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## Modelisation of a gas phase polarization induced by a 200 GHz chirped pulse

Robin Bocquet, Daniele Fontanari, C. Bray, A. Cuisset, Guillaume Dhont, K.  
Hickson, Gaël Mouret, Francis Hindle

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and find the solutions by successive approximations:

$$\begin{aligned} z^{(n+1)}(t) &= -i \int_0^t e^{-i(\omega_0 + \tau)(t-x)} \varepsilon(x) W^{(n)}(x) dx \\ W^{(n+1)}(t) &= \frac{1}{2i} \int_0^t e^{-\tau(t-x)} (\varepsilon^*(x) z^{(n)}(x) - \varepsilon(x) z^{(n)*}(x)) dx \end{aligned} \quad (5)$$

Taking the initial conditions ( $z(0)=0$  and  $W(0)=W_{eq}$ ), only the odd terms of the  $z$ -series and the even terms of the  $W$ -series remain. We will work with  $z^{(1)}(t)$  and  $W^{(0)}(t)=W_{eq}$  and do a changing variable to simplify the equations (rotating frame):

$$\zeta^{(1)}(t) = z^{(1)}(t) e^{i[(\omega_0 - c \Delta\omega)t + \frac{\Delta\omega}{2T_c} t^2]} \quad (6)$$

Then we can show that for  $t=T_c$ :

$$\zeta^{(1)}(T_c) \approx \rho_{app} e^{i\theta_{app}} + \tilde{\zeta} \quad (7)$$

with

$$\begin{aligned} \rho_{app} &= \frac{\sqrt{2\pi} \Omega_0 W_{eq} \sqrt{T_c} \exp[-\frac{T_c}{T_2}(1-c)]}{\sqrt{\Delta\omega}} \\ \theta_{app} &= (1-c)^2 \frac{\Delta\omega}{2} T_c - \frac{3}{4}\pi \\ \tilde{\zeta} &= \frac{W_{eq} \Omega_0}{\Delta\omega} \left( \frac{1}{1-c} + \frac{1}{c} \exp[i \frac{\Delta\omega}{2} T_c (1-2c)] \right) \exp[-\frac{T_c}{T_2}(1-c)] \end{aligned} \quad (8)$$

$\rho_{app}$  (which is positive) represents the amplitude of  $\zeta^{(1)}(T_c)$ ,  $\theta_{app}$  its phase and  $\tilde{\zeta}$  induces a small oscillation. In particular  $\rho_{app}$ , tends to 0 when  $T_c$  tends to 0, increases until it reaches a maximum when  $T_c = \frac{T_2}{2(1-c)}$  with

value  $\frac{\sqrt{2\pi}}{e} \frac{\sqrt{T_c}}{\Delta\omega} W_{eq} \Omega_0$  and then it decreases toward 0 as

$T_c \rightarrow +\infty$ . The approximation which retains  $\rho_{app}$  is already very good to describe the system behavior.

**Free Induction Decay (FID)** – In this phase, the Rabi frequency vanishes and the optical Bloch equations simplify [3]. Using an heterodyne detection scheme, we can demonstrate that the output signal is :

$$S(t') = B \sqrt{\frac{T_c}{\Delta\omega}} e^{-\frac{T_c}{T_2}(1-c)} \left[ e^{\frac{-t'}{T_2} - \frac{t'^2 \Delta\omega_0^2}{4}} \cos(\omega_{IF} t' + \Phi) \right] \quad (9)$$

where  $t' = t - T_c$  and  $B$  is a constant depending of the experiment (amplifiers, mixers ...). Applying a FFT, we can access the signal amplitude at the intermediate frequency:

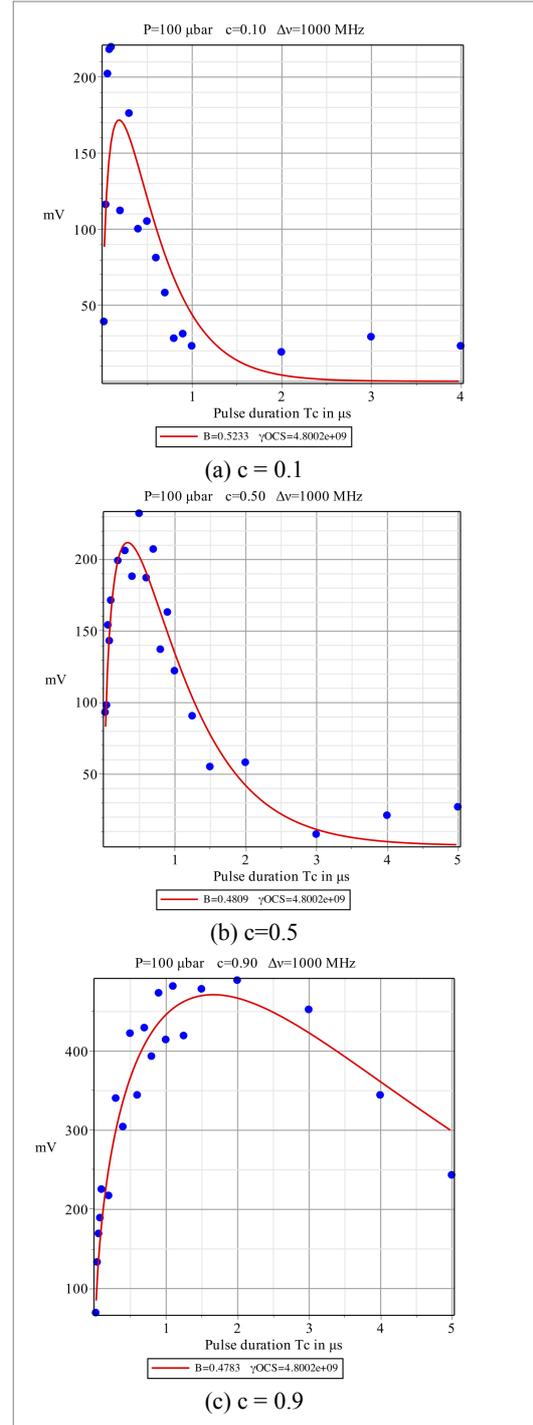
$$A_{TF}(\nu_{IF}) = \frac{B}{\sqrt{\Delta\omega}} \sqrt{T_c} \exp\left[-\frac{T_c}{T_2}(1-c)\right] \quad (10)$$

### Experimental results

We compared the theoretical model we obtained with experimental data recorded with carbonyl sulfide gas (OCS). We chose the 17->16 rotational line centered at 206.745 GHz. The output signal amplitude at the IF have been measured for different gas pressure and different positions of the molecular resonance in the CP described by the  $c$  parameter. The figure 2 compares experimental measurements and analytical modelisations for 3 different  $c$  values. A very good agreement is obtained between experimental data (blue points) and model (red curve) for a 1 GHz extension chirp and for a 100  $\mu$ bar gas pressure (collisional width larger than Doppler width).

### Conclusion

We used a mathematical method by successive approximations to solve the optical Bloch equations and to get an analytical formula giving the output signal of a chirped pulse experiment. This will be useful for absolute intensity lines measurements and to optimize the pulse duration to obtain the best signal to noise ratio.



**Fig. 2.** Output signal amplitude  $A_{TF}(\nu_{IF})$ , function of pulse duration  $T_c$ , recorded for OCS at 206.745 GHz with line resonance position  $c$  as parameter. Comparison of experimental data (blue points) and model (red curve) from equation (10)

### References

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