Frequency-Resolved Optical Gating:
The Measurement of Ultrashort Laser Pulses

If you haven't measured it, you haven't made it.

Wayne Knox

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The Measurement of Ultrashort Laser Pulses

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To Linda Leigh, whose love and enthusiasm made this work—and everything else—possible.

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0. Read Me

I don't know about you, but whenever I try to read a scientific book for more than a few minutes, I fall asleep. Scientific writing is dull. Indeed, scientists have a well-deserved reputation for being dull people who write dull books for other dull scientists.

I hate this.

So in writing and editing this book, I've tried something different. I've tried, not only to teach you the science of ultrashort-laser-pulse measurement, but also to convey to you the excitement and fun I've experienced in learning and discovering these ideas. For example, when I heard about the idea of phase retrieval, I dropped everything I'd been doing and spent an entire week in the library reading everything I could find on this amazing topic. So I've tried to convey that enthusiasm to you whenever possible. I've also tried to use some writing techniques that make reading a novel fun. For example, you may notice some recurring themes and direct addresses. (Lucky for you I don't know anything about symbolism.) And I've just written whatever I've felt like writing in places and not bothered to edit it out. Fortunately, the wonderful folks at Kluwer Academic Publishers have encouraged this. I've had a lot of fun writing my share of this book, and I hope this means that you'll have some fun reading it.

But that's not all that makes this book different from the other scientific books on your shelf. I've included a CD that's full of cool full-color stuff for you to play with and lecture from.

The CD contains the FROG software (for the PC and Mac) that retrieves pulses from traces. This software is fun to play with even if you haven't set up a FROG; you can have it try theoretical (or experimental) traces and watch it work. Ken DeLong and Marco Krumbuegel (the authors of this software and both former post-docs in my group) sell this software for a few hundred dollars, so they've disabled the 'save' functions on these free versions. Please buy a copy from their companies (Femtosoft or MakTech) if you plan to do anything serious with it. They're nice guys who aren't making much money from this endeavor; they're mostly doing it for the benefit of humankind.

The CD also contains five hours of full-color PowerPoint lectures (for the PC and Mac in English and in French) that I give to my Ultrafast Optics class on ultrashort pulses and their measurement. So if you're teaching such a class, you just saved 200 hours of lecture preparation time when you bought this book! Indeed, I'm hoping that my supplying these polished files might persuade you to make the transition to high-tech teaching. Gone are the days when the most important attribute of a professor was his penmanship on the blackboard. Why don't all textbooks do this? I don't know. Okay, it might have something to do with those 200 hours it took to create them. Nevertheless, I think they should. Supplying prepared lectures along with a textbook could free up some time for teachers to actually improve the lectures, help students, do research, or maybe just relax.

And there's yet a third innovation in this book. I've written a couple of hundred pages, but the life of a professor is in busy one, and I don't have time to write about everything I'd like included in this book. So I've asked several other scientists to supply chapters on important relevant subjects on which they're the world's leading experts. In this way, we can cover everything, but still get this book to you in a reasonable amount of time. The result is that this is not a single-author book, but it's also not an edited book of independent chapters; it's a hybrid. Whatever works. I've tried to edit the style of these additional chapters to better match mine, but my style is sufficiently weird that I exhibited some restraint here to avoid irritating these wonderful folks who were kind enough to provide chapters.

The result is that roughly the first half of the book—which I mainly wrote—is more general, simpler, more informal, and about right for an advanced undergrad or a first- or second-year grad student, who's just learning about the fascinating world of ultrafast optics and who'd like to know the basic concepts of ultrashort pulses and their measurement. The second half—by the additional authors—is more specialized, more advanced, more formal, and about right for an older grad student or researcher who has to worry about the details of a specific pulse-measurement project. The book is so long that, if you start it your first year in grad school, you'll probably not get through it until a few years later when you're about to graduate, so things may work out just right.

Coincidentally, that's about the same time scale over which the research described in this book occurred. Only a decade ago, it wasn't possible to measure an ultrashort pulse. Autocorrelators provided a rough measure, but that was about it. FROG emerged in 1991, and it's changed the way ultrafast scientists think about their lasers and helped to provide an understanding of these lasers that has led to ever-shorter pulses.

This book is mainly about FROG, which has allowed us to measure an ultrashort laser pulse's complete intensity and phase vs. time and to do it very well and in a very general way. But it also discusses in some detail autocorrelators, partly for historical reasons, but also because an autocorrelator is a key component of a FROG. There's also some discussion of spectrometers for the same reasons. We also cover spectral interferometry (SI) because it nicely complements FROG: it's extremely sensitive (FROG isn't); it's linear (FROG is nonlinear); it requires a well-characterized reference pulse, and FROG, which doesn't, is the best way to obtain one.

Some people have asked me about including other methods, which (in my completely unbiased opinion...) are less well known, less general, less accurate, and more complex. Actually, that *was* my original plan, but I realized that all such methods are used by at most a few groups (usually just the group that invented it) for highly specialized purposes, and they already know about them. Typically, these methods are prohibitively complex: they often begin with a FROG, and then add numerous additional components—including such complex devices as interferometers and pulse stretchers!—seriously complicating an already nontrivial measurement. One involves an interferometer within an interferometer! (If you know someone who's using such a method, ask him which device he re-aligns when his pulse isn't short enough, the laser or the measurement device. I'll bet it's the latter...) Since these other methods are not in general use, there are only a few papers on each of them, and it's easy to do a quick literature search and read everything there is to know about them; a book on them is unnecessary. Many of them don't actually work or only work on a limited class of pulses—a fact that might not be evident from the papers—so consider yourself warned!

Also, FROG isn't just one technique; it's a class of powerful techniques, each with many variations. In addition, there are many clever things you can do with FROG that you can't do with other methods. For example, FROG has reliable independent checks on the measurement, something not present in any other method. These independent checks are very important because the corollary to Wayne's quotation on the first page of this book is that "If you measured it badly, you probably made it badly, too." Which suggests the following joke:

> Question: What's a poorly measured 5-femtosecond pulse?
> Answer: A 10-femtosecond pulse.

FROG can even measure the most complex ultrashort pulses ever generated (with a time-bandwidth product in excess of 1000); this is about three orders of magnitude more complex than the most complex pulse ever measured with any other method. Even its alleged weaknesses are in fact advantages: FROG's relatively slow (few-second) iterative algorithm makes it much more versatile than any other method. And its over-determination of the pulse allows such niceties as automatically calculated error bars and the correction for systematic error. More than 300 scientific papers describe FROG and its variations, features, and applications. And, as you can see, just covering FROG has required more than 400 pages—and we had to leave lots of stuff out! In the final analysis, I'd rather do one or two things well than a bunch badly.

In fact, if you feel that I've omitted something—like a reference to a paper—let me know, and I'll include it in the next edition. Keeping up with the literature—even just the FROG literature!—is becoming harder and harder everyday, so I'd appreciate the help.

Finally, when a professor writes a book, the folks who really pay the price are his grad students, who, as a result, are neglected so badly that their graduations can be delayed by as much as a year or more. I'm sensitive to this issue, so I've carefully avoided doing that. In view of the fact that scientists are even duller public speakers than they are writers, I took a different approach, and here's the resulting disclaimer:

> No graduate-student careers were harmed in the writing of this book. I wrote my share while pretending to take notes during dull

conference talks when the rest of the audience—and in some cases the speaker—were asleep.

Rick Trebino
Georgia Research Alliance-Eminent Scholar
Chair of Ultrafast Optical Physics

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1. The Dilemma

Rick Trebino

In order to measure an event in time, you must use a shorter one. But then, to measure the shorter event, you must use an even shorter one. And so on. So, now, how do you measure the shortest event ever created?

No, this isn't one of those age-old unresolvable dilemmas, the kind that frustrated ancient Greek philosophers. True, it's reminiscent of Zeno's paradox, which considered how finely one may divide distances, rather than durations of time. And it's equally confounding. But, in fact, the above dilemma is a recently solved optical measurement problem, which, until a few years ago, badly frustrated modern laser scientists.

And, unlike the conundra pondered by the ancient Greeks, which were of little practical value, the above dilemma has proven eminently practical. Indeed, to see the action in any fast event, whether it's a computer chip switching states, dynamite exploding, or a simple soap bubble popping, requires a strobe light with a shorter duration in order to freeze the action. But then to measure the strobe-light pulse requires a light sensor whose response time is even faster. And then to measure the light-sensor response time requires an even shorter pulse of light. Clearly, this process continues until we arrive at the shortest event ever created.

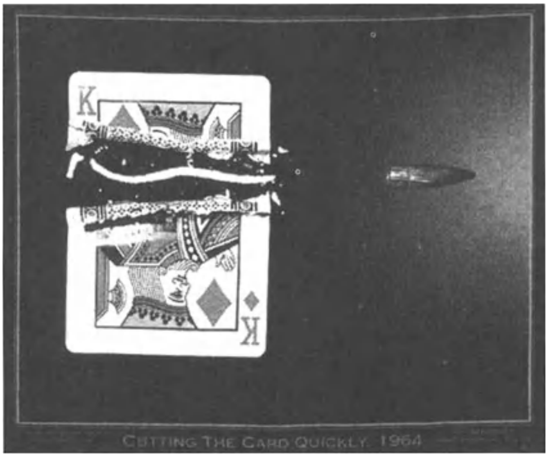

Fig. 1.1: A very short event. Measuring it requires an even shorter event: a strobe light only a few microseconds long. So then, how do you measure the strobe light? (Figure reprinted courtesy of Harold Edgerton collection).

R. Trebino, *Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses*

And this event is the ultrashort light pulse.

Ultrashort light pulses as short as a few femtoseconds (1 femtosecond = 1 fs = 1×10^{-15} sec) have been generated with lasers, and it is now routine to generate pulses less than 100 fs long. Here's some perspective on the mind-boggling brevity of these durations: 30 fs is to 1 second as 1 second is to a million years. Or, recalling the well-known fact that time is money, if one second corresponds to the current U.S. national debt ($5 trillion), then 10 fs corresponds to a mere nickel!

Now you might think that events this short would have little use; what happens on such short time scales that needs to be measured? The answer is: *A lot!* Key processes in biology—photosynthesis, vision, protein-folding, to name a few—all contain events that occur on fs time scales. Key processes in chemistry—molecular vibrations, re-orientations, and liquid-phase collisions, to name a few—also occur on this time scale. And key events in physics and engineering—high-lying excited-state lifetimes, photo-ionization, and electron-hole relaxation times that determine the response times of light detectors and electronics—are also ultrafast. The scientific literature of all of these fields contains many more.

Okay, so there's much to measure with these pulses. But why worry about the pulses themselves? Isn't that a problem of interest only to philosophers? The answer is: *No!* To begin with, we always need to check that a light pulse is in fact shorter than the event we're measuring with it. And if we actually know the precise pulse shape, we can use a pulse only slightly shorter than the event we're measuring with it, rather than one significantly shorter. Second, in many experiments—studies of molecular vibrations, for example—additional details of the pulse's structure play an important role in determining the outcome of the experiment. Of particular importance is the variation of color, or frequency, during the pulse, known as *chirp*. For example, chirped pulses can cause much greater molecular photo-dissociation than unchirped pulses [1]. Also, when a batch of molecules are excited, they make transitions to an excited state and then emit light whose color depends on the separation in energy between the excited state and ground state. Molecules are best described by *potential surfaces*, which are functions of the separation between nuclei in the molecule. As shown in Fig. 1.2, the color of the emitted light will change with time as the molecule vibrates or dissociates. Measuring such light tells us a great deal about the molecule. Third, we'd like to understand the physics of the lasers that emit these pulses, and, to verify theoretical models, we require precise knowledge of the pulse's properties [2–5]. And, in particular, to make even shorter pulses, we must understand the distortions that limit the length of currently available pulses [4,5]. Fourth, many new material-characterization techniques depend heavily on the ability to precisely characterize an ultrashort pulse experimentally. More detailed material information can be discerned by fully characterizing the input and output pulses in such methods [6,7]. Fifth, numerous applications have emerged for *shaped* ultrashort pulses [8,9]. A particularly interesting example of such an

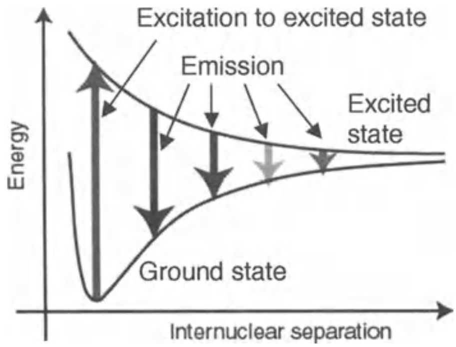

Fig. 1.2: Potential surface diagram for a generic molecule, showing that the emission color (here various shades of gray) changes with time after excitation (the upward-pointing arrow) from the ground to an excited-state surface. Knowledge of the time-resolved luminescence frequency yields important information about the potential surfaces, not available from a mere spectrum or intensity vs. time. This is especially the case for complex molecules—with more complex surfaces than those shown here.

application is the use of chirped pulses to generate novel states of matter unique to the quantum world and having no classical analog. Of course, in all such applications, one must verify that the correct pulse has been used. In general, *any optical measurement of a medium is ultimately limited by the ability to measure the light into and the light out of the medium*, so better light measurement techniques are a generally good idea.

So being able to measure ultrashort light pulses is of great practical value. But philosophical interest is nothing to be ashamed of. And we're not short of that here. Indeed, the measurement of fast events has fascinated humans since the dawn of time [10]. The ancients measured time intervals in days and developed devices such as sundials to measure shorter intervals. The hourglass and dripping-water methods eventually improved temporal resolution to better than 100 seconds. In the seventeenth century, Galileo Galilei used his heartbeat as a clock in his classic pendulum experiments, achieving an accuracy of close to 0.1 seconds. In 1819, de la Tour devised a standard of time based on sound. He noted that, because the human ear can hear sonic frequencies of greater than 10^4 Hz, periodic intervals transformed into sound waves by some means could be detected by ear to achieve a resolution of 0.0001 seconds. This method transformed the problem of time-measurement into the frequency domain. Many subsequently developed methods also made use of the frequency domain, reducing the problem of time-interval measurement to the often easier measurement of differences in frequency. Charles Wheatstone used electric discharges to ionize air and produced a momentary spark that could "freeze" motion. Henry Fox Talbot invented "instantaneous" photography in 1851, when he made an image of a newspaper on a spinning disk using a spark-discharge flash. Mid-nineteenth-century rotating-mirror streak techniques and excite-probe spark photography achieved microsecond resolution, largely due to the work of Ernst Mach (of Mach-number fame). By