Observation of Nonlinear Optical Coupling in the Kiloelectronvolt X-Ray Regime

Shahab Firasat Khan, Ervin Rácz, Sankar Poopalasingam, John Charters McCorkindale, Alex Boris Borisov, James William Longworth, and Charles Kirkham Rhodes, *Fellow, IEEE*

(Invited Paper)

***Abstract*— Experimental findings with Xe(M) radiation in the ~1 keV X-ray region have confirmed the presence of a predicted zone of anomalously strengthened radiative coupling operative at sufficiently high intensity (I > 10^{15} W/cm^2) and frequency ($\hbar\omega$ > 5 eV). These new results herald the general existence of a strongly enhanced modality of radiative interaction that is based on ordered-driven electron motions in the attosecond regime.**

***Index Terms*— Nonlinear optics, plasma channels, X-ray emission.**

I. Introduction

STUDIES of the simultaneous production of Xe(M) and Xe(L) X-rays produced in plasma channels in a xenon cluster target have been conducted with an enhanced diagnostic suite. Overall, the aggregate of the recorded data contains approximately ten thousand files of *single pulse multi-component* information. Typically, each recorded file includes (1) the 248 nm pulse energy (mJ), (2) the xenon nozzle backing pressure (psi), (3) the nozzle temperature (K), (4) Xe(M)/Xe(L) joint pinhole camera images, (5) a Thomson image of the electron density from scattered 248 nm light, (6) a Xe(M) or Xe(L) transversely observed spectrum, and (7) an axially observed Xe(M) or Xe(L) spectrum. This array of diagnostics is illustrated in Fig. (1). Importantly, the pinhole camera images and the 248 nm energy measurement are calibrated. Furthermore, in comparison to all earlier work, the Xe(M) and Xe(L) data are now obtained with an instrumental sensitivity and dynamic range approximately 100-fold greater than earlier results. Accordingly, with this sea change in capability, (A) new unexpected phenomena are being observed and (B) the pace of experimental discovery has been conservatively lifted by at least one order of magnitude. Although much has been learned, there is a single chief discovery of high significance that is the subject of this study.

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S. F. Khan was with the Department of Physics, Laboratory for X-Ray Microimaging and Bioinformatics, University of Illinois, Chicago, IL 60607 USA. He is now with the National Ignition Facility, Lawrence Livermore National Laboratory, Livermore, CA 94550 USA (e-mail: khan9@llnl.gov).

E. Rácz is with the Department of Physics, Laboratory for X-Ray Microimaging and Bioinformatics, University of Illinois, Chicago, IL 60607 USA, and also with the KFKI Research Institute for Particle and Nuclear Physics, EURATOM Association, Budapest H-1525, Hungary (e-mail: eracz@uic.edu).

S. Poopalasingam, J. C. McCorkindale, A. B. Borisov, and J. W. Longworth are with the Department of Physics, Laboratory for X-Ray Microimaging and Bioinformatics, University of Illinois, Chicago, IL 60607 USA (e-mail: psanka1@uic.edu; johnmc@uic.edu; alexbor@uic.edu; zepto@uic.edu).

C. K. Rhodes is with the Department of Physics, Laboratory for X-Ray Microimaging and Bioinformatics, University of Illinois, Chicago, IL 60607 USA, and also with the Departments of Bioengineering, Computer Science, and Electrical and Computer Engineering, University of Illinois, Chicago, IL 60607 USA (e-mail: rhodes@uic.edu).

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II. Key Finding and Significance

A. Intensity-Frequency Interaction Landscape: Experimentally Determined Zone of Anomalous Coupling

Based on a long sequence of experimental studies [1–19], it is possible to forecast a zone of anomalous strong electromagnetic coupling that stands in sharp contrast to a corresponding region of conventional weaker interaction. These two regions are presented in the intensity (I) –quantum energy ($\hbar\omega$) plane illustrated in Fig. (2). As described below, new data obtained through studies of plasma channel production in a xenon cluster medium confirm this prediction through measurements made in the keV region. The lower boundary of the intensity for the anomalous coupling shown in Fig. (2) is given approximately by I $\sim 10^{15} - 10^{16}$ W/cm^2, a range corresponding to an electric field slightly less than one atomic unit ($e/a_o^2 \sim 5.14 \times 10^9$ V/cm). The corresponding lower limit in the frequency, estimated on the basis of measurements with N_2 and Xe, is h$\omega \sim 5$ eV, a value slightly less than one half of a Rydberg. Therefore, as an overall measure, the region of anomalous interaction is bounded by the characteristic values of electric field strength (intensity) and frequency (energy) associated with the hydrogen atom, the paramount fundamental atomic entity.

Key supplementary data are also placed on Fig. (2). They include (a) the line

$$\frac{eE}{m\omega c} = 1 \quad (1)$$

that defines the peak electric field E for the onset of relativistic motions for elections with mass m and charge e driven at angular frequency ω, (b) the vacuum pair creation limit, the upper value bounding the achievable intensity that is associated with ultrarelativistic electron/positron cascades [20], (c) the intensity I $\cong 4.6 \times 10^{29}$ W/cm^2 corresponding to the Schwinger/Heisenberg limit [21, 22], and (d) the intensity point characterizing saturated amplification of the Xe(L) hollow atom system [12–14].

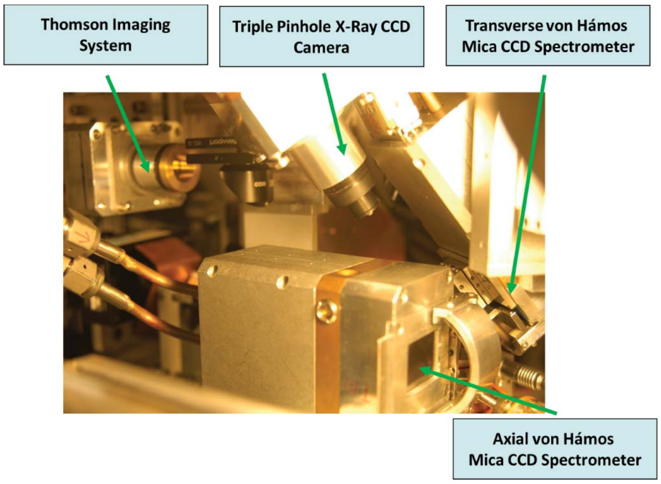

Fig. 1. Illustration of diagnostic array. The Thomson system images scattered laser light from electrons in the plasma. The triple pinhole X-ray camera allows the simultaneous imaging of Xe(M) and Xe(L) X-rays. The transverse and axial von Hámos spectrometers measure the spectra of the emitted X-rays in the side and forward directions, respectively.

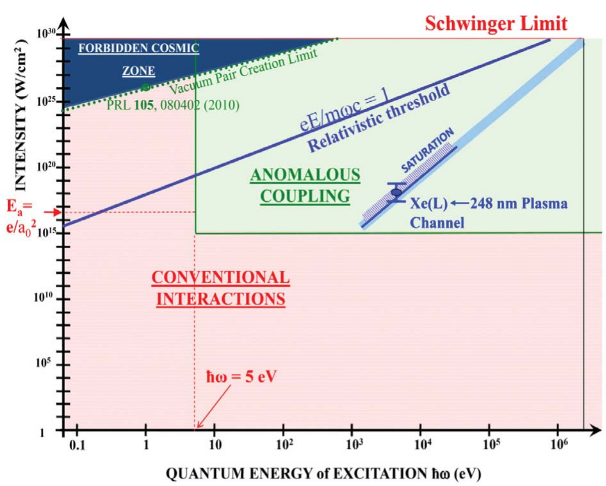

Fig. 2. Intensity–quantum energy interaction plane. Experimental data define a region of anomalous coupling that is designated by the green shading.

The quadrant of Fig. (2) that corresponds to the zone of anomalous coupling that is estimated to be free of the cosmic e^+/e^- cascade intensity limit [20] is highlighted in Fig. (3). The lower boundaries are given roughly by

$$\hbar\omega > 1\ keV \tag{2}$$

and

$$I > 10^{15}\ W/cm^2. \tag{3}$$

This figure also designates a projected zone for intensities, $I > 10^{27}$ W/cm^2 as a region in which nuclear reactions are anticipated to be observed [23, 24].

B. Confirmation of Anomalous Zone With Evidence for Nonlinear Coupling in the X-Ray Range

Simultaneous single-pulse X-ray pinhole images recording Xe(M) and Xe(L) emission, respectively at $\sim$ 1 keV and

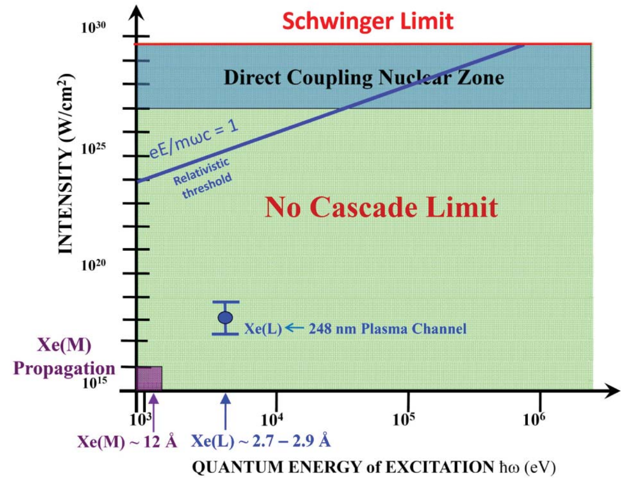

Fig. 3. Zone of anomalous interaction presented in Fig. 2, which is free of the cascade limit for intensities up to the Schwinger/Heisenberg value. An estimated zone associated with direct coupling to nuclei is also indicated [23], [24]. The datum associated with the Xe(M) intensity illustrated in Fig. 4 is given by the purple rectangle with coordinates of $\hbar\omega \cong 1$ keV and $I = 10^{15}$ to 10^{16} W/cm^2.

$\sim$ 4.5 keV from 248 nm channels produced in a xenon cluster medium, give evidence for an anomalously strong nonlinear coupling of the Xe(M) radiation to *unexcited* clusters whose signature matches the predicted zone presented in Fig. (3). The experiments demonstrate the following phenomena. Specifically, the outcome of strong Xe(M) production is the clear observation of both (1) subsequent spatially overlapping Xe(L) emission in the $\lambda \sim 2.8$ Å range and (2) heavily modified anomalous propagation of the Xe(M) radiation. The propagation, as shown in Fig. (4), manifestly illustrates the phenomena of self-focusing of the Xe(M) radiation. The Xe(L) signal presented in Fig. (5) reveals a spatially broad zone of Xe(L) emission well matching the area of Xe(M) emission, a fact that shows that this more energetic radiation is excited directly by the strong Xe(M) signal. Specifically, the overall nonlinear absorptive process observed is

$$n\gamma(M) \rightarrow Xe^*(L) \rightarrow \gamma(L) \tag{4}$$

in which the minimum value of n is 5 based on the energies of the states involved. We note that, on the basis of the known measured threshold intensity of $\sim 2 \times 10^{17}$ W/cm^2 for the production of Xe(L) emission by 248 nm irradiation of xenon clusters [25], the 248 nm power used in this experiment of $\sim$ 2 TW could not produce this required intensity over the large spatial scale of $\sim$ 400 μm in breadth observed for the Xe(L) signal. Hence, the source of the Xe(L) excitation is the Xe(M) radiation through the amplitude specified by Eq. (4), a reaction confirmed by the spatial overlap of the Xe(M) and X(L) signals illustrated in Figs. (4) and (5).

We note that related studies of nonlinear phenomena at short wavelengths, such as the investigation of multiphoton absorption in the soft X-ray regime [26–30] and the examination of hidden resonances appearing with ultrashort high-intensity X-ray laser irradiation [31], have been recently performed at the LCLS (Linac Coherent Light Source) [32],

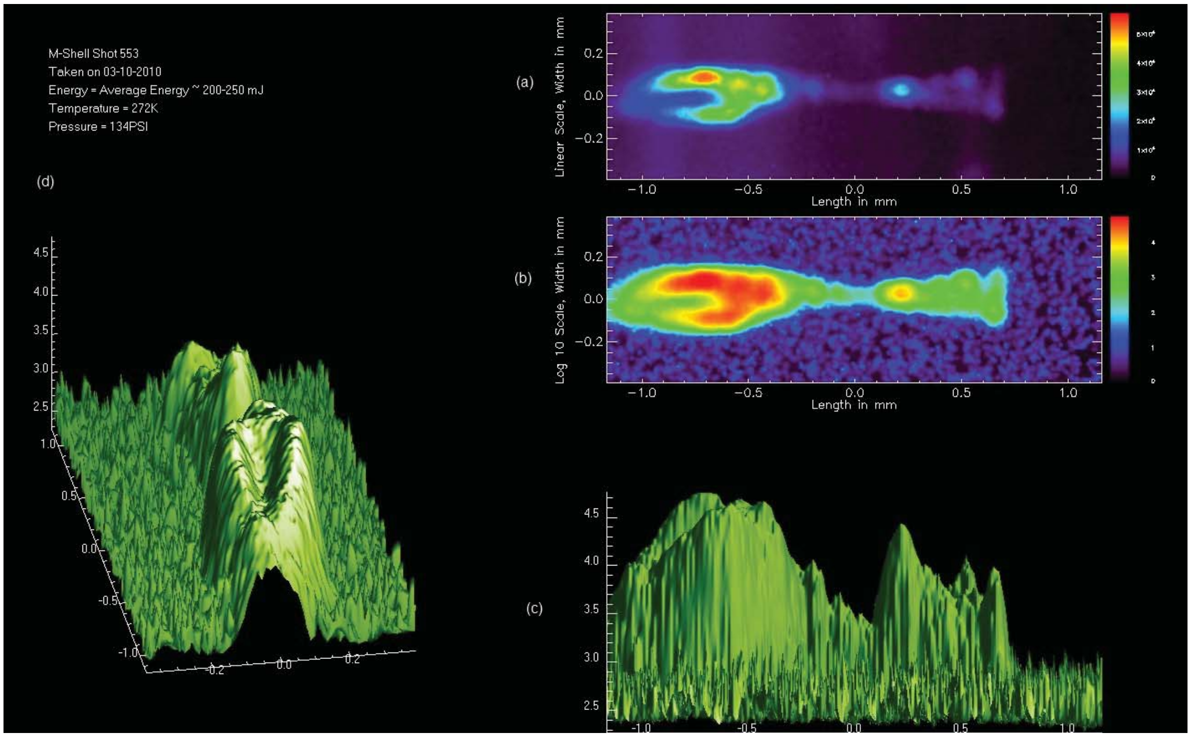

Fig. 4. Transverse pinhole camera image of Xe(M) radiation from 248 nm channel region, the direction of 248 nm propagation is right to left. (a) Linear scale of observed emission. (b) Log scale of observed emission. (c) Panel (b) presented as a pixel array. (d) Isometric view of panel (c) clearly illustrating the focusing collapse of the central zone as the Xe(M) beam propagates, downward in the image corresponds to increased distance of propagation, from right to left corresponding to panels (a)–(c). Data are from 10 March 2010, shot #553, Xe(M).

the world's first hard X-ray free-electron laser. Similar work had been conducted earlier at the FLASH (Free-Electron Laser in Hamburg) facility on xenon involving the giant 4d resonance [18].

The two observed nonlinear couplings, one dispersive and the other absorptive, as illustrated above in Figs. (4) and (5), can be explained by the nonlinear Kramers-Krönig relation [33]; the governing statement is

$$\Delta n(\omega;\zeta) = \frac{c}{\pi} P \int_0^\infty \frac{\Delta\alpha(\omega';\zeta)}{\omega'^2 - \omega^2} d\omega'. \tag{5}$$

The direct comparison of the Xe(M) and Xe(L) emissions illustrated in Figs. (4) and (5) simply expresses both sides of Eq. (5). Since Eq. (5) is a fundamental statement based only on casuality, the presence of one effect necessarily implies the presence of the other. Hence, the dual observation shown in Figs. (4) and (5) is imperative. Estimates of the maximum intensity of the Xe(M) X-ray emission shown in Fig. (4) give values in the $\sim 10^{14} - 10^{16}$ W/cm^2 region. This range of intensity falls in the anomalous interaction zone described above and is presented in Fig. (3) as the purple region.

The overall lesson is clear; if the frequency is sufficiently high, an intensity of $\sim 10^{15}$ W/cm^2, or equivalently, if the radiative electric field E of the wave falls in the range

$$E > \frac{e}{5a_o^2}, \tag{6}$$

the chief outcome is the induction of very strong nonlinear behavior that includes the production of inner-shell vacancies.

C. Significance

The significance of the nonlinear coupling associated with Eq. (5) is highlighted by the location of the purple Xe(M) rectangle shown in Fig. (3); it falls essentially at the lower limit of the zone given by $\sim 10^{15}$ W/cm^2 in Fig. (2). Furthermore, on the basis of the measured threshold of $\sim 2 \times 10^{17}$ W/cm^2 for the corresponding production of Xe(L) emission with 248 nm radiation $\hbar\omega \cong 5$ eV, we see that the threshold value has decreased by a factor of $\sim 10^2$ at the higher quantum energy of ~ 1 keV. It follows that the circumstances for the production of deep inner-shell vacancies could hardly be more favorable; the minimally required intensity provides strong excitation of the main states of interest. Furthermore, since the greatly reduced threshold now enables the consideration of excitation of correspondingly increased volumes of material, X-ray pulse energies in the 1-10 J range can now be practically contemplated. Although a complete theoretical picture of this phenomenon is not in hand, the experiments combine two conditions in an incomparably new situation; specifically, it is the alliance of (1) a strong radiation field on the order of e/a_o^2 and (2) a high frequency ω whose period is less than 5 as. Since a time of ~ 5 as is far less than an electron dephasing time in all materials, this situation is well suited for the production of unconventional highly organized electronic motions and we can anticipate the production of

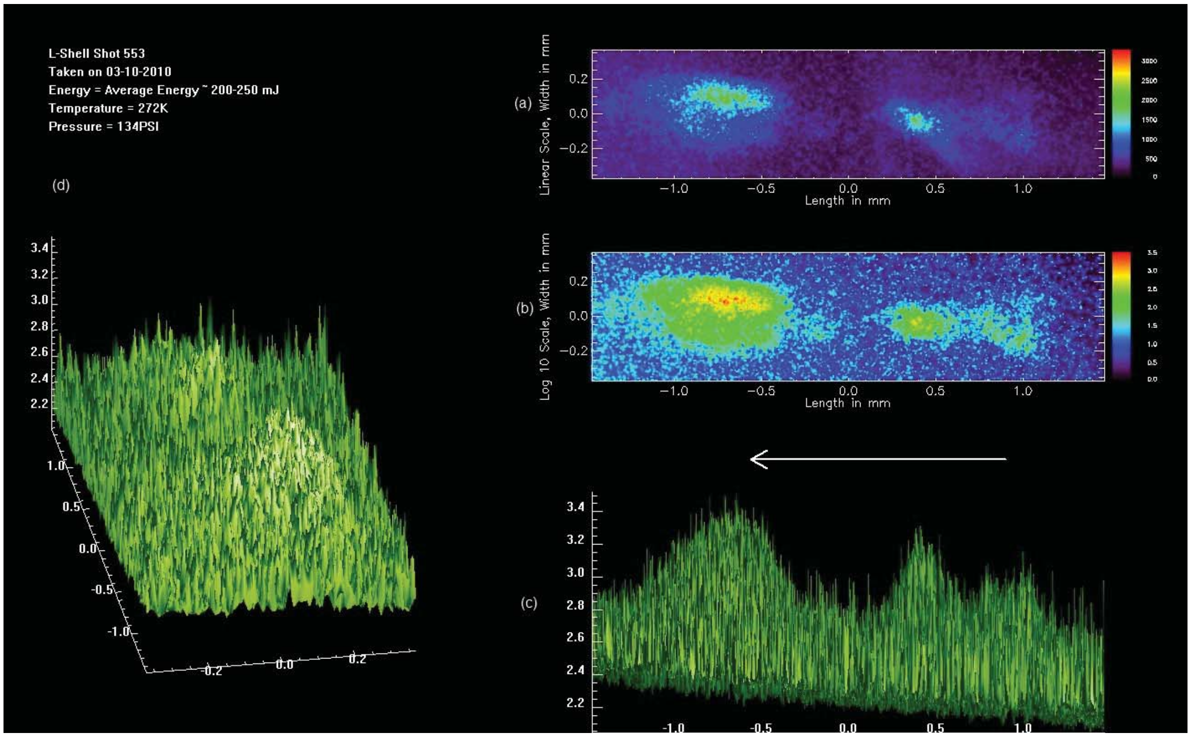

Fig. 5. Transverse pinhole camera images of the Xe(L) radiation from the 248 nm channel, which correspond spatially to the images shown in Fig. 4. The key finding is the large broad zone (~400 μm) of Xe(L) emission presented in panel (b) that matches the pattern of Xe(M) emission illustrated in Fig. 4(b). Data are from 10 March 2010, shot #553, Xe(L).

new exceptional forms of energetically excited matter that are analogous to the examples [6, 8] provided by N_2 and Xe.

III. Conclusion

Experimental findings with Xe(M) radiation in the ~1 keV X-ray region have confirmed the presence of a predicted zone of anomalously strengthened radiative coupling operative at sufficiently high intensity ($I > 10^{15}$ W/cm^2) and frequency ($\hbar\omega > 5$ eV). These new results herald the general existence of a strongly enhanced modality of radiative interaction that is based on ordered driven electron motions in the attosecond regime.

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Shahab Firasat Khan was born in Chicago, IL, in 1977. He received the B.S. and Ph.D. degrees from the University of Illinois, Chicago (UIC), in 2003 and 2011, respectively.

He is currently a Research Scientist with the National Ignition Facility, Lawrence Livermore National Laboratories, Livermore, CA, working to achieve laser-driven inertial confinement fusion. He was a Post-Doctoral Fellow with the Laboratory for X-Ray Microimaging and Bioinformatics, UIC, where he worked toward the development of a super intense guided X-ray source produced by laser irradiated xenon clusters. His current research interests include the development and characterization of gated X-ray framing cameras that are used to capture time-dependent images of inertial-confinement-fusion implosions.

Ervin Rácz was born in Budapest, Hungary, in 1973. He received the M.Sc. degree in physics and mathematics and the Ph.D. degree in physics from the University of Szeged, Szeged, Hungary, in 1999 and 2006, respectively.

He was with the University of Szeged in 1999, after he graduated as a Teacher of physics and mathematics. From 1999 until 2002, he was with KFKI Research Institute for Particle and Nuclear Physics, Budapest, Hungary, as an Assistant Research Fellow and as a Research Associate from 2011. In 2006, he joined the Department of Physics, Charles Rhodes' Laboratory, University of Illinois, Chicago (UIC). He started his work as a Research Specialist of high intensity laser systems. Later in 2008, he was a Visiting Research Assistant Professor and has been a Research Assistant Professor at UIC since 2010. From 1999 until 2006, he was working on high harmonics generation on solid surfaces using high intensity ultraviolet dye-KrF excimer laser systems. He experimentally investigated ultrashort and ultraintense X-ray pulses by von Hamos crystal spectrometers and other useful X-ray detectors. His current research interests include experimental laser-plasma physics, high-intensity excimer-based lasers, high intensity Ti:Sa-KrF excimer hybrid laser systems, and hard X-ray generation in gas jets by high intensity laser systems.

Dr. Rácz received the Young Ph.D. Scholarship of Physics from the Hungarian Academy of Sciences at the KFKI Research Institute for Particle and Nuclear Physics, Department of Plasma Physics.

Sankar Poopalasingam was born in Jaffna, Sri Lanka. He received the B.Sc. degree in physics from the University of Jaffna, Jaffna, the M.Sc. degree in material science from the Postgraduate Institute of Science, University of Peradeniya, Peradeniya, Sri Lanka, and the M.S. degree in physics from the University of Illinois, Chicago. He is currently pursuing the Ph.D. degree in physics with the University of Illinois, Chicago.

He is currently a Research Assistant with the Laboratory for X-Ray Microimaging and Bioinformatics, University of Illinois, where he is involved in the field of operation and maintenance of ultrafast hybrid ultraviolet excimer laser systems, beam alignment for experiments, designing and aligning diagnostics for tabletop X-ray laser development, and the study of laser matter interactions. His current research interests include the development and characterization of tabletop X-ray lasers and the study of laser matter interactions.

Mr. Poopalasingam is a recipient of the Gold Medal from the University of Jaffna for his best performances in the B.Sc. degree in physics.

John Charters McCorkindale was born in San Francisco, CA, in 1981. He received the B.Sc. degree in physics with a minor in astronomy and planetary science from the University of California, Santa Barbara, and the M.S. degree from the University of Illinois, Chicago, in 2004 and 2008, respectively. He is currently pursuing the Ph.D. degree with the Laboratory for X-Ray Microimaging and Bioinformatics, University of Illinois.

He is currently a Research Assistant with the Laboratory for X-Ray Microimaging and Bioinformatics.

Mr. McCorkindale was a two-time recipient of the Directed Energy Professional Society Graduate Scholarship in 2005 and 2006 and a recipient of Graduate Assistance in Areas of National Need Fellowship in 2007.

Alex Boris Borisov received his degrees from Moscow State University, Moscow, Russia.

He is a Research Associate Professor with the Department of Physics, University of Illinois, Chicago. He has made numerous significant contributions to the fields of ultraintense laser-plasma interaction, X-ray laser development, X-ray spectroscopy, and computational nonlinear optics. He is the author or co-author of more than 100 publications. His current research interests include laser physics, physics of plasmas, X-ray physics, and mathematical and computational physics.

Prof. Borisov is a member of the American Physical Society and the Optical Society of America.

James William Longworth was born in Stockton Heath, Cheshire, U.K., on September 16, 1938. He received the B.Sc. and Ph.D. degrees from Sheffield University, Sheffield, U.K., in 1959 and 1962, respectively, all in biochemistry. His Ph.D. thesis was based on the phosphorescence of proteins and nucleic acid while working with G. Weber.

He held a Post-Doctoral Fellowship with R. Livingston at the Department of Chemistry, University of Minnesota, Minneapolis, from 1962 to 1963, and became a Technical Staff Member with the Department of Chemical Physics, Bell Laboratories, Murray Hill, NJ, from 1963 to 1965, working with F. Bovey. At Bell Labs he, in collaboration with R. Shulman, R. Rahn, and J. Eisinger, identified thymine as the lowest triplet state of DNA. He joined the Biology Division, Oak Ridge National Laboratories (ORNL), Oak Ridge, TN, from 1965 to 1983, where he became a Senior Biophysicist. While at ORNL, he showed there was singlet electronic energy transfer in poly-L-tyrosine, tryptophyl fluorescence quenching by disulphide linkages in models and proteins, triplet electronic energy transfer in polyadenylic acid, and exciplex tryptophyl fluorescence in proteins. From 1984 until 2009, he was an Associate Professor of physics with the Illinois Institute of Technology (IIT), Chicago. While at IIT, he studied excited singlet electron transfer from tryptophan in the copper protein azurin and the dynamics of tryptophan in endonuclease with G. Fleming at the Department of Chemistry, University of Chicago, Chicago, and developed an optical holographic microscope with C. Rhodes at the Department of Physics, University of Illinois, Chicago. From 1988 to present, he is a Research Professor of physics with the University of Illinois, and is working with C. Rhodes on the construction and characterization of a multiterawatt KrF 248 nm laser system and the production of hard X-ray lasing in xenon gas puffs. He is involved in developing the application of a single shot nondegenerate deep UV four-wave mixing to identify the origin of fluorescence of DNA, which is still unknown. The design is to use a nondegenerate four-wave mixing that forms a tunable deep UV 10 fs pulse as a source of optical radiation excitation. He collaborates with A. Schroeder at the Department of Physics, University of Illinois, where both apply four-wave mixing to define the role of quantum coherence in excited singlet electronic transfer in DNA in the inherently corrugated electronic energy level system that constitutes natural DNA and may play a crucial role in the protection of DNA from cancer-forming DNA photoproduct formation. His current research interests include characterizing multiterawatt 248 nm laser pulses by using a degenerate single shot four-wave mixing frequency resolved optical gating.

Dr. Longworth has been a member of the American Society of Biological Chemistry and the Biophysical Society, and part of the Foundation Group of the American Society for Photobiology. He has been the eighth President of the American Society for Photobiology and a Chairman of the Committee on Photobiology of the National Research Council.

Charles Kirkham Rhodes (SM'79–F'89) was born in Mineola, NY, on June 30, 1939. He received the B.S.E.E. degree from Cornell University, Ithaca, NY, in 1963, and the M.S.E.E. and Ph.D. degrees in physics from the Massachusetts Institute of Technology, Cambridge, in 1965 and 1969, respectively.

He is the A. A. Michelson Professor of physics, Emeritus, a Distinguished Appointment in the University of Illinois system, Chicago, the Director of the Laboratory for X-Ray Microimaging and Bioinformatics, Department of Physics. He currently holds additional appointments with the Departments of Bioengineering, Computer Science, and Electrical and Computer Engineering, College of Engineering. He has a professional record of over 300 publications.

Dr. Rhodes is a fellow of the American Physical Society, the Optical Society of America, and the American Association for the Advancement of Science. He is the recipient of an Alexander von Humboldt Award for Senior U.S. Scientists, a Foreign Member of the Russian Academy of Engineering, and a recipient of the Inventor of the Year Award at the University of Illinois, Chicago, for the development of a patented concept for multikilovolt X-ray amplification in 2000.