Ultrafast Time-Resolved Spectroscopy 2015
**Exam questions with model answers**

There are 7 questions in total with the maximum of 35 points to score
The final score is calculated as (Number of scored points)/35*10

1. Make a comparison between long and short pulses in time and frequency domains. How are these two domains connected? (3 points)

Model answer:
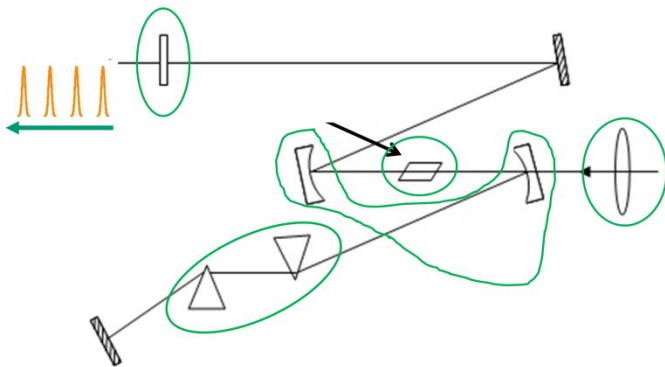1 point for each domain and 1 point for FT

The uncertainty principle says that the product of the temporal and spectral pulse widths is greater than ~1
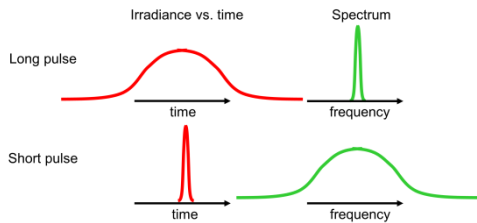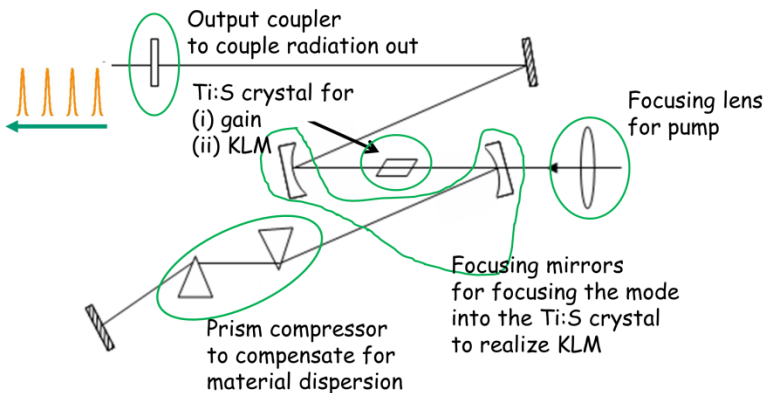

Time and frequency domain representations are linked via Fourier-transformation:

$$E(t) = \int_{-\infty}^{+\infty} \tilde{E}(\omega) e^{-i\omega t} d\omega \qquad \tilde{E}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} E(t) e^{i\omega t} dt$$

2. In picture below, schematics of a femtosecond Ti:Sapphire oscillator is shown. Name its main components (as indicated in the diagram below) and explain their functions. (5 points)

Model answer:

Output coupler to couple radiation out

Ti:S crystal for
(i) gain
(ii) KLM

Focusing lens for pump

Focusing mirrors for focusing the mode into the Ti:S crystal to realize KLM

Prism compressor to compensate for material dispersion
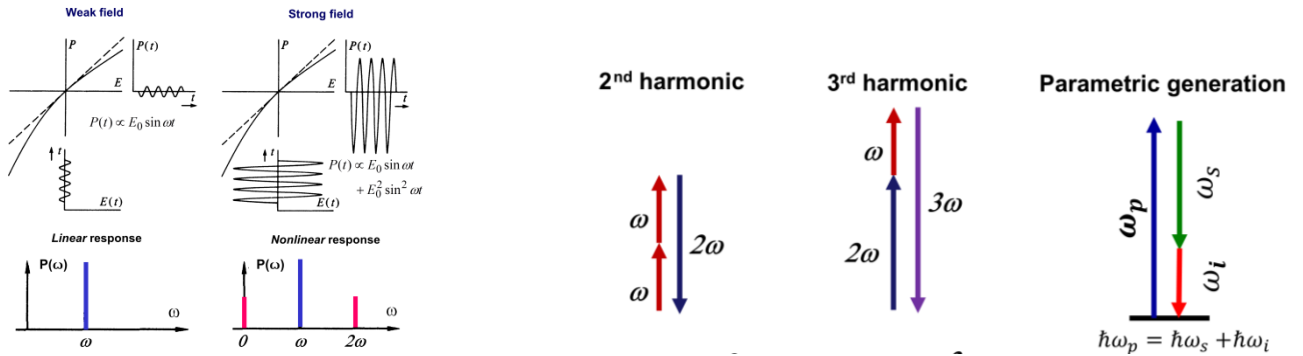
3. What is the base for nonlinear frequency conversion? Illustrate your answer by providing medium responses for linear and nonlinear approximations. Draw the energy (photon) diagrams for processes of second harmonic generation, third harmonic generation, and optical parametric generation (amplification). (5 points)

Weak field   Strong field

P $P(t)$ P $P(t)$
E E
$P(t) \propto E_0 \sin \omega t$

$E(t)$ $P(t) \propto E_0 \sin \omega t$
                              $+ E_0^2 \sin^2 \omega t$
                              $E(t)$

Linear response   Nonlinear response

$P(\omega)$ $P(\omega)$

ω $\quad \omega$ $0 \quad \omega \quad 2\omega$

2nd harmonic   3rd harmonic   Parametric generation

ω ω ω_p ω_s
ω 3ω
            2ω 2ω ω_i
ω

$\hbar\omega_p = \hbar\omega_s + \hbar\omega_i$

4. When performing time resolved measurements at synchrotrons users can 'request' specific time structures for their x-ray pulses, which fall into three general categories – natural bunch length, low-α, and slicing. What are the respective x-ray pulse durations for these three operational regimes? What are the drawbacks to these three regimes? (3 points)

5. Consider a pump-probe experiment where two beams intersect at the sample surface at an angle θ (as shown in the figure). If we consider our pulses of light to be 'flying pancakes,' one can see that we encounter a 'geometric smearing' i.e. the time resolution of our measurements may be determined by this angle. Determine the time resolution in terms of the variables given. (This is a geometry problem) (8 points)

d_{pump} Pump (800nm)

θ

Probe (800nm)
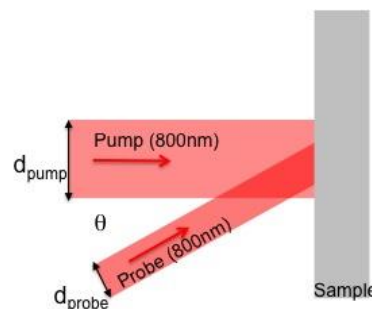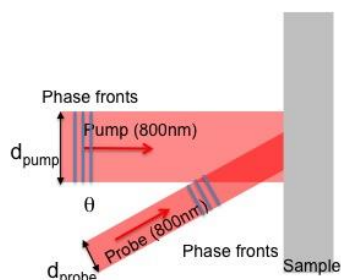
d_{probe} Sample

In considering how the 'pancakes' interact at the sample surface, on can see that the bottom side of the probe hits the sample before the upper portion. The 'geometric time smear' can be calculated by considering the time it takes to transverse the extra distance for the top of the beam. It can be calculated to be $\Delta t = \left(d_{probe} \, {}^* \tan q \right) / c$. In the example shown here, the smear is determined by the diameter of the smaller of the pump or probe, and the angle. For a typical experiment with d_{probe} =100µm and an angle of 20deg we get a smear of 120fs. This is significant if we want 20fs resolution.

6. (i)You would like to measure thermal diffusion along the surface of an opaque 2D material (e.g. a thin film or bulk graphite). Construct a measurement scheme to do this.
(ii) Now you would like to measure thermal diffusion perpendicular to the surface of an opaque 2D materials (e.g. a thin film or bulk graphite). Detail an experimental geometry that could accomplish this task.
(6 points)

Model answer:
(i) This is a classical example of using transient grating spectroscopy. We measure the diffraction signal as a function of different excitation grating periodicities, which we plot to extract the diffusion constant. (3 points)
(ii) We could continue to employ the TG method, but it is not necessary. In reality, one needs to consider experiments that pump from one side, and probe the optical reflectivity from the other side. If one does this as a function of film thickness, then we could extract perpendicular diffusion.
If we deal with a bulk sample like graphite, we could also perform a standard TG measurement perpendicular to the layer direction. (3 points)

7. What is the time-dependent Stokes shift? What are the typical time scales of this shift? How can it be used to study properties of the molecule and/or solvent? Provide an illustration for the time-dependent Stokes shift. (6 points)

Model answer:
Time-dependent Stokes shift is the red shift of the fluorescence in time. (1 point)
It is caused by two processes: (i) intramolecular vibrational relaxation (i.e. the properties of the molecule can by studied) (1 point) and (ii) reorganization of the surrounding (i.e. the surrounding properties – e.g. solvent -- can be studied) (1 point). The typical timescales are from ps to 100's ps. (1 point)
Illustrations 1 point each.