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Final Report of LDRD Project:

Compact Ultrabright Multikilovolt X-Ray Sources for Advanced Material Studies, 3D Nanoimaging,and Attosecond X-Ray Technology

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Abstract

Experimental evidence and corresponding theoretical analyses have led to the conclusion that the system composed of Xe hollow atom states, that produce a characteristic Xe(L) spontaneous emission spectrum at l @ 2.9 Å and arise from the excitation of Xe clusters with an intense pulse of 248 nm radiation propagating in a self-trapped plasma channel, closely represents the ideal situation sought for amplification in the multikilovolt region.

The key innovation that is central to all aspects of the proposed work is the controlled compression of power to the level (~ 10^{20} W/cm^3) corresponding to the maximum achieved by thermonuclear events. Furthermore, since the x-ray power that is produced appears in a coherent form, an entirely new domain of physical interaction is encountered that involves states of matter that are both highly excited and highly ordered. Moreover, these findings lead to the concept of "photonstaging," an idea which offers the possibility of advancing the power compression by an additional factor of ~ 10^9 to ~ 10^{29} W/cm^3. In this completely unexplored regime, g-ray production ($\hbar\omega_\gamma \sim 1$ MeV) is expected to be a leading process. A new technology for the production of very highly penetrating radiation would then be available.

The Xe(L) source at $\hbar\omega_X \sim 4.5$ keV can be applied immediately to the experimental study of many aspects of the coupling of intense femtosecond x-ray pulses to materials.

In a joint collaboration, the UIC group and Sandia plan to explore the following areas. These are specifically, (1) anomalous electromagnetic coupling to solid state materials, (2) 3D nanoimaging of solid matter and hydrated biological materials (e.g. interchromosomal linkers and actin filaments in muscle), and (3) EMP generation with attosecond x-rays.

The specific tasks/milestones were:

A. Examine the coupling to solids in two stages; initially, with readily performed screening studies utilizing foils, to examine the gross response, and subsequently with more detailed experimental analyses. Specifically, we plan to examine Al, Ti, Fe, Cu, Sn, Pb, Ta, Au, and U. For sufficiently thin foils, film packs would be constructed similar to that shown in Fig. (1). A main point of interest is the detection of a strengthening of the radiative coupling to the solid over that conventionally predicted. The materials Sn and U are particularly interesting, since they respectively have edges at 4465 eV (L_1) and 4304 eV (M_3) that fall within the 2.7 – 2.9 Å range.

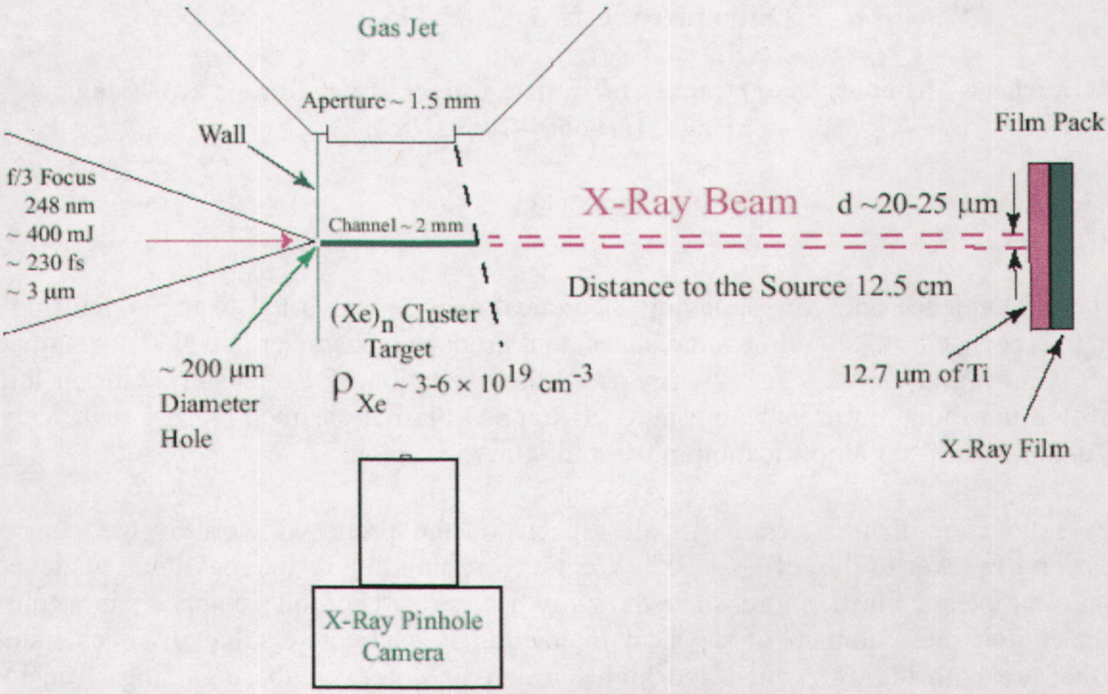

Fig. (1): Experimental configuration used for the observation of amplification of Xe(L) radiation in self-trapped channels inside an evacuated chamber. The x-ray pinhole camera was equipped with a ~ 10 μm thick Be foil enabling the morphology of the channel to be visualized by the Xe(M) emission (~ 1 keV). The observed channel length typically is { ≅ 1.5–2.5 mm. The wall defining the entrance plane having the 200 μm aperture was fabricated from ~100 μm thick steel and the incident 248 nm pulse was focused with an f/3 off-axis parabolic mirror to a spot size of ~ 3μm. The average Xe density in the target was estimated to be $\rho_{Xe} \sim 3 - 6 \times 10^{19}$ cm^{-3}. The location of a film pack used for measurement of the amplified x-ray beam composed of a 2 cm square 12.7 μm thick foil backed by a matching piece of RAR 2492 film is shown. This detector was placed on the channel axis in a perpendicular orientation at a distance of 12.5 cm from the cluster target. For a channel diameter of 2–3 μm, the location of the film pack corresponds to the far field of the source. Typical single-pulse damage caused on the Ti foil by an incident x-ray pulse had a diameter d ≅ 20–25 μm.

B. Perform x-ray imaging studies of hydrating (living) biological structures. We plan to conduct our initial experiments on chromolinkers, presently hypothesized entities that are believed to play a key role in chromosomal organization.

C. Extend the single-pulse measurements to evaluate the ability to produce attosecond x-ray pulses with a length < 100 as for studies of EMP generation. We expect that the EMP effect can be calibrated. This calibration, along with the ability to record the pulse energy, gives a way to derive the temporal pulse duration. This would enable a new time measurement technology for short x-ray pulses to be developed that is applicable to the attosecond regime.

D. Prepare a SAND report.

CONTENTS

1) Project Overview

Scientific and Technical Soundness

Experimental evidence and corresponding theoretical analyses have led to the conclusion that the system composed of Xe hollow atom states, that produce a characteristic Xe(L) spontaneous emission spectrum at $\lambda \cong 2.9$ Å and arise from the excitation of Xe clusters with an intense pulse of 248 nm radiation propagating in a self-trapped plasma channel, closely represents the ideal situation sought for amplification in the multikilovolt region.

Experimental data of several forms have been obtained which constitute conclusive concordant evidence of amplification in a self-trapped plasma channel on several transitions of the Xe(L) hollow atom spectrum at wavelengths in the range $\lambda \sim 2.71$–2.93 Å [Borisov *et al.*, "Ultrabright Multikilovolt Coherent Tunable X-Ray Source at $\lambda \sim 2.71$–2.93 Å," *J. Phys.* B 36, 3433 (2003)]. Specifically, they are (α) strongly enhanced spectra arising from Xe^{31+}, Xe^{32+}, Xe^{34+}, Xe^{35+}, Xe^{36+}, and Xe^{37+} ions recorded from the channel in the forward (axial) direction, as shown, for example, for the Xe^{34+} array at $\lambda \sim 2.88$ Å and the Xe^{32+} array at $\lambda \cong 2.71$ Å, (β) the measurement of a corresponding spectral narrowing on the directionally enhanced lines emitted from the channel, (γ) evidence for saturation of three of these transition arrays (Xe^{34+}, Xe^{35+}, Xe^{36+}) given by the simultaneous quenching (spectral hole-burning) of the corresponding spontaneous emission from them in transversely recorded spectra only when amplifying channels are present, observations that correlate fully with the measured spectral narrowing, and (δ) small-scale structural damage to both (i) the 12.7 μm thick Ti foil located at a separation of ~ 2.5 cm from the source at the entrance to the axially located von Hámos spectrometer and, with the von Hámos spectrometer removed, (ii) similar Ti foils protecting film packs axially positioned at a distance of 12.5 cm from the cluster target. In the case involving the Ti foil shielding the spectrograph, a canonical spatial mode pattern has been recorded consisting of a regular circular array of holes pierced in the foil having an individual feature size of ~ 1 μm and an overall diameter of ~ 5 μm. This observation (a) indicates a divergence of $\delta\theta_x \cong 0.2$ mr, a value approximately twice that expected from a coherent aperture formed by a channel with a diameter of 2–3 μm that radiates at a wavelength ~ 2.9 Å, and (b) demonstrates the spatial coherence of the amplified x-rays. The detection of the amplified beam with the film packs at a distance of ~ 12.5 cm from the source confirms the divergence of $\delta\theta_x \cong 0.2$ mr. In this case, since the damage to the Ti foil is not fully penetrating, the morphology of the physical damage produced on the Ti surface is matched by the shape of the exposure of the transmitted x-rays on the film located directly behind it. Further, the observed damage to the Ti foils is consistent with both the estimated saturation flux of ~ 10 J/cm^2 for the Xe(L) transitions and the observation of the spectral hole-burning [Borisov *et al.*, "Saturated Multikilovolt X-Ray Amplification with Xe Clusters: Single-Pulse Observation of Xe(L) Spectral Hole-Burning," *J. Phys.* B 36, xxx (2003)]. An estimate of the peak spectral brightness achieved in these experiments gives the value of ~ 10^{31}–10^{32} photons·s^{-1}·mm^{-2}·mr^{-2} (0.1% Bandwidth)$^{-1}$.

Of particular significance are the single-pulse measurements of spectral hole-burning of the Xe(L) 3d→2p hollow atom transition arrays observed from a self-trapped plasma channel, since they provide new information on the dynamics of saturated amplification in the $\lambda \sim 2.8$–2.9 Å region. Specifically, these results (a) demonstrate a very favorable energy efficiency and (b) point to the potential for development of a new attosecond x-ray technology. Of chief importance is the fact that the spectral hole-burning on transitions in the Xe^{34+} and Xe^{35+} arrays reaches full suppression of the spontaneous emission and presents simultaneously a corresponding width $\Delta\hbar\omega_x \cong 60$ eV, a value adequate for efficient amplification of multikilovolt x-ray pulses down to a limiting length $\tau_x \sim 30$ as. The depth of the suppression at 2.86 Å indicates that the gain-to-loss ratio is ≥ 10. Further, an independent determination of the x-ray pulse energy from damage produced on the surface of a Ti foil in the far field of the source gives a pulse energy of 20–30 μJ, a range that correlates well with the observation of the spectral hole-burning and indicates an overall extraction efficiency of ~ 10%.

Creativity and Innovation

The key innovation that is central to all aspects of the proposed work is the controlled compression of power to the level (~ 10^{20} W/cm^3) corresponding to the maximum achieved by thermonuclear events. Furthermore, since the x-ray power that is produced appears in a coherent form, an entirely new domain of physical interaction is encountered that involves states of matter that are both highly excited and highly ordered. Moreover, these findings lead to the concept of "photon-staging," an idea which offers the possibility of advancing the power compression by an additional factor of ~ 10^9 to ~ 10^{29} W/cm^3. In this completely unexplored regime, γ-ray production ($\hbar\omega_\gamma \sim 1$ MeV) is expected to be a leading process. A new technology for the production of very highly penetrating radiation would then be available.

Project Plan

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In a joint collaboration, the UIC group and Sandia plan to explore the following areas. These are specifically, (1) anomalous electromagnetic coupling to solid state materials, (2) 3D nanoimaging of solid matter and hydrated biological materials (e.g. interchromosomal linkers and actin filaments in muscle), and (3) EMP generation with attosecond x-rays.

Impact

Experimental evidence definitely exists for amplification of ~2.9 Angstrom radiation form Xe(L) hollow atom states using self-focussing ultraviolet terawatt excitation. This new physics is a revolutionary step in developing a new class of table-top coherent imaging tools with high spectral brightness for materials analysis and biological studies. Extremely promising for x-ray production and selective atomic coupling, detailed experiments investigating the complex interactions of short-pulse self-focusing laser pulses with ordered matter have assessed the viability of this technology for scaling to higher brightness and quantum energies suitable for probing high-energy density environments.

The proposed work is designed to take advantage of the unique high brightness x-ray source now available at UIC to explore several areas important to (1) the microstructure analysis of materials and plasmas, (2) new 3D modalities of nanoscale imaging applicable to biological materials, (3) new anomalous modes of electromagnetic coupling to materials, and (4) EMP generation with pulses in the attosecond range.

2) Multikilovolt Channeled X-Ray Propagation in Water

Water is a nearly ideal medium for the production of confined modes of propagation with the relativistic/charge-displacement mechanism in the multikilovolt x-ray region. Calculations for λx ~ 2.9 Å anchored to experimental data in the ultraviolet (248 nm) region yield stable channels having a characteristic diameter of ~ 190 Å, a propagating intensity of ~ 1026 W/cm2, and a loss parameter due to photoionization of ~ 0.3 mJ/cm. These values are consistent with the generation of x-ray channels conducting coherent x-ray fluences > 1010 J/cm2 in water over a length of ~ 1 m, a range a factor of ~ 104 greater than the distance associated with linear propagation.

The ability to produce stable self-channeled propagation of intense pulses of radiation in an underdense plasma is a well established phenomenon at quantum energies below ~ 5 eV [1-5]. The recent observation [6,7] of strong amplification on Xe(L) hollow atom transitions in the multikilovolt spectral region (~ 4400 eV) at λ_x ~ 2.71–2.93 Å introduces the possibility of extending the generation of such stable highly confined ordered modes of deeply penetrating propagation to x-ray wavelengths in materials at solid density. The importance and abundance of water, in either liquid or solid form, nominate it as a leading system for study. Indeed, recent experiments [8] in water have revealed the existence of an unexpected mode of propagation involving chaotic filamentation at 527 nm based on a dynamic process of pulse evolution [9].

In order to achieve channeled propagation of intense x-rays in condensed matter over an appreciable scale length, we require a physical process that dynamically combines (1) a very high electron density $N_{e,0}$ characteristic of the solid state, (2) a high propagating intensity I_x, (3) a stable mechanism of power compression that naturally seeks a small transverse scale length Δ for the confinement of the radiative energy, and (4) a small loss parameter. The demonstrated properties [1-4] and scaling behavior [10] of relativistic/charge-displacement self-channeling in the underdense plasma regime ($N_{e,0} < N_{cr}$) in a low-Z solid medium fully satisfy these four demands.

At a wavelength $\lambda_x \sim 2.9$ Å, for which the critical electron density is $N_{cr} = 1.33 \times 10^{28}$ cm^{-3}, all fully ionized condensed matter is underdense, including uranium, even at a compression of ~10^3-fold over the normal metallic state, a condition that corresponds to $N_{e,0} \sim 4.38 \times 10^{27}$ cm^{-3}. Importantly, the characteristics of the relativistic/charge-displacement process are well established both theoretically and experimentally in the underdense regime ($N_{e,0} / N_{cr} \leq 0.05$) by several previously conducted studies [1-5,10-12] at the wavelength of 248 nm ($\hbar\omega$ = 5 eV). Accordingly, the observational findings available from the work at 248 nm act as a firm physical anchor for projections into the x-ray regime.

The key requirement [1-4,11] for the production of a channel is the generation of a peak power P_o exceeding the critical power P_{cr} necessary for the development of the confined propagation with the relativistic/charge-displacement mechanism given by

$$P_{cr} = (m_{e,0}^2 c^5 / e^2)\int_0^\infty g_0^2(\rho)\,\rho d\rho\,(\omega/\omega_{p,0})^2 = 1.6198 \times 10^{10}\,(\omega/\omega_{p,0})^2 \text{ W}. \qquad (1)$$

In Eq.(1), $m_{e,0}$, c, and e have their standard identifications, $g_0(\rho)$ is the Townes mode [13] representing the lowest eigenmode for cubic Kerr self-focusing, and ω, $\omega_{p,0} = (4\pi e^2 N_{e,0} / m_{e,0})^{1/2}$, respectively denote the angular frequency at electron density $N_{e,0}$

corresponding to the propagating radiation and the angular frequency of the unperturbed plasma. For the case of $\lambda_x \cong 2.9$ Å and fully ionized H_2O, Eq. (1) gives $P_{cr} \cong 434$ TW.

With exclusion of experimentally unbecoming materials (i.e. liquid/solid H_2, liquid He, and solid Li), the optimal medium for the formation of high power x-ray channels of an extended length in condensed matter is the low-Z solid Be with Z = 4, since the losses associated with ionization would be minimal. However, water (H_2O) with 10 electrons and a maximal value of Z = 8, falls very close to this ideal. Full ionization of the liquid corresponds to an electron density $N_{e,0} \cong 5 \times 10^{23}$ cm^{-3}. Calculations for H_2O have been performed for a quantum energy $\hbar\omega_x \cong 4.5$ keV, as illustrated in Fig. (1), that establish (1) the transverse scale size Δ of the channel, (2) the characteristic intensity I_x of the confined propagation, (3) the distance over which the incident energy reaches the eigenmode [2,4], (4) the efficiency of the confinement of the incident power in the channel, and (5) the stability of the channel [4,5,10]. The chief results are a channel width $\Delta \sim 190$ Å, a characteristic propagating intensity $I_x \sim 1.1 \times 10^{26}$ W/cm^2, a confined power of 655 TW, a value that corresponds to a trapping efficiency of 0.744, and the stable evolution of the incident pulse to the eigenmode [2,4,10] in a length of $\ell \sim 10$ Rayleigh ranges, a distance equivalent to $\ell \sim 2000$ μm. The computations were performed in both the collisional and non-collisional regimes. The influence of the collisions on the results shown in Fig. (1) was found to be entirely negligible, an outcome whose basis is the extreme thinness ($N_{e,0} / N_{cr} \cong 3.76 \times 10^{-5}$) of the plasma. Furthermore, on the basis of the measurements [6,7] of the properties of the Xe(L) system, the very favorable power scaling implied should enable power levels well above the critical power of $P_{cr} \cong 434$ TW necessary for the onset of channeling in water to be practically produced.

Since the channel has a very small diameter $\Delta \sim 190$ Å, a size considerably less than a typical virus, the losses due to photonionization, even at full ionization, are correspondingly small. With the assumption of $\sim 10^3$ eV/e$^-$ as an average value for deposition at full ionization, the energy loss rate is ~ 0.3 mJ/cm after formation of the channel on the eigenmode designated by point B in Fig 1(b). For these parameters, the energy loss associated with the evolution of the channel over length ℓ to the eigenmode has an upper bound of ~ 1.6 mJ. Hence, the propagation on the eigenmode of the pulse illustrated in Fig. 1(a) over a length of ~ 0.71 m would not reduce the propagating power below the critical level necessary for channel formation [1-4,10]. In contrast, conventional linear propagation of $\lambda_x \cong 2.9$ Å radiation in water would limit the length to ~ 50 μm, a distance more than a factor of $\sim 10^4$ less than that characteristic of the channeling mechanism.

4.5 keV Stable Self-Channeling in Water

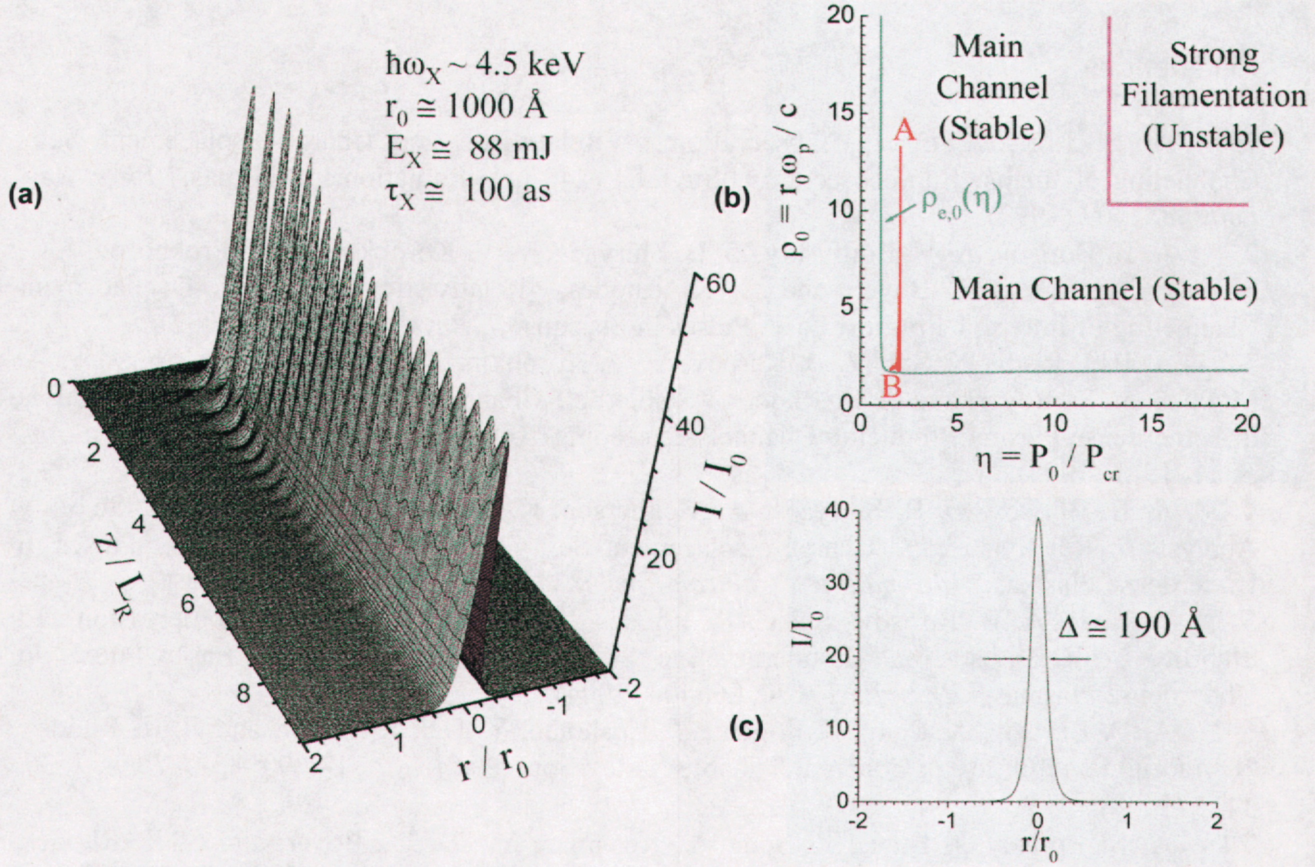

Fig. 1: (a) Evolution in water of the incident beam, designated by point A in panel (b), to the eigenmode $\rho_{e,o}(\eta)$, denoted by point B in panel (b). A pulse width $\tau_x \cong 100$ as is assumed for this calculation. The parameter r_o designates the radius of the incident pulse. The incident power P_o is ~ 880 TW and the medium is assumed to be fully ionized with an electron density $N_{e,0} = 5 \times 10^{23}$ cm^{-3}. The critical power for these conditions is $P_{cr} \cong 434$ TW. The pulse evolves to point B in a distance of approximately ten Rayleigh ranges ($Z/L_R \cong 10$). The power trapped in the channel P_x is ~ 0.744 of that incident, giving $P_x \sim 655$ TW. (b) Stability map corresponding to the relativistic/charge-displacement mechanism [4]. The incident pulse, point A, evolves in the zone of stability to point B on the eignemode $\rho_{e,o}(\eta)$. (c) The transverse profile of the channeled x-ray pulse at point B gives the characteristic width of the channel $\Delta \cong 190$ Å. This value yields a characteristic propagating intensity $I_x \sim 1.1 \times 10^{26}$ W/cm^2.

Although the results shown in Fig. (1) correspond to the pulse width $\tau_x \cong 100$ as, we note that the observation [7] of single-pulse spectral hole-burning on the Xe(L) system at $\lambda \sim 2.85$ Å with a width $\Delta\hbar\omega_x \sim 60$ eV, a value that corresponds to a temporal duration of $\tau_x \sim 30$ as, suggests that x-ray pulse durations on the order of an atomic time ($\hbar/\alpha^2 m_e c^2 \cong 24$ as) may be available in the future to produce high power x-ray channels in condensed matter.

In conclusion, relativistic/charge-displacement is the ideal mechanism for production in the multikilovolt x-ray region of stable high power confined modes of deeply penetrating radiation in condensed matter. The example of H_2O indicates that channels possessing lengths of ~ 1 m can be produced that conduct coherent x-ray fluences > 10^{10} J/cm^2 at an intensity of ~ 10^{26} W/cm^2 through a structure whose transverse width is ~ 190 Å.

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3) Ultrabright Multikilovolt Coherent Tunable X-Ray Source at ~ 2.71 – 2.93 Å for Biological Microimaging

The recent observation of strong amplification on multikilovolt Xe(L) hollow atom transitions in the ~2.8 Å spectral region can be seen as a consequence of the combination of (1) a new concept for amplification that involves the creation of a highly ordered state combining ionic, plasma, and coherent radiative components and (2) the use of two recently discovered (c. ~1990) forms of radially symmetric energetic matter, namely, hollow atoms and self-trapped plasma channels. This approach enables the demanding power densities necessary for x-ray amplification (~10^{19} W/cm^3) to be reached under conditions for which (α) the effective phase space volume of the interaction is profoundly limited and (β) the energy transfer is radiation dominated. A leading application will be the realization of a new mode of microimaging of living biological matter having a spatial resolution ~1000-fold superior to conventional light microscopy.

Xe(L) Emission Spectrum (no channel)
2p ← 3d

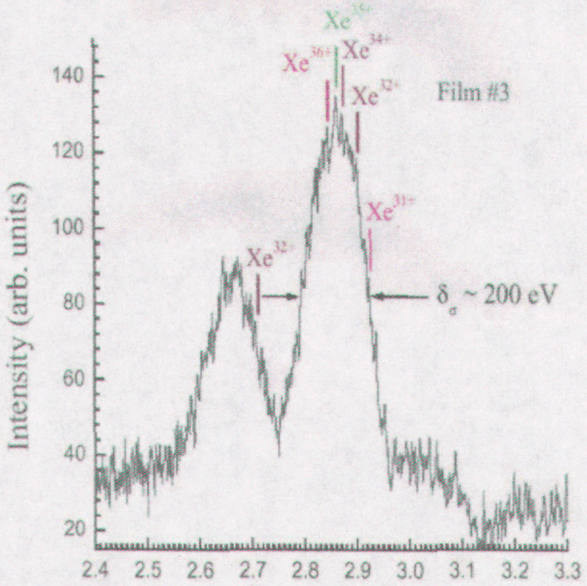

FIGURE 1. Spontaneous emission profile of Xe(L) hollow atom states.

Xe(L) Emission Spectrum in Channel Axis Direction 2p ← 3d

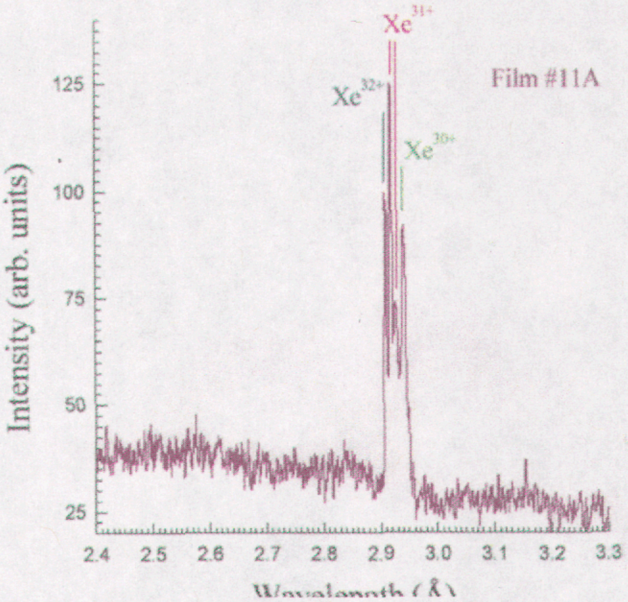

FIGURE 2. Amplified components of the Xe(L) emission on the Xe^{30+}, Xe^{31+}, and Xe^{32+} arrays observed from the plasma channel in the axial direction.

A

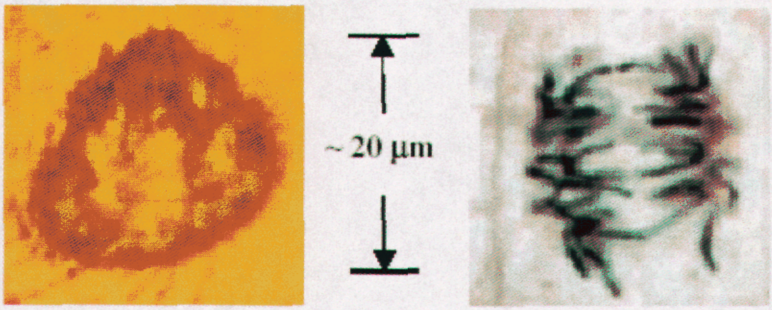

FIGURE 3. (A) Surface damage to the 12.7 μm thick Ti foil used to shield the film pack at an axial distance of ~12.5 cm from the source as viewed under a light microscope with front surface illumination. The damage does not fully penetrate the Ti foil. (B) Photograph of a cell in the early anaphase stage of cell division obtained with a light microscope that illustrates the favourable spatial match of the x-ray beam with the requirements of an illuminator for biological microimaging. This cellular image is taken with permission from Sobotta/Hammersen, Third edition, *Histology, Color Atlas of Microscopic Anatomy* (Urban and Schwarzenberg, Baltimore-Munich, 1985), figure no. 73f on page 45.

Detailed molecular structural information of the living state is of enormous significance to the medical and biological communities. Since hydrated biologically active structures are small delicate complex three-dimensional (3D) entities, it is essential to have molecular scale spatial resolution, high contrast, distortionless, direct 3D modalities of visualization of naturally functioning specimens in order to faithfully reveal their full molecular architectures. An x-ray holographic microscope equipped with an x-ray laser as the illuminator would be uniquely capable of providing these images [1,2,3]. A concordance of physical evidence [4,5], that includes (a) the observation of strong enhancement of selected spectral components of several Xe^{q+} hollow atom transition arrays (q = 31, 32, 34, 35, 36, 37) radiated axially from confined plasma channels, (b) the measurement of line narrowing that is spectrally correlated with the amplified transitions, (c) evidence for spectral hole-burning in the spontaneous emission, a manifestation of saturated amplification, that corresponds spectrally with the amplified lines, and (d) the detection of an intense narrow ($\delta\theta_x \sim 0.2$ mr) directed beam of radiation, (1) experimentally demonstrates in the $\lambda \cong 2.71$–2.93 Å range ($\hbar\omega_x \cong 4230$–4570 eV) the operation of a new concept capable of producing the ideal conditions for amplification of multikilovolt x-rays and (2) proves the feasibility of a compact x-ray illuminator that can cost-effectively achieve the mission of biological x-ray microholography. The development of this new mode of seeing of living systems will represent the third major advance in three centuries in biological microimaging. The suitability of the x-ray source for the illumination of small biological entities is illustrated in Fig. (3) above by the matching of the beam size (A) with a typical specimen of interest (B). The measurements also (α) establish the property of tunability in the quantum energy over a substantial fraction of the spectral region exhibiting amplification ($\Delta\hbar\omega_x \sim 345$ eV) and (β) demonstrate the coherence of the x-ray output through the observation of a canonical spatial mode pattern. An analysis of the physical scaling revealed by these results indicates that the capability of the x-ray source potentially includes single-molecule microimaging, the key for the *in situ* structural analysis of membrane proteins, a cardinal class of drug targets. An estimate of the peak brightness achieved in these initial experiments gives a value of ~ 10^{31}–10^{32} photons $\cdot s^{-1} \cdot mm^{-2} \cdot mr^{-2}$ (0.1% Bandwidth)$^{-1}$, a magnitude that is ~ 10^{7}–10^{8}-fold higher than presently available synchrotron technology, as shown in Fig. (4).

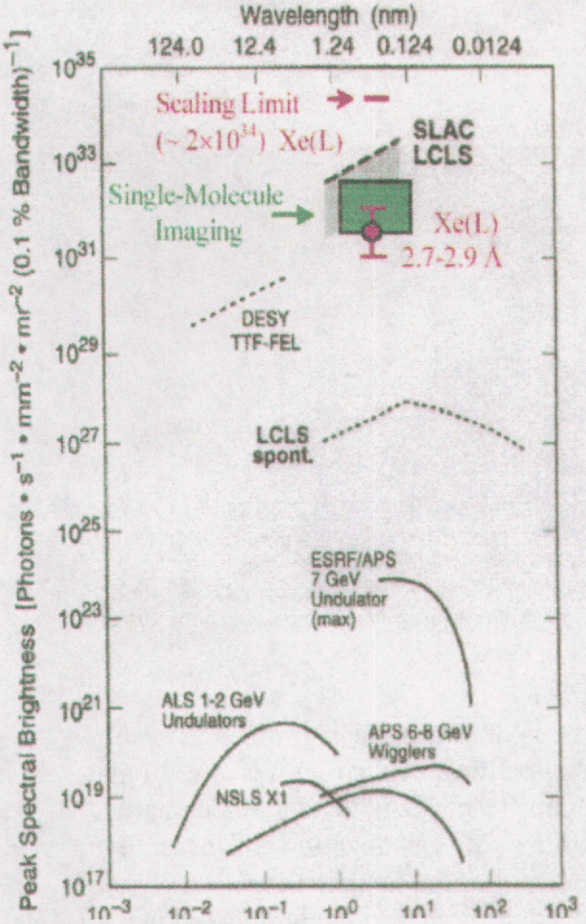

FIGURE 4. Peak spectral brightness comparisons of the present Xe(L) source at 2.7–2.9 Å with existing (solid contours) and projected (dashed contours) facilities. The present performance is given, the estimated requirement for single-molecule imaging is represented by the green zone, and the scaling limit for a compact laboratory instrument based on the findings of this study is indicated. Figure used with permission and adapted from Tatchyn, R. et al, "X-ray optics design studies for the SLAC 1.5–15 Å Linac coherent light source, *Nucl. Instrum. Methods Phys. Res.* A **429,** 397 (1999).

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