

1. The Dilemma

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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.

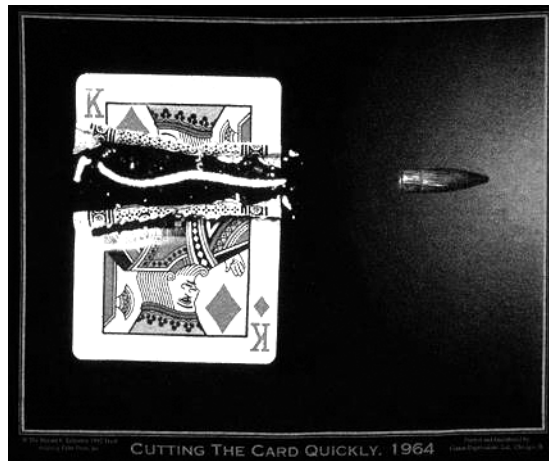


Fig. 1.1. A very short event. Measuring it requires an even shorter event: a strobe light only a few microseconds long. So then, how do you measure the strobe light? (Figure reprinted courtesy of Harold Edgerton collection)

1. The Dilemma

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.

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 emit-

1. The Dilemma

ted 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 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.

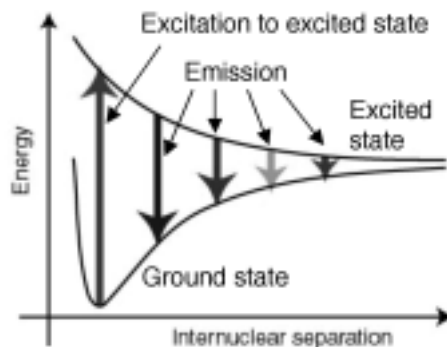


Fig. 1.2. Potential surface diagram for a generic molecule, showing that the emission color (here various shades of gray) changes with time after excitation (the upward-pointing arrow) from the ground to an excited-state surface. Knowledge of the time-resolved luminescence frequency yields important 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.

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 fasci-

1. The Dilemma

nated 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 the turn of the century, Abraham and Lemoine had demonstrated nanosecond resolution with a electrical-gate technique employing the dc Kerr effect. And in the middle years of the 20th century Harold Edgerton at MIT developed this work to a fine art (literally) with the development of high-speed strobe lights.

Modern electronic light detectors have pushed the limit of experimental temporal accuracy to picoseconds (1 picosecond = 1 ps = 1×10^{-12} sec). And finally, ultrashort-laser-pulse techniques, first developed in the 1970’s, are rapidly approaching single-fs resolution. A good fraction of these methods were developed simply because “they were there.” Indeed, nineteenth-century photographer, Muybridge, developed the first high-speed movie technology simply to *settle a bet* as to whether a galloping horse’s feet all left the ground at once. Many ultrafast scientists, myself included, proudly trace the history of their field back to this event.

So how do we measure ultrashort laser pulses? Obviously, we don’t have the required shorter event—a shorter pulse. And even if we did, we’d have the even harder problem of measuring this even shorter pulse. So it wouldn’t really help. Clearly, the shortest event available for measuring the pulse is *the pulse itself*. Indeed, early on, it was realized that we must use the pulse to measure itself. But, of course, that’s not good enough. As you might expect, techniques that have used the pulse to measure itself have yielded blurry pictures of the pulse: smeared out quantities that mask dips and bumps in the temporal shape of the pulse, just as the use of too slow a camera shutter speed yields a blurry picture of a moving object.

1. The Dilemma

As a result, the development of a technique simply to measure the pulse intensity vs. time remained an unsolved problem for many years, lagging badly behind humankind's ability to create such pulses. The problem of measuring the pulse phase (or, equivalently, its frequency or color) vs. time seemed beyond reach. As late as 1990, these two problems, which together comprise the task at hand, remained unsolved, despite the publication of hundreds of scientific papers on this topic by then.

Interestingly, these problems have recently been solved. The *Frequency-Resolved Optical Gating (FROG)* class of techniques, introduced in 1991 by Daniel J. Kane and Rick Trebino, can measure the full time-dependent intensity and phase of ultrashort light pulses in a wide variety of circumstances. FROG is rigorous, robust, and general; it works over a wide range of wavelengths, pulse energies, and pulse lengths. Using FROG, it's now possible to measure even the shortest pulse. It's possible to measure extremely complicated pulses. It's possible to measure a single pulse. It's even fairly easy, and it just recently became even easier. To give an idea of the current level of sophistication, FROG in conjunction with another technique, spectral interferometry, has measured a train of pulses with considerably less than a photon each.

And all without a shorter event.* It's an interesting story. It's a story with twists and turns, of seemingly unrelated ideas in fields ranging from acoustics to astronomy to number theory, working together to yield an elegant and robust solution. A key role will be played by the musical score, or rather, its mathematically rigorous cousin, the spectrogram. And an unlikely hero will emerge: *a theorem that fails in higher dimensions*. In fact, it's this failure that saves the day. It's almost an adventure story.

It's a story that involves FROGs, TADPOLES, and POLLIWOGs. No it's not a nature story; these are actually the rather frivolous acronyms for some of the more successful techniques. Despite their silly names, however, these techniques offer great measurement potential and capability unavailable previously. And they're finding applications in many labs around the world.

In order to tell this story, we'll first describe ultrashort laser pulses and define just what it is that we need to measure. This comprises the intensity and phase of a pulse vs. time or frequency. We'll also mention some assumptions that are implicit in pulse-measurement techniques and some properties of the temporal and spectral phase. That'll be Chapter 2.

In Chapter 3, we'll introduce the basic concepts of nonlinear optics in case you haven't seen these ideas before; this is essential because all ultrashort-light-pulse measurements require the use of nonlinear optics.

In Chapter 4, we'll describe the autocorrelation and spectrum, the old standards of short-pulse measurement, and the limited information available from them. These techniques are important because they're the building blocks for FROG, which is simply the spectrum of the autocorre-

1. The Dilemma

lation (no, I don't know why no one thought of it before, either**). It turns out that the retrieval of the pulse intensity and phase from the spectrum is an example of the *one-dimensional phase-retrieval problem*. It's the attempt to reconstruct a function of one variable from the magnitude of its Fourier transform (without knowing the phase). This isn't possible unless additional information is available. Typically, we might also know that the function is of finite duration. Unfortunately, even with this additional information, it's generally not possible to reconstruct the function. Indeed, the one-dimensional phase-retrieval problem is a notorious problem, well known to be unsolvable in almost all cases. Its unsolvability is very fundamental: it follows from the *Fundamental Theorem of Algebra*, which guarantees that we can factor polynomials of one variable. Coincidentally, the retrieval of the pulse intensity from the autocorrelation is also an example of the one-dimensional phase-retrieval problem.

In Chapter 5, we'll introduce the important notion of the *time-frequency domain*,^[11] the domain in which the most successful new techniques operate. Unlike the autocorrelation and the spectrum, which are pure time- or frequency-domain quantities, the time-frequency domain involves making measurements with both time- and frequency-resolution, *simultaneously*. We'll do some serious pulse measurement in Chapter 5, when we discuss the specifics of the FROG technique. We'll also discuss the *two-dimensional phase-retrieval problem*. This problem commonly occurs in astronomy and x-ray diffraction. Quite counter-intuitively, two-dimensional phase retrieval, unlike its one-dimensional cousin, *is* a solvable problem when some additional information is available. Two-dimensional phase retrieval turns out to be possible because the Fundamental Theorem of Algebra *fails* for polynomials of two variables. This fascinating and highly unintuitive fact was only discovered in the late 1970's.^[12] And it'll turn out that FROG succeeds because the *retrieval of the pulse intensity and phase from the FROG trace is equivalent to the two-dimensional phase-retrieval problem*. We'll show how to make a FROG trace of a pulse and how to interpret it. And we'll discuss how FROG avoids the dilemma at the beginning of this chapter.

In Chapter 6, we'll discuss various beam geometries for FROG, which make it the versatile technique that it is. We'll also give the details you need to decide which geometry is right for you and how to set it up.

It's also possible to measure the intensity and phase of a *single* ultrashort laser pulse. This is accomplished simply by crossing beams at a large angle. The entire trace can then be obtained on a single camera image (Chapter 7). Such geometrical effects are not always desirable, however, and Chapter 7 also includes a discussion of these effects, which can almost always be avoided but should nevertheless be understood.

1. The Dilemma

FROG traces can often be interpreted by simple visual inspection. Nevertheless, it's important to be able to retrieve the mathematical form of the pulse intensity and phase from them. Doing so requires an iterative algorithm. Fortunately, there are several, most derived from phase-retrieval algorithms. These algorithms are the subject of Chapter 8.

FROG is extremely insensitive to noise. This results from the great over-determination of the pulse. The FROG trace is an $N \times N$ array, used to determine only N intensity and N phase points, or $2N$ points. This over-determination has many advantages, one of which is noise reduction. Even better, it allows simple filtering techniques, which can improve the signal-to-noise ratio of retrieved pulses tremendously. This work is the subject of Chapter 9, and you shouldn't do FROG measurements without first having read this chapter!

Measurements of ultrashort laser pulses are prone to many types of errors. As a result, John Dudley likes to say that there are two kinds of ultrashort-laser pulse measurements: *those with independent checks and those that are wrong*. In Chapter 10, we'll discuss some practical issues in making FROG measurements, such as independent checks. We'll show how to verify that measured traces are consistent with other data, such as the spectrum and autocorrelation. An important aspect of FROG is that such checks are available; they're not in autocorrelation and spectral measurements or in other intensity-and-phase measurement techniques, where it isn't possible to know whether the measurement you just made is correct and free of systematic error.

A number of additional simple tricks exist that make building and using a FROG not only easy, but also more accurate, and they're the subject of Chapter 11. One such trick allows us to automatically calibrate a FROG. It also removes the ambiguity in the direction of time that occurs in one of the FROG beam geometries.

FROG is a relatively simple measurement technique experimentally, only slightly more complex than an autocorrelator. But that doesn't mean that FROG can't be simplified further. Quite surprisingly, a FROG trace of a pulse can be produced by an almost trivial device composed of as few as five simple optical elements. This extremely simple device is called GRating-Eliminated No-nonsense Observation of Ultrafast Incident Laser Light E-fields (GRENOUILLE). Whereas an autocorrelator has four sensitive alignment parameters, GRENOUILLE has *no sensitive alignment parameters at all!* It can be set up in minutes, making it the simplest ultrashort-pulse-measurement device in history. It almost fits in your pocket. And it works. Furthermore, it has much better sensitivity than any other technique, including FROG. Oh, by the way, if you don't speak French, "grenouille" is French for "frog," but you probably could've guessed that. Chapter 12 will discuss this recent development.

1. The Dilemma

Okay, so we've told you everything you can possibly need to know about the FROG technique, in general. What about its use in practical—and difficult—situations, such as measuring UV and high-power pulses? The numerous beam geometries available for FROG measurements become important in these regimes, and Chapter 13 summarizes this issue.

FROG is also ideal for measuring extremely, incredibly, unbelievably short pulses (that is, shorter than merely ultrashort; clearly, we're running out of superlatives), only a few fs long. Indeed, at the time of this writing, the shortest event ever created is an ultrashort laser pulse only 4.5 fs long, and it was measured using FROG. Chapter 14 discusses in detail the issues associated with measuring such extremely short pulses.

Most researchers assume that their pulses are smooth and single-peaked. This is usually not true, however, and it was not until the development of FROG that this has become apparent because autocorrelation tends to mask and smooth out pulse structure. FROG is the only method that is capable of measuring complex pulses. This is because a FROG trace's large number of points (~10,000 in a 100 x 100 trace) give it the information capacity to measure a pulse with a large amount of structure. Indeed, no other technique has ever measured the intensity and phase of a pulse with a time-bandwidth product greater than about 3. And FROG has measured pulses with time-bandwidth products approaching 100. Chapter 15 will discuss the issues involved in such measurements.

The big advantage of FROG is that it requires no reference pulse—the pulse can measure itself. This is great because there usually aren't any reference pulses lying around. But occasionally there are. Occasionally, a laser emits a fairly smooth pulse, which can be measured using FROG, and which then undergoes some sort of optical torture, becoming a ragged and disheveled mess as a result. As we mentioned above, such a pulse can often be measured using FROG, but why bother? A much more intuitive trace results when measuring the ragged pulse with the smooth, known reference pulse. This variation on FROG, which yields a traditional spectrogram, is usually referred to as *Cross-FROG (XFROG)*, and it's the subject of Chapter 16.

When you need to measure really complex pulses, it's better to use XFROG. Interestingly, it's only recently become possible to generate extremely complex pulses—with time-bandwidth products in excess of 1000. While the measurement of such pulses may seem impossible, in fact, XFROG has been used to measure such pulses; this is the subject of Chapter 17.

Amazingly, it's even possible to measure pulses using a medium with an arbitrary response. Because FROG uses a very versatile iterative algorithm, it can easily be modified to deal with almost any situation, including one in which the nonlinear-optical medium is slow. Chapter 18 will show how to do this.

1. The Dilemma

An example of such a situation is *Fiber-FROG*, which makes FROG measurements inside an optical fiber. Chapter 19 will show how. Fiber-FROG is most useful for measuring pulses of 1.5 μm in wavelength.

Okay, so measuring an ultrashort pulse is becoming almost routine. So how about a much harder problem. How about measuring two pulses? At the same time? And how about doing so using not much more than the apparatus already sitting on the table? And just to really complicate the problem, let's do so at the sample medium in an ultrafast spectroscopy apparatus. In this case, we can actually measure both pulses in a manner that actually takes advantage of the apparatus that's being used to do an ultrafast spectroscopy experiment. This method is often called *Blind FROG*, because it's equivalent to the mathematical problem called *Blind De-convolution*. It's the subject of Chapter 20.

The algorithm for Blind FROG is similar to the standard FROG algorithm, but Dan Kane has developed a technique called *principal components generalized projections*, which can, not only retrieve pulses from Blind FROG traces, but also retrieve them from standard FROG traces and do so very rapidly. In fact, his new algorithm (Chapter 21) is so fast that it can retrieve pulses in real time.

What about the measurement of very weak ultrashort pulses? Making a FROG trace of a train of ultrashort laser pulses with less than about a picojoule of energy each is difficult because FROG (and any other such technique) requires the use of a nonlinear-optical process. And nonlinear-optical processes require fairly intense pulses and hence cannot work for such weak pulses. A *linear*-optical method is thus required. But it can be shown that linear-optical methods cannot completely measure ultrashort pulses. So in Chapter 22 we discuss the problem of measuring weak pulses. The solution in most cases is to realize that weak pulses are not created "in a vacuum;" they are almost always created from stronger pulses. Indeed, in order to create an ultrashort laser pulse in the first place, the laser must use nonlinear-optical processes. As a result, the pulse directly from the laser is almost always intense enough to be measured using FROG, and then it can act as a reference pulse for measuring the weak pulse. One option is XFROG. But when such a well-characterized reference pulse is available, linear-optical methods also suffice. A particularly simple and useful method, *spectral interferometry*, is available. Use of spectral interferometry in this manner, in conjunction with FROG to measure the reference pulse—a technique called *Temporal Analysis by Dispersing a Pair Of Light E-fields, or TADPOLE*—has succeeded in measuring a train of pulses with less than a photon per pulse!

Chapter 23 discusses an even more difficult-sounding problem: the measurement of the ultrafast variation of a pulse's polarization state. Indeed, this problem sounds impossible—isn't this what is meant by "unpolarized light," that is, light whose polarization state varies too quickly to

1. The Dilemma

be resolved? If we throw in the additional difficulty that the light happens to be very weak, as well, the problem becomes quite a challenge. But a very simple approach, involving simultaneous TADPOLE measurements of both polarizations of the light, yields the solution. This relative of TADPOLE is appropriately referred to as *POLarization-Labeled Interference vs. Wavelength for Only a Glint (POLLIWOG)*. Okay, I admit it; I stayed up really late one night coming up with that name.

In addition, it's occasionally important to combine the advantages of both FROG and spectral interferometry. FROG has the advantage of incorporating an ultrafast gate, which eliminates any continuous background light that may be present. But because FROG involves the pulse gating itself, it doesn't measure the arrival time of a pulse. Usually, this information is undesirable; who cares how far it is from the laser to the FROG? And who would like to take the massive effort to accurately stabilize this distance? But occasionally, for example, in spectroscopic measurements in plasmas, these quantities are crucial. Spectral interferometry, can yield the arrival time easily, but it does not involve any gating, and so spectral interferometry measurements can be badly contaminated by continuous background. A combination technique, called *Multipulse-Interferometric FROG (MIFROG)*, offers the best of both worlds and is the subject of Chapter 24.

Finally, in Chapter 25, we conclude by mentioning a few issues to be considered in the future: future applications for FROG, variations that would be welcome, and unsolved pulse-measurement problems.

Many interesting applications have been found for FROG, but it is important to limit the length of this book, and so we've chosen to limit our discussion to the technique, its implementation, and its variations, and leave it to you to find additional clever applications for it.

* Actually, a medium with a more rapidly responding event (usually, a non-resonant electronic response) is, in fact, required. But this event is not harnessed in the same sense as a strobe light. The strobe in these techniques is, in fact, the convolution of the electronic response and the light pulse, that is, an event on the order of the pulse itself.

** Actually, Ishida and coworkers[13, 14] made spectrally resolved autocorrelations (i.e., FROG measurements) in the 1980's but did not attempt to extract the pulse intensity and phase from them.

1. The Dilemma

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