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# Development of Laser-Plasma Diagnostics Using Ultrafast Atomic-Scale Dynamics

## 96-ERD-046 Final Report

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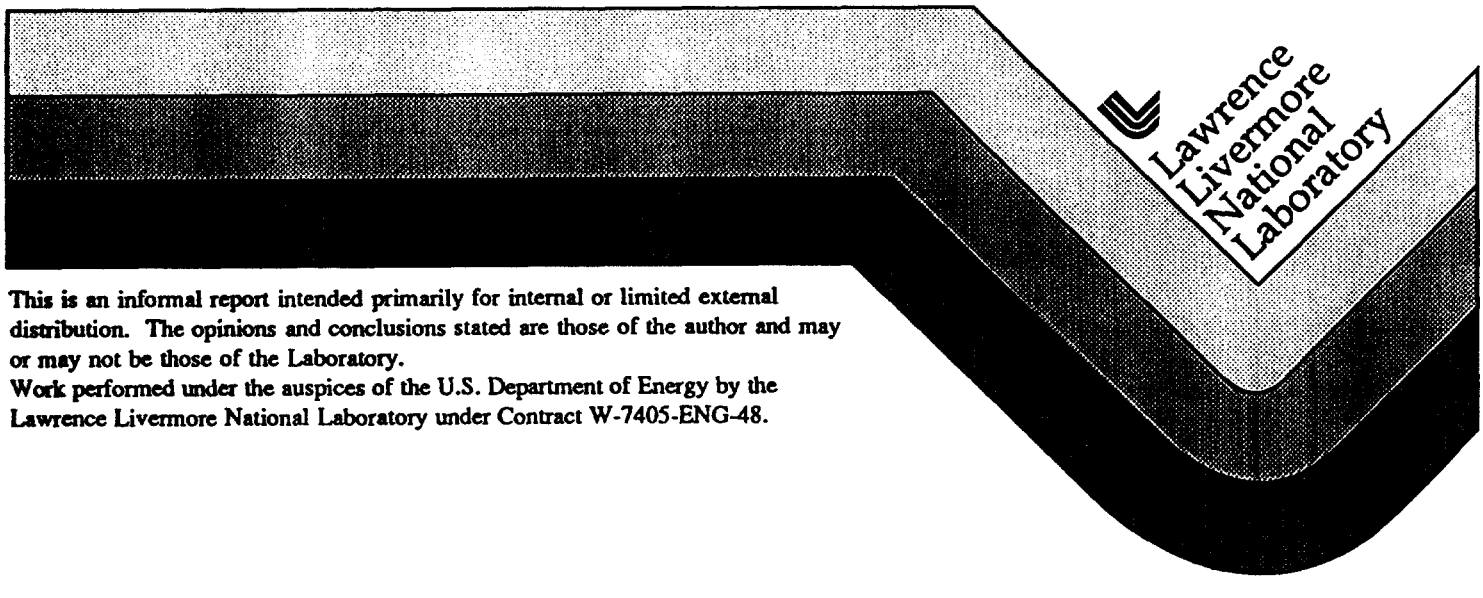


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# Development of Laser-Plasma Diagnostics Using Ultrafast Atomic-Scale Dynamics * (96-ERD-046 Final Report)

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## 1. Introduction

Ultrashort laser pulse systems allow examination of intense, ultrafast laser-plasma interactions. More specifically, intense laser irradiation can induce short xuv/x-ray bursts from the surface of condensed phase targets. Ultrafast xuv/x-ray detection is needed to understand laser-plasma interactions in this dynamic regime. Support of the Stockpile Stewardship and Management Program requires this critical understanding. Our effort here has been to extend understanding of atomic-scale dynamics in such environments with the goal of developing next generation ultrafast xuv/x-ray diagnostics where the sensors will be the atoms and ions themselves and the time resolution will approach that of the induced atomic transitions (~ a few femtoseconds). Pivotal contributions to the rapidly developing field of highly nonperturbative interactions of ultrashort pulse lasers with atoms/ions have been made at this laboratory.

In the visible/infrared wavelength regions the temporal and spectral content of ultrashort laser pulses are now reliably monitored within a single pulse using frequency resolved optical gating (FROG) which is based on rapid nonlinear optical processes such as the Kerr effect [1,2]. New applications of this basic concept are still being developed. Corresponding detection for the xuv/x-ray wavelengths does not exist and is urgently needed in many laboratory programs. The FROG technique cannot be applied in the xuv/x-ray region. Current x-ray streak camera technology is limited to ~ 0.5 picosecond resolution.

## 2. Concept and Experimental Approach

We focus on developing next generation ultrafast xuv/x-ray diagnostics. Intrinsically rapid Auger decay processes in atoms and ions are well suited to the broad band xuv/x-ray emission typical of irradiated condensed phase targets. A series of pump-probe experiments using high intensity, ultrashort laser pulses (initially of duration near 100 femtoseconds) were planned. The goal was to measure temporal and spectral characteristics of the laser-induced xuv/x-ray emission. Xuv/x-ray photons generate inner shell vacancies in the atoms of a sensor gas target (a noble gas) placed in close proximity to the xuv/x-ray source. Intrinsically rapid Auger decay of the core vacancies produces characteristic electron energy spectra. These Auger electron spectra are uniquely associated with levels of the core excitation/ionization following the absorption of a single xuv/x-ray (pump) photon. Auger electron emission by the excited atoms/ions can occur on the femtosecond time scale. For example incident photons of energy above 248 eV can induce LMM transitions in Ar that produce electron spectra in the energy range 200 - 220 eV within 10 - 15 femtoseconds. In Kr, 222 eV LLN Auger electrons are released within about 5 femtoseconds following excitation by 1.9 KeV photons. These characteristic decay spectra can be examined with eV resolution using long time-of-flight spectrometry.

Ultrafast detection would be accomplished by the modulation of Auger spectra with ultrashort laser probe pulses whose arrival time at the sensor target is varied relative to that of the xuv/x-ray pulse[3]. The probe laser field 'dresses' the sensor atoms and generates Auger spectral sidebands observable only when both the xuv/x-ray pump and laser probe fields coincide within a sensor target. Sidebands are spaced by the probe photon energy, $h\nu$ which is independent of the xuv/x-ray spectrum being monitored. A simple classical calculation can be used to estimate the number of sidebands, N as follows [3]:

$$N \approx \frac{2\sqrt{8U_aU_p}}{h\nu} \qquad (1)$$

where $U_p$ is the ponderomotive (average oscillatory) energy of the electron in the laser field and $U_a$ is the intrinsic Auger electron energy.

Figure 1 illustrates the concept of the proposed series of experiments. Temporal resolution is defined by the pulse length of the probe which will initially be of order 100 femtoseconds. Resolution can be improved by using much shorter probe pulsewidths. It has also been shown that a variant of this pump-probe technique can be applied to the measurement of high order harmonic pulsewidths generated by some fundamental intense laser pulse[4,5]. In this case one observes sidebands in photoelectron energy spectra (revealed as even harmonics) which are rapidly generated by the ultrashort (odd) harmonics of the incident laser pulse.

In FY'96 we assembled two parallel experimental assemblies. Because it is important to understand photoelectron energy spectra as a potential source of background signal the first experimental setup was dedicated to the measurement of photoelectron energy from the optical field ionization (OFI) typical of intense, ultrashort laser pulses. Furthermore, measurement of the laser intensity dependence of these spectra provides threshold information

needed to guarantee that lower intensity probe pulses would not cause unwanted ionization in the sensor gas target (which serves as the source of Auger electrons)[6,7]. Progress with this investigation is discussed in the section four.

The second experimental setup was configured for conducting the pump/probe experiment in which ultrafast xuv/x-ray detection was to be accomplished by modulation of the Auger electron energy spectra. Examination of background sources relevant to Auger spectrometry and the development of the molten gallium target were completed using this second configuration. Development of the gallium target is discussed in section three and background measurements are described in section five.

Argon was chosen as the first sensor gas to be used. $L_{23}M_{23}M_{23}$ Auger transitions in argon generate decay electrons in the energy range, 200 to 220 eV , leaving behind doubly charged argon ions (mainly in the ground state). The electron energy spectrum associated with this decay channel is well known. In the pump-probe technique these electrons are modulated by 1.55 eV photons (800 nm probe). Our work with gallium indicates that this is a suitable source of soft x-rays yielding photons with energy above the 248 eV excitation level (argon L edge for ejection of $2p^6$ electrons) of the LMM Auger channel.

### 3. Molten Gallium as a Soft X-ray Source

A significant development to emerge from this project is the design of a molten target which is self-regenerating under repetition-rated laser irradiation. Molten gallium has proven to be a viable regenerative target for xuv/xray production at repetition rates of a few hertz where emission of soft x-rays with photon energy above 250 eV has been verified[3]. Figure 2 illustrates the target configuration. A small sample of solid gallium is placed onto a heated cradle. Gallium is a suitable choice because of its low vapor pressure and low melting point of 85.6 $^{o}$ F ( 29.8 $^{o}$ C). Cradle heating is

provided by a heater coil wrapped around the supporting post below the cradle. Cradle temperature is controlled to within 0.1 $F^{o}$ by using a thermal probe mounted inside the heated post. A saucer type catcher is positioned between the cradle and heated post to intercept any gallium which falls from the cradle during experiments (with adequately high incident laser intensites the molten target can be completely removed from its cradle after a single shot). This entire assembly was mounted on a three dimensional translation stage for the x, y, and z motion required to bring the target to the position of best focus for the laser irradiation in a vacuum environment. We operated the gallium target under a vacuum pressure levels of $10^{-6}$ to $10^{-5}$ torr.

The molten gallium surface is irradiated with intense, ultrashort 800 nm laser pulses at a maximum repetition rate of 5 Hertz. Typical laser pulsewidths were 120 femtoseconds with pulse energy ranging from about 0.1 to 25 millijoules. While exposed to laser irradiation the movement and depletion of the gallium sample (by ablation) can be observed in shadowgraphs displayed on a video camera. An incandescent source is used to backlight the gallium target at 90 $^{o}$ relative to the direction of laser pulse propagation. The prepared target surface is of smooth curvature and debris is easily directed by target positioning relative to incident irradiation. Energy absorbed from the first several laser pulses (at the few millijoule level) is adequate to reshape a molten sample of initially arbitrary shape to a smooth, well behaved curvature. It is this conditioned molten sample which serves as the xuv/x-ray production target.

Under vacuum operating conditions the stability of the target material is dependent on its temperature, the incident laser pulse energy and the laser repetition rate. Optimum conditions for material stability are : temperature setting in the range 85 to 87 $^{o}$F, laser pulse energy in the range 1 to 13 millijoules, and maximum repetition rates of 3 Hertz. Under these conditions the gallium surface was observed to quiver (at the amplitude level of a few microns) after each laser shot. Conservative target recovery times in excess of 200 milliseconds were used. With each laser shot some gallium is lost as debris. A sample with mass of order 10 mg can last for thousands of laser

shots (typically 3000 to 5000). With laser pulse energies above 13 millijoules the target material would quiver with greater amplitude with a significant possibility at the 20 millijoule level of being physically removed from the cradle onto the catcher. Similar unstable behaviour was observed when repetition rates above 3 Hertz were used not allowing sufficient time for the molten surface to recover its smooth shape between shots. The molten target was more flexible at higher temperature. The optimum operating temperature range represents a compromise of viscosity which minimizes target recovery time yet preserves the smooth curved target surface for each laser shot.

We were also able to operate the target optimally while ejecting gas (argon or neon) from a nozzle located near the molten gallium. Furthermore, the ability to direct gallium debris away from the Auger electron spectrometer protected the microsphere plate detector from damage due to breakdown.

Experiments have been conducted to determine the xuv/x-ray emission spectrum from gallium. This was done by monitoring the transmission through a simple filter array mounted between the gallium target and a thinned, back-illuminated CCD detector [8]. This data is currently being analyzed. For coarse comparison we have also calculated the xuv/xray yield expected from a germanium surface (shown in figure 3 [9]) which is expected to be similar to that of gallium.

Regenerative target development is important for applications requiring repetition-rated irradiation by intense, ultrashort laser pulses.

**4. Determination of Photoelectron Energy Spectra**

It is important to distinguish Auger electrons from photoelectrons which can be present in the pump-probe experiments. Photoelectrons can be formed via optical field ionization driven by the intense irradiation incident on the gallium target[6,7]. Therefore ionization of the gallium or the sensor gas could occur. Similarly the lower intensity probe pulse may also generate

photoelectrons. In this case care should be taken to keep the probe intensity below the established OFI threshold for single ionization.

An experimental configuration as shown in figure 4 was assembled in which photoelectrons were monitored with a retarding potential spectrometer employing a multigrid (four) analyzer section. The use of multiple analyzer grids provides high energy resolution[10]. The detector is a matched pair of microchannel plates. Data has been taken to examine the dependence of photoelectron energy spectra on laser intensity and the orientation of linear polarization (horizontal and vertical cases with repsect to the spectrometer axis). Ultrashort laser pulses of 800 nm wavelength and 120 femtosecond duration were focussed to a 16 micron diameter spot at the entrance aperture of the spectrometer . The intensity (energy) range examined was $2 \times 10^{14}$ to $6 \times 10^{16}$ W/cm$^2$ (0.05 to 14 mjoules). Analysis of this data is underway.

A typical photoelectron spectrum, formed by optical field ionization (OFI) of helium, is shown in figure 5 where experimental data (solid line) is compared to the prediction obtained using our semiclassical ionization model [7]. The data was obtained at a laser intensity of $1.2 \times 10^{15}$ W/cm$^2$ for a 150 femtosecond pulsewidth at a 780 nm central wavelength. Noteworthy are the energy range of the spectrum and its dynamic range. The two main components of this OFI spectrum can be described in terms of the ponderomotive energy, $U_p$.

Electron energies up to $2U_p$ represent the lower energy component. This part of the spectrum is accurately predicted based on the interaction of the liberated electron with the laser field where a simple quasi-classical model is adequate. Most of the photoelectrons occupy this part of the spectrum.

By contrast, relatively few electrons are produced in the higher energy tail region between $2U_p$ and $10U_p$. This second spectral component is attributed to rescattering of the electron (following ionization) by the remaining ion core in the combined laser and ion fields. It has been shown that incorporation of this subsequent electron-ion scattering is needed to accurately predict the higher energy spectra over the large dynamic range

shown in figure 5. Knowledge of such OFI photoelectron spectra is needed to assess potential background contributions in Auger spectroscopy experiments.

**5. Tests with Auger Electron Spectrometer**

The Auger spectrometer design is shown in figure 6. A five-grid analyzer section is placed above two extraction grids[10]. The remaining grids above the analyzer section comprise the drift section (approximately 1 meter long) which provides temporal dispersion. For baseline experimental tests the gallium target was placed below all grids of the spectrometer as seen in the figure and the gas nozzle directed sensor gas into the region between the two extraction grids. At the end of the drift section we used a microsphere plate detector (MSP).

Operation of the sensor gas valve is limited by the moderate vacuum requirement for operation of the MSP detector. For the Auger spectroscopy the sensor emission occurs synchronously with the laser pulse. In summary it was experimentally determined that a vacuum environment suitable for MSP operation could be maintained while the sensor gas is introduced under the following conditions:

| | |
|---|---|
| repetition-rate | < 5 Hz |
| gas valve open time | < 250 microseconds |
| plenum pressure | < 80 psig |

With the above cconstraints we could eliminate the need for enhanced collection (i.e. into a collector funnel) of sensor gas to facilitate pumping.

Synchronous timing of the laser pulse and gas emission was set using frequency-resolved optical-gating (FROG). The FROG trace of ultrashort laser pulses transmitted through the sensor gas was monitored. The firing time of the gas valve was varied until 'blue' spectral shifting of the transmitted laser pulse was observed. This shift toward shorter wavelengths is characteristic of gas ionization (in this case OFI) induced by the laser pulse[2].

Gaseous ionization is also responsible for significant off-axis scattering of laser light as well as emission of light from gallium during recombination. In the course of the gas timing measurements we routinely detected such scattered light with the MSP detector. In this way scattered light directed up the drift tube of the Auger spectrometer provided a convenient optical fiducial marker for determination of electron energy (using drift times). This was particularly effective when the gallium target was in place. This early optical fiducial marker can be seen in the oscilloscope trace of figure 7 taken with a gallium target. The addition of a gallium target at the laser focus (under the same spectrometer bias settings and laser irradiation conditions as in the vacuum case) resulted in a significant enhancement of both the optical fiducial and electron signal levels.

The placement of the gallium target also enabled us to direct gallium debris away from the electron spectrometer grids and drift tube. Debris was routinely collected by the upper extraction grid due to its proximity. Following thousands of laser shots on target there was negligible debris on the analyzer grids and above. Meanwhile, a configuration of debris shields was kept in place to protect all optics in the plane of incidence of the laser pulse.

To determine the energy spectrum of the potential photoelectron background in the experiments we had examined time-of-flight (TOF) data for two cases; a vacuum target (ambient density), and a gallium target in vacuum. This was done at laser irradiation levels of order $10^{17}$ W/cm$^2$ (laser pulse energy of 20 mjoules with a 10 micron spotsize, 130 femtosecond

pulsewidth, and 800 nm central wavelength) at a 5 Hz repetition rate. Horizontal orientation of the linearly polarized pulse was used.

With a vacuum target both electron and ion signals could be seen on the TOF spectra. The ion contribution was significantly delayed (relative to the electron contribution) with flight times of order microseconds. We verified that the electron flight time (TOF), for a given energy, could be adjusted by the bias level on grids 8,9 and 10. Electron TOF is determined relative to the optical fiducial. The spectrometer grid bias settings used are listed below :

| grid # | bias (volts) |
|---|---|
| 1 | 0 |
| 2 | 2 |
| 3-7 | -200 |
| 8-10 | -150 |
| MSP | -4500. |

This spectrometer design was set (i.e. the biases on the analyzer grids 3 to 7) to allow only transmission of electron energies above 200 eV. This is the energy of interest for Auger electrons generated in the sensor gas. Simulations of this design (using the application, SIMION) indicate that the flight time for 210 eV Auger electrons would be 208 nsec and that these electrons would arrive at the MSP detector with 60 eV kinetic energy. Figure 8 illustrates the calculated dependence of electron flight time and final kinetic energy (i.e. at the MSP detector) on the bias of grids 8,9, and 10.

## 6. Summary

Design, fabrication, and testing of critical apparatus components have been completed in preparation for conducting the proposed pump-probe experiments to develop ultrafast xuv/xray detection. Tuning of the Auger spectometer has been demonstrated for this work using photoelectrons and optimum operating conditions for the regenerative molten gallium target as a viable xuv/xray source have been established. Photoelectron spectra have been examined experimentally and theoretically to verify spectrometer performances and to characterize potential background contributions to sought Auger spectra. Detailed analysis of OFI photoelectron data and xuv/xray yields from gallium are ongoing. These developments are critical and serve as the technical basis for any continuation of this work.

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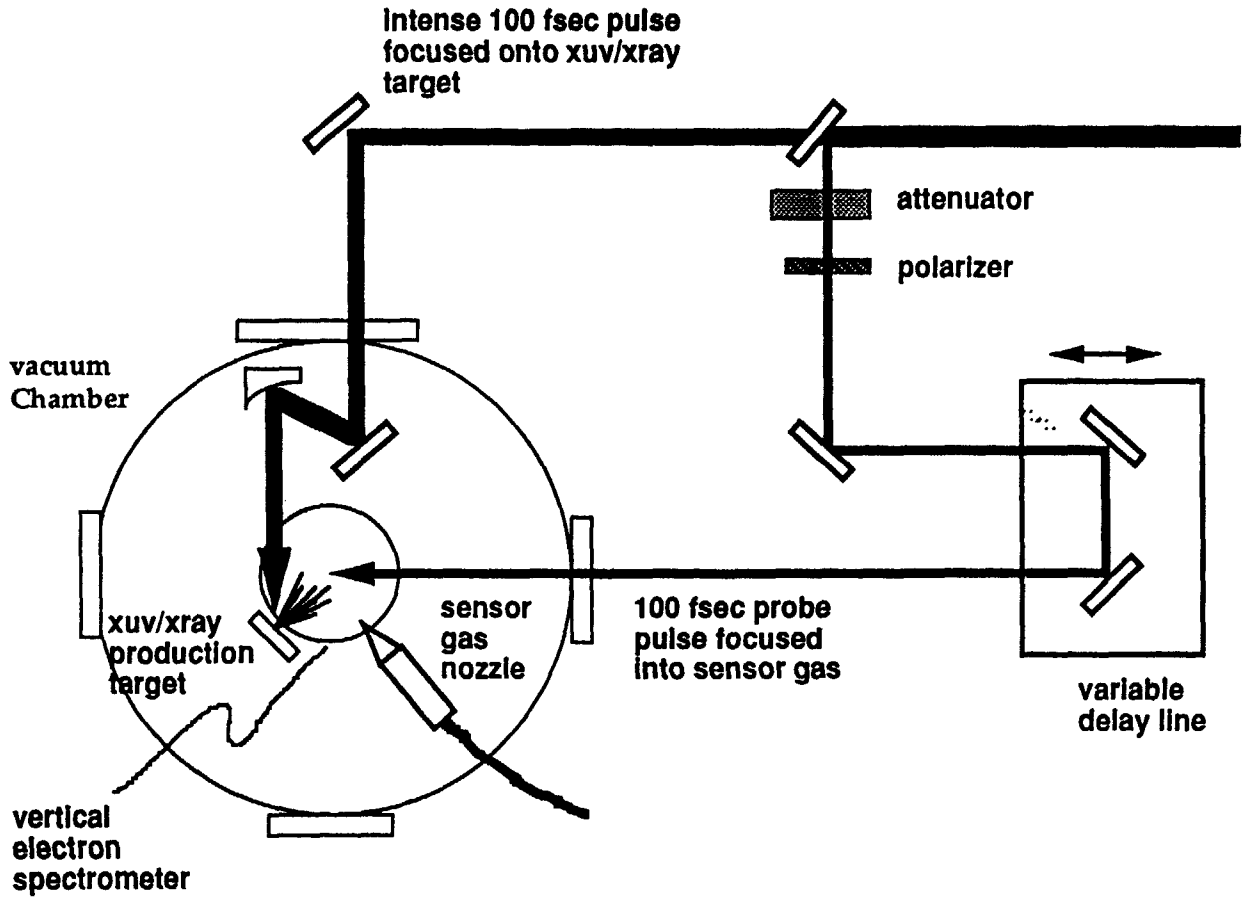


Figure 1. Experimental configuration for pump/probe studies of modulated Auger spectroscopy and consequent ultrafast xuv/xray detection.

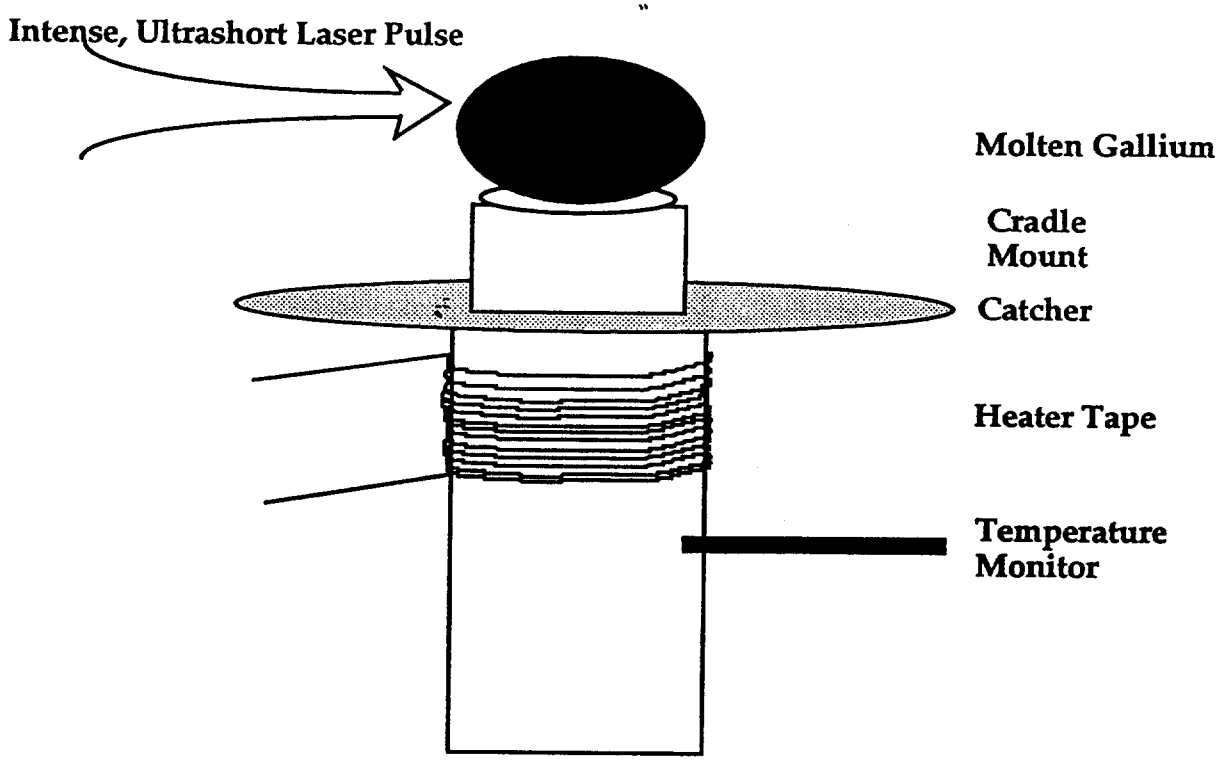


**Figure 2. Molten gallium target design**

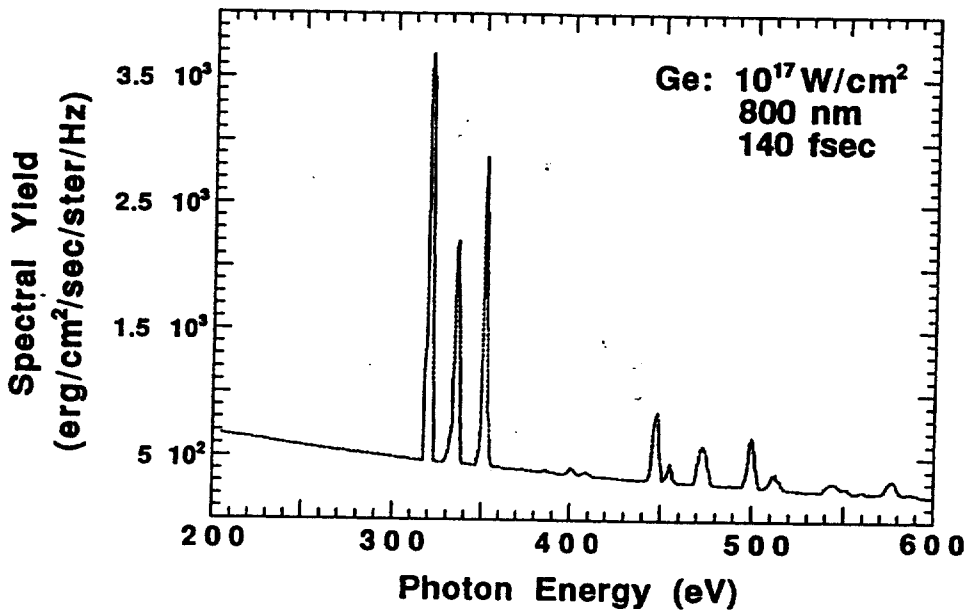


Figure 3. Spectral Yield from germanium irradiated by 800 nm laser pulses of 140 femtosecond at $10^{17}$ W/cm$^2$.

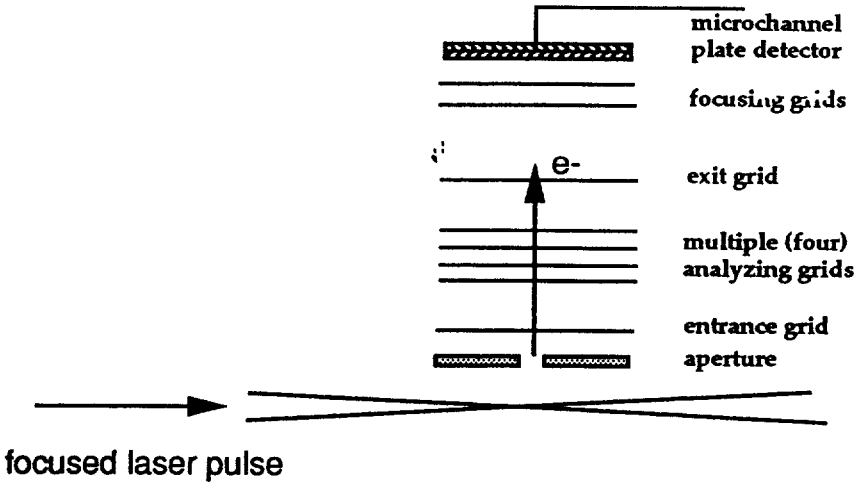


**Figure 4. Multigrid retarding potential spectrometer for photoelectron detection.**

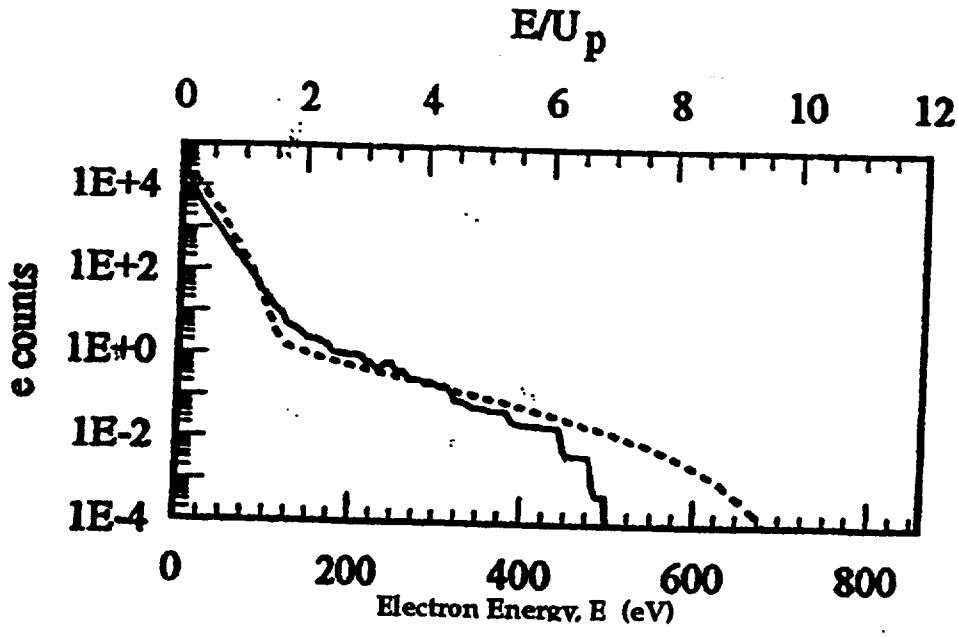


Figure 5. Photoelectron spectrum produced by optical field ionization.

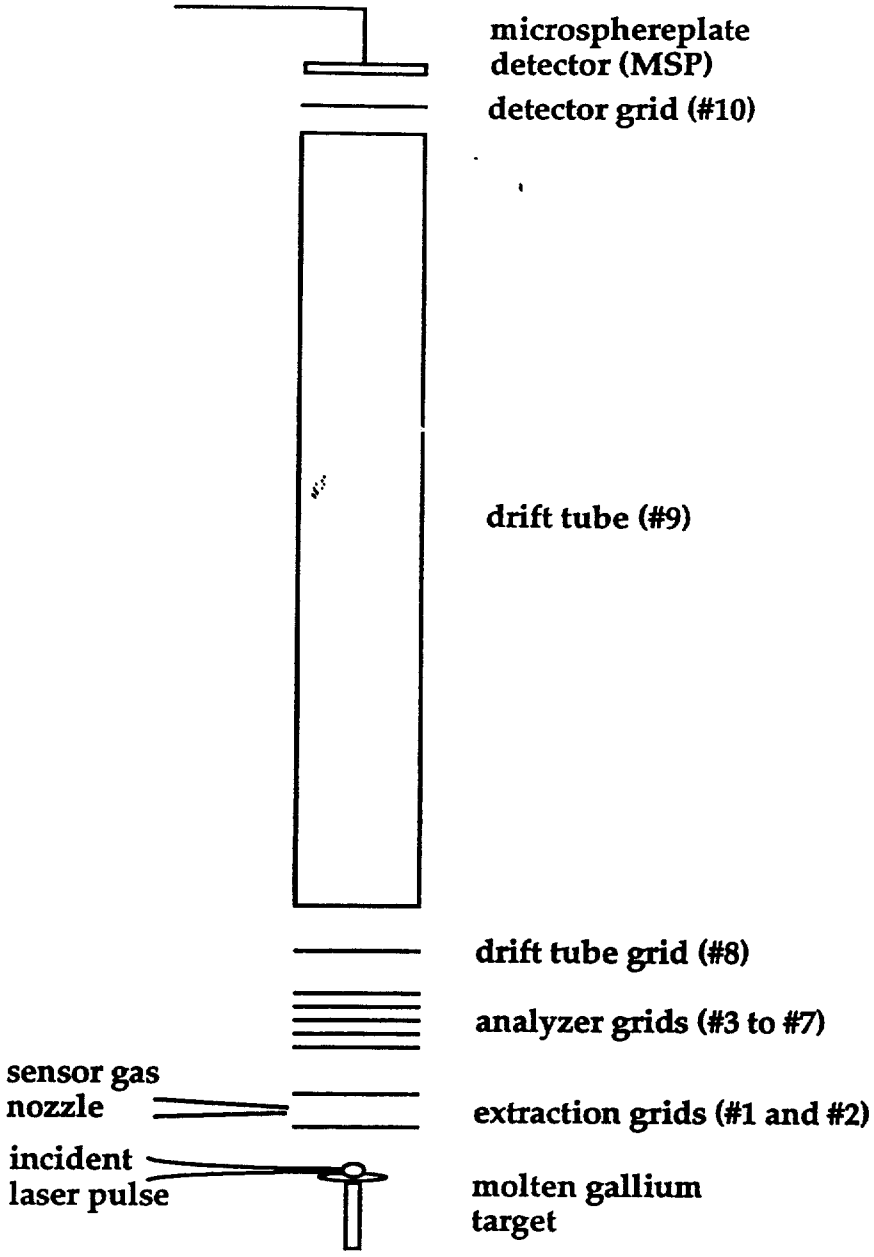


**Figure 6. Time-of-flight Auger spectrometer with multigrid analyzer section.**

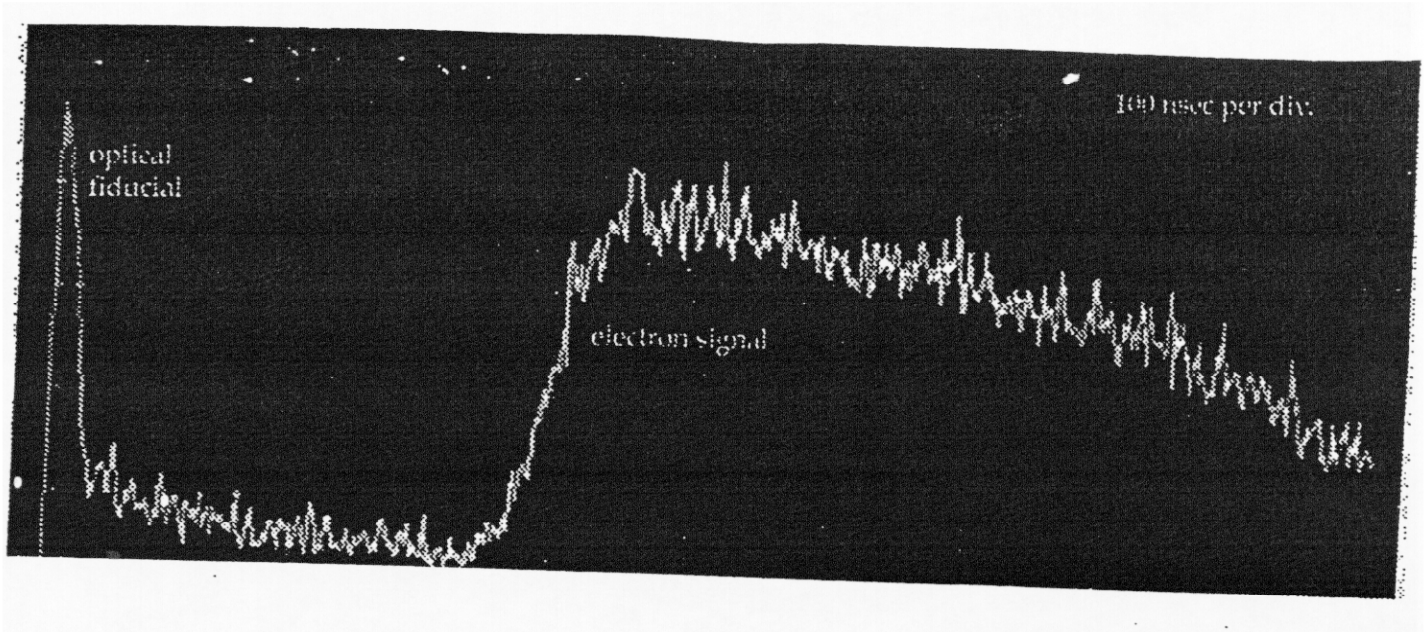


**Figure 7. Electron flight times determined relative to the optical fiducial signal with the Auger spectrometer**

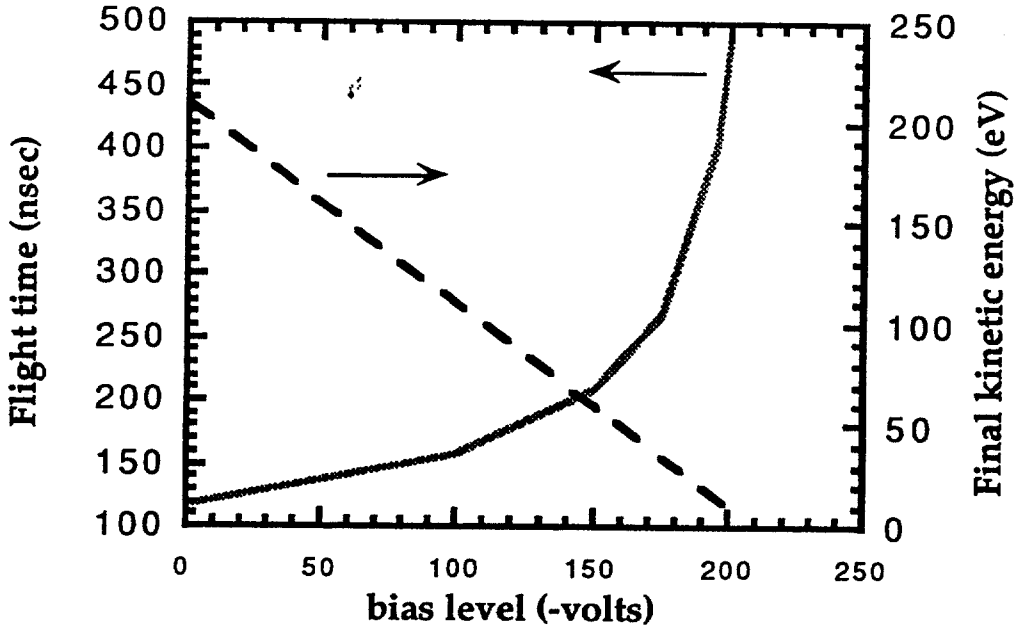


**Figure 8. Bias level tuning of Auger spectrometer.**

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