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Introduction

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What is ultrafast?

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What is ultrafast?

Why?

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What is ultrafast?

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Optical Pulses

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Introduction

What is ultrafast?

Why?

Optical Pulses

Significance for Chemistry

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Introduction

- What is ultrafast?
- Why?
- Optical Pulses
- Significance for Chemistry
- The new frontier

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Introduction

- What is ultrafast?
- Why?
- Optical Pulses
- Significance for Chemistry
- The new frontier
- What will this course cover?

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Introduction

What is ultrafast?

Why?

Optical Pulses

Significance for Chemistry

The new frontier

What will this course cover?

If I take the course how do I get a grade?

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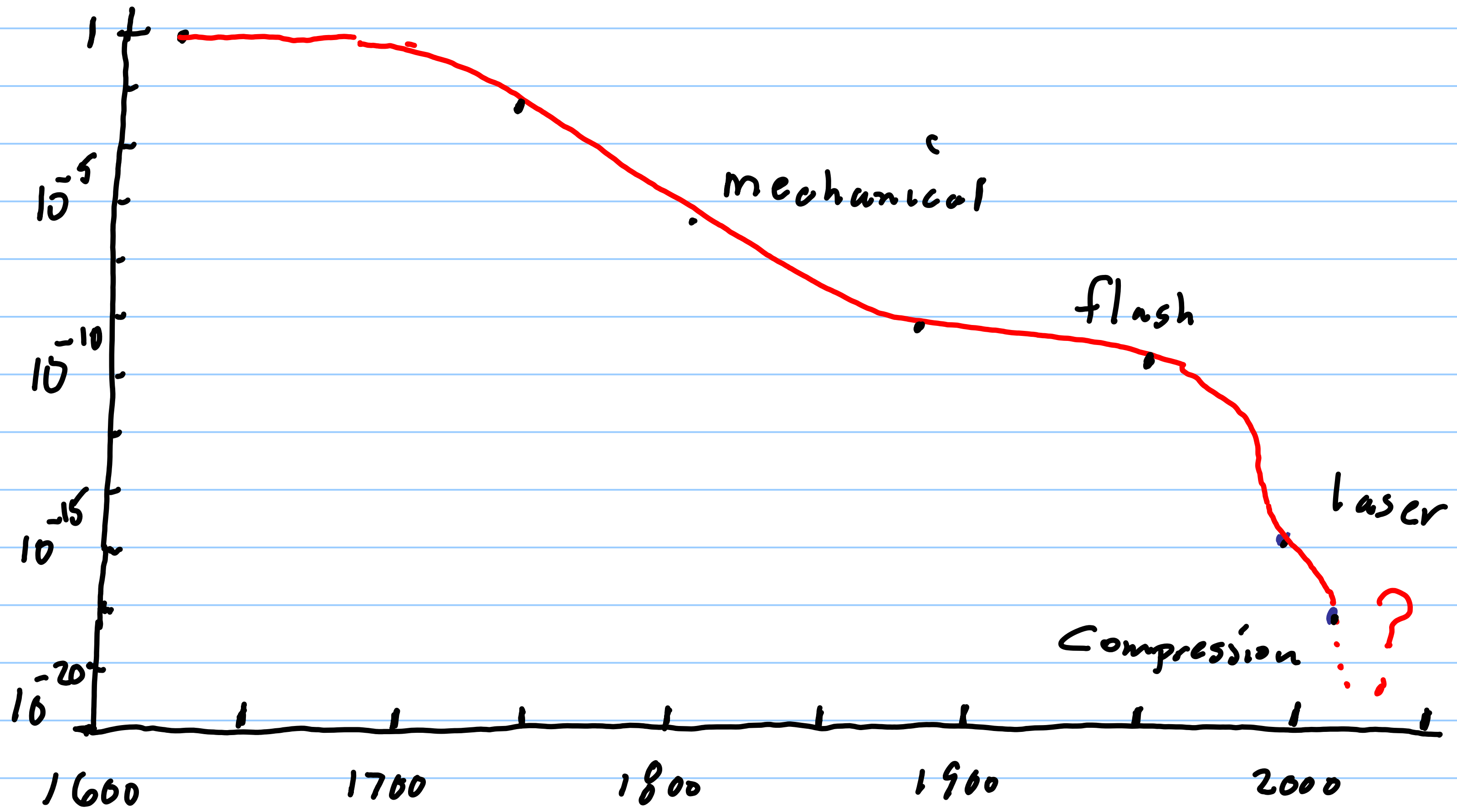
What is ultrafast?



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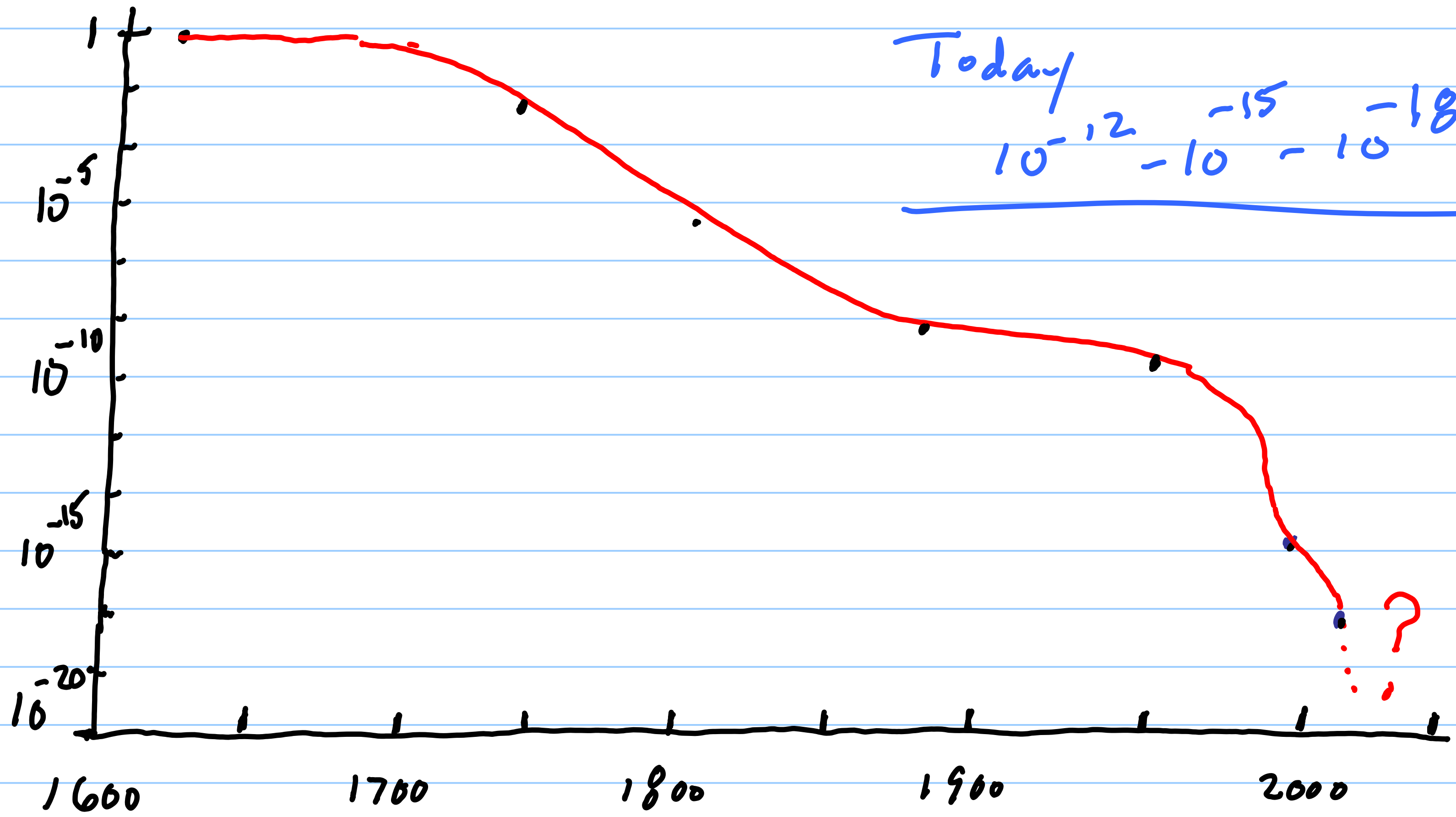
What is ultrafast?



Today

$10^{-12} - 10^{-15} - 10^{-18}$

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Why?

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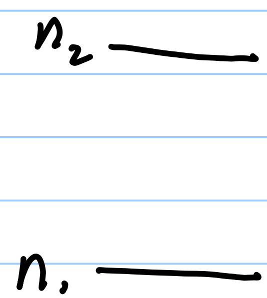
Why?

Consider a simple two level system



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Why?

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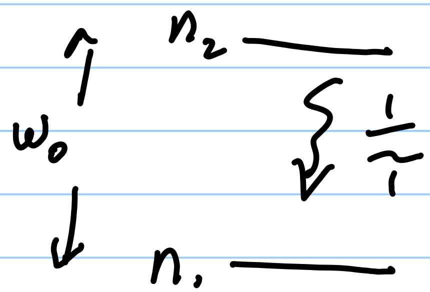

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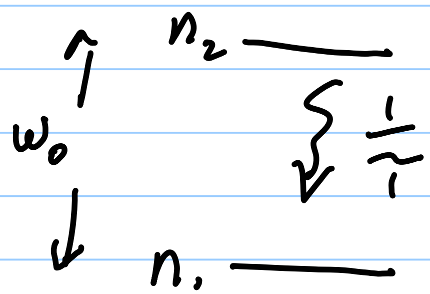
Why?

Consider a simple two level system



Spontaneous emission

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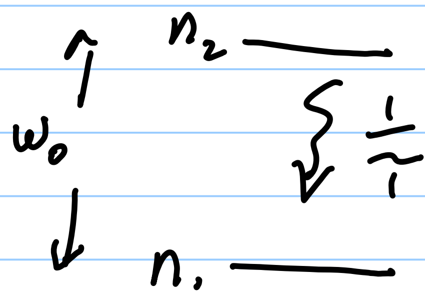
Why?

Consider a simple two level system



Spontaneous emission

$$N_2(t) = n_{20}\, e^{-t/\tau}$$

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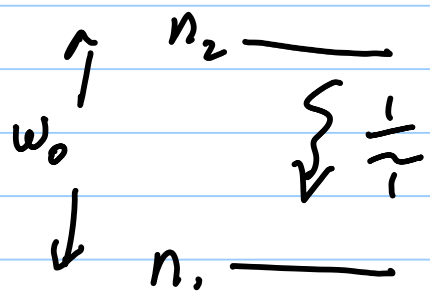
Why?

Consider a simple two level system



Spontaneous emission

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$$n(\omega) \sim \frac{1}{1+\omega^2\tau^2}$$

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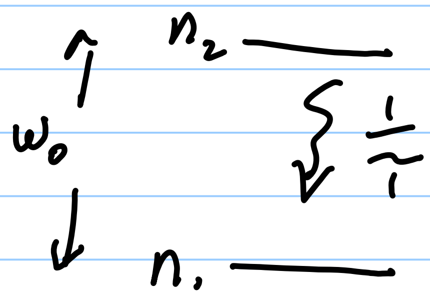
Why?

Consider a simple two level system



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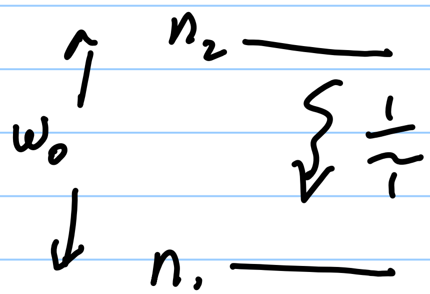

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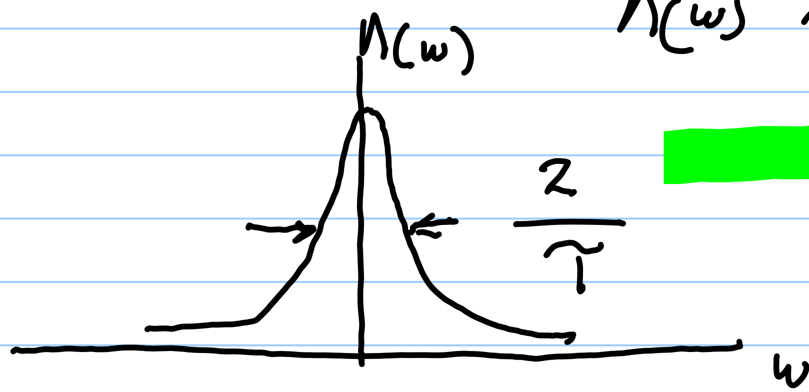

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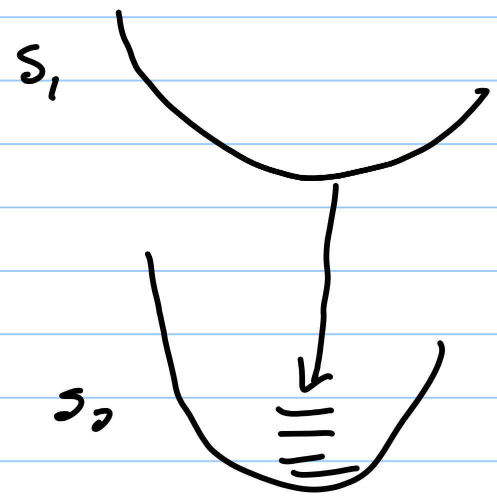
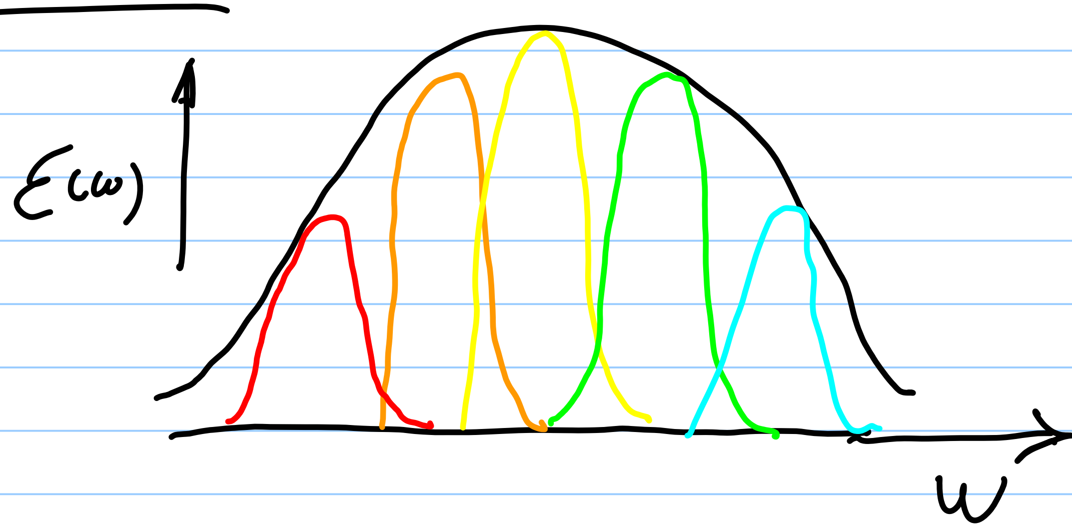
Molecular Emission





Temporal Dynamics Useful!

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Optical Pulses

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Optical Pulses

 light bulb

Lecture 1 August 29

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Optical Pulses

light bulb

$i(\omega)$

Frequency Domain

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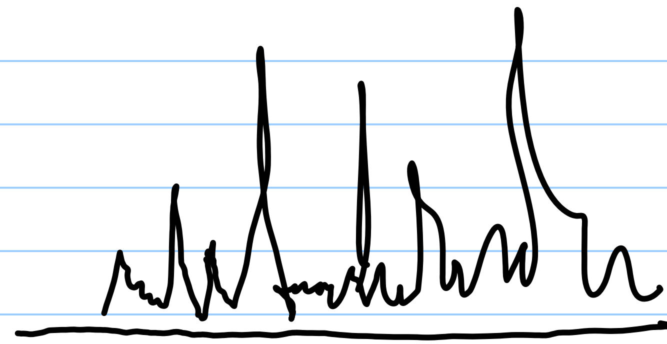
Optical Pulses

light bulb

$i(\omega)$

Frequency Domain



$I(t)$

Time Domain

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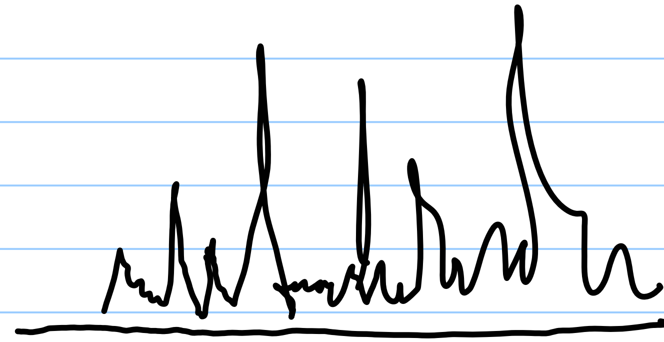
Optical Pulses

light bulb

$i(\omega)$

Frequency Domain



$I(t)$

Time Domain

- Associated with each frequency is a phase

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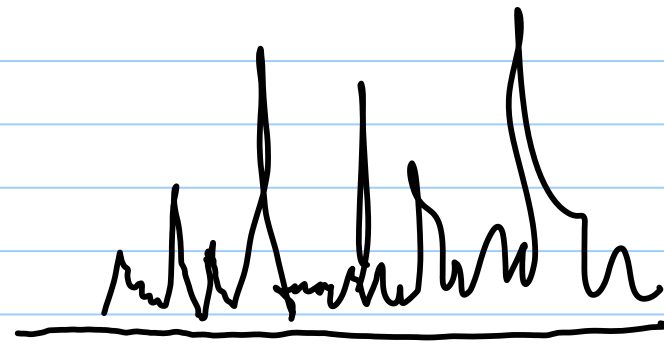
Optical Pulses


light bulb


$i(\omega)$


$I(t)$

Frequency Domain **Time Domain**

- Associated with each frequency is a phase
- In a light bulb the phase is random

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Note Title 8/20/2006

laser pulse

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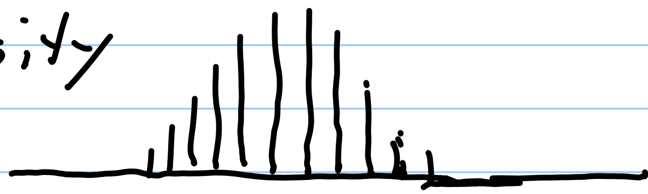
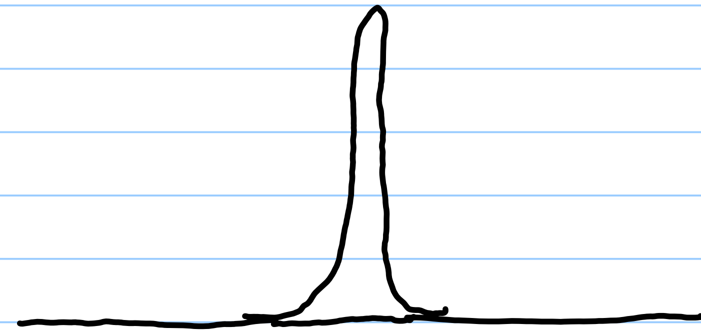
laser pulse (ideal)

Intensity



i(ω)



I(t)

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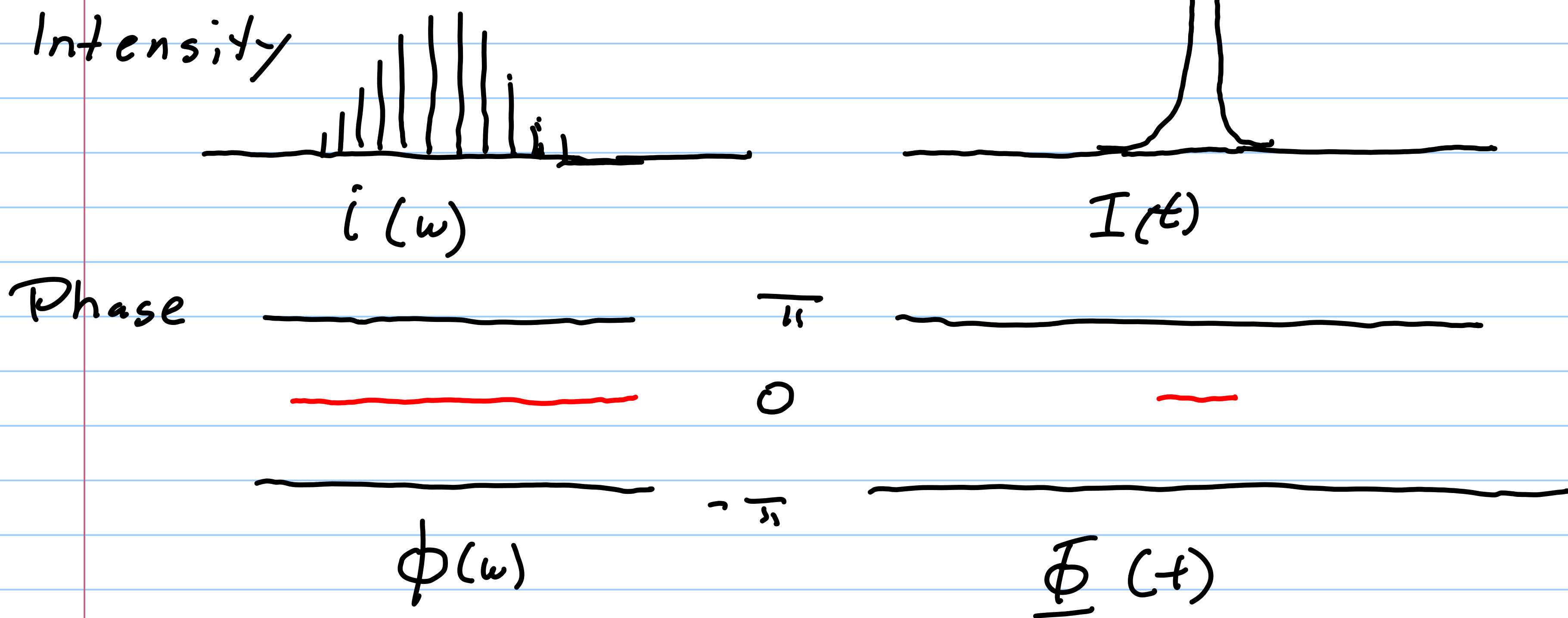
laser pulse (ideal)



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Electric Field

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Electric Field

Time

$$E(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \varepsilon(\omega)\, e^{-i\omega t}\, d\omega$$

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Electric Field

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Slowly varying amplitude approx

$$\Delta\omega < \omega_0$$

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Slowly varying amplitude approx

$$\Delta\omega < \omega_0$$

$$\varepsilon(\omega) = a(\omega)\, e^{-i\phi(\omega)}$$

$$E(t) = A(t)\, e^{i[\phi(t) - \omega_0 t]}$$

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Instantaneous Intensity

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Note Title 8/20/2006

Instantaneous Intensity

$$I(t) = E(t)E^*(t) = A^2(t)$$

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Note Title 8/20/2006

Instantaneous Intensity

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Similarly the spectrum becomes

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Parseval's theorem

$$\int_{-\infty}^{\infty} I(t)\,dt = \int_0^{\infty} a^2(\omega)\,d\omega$$

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Note Title 8/20/2006

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Pulse energy proportional to are under spectrum or intensity profile

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Uncertainty Principle

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Uncertainty Principle

$$\Delta\omega \, \Delta t / 2\pi \geq K$$

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Uncertainty Principle

$$\Delta\omega \, \Delta t / 2\pi \geq K$$

A transform limited pulse width

$$\Delta t = 2\pi K / \Delta\omega$$

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Uncertainty Principle

$$\Delta\omega \, \Delta t / 2\pi \geq K$$

A transform limited pulse width

$$\Delta t = 2\pi K / \Delta\omega$$

K depends on pulse shape

For a gaussian

$$i(\omega) = e^{-(\omega-\omega_0)^2/\alpha}$$

$$I(t) = \alpha e^{-\alpha(t-t_0)^2}$$

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Uncertainty Principle

$$\Delta\omega \, \Delta t / 2\pi \geq K$$

A transform limited pulse width

$$\Delta t = 2\pi K / \Delta\omega$$

K depends on pulse shape

For a gaussian

$$i(\omega) = e^{-(\omega-\omega_0)^2/\alpha}$$

$$I(t) = \alpha e^{-\alpha(t-t_0)^2}$$

and $K = \dfrac{2\ln 2}{\pi} = .441$

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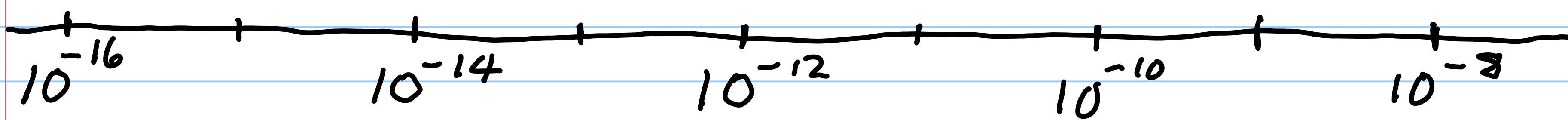
Chemistry



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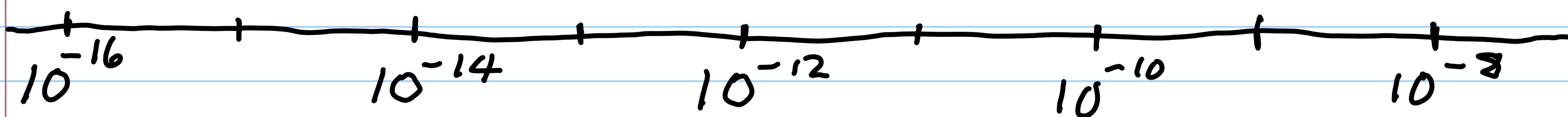
Chemistry

Liquid Collisions   Molecular Rotation



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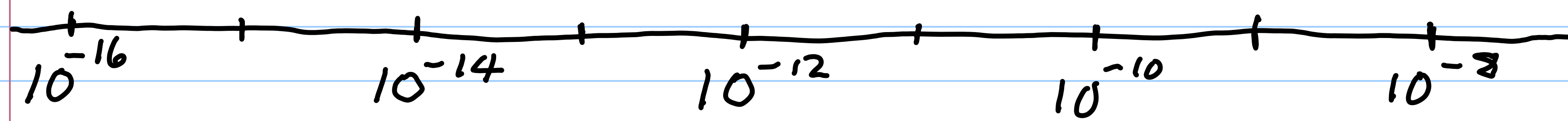
Chemistry

Liquid Collisions Molecular Rotation

Solvent Relaxation



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Chemistry

Liquid Collisions

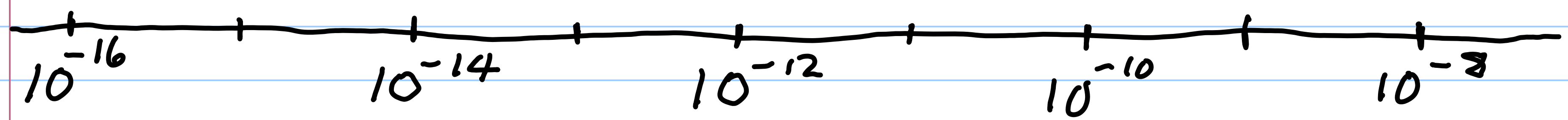
Molecular Rotation

Solvent Relaxation

Electronic Dephasing

Vibrational Relaxation

Vibrational Motion



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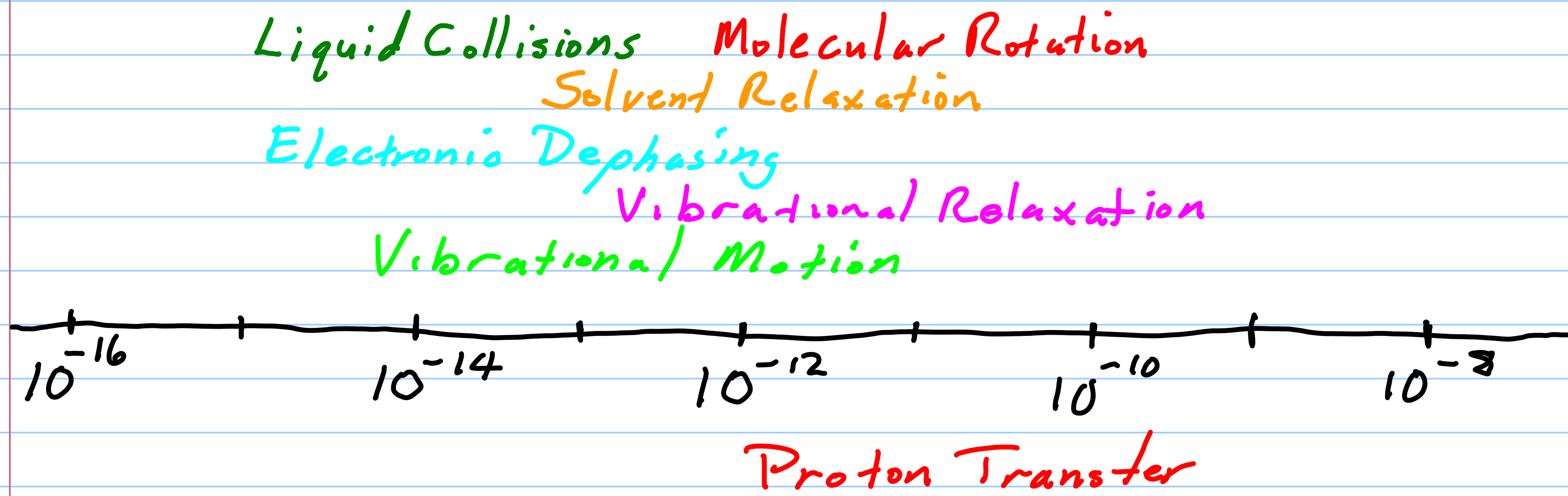
Chemistry



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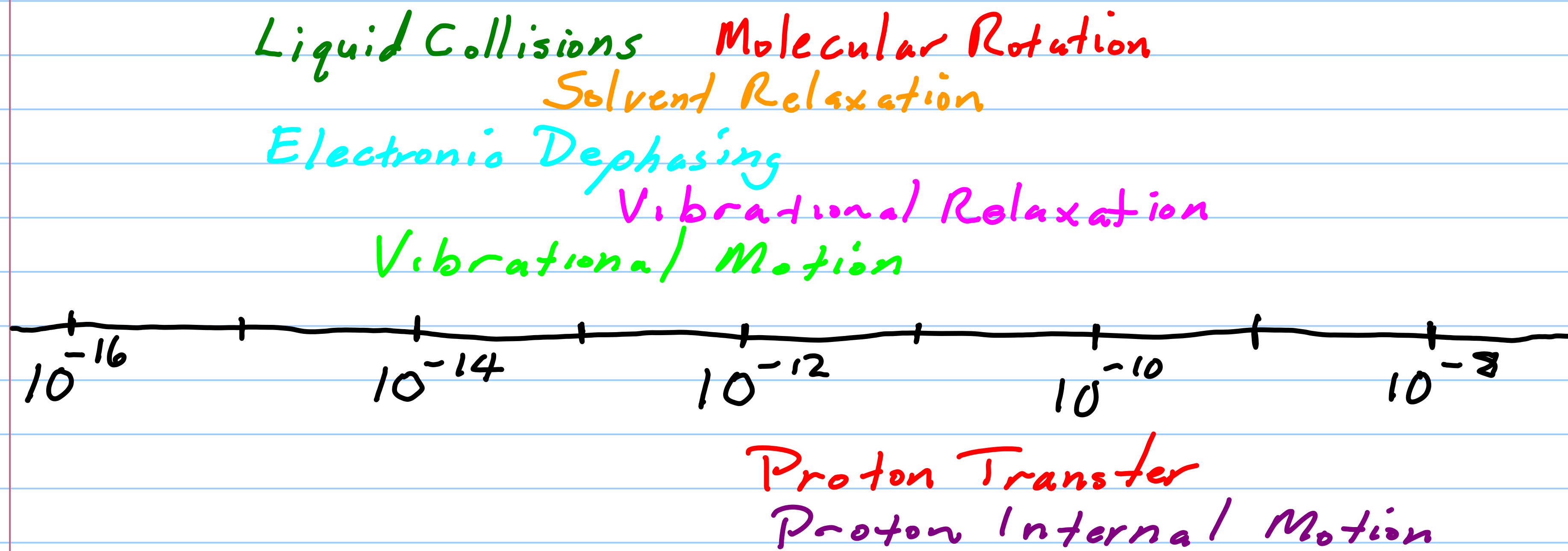
Chemistry



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Note Title 8/20/2006

Chemistry

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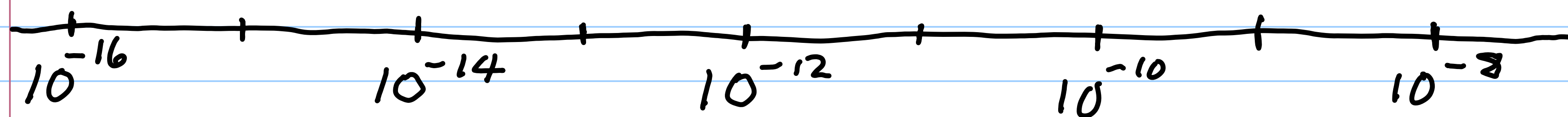
Solvent Relaxation

Electronic Dephasing

Vibrational Relaxation

Vibrational Motion



Proton Transfer

Proton Internal Motion

Photo Isomerization

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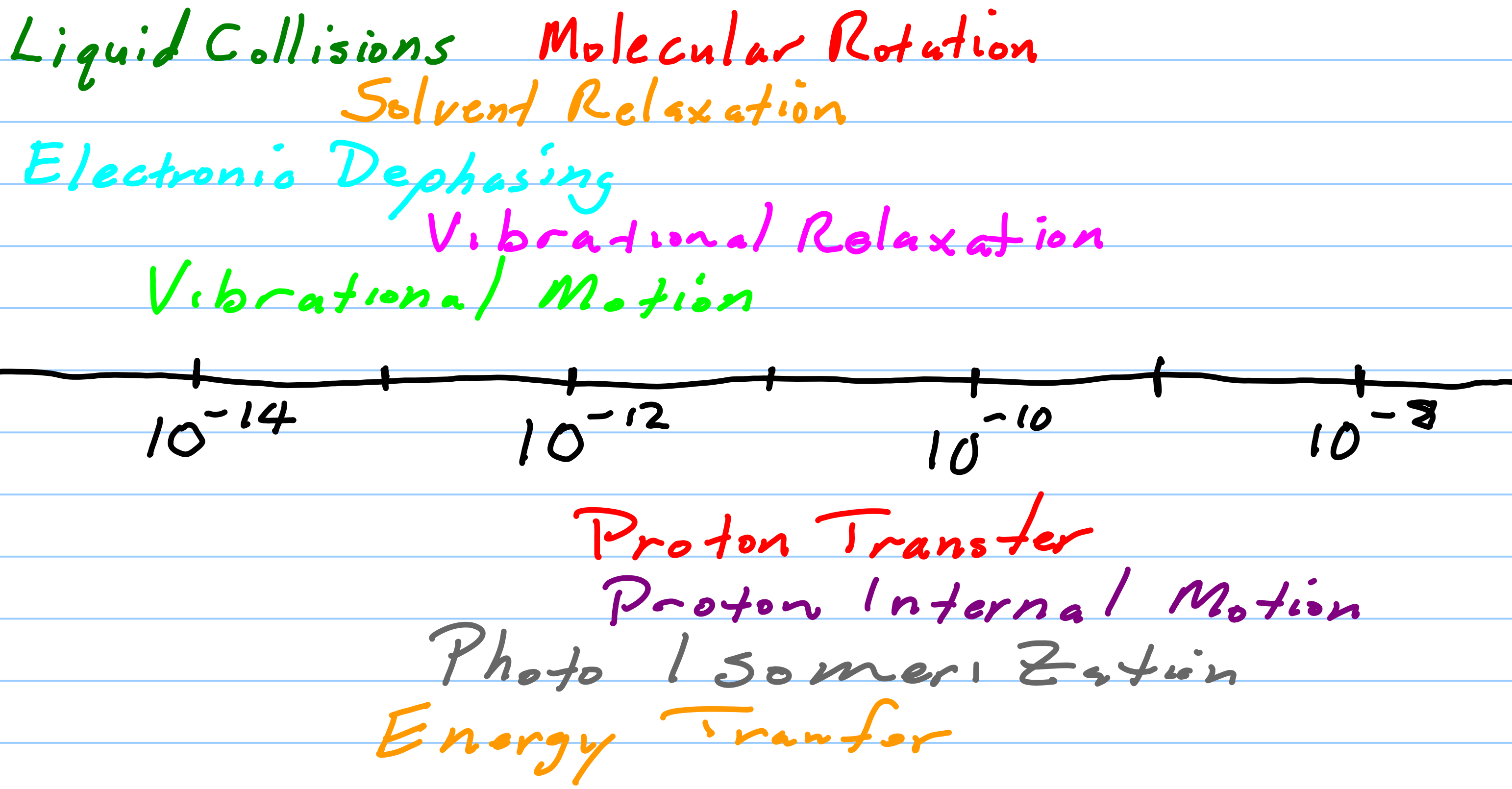
Chemistry



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Chemistry

Liquid Collisions Molecular Rotation

Solvent Relaxation

Electronic Dephasing

Vibrational Relaxation

Vibrational Motion

10^{-16} 10^{-14} 10^{-12} 10^{-10} 10^{-8}

Proton Transfer

Proton Internal Motion

Photo Isomerization

Energy Tranfer

Inner shell Auger Processes

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The new frontier

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The new frontier

- time resolved structure

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The new frontier

- time resolved structure
    - synchrotron sources

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The new frontier

- time resolved structure
  - synchrotron sources
    - ALS slicing

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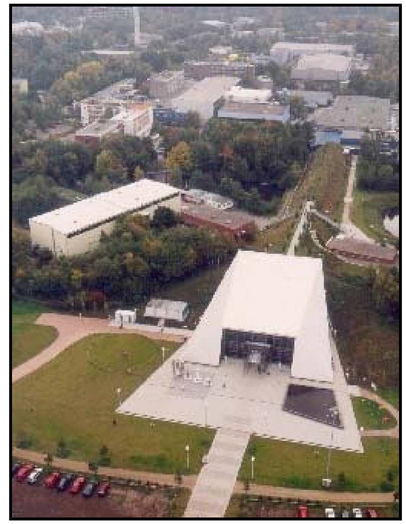
The new frontier

- time resolved structure
  - synchrotron sources
    - ALS slicing
  - VUV free electron lasers

Free Electron Laser

Update: May 3, 2006
webmaster

A "Free Electron Laser" for wavelengths down to 6 nm in the vacuum-ultraviolet and soft X-ray regime (**FLASH**, the former VUV-FEL) is under construction at the TESLA Test Facility (TTF) at DESY. It is operated in the "self-amplified spontaneous emission" (SASE) mode and delivers sub-picosecond radiation pulses, with gigawatt peak powers. At present, lasing has been observed down to about 30 nm in the fundamental and 10 nm in the third harmonic, the shortest wavelength ever achieved with a free electron laser. User experiments using this unique radiation started in August 2005 and were up to now carried out around 45 nm and 32 nm. For these wavelengths, FEL pulse intensities from typically a few µJ up to more than 100 µJ have been obtained with pulse lengths between 20 and 50 fs.



Aerial view of the experimental hall (Bldg. 28c) for the FLASH User Facility (center) and the tunnel for the TTF phase 2 extension behind it (covered with grass). The hall in the upper right corner was initially housing the TTF phase 1 FEL.

In addition, an **European X-ray FEL (XFEL) project** for wavelengths just below 0.1 nm is prepared at DESY.

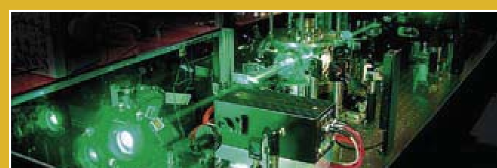
Due to the short pulse length and their high peak brilliance these FELs will open up exciting new paths for basic research and application-oriented studies, giving scientists, for example, insight into hitherto unknown properties of materials.

THE FUTURE: A CASCADE OF LIGHT

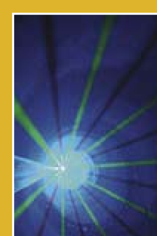
FERMI@ELETTRA (Free Electron Laser Radiation for Multidisciplinarity Investigations) is a project for the realization of a free electron laser. It will be a unique fourth generation source, able to produce very intense and short flashes of light in the wavelength interval between the far ultraviolet and x-rays.

FERMI @elettra

The electron bunches, accelerated by a Linac (a 120 meter long linear accelerator), will pass through a sequence of undulators (magnetic structures that force the electrons to wiggle) emitting ultra-short light pulses of great spectral pureness and with extraordinary peak power. The new light source will allow researchers to explore even farther in space and time, following the evolution of chemical reactions in time scale of a tenthousandth of a billionth of a second and scanning matter of microscopic dimensions, down to the nanometer. These will be the new frontiers for basic and applied research.



www.elettra.trieste.it/FERMI/



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The new frontier

- time resolved structure
  - synchrotron sources
    - ALS slicing
  - VUV free electron lasers
  - X-ray lasers

LCLS Linac Coherent Light Source

Stanford Linear Accelerator Center

Stanford Synchrotron Radiation Laboratory

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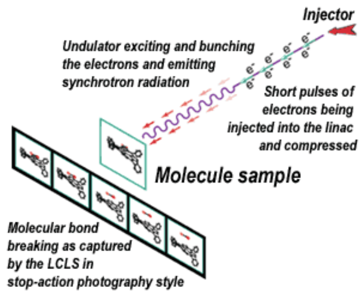
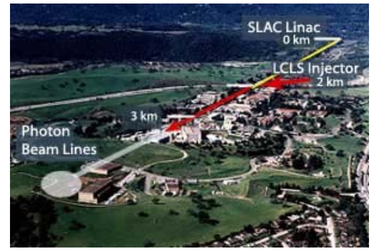
LCLS FAC

LCLS Science

LCLS Machine

FEL Resources

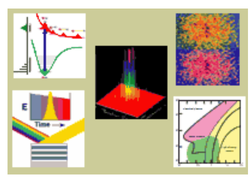




Downloadable LCLS brochure

The Linac Coherent Light Source (LCLS) will be the world's first x-ray free electron laser when it becomes operational in 2009. LCLS is currently in the detailed project engineering and design phase, with a construction start planned in FY2005. Pulses of x-ray laser light from LCLS will be many orders of magnitude brighter and several orders of magnitude shorter than what can be produced by any other x-ray source available now or in the near future. These characteristics will enable frontier new science (click box below to explore LCLS science) in areas that include discovering and probing new states of matter, understanding and following chemical reactions and biological processes in real time, imaging chemical and structural properties of materials on the nanoscale, and imaging non-crystalline biological materials at atomic resolution. The LCLS project is funded by the U.S. DOE and is a collaboration of six national laboratories and universities.

LCLS Science



New horizons in the domain of the ultra-fast and ultra-small world

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EPICS Meeting April 27-29, 2005

LCLS Diagnostics & Commissioning Workshop Sept 22-23, 2004

Ultrafast Science Workshop Oct 25-26, 2004

Past Workshops

Reviews

LCLS FAC Meeting Oct 12-13, 2004

LCLS SAC Meeting Oct 27-28, 2004

Past Reviews

Sub-Picosecond Pulse Source

SPPS

Guest House

Collaborating Institutions

LLNL

UCLA

Los Alamos

ANL

BNL

XFEL

The European X-Ray Free-Electron Laser

Technical Design Report

DESY 2006-XXX

JULY 2006

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The new frontier

- time resolved structure
  - synchrotron sources
    - ALS slicing
  - VUV free electron lasers
  - X-ray lasers
- Attoseconds

Subfemtosecond XUV Pulses: Attosecond Metrology and Spectroscopy

Reinhard Kienberger and Ferenc Krausz

Institut für Photonik, Technische Universität Wien, Gusshausstr. 27/387,
1040 Wien, Austria
reinhard.kienberger@tuwien.ac.at

Abstract. The generation of ever shorter pulses is a key to exploring the dynamic behavior of matter on ever shorter timescales. Recent developments have pushed the duration of laser pulses close to its natural limit, to the wave cycle, which lasts somewhat longer than one femtosecond (1 fs = 10^{-15} s) in the visible spectral range. Time-resolved measurements with these pulses are able to trace dynamics of molecular structure but fail to capture electronic processes occurring in atoms on an attosecond (1 as = 10^{-18} s) timescale. The generation of high-order harmonic radiation in the extreme ultraviolet and soft X-ray regime from atoms exposed to intense few-femtosecond laser pulses comprising just a few wave cycles opened the way to the generation of isolated XUV/X-ray pulses shorter than 1 fs. This chapter will focus on the generation and measurement of these pulses (metrology) and discuss possible ways of using these atomic-timescale bursts of electromagnetic radiation for tracking ultrafast electronic processes with attosecond resolution (spectroscopy).

1 Generation of Atomic-Timescale Pulses

Electrons inside atoms and molecules can be excited and subsequently relax or interact with each other within attoseconds, requiring subfemtosecond pulses for capturing these ultrafast motions. As nature limits the shortness of light pulses to one wave cycle, which lasts somewhat longer than one femtosecond in the visible spectral range, one has to draw on XUV or X-ray radiation for producing attosecond pulses. Excellent coherence and relatively low pump-energy requirements, which can be readily met by table-top-femtosecond laser systems, make high-order harmonic generation (HHG) an attractive approach to developing a laboratory short-wavelength source [1, 2, 3]. The XUV radiation emerges as an electron, is detached from an atom by a high laser field, and then driven back to its parent ion where it can recombine into its original bound state. This process is repeated quasi-periodically, each half cycle of the driving laser wave, resulting in a discrete frequency spectrum made up of high-order odd harmonics of the driving laser. The highest order harmonics are produced near the intensity peak of the laser pulse, as the recollision energy of the electrons is highest when the laser field is largest. Using a few-cycle driver [4, 5, 6] (see also Chapter by *De Silvestri* in this volume) and passing the harmonics through a filter that transmits only the highest energy photons, we select radiation produced within only a fraction of the laser

F. X. Kärtner (Ed.): Few-Cycle Laser Pulse Generation and Its Applications,
Topics Appl. Phys. 95, 343–379 (2004)

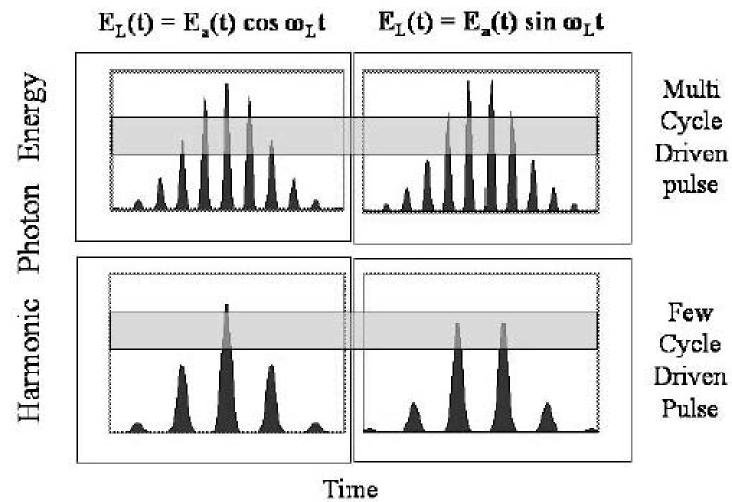

Fig. 8. Comparison of the temporal structure of XUV pulses emerging from an HHG driver by multicycle or few-cycle pulses for different values of the carrier–envelope phase. The peaks sketch the temporal structure (*horizontal axis*) of emitted XUV radiation as a function of XUV photon energy (*vertical axis*)

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Note Title 8/20/2006

The new frontier

- time resolved structure
  - synchrotron sources
    - ALS slicing
  - VUV free electron lasers
  - X-ray lasers
- Attoseconds
- Imaging