Coherent spectroscopy with quantum cascade lasers

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High power single mode quantum cascade lasers (QCLs) open up the possibility for rovibrational state preparation of gas phase molecules in low frequency vibrational modes. Examples of pump and probe experiments with cw QCLs with these aims will be presented.

Recently we have used two 5 μm cw QCLs to perform a counter-propagating pump and probe experiment on a low pressure sample of nitric oxide [1]. The strong pump field excites a fundamental rovibrational transition and the weaker probe field is tuned to an appropriate rotationally resolved hot band transition. When both light fields are in resonance, rapid passage is observed in the hot band absorption lineshape arising from a minimally damped and velocity-selected sample of molecules in the v = 1 state.

In these first experiments a commercial external cavity QCL (140 mW power, linewidth = 2 MHz Daylight Solutions) was used as the pump laser while the probe laser was a distributed feedback QCL (7 mW, linewidth = 16 MHz, Alpes Lasers). Most recently, the weak probe has been replaced with a higher powered (up to 190 mW) narrow bandwidth (800 kHz) device. Figure 1 adjacent shows example data obtained with 35 mTorr of NO in a 70 cm long cell in which the $R(14.5)_{1/2}$ v = 1←0 and $R(15.5)_{1/2}$ v = 2←1 transitions are used as the pump and probe transitions respectively. In these experiments, the pump and probe powers were 80 mW and 41 mW respectively and their chirp rates were 150 Hz ns^{-1} and 145 KHz ns^{-1}. The data clearly shows the effect of rapid passage as the probe interrogates a velocity selected vibrationally excited sample.

The rapid passage structure is well fit by a (exponentially) damped sinusoidal function and the fitted decay constants, τ, are a function of the total sample pressure – see figure 2. As expected the rapid passage signal decays more rapidly with increasing pressure of NO as the increased collision frequency leads to more rapid dephasing and relaxation of the induced polarisation. It is also noted that the decay constants are subtly different for each Λ doublet of the pumped rotational state indicating that the underlying hyperfine structure of the transition plays a role in determining the dephasing of the observed signals.

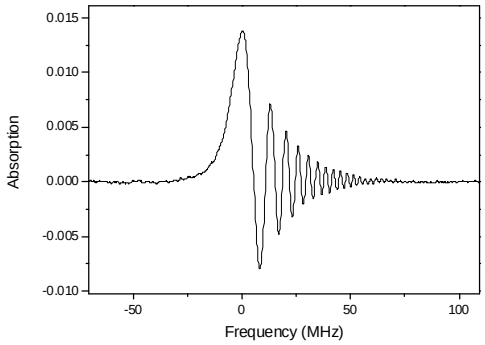

Fig. 1: *An example of the velocity selected hot band transition lineshape observed when pumping NO on a fundamental ro-vibrational transition.*

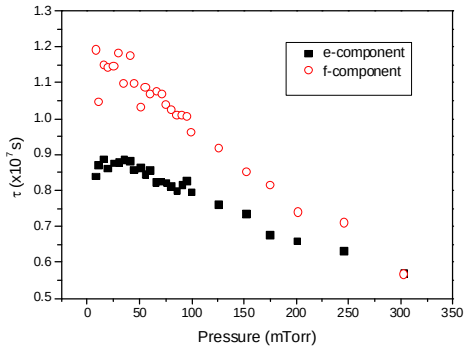

Fig. 2: *Measured decay constants for each of the velocity selected Λ doublets prepared in v = 1.*

References

[1] R.J. Walker, J.H. van Helden, J. Kirkbride, E.A. McCormack, M.T. Bell, D. Weidmann, G.A.D. Ritchie – *Optics Letters* 36 (2011) 4725.