53. The Dawn of Ultrafast Science and Technology at the University of Rochester

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The University of Rochester in the 1980s played an important role in the development of the field of ultrafast science and technology as one of the most active field of optics today. The Institute of Optics and the Laboratory for Laser Energetics (LLE) occupied the central stage. The Institute of Optics provided exceptional students and LLE a unique technical platform. Many of the techniques that the researchers in the field use today, like THz generation, picosecond electron diffraction (PED), electro-optic sampling (EOS), chirped pulse amplification (CPA), and jitter-free synchronization were conceived and demonstrated by the ultrafast science group. The Institute of Optics students—Wayne Knox, Theodore Sizer, Irl Duling, Janis Valdmanis, James Kafka, Donna Strickland, Maurice Pessot, Jeffrey Squier and John Nees—formed the core. Their enthusiasm was infectious and contributed much to attract students from physics, such as Steve Williamson, Theodore Norris, and Kevin Meyer, and from electrical engineering, including Daniel Blumenthal, John Whitaker, and Doug Dykaar, as well as faculty like C. Gabel, R. Knox, C. Stancampiano, T. Hsiang, R. Sobolewski, A. Melissinos, J. Eberly, and D. Meyerhofer.

A group picnic in France.

In the early 1980s ultrafast science was dealing with eV phenomena. Our group extended its range from the meV on one side, with the introduction of THz beams and electro-optic sampling (EOS) techniques, to the MeV-GeV on the other side, with chirped pulse amplification (CPA) and its ability to produce relativistic intensities.

I. Switching High Voltage with Picosecond Precision: Applications to Active Pulse Shaping, Jitter-free Streak Camera, Time-resolved Spectroscopy

Work in this area mainly started in 1978 after my arrival at LLE. LLE was running a highly programmatic effort on inertial confinement fusion. At that time the director and founder, M. Lubin, and later R. McCrory understood the importance of creating and supporting in parallel to the main research activity a group that would work on weakly related laser fusion projects, that could offer the flexibility and the type of environment that Ph.D. research demands. I would be in charge of this group, known as the ultrafast science group. I was impressed by the work of Dave Auston at AT&T Bell Laboratories that demonstrated that electrical signals could be switched with picosecond precision. I was convinced that this technique could find some important applications in laser fusion, where the program had a need for synchronized high voltage pulses for active pulse shaping or for jitter-free streak cameras. An exceptional undergraduate student, Wayne Knox, shared the understanding of the importance of this line of research. At Wayne's high speed of progress we extended Auston's work to very high voltage [1] and applied it to the synchronization of streak cameras [2]. For the first time the streak camera could be used in accumulation mode. Weak luminescence signals could be accumulated, improving their signal/noise ratio. The jitter-free streak camera found immediate applications in photobiology with the group of Wayne's father, Professor R. Knox. This technique is now routinely used in synchrotron-based femtosecond x-ray diffraction experiments [3]. High voltage switching applications were also in active pulse shaping [4], as demonstrated in collaboration with J. Agostinelli (student of C. Gabel), and in contrast improvement [5] with W. Seka. This technique is still used today [6] in high field science.

II. Opening the Field of Picosecond Electron Diffraction

A streak camera is a beautiful photon-electron transducer. It makes an electron replica of the photon pulses. The electrons are deflected across the phosphor screen, leaving a phosphorescent track. I was mesmerized by the thought that we could use this perfectly synchronized photoelectron pulse to perform time-resolved electron diffraction in the picosecond time scale by simply locating a sample under study in the camera drift region. We could study solid-liquid transformation simply by using a short optical pulse to produce the phase transition and the electron pulse to probe the structural change that would follow.

I asked a new student with great passion for research, Steve Williamson, if he would be interested in this project. This was an enormous challenge, as none of us had any kind of electron diffraction experience in steady state let alone in the transient regime. But Steve was a superb experimentalist, and in one year he built a complete "streak camera" and demonstrated the concept [7]. We applied it by performing the first time-resolved structural transformation in the picosecond domain. It was the solid-liquid phase transformation of aluminum [8]. Further works were conducted by H. Elsayed-Ali, notably on surface melting [9]. The activity was extended later to gas electron diffraction by A. Zewail [10]. More recently—twenty years later—our picosecond electron diffraction experiment on Al was repeated by Dwayne Miller from the University of Toronto with a superior laser and shorter pulses [11]. Note that Dwayne was at the University of Rochester in the chemistry department with a joint appointment in Optics when, in 1982, Steve did his seminal experiment. Today, time-resolved electron diffraction is becoming a very active field, rivaling with time resolved x-ray diffraction.

III. The First Steps of THz Generation

We knew that the picosecond rise time produced by photoconductive switching could be used to produce THz transients either from the gap itself, by putting a switch in a coaxial waveguide transition, or by exciting a microwave antenna [12]. This simple experiment was performed by a dedicated undergraduate student, Daniel Blumenthal from the electrical engineering department, in collaboration with his adviser, C. Stancampiano, and A. Antonetti from the Ecole Nationale Supérieure de Techniques Avancées in France [13]. The THz field became a very important domain, once it was realized by Auston [14] that the electric field could be time-resolved by the laser pulse itself. The field amplitude and phase could be measured and a new THz spectroscopy technique was born that would replace infrared Fourier-transform spectroscopy [15]. Besides spectroscopy, applications of these transients include THz imaging. Also, the methods of generation have been vastly advanced as demonstrated by X-C Zhang [16].

IV. Electro-optic Sampling (EOS): Opening the Field of THz Electronics

We could switch electrical signals with risetimes in the subpicosecond domain, but it was difficult to measure them. Wide band sampling oscilloscopes could only go to 25 ps., and the only way to measure the picosecond pulses was to use a second photoconductive gap with a fast photoconductive semiconductor. Of course, one solution was to try to use the electro-optic effect. The EO effect can have a purely electronic reaction with a subfemtosecond response. But there is no free lunch, and this ultrafast response is paid for in terms of sensitivity. Kilovolts are usually necessary to detect a signal. So, it appeared the EO effect could not be a contender for fast measurements, as it was not sensitive enough. Janis Valdmanis, who had the idea to use lock-in detection in conjunction with the electro-optic

effect, demonstrated this to be false. With his "golden hands," Janis showed that submillivolt, subpicosecond signals could be measured [17]. The EOS technique became an indispensable tool to visualize THz electrical signals. For the first time, direct propagation of picosecond electrical pulses on transmission lines, both normal and superconducting (with low and high-Tc) [18, 19] could be investigated. EOS was also used in the measurement of the fastest transistor risetimes [20] and the switching of Josephson junctions [21]. It was also used in the direct investigation of subpicosecond carrier dynamics in semiconductors, such as velocity overshoot [22]. Most of the activity was coordinated by D. Dykaar and involved many students, like J. Whitaker, visiting scientist R. Sobolewski and professor T. Hsiang, from electrical engineering, as well as K. Meyer, a student from physics.

V. CPA for Chirped Pulse Amplification, not for Certified Public Accounting

The generation and amplification of short pulses was, however, our main activity. Short pulses were used for everything. At that time, Ti:sapphire had not been invented, and dyes like rhodamine 6G were the main amplifier media. The leading laboratories were at AT&T Bell Laboratories with the group of C. V. Shank, and with E. Ippen and H. Haus at MIT. In our group, outstanding students were working on dye-based generation and amplification of ultrashort pulses. They were T. Sizer [23], I. Duling [24], J. Kafka [25] and T. Norris [26]. During one of our constant and endless discussions about novel ideas and concepts, we discussed in 1982 with Steve Williamson a possible way to get larger energy per pulse by using good energy storage media. We were at LLE and we knew that Nd:glass was a vastly superior energy storage medium—by a factor 10^3–10^4—than dyes. From a bandwidth point of view, Nd:glass can in principle amplify subpicosecond pulses. However, unlike in dye, Nd:glass is almost too good of an energy storage medium. The major problem is that the pulse energy becomes too large, leading to high intensities and nonlinear effects. The nonlinear effects contribute to destroy the beam quality and ultimately lead to the "destruction" of the optical amplifier. Dyes, on the other hand, do not have this problem. They are mediocre energy storage media, due to their large cross-section. Therefore, the pulse energy stays below the critical intensity level where the nonlinear effects dominate. We were greatly influenced by the work of D. Grischkowsky (IBM Yorktown Heights) and A. Johnson (AT&T Bell Laboratories) that demonstrated that by propagating a relatively long pulse in a fiber, the pulse will be the subject of broadening and stretching by a combination of self-phase modulation (SPM) and group velocity dispersion (GVD). As a result, the pulse is stretched with the spectral content of a much shorter pulse. It exhibits a linear chirp. At this point it can be compressed by using a Treacy grating pair, which exhibits a negative GVD to a value one hundred times the value of the input pulse. It looked to me that it would be simple to try to amplify the pulse in order to extract the amplifier energy and compress it later when the energy would be fully extracted. I asked a new student, Donna Strickland, if she would like to do this experiment. Donna was excited about it but also concerned that it might not be good enough for a Ph.D. thesis. She quickly demonstrated that this concept was working to the millijoule level [26].

The key to CPA: the matched stretcher-compressor. The first approach to CPA was rudimentary and relied on an unmatched stretcher-compressor system. It was not perfect. After a

certain amount of stretching, the compressor could not compress the pulse without causing significant wings on the pulse. The fiber-grating pair system was not matched over all orders. What we needed was a matched stretcher-compressor system so we could extract the energy better and compress it better. The matched stretcher compressor became our "Holy Grail." I was continuously thinking about it. One day I was skiing at Bristol Mountain with my wife Marcelle, and on the chairlift I started to think about a paper I read the day before from O. Martinez [27]. This paper was describing a compressor for communication applications at 1.5 μm. At this wavelength the GVD in fiber is negative and the pulses exhibit a negative chirp where the blue frequencies lead the red ones. To recompress the pulse at the fiber output, Martinez proposed a compressor with positive GVD that was a combination of a grating pair and a telescope of magnification unity. I realized that the Martinez compressor in the positive GVD region was in fact the matched stretcher of the Treacy compressor. This was exactly what we were looking for. I interrupted my day of skiing and went back to the laboratory, where I met Maurice Pessot, a new student in my group. I asked Maurice to drop what he was doing and show that the Martinez stretcher and the Treacy grating pair were matched. In a beautiful experiment, Maurice showed that an 80 fs. pulse could be arbitrarily stretched 1000 times by the Martinez device and recompressed by the same factor to its initial value [28]. A major hurdle in CPA was overcome. Fifteen years later, this stretching-compression system is still part of the standard CPA architecture.

The Petawatt. The stretcher-compressor was integrated in our first Joule level Nd:glass system by a visiting scientist, P. Maine, and a post doctoral fellow, P. Bado [29]. With Donna they demonstrated a pulse with one joule in 1 ps., i.e. 1 terawatt on a table top—called the "Maine event" since. It was at night and we were jubilant. R. McCrory, the LLE director, was as usual working late and heard our noisy celebration. He poked his head in the laboratory curious to know what was going on. I told him that we just had demonstrated the generation of one TW with a new amplification technique. It was a thousand times improvement in power over standard techniques, and moreover, this technique could be scaled to a much higher energy than the 1 kJ level using the glass development laser (GDL), a prototype chain at LLE. At that time we paused and asked ourselves what the next scientific prefix after "tera" was. Nobody knew. We went to Bob's office and discovered that it was "peta." So from now on, our next goal would be the petawatt. The first article on the possibility of producing petawatt level pulses was described in a French scientific journal, "En Route Vers Le Petawatt" [30] and the first petawatt pulse was demonstrated by M. Perry at Livermore ten years later.

The name T^3, for table top terawatt, was coined by Jonathan Heritage as he was visiting our laboratory. At that time we decided with Patrick and Donna to call this new amplification technique chirped pulse amplification (CPA). CPA appeared for the first time in reference [29]. Of course Wayne, who was at AT&T Bell Laboratories by that time and always has something to say, called me to argue that people would get the acronym mixed up with "certified public accounting."

We worked a lot to extend the technique to other materials, such as Alexandrite with Jeff Squier and Don Harter [31]. That was before the Ti:sapphire. Alexandrite was at that time the only broadband high-energy storage material available. A lot of the CPA work continued after our move to the University of Michigan with Ted Norris [32], Jeff Squier, Francois Salin [33] and Gary Valliancourt producing the first kHz Ti:sapphire source, which is the workhorse of many ultrafast optical laboratories today. Let's also not forget

the indispensable role of Marcel Bouvier, our dedicated electronics engineer who devised the first kHz Pockels cell, the key element of any CPA system.

VI. CPA and Its First Steps in Science

The first paper on the CPA and T^3 was presented at the 1986 ICOMP meeting in Boulder [34], where I met Mike Perry from Livermore and Gérard Mainfray from the Commissariat Atomique Energie at Saclay France. Because of the possibility of producing intensities well into the 10^{18} W/cm^2 regime, the relativistic regime, it was recognized that the CPA technique would revolutionize atomic, plasma and high-energy physics. In a sense the University of Rochester was the perfect cradle. The potential of the T^3 and CPA was understood by a number of people. Among the early ones were the tandem of Mike Campbell and Mike Perry from Lawrence Livermore National Laboratory (LLNL). Together we dreamt of the possibility of building a petawatt at Livermore. That dream was fulfilled in 1997 [35], ten years after the first TW, and a few years after the concept of fast ignition was conceived. In 1988 we wrote together the first proposal on petawatt physics) for a LLNL laboratory director's grant. The requested amount was \$590 k. Certainly considered too early or too revolutionary, this proposal was not funded. The first experiments on T^3 were done in plasma physics by the group of the Institut National de la Recherche Scientifique in Montreal (Quebec) led by H. Pepin [37] in collaboration with a new young professor, D. Meyerhofer. The first atomic physics experiment [38] was performed under the direction of S. L. Chin and J. Eberly (S. L. Chin was on sabbatical with J. Eberly). With A. Melissinos we were thinking about high-energy accelerators by combining fast switching and CPA as well as with K. McDonald from Princeton on nonlinear QED [39]. In 1988 the first CPA Nd:glass was reproduced outside the University by the group of G. Mainfray at the CEA Saclay [40] as well as at the Institute for Laser Engineering (ILE) in Japan by Y. Kato's group with C. Barty and K. Yamakawa [41]. I was relieved to see that our work could be duplicated elsewhere. They were the first of a long series of CPA systems and the birth of a highly active and competitive field involving atomic, plasma, nuclear and high energy physics, astrophysics and cosmology.

Thanks to The Institute of Optics' exceptional students and the Laboratory for Laser Energetics' facilities, the University of Rochester has been the cradle of a number of ultrafast disciplines, notably the thriving high field science area that includes over a thousand researchers and published papers, as well as dozens of conferences per year.

Professor Mourou is currently director of the Center for Ultrafast Optical Science, University of Michigan.

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