based Time-of-Flight Electron Spectrometer with Ultra-High Resolution

Project Participants

Senior Personnel

Name: Frederick, B.
Worked for more than 160 Hours: Yes
Contribution to Project:

B. Frederick has been responsible for overall management of the project, coordinating the design, construction and testing of the individual components of the time of flight electron spectrometer. Since this is a development project for an instrument which will be the first of its kind, the close coordination between the theoretical simulation and experimental measurement of the dependence of performance on design parameters is necessary to ensure that the overall instrumental specifications can be achieved.
He is supported through University matching funds.

Name: Kleban, Peter
Worked for more than 160 Hours: Yes
Contribution to Project:

Peter Kleban has been doing theoretical simulations to evaluate the performance of the electron beam chopping device, which is an electrode array to which time-varying potentials are applied. His work includes consideration of how the geometrical arrangement and size of the electrode array affect the potential energy surface as well as dynamical effects in the interaction of the electron beam with the chopper. Kleban plays a significant role in advising a third year, Physics graduate student, Zhongyu Yang, whose thesis work relates experimental measurements of the chopper performance to detailed theoretical simulations.
Kleban is supported through University matching funds.

Post-doc

Name: Jackson, Robert
Worked for more than 160 Hours: Yes
Contribution to Project:

Robert Jackson, a graduate student in Physics who was appointed as Post-doc effective June 1, 2000, has been involved in the entire range of activities since the early work described in the grant proposal. While completing his Ph.D. thesis, involving design of an unrelated UHV instrument, Jackson has not been supported by NSF. Nevertheless, he has made significant contributions at key stages of the chopper development, including theoretical analysis, component design and experimental verification work.

Name: LeGore, L.
Worked for more than 160 Hours: Yes
Contribution to Project:

Jay LeGore has recently completed a Ph.D. in Materials Science at the University of Maine and been appointed as a post-doc effective May 1, 2000. An advisee of Frederick over the last two years, LeGore has gained experience with lithography and materials characterization as well as UHV instrumentation and electronics. LeGore is continuing the work of LeCursi (who is returning to graduate school in prosethics), involving the construction and testing of the chopper device itself.

Graduate Student

Name: Yang, Zhongyu
Worked for more than 160 Hours: Yes
Contribution to Project:

Zhongyu Yang has completed all requirements for candidacy for the Ph.D. program in Physics, and has received recognition for his outstanding graduate record, including his research contribution to this project and his coursework. Since Sept. 1, 1999 he has received a research assistantship and tuition through matching funds associated with this grant. His thesis work involves comparison of chopper performance (experimental measurments) with theoretical simulation (trajectory calculations) of deflection and magnetism, a course in probability and stochastic processes, and audited a course in digital signal processing, which provide a solid foundation for continued work on this development project.

Undergraduate Student

Name: Kresge, Wendy

Worked for more than 160 Hours: No

Contribution to Project:

Wendy Kresge is an undergraduate student in electrical engineering. She assists in deposition of metals in other, sensor related work at LASST, and assisted George Bernhard in the gold deposition. She received no funding from this grant, but has gained experience with RF magnetron sputtering through the work study program.

Name: Davis, Clifford

Worked for more than 160 Hours: Yes

Contribution to Project:

Clifford 'Mike' Davis, a senior chemical engineering major, took a major role in the design and construction of the sample transfer system and linking together the TOF-HREELS vacuum chamber with three other vacuum chambers: a VG preparation chamber with a fast entry lock; a vacuum chamber used for sample analysis; and a photoemission spectrometer designed for operation under elevated pressures at the sample. Mike specified the additional components required to link the systems together, handled the bidding for components with suppliers, interfaced with a UK manufacturer of our custom-designed sample carriers and sockets, and was primarily responsible for assembling the entire system. Mike was nominated for Student Employee of the Year, and was chosen at the University, State, and New England regional levels. Mike was supported partially through the Supplemental funding for an undergraduate and partially through UMaine funds as a work study student.

Technician, Programmer

Name: LeCursi, Nicholas

Worked for more than 160 Hours: Yes

Contribution to Project:

Nick LeCursi is a research engineer working on design and construction of the chopper grid. He has designed the masks for patterning the ceramic substrates, coordinated the lithography and metalization, and performed the wire bonding of the device. For precision alignment of the electrode array, Nick has designed a jig for winding and aligning delicate gold wires (0.001' and 0.002' dia.) which accurately tensions the wire prior to bonding. Nick has received partial support through this NSF grant.

Name: Crothers, C.

Worked for more than 160 Hours: Yes

Contribution to Project:

Bronson Crothers is a soft-money, staff electronics technician at LASST, who has been support part time by NSF. In the first 9 months of the project, he has been responsible for the development of the fast rise-time (~125 ps) output stage driver electronics and the PRBS sequence generator. A simple TTL-based circuit was tested at 20 MHz to modulate the electron beam and then Bronson has used that information to design an ECL-based, microprocessor controlled PRBS modulator card which will operate at 100MHz.

Name: Silvestre, Conrad

Worked for more than 160 Hours: No

Contribution to Project:

Conrad Silvestre is a staff scientist at LASST, responsible for operation of our clean room facilities. He has performed the patterning of the ceramic substrates, using lift-off technology, in aid of the chopper development work coordinated by Nick LeCursi. He has been supported through other funding.

Name: Bernhardt, George

Worked for more than 160 Hours: No

Contribution to Project:

George Bernhardt is a staff scientist at LASST with experience in oxide and metal film deposition on ceramic substrates. George has assisted in the chopper construction by at the gold deposition stage. He has not received support from this grant.

Other Participant

Research Experience for Undergraduates

Name: NgueMba, Joel

Worked for more than 160 Hours: Yes

Contribution to Project:

Years of schooling completed: Junior

Home Institution: Same as Research Site

Home Institution if Other:

Home Institution Highest Degree Granted(in fields supported by NSF): Doctoral Degree

Fiscal year(s) REU Participant supported: 2001

REU Funding: REU supplement

Organizational Partners

Cutting Edge Technology

Cutting Edge Technology (Orono, ME) is a highly specialized, scientific software development company, which markets a graphics interface for a library of statistical algorithms. The library is produced by Spectrum Square Associates, Inc., Ithaca, NY, and includes Bayesian and maximum entropy methods for Fourier deconvolution, smoothing, peak picking, and non-linear least squares fitting.

During the second year of the grant, Cutting Edge Technologies completed the first of four tasks detailed under a subcontract through the University. The company wrote a number of functions for initiating computer control of the University's LeCroy digital storage oscilloscope (DSO), which were combined with their existing graphics interface, Cutting Edge®, to form a new program, TOF-PPC. Specifically, codes were written to: i) download large waveforms (up to 2 billion points) and store to disc file; ii) download waveforms and histograms, store them on disc and display in a graphics interface; iii) download DSO instrument control settings and store to disc; and iv) upload instrument control settings to the DSO. These tools are now available for instrument diagnostic and data collection purposes.

During the third year of the grant, Cutting Edge implemented the data recovery algorithm from Spectrum Square Associates, a version of the Lucy algorithm adapted for use with PRBS modulated data. In addition, utility functions, including autocorrelation, cross correlation, smoothing, and a phase shift, all for the PRBS type of data, were implemented.

Interfacing functions were developed for USB, GPIB (IEEE-488), and RS-232 to allow direct control of external devices.

The research group has also collaborated with Cutting Edge Technologies on the development of new methods for production of choppers. This collaborative work has been supported in part through a Seed grant from the Maine Technology Institute. Cutting Edge Technologies has negotiated an option to license University IP related to the TOF project and seeks to produce chopper hardware and software for a variety of time-of-flight applications.

Millbrook Instruments, Ltd.

Millbrook Instruments, Ltd. (Blackburn, Lancashire, England) has developed an award winning SIMS (secondary ion mass spectrometry) instrument and holds intellectual property in the area of time-of-flight mass spectrometry using advanced beam modulation techniques similar to the methods for electron spectroscopy being developed under this MRI grant. Several fundamental issues are common to both applications, such as methods for gating charged particle beams and recovering the underlying TOF spectrum from the modulated (raw) data.

During the second year of the grant, the research group collaborated with Millbrook in several ways. Millbrook wishes to utilize advanced chopper hardware and software technology in its SIMS product line and to develop new TOF-MS products. To this end, Millbrook have negotiated the basic guidelines of a cooperation agreement with Cutting Edge Technologies to utilize products developed under license agreement with the University of Maine. In addition, Millbrook committed $100,000 in matching support for a Development Award submitted by Cutting Edge Technologies to the Maine Technology Institute (MTI) for commercialization of TOF chopper hardware and software.

In June 2001, Frederick (P.I.), Jackson (post-doc) and Yang (Ph.D. student) visited Millbrook Instruments while en-route to the 10th International Conference on Vibrations at Surfaces in France. Theoretical and experimental work completed under the NSF grant, as well as related work performed under the MTI Seed grant, was presented and discussed.

The Development Award, submitted by Cutting Edge Technologies to MTI, was successful and awarded to a new company, Stillwater Scientific Instruments, formed as a spin-off of the University research personnel. Stillwater has continued to develop the technology under MTI funding in collaboration with Millbrook Instruments. The collaborative work with Millbrook has influenced fundamental work at the University in the specific areas of the chopper design and quantifying the performance of the recovery algorithm.

Spectrum Square Associates, Inc.

Spectrum Square Associates, Inc. (Ithaca, NY) is a highly specialized software development company providing a numerical library of statistical algorithms for spectral processing. They license their codes to original equipment manufacturers as well as to basic research groups around the world, including that of the P.I.

The research group has generated simulated data, based upon the statistical properties and theoretical and experimental understanding of the FT-TOF instrument. Spectrum Square have then tested customized versions of their deconvolution algorithms using the simulated data sets.

Results demonstrating significant advantages in using maximum likelihood methods to recover the TOF spectrum in HREELS were coauthored by University and Spectrum Square Associates personnel. A manuscript, coauthored by Frederick (P.I.) and coworkers, and DeNoyer (of Spectrum Square) demonstrating the use of the recovery methods was published in the proceedings of the 10th International Conference on Vibrations at Surfaces in France in June 2001.

The University disclosed the work as part of a provisional patent in June 2001. The University of Maine, Spectrum Square Associates, and Stillwater Scientific Instruments filed jointly for US and PCT patent protection in June

2002.

Further refinements to the software and functions were completed by Spectrum Square Associates, a final version of the algorithm was delivered, and final payment of the subcontract was completed in August 2003.

Stillwater Scientific Instruments

Stillwater Scientific Instruments was formed in February 2001 to commercialize the PRBS-TOF technology developed at UMaine through partial NSF support under this grant. Personnel transitioned from the University onto Stillwater's funding to apply the technology to mass spectrometry. The company was awarded a Maine Technology Institute Development Award in February 2001 and moved into newly constructed space in the Target Technology Center, a business incubator for Information Technologies. The center is a collaboration between the State of Maine's Applied Technology Centers, the University of Maine, the Bangor Target Development Corporation, and the Town of Orono.

During this time, Stillwater has provided in-kind support to the TOF-HREELS project, primarily in the areas of trouble shooting electronics and software development.

The University and Stillwater have collaborated on further development work and in June 2002 jointly filed two separate patents, in both the US and under the PCT, for chopper fabrication methods and the design of a spectroscopy instrument. The University has continued to provide in-kind support (not using NSF-MRI funds) to Stillwater to promote the commercialization of the technology in the areas of TOF-HREELS and mass spectrometry.

Other Collaborators or Contacts

The TOF detector is being designed essentially as a 'bolt-on' analyzer that could be added to existing HREELS instruments. The University of Maine instrument is based upon an LK Technologies spectrometer, with custom designed features to accommodate the TOF detector and the development work
that took place. The LK 3000 spectrometer was delivered April 30, 2001, and informal discussions with personnel at LK Technologies have continued throughout the grant.

The P.I. hosted a visit from Prof. R. Palmer (Dept. Physics, Univ. of Birmingham, England) who is developing a scanning probe electron energy loss spectroscopy (SPELS) instrument. The concept involves using an STM tip in field emission mode and a conventional dispersive electron energy analyzer as a detector. The advantages of the FT-TOF electron analyzer being developed at the University of Maine for the SPELS instrument have been discussed.

Activities and Findings

Research and Education Activities: (See PDF version submitted by PI at the end of the report)

Our development project culminates with proof of concept of an instrument that is the first of its kind. Thus, there has been an intentional effort to provide a close link between theoretical design work, construction, and testing. This concerted effort provided a solid foundation in which theoretical descriptions guide the choice of instrumental design parameters; likewise, systematic experimental data validated theoretical models.

This MRI grant has provided full-time support to a post-doc and graduate student, and partial support for a second post-doc, an electronics design engineer, an undergraduate student, and the P.I. and Co-P.I., enabling a high level of activity to be maintained during the project. We have refined our experimental and theoretical understanding of the spectrometer and signal processing techniques. Through several generations of improvements in the construction of the vacuum hardware, electronic circuitry, and software methods, the Fourier transform electron velocity analyzer has largely achieved the specifications described in the grant proposal.

As described in the proposal, the critical components that needed to be designed and fabricated were the chopper assembly at the entrance to the drift tube, the detection electronics at the exit of the drift tube, and the software required to recover the underlying TOF spectrum from the modulated signal.

The description of the major activities during the project are organized among three different areas:
o design, construction and testing of the chopper assembly;
o design, construction and testing of electronics; and
o simulation of instrument operation and testing of maximum likelihood recovery software.

A. Chopper design/fabrication/testing:

The chopper development work has proceeded in parallel along several paths:

o theoretical simulation and analysis of the interactions between electrons and the electrode array;
o fabrication of devices using lithographic methods to accommodate smaller electrode size and spacing and to achieve higher precision in electrode alignment; and
o testing of device performance and comparison with theory

The major accomplishments in each of these areas will be described in 'bullet' style below; figures will be attached in a separate pdf document.

1. Theoretical simulation and analysis of the chopper:

Figure 1 illustrates the analytical potential derived using conformal mapping that was used in trajectory calculations. This work has been carried out primarily by a fourth year physics graduate student, Zhongyu Yang, with the guidance of the P.I.'s and Robert Jackson, post-doc. Using this potential energy surface, Yang refined his simulations to quantify how the deflection of the beam depends upon applied voltage and wire diameter and spacing. Using time varying potentials, he also was able to specify how the 'energy corruption', i.e. the energy changes that occur when the electron is near the wires when the potential is switched, depend upon the acceptance angle of the detector. This in turn led to a method for evaluating the statistical distribution of energy corruption and other artifacts of the chopper that were incorporated into simulations of overall instrument performance. This work formed the basis for a paper that was submitted and a poster that Yang and Jackson presented at the 10th International Conference on Vibrations at Surfaces in France, June 17-21, 2001. Their involvement in the theoretical development and their presentation of the results at an international conference represents a significant educational activity. The conclusions from this work will be presented in more detail in the 'Findings' section below.

The trajectory calculations methods were refined and extended to modeling of the entire PRBS sequence for arbitrary energy distributions. This allowed the experimental data for the azimuthally oriented PTFE (Teflon) films to be compared directly with theoretical predictions of the non-ideal effects of the chopper.

The mathematical description of the instrument response function was further refined to include the energy corruption histograms in a space and time-dependent formalism. The method allows the response function to model the entire PRBS sequence. The work then led to an approximate analytical expression for the critical boundaries which were then implemented numerically to obtain response functions. This allowed the dependence of the response function on the kinetic energy of the electron to be quantified much more accurately. This analysis was further refined using the conformal mapping methods described in the 'Findings' section below.

2. Fabrication of chopper devices

To optimize the performance of the TOF instrument, both design and fabrication aspects of the chopper at the entrance to the flight tube have been improved to achieve sub-nanosecond time resolution. The prototype device, described in the proposal, was a hand wired grid built upon a laser-cut ceramic frame, in which a wire-to-wire spacing of 0.6 mm (0.024') was achieved. While this design demonstrated a time response of several nanoseconds, construction of more sophisticated chopper designs and finer spacing with smaller diameter wires would be difficult with this method. Likewise, this device was not optimal for delivering high frequency signals cleanly. Therefore, techniques have been developed for chopper construction which allow impedance matching into the 1 GHz frequency range and the ability to modify geometrical design parameters to take full advantage of theoretical predictions. Additionally, a number of practical aspects of device fabrication have been solved. The various aspects of this activity are as follows:

o The construction of choppers using lithographic techniques has proceeded by purchasing square ceramic substrates (polished alumina) upon which a gold layer was deposited via the liftoff technique. The gold pattern provides bond pads for attaching gold electrode wires. The dimensions of the conduction paths, which feed the signals to the electrode array, were chosen from a consideration of their RF characteristics based upon microstrip techniques. Wire-bonded choppers have been constructed with 90% transmission and spacings of 250 um, 500um and 1 mm. An example of a chopper fabricated with the finest wire spacing (250mm) and wire diameter (25mm) is shown in Figure 2.

ò The artwork for the lithography was designed at the Laboratory for Surface Science and Technology (LASST), University of Maine, by LeCursi with AutoCAD and the masks were produced photographically by a local vendor. The ceramic substrates, specified by LeCursi, were produced by Accumet. Silvestre used the masks and LASST clean room facilities to pattern the substrates, via the lift-off technique. Bernhardt and Kresge performed RF magnetron sputtering of a zirconia adhesion layer followed by the gold layer on the ceramic substrate.

ò LeCursi designed and constructed a jig to accurately align and uniformly tension the electrode wires, suitable for gold wire diameters down to 0.001' dia. The jig is shown in Figure 3. After winding, the device is placed in a holder to support the ceramic substrate while bonding the gold wires to the metallization via parallel gap welding techniques.

ò Testing of the adhesion of the gold metallization and the strength of wire bonds was performed on test samples of the ceramic materials provided by vendors prior to selection of the vendor and specification of the ceramic substrate. This work utilized a Vitrodyne pull-tester at LASST and benefits from similar metal-ceramic lithography work in progress for other research programs.

ò Preliminary design work was done to allow for interchangeable grids in the testing configuration as well as in the chopper assembly itself. The considerations include termination of signals and flexibility to implement multi-level electrode arrays.

ò LeGore continued to develop alternate chopper fabrication methods using silicon micromachining, in part through MRI funding and partly under MTI funding through Cutting Edge Technologies. The structures have advantages in allowing new electrode geometries to be made which are anticipated to have superior beam modulation characteristics as compared to the wire-bonded choppers. Additionally, the micromachining approach allows smaller electrode sizes and should be less labor-intensive during fabrication. A number of materials compatibility issues, such as gold delamination, are being addressed in developing the fabrication methods.

ò Jackson designed a slit assembly that precisely defines the angular distribution of the beam feeding the chopper and determines the detector acceptance angle. The compact assembly, shown in Figure 4, allows for easy exchange of chopper grids, precise alignment of slit assemblies, and the ability to change aperture sizes.

3. Testing of device performance

The design of high performance electron spectrometers requires careful control of the spatial and angular distributions of electron beams. In our case, the design constraints on the input lens are critically dependent upon the characteristics of the chopper. Time-dependent phenomena are also important. In parallel with theoretical design and practical fabrication aspects, we have continued to test the response of an electron beam to the chopper. These activities are summarized below; results are described in the 'Findings' section:

ò We purchased a LeCroy LC584AM digital storage oscilloscope (DSO), and programmed a host computer for acquisition of time-of-flight histograms at acquisition rates of up to 30,000 events per second. This instrument provided the data acquisition requirements for the medium term, as well as the test and measurement facilities for development of dedicated electronics for the chopper driver and detection circuitry (see below). The DSO allowed us to record the time dependent signals applied to the chopper device, accumulate the TOF histogram, and, using the 'Jitter Package' option, allowed variation in signal timing (e.g. cycle-to-cycle chopper on-time duration) to be quantified. Thus, estimates of the driver electronics contributions to the rise time could be separated from the fundamental limitations of the chopper interactions with the electron beam.

ò Measurements have been performed with the original prototype chopper to determine the dependence of the beam energy and electrode potentials on the rise/fall time (in the TOF configuration) and the angular distributions (using static potentials on the chopper with the HREELS instrument described in the proposal). These measurements are in qualitative agreement with theoretical predictions, for example as shown in Figure 5.

o Measurements have been performed with the three different size wire bonded choppers to determine the dependence of the angular distributions (using static potentials on the chopper with the HREELS instrument described in the proposal) on wire spacing, beam energy and applied potentials. More careful investigation of the HREELS instrument revealed a number of peculiarities, such as unshielded lens potentials which were distorting the angular distributions, that required appropriate modifications.

ò To demonstrate the inherent time resolution of the TOF detector, we have designed and fabricated a gas cell, with liquid nitrogen cooling to achieve differential pumping. The gas enters as a molecular beam from above and intersects the electron beam at roughly 90?. The first test will be in a single pulse mode to demonstrate our ability to resolve vibrational losses in the TOF spectrum by comparison with data in the literature using conventional analyzers. Then, we will begin to apply psuedo random modulation sequences to the chopper and test the performance of data recovery algorithms to recover the underlying spectrum from the modulated signal. (See example in the theoretical simulation and analysis section below.)

ò We have performed psuedo-random modulation of the electron beam itself in a test which comprised the chopper, a TTL-based PRBS generator card, the output stage driver cards, and the LeCroy digital storage oscilloscope. A section of the TOF histogram is compared with the applied 1024 bit sequence in Figure 6. The exercise, at a reduced modulation frequency of 4 MHz, was useful as a 'dry-run' for several reasons:

i) to verify design concepts and challenges in the PRBS and driver electronics;
ii) to learn how to make complex timing measurements efficiently with the LeCroy DSO; and
iii) to demonstrate the overall compatibility of the individual components being designed by members of the development team.

Experience gained in this exercise guided the design of next-generation chopper electronics, which run at 125 MHz (see below).

o Using the LeCroy LC584AM digital storage oscilloscope (DSO), we have measured time-of-flight histograms with square wave potentials applied to the chopper. Data was obtained for a range of applied voltage to acceptance angle ratios for choppers of different wire spacing and diameter. We compared the energy corruption and lead/lag effects to theoretical simulations. We have characterized factors such as the rise time and the dynamic range between the open and closed states, as discussed below in the 'Findings' section.

B. Electronics design/fabrication/testing

Electronics design and fabrication focussed primarily on those components necessary for the 'front-end' chopper assembly. The electronics comprise the signal generator, which could be either a single square pulse or an advanced modulation sequence (e.g. PRBS); the output stage driver circuitry; and the transmission lines carrying the signals to the chopper grid itself.

1. Signal Generators

ò A first generation pseudo-random binary sequence (PRBS) generator circuit, based on FAST-TTL was designed and constructed. The prototype, shown in figure 7 with a 20 MHz on-board clock, was constructed using wire wrap techniques and was therefore limited to a clock speed of 4 MHz. Nevertheless, the circuit demonstrated the design concepts and requirements for a second generation, microprocessor controlled PRBS generator. This board was used in the measurements referred to in Figure 6 above.

o A second generation, microprocessor-controlled 'smart' clock board and pseudo-random binary sequence (PRBS) generator card, again based on FAST-TTL was designed and constructed, in collaboration with Cutting Edge Technologies. This version, shown in figure 8, was constructed using printed circuit board techniques and was operated at a clock speed of 10 MHz. The circuit demonstrated the design concepts and requirements for a third generation, ECL-based PRBS generator that needed to operate in excess of 100 MHZ. The design specifications of the 10 MHz system included:
i) microprocessor control of clock and PRBS generator, including loading the PRBS sequence, starting and stopping operation, opening and closing the gate, and selecting the internal 10 MHz or external clock;
ii) detection of the roll-over of the PRBS sequence, supplied to detection circuitry as a synchronization pulse output;
iii) sequence length selectable from 128 to 4096 bits by swapping PCB cards; and,
iv) front-panel control circuitry.

o Design of the 3rd generation PRBS generator, shown in Figure 9, was based on emitter coupled logic (ECL) was then completed in the third year. The chopper driver circuitry consisted of a 125 MHz clock, generated by the Time-to-Digital Converter (TDC), which provided the time base for the PRBS sequence generator. At the core of the PRBS Controller is the Sequence Generator Board. It was designed to accept daughter cards to select the feedback for different shift register lengths. Only the 8-bit shift register length, generating a sequence of period 2^8 û1 = 255 clock cycles, was implemented. The output of the shift register was inverted and then coupled, via a balun, to generate two synchronous output signals of equal and opposite magnitude. Each output signal provided the input to a pulse generator (E&H 1560, E&H International) modified to utilize only the output stage of the pulse source. The 50 ohm output signals could be tuned to balance the amplitude of the drive potentials.

2. Output driver circuitry

ò A pair of first generation driver cards, like the one shown in Figure 10, achieved rise times of approximately 2 ns. These drivers provided valuable data on the performance of the chopper in experiments with electron beams. However, the experience working with these cards and comparison with circuit simulators revealed several limitations of the initial design.

ò A second generation driver card was simulated and fabricated. This version was designed to overcome problems associated with high-speed low distortion electric field generation and simplified the output circuit in order to provide greater power. Additionally, it was compatible with the ECL output of the second generation sequence generator card. Simulations indicated rise times should be improved by a factor of four, which would present no limitation to instrumental time resolution as specified in the grant proposal. Testing of this prototype card revealed coupling between the output and feedback circuitry in the constant current sources. Revisions to the support circuitry design were made and implemented.

o A third generation, double-sided output driver card, shown in Fig. 11, was fabricated and tested, having a number of new capabilities. The signal driver input is isolated with optocouplers to allow the chopper and flight tube to float at potentials up to 700 V. The dual output voltages are generated from a single ended input, simplifying the PRBS generator card requirements. A section at the beginning and end of the PRBS sequence is shown in Fig. 12.

In the final configuration, E&H supplies were modified and replaced the pulse driver output cards.

3. Transmission lines

For optimal performance of the instrument, the amplitude matching and synchronization of voltages applied to opposing elements of the chopper grid is critical. Additionally, the impedance matching from driver card output, through electrical feedthroughs at the vacuum interface, to the chopper device itself are critical to the realization of clean signals on the electrode elements. Jackson designed a custom feedthrough, in which a glass seal is inverted to provide extremely short distances between the output driver and chopper hardware. A set of feedthroughs were manufactured to this specification. Fig. 13 shows the glass feedthrough with the driver card and chopper mounted.

C. General Aspects of instrument design

1. Theoretical simulation and analysis of instrument performance

We have developed methods to simulate the instrument to assess the ability of the maximum likelihood software to recover the underlying TOF spectrum. The simulator is based upon Monte Carlo methods and includes the initial kinetic energy distribution of the beam, details such as the rise and fall times of the PRBS sequence, the drift tube length, and the Poisson noise inherent in the counting experiment. Two data sets, generated with the instrument simulator over the past year, have been used for testing by Spectrum Square Assoc. and have provided very encouraging results, which contribute to chopper and instrument design choices. Figure 14 shows an example, simulating a single pulse TOF spectrum and after modulation with a PRBS sequence. Details of our findings are given in the 'Findings' section and were submitted for publication in the proceedings of the 10th International Conference on Vibrations at Surfaces and was presented by Frederick (P.I.) at the conference in France, June 17-21, 2001.

To demonstrate the inherent time resolution of the TOF detector, we designed and fabricated a gas sample cell, with liquid nitrogen cooling to achieve differential pumping. The gas enters as a molecular beam from above and intersects the electron beam at roughly 90?. The first tests, in a single pulse mode, were not able to achieve sufficiently high sensitivity. In addition, the large spatial interaction region inside the gas cell implied an uncertainty in the position of the loss process, leading to a loss of resolution. We therefore re-designed the gas cell, placing the beam at the center of the mono rotation and the chopper assembly at the entrance to the flight tube. Benzene was used since it could be pumped by the liquid nitrogen trap and has negative ion resonances at low energies where sufficient time resolution could be achieved. Although this configuration still did not have the sensitivity to see loss features, the exercise pointed out the importance of the dynamic range of the instrument and the acceptance angle for isotropic scattering. The main limitation to the dynamic range is a tail on the loss side of an electron pulse, which appears to be due to inelastic scattering. Since the design of the chopper slit assembly and estimates of the scattering probability from the grid wires do not account for the tail, the most likely explanation is that the 1st generation HREELS monochromator does not produce a clean monochromatic beam at levels of less than 1% of the primary beam intensity. New measurements with the state-of-the-art instrument (delivered April 31, 2001) will therefore be critical to resolving this issue.

The high resolution electron energy loss (HREEL) spectrometer comprising a conventional dispersive sector electron monochromator and analyzer was ordered in June of 1999 with University start-up funds through LK Technologies (Bloomington, IN). The specification of > 14 pA current with < 1meV FWHM energy resolution is considered to be the current state-of-the-art. Custom design modifications included an increased monochromator rotation range with stepper motor control. The ultra-high vacuum (UHV) chamber was modified for addition of the TOF detector to allow direct comparison with measurements using the conventional analyzer, as well as ports to accommodate future development work. A schematic of the chamber is shown in Figure 15.

The TOF Analyzer consists of the chopper assembly, a deflection lens directly following the chopper at the entrance to the drift tube, and a micro-channel plate detector. The flight tube is shielded from the earthÆs magnetic field via a double mu-metal shield. A functional diagram of the electronics is also included in Figure 15.

Findings: (See PDF version submitted by PI at the end of the report)

(The section numbering is the same as in the Research Activities section to aid in cross-referencing.)

A. Chopper design/fabrication/testing:

1. Theoretical simulation and analysis of the chopper:

Results of trajectory calculations characterizing chopper behavior:

Software was developed to perform trajectory calculations over the model potential energy surface (Fig. 1, Activities section) and analyze the chopper response function:

i) Figure 16 shows that the angular distribution of electrons, deflected out of the
beam as a function of applied voltage agree very well with the experimental measurements. The deflection angle increases linearly with applied voltage for high transmission (R<d). The alternating potentials on the grid are designed to reduce the field penetration along the flight tube, which decreases with the spacing, d, at constant transmission. Smaller wire spacing leads to stronger fields for the same applied voltage; however, a detailed analysis revealed that, because the transverse field exists over a shorter spatial range, to achieve the same deflection angle the applied voltage cannot be reduced with wire spacing.

ii) Figure 17 illustrates a simulation of the response of an electron beam to square wave potentials applied to the chopper. The spikes and tails, due to energy corruption effects (changes in energy for electrons close to chopper wires when the potentials are switched), and 'lead/lag effects' of the chopper (differences between the time the potentials are off and the time the electron beam is transmitted), decrease significantly as the chopper spacing is reduced.

iii) Figure 18 shows the critical boundaries, beyond which the electron must pass to reach the detector when the potential is turned off, for various ratios between the applied voltage and detector aperature. Monte Carlo integration of the potential (contours shown) is used to determine the distribution of energy corruption for a particular chopper size, beam energy and deflection voltage. We have also found an approximate analytical expression for this boundary (see below).

iv) Figure 19 shows energy corruption histograms for electrons in various regions away from the gate at the time the potentials are turned off, which are used in simulation of the instrument response. We find that the energy corruption places a greater limitation on the ultimate energy resolution of the instrument than the rise and fall times during beam modulation.

v) Figure 20 shows that the change in the shape of the response function, at kinetic energies of 4.0 and 3.6 eV, is negligible. This is critical to the assumption that the data is a convolution of the underlying time-of-flight spectrum with the instrument response function over the entire width of the energy loss spectrum.

3. Testing of device performance

The measured angular distributions have been compared with theoretical simulations (Fig. 16). The results show that the deflection angle as a function of wire spacing and the ratio of kinetic energy to applied voltage is in good agreement with our analytical model when the beam fills the region between consecutive wires uniformly. Several more subtle effects have also been observed, as illustrated in Fig. 21. The deflection angle depends upon the position at which the electron crosses between the wires, consistent with the theory, and for more highly focussed beams, the magnitude of the deflection angle is different if the applied voltages are reversed. This can in principle be used to determine the position and spread of the electron beam. Additionally, we find that the width of the angular distribution, measured with the conventional analyzer, is narrower for the deflected beams than for the straight-through distribution. Comparison with theory confirms that the interleaved comb chopper behaves like a lens, with a poor focus at relatively short distances from the chopper under our conditions. By comparison, when the angular distribution is measured at larger distances, as is the case with the TOF detector, the angular spread of the deflected beams is significantly larger than the straight through angular distribution.

The time dependent response is illustrated in Fig. 22. The dependence of the modulation depth (dynamic range between open and closed states) on applied voltage is simply due to the fraction of the beam which is excluded from the acceptance apperture, and is consistent with DC measurements and simulations. With the higher data collection efficiency of the LeCroy DSO, we were able to collect sufficiently good data to show that the spikes and tails on the edges, most clearly seen in the data for the chopper with largest wire spacing, are real and are consistent with the theoretical simulations. The data was measured in 250 ps bins, which shows that rise times below 500 ps can be achieved experimentally. The observed time resolution therefore substantiates our measurements with the LeCroy DSO Jitter & Timing software that variation in the electronics output is in the sub 10ps regime. Careful analysis of the difference between the time that the voltages are low (measured as the FWHM of the driver waveforms) and the time the beam is 'on' (FWHM of the TOF histograms) shows that there is a lag in turning on and a lead in turning off. The voltage dependence of this effect is shown in the lower left graph of Fig. 22 and can be understood with the aid of Fig. 18. The chopper artifacts were measured also as a function of detector position. The tails which are observed on the left (loss energy) side of the square pulse decay over a time corresponding to an energy range of several eV. The origin of this is not certain, but is most likely due to artifacts of the early generation HREELS monochromator utilized in these measurements.

C. General Aspects of instrument design

1. Theoretical simulation and analysis of instrument performance

The technique of psuedo-random binary sequence (PRBS) modulation has been utilized for some time in a number of different areas; however, to our knowledge, no attempts have been made to directly deconvolute the modulated data. The standard analysis method is to cross correlate the data with the original PRBS modulation function. Figure 23 shows a set of simulated data and illustrates schematically the difference between the traditional cross correlation analysis and the direct deconvolution methods we have developed. The special property of an ideal PRBS is that its autocorrelation function is a delta function. Therefore, cross correlation of the the data with the PRBS is equivalent to convolution of the data with a delta function. Due to limitations of the charged particle gate, it is not possible to operate the gate in a mode where the open time is small compared to the required time resolution. Under these conditions, the autocorrelation function of an ideal PRBS is a triangle wave, and the throughput advantage due to the 50% duty cycle of the PRBS leads to a degredation of the instrumental resolution. Therefore, we oversample the signal by a factor of 16-64, and deconvolute the PRBS from the data directly.

Several sets of simulated data were generated to investigate the effects of the Poisson noise distribution, the finite rise time of the chopper response, the lead/lag effects, and the energy corruption problems. The results demonstrated that the PRBS modulated data could be recovered with maximum likelihood methods, improving the resolution significantly over the traditional cross-correlation methods while maintaining the throughput advantage anticipated.

The effects of the Poisson noise distribution, due to the low signal intensities in HREELS and consequent need for pulse counting electronics, is illustrated in Fig. 24. The results indicate that, although the cross correlation analysis is not strictly valid in the presence of a Poisson noise distribution, the artifacts are sufficiently small to obtain an initial estimate of the underlying spectrum for further maximum likelihood signal processing.

Figure 25 illustrates the performance of the maximum likelihood algorithm for direct deconvolution of the modulated data in the case that the chopper response is not an ideal PRBS, but has linear, 1ns rise and fall times. The result shows that, for simulated data containing a total of 256 million counts (corresponding to data acquisition times of several minutes, as compared to several hours with a conventional instrument), a significant improvement in resolution vs. the cross-correlation method is obtained. The achievable resolution was limited in this case by the size of the sampling time bins.

More realistic simulations of the instrument were performed that included the energy corruption effects of the chopper. These effects were based upon the trajectory calculations for the chopper potential model developed. The resulting data was provided to Spectrum Square Associates and the results (Fig. 26) show that the TOF spectrum can be recovered with the performance (throughput) advantage expected, while achieving significantly improved resolution, compensating for the energy corruption and lead/lag effects of the non-ideal chopper response, and accounting for the Poisson noise distribution. The results (patent pending) have been submitted in a second paper on application of maximum likelihood methods to PRBS modulated data. (See papers submitted.)

Simulations of the instrument operated with limiting cases of non-ideal response functions were used to determine the types of artifacts that the data recovery algorithms could and could not handle. An example, shown in Figure 27 shows that when the height of the a single bit (1Æs) in the response function is reduced relative to the height of multiple ôonö bits in the PRBS sequence, the data recovery algorithm can generate artifacts.

KlebanÆs experience in conformal field theory (and more pertinently, conformal mapping) led to some useful results for the analysis of the chopper response using techniques from a seemingly very different area under grant NSF-DMR prop. no 0203589, ôExact Results in Model Statistical Systemsö.
The Co-PI
has derived new analytical formulas for small electron scattering angles and also for the number of ions with a given excess voltage (ôenergy corruptionö) when the voltage is turned on or off. These quantities are of central
importance in understanding the performance of the instrument.

2. Measured performance of instrument

Figure 28 shows results of the output drive signals and the associated response function when the monochromator was directed into the TOF analyzer. A number of non-ideal characteristics became evident that were not present under the conditions of the chopper tests. The limitations of the rise time lead to a decrease in the transmission of single 1Æs relative to multiple 1Æs in the PRBS sequence.

The limitations of the instrument were thoroughly analyzed, and further refinements made to improve the instrument response function. In Figure 29, a response function at a primary beam energy of 4 eV shows that very clean responses can be achieved. In this data, the noise is dominated by the Poisson noise distribution. Synthetic data sets were also obtained as a means to characterize the instrument using the monochromatic beam without a sample. As shown in Figure 30A, we superimposed PRBS modulated data, acquired at energies of 4.0 and 3.87 eV, to simulate a two-featured spectrum, corresonding to an elastic peak at 4.0 eV and a loss feature at 3.87 eV of 5% intensity. The longer flight time results in a phase shift of the loss feature and contributes intensity in the troughs of the elastic peak PRBS sequence. Deconvolution of similar data indeed reveals the the two featured spectrum. A similar measurement, made with a 2.0 eV ôelastic peakö and a 60 meV loss with a resolution of less than 2.5 meV, is shown in Figure 30B. This demonstrates the resolution goal proposed in the original grant proposal.

Further work with real samples is shown in Figure 31. A section of the PRBS modulated data, measured off an oriented PTFE (Teflon) sample is compared with the response function estimated from directing the monochromator into the TOF analyzer and using signal processing methods. Figure 31B compares the deconvoluted spectrum with a spectrum obtained with the conventional HREELS analyzer, after conversion to a time-of-flight spectrum. The results demonstrate that the features of the loss spectrum can be obtained with a high degree of fidelity and perhaps slightly higher resolution than with conventional instruments. We take these results to demonstrate proof-of-concept for the development of the worldÆs first PRBS-modulated time-of-flight HREELS spectrometer.

Training and Development:

The research has been a central part of the graduate education of the Ph.D. graduate student, Zhongyu Yang, who received his Ph.D. in May 2003, and is pictured in Fig. 32. He has been able to apply the methods and knowledge of his two graduate electricity and magnetism courses to calculation of the potential energy surfaces. He has also gained experience with numerical methods through the trajectory calculation work. By making direct comparisons between his theoretical predictions and his experimental measurements, this project provides a unique opportunity to connect mathematical abstractions to physical phenomena.

The project also provides research experience to two post-docs, Jackson (also shown in Fig. 32) and LeGore, three staff engineers, Crothers, LeCursi, and Silvestre, and three undergraduates, Ngue Mba, Davis and Kresge. Each have gained experience in new areas as a direct result of this project. The integration of the UHV systems to allow sample transfer between the TOF-HREELS, preparation chamber, and controlled atmosphere photoemission spectrometer (CAPES) (Figure 33) was spearheaded by Mike Davis, who received University, State, and regional awards for Student Employee of the Year in May 2003 for this work.

Outreach Activities:

The PI has been involved in K-12 education through one graduate student's involvement in the GK-12 Sensors program. In addition, the PI and several group members have usually volunteered as judges for the Maine Science Fair held at UMaine each Spring.

Journal Publications

R. H. Jackson, L.J. LeGore, Z. Yang, P. Kleban, B. G. Frederick, "Application of the Interleaved Comb Chopper to TOF Electron Spectrometry ", Surface Science, p. 240, vol. 502-503, (2002). Published

L.J. LeGore, R. H. Jackson, Z. Yang, P. Kleban, L.K. DeNoyer, B. G. Frederick, "Advantages of Maximum Likelihood Methods for PRBS Modulated TOF
Electron Spectrometry", Surface Science, p. 232, vol. 502-503, (2002). Published

Books or Other One-time Publications

Web/Internet Site

URL(s):
http://www.ume.maine.edu/LASST/research/projects_research.htm
Description:

Other Specific Products

Product Type: Instruments or equipment developed

Product Description:

The time-of-flight (TOF) high resolution electron energy analyzer developed under this grant achieves a throughput advantage of 500-1000 over conventional HREELS spectrometers while maintaining state-of-the-art resolution. Two patents have been filed based upon this work, both in the US and under the PCT:

1. US Patent Application No: 10/165,852 L.J. LeGore, R. H. Jackson III, Z. Y.Yang, L. K. DeNoyer, P. H. Kleban, B. G. Frederick "Spectroscopy Instrument Using Broadband Modulation and Statistical Estimation Techniques to Account for Component Artifacts", filed June 7, 2002.

2. International Application No: PCT/US02/18006 L.J. LeGore, R. H. Jackson, Z. Y.Yang, L. K. DeNoyer, P. Kleban, B. G. Frederick "Spectroscopy Instrument Using Broadband Modulation and Statistical Estimation", filed June 7, 2002.

3. US Patent Application No: 10/165,851, N. LeCursi, L. J. LeGore, R. H. Jackson III, C. B. H. Crothers, P. H. Kleban, B. G. Frederick, "Fabrication of Chopper for Particle Beam Instrument", filed June 7, 2002.

4. International Application No: Claims Benefit of 60/296,850, filed June 8, 2002, N. LeCursi, L. J. LeGore, R. H. Jackson III, C. B. H. Crothers, P. H. Kleban, B. G. Frederick, "Fabrication of Chopper for Particle Beam Instrument", filed June 7, 2002.

Sharing Information:

HREELS instruments are largely research based instruments, but can be applied to a broad range of surface science studies, primarily with single crystal samples. Therefore, they are typically used in model studies. The technique has advantages of high dipole sensitivity and the possibility of non-dipolar interactions. Typical uses are in the area of catalysis and surface chemistry where vibrational spectra identify and in some cases determine the orientation of adsorbed molecules; semiconductor physics where plasmon and phonon modes can be mapped; and solid state physics of metals and insulators in which phonon band structure is mapped.

Contributions

Contributions within Discipline:

This development project spearheads a new generation of electron spectrometers in the field of surface science, with performance up to 500 times better than current instruments. The advantages of faster acquisition rate or better signal to noise will mean that analysis of the elemental composition, chemical bonding or electronic states at the surface of materials, such as metals or semiconductors, can be performed more quickly and less expensively. The performance advantages will allow a host of new experiments to be performed, such as our recent example of inelastic diffraction {Frederick, et al. Surf. Sci.418 (1998) 407}.

Contributions to Other Disciplines:

The demonstration of the technique for application to electron spectroscopy is likely to provide spinoffs in mass spectrometry. The widespread use of time of flight (TOF) methods for mass spectrometry suggests possible contributions to other fields, from biology and bio-medical research to materials science and environmental applications.

Stillwater Scientific Instruments, a spin-off our research at UMaine, was founded to develop and commercialize the technology for mass spectrometry.

Contributions to Human Resource Development:

My research group, consisting of the P.I. and Co-P.I., two post-docs, two Ph.D. students, one M.S. student, one B.S.(Physics) and two technicians (GC/MS tech, electronics engineer), meets weekly for 1-2 hours. During this time, members present on-going work and important papers in the literature. I strive to have an engaging and supportive discussion where all members, from the B.S. to P.I. level can ask questions and learn new ideas. The B.S. student, Joel Ngue Mba, is of West African descent, and has tremendous potential. I hope that his experience is a positive influence on his future career choices and that he will be one of the 'retained' statistics as a minority in the field of physics. I also have one female technician and will be taking on a new female Ph.D. student. I believe that the level of respect within the group for all members is conducive to improving diversity in my field.

Contributions to Resources for Research and Education:

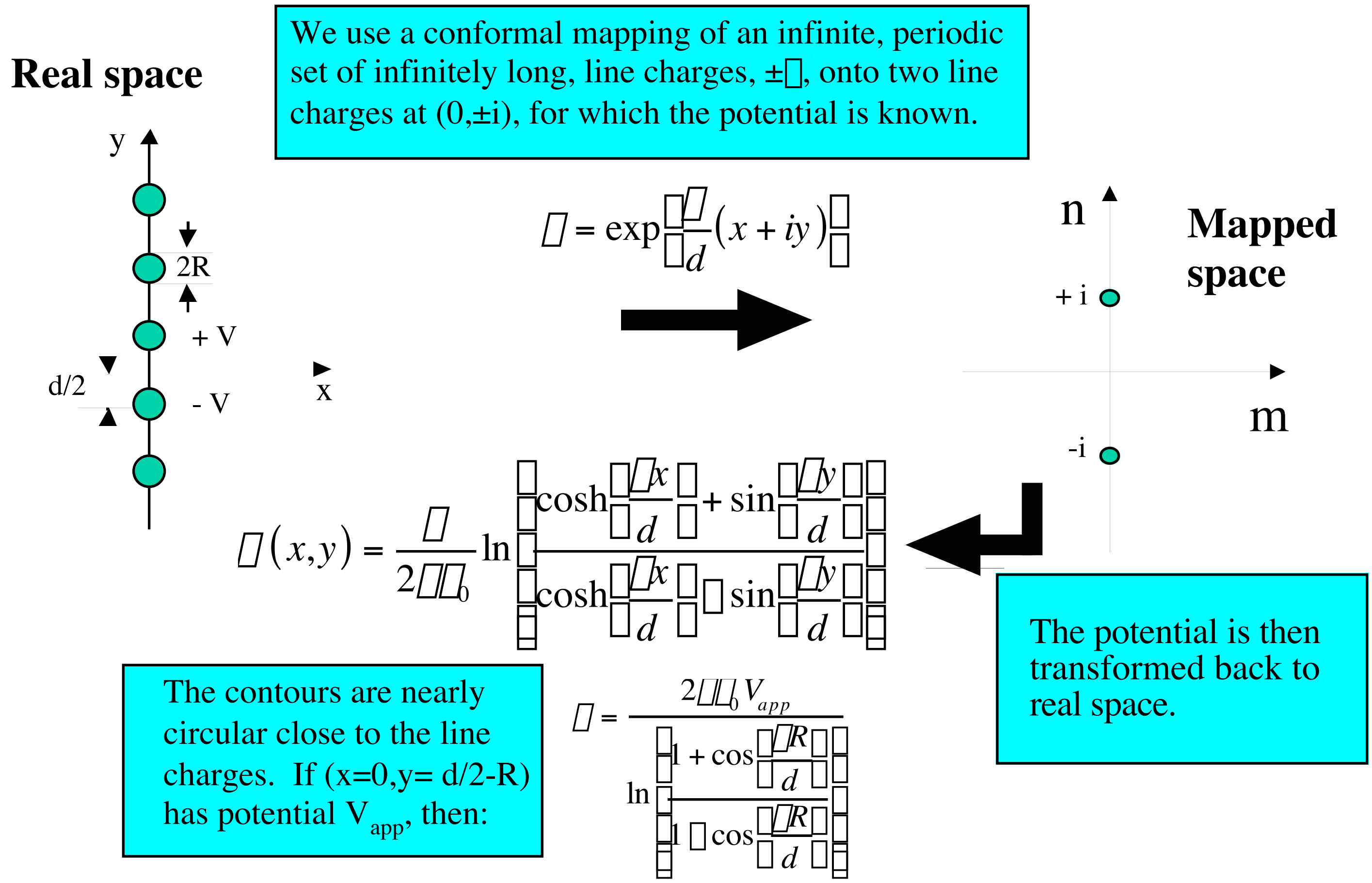
One of the post-docs created a TOF-based lab for the Senior Laboratory course in Physics, based upon early developments in this MRI project. The B.S. physics student, now supported under a supplement, has also been involved in the NSF-REU program administered in Electrical Engineering and has been working on a Senior project within my group. In the near future, I expect that the instrument being built will begin to be used by members of my group and other groups in LASST.

Contributions Beyond Science and Engineering:

I have been heavily involved in the start-up of Stillwater Scientific Instruments and have represented both the University and the company at State and National events, including a National Academy of Sciences workshop on 'Reducing the Time from Basic Research to Innovation in the Chemical Sciennces'.

Categories for which nothing is reported:

Any Book



We use a conformal mapping of an infinite, periodic set of infinitely long, line charges, ±λ, onto two line charges at (0,±i), for which the potential is known.

$$\zeta = \exp\left[\frac{\pi}{d}(x+iy)\right]$$

The potential is then transformed back to real space.

$$\phi(x,y) = \frac{\lambda}{2\pi\varepsilon_0}\ln\left[\frac{\cosh\left(\frac{\pi x}{d}\right)+\sin\left(\frac{\pi y}{d}\right)}{\cosh\left(\frac{\pi x}{d}\right)-\sin\left(\frac{\pi y}{d}\right)}\right]$$

The contours are nearly circular close to the line charges. If (x=0,y= d/2-R) has potential V_{app}, then:

$$\lambda = \frac{2\pi\varepsilon_0 V_{app}}{\ln\left[\frac{1+\cos\left(\frac{\pi R}{d}\right)}{1-\cos\left(\frac{\pi R}{d}\right)}\right]}$$

Fig. 1. Theoretical model based upon an infinite, periodic array of wires of finite diameter and infinite length which has been solved exactly to obtain an analytical expression for the potential.

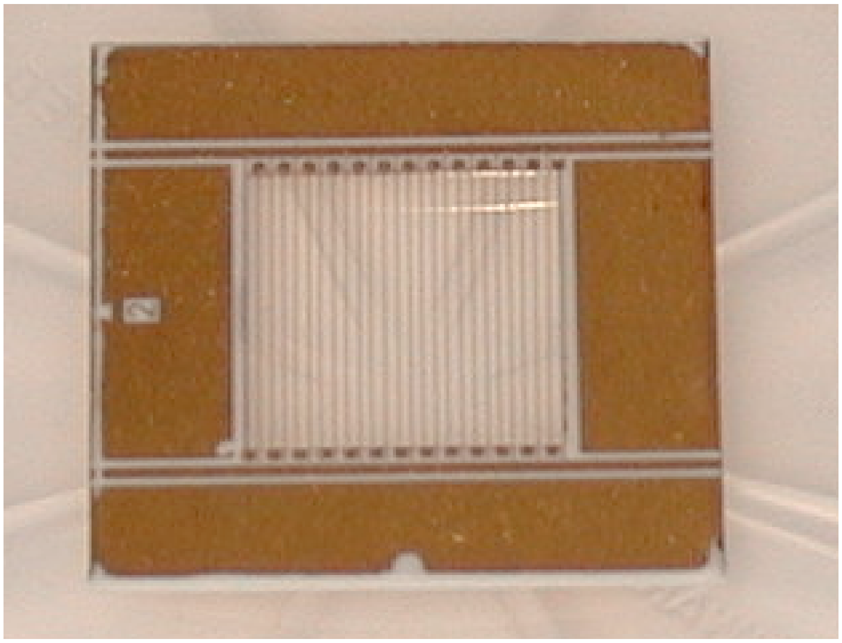


Figure 2. A completed chopper grid, constructed from a 1" square polished alumina substrate, with a 0.5" square hole laser cut from the center. Gold wires, 0.002" dia and spaced 0.020" apart, are wire bonded to pads on one side and the supply line microstrip on the other.

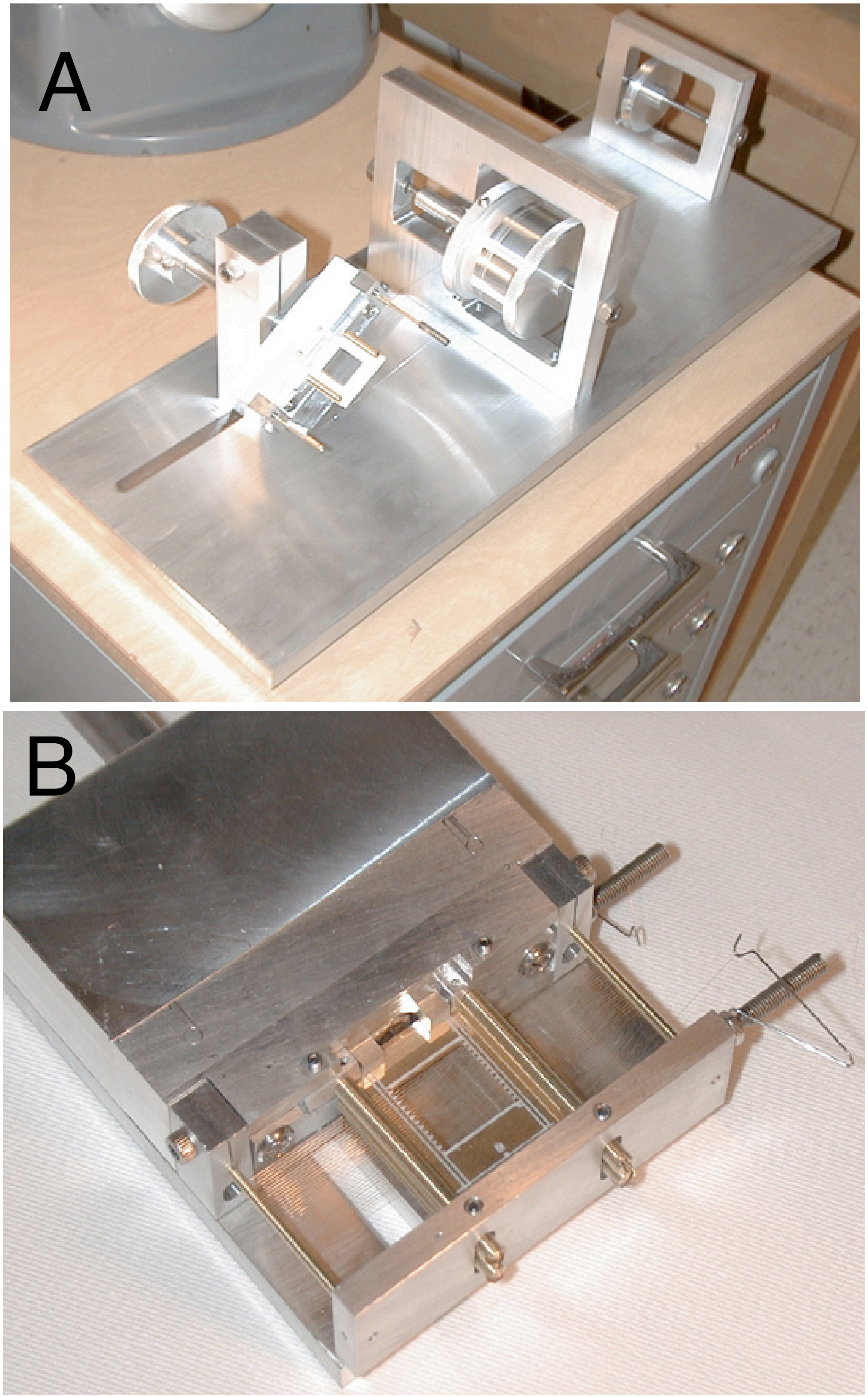


Figure 3. A) Jig and tensioning system designed at LASST for winding wires and alignment during parallel gap welding. B) Ceramic substrate with 0.002” dia wires spaced on 0.02” centers, is shown clamped in holder ready for welding.

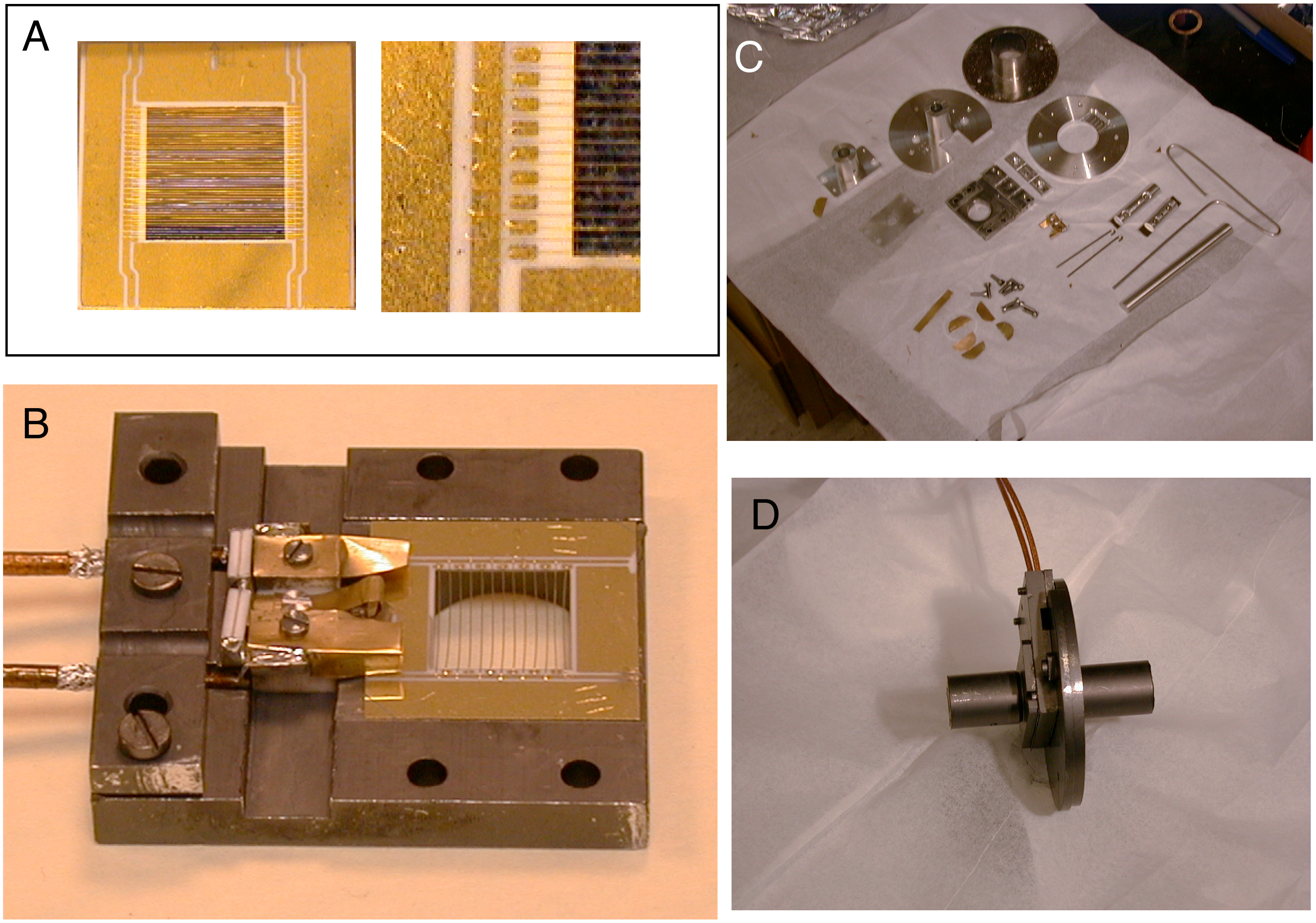


Fig. 4. A) An example of a wire-bonded chopper with 80 wires of 25mm diameter on a 250mm spacing. B) Chopper mounted in socket providing 50 W termination. C) Slit housing parts ready for assembly and D) fully assembled.

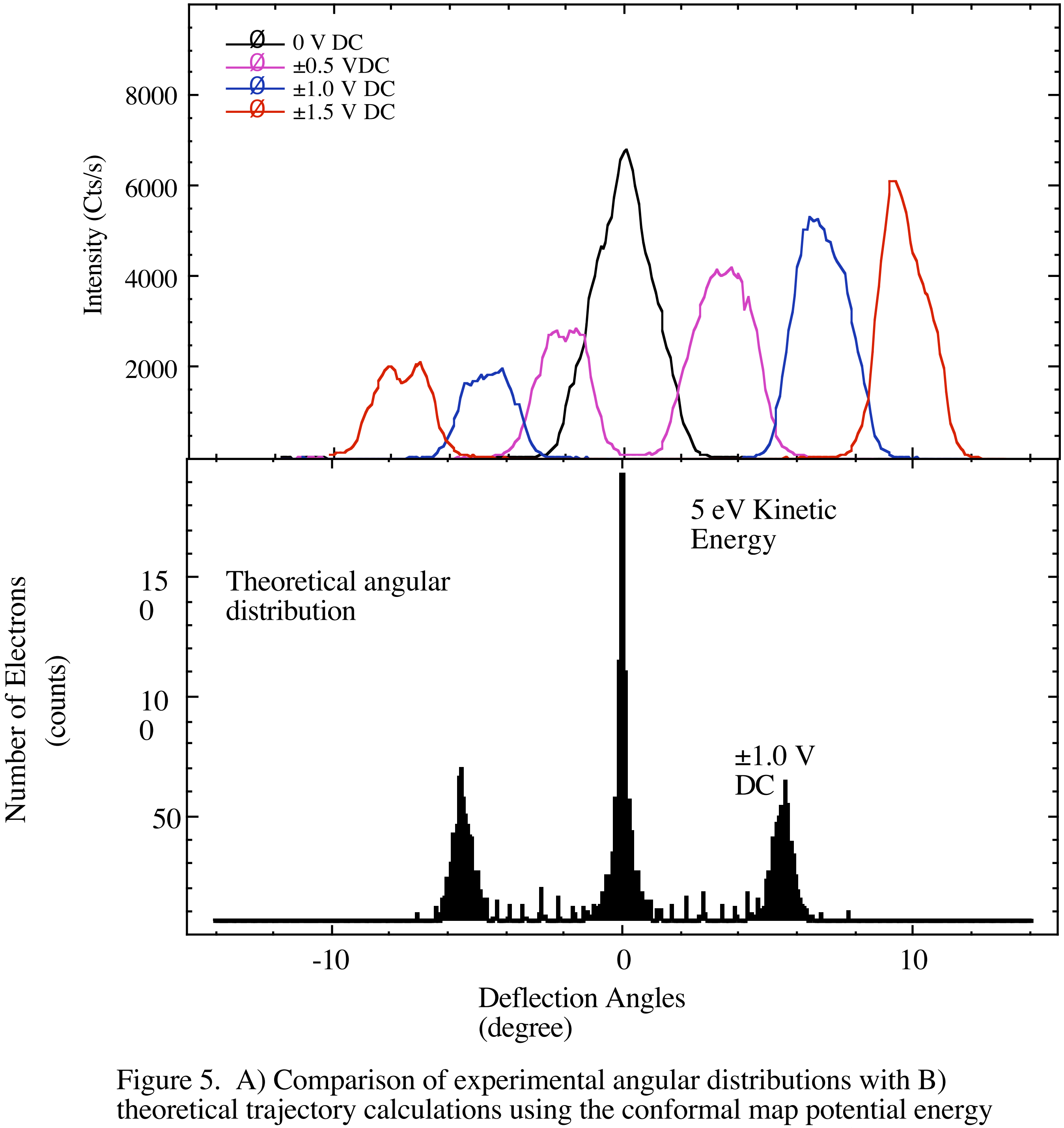


Figure 5. A) Comparison of experimental angular distributions with B) theoretical trajectory calculations using the conformal map potential energy surface (infinite number of wires of finite diameter and infinite length) The broader width of the peaks in the measured distribution (blue curves) is due in part to the angular distribution of the monochromatic beam.

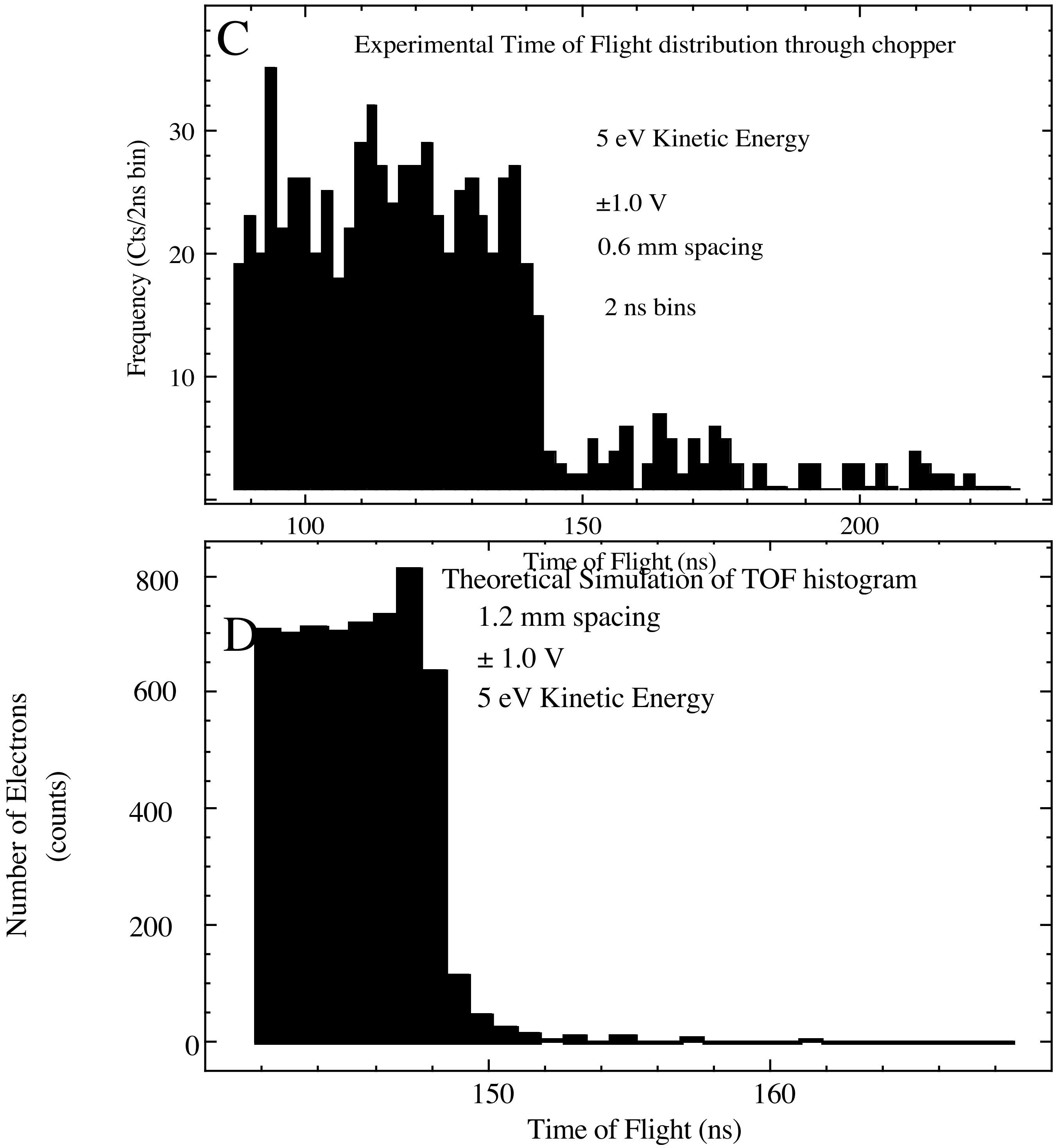


Figure 5. C) Comparison of experimental time of flight distribution with D) theoretical distribution for similar parameters.

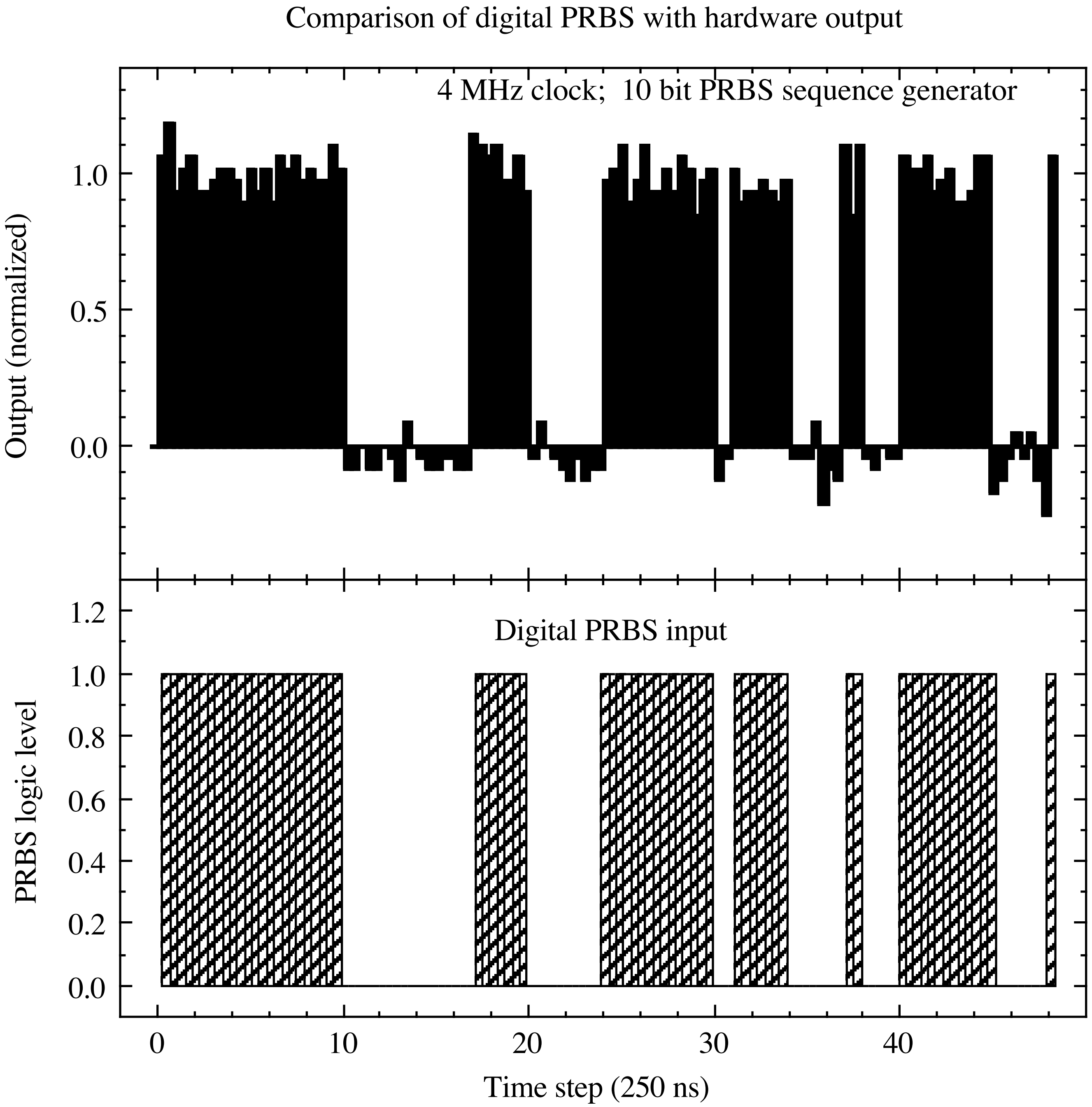


Figure 6. A section of the TOF histogram obtained while modulating the electron beam with the chopper in a 1024 bit pseudo random sequence. The applied sequence is shown for comparison.

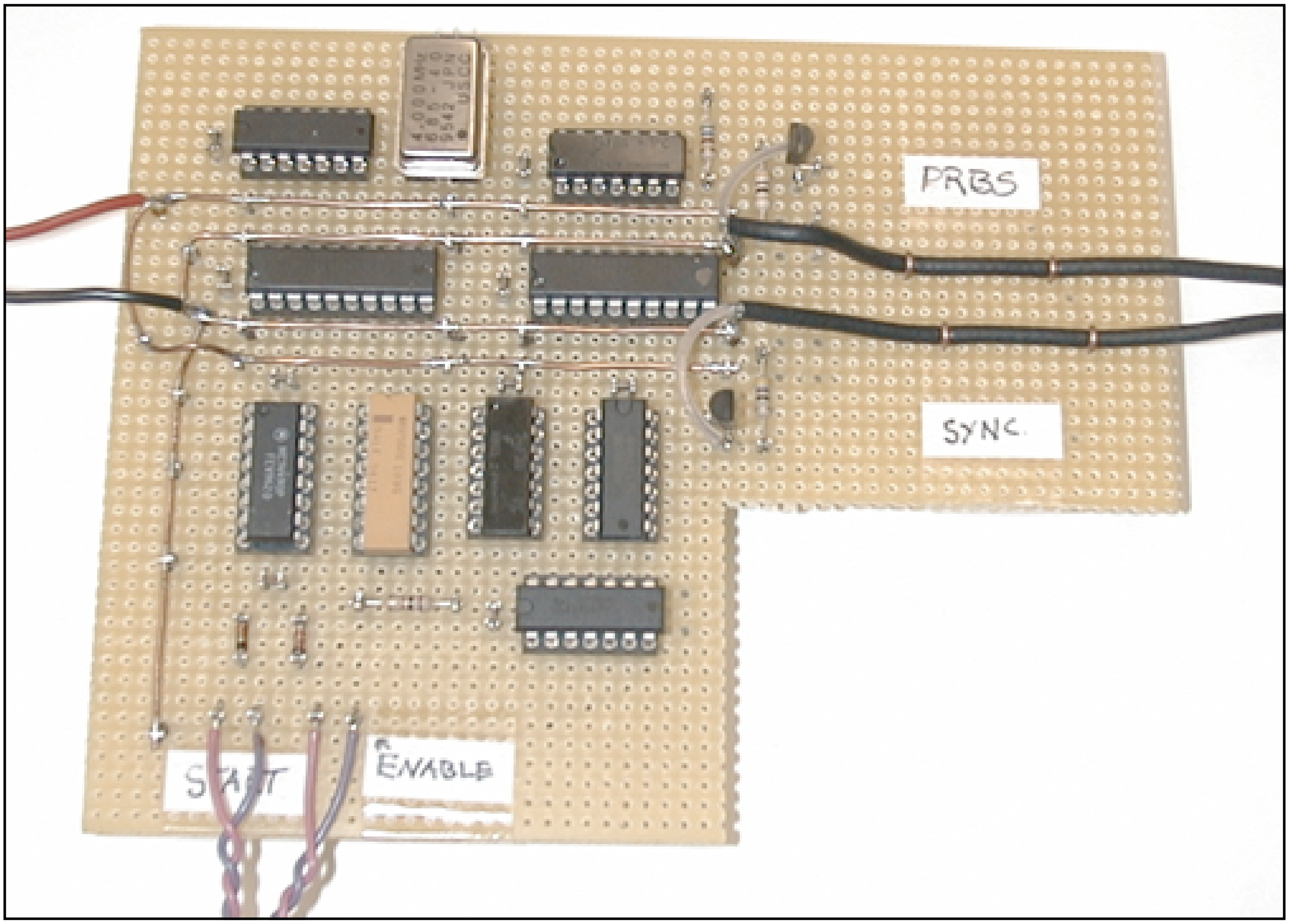


Figure 7. The first generation PRBS sequence generator card constructed using wire wrap techniques.

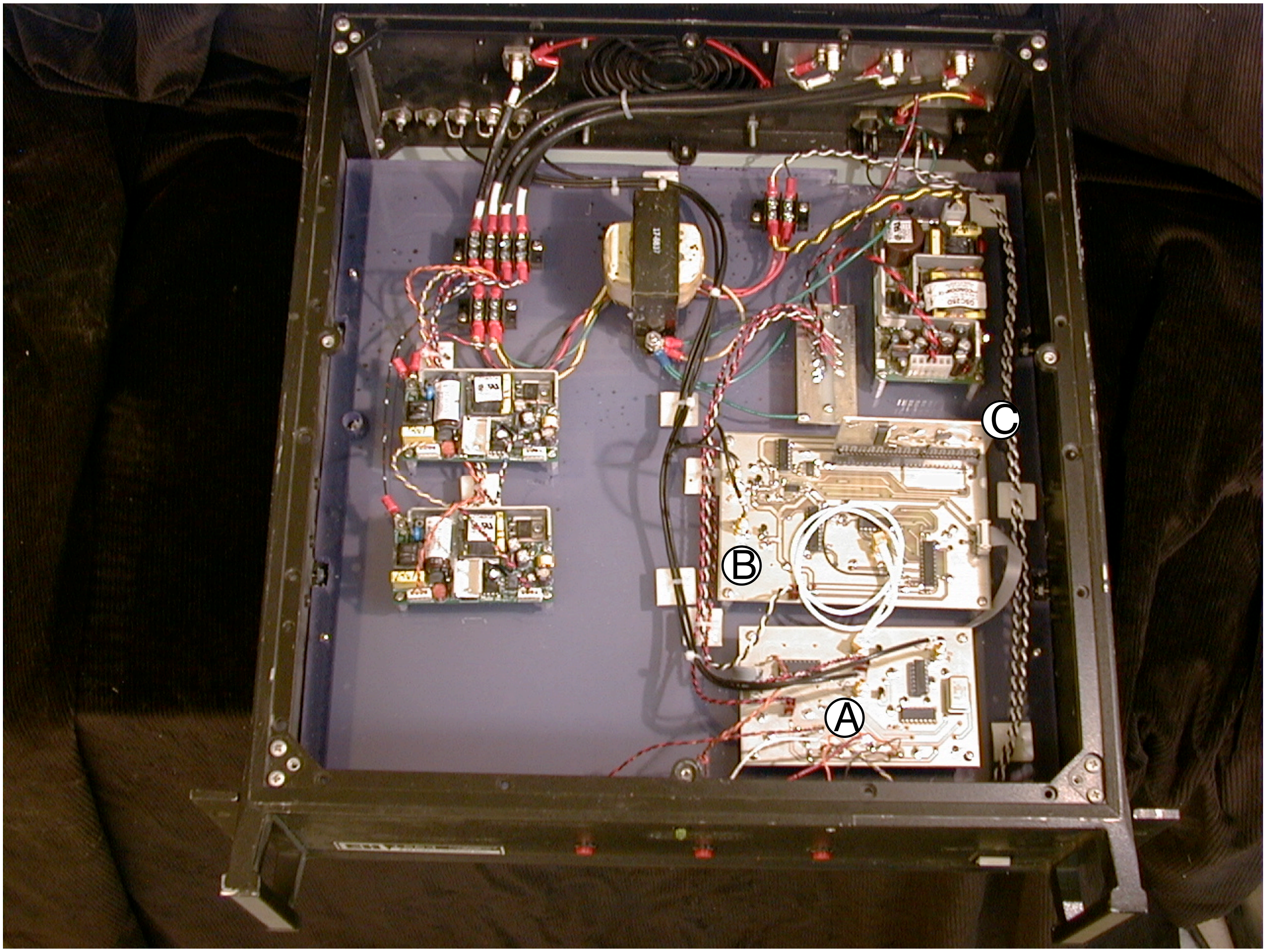


Fig. 8. Microprocessor controlled "smart clock" board (A, lower right) and PRBS generator board (B) with 10-bit sequence card (C) inserted. The unit, including front-panel controls and power supplies for the output driver card, is based upon FAST-TTL and is designed to operate at 10 MHz.

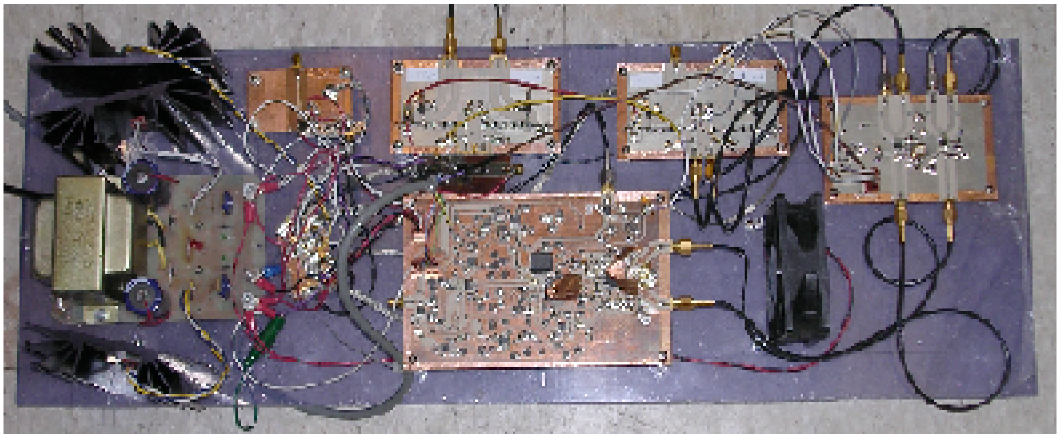


Fig. 9. The 125 MHz ECL based PRBS generator board.

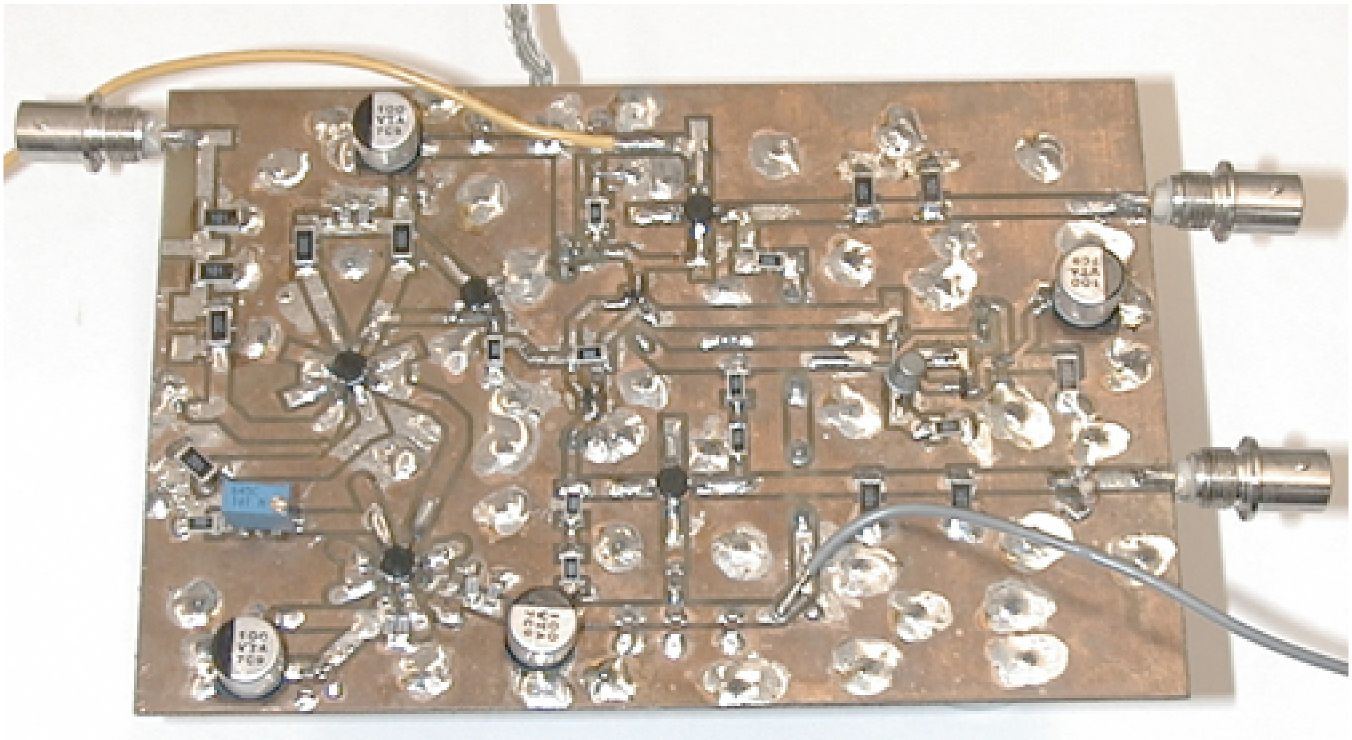


Figure 10. First generation driver output card, achieving 2 ns rise time.

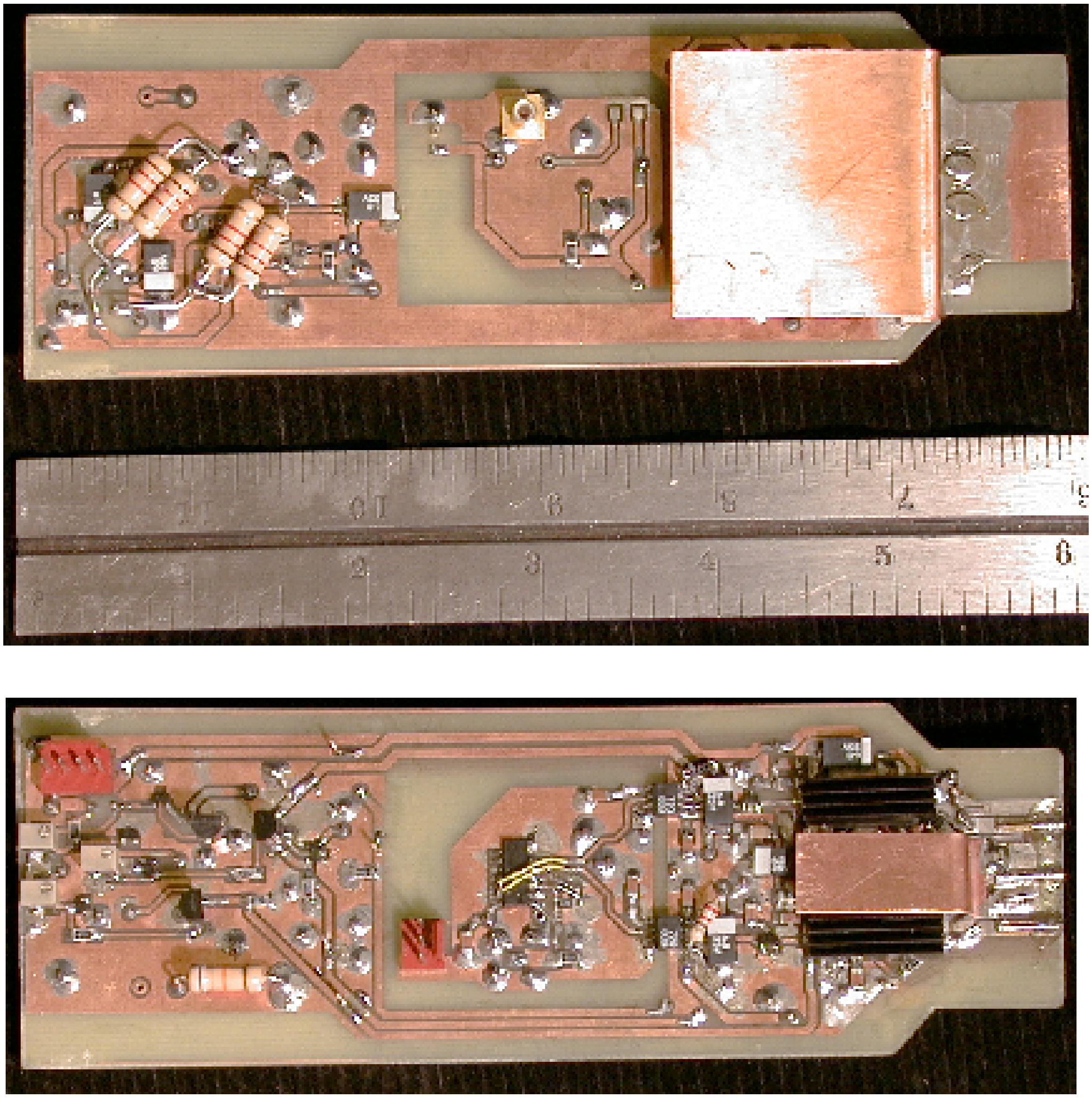


Fig. 11. Top and bottom side of output driver card designed for a single-ended input, optically isolated to allow the chopper and flight tube to be biased at up to 700V.

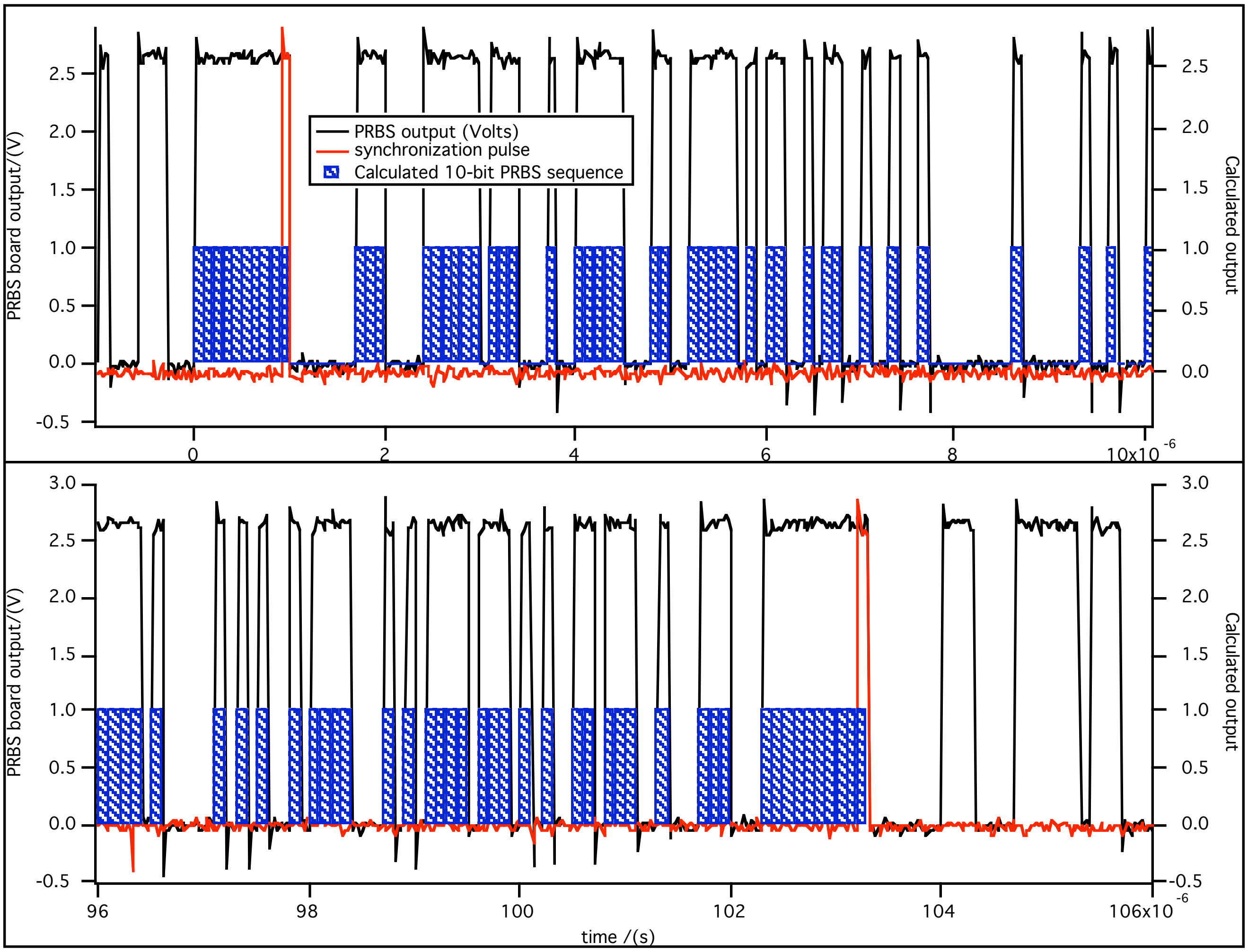


Fig. 12. A comparison of sections at the beginning and end of the PRBS sequence between the ideal and measured PRBS Generator card output is shown below. The rollover occurs at the end of the set of 10 1's. The driver output, measured with a 1 GHz bandwidth DSO, illustrates the time-dependence of the positive and negative going gate driver signals.

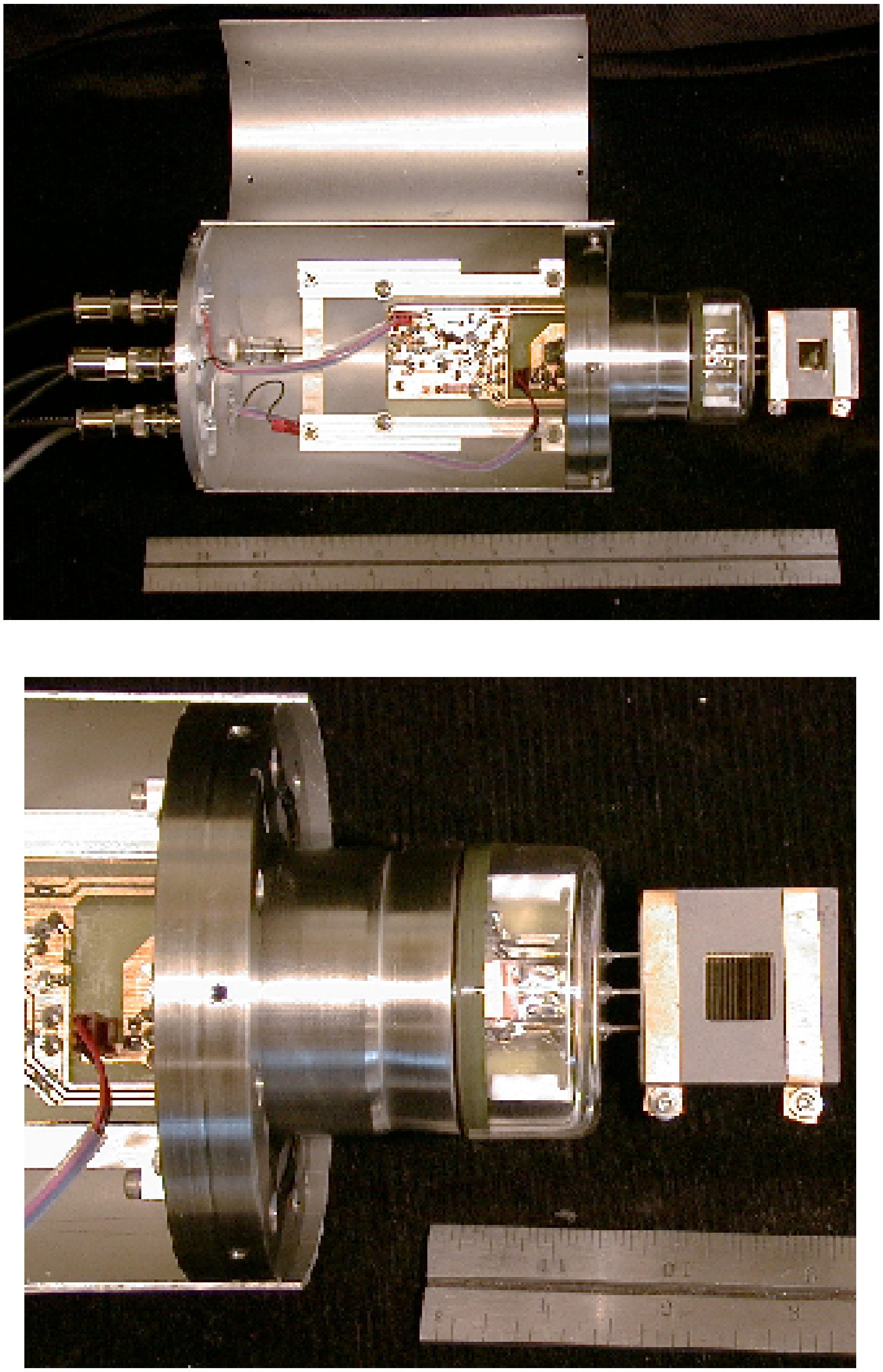


Fig. 13. Custom designed, inverted glass feedthrough allowing the output driver card to be mounted in close proximity to the chopper.

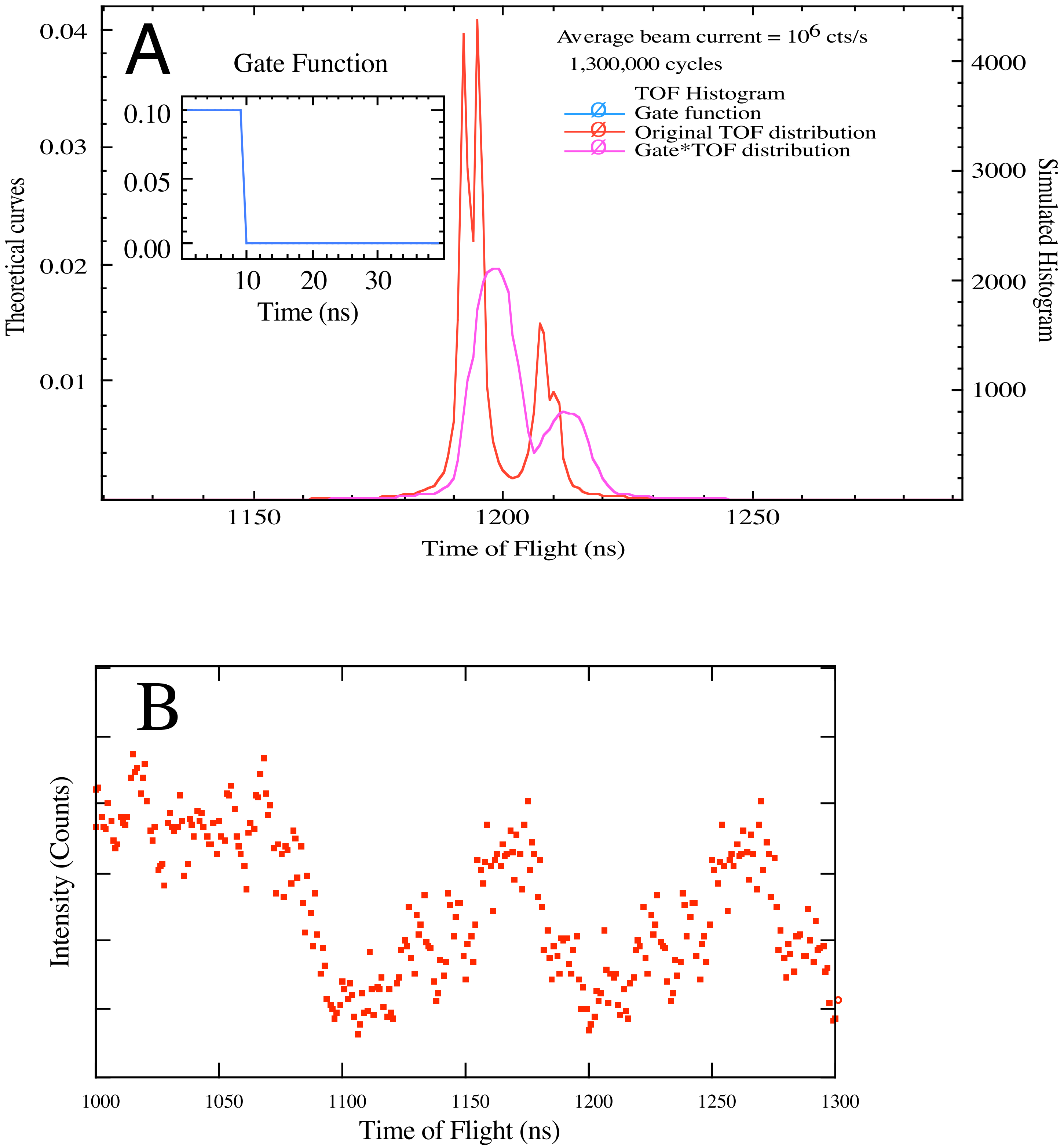


Figure 14. Example of output of the Monte Carlo simulation of the instrument. A) The underlying energy distribution is represented by the “Original TOF distribution”, which when convoluted with the Gate Function, gives the purple curve. The simulator produces a TOF spectrum with a Poisson noise distribution (dots). B) A section of the corresponding TOF distribution when the gate function uses a PRBS for modulation.

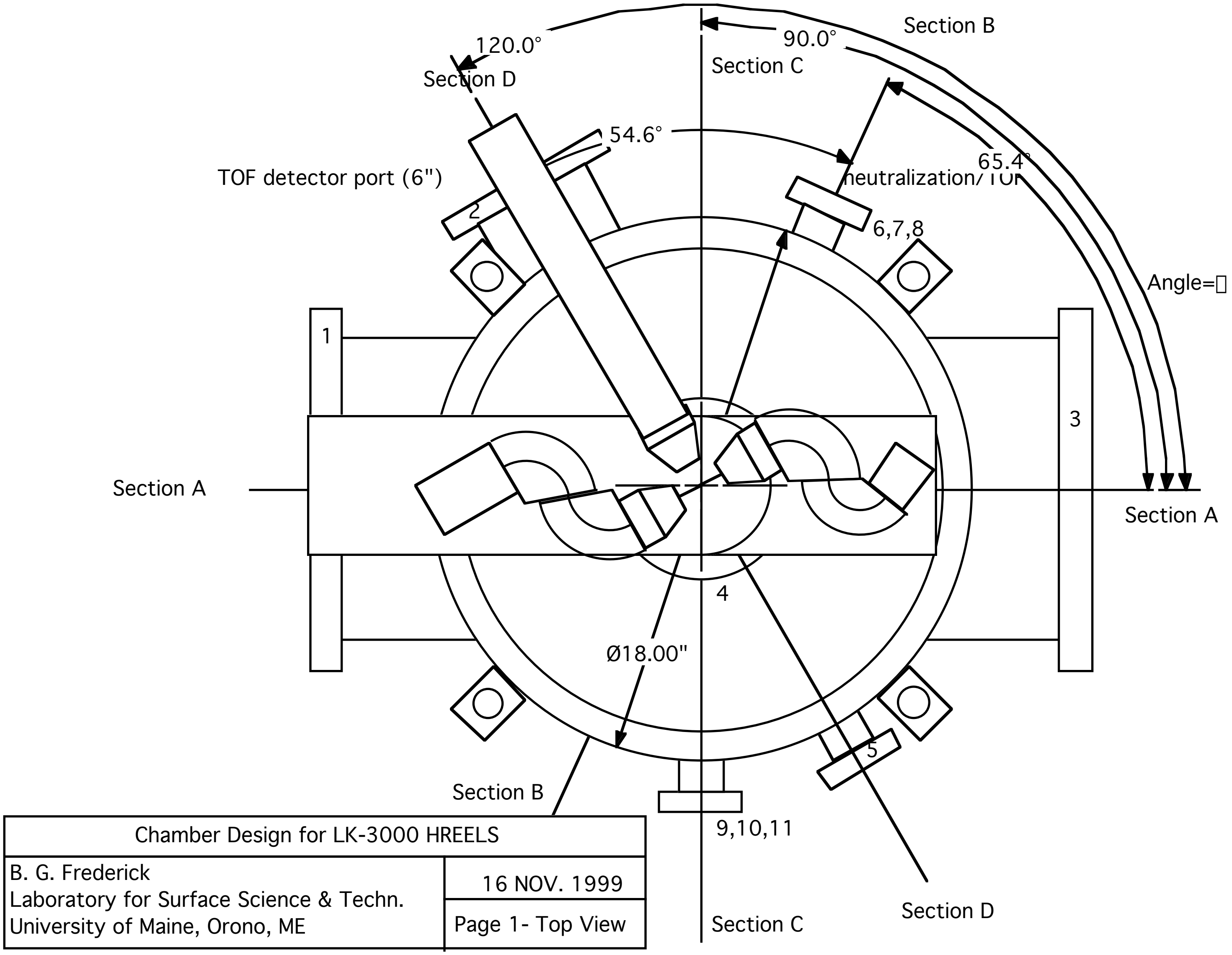
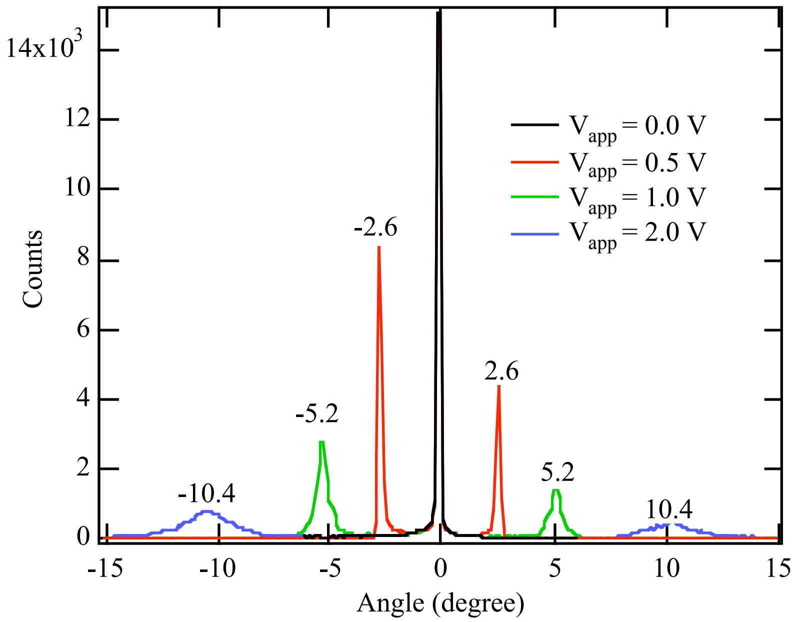


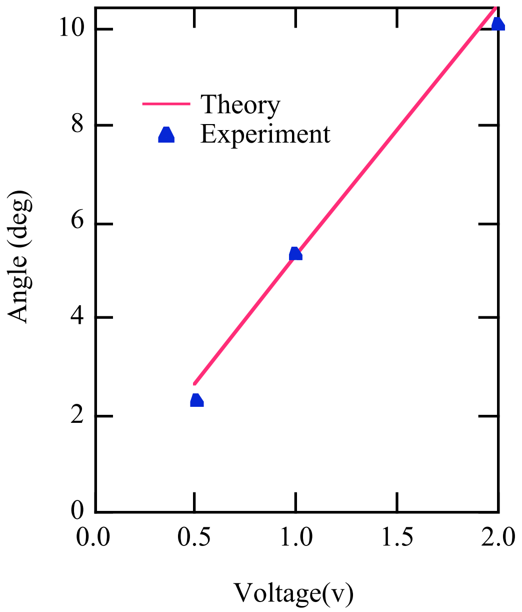
Figure 15. Top view schematic drawing of the new HREELS chamber showing conventional double-pass monochromator and analyzer and flight tube of the TOF electron detector.

Angular Distribution: Simulation vs. Expt

Trajectory calculations using our model potential show good agreement with experiment for the deflection as a function of applied voltage, V_{app}:



The ratio of the intensity of the peaks at ±□ depends on the fraction of the beam passing between (+V/-V) and (-V/+V) wire pairs.



Data shown for a 5 eV beam.

Fig. 16. Comparison of angular distributions predicted from trajectory calculations and measured experimentally.

TOF Histogram Simulation

Calculated time-dependent response as a function of applied voltage, V_{app} (a) 0.5 V, (b) 1.0 V and (c) 2.0 V; and (d,e,and f) predictions for smaller wire spacing at constant transmission (1-2R/d) and applied voltage V_{app} = 1.0 V.

The spikes and the time lag effects increase with applied voltage and large wire spacing, similar to the experimental results.

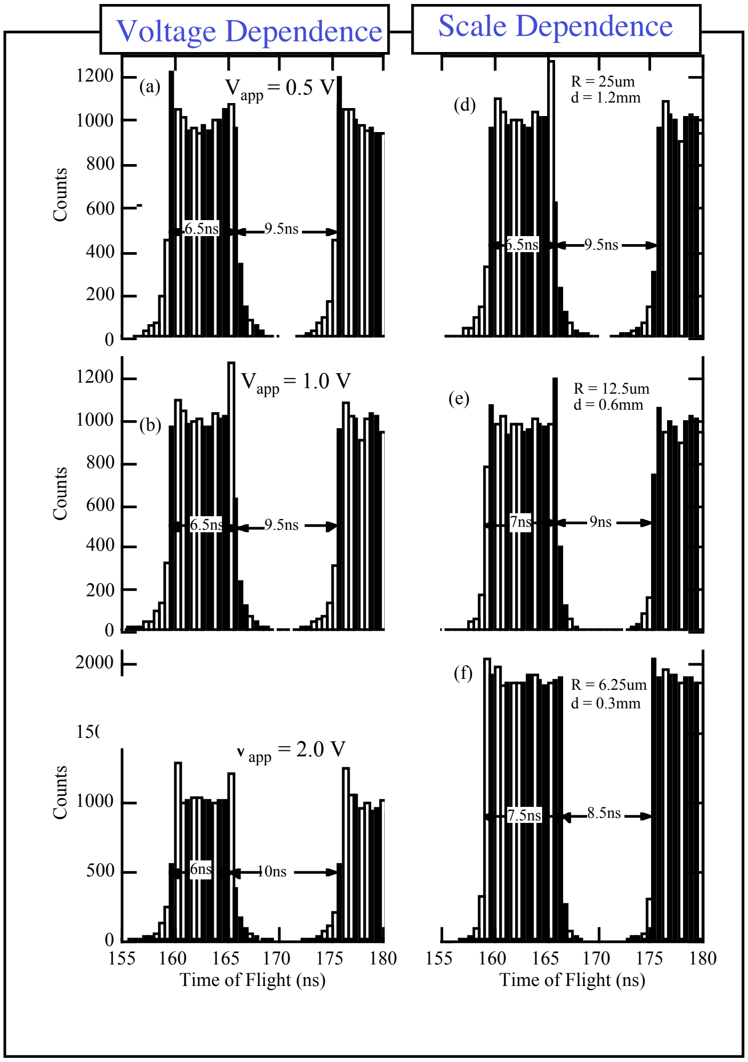


Fig. 17. Time-dependent response from trajectory calculations as a function of applied voltage and wire spacing at constant transmission.

Closing the gate

(Turning on the voltages)

Contour map of potential,□, is shown with critical boundaries, $f(x,y,\square_{acc},V_{app})$, (colored curves) as a function of the acceptance angle and applied voltage.

For an electron flying upward, if the electron is beyond the curve at the time when the potential is applied, the electron achieves an angle $\square_f < \square_{acc}$; i.e. the electron reaches the detector.

The chopper can close early or late. For example, if $\square_{acc}$= 1°/$V_{app,}$ for an electron to be detected, it has to be beyond the red curve when the potentials are applied, so the chopper appears to close early.

Switching the potential also leads to “energy corruption”. The total energy change is the change in the potential at the position of the electron when the voltages are switched. Statistical corruption distributions can be determined from the potential.

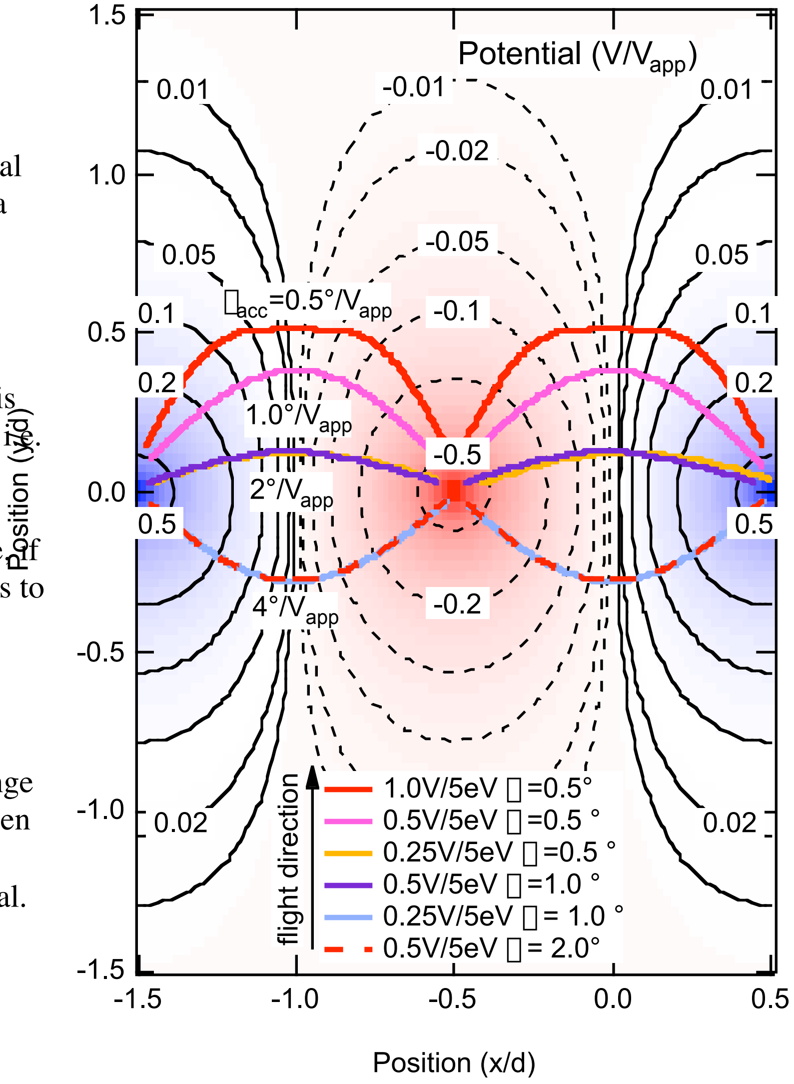


Fig. 18. Relationship between the acceptance angle and the spatial location of the electron when the gate is “opened”. The potential (overlaid) is used to determine the statistical distribution of energy corruption effects.

Energy Corruption Distributions

These energy corruption histograms were calculated for a 2 eV electron passing a chopper with dimensions 1.2mm spacing and 50 μm diameter, acceptance angle 1° and $V_{app} = 0.4V$. The energy corruption histograms represent temporal/spatial regions corresponding to each 0.5 ns before or after the potential has been changed. The magnitude of corruption is greatest initially (curve a) and decreases with time.

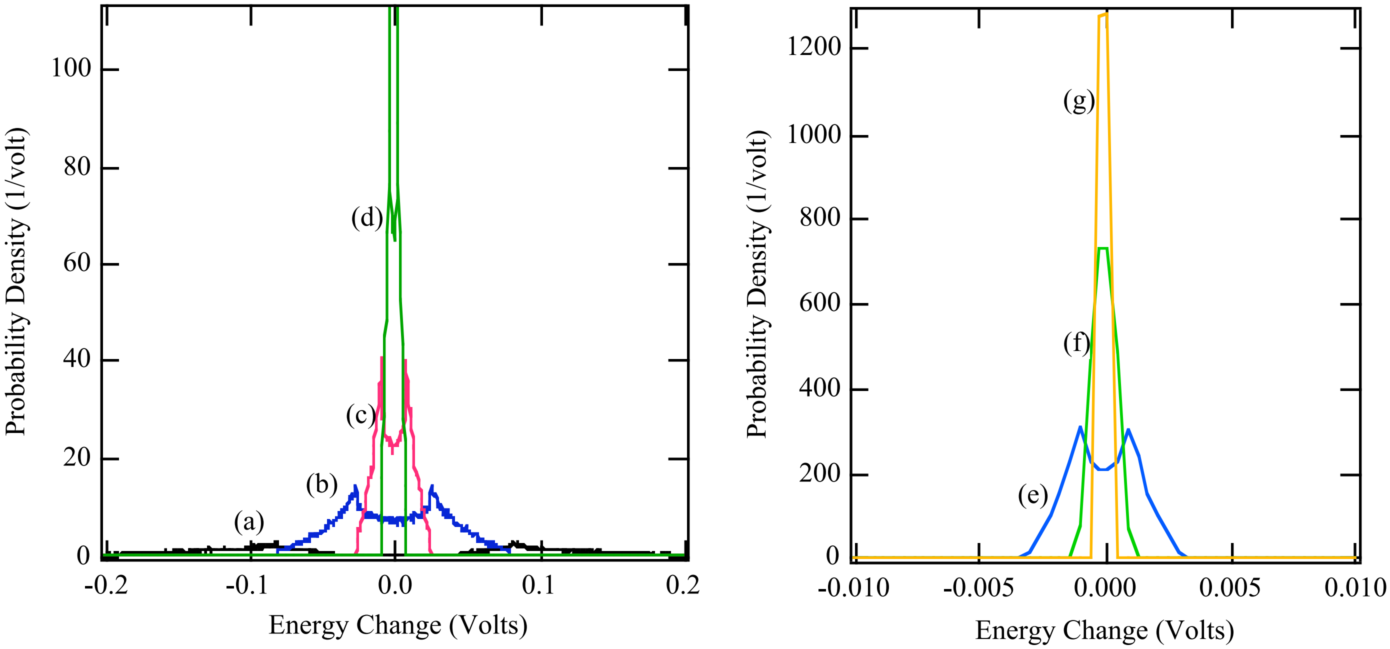


Fig. 19. Energy corruption histograms as a function of time (or distance) from the chopper when the potential is switched, which are used in statistical simulation of instrument operation.

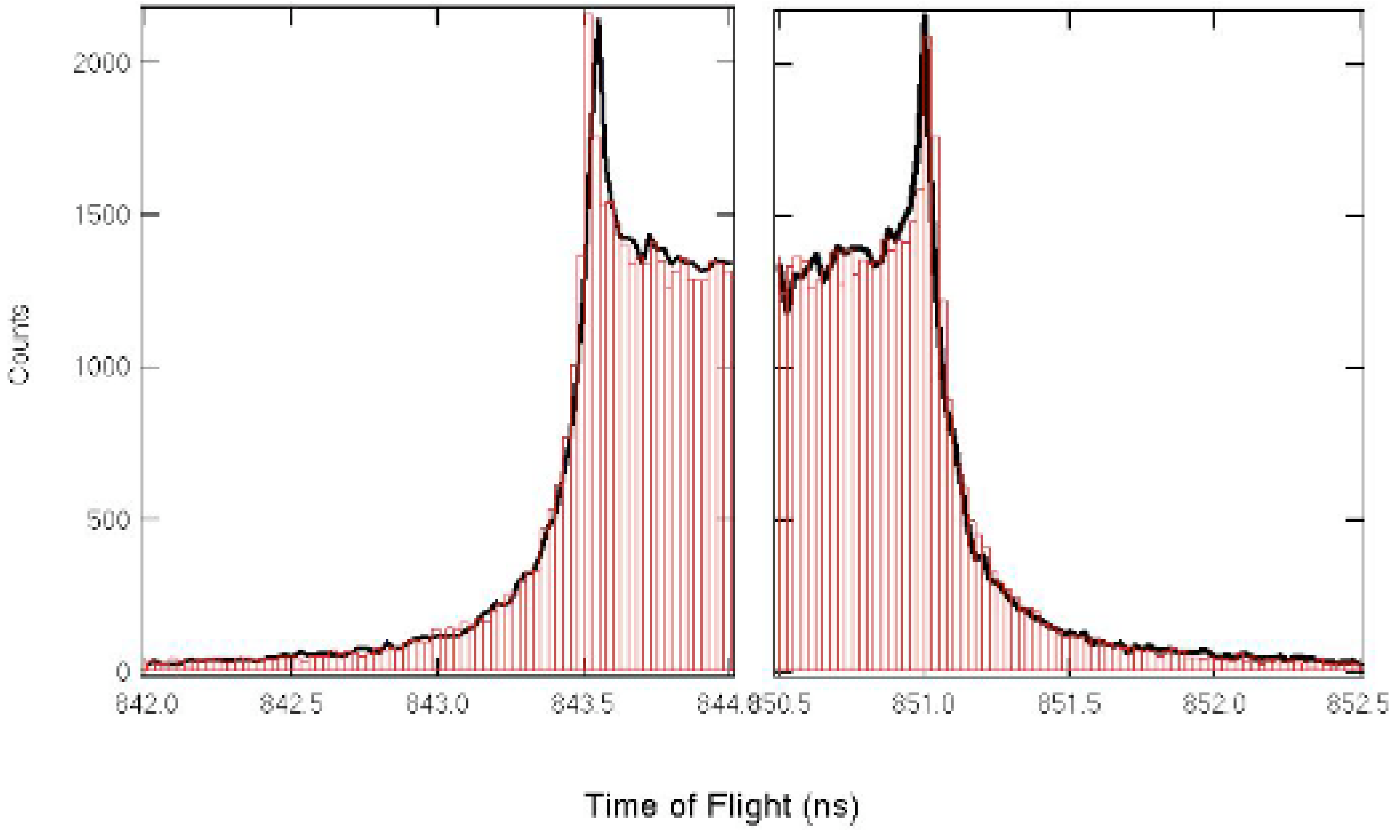
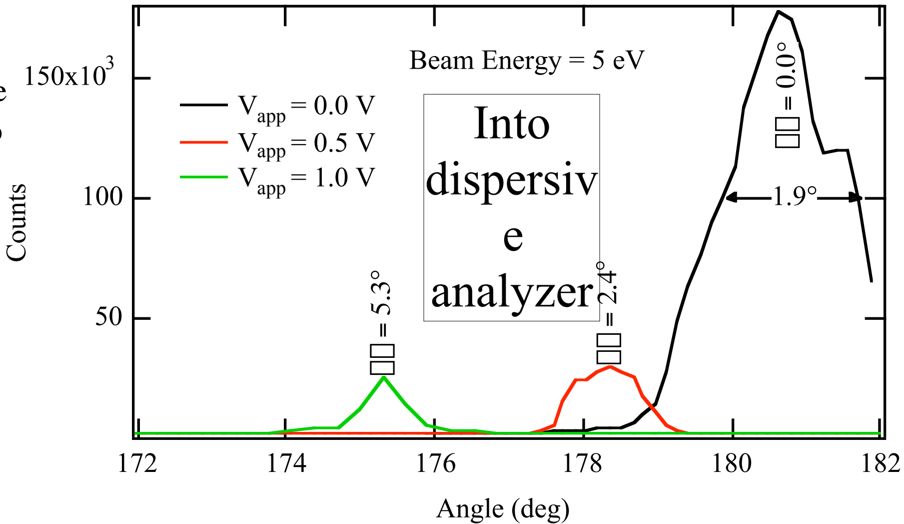


Fig. 20. **Comparison of the shaper of the A) rising edge and B) falling edge of a single pulse calculated with the Monte Carlo method on a 25 ps time bin size for a kinetic energy of 4 eV (bars) and 3.6 eV (solid curves) to show that the energy dependence of the shape of the response function is negligible under the conditions of this experiment. This simulation is based on a chopper size of $d = 250\ \mu m$, $V_{app} = \pm 0.4\ V$ and $\theta_{acc} = \pm 1°$.**

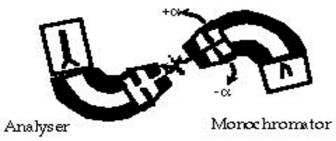
We measured the deflection angle of the beam as a function of applied voltage to compare with theoretical calculations and to determine the depth of modulation of the electron beam.

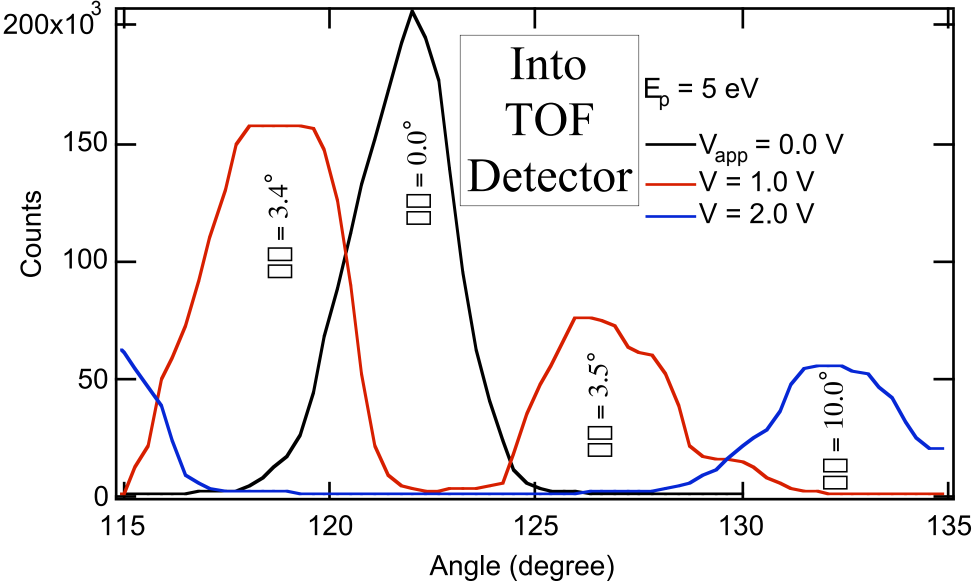
For a chopper with 96% transmission, we find that the deflection angle is approximately linear in the ratio of the applied voltage, V_{app}, to the electron's kinetic energy, with the dependence:

$$\theta_{defl} = 26 \left(\frac{eV_{app}}{KE} \right)$$



Into dispersive analyzer:





Into TOF Detector:

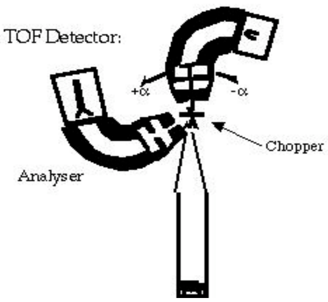


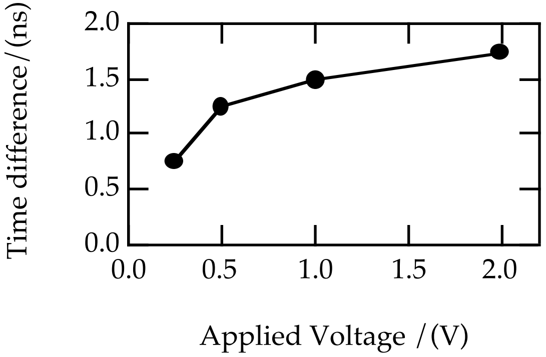
Fig. 21. Experimental measurements of angular distributions.

The Time of Flight (TOF) Spectrum

The following results characterize the ability of the chopper to open and transmit a pulse of 5 eV electrons. The voltages are held at ±V_{app} and dropped to zero periodically for 100 ns. The flight times of detected electrons are measured relative to a synchronization pulse with a digital oscilloscope and are shown in a histogram with 250 ps bins.

The data below shows that the gate can be opened with rise times in the sub-nanosecond regime. However,there are two types of artifacts:

- The time that the beam is on is less than the time that the voltages are off
- There are spikes and tails in the TOF spectrum when the voltages are changed



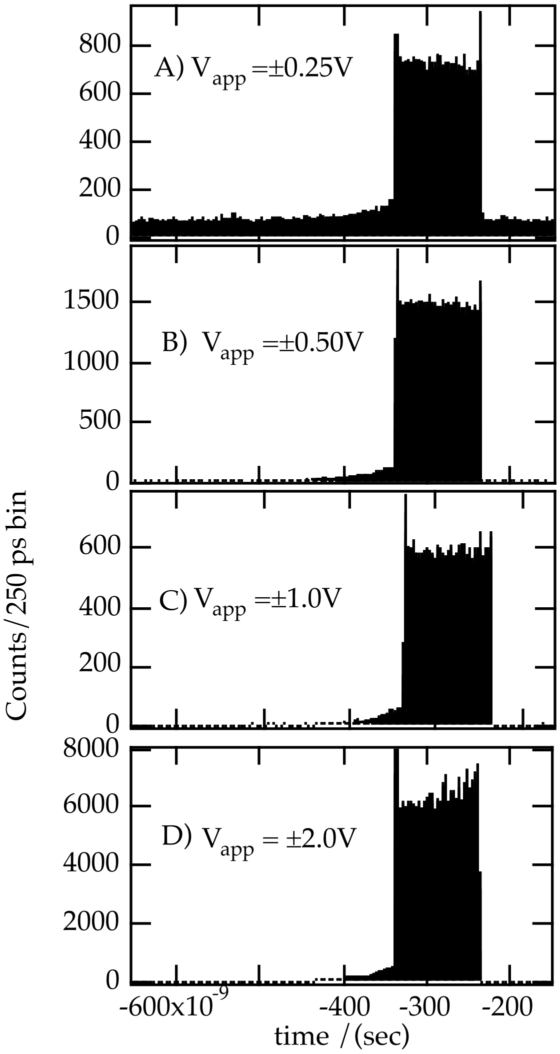


Fig. 22. Experimental measurements of the time-dependent response of the largest spacing (100μm wire diameter) chopper as a function of applied voltage.

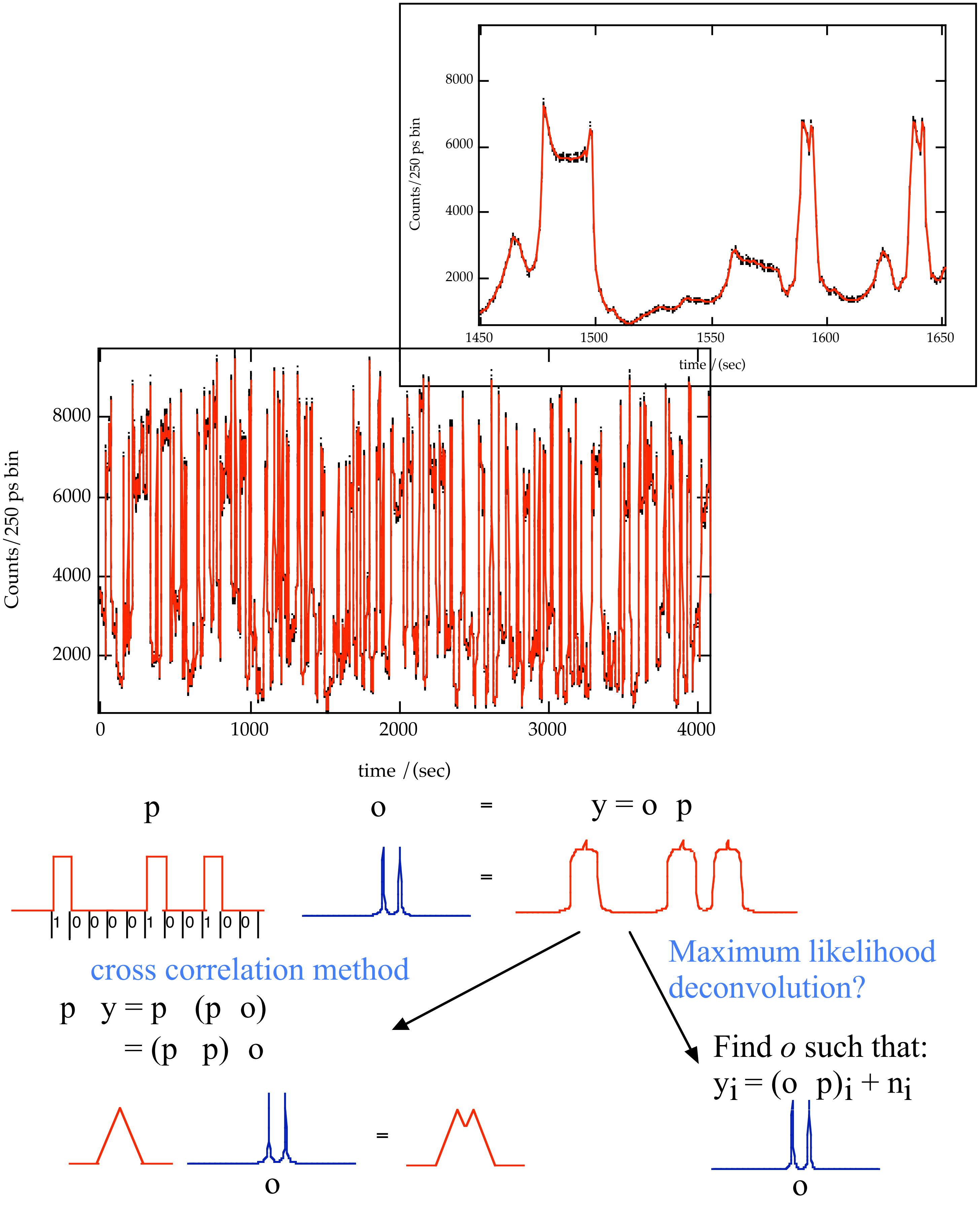


Fig. 23. Simulated data and methods of recovering the PRBS modulated data.

Effect of Poisson noise
Throughput advantage of PRBS

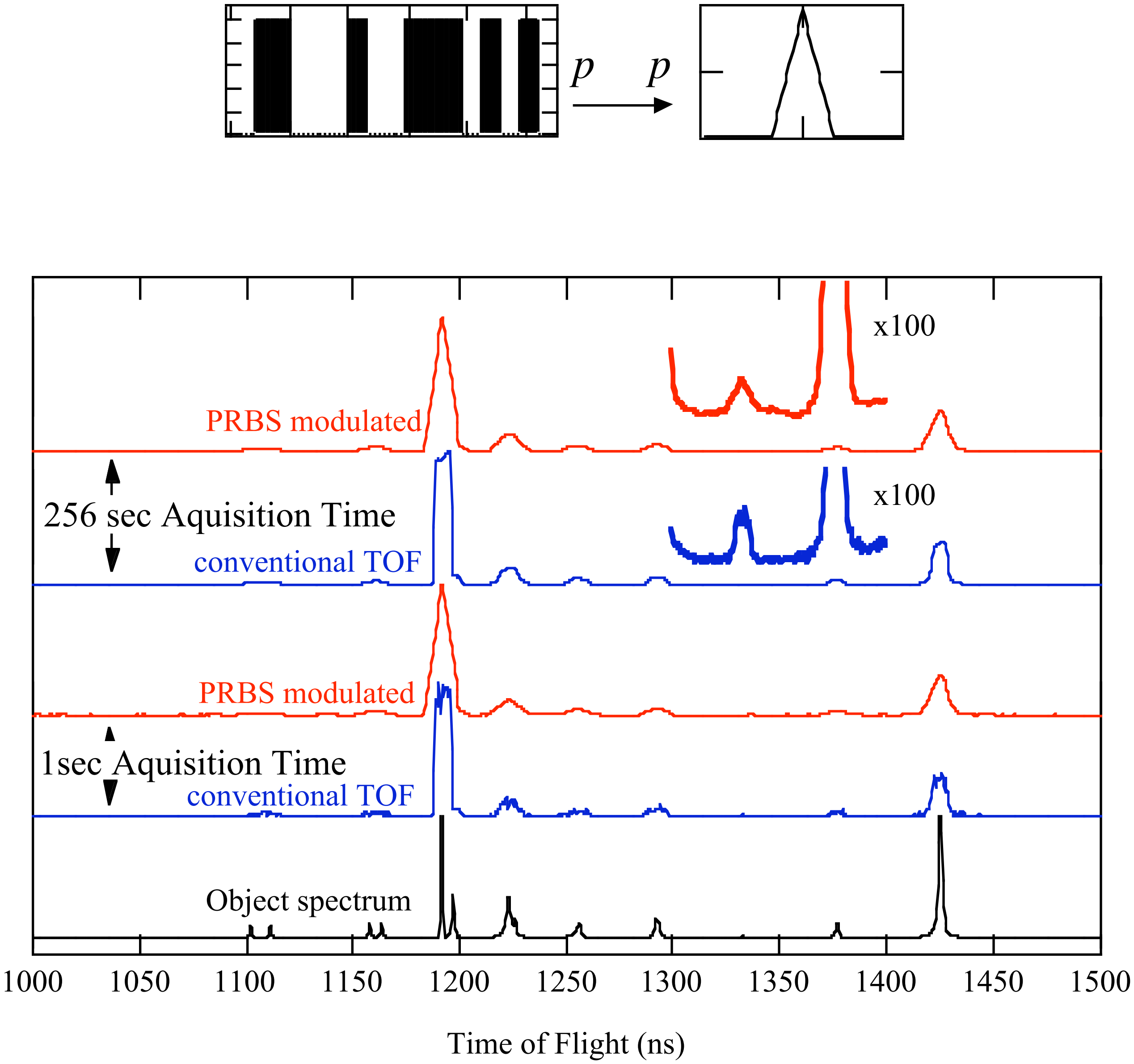
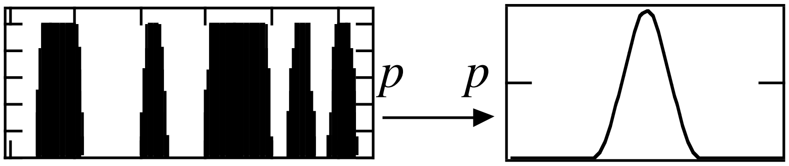


Fig. 24. Effect of the Poisson noise distribution on the standard cross correlation analysis compared to a conventional TOF experiment. The throughput advantage is clear from comparison of the signal to noise level, particularly in the feature at 1330 ns with intensity 0.1% of the elastic peak (at 1190 ns). Although strictly not valid when the noise power correlates with the signal power, the artifacts due to the Poisson distribution are relatively small compared to the magnitude of typical loss features in HREELS.

- PRBS with linear rise/fall with 50% duty cycle
- Resolution enhancement depends on number of iterations and total counts (S/N)



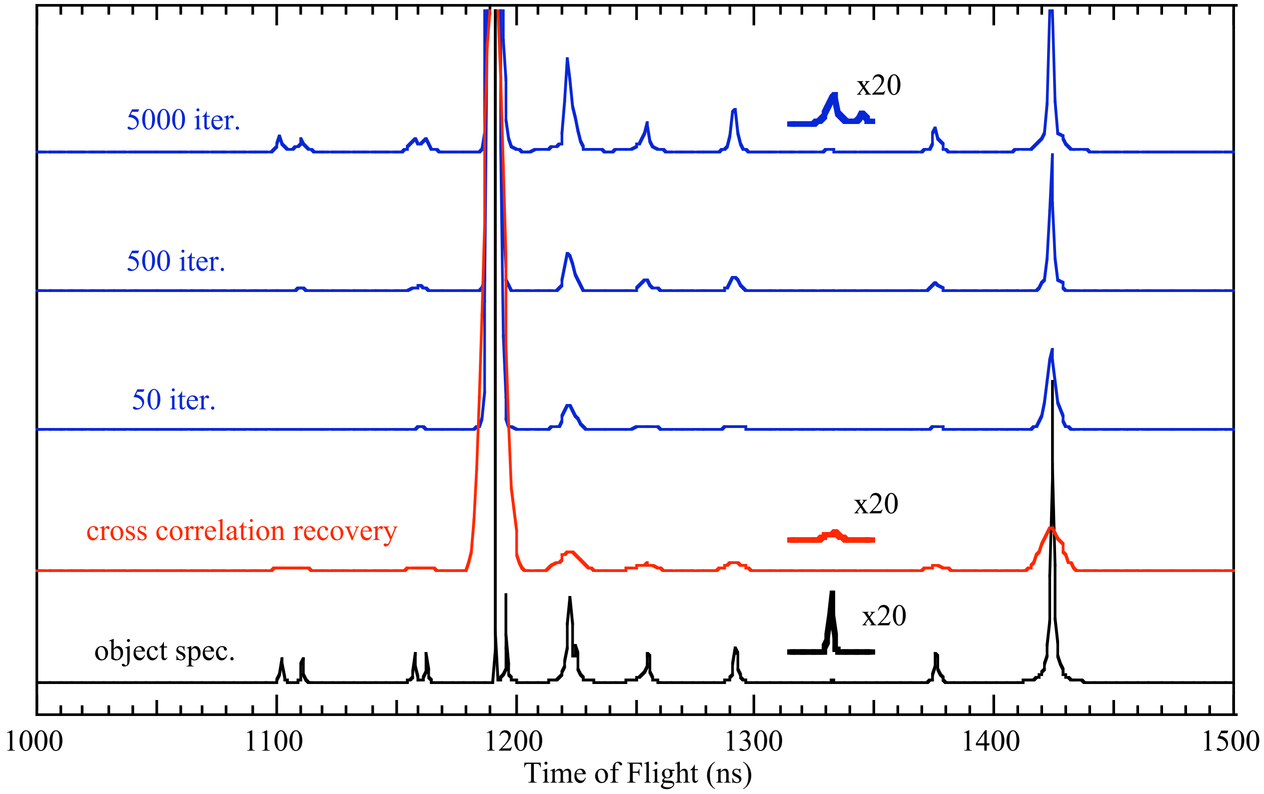


Fig. 25. Performance of the maximum likelihood algorithm in recovering the underlying TOF spectrum for the case of a PRBS sequence with finite (1ns) rise and fall times and the inherent Poisson noise distribution. The cross correlation recovery is used as the initial estimate; further iterative processing leads to improved resolution. The 1 ns sampling rate (oversampling by 8x) limited the achievable resolution in this case.

- Cross correlation yields artifacts
- ML with Ideal PRBS also gives artifacts
- ML with actual PRBS is excellent

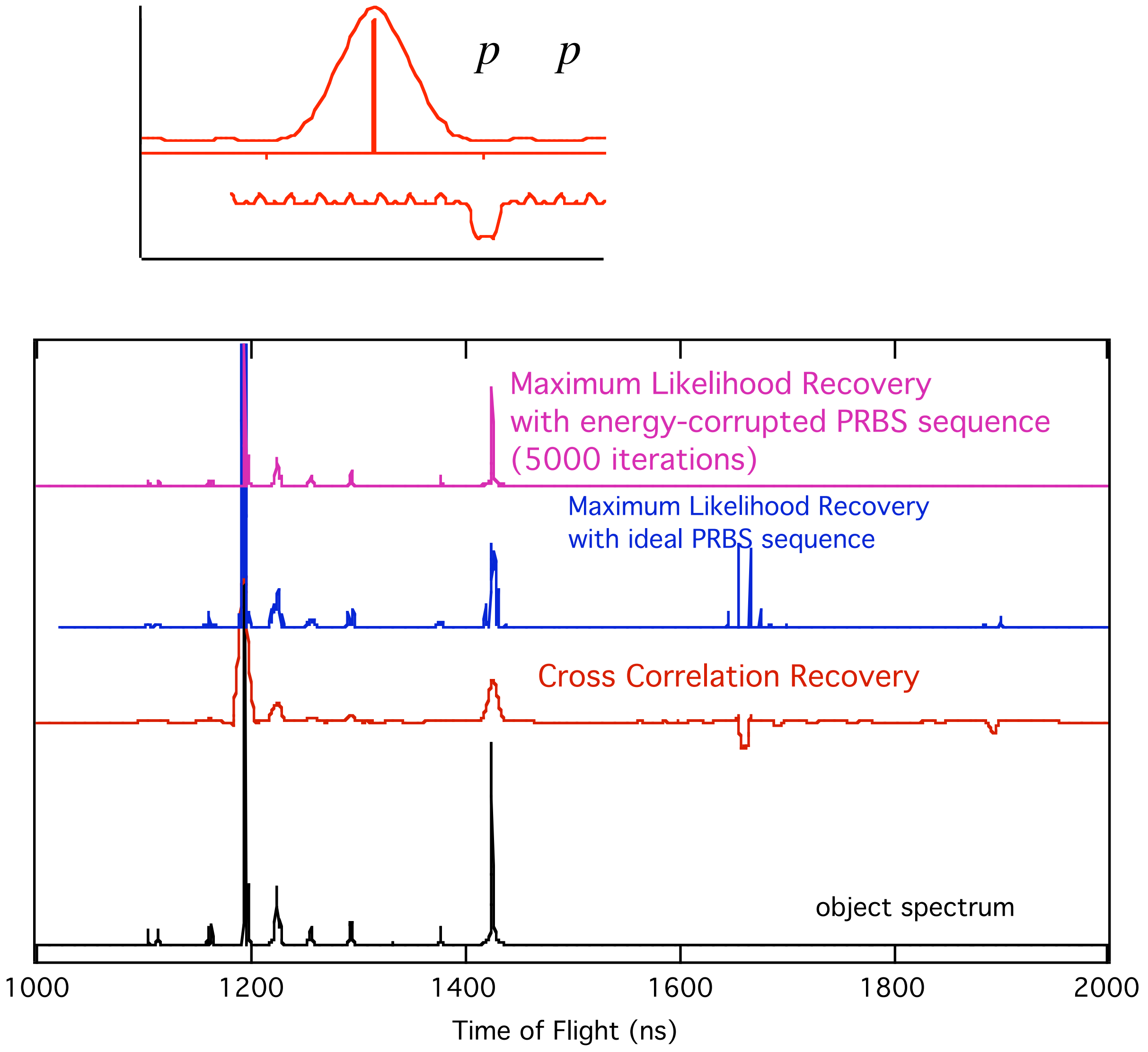


Fig. 26. Performance of the maximum likelihood algorithm in recovering the underlying TOF spectrum for the case of a PRBS sequence with rise and fall times, lead and lag effects, and the energy corruption effects obtained from the trajectory calculations for a 500 μm wire spacing 50 μm diameter chopper, as well as the inherent Poisson noise distribution. The cross correlation recovery, used as the initial estimate contains large artifacts as expected from inspection of the autocorrelation function (upper right corner). Iterative processing with the ideal PRBS sequence also leads to artifacts (middle curve). By using the actual chopper response function, however, the TOF spectrum is recovered (top curve) with resolution improved by a factor of 6-8 over the cross correlation analysis while avoiding artifacts. Peaks at a level of 0.1% are reproduced with a signal to artifact ratio of approximately 10.

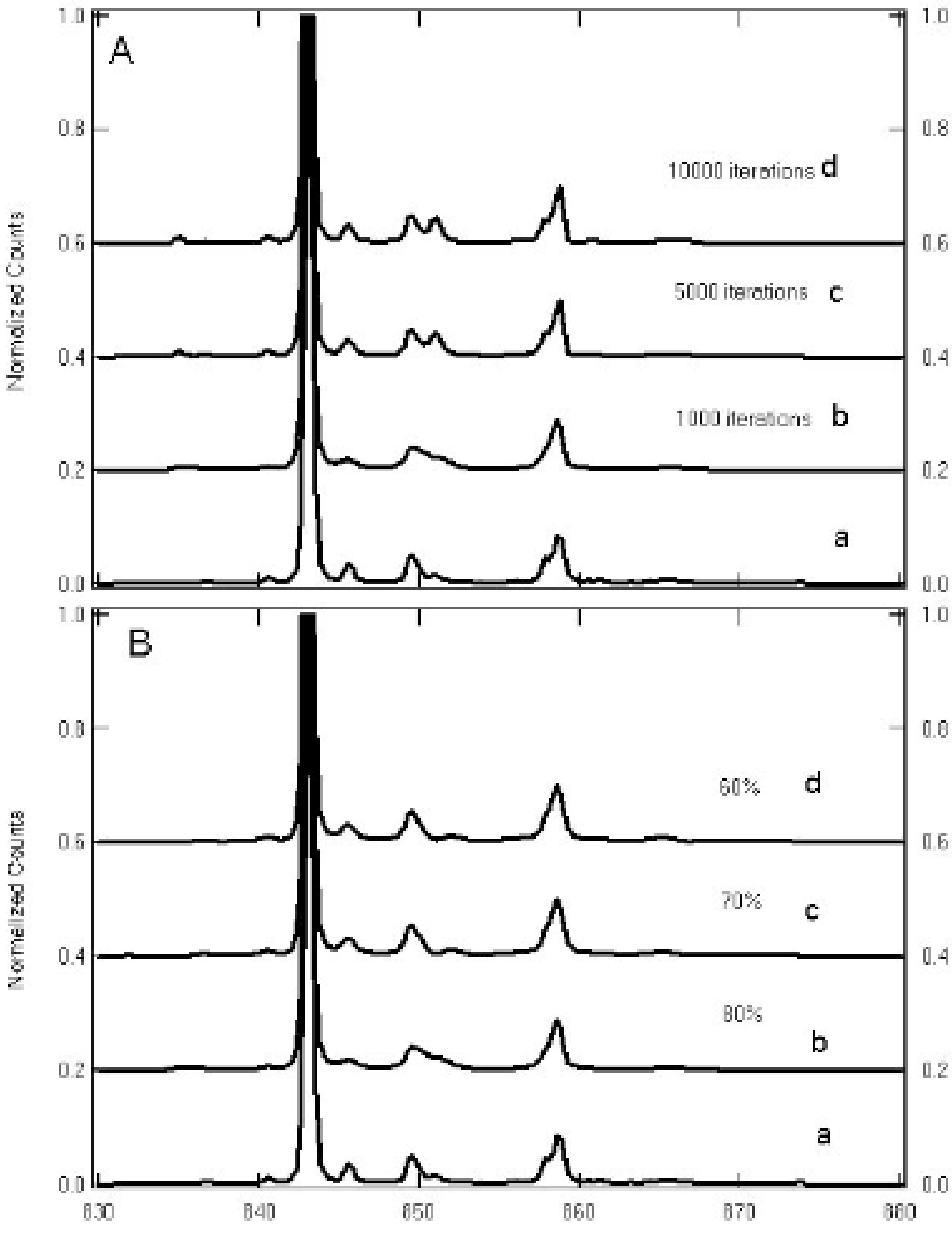


Fig. 27. A) Comparison of a) the object function used to generate noise-free modulated data by convolution with a PRBS response function that is ideal except that the height of single 1’s is reduced to 80%, with recovered spectra using the Lucy algorithm after b) 1000 iterations, c) 5000 iterations, and d) 10,000 iterations. Notice that loss of information results in artifacts and re-distribution of the peak intensities. B) Results after 1000 iterations for data generated as in A) but for the height of single 1’s reduced to b) 80%, c) 70%, and d) 60%.

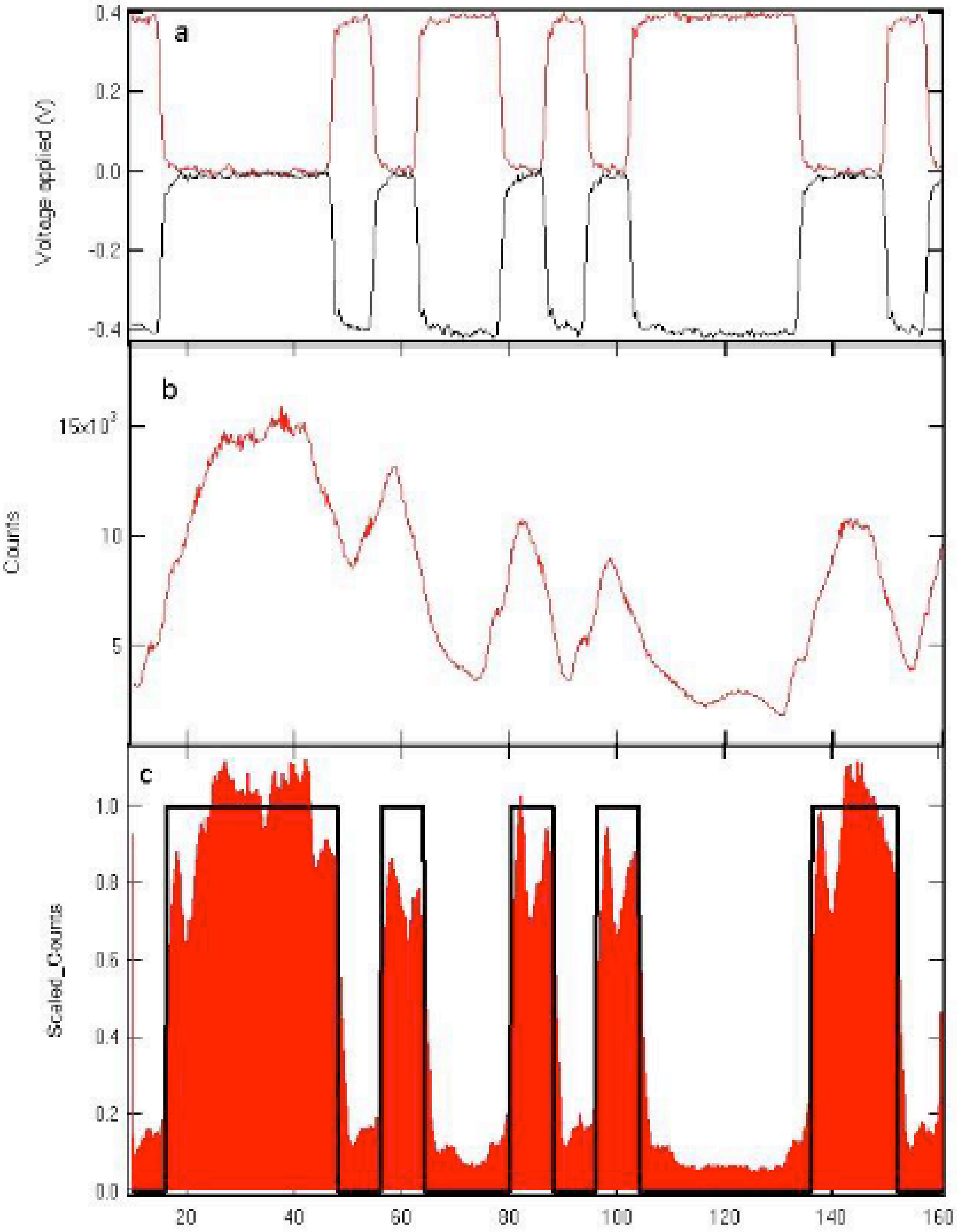


Fig. 28. A section of the 255-bit PRBS modulation function showing a) the time dependence of the voltages applied to the chopper wires, b) the corresponding section of the TOF histogram measured at the detector with the 4eV electron beam from the monochromator aimed directly into hte chopper with the lens potentials off to illustrate the effects of scattering from the flight tube, and c) the improved TOF spectrum with the lens after the chopper is optimized. The effects of several non-ideal factors in the instrumental response function, as compared to the ideal PRBS modulation function (solid curve), are discussed in relation to the ability of the Lucy algorithm to recover the TOF data.

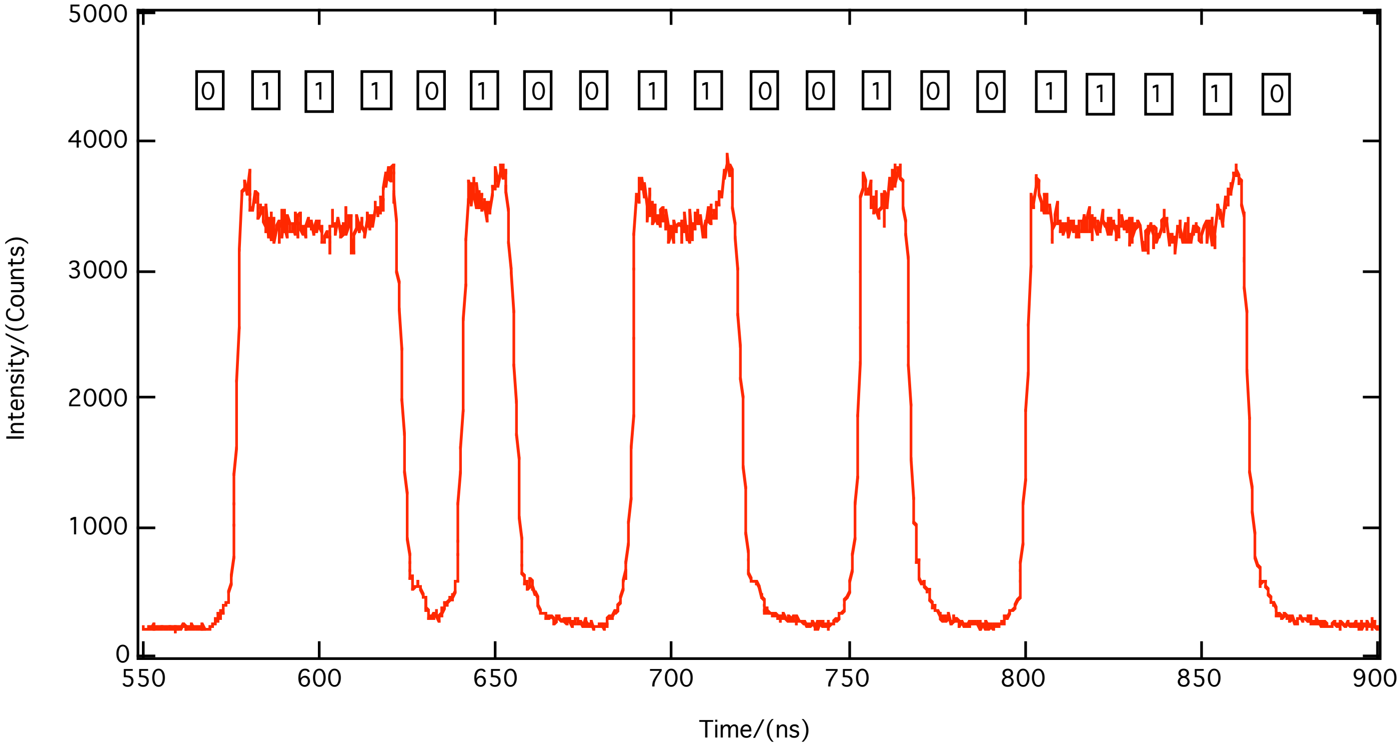


Fig. 29. Experimental response function with a 4 eV kinetic energy beam directly aimed into the TOF analyzer.

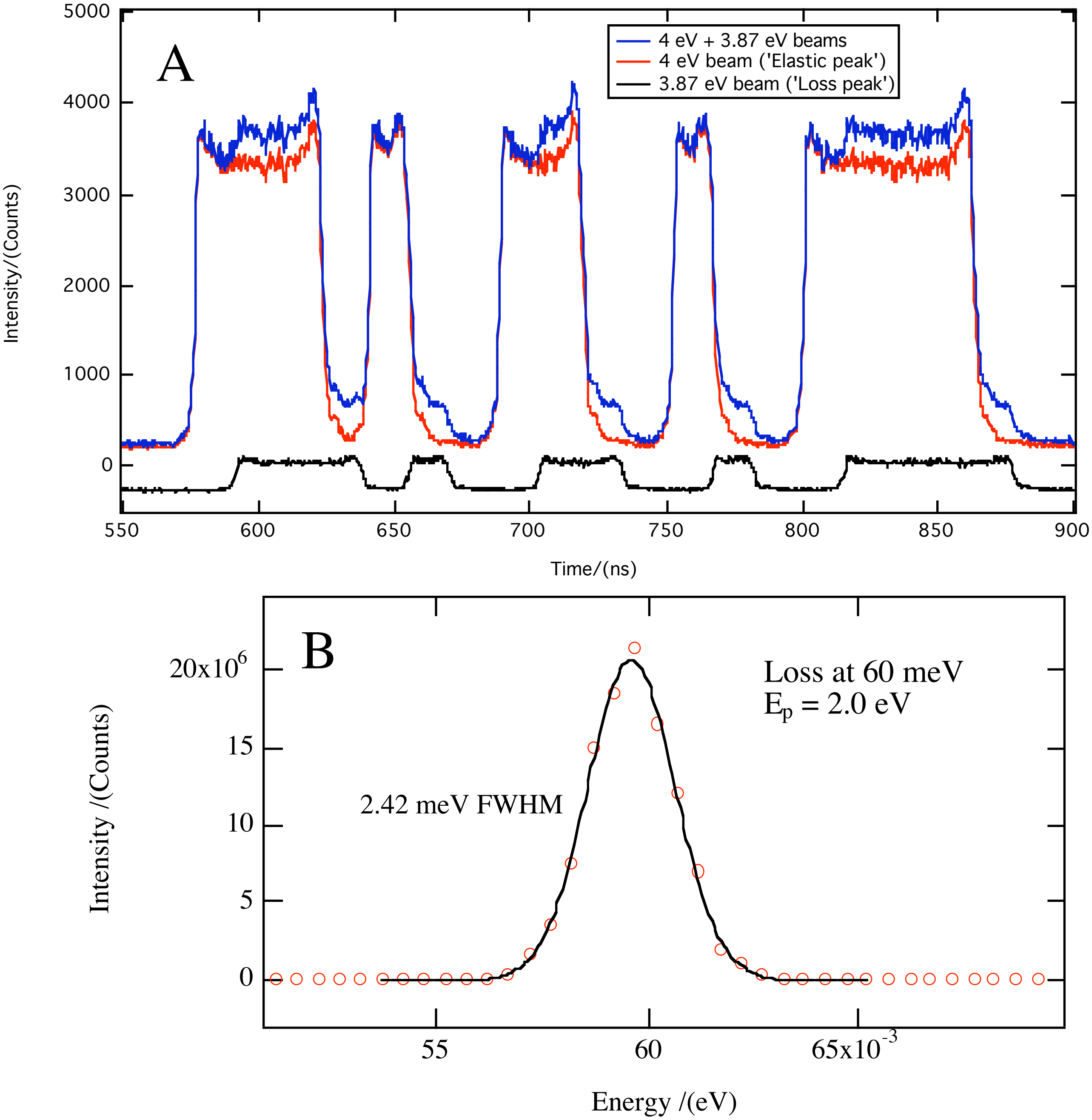


Fig. 30. A) A section of PRBS modulated data measured at beam energies of 4 eV (red curve) and 3.87 eV (black curve). Their sum (blue curve) represents the superposition of an elastic peak at 4 eV and a loss feature at 130 meV. B) Deconvolution of data similar to that shown in A) demonstrates the state-of-the-art resolution obtained with the ToF analyzer.

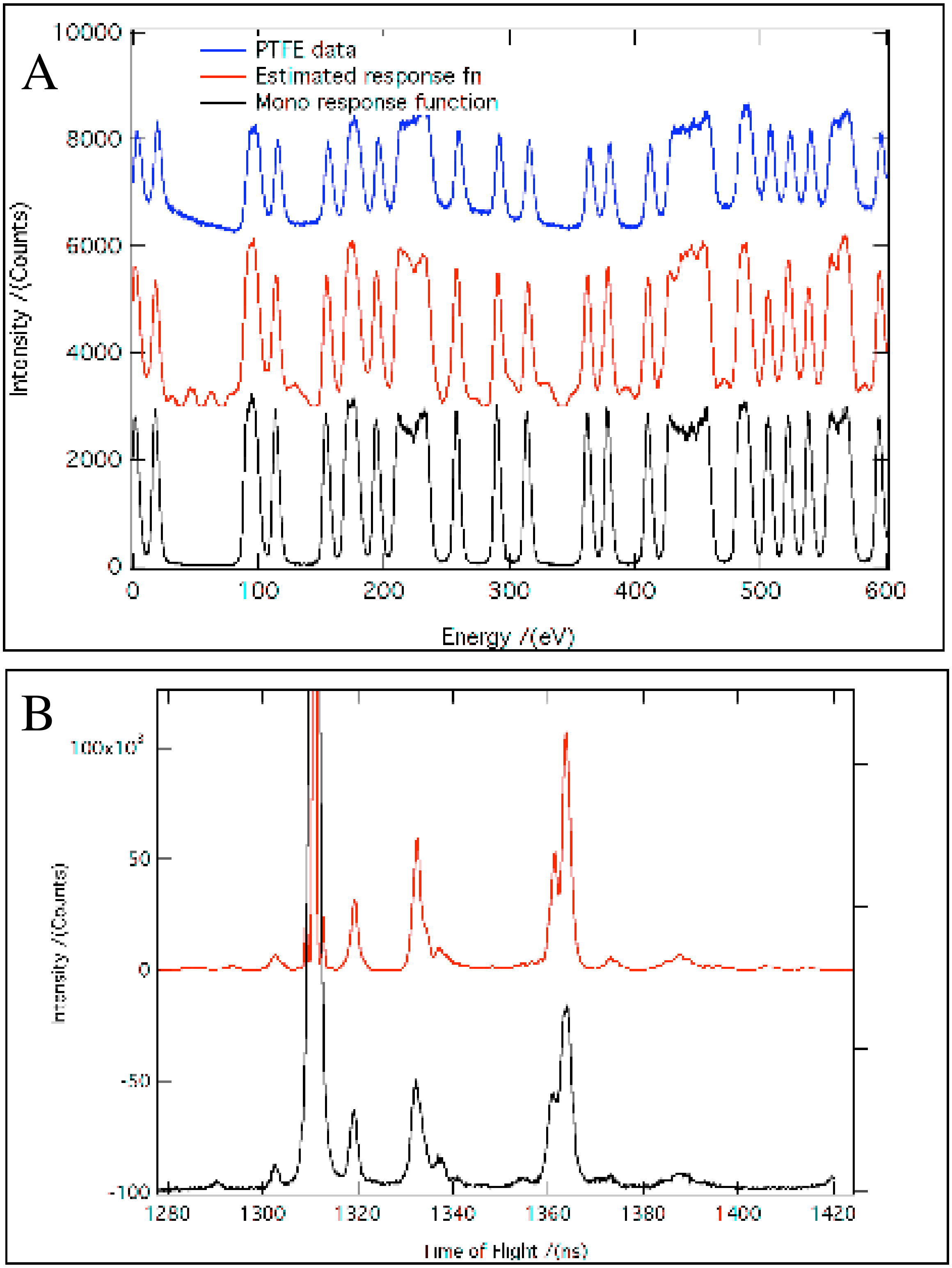


Fig. 31. A) A section of PRBS modulated data measured on a PTFE sample (blue curve), is compared to the response functions estimated from measuring the monochromatic beam (black curve) and estimate from digital signal processing. B) The recovered spectrum (red curve) using the estimated response function (red curve in A) compared to the spectrum measured with the conventional analyzer and converted to time-of-flight (black curve).

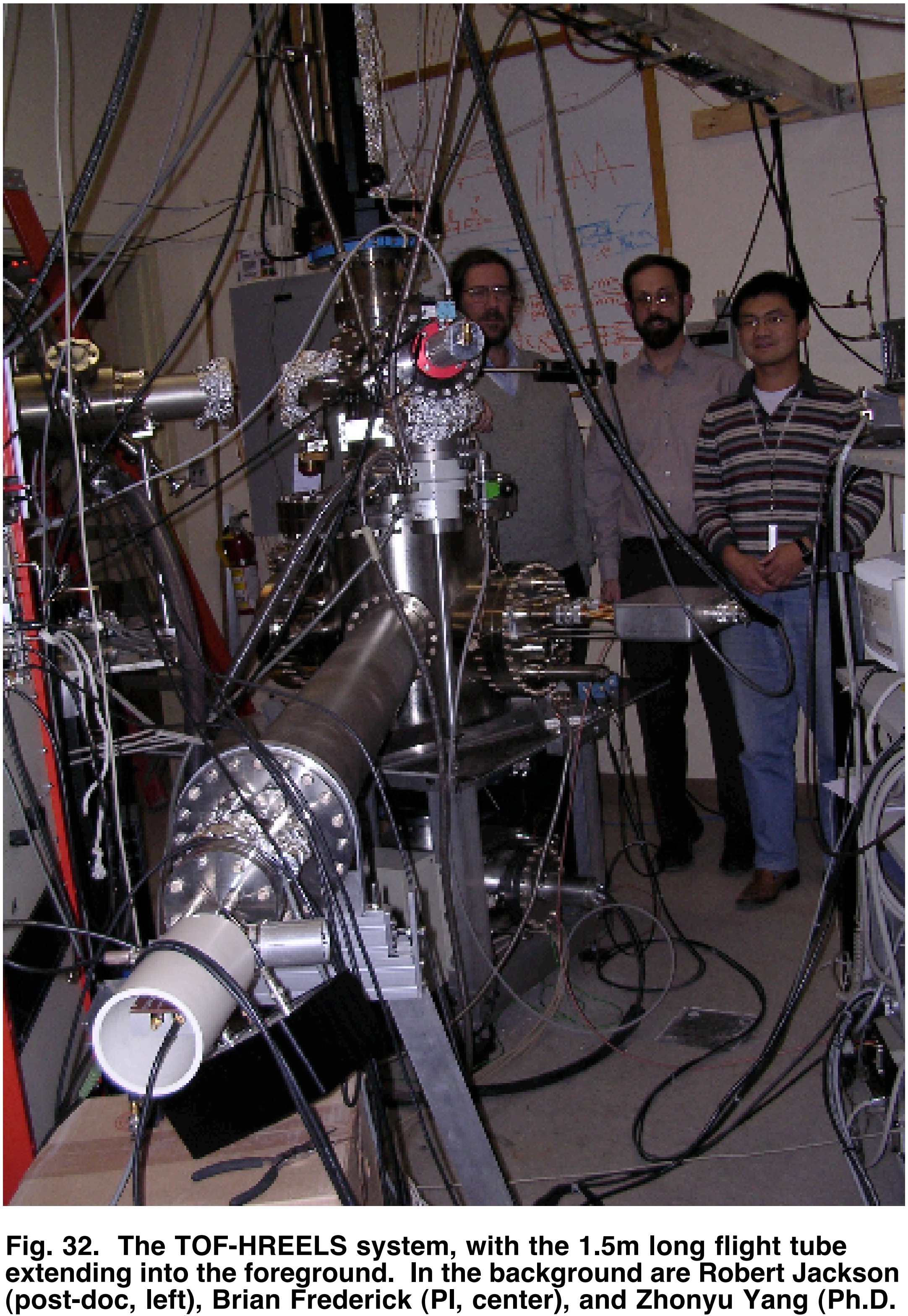


Fig. 32. The TOF-HREELS system, with the 1.5m long flight tube extending into the foreground. In the background are Robert Jackson (post-doc, left), Brian Frederick (PI, center), and Zhonyu Yang (Ph.D. student, right).

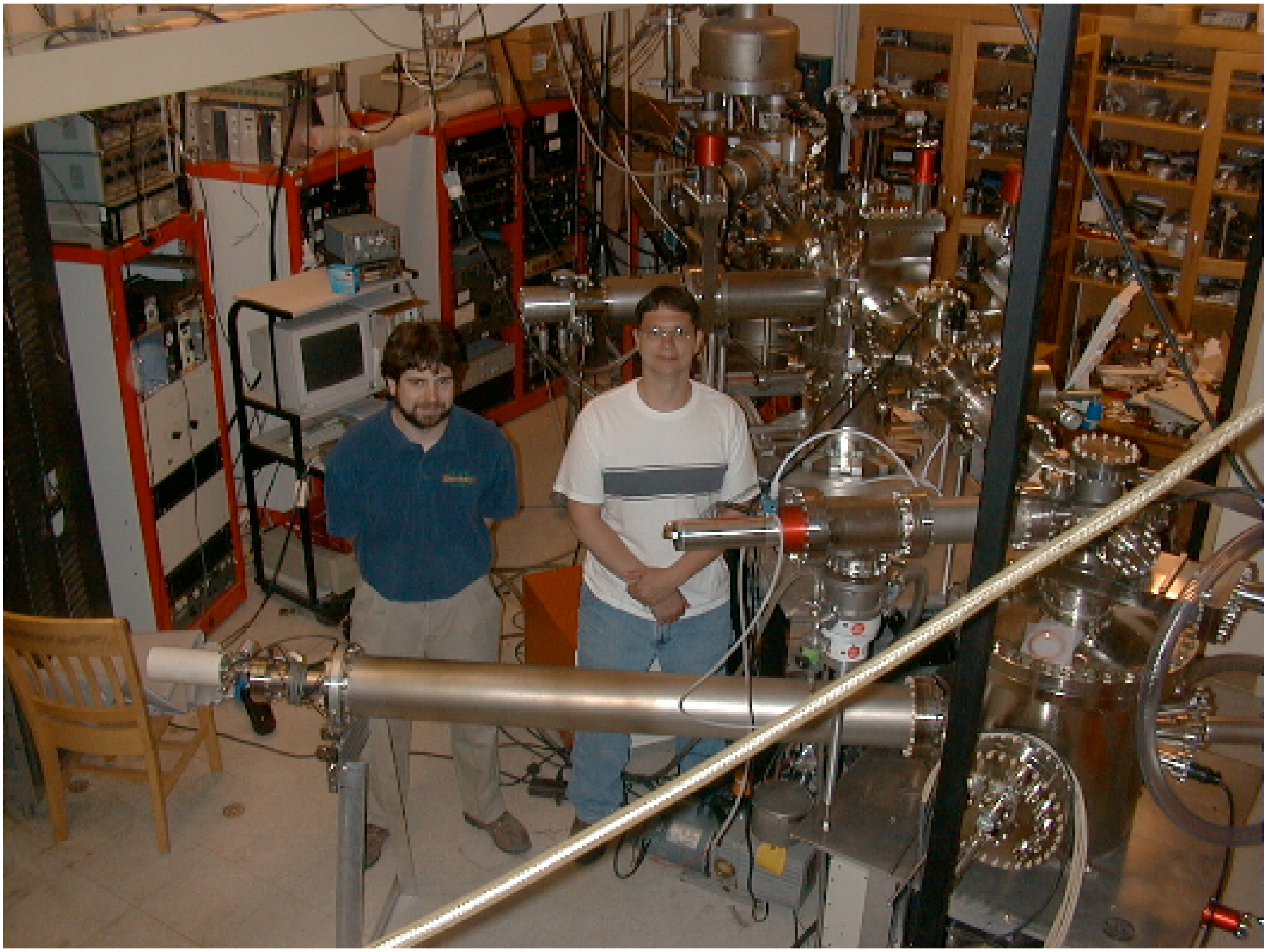


Fig. 33. The TOF-HREELS system, with the 1.5m long flight tube extending across the foreground. The TOF-HREELS instrument is connected to a preparation chamber with load lock capabilities and a controlled atmosphere photoemission spectrometer. The sample transfer and UHV system integration work was carried out by Mike Davis (right, undergraduate) and Eric Martin (Research Engineer, left).