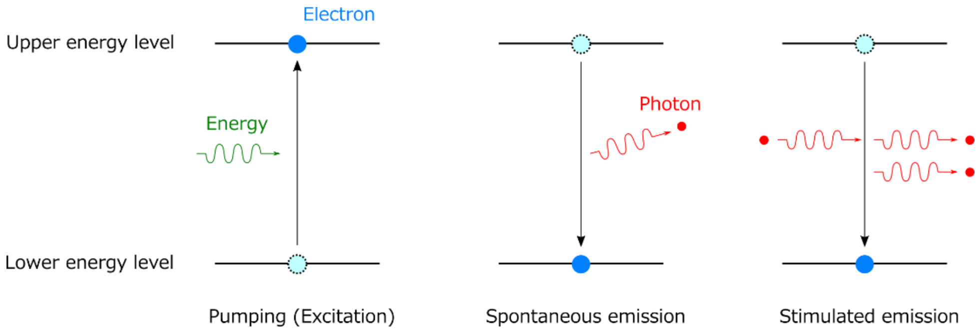
Exercise 10.1 : Lasers

1) Discuss the operational principle of a laser: Sketch the energy states involved and explain how optical gain can be achieved.
2) What is the main requirement for an efficient semiconductor laser?

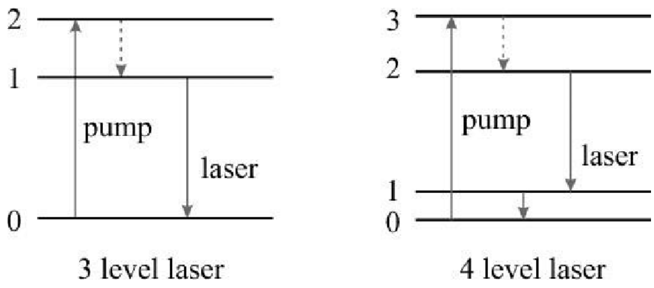
Exercise 10.1 solution.

1) See slides 29-32 of lecture 5.1-5.2.

   In a laser we use stimulated emission to amplify light. An excited atom or molecule decays to a lower energy state when stimulated by an incoming photon, emitting a second photon in the process. This emitted photon has the same energy, phase, and direction as the incident photon, making the light coherent.
   Stimulated emission requires that the excited energy states are always populated. To achieve this, the rate of excitation must exceed the rate of decay. However, in a two-level system, this is fundamentally impossible because absorption and stimulated emission occur at the same rate, preventing a population inversion.
   Therefore, a three-level system is used, where atoms are pumped from the ground state (level 0) to a high-energy state (level 2), which rapidly decays non-radiatively to an intermediate state (level 1). If level 1 has a long lifetime, a population inversion between levels 1 and 0 can be achieved.



   Four-level systems are even more efficient. In this scheme atoms are excited to level 3, decay quickly to level 2, emit a photon by transitioning to level 1, and then rapidly decay to the ground state 0. Because level 1 is ~always empty, there is no reabsorption, which makes it more effective.



The lasing medium is placed in an optical cavity, where the light reflects back and forth, passing through the medium multiple times and resulting in exponential amplification. The geometry of the cavity supports only certain discrete resonant frequencies. Among these, only those that coincide with frequencies for which the gain exceeds the losses (i.e., gain > 1) will "lase".

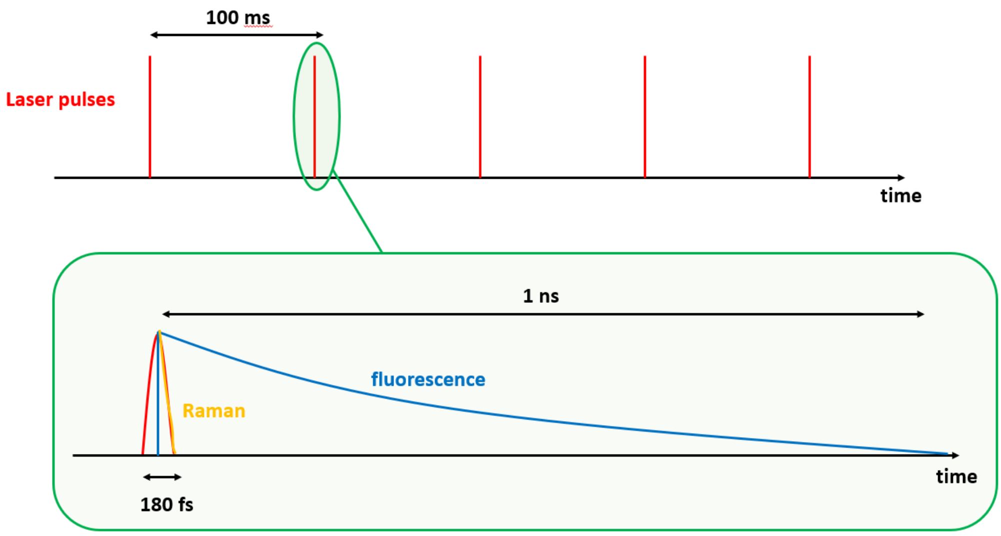
2) Semiconductor (SC) lasers work with the same principle, except we use the optical transitions between the valence and conduction bands. A current is injected to populate the conduction band. <u>The main requirement for the SC is to have a direct bandgap.</u>
In the process of stimulated emission, for a direct bandgap material the incident photon interacts only with an electron and a hole. While for an indirect bandgap material there must be interaction with a phonon, which makes the process less probable. Besides, indirect bandgap creates a strong temperature dependence.

Exercise 10.2: Photoluminescence lifetimes

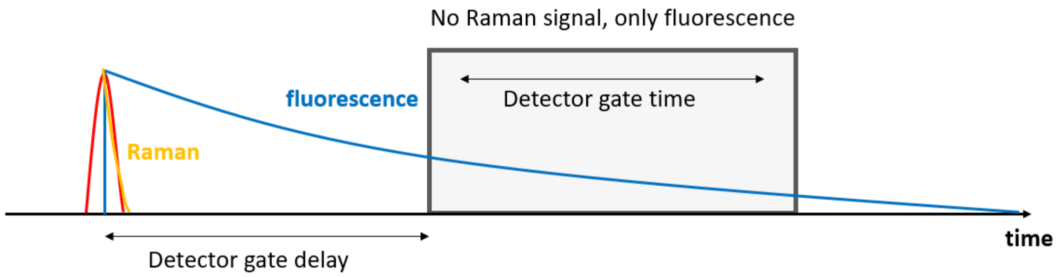
A pulsed laser with 180 femtosecond pulses with a repetition rate of 10 Hz is used to excite a semiconductor. The sample's photoluminescence decays over 1 ns. The sample also exhibits some Raman scattering.

On a time axis, sketch the laser pulses, the Raman signal and the photoluminescence (PL) of the sample. Knowing that the Raman signal and the PL occur in the same spectral region, what strategy could you use to separate the Raman signal and the PL?

Exercise 10.2 solution.



Raman signal is instantaneous, while fluorescence has a long decay time. We can eliminate all the Raman signal if we probe the signal later in time:



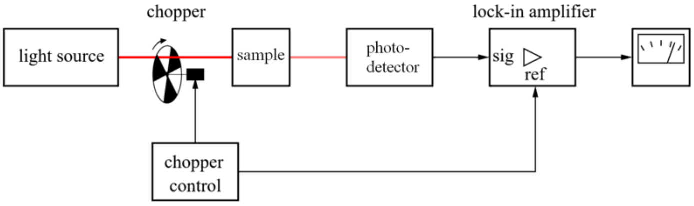
In practice, we use detectors that can be gated in time. The detector operates at the same frequency as the laser, with a gate time and a gate delay that can be chosen. However, note that with modern electronics, the smallest possible gate time is typically nanoseconds (state-of-the-art: tens of picoseconds).

Exercise 10.3: Absorption of excited states

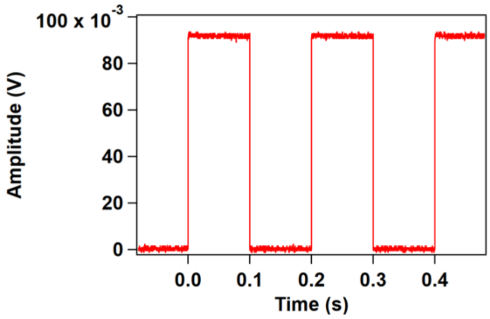
1) Which state is probed by absorption spectroscopy? Which states are probed by transient absorption spectroscopy?

2) Explain why it is not straightforward to measure the absorption of a material's excited state.

3) Assuming the following lock-in experiment:



The reference signal coming from the chopper is represented below:



What is the frequency of the signal that will be amplified in this lock-in experiment?

Exercise 10.3 solution.

1) Absorption spectroscopy probes the fundamental state population by monitoring the transition between the ground state and the excited state(s). Transient absorption spectroscopy probes the excited state(s) population(s) by monitoring the transition between an excited state and other excited states. Note that in addition to excited states, transient absorption spectroscopy can probe the ground state because of the bleaching process, which corresponds to the ground state being depopulated. Additionally, stimulated emission or fluorescence directly probe the excited state population by monitoring the transition between the excited state back to the ground state.

2) It is not straightforward to probe excited states because one needs to generate a sufficient amount of excited state population that can be detected by a specific technique. If the rate of decay of the excited state population is faster than the

excitation rate, one will not generate a sufficient amount of excited state population. In order to increase the amount of excited state population, one needs to have an excitation rate larger than the decay rate, and this is why laser sources are generally used to photoexcite samples.

3) Sometimes a signal of interest is extremely small, such that the signal cannot be easily distinguished from noise, for example. If the signal of interest is periodic with a certain frequency (different from the typical noise frequency), then we can isolate the signal by selectively amplifying this frequency.

   Therefore, in this experiment, we modulate the excitation (and thus the final signal of interest) at a frequency determined by the chopper's rotation speed.

   The chopper having a well-known and well-controlled frequency, one can selectively amplify this frequency to extract the signal out from the noise with a lock-in. Here, the frequency of the signal is $1/T = \frac{1}{0.2} = 5\ \mathrm{Hz}$.