Fathoming ice

Using non-linear ultrafast spectroscopy to look at interfacial properties

Sudera, P.

Publication date
2021

Chapter 3

Generation of ultrashort high-intensity pulses

Vibrational dynamics influence molecular reactions and occur on a timescale of a few femtoseconds to picoseconds. To resolve such ultrafast dynamics, a probing methodology of a similar time resolution, if not shorter, is required. It took only two decades after the laser was invented for the duration of light pulses to shrink down by six orders of magnitude, from nanosecond to the femtosecond regime. So far, the shortest light pulse being generated is a mere 80 attoseconds. To put things into perspective, light only travels 300 nanometers in 1 femtosecond.

Laser basics

Stimulated emission

Einstein postulated that three different processes can take place when light interacts with matter (figure 3.1): Absorption – a photon with a radiation field transfers its energy to an electron, which transfers its state from E_m to E_n, with the probability B_{mn}; Spontaneous emission – From the excited state E_n, an electron with a probability A_{nm}, spontaneously falls to the lower state E_m, with simultaneous emission of a photon with energy $h\nu = E_n - E_m$, with a random direction, phase and polarization; and Stimulated emission – wherein a photon with energy $h\nu$ passes by an excited atom causing a stimulated emission of a twin photon, with the same energy, phase, polarization state, and direction of propagation, with the probability B_{mn}.

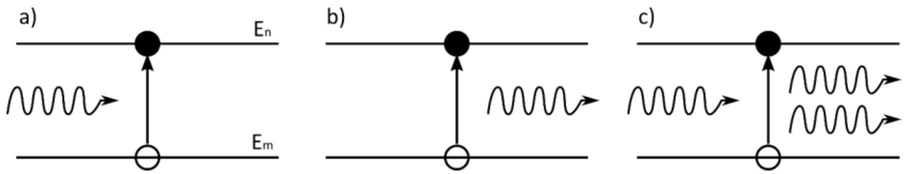

Figure 3.1: elementary electron-photon interaction processes in atoms: a) absorption, b) spontaneous emission, c) stimulated emission

effects like spatial and temporal focussing, there is a change in the carrier frequency on both the leading and trailing edges of the pulse, leading to broadening of the spectrum.

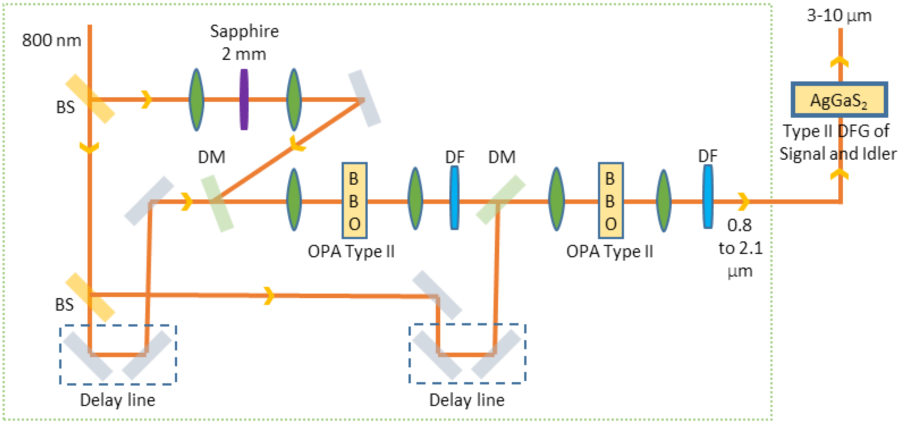

Figure 3.8: Working principle of a near-IR OPA setup to generate near-IR wavelengths in dashed green box; DF: dichroic filter, DM: dichroic mirror, BS: beam splitter. Figure adapted[40]. Further DFG process to produce mid-IR wavelength beams.

Following the seed generation, the pump and the seed pulses undergo amplification in two stages, via spatial and temporal overlapping in a non-linear crystal, by the non-linear parametric amplification process, where the pump beam (ω_3), amplifies the lower frequency signal beam (ω_2), also additionally producing a lower frequency idler beam (ω_1). The first amplification stage is used to compensate for the group velocity mismatches between the pump and signal pulses using a delay line. The second amplification stage, called the power stage, causes significant pump depletion with a conversion efficiency above 30%. The second stage is able to produce much more stable pulses as the seed fluctuations are minor after the first amplification. After the second amplification process, the signal and idler beams are separated from the pump beam using dichroic mirrors or filters.

The signal and idler produced in the near-IR region are subsequently separated using dichroic mirrors to induce time-delay changes, and go through a Type II DFG process in an $AgGaS_2$ crystal to produce pulses in the mid-IR frequency range. The whole process is depicted in Figure 3.8.