
The research work of Martin Mičica is centered on optically pumped molecular lasers/amplifiers for THz emission with the approach of using a mid-infrared laser – a quantum cascade laser – with a tunable spectral emission to pump a system for THz gain. This is studied initially in the gas phase where THz amplification is realized in existing optically pumped molecular laser (OPML) to show the large gain of these systems. This is followed by the characterization of the OPML in a new geometry where the reflectivity of the output couplers can be varied, as well as laser action of a multitude of spectral lines by simply tuning the MIR pump to other molecular transitions within the gas system. This is an important highlight as this could not be achieved previously when using CO$_2$ pump lasers in OPMLs. A second more perspective works looks at the very interesting and novel approach of using solid state materials as the material with THz gain. The work in the thesis is a recent and important subject, only first realized in 2016, showing the unique features of OPML and will influence the uptake of THz laser technology. The thesis itself is organized in three distinct chapters that are concise, well planned and has the necessary information to understand the main concepts of the research. The chapters are complemented by an introduction and conclusion. The work is very good, with a complete study that permits to understand OPMLs and the scientific advances made during the thesis.

Following the introduction, which briefly presents the state-of-the-art and the thesis organisation, the second chapter describes current THz sources, working towards presenting OPMLs. Molecular transitions are presented within the anharmonic potential, and for symmetric and asymmetric molecules with increasing complexity in determining the rotational energies. THz spectroscopic techniques are presented with the applications of THz time domain spectroscopy applied to determine the refractive index of materials. The importance of taking into account the Gouy phase shift is then presented. This is important especially when measurements are taken between the sample and reference, where different positioning can result in incorrect index determination. Correction of this phase shift in presented in clear steps and shows that reliable data can be achieved.

Chapter 3 enters into the heart of the thesis where the gain of an OPML is investigated. These I believe are the first such investigations with a MIR QCL as pump, with high gains for OPMLs shown and highlights the strong potential of these OPMLs. NH$_3$ is initially studied in a new geometry for the study of the gain, where Brewster angled windows are used to inhibit any feedback and hence laser action. This involves a considerable experimental development where a THz ‘probe’ is used with the MIR pump. Two pumping transitions in particular were targeted and probed around 1THz. Further details of other transitions with gain would have been useful as well as mentioning of the dynamics associated with the laser states involved in the transitions and the effect of the pressure on the interactions. The gain bandwidth is shown and related to Doppler broadening of the transition. The effect of pressure is also shown with an optimum pressure around 25µbar. High gain values, the highest reported for OPML, are demonstrated. (A comparison with other THz lasers would have been welcomed). A similar study is performed on D$_2$O but at lower frequencies, permitting a VNA to be used, accessing the full set of S-parameters. This interestingly showed the shift of the gain depending on the S-parameter used owing to the Doppler effect.

The last section on Chapter 3 then presents the OPML and shows the emission at a range of THz frequencies that would not been possible with a CO$_2$ pump laser owing to its lack of fine tuning. Here the
new laser system is shown where the waveguide is placed in a vacuum system with the output coupler based on a FP interferometer to tune the output reflectivity (that would benefit from being shown as a function of separation distance). The intensity distributions of the different modes were visualised using a THz camera and varying the resonator length showing likely the presence of the low order modes of the cavity. The laser lines that were detected were then shown (but no spectra) showing a wide range of frequencies that could be accessed, highlighting the nice frequency flexibility of the system. At the end of this chapter, an interesting study is performed in mixing gases and permitting to play with the dynamics of the system. Although no power improvements were observed, there is obviously an effect of different gas mixtures and opens up an interesting avenue to investigate in further studies.

The last experimental chapter describes a new approach in taking the concepts of gas lasers and applying them to the solid state. This I believe is an exciting research direction which could potentially lead to compact systems where the complete laser properties can be engineered from the tunability, the spectral regions targeted, the relaxation dynamics to saturation. Here the idea of using molecules or groups of atoms in a crystal structure is presented. However, there is a considerably amount of material research that needs to be done first, which the thesis tackles. The first randomly orientated crystals were prepared and characterised spectrally in the MIR by ATR measurements and THz by pellet preparation with THz TDS. A variety of samples are tested, each showing complex absorption features in the MIR (~5µm to 20µm), with I believe a target the type of spectra equivalent to the gas phase measurements in Chapter 3. Single crystals of L-tartaric were also realised and spectrally characterised in mostly the THz, showing clear absorptions with narrow linewidths. The peaks could also be classified in terms of their vibrational modes. MIR spectral features were also seen with different absorption properties depending on the orientation. Although a ‘road-map’ would have been useful in determining what is exactly needed in solid state OPML (linewidths, absorptions, losses, interactions between molecules etc.) and its comparison to the gas state, the strong material development provides a solid basis for future work of new types of THz lasers.

To conclude, I believe that this research has shown important advances in the investigations of these types of OPMLs. The gain has been characterized for the first time, and it has demonstrated high values for OPMLs. The works also showed that MIR QCL pump sources can access transitions that cannot be accessed with spectral fixed CO2 sources. This should not be underestimated as it prevents laser-pump slavery that has limited the uptake of this technology towards more compact geometries, as well as very interesting perspectives in engineering the absorption spectra using gas mixtures. The idea of using the solid state is a further exciting prospect and the work here shows the first steps in realizing molecular crystals for THz lasers. Importantly, Mr. Mičica has shown the ability to drive two very distinct parts to his research – the photonic and material development – and demonstrated research beyond the state-of-the-art. I therefore highly recommend that Mr. Mičica be permitted to make an oral defense in view of obtaining his doctorate degree.

Yours sincerely

Dr. Sukhdeep DHILLON
CNRS Research Director
dhillon@ens.fr