

# ELEMENTARY ATOMIC AND MOLECULAR PROCESSES IN AN INTENSE X-RAY FIELD

AMOS team (Argonne National Laboratory, Lawrence Berkeley National Laboratory, U. Colorado, U. Michigan, Ohio State U., Texas A&M and U. Texas, Austin)

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## 1. INTRODUCTION

The first x-ray FEL will be realized with the 2008 commissioning of the LCLS at SLAC. The LCLS will achieve an extraordinary set of output parameters but the unparalleled advance in comparison to all existing x-ray sources is the unprecedented peak power. The LCLS beam intensity is many orders of magnitude ( $10^{13}$  x-rays/200 fs) greater than the current 3<sup>rd</sup> generation sources ( $10^8$  x-rays/100 ps). Nonlinear and strong-field effects are expected when the LCLS beam is focused to a spot diameter of 100 nm, as is required for the structure determination of single biological molecules in the gas phase<sup>1</sup>. With this extreme focusing, the intensity is  $\approx 5 \times 10^{35}$  photons/s/cm<sup>2</sup> ( $\approx 0.6 \times 10^{20}$  W/cm<sup>2</sup> for 800 eV x rays)! The LCLS will be the first tool that unequivocally moves the basic physics into a new and unexplored interaction regime where both the external field strength and frequency can exceed an atomic unit.

In addition the short pulse duration, and in particular the possibility of decreasing the duration to sub-fs, provides an important tool for studying dynamics on the time scale ranging from atomic motion in solids to electron configuration changes in atoms, using hard x-rays. Prior to the arrival of first light from the LCLS, technology has restricted the exploration of the intense laser-atom interaction to the response of valence electrons in low-frequency fields ( $h\nu \ll E_b$ , atomic binding energy). These investigations have documented many remarkable discoveries, e.g. cluster-driven fusion and attosecond pulses. The atomic response to intense fields will fundamentally differ at high frequencies ( $h\nu \gg E_b$ ). For one, the electron's ponderomotive or quiver motion so important at low frequencies will be negligible in the x-ray regime due to its  $\lambda^2$ -scaling. Furthermore in the x-ray regime, unlike the low-frequency regime, the valence electrons will be transparent while the inner-electrons will have a strong response. In a strong field, single electron approximations have been shown to yield an adequate description of the optical excitation of valence electrons but the description of inner shell ionization is less straightforward since post-collision and Auger processes will become important. Thus the fundamental difference between intense light-matter interactions in the x-ray regime and all previous investigations is that atomic ionization will occur from the inside out and accordingly the multiple-ejection electron dynamics should be dominated by the short range physics and correlation.

In this document we propose to investigate the elementary process of *intense* x-ray light interacting with simple isolated systems, e.g. atoms, molecules, and clusters. As discussed below, from a fundamental perspective the opportunity for new discovery is high since prior experimental studies are nonexistent. Furthermore the understanding acquired from the proposed program will have an impact on other LCLS experimental programs involving more complex systems or concepts. In addition the controlled nature and simplicity of the proposed experiments will provide a benchmark of LCLS performance and advance new methods of characterization. This letter of intent (LOI) organizes a multi-institutional collaborative team in atomic, molecular and optical science (AMOS) for accomplishing the objectives of the proposed program. The AMOS membership encompasses broad and demonstrated expertise in intense laser-matter physics, x-ray and nonlinear optics, two-color processes, coherent control and atomic theory. Part of the membership (Bucksbaum, Young) was responsible for developing the AMO science case for the LCLS "first experiments" document while others (DiMauro, Ditmire, Paulus) have participated in the preliminary design of the LCLS AMOP Near Hall experimental hutch. Our expanded membership also includes expertise in atomic theory (Greene, Reading) which will require combining aspects of correlated many-body theory, including inner-shell resonance and threshold effects and nonlinear interactions with the radiation field, in order to interpret and model observations. The aim

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<sup>1</sup> R. Neutze, R. Wouts, D. van der Spoel, E. Weckert, and J. Hajdu, *Nature* **406**, 752 (2000).

of the science program is to gain fundamental insights while providing benchmark studies for the LCLS project by concentrating on the following AMOP thrust areas:

1. Elementary intense field atomic & molecular inner-shell photoionization
2. X-ray nonlinear processes
3. X-ray driven cluster dynamics
4. Static & time-resolved x-ray imaging of molecular structure.

As a *category A* LOI, the AMOS team will develop the instrumentation needed for the science program via two paths.

- Close coordination with LCLS personnel for design and construction of the Near-Hall atomic physics experimental end station hutch and x-ray optics planned as part of the project baseline.
- Seek DOE/BES funding for additional instrument development and end station operations.

The conceptual design of the atomic physics end stations will not only function for the proposed AMOS program but also will be adaptable for future developments in the LCLS capabilities and provide flexibility for different AMOP proposals.

## 2. PROPOSED STUDIES

### *ELEMENTARY INTENSE FIELD ATOMIC INNER-SHELL PHOTOIONIZATION*

All LCLS experiments will involve the interaction of an intense x-ray beam with matter. Therefore establishing an early program aimed at achieving a thorough understanding of the basic atomic processes in strong x-ray fields is an essential step on the roadmap leading to more complex processes. Our present understanding of x-ray interactions with atoms is largely based upon extensive research using synchrotron radiation, where the interaction is clearly in the weak-field limit.

We propose a suite of experiments, initially on gas phase atoms, to understand the transition from the weak- to the strong-field regimes using focused x-rays. Some striking phenomena (for x-ray wavelengths) are expected due to the increased x-ray intensity: 1) saturation of photoionization, such that the photoabsorption rate is comparable to the Auger decay rate, leading to multiple core hole formation, and 2) two-level population trapping when the x-rays are resonant with a transition to an isolated Rydberg level, leading to a decreasing ionization probability with increasing intensity. Less clear but extremely interesting is whether the presence of the intense x-ray field will alter the Auger process.

To illustrate this point consider atomic neon interacting with the LCLS beam near the K-edge, 870 eV. Double-K ionization due to two-photon absorption in a single atom, is energetically allowed for photon energies above  $1863 \text{ eV}/2 = 932 \text{ eV}$ , one half of the KK threshold. Above this energy, KK-dominates the single K- vacancy production via two-photon absorption<sup>2</sup>. For the focused intensity of  $5 \times 10^{35} \text{ photons/s/cm}^2$ , the probability for KK ionization is 100%. This can be contrasted to the KK ionization probability of 0.32(4)% measured for single photon absorption of 5 keV x-rays<sup>3</sup> using synchrotron radiation. The decay of the KK-vacancy state will produce higher charge states and will be an important damage consideration in experiments on molecules, where extensive fragmentation will take place. In fact, the central assumption of the biomolecular imaging experiment, as yet unproven, is that the photoabsorption cross-section decreases during the x-ray pulse due to the production of multiple K-holes, effectively increasing the damage threshold. It is crucial for these and other experiments to understand the timescale of the fragmentation via pump-probe techniques.

Another interesting scenario can arise when Auger decay occurs in a strongly driven resonant transition. For example, the x-ray absorption spectrum of neon near the K-edge shows clear Rydberg structure prior to the onset of the continuum. After excitation of the 1s electron to one of these Rydberg states, spectator Auger decay is dominant, leaving  $\text{Ne}^+$  as the most abundant charge state. Rapid cycling of the  $1s \rightarrow \text{Rydberg}$  transition can inhibit the Auger decay, leading to a decreasing ionization probability

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<sup>2</sup> S. A. Novikov and A. N. Hopersky, J. Phys. B **33**, 2287 (2000) and **35**, L339 (2002).

<sup>3</sup> S. H. Southworth *et al.*, Phys. Rev. A **67**, 062712 (2003).

with increasing intensity. This is one example of interesting and potentially useful resonant phenomena that can be observed with sufficient monochromatization and tunability of the LCLS beam.

The significance of inner-shell processes involving several electrons immediately raises the question of collective effects. Inner-shell ionization might be used to probe molecular potential surfaces by the outgoing photoelectron: the molecule gets rapidly illuminated from within while it dissociates. We propose to use pump-probe techniques for setting an internal molecular clock and analyze the many particle events using a reaction microscope coincidence technique. A new regime will be realized as the LCLS pulse duration is decreased to 10 fs since the molecular motion will be frozen.

### *X-RAY NONLINEAR OPTICS*

The LCLS x-rays will differ with traditional nonlinear optics since multiphoton excitation will proceed through inner-shells. In contrast to low-frequency light, x-ray nonlinear excitation will cause a change in the absorption and scattering as the core configuration is modified. In other words the nonlinear process does not see a frozen core. At the highest LCLS intensities, the presence of the x-ray field could also influence the excitation dynamics. Modified inner-shell processes can also be investigated by quivering the valence electrons with a low-frequency laser light.

A simple perturbative calculation illustrates the efficacy of the LCLS for driving nonlinear processes. The 2-photon transition probability is given as  $\sigma_2 F^2 \tau$ , where  $\sigma_2$  is the generalized two-photon cross-section (units,  $\text{cm}^4 \text{s}^{-1}$ ),  $F$  is the photon flux (units,  $\text{cm}^{-2} \text{s}^{-1}$ ) and  $\tau$  is the pulse duration (for LCLS  $\tau \cong 100$  fs). Assuming a 1-photon non-resonant cross-section of  $10^{-18} \text{cm}^2$  and a lifetime of the virtual state of 1 as, one derives a value of  $\sigma_2 = 10^{-54} \text{cm}^4 \text{s}^{-1}$ . A conservative estimate of the focus spot size (radius  $\approx 1 \mu\text{m}$ ) of the LCLS yields a photon flux of  $10^{33} \text{cm}^{-2} \text{s}^{-1}$ , which yields a value of the 2-photon transition probability of 10%. By similar arguments, it can be shown that the LCLS can also drive higher-order processes. The ability to study inner shell processes using nonlinear spectroscopy as a routine probe will advance our knowledge of complex electron systems.

In addition, we will explore the application of x-ray nonlinear processes for advancing the direct characterization of the LCLS x-ray pulse using a variety of known temporal metrological schemes, e.g. autocorrelation and FROG. Cross-correlation methods can also be envisioned using a synchronized CPA laser to provide critical timing information.

Further, new approaches to x-ray pulse metrology will be explored. X-ray plus visible-light sum frequency generation, for instance, can lead to temporally shaped x-ray pulses (using shaped visible pulses) offering new approaches to coherent control of chemical reactions while also offering the new capability of probing valence charge distributions via (x-ray/visible) nonlinear diffraction (see Molecular Structure section).

The AMOS team will perform preliminary nonlinear studies using different venues, e.g. SPPS, APS and optimized harmonic sources. These sources, although not completely equivalent to the LCLS output, will provide important benchmarks.

### *X-RAY DRIVEN CLUSTER DYNAMICS*

One of the most important and exciting ultimate applications of the LCLS will be to image single, large biological molecules. The calculations performed by Neutze et al.<sup>1</sup> indicate that such single molecule imaging will be feasible at LCLS provided that the x-ray pulse can scatter enough photons prior to the radiation driven disassembly of the molecule. Their simulations indicated that pulse durations as short as 10 fs may be needed to achieve this. This conclusion was based on very simple modeling of the disassembly of the molecule assuming that all photoionization events were accompanied by an immediate ejection of the freed electron well out of the molecule. The consequence of the large x-ray flux needed for imaging is that rapid photoionization leads to a very fast Coulomb explosion which drives the molecule apart in a few tens of femtoseconds. This simulation suggests that the base line LCLS parameters will not be adequate for this kind of imaging and that the 200 fs LCLS pulse is too long.

We propose to simulate the conditions relevant to the explosion of large biological molecules (thousands or tens of thousands of atomic constituents) by studying simpler atomic clusters such as van der Waals bonded rare gas or small molecular clusters. In recent years, considerable progress has been

made in understanding the explosion of large (many thousand atom) clusters when irradiated by intense optical or near IR ultrafast laser pulses. These studies have shown that the dynamics of a cluster explosion can be more complicated than the simple Coulomb explosion picture assumed in the bio-molecule calculations of Neutze et al.<sup>1</sup> It is known that in small, low Z clusters (such as thousand atom or less hydrogen clusters) irradiated at high intensity, a Coulomb explosion does occur as the strong laser ponderomotive potential is sufficient to eject all of the optically ionized electrons from the cluster soon (few fs) after they are produced. However, with larger clusters, space charge forces and multiple scattering from the cluster atoms can confine electrons to the cluster after optical ionization. This yields a nanoplasma during the laser pulse. The explosion time of this hot nanoplasma can be quite different than that of ions repelling each other by Coulomb forces; the confined electrons can serve to shield the ion charges and the expansion is ultimately driven, instead, by the electron pressure (which is a function of the electron temperature in the nanoplasma). This explosion time can be substantially longer than would be predicted by a simple Coulomb explosion, confirmed by optical pump-probe experiments<sup>4</sup>.

X-ray irradiation of a cluster may undergo similar steps, however the very high photon energy and very low associated ponderomotive energy of the pulse will certainly lead to dynamics quite different than those of IR irradiation. How an intense x-ray pulse will drive the explosion of a cluster, or similarly sized bio-molecule is not well understood but is, no doubt, more complex than the simple Coulomb explosion picture. In fact, a simple consideration of the physics of an x-ray driven explosion indicates that the Neutze calculation may have overestimated the velocity of the large molecule disassembly. In an x-ray irradiation of the cluster, photoionization will release electrons with energy of a few keV. The initial electrons will leave, charging the cluster sphere to a potential of a few keV. Further ionization events will create electrons that cannot overcome the space charge confinement of the cluster. This will lead to the formation of a nanoplasma, which will equilibrate through inelastic collisions on a time scale of perhaps a few tens of femtoseconds. Because of the low ponderomotive potential of the x-ray pulse, the nanoplasma will probably persist for some time, though, just how long it will persist will depend on electron heating rates in the x-ray pulse (which are not well known). Initial experiments by Wabnitz et al.<sup>5</sup> on exploding clusters driven by the DESY VUV-FEL show evidence that a warm nanoplasma was indeed formed in irradiated Xe clusters. The observed response of the cluster to VUV-FEL radiation was modeled by including atomic-structure, plasma-screening and inverse bremsstrahlung effects.<sup>6</sup> It is also known that collective electron effects are important in the dynamics of IR irradiated large clusters. Whether these will play a role in short wavelength interactions remains another open question.

#### *STATIC & TIME-RESOLVED X-RAY IMAGING OF MOLECULAR STRUCTURE*

Characterization of the dynamics of molecular bonds in the process of chemical transformation is one of the most interesting and challenging problems in chemistry and molecular physics. Transition state chemistry (i.e. understanding intermediate chemical structures) is an important scientific frontier as evidenced by the 1999 Nobel Prize in chemistry, which was awarded for the use of visible light to probe the transition state. Because it primarily probes the delocalized valence electrons in a molecule, visible light provides only indirect information regarding structure. In contrast, x-rays can provide direct structural information via x-ray diffraction and absorption, and the application of these techniques on an ultra-fast timescale represents an important scientific frontier in the study of chemical and biochemical reactions. The potential for increased knowledge in an area of great importance is difficult to overestimate. Thus, the ultra-fast x rays of the LCLS are expected to play a unique and unparalleled role in probing transient structural dynamics in systems of chemical and biochemical importance. Furthermore, if the time-scale of these intense x-ray sources is short enough ( $\leq 5$  fs), it may be possible to image<sup>1</sup> “single” complex macromolecules, e.g. proteins, thus circumventing the need for crystallization.

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<sup>4</sup> J. Zweiback, T. Ditmire, and M. D. Perry, *Opt. Express*, **6**, 236 (2000).

<sup>5</sup> H. Wabnitz, L. Bittner, A. R. B. De Castro, *et al.*, *Nature* **420**, 482 (2002).

<sup>6</sup> R. Santra and C.H. Greene, *Phys. Rev. Lett.* **91**, 233401 (2003).

X-rays from synchrotron sources are the tool of choice for structural determination since scattering occurs mostly from the tightly bound electrons around the cores, thus mapping the atomic positions. Furthermore, resonant enhanced scattering provides atomic specificity. The LCLS's high-flux, ultra-fast x-rays will revolutionize our understanding of transition state chemistry by providing snapshots of the molecule as it transforms. For example, one area of particular interest is the study of conical intersections between the ground and excited state manifolds. These are the regions along the dynamical reaction coordinate(s) that enable a broad class of non-radiative chemical reactions. Conical intersections are very poorly understood, and have been measured in only a few cases, involving valence, Rydberg, or low energy continuum electrons. High time resolution x-ray studies could provide an extraordinary probe of these critical regions for inner-shell processes.

We believe that proper choice of molecular samples, e.g. organometallics or halogenated organics, will allow quick realization of a molecular dynamics program using baseline LCLS parameters. As one example, the LCLS wavelength tuning accesses a number of absorption edges for atomic specificity. The number of photons will be high enough to collect information about structure easily in a few seconds, or even with a single x-ray pulse. The pulse duration, while specified at an uncomfortably long 200 fs, will likely be much shorter through improvements of basic techniques of manipulating the electron beam. Proposals using emittance-spoiling insertion devices on the LCLS could result in x-ray pulses as short as 2 fs<sup>7</sup>. This would be sufficient to cover the range of principal interest in chemical dynamics.

There have been a number of recent advances in optical techniques for aligning and orienting molecules.<sup>8</sup> In general, the interaction of a molecule with an anisotropic polarizability with a sufficiently intense laser pulse (usually in the near infrared) will result in the alignment of the molecule. The development of these techniques will ultimately allow the preparation of a molecular sample in a specified orientation, thus simplifying the analysis of the resulting x-ray experiments. Scattered x-rays (in the Laue configuration) as well as the momentum of ions originating from Coulomb explosion will be detected as a function of orientation. In addition, the dynamics of molecular behavior and alignment in a laser field will become accessible with unprecedented temporal and spatial resolution. As compared to experiments with single molecules, the ordered structure of aligned molecules will greatly enhance the sensitivity for x-ray structure analysis for small molecules, while alignment of individual large molecules would provide well-defined initial conditions.

### 3. AMO PHYSICS END-STATION CONCEPTUAL DESIGN

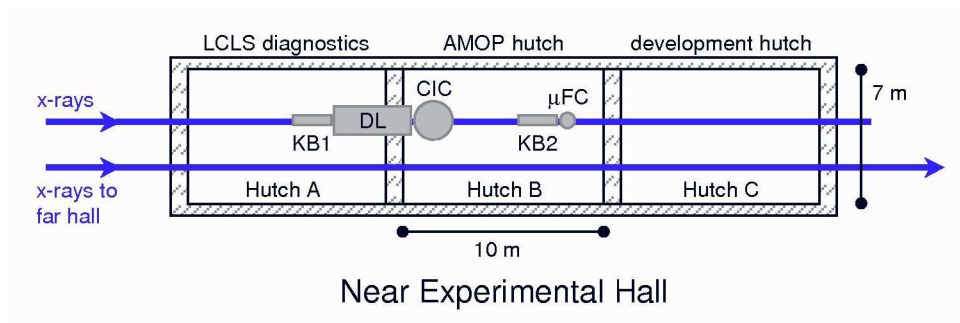


Figure 1: Schematic layout of the atomic physics end-stations in Hutch B of the Near Experimental Hall. The cluster interaction chamber (CIC) will utilize a long focal length (5 m) Kirkpatrick Baez (KB1) mirror pair in conjunction with the x-ray beam two beam pump probe delay line module (DL). The  $\mu$ -focus AMO chamber ( $\mu$ FC) will use a short focal length Kirkpatrick Baez (KB2) mirror pair to achieve tight focusing conditions.

The AMO physics end station will utilize the initial output parameters of the LCLS, and also have significant flexibility to accommodate future experiments. Technical constraints dictate two independent

<sup>7</sup> P. Emma, K. Bane, M. Cornacchia *et al*, *Phys. Rev. Lett.* **92**, 074801 (2004).

<sup>8</sup> H. Stapeldeldt and T. Seideman, *Rev. Mod. Phys.* **75**, 543 (2003).

chambers located in tandem on the same x-ray beam line, referred to as the  $\mu$ -focus chamber ( $\mu$ FC) and the cluster interaction chamber (CIC). The required x-ray beam size and near proximity to the electron beam dictate that the AMOP end-station is located in the Near Experimental Hall, as shown in Fig. 1. The CIC is placed adjacent to the x-ray pump-probe delay line (DL) and long focal length Kirkpatrick-Baez (KB1) mirror pair that are planned for location in LCLS Near Hall Hutch A.

### GENERAL END-STATION ISSUES

We identified a number of general issues that will not only be critical to the success of the AMOP program but will also have an impact on overall LCLS operations.

1. Effective x-ray beam attenuator and harmonic rejector.
2. Single-shot x-ray carrier frequency & bandwidth monitor
3. Single-shot x-ray energy/intensity monitor, position and spot size diagnostics
4. Monochromatized x-ray beam,  $10^{-3}$  at 800 eV and  $10^{-4}$  at 8000 eV for resonant studies
5. X-ray beam energy should be stepwise tunable, e.g. 800 - 1000 eV, with stepsize  $\leq$  bandwidth
6. Precision trigger ( $\Delta t \sim 100$  fs) relative to x-ray beam available to experimental end-stations. Many experiments will require a substantial pre-trigger, for pre-excitation, molecular alignment, or similar preparation of the sample. In some cases such as impulsive alignment, the pretrigger timing jitter is just as important as pump-probe jitter.
7. A single-shot monitor of the relative time delay between x-ray beam & laser beam: Jitter measurements down to femtoseconds or even below are highly desirable, and the level of jitter is an important trade-off in design.
8. Delay line for x-ray dual beam pump-probe experiments, time delay  $\pm 20$  ps, resolution 0.1 fs
9. Focusing element (Kirkpatrick-Baez mirror pair) to produce 100 nm spot size
10. Terawatt CPA laser system located above Hutch B in Near Experimental Hall
  - (a) synchronization to x-ray beam  $< 500$  fs
  - (b) 40 fs pulse duration and 5 fs option with gas filled hollow core fiber
  - (c) tunable wavelength operation via nonlinear conversion, e.g. NOPA
  - (d) shielded optical access ports between laser room and Hutches A-C
  - (e) transport between laser room and hutch should minimize pointing instability via relay imaging
  - (f) high powered laser beams must be present in some experiments. In many cases it will be desirable to do the final optical pulse compression in the x-ray hutch
  - (g) full characterization of the sub-femtosecond structure of the XFEL would be facilitated by a carrier-stabilized femtosecond laser source.
11. Vibration isolated footings near hutches for high throughput pumps.

### 3.1 $\mu$ -FOCUS AMO CHAMBER DESIGN

The primary purpose of the  $\mu$ -focus chamber is to explore the interaction of high intensity x-ray radiation with matter. This consideration drives the design of this chamber and differentiates it from the cluster interaction chamber that has less stringent focusing and vacuum requirements. Estimates to limit the number of allowable background ionization events to one per shot, indicate a permissible background pressure of  $10^{-10}$  torr, i.e. UHV.

We plan to use the standard focusing method for highest flux density, Kirkpatrick-Baez (KB) mirror pairs. State-of-the-art focusing has achieved spot diameters of 100 nm, with an input acceptance of  $\approx 200$ -400  $\mu$ m. This is well matched to the x-ray beam size in the Near Experimental Hall. The small spot size necessitates a short working distance of  $\approx 50$  mm, which in turn forces the KB mirrors to be reentrant to the UHV  $\mu$ -focus chamber. This is a major design consideration, the details of which are to be worked out in partnership with the LCLS optics designers as part of baseline operation. While the goal is to have a focused beam diameter  $\approx 100$  nm, nonlinear processes will already be apparent at beam sizes  $\approx 10$   $\mu$ m.

The layout for  $\mu$ -focus experiments is shown in Fig. 2. Proceeding downstream, there is a differential pumping region, the KB mirror pair, the interaction region and the x-ray beam diagnostics. The KB

mirror pair focuses and deflects the x-ray beam (roughly 4-6 mrad for 8 keV x rays). The chamber/interaction region must follow the deflected x-ray beam and thus is mounted on a remotely controllable table, similar to those used at the APS. The x-rays exit the chamber through a small aperture to beam diagnostics.

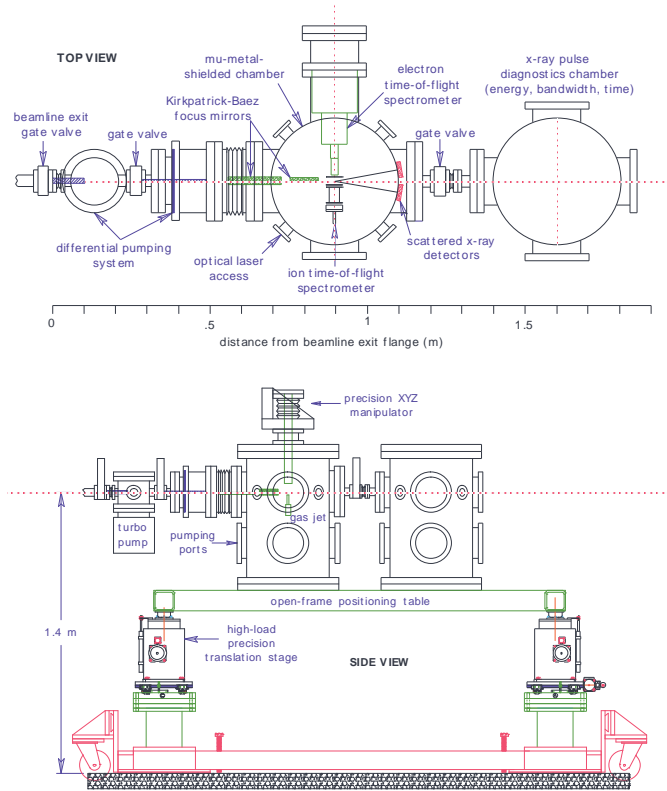


Figure 2: Top and side view of the proposed experimental layout for high intensity atomic physics experiments using the  $\mu$ -focus chamber.

Detection of the products (ions, electrons and photons) and provision for optical access with lasers define the remaining body of the chamber. A multi-port octagon chamber configuration allows access from many angles for electron TOF, x-ray detectors and lasers. Ion momentum imaging with  $4\pi$  collection efficiency for coincidence and singles measurements will be incorporated. Typical operating parameters follow: the electron TOF,  $\Delta E/E \approx 10^{-4}$  at  $E = 1$  keV; x-ray spectrometers in the 300-1000 eV range, resolving power of 1300, ion imaging with sub-meV resolution and multi-hit capability. The large port sizes can accommodate novel sources, such as skimmed supersonic jets for cooled molecules and chromophore-containing helium droplets.

### 3.2 CLUSTER INTERACTION AMO CHAMBER DESIGN

Since one of the most important scientific questions is understanding the time scale of the cluster explosion, temporal pump-probe diagnostics will be the central design driver of this chamber. The chamber will require the focusing of the LCLS beam into a cluster beam, formed by a pulsed gas jet. It should be possible to derive some information on explosions by probing the x-ray irradiated clusters with a synchronized femtosecond laser pulse to do time-resolved Rayleigh scattering and optical absorption (akin to that of ref. 4). However, limitations in the time synchronization between x-ray pulse and laser as well as the limited information that can be derived from the scatter of an optical pulse indicate that the *optimum experiment will involve x-ray pump and x-ray probe*. For this reason, we propose to develop an x-ray optical package that will allow us to irradiate the clusters with the x-ray pulse and then interrogate them with a delayed part of the initial pulse. Using spatially resolving x-ray detectors, it will be possible

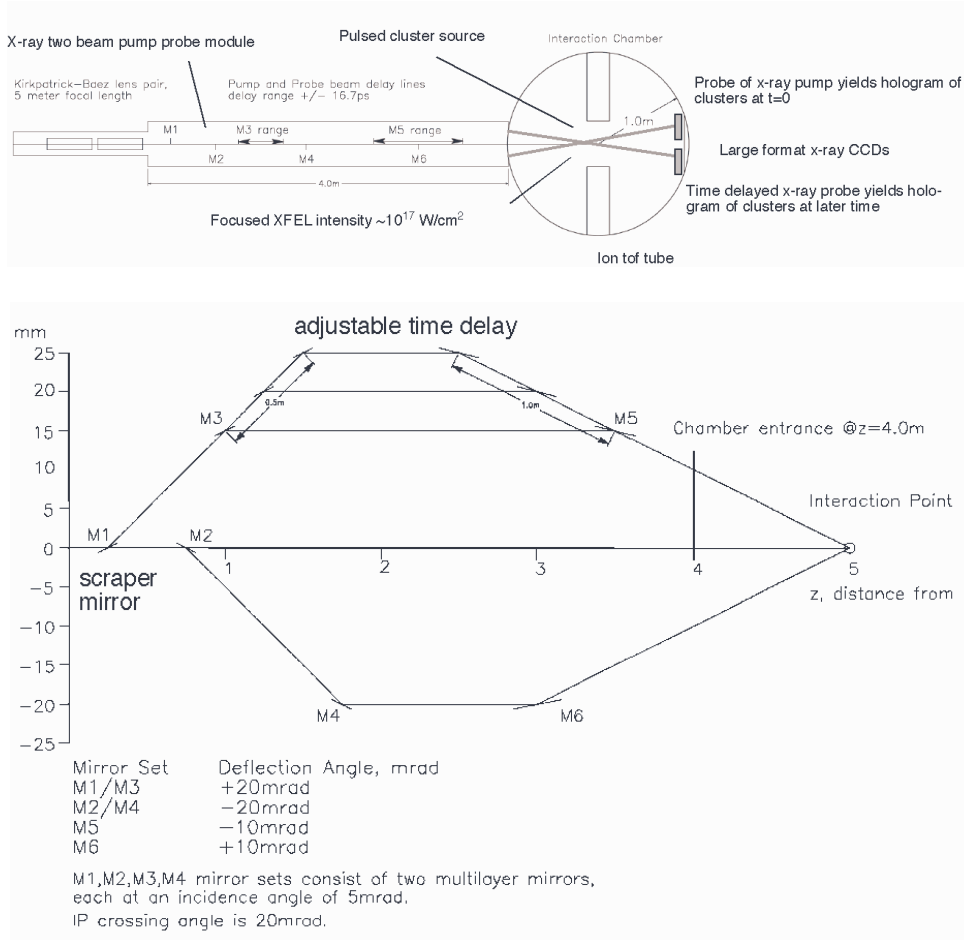


Figure 3: Proposed (top) cluster interaction chamber and (bottom) two-beam x-ray pump-probe module for the LCLS AMOP studies.

to derive information about the size of the cluster both initially (by detection of the forward angle scatter of the x-ray pump) as well as the size of the exploding cluster at some time delay (by observation of the forward scatter of the x-ray probe).

The target chamber proposed for this class of experiment is illustrated in the top of Fig. 3. The chamber will be designed to field a flexible set of diagnostics. It will be a saucer-shaped chamber of 1 m radius. The x-rays will be focused with a KB pair into the chamber. A two-beam x-ray pump-probe module will be inserted between the focusing optics and the interaction region. While the specific design of the device will require R&D, one potential configuration is illustrated in the bottom of Fig. 3 in which the x-ray beam is split with a “scraper optic” which sends part of the x-ray pulse into a temporally delayed arm which crosses the x-ray pump pulse at a small angle at the interaction point in the center of the chamber. With this geometry, forward angle x-ray scatter detectors can be fielded on both the pump and probe beams. Ion and electron TOF spectrometers will be situated on axes perpendicular to the incoming x-ray beams. The chamber will also have sufficient access to allow laser probing of the clusters from a number of different angles. This chamber will also be useful for future, solid target experiments e.g. those interested in warm dense matter.

#### 4. LCLS AMOS TEAM

There are seven institutions in the AMOS team with 18 total members:

**Ohio State University:** Louis DiMauro (*dimauro@bnl.gov*), Linn Van Woerkom & Pierre Agostini

**University of Michigan:** Philip H. Bucksbaum (*phb@umich.edu*), David Reis & Roy Clarke

**Argonne National Laboratory:** Linda Young, Steve Southworth, Paul Fuoss, Bertold Kraessig, Elliot Kantor & Steve Pratt

**Lawrence Berkeley National Laboratory:** Ernie Glover

**Texas A&M:** Gerhard Paulus, Alexei Sokolov & John Reading

**University of Texas, Austin:** Todd Ditmire

**University of Colorado, Bolder:** Chris Greene

**AMOS Tasks:** The AMOS team has considerable depth of expertise in all aspects of the proposed experiments. The list below shows the principal responsible investigators (PRI), but it should be understood that we will work as a team, and all sub-groups are interdependent.

The main development and implementation of the experiments will be divided into three sections: Theory, X-ray Physics, and Ultra-fast Laser Physics. The divisions are not sharp in many areas, but they help to organize the work.

**Project co-Directors:** Louis DiMauro & Phil Bucksbaum

**AMO Theory:** Chris Greene & John Reading

**X-ray Physics:**

*Inner shell atomic physics:* Linda Young, Steve Southworth, Bertold Kraessig & Elliot Kantor

*Molecular physics:* Steve Pratt

*X-ray transport physics:* Paul Fuoss & Roy Clarke

**Ultrafast Laser Physics:**

*Coherent control physics:* Phil Bucksbaum

*Cluster and plasma physics:* Todd Ditmire

*Nonlinear optics:* Ernie Glover

*Laser-X-ray pump-probe physics:* David Reis

*Strong-field physics:* Pierre Agostini, Linn Van Woerkom & Louis DiMauro

*Attophysics:* Alexei Sokolov & Gerhard Paulus

## 5. BUDGET SUMMARY

The AMOS collaboration will make full use of the atomic physics end-stations in the LCLS baseline design. Equipment needs over and above the budget for the baseline stations will enable expansion of the capabilities of those instruments beyond the baseline needs of LCLS, and help to launch the field of ultra-fast and high field AMO x-ray physics.

### *BASELINE EQUIPMENT AND OPERATION BUDGET:*

Two chambers are available for AMO activities in the baseline design of LCLS. The  $\mu$ -focus AMO chamber is designed for utilizing the unprecedented intensity capabilities of the LCLS x-ray beam. Interaction of a tightly focused x-ray beam with atoms and molecules will provide groundbreaking experiments in the high frequency limit. The general design of the  $\mu$ -focus chamber allows complete kinematic determination of the photofragmentation process and places rigorous requirements on the vacuum integrity, e.g. UHV.

The cluster interaction AMO chamber will explore the interaction of intense, ultra-fast x-rays with large clusters and solid targets. Analysis of the energetic photofragments will be accomplished with charge particle analyzers and a suite of x-ray diagnostics will monitor the scatter beam from x-ray pump-probe experiments. The need for a high backing pressure nozzle for large cluster size production and the broader suite of diagnostics requires a larger vacuum workspace with reduced vacuum pressure requirements as compared to the  $\mu$ -focus AMO chamber.

The projected expenses for the first three years permit a build-up period for the AMOP experimental program. These funds will be raised by grant requests, primarily to DOE and NSF. In addition, we will take advantage of considerable existing infrastructure at the Michigan FOCUS Center, the proposed Ohio

State Attoscience Center, and the research groups at the other institutions. Matching funds will be sought from the participating institutions.

**ONE YEAR BUDGET AMOS TEAM:**

<i>Project Direction (DiMauro, Bucksbaum)</i>	<i>Cost, k\$</i>
Partial summer salaries (incl. benefits)	\$30

<i>Elementary intense field atomic inner-shell photoionization</i>	<i>Cost, k\$</i>
Summer Salaries (Young, Southworth, Kantor, Kraessig)	\$40
Students and postdocs (includes benefits) (2+1)	\$120
Materials and supplies	\$50
Travel and misc.	\$10

<i>X-ray nonlinear processes</i>	<i>Cost, k\$</i>
Summer Salaries (Clark, Fuoss, Glover, Agostini)	\$40
Students and postdocs (includes benefits) (2+1)	\$120
Materials and supplies	\$50
Travel and misc.	\$10

<i>X-ray driven cluster dynamics</i>	<i>Cost, k\$</i>
Summer salaries (Ditmire, Reis, Van Woerkom, Greene)	\$40
Students and postdocs (includes benefits) (2+1)	\$120
Materials and supplies	\$50
Travel and misc.	\$10

<i>Static &amp; time-resolved x-ray imaging of molecular structure.</i>	<i>Cost, k\$</i>
Summer Salaries (Sokolov, Paulus, Reading, Pratt)	\$40
Students and postdocs (includes benefits) (2+1)	\$120
Materials and supplies	\$50
Travel and misc.	\$10

<b>Subtotal</b>	<b>\$910</b>
IDC (estimate 40%)	\$364
<b>Total 1 Year</b>	<b>\$1,274</b>
<b>Subtotal 3 Years:</b>	<b>\$3,822</b>
Equipment	\$1,000
Less cost sharing	\$600
<b>TOTAL 3 YEARS</b>	<b>\$4,220</b>

**We require the LCLS baseline chambers for the AMOP program<sup>†</sup>:**

μ-focus chamber	\$1,138
cluster chamber	\$1,725

<sup>†</sup> The AMO chambers are part of the requested end station equipment for the LCLS baseline project. At the time of this writing the budget has not been fixed. Once resolved, the AMOS team will coordinated the construction with the LCLS design team and if necessary will seek additional external funds for completion of the AMOP end station.