„Particularities of transit time broadening for Gaussian laser beam in the weak excitation limit”

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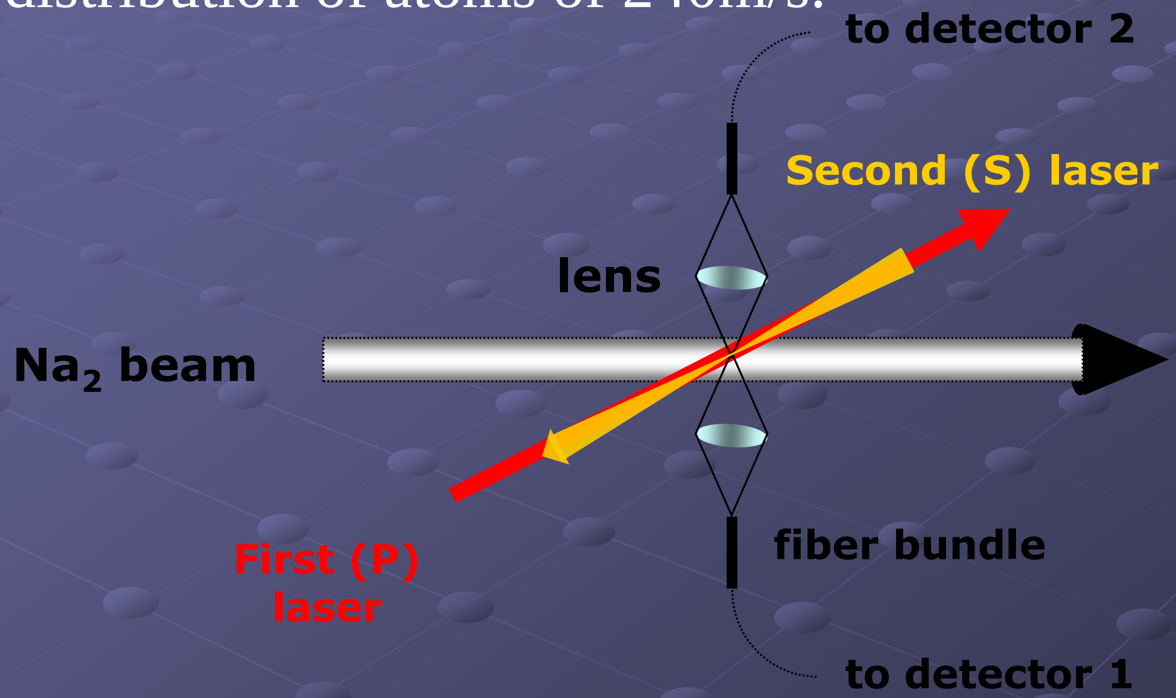
Abstract

We revisit transit time broadening for one of the typical experiment designs in molecular spectroscopy- that of a collimated supersonic beam of particles crossing a focused Gaussian laser beam.In particular, we consider a Doppler-free arrangement of a collimated supersonic beam of Na2 molecules crossing two counter-propagating laser beams and exciting a two-photon transition in a three-level ladder scheme. We propose an analytical two-level model with virtual intermediate level to show that the excitation lineshape is described by a Voigt profile and provide the validity range of this model with respect to significant experimental parameters. The model also shows that line broadening due to the curvature of laser field wavefronts on the particle beam path is exactly compensated by increased transit time of particles further away from the beam axis, such that the broadening is determined solely by the size of laser beam waist. The analytical model is validated by comparing it with experimental results and with numerical simulations of density matrix equations of motion using split propagation technique.

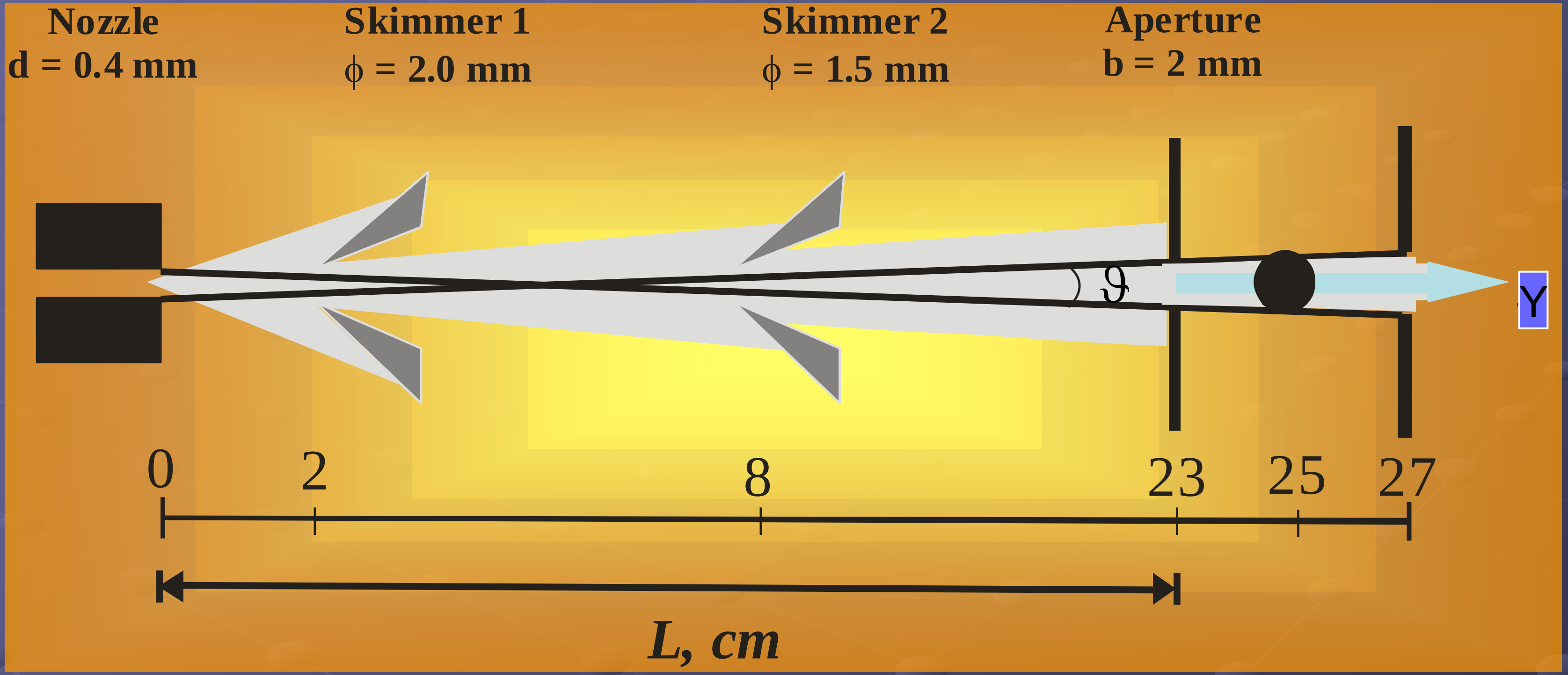
Experimental Set Up

The experiment uses a single collimated supersonic **Na2** beam with flow velocity of v_f = 1340m/s and longitudinal 1/e-widths of the velocity distribution of atoms of 240m/s.



Na2 supersonic beam: 2x10cm-3 molecules; collimated to 0.73^0 , Doppler widths 25MHz (P laser), 27Mhz (S-laser);
Lasers: cw, lineary polarized, laser linewidth 1 MHz

A Beam Formation



Collimation of the Na2 beam through skimmers and aperture

Gaussian laser beam

$$I_{laz}(z) = I_0^{(laz)} \exp\left(-2y^2 / \omega^2\right)$$

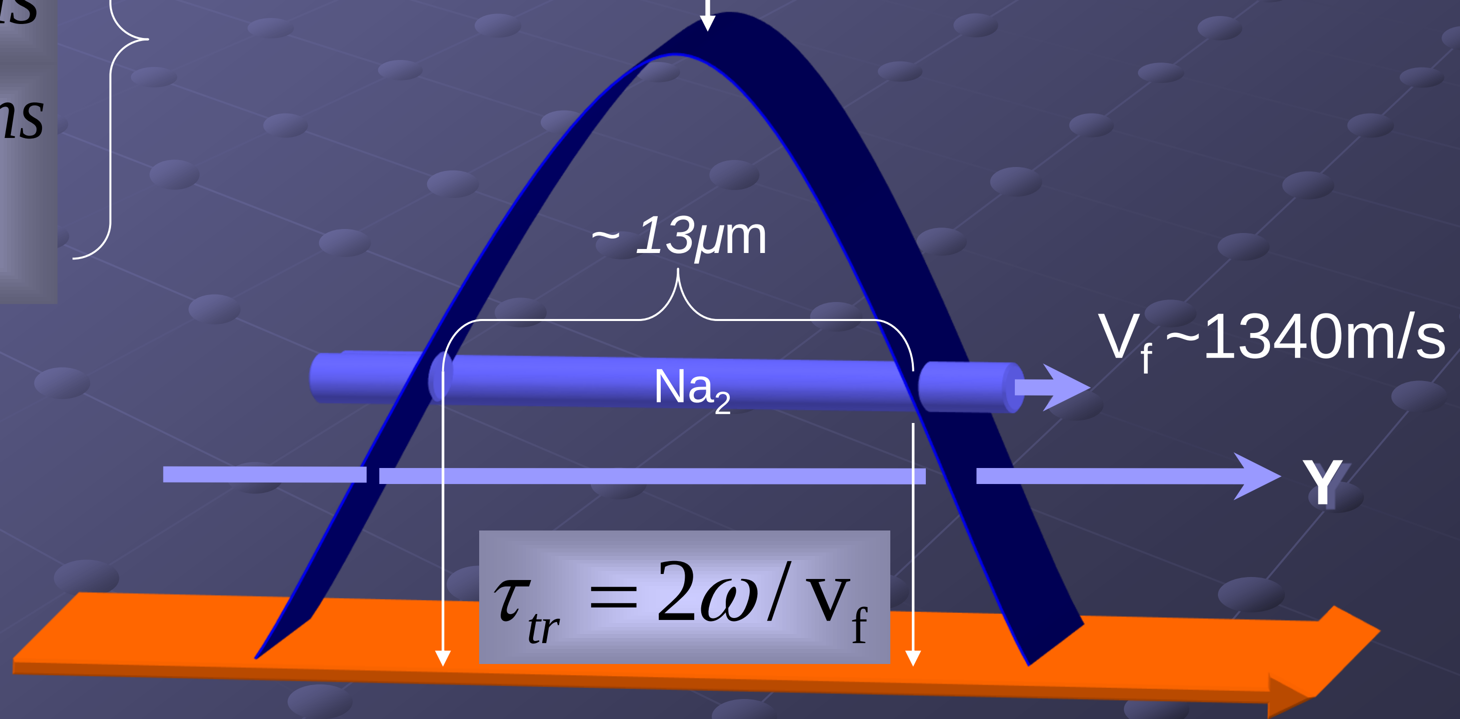
$$\tau_{tr}^{P} = 19ns$$

$$\tau_{tr}^{S} = 63ns$$

$$\tau_e = 12.45ns$$

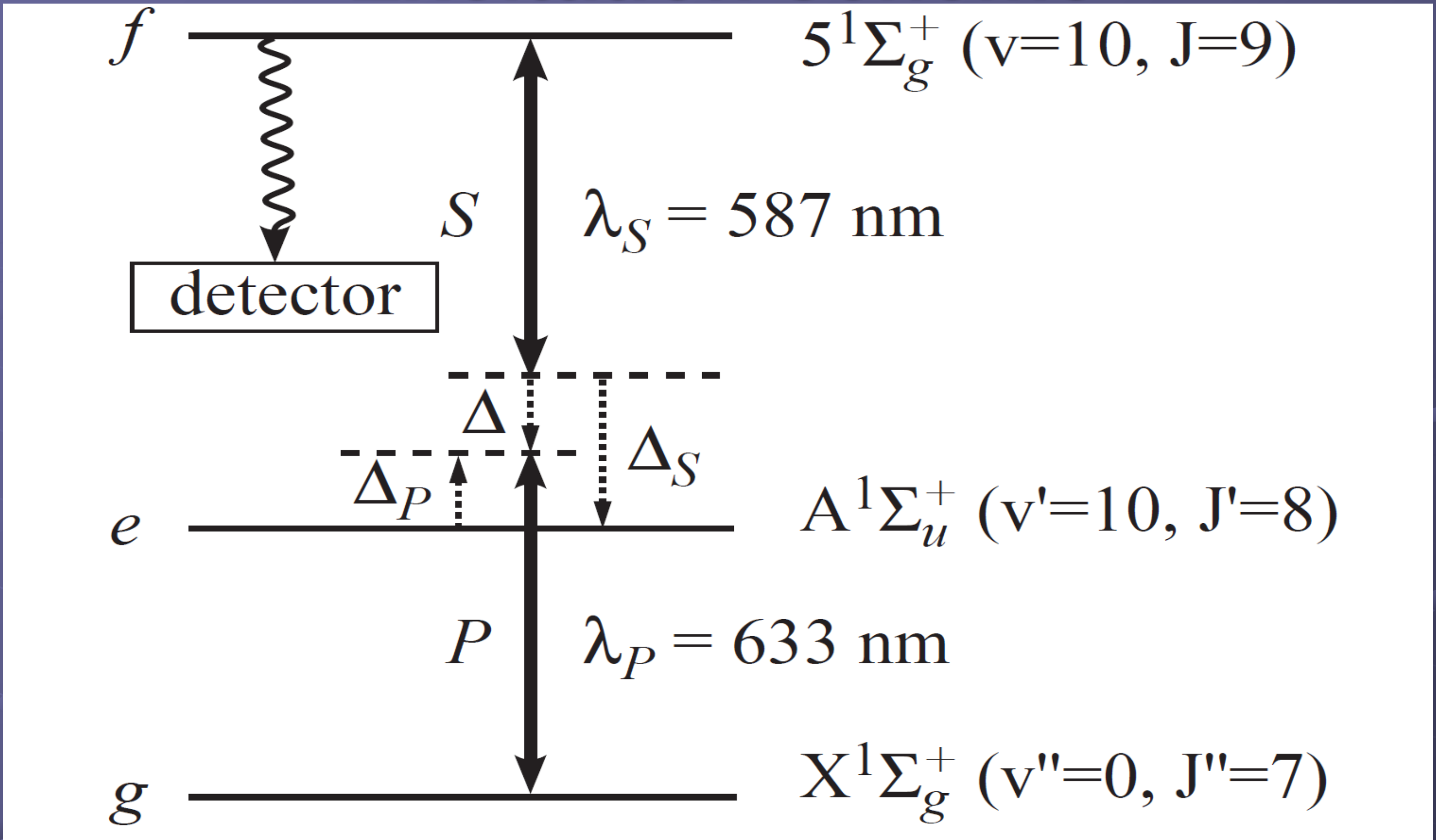
$$\tau_f = 35ns$$

Interaction time!



$$\tau_{tr} = 2\omega / \mathrm{v_f}$$

Excitation scheme



Three-level ladder scheme in Na2. The S laser field with λ_S = 587nm couples levels e and f and is detuned off from the one-photon resonance by ΔS. Levels g and e are coupled by the P laser field with ΔP = 633nm, which is scanned across the two-photon resonance at ΔP = −ΔS. The measured total fluorescence from level f as a function of Δ= ΔP + ΔS represents the excitation spectrum of the two-photon transition.

Transit time broadening

Intuitively, the transit time broadening can be understood from the Heisenberg's uncertainty principle:

$$\Delta\tau\Delta\varepsilon \sim 1$$

Uncertainty of energy $\Delta\varepsilon$ of an excited state is affected not only by its spontaneous lifetime ($\Delta\tau \approx \Delta\tau_{sp}$), but also by the transit time ($\Delta\tau \approx \Delta\tau_{tr}$).

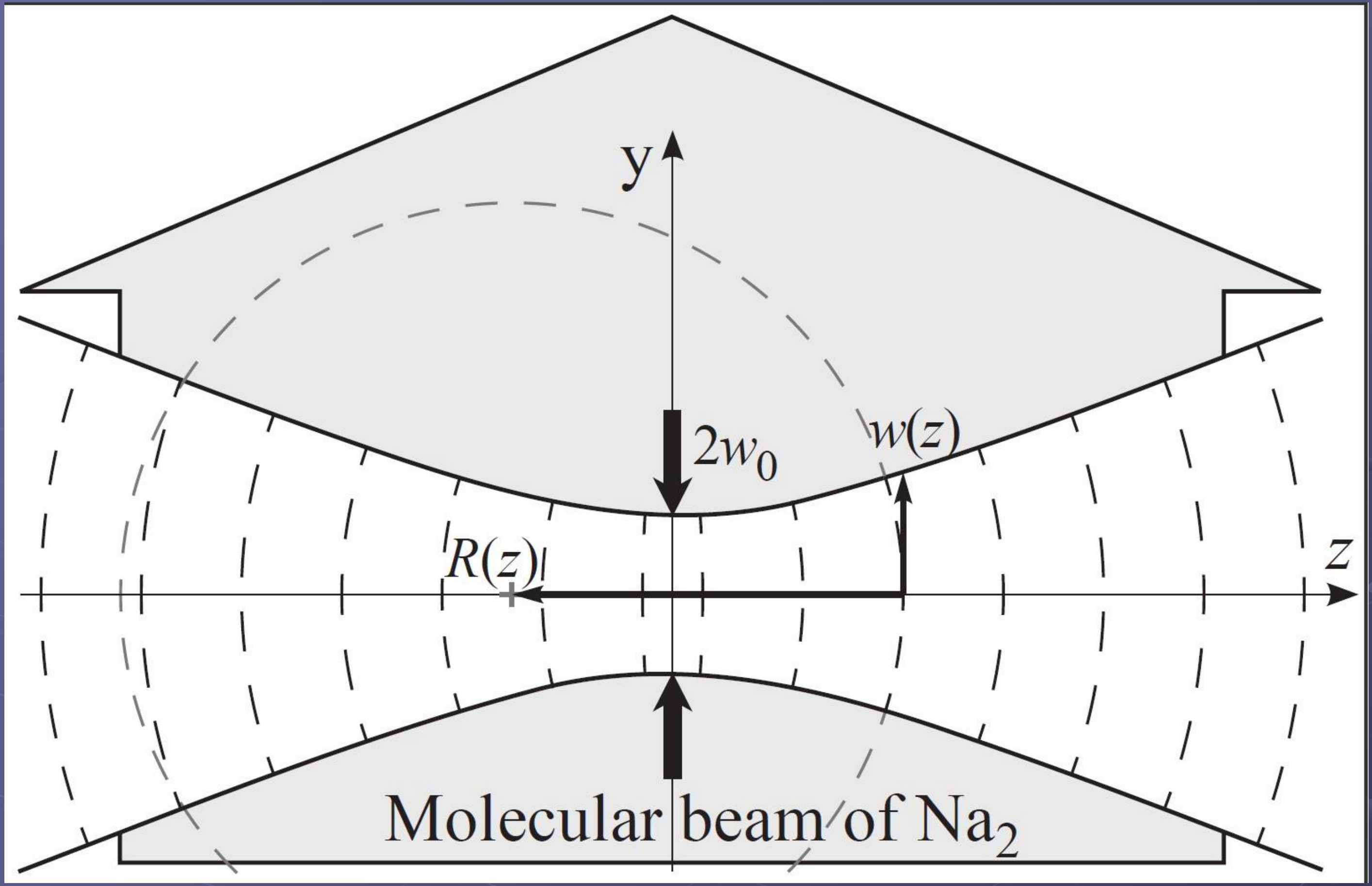
Common representation of transit time

Transit time effects usually is considered in two forms. In both cases an atom is flying through an laser beam of Gaussian intensity distribution. For atom excited in an infinitely long livingstate the spectral lineshape takes form of a Gaussian line profile. However, if the transit time τ_{tr} is comparabe to the natural lifetime $\tau_{sp} = 1/\Gamma_{sp}$ of the excited state the spectral lineshape is usually described by Lorentz profile [2].

$$P(\Delta) = \frac{\pi\tilde{\Gamma}}{\Delta^2 + \tilde{\Gamma}^2}$$

where the spectral line width (HWHM) is

$$\Delta\omega = \frac{\tilde{\Gamma}}{2} = \frac{1}{2}\left(\Gamma_{sp} + \frac{1}{\tau_{tr}}\right)$$



Laser field with Gaussian intensity distribution described by Eqs. (1, 2) propagates along the z axis. It is focused by a cylindrical lens with the focal plane at z = 0 and beam waist w0, and it crosses the molecular beam propagating along the y axis. Molecules moving at distances |z| > 0 experience longer interaction times with the laser field and hence exhibit a smaller transit time broadening compared to molecules moving at z = 0. At the same time, the molecules moving at |z| > 0 are crossing curved wavefronts of the light field, hence they experience phase shifts of the light field along their trajectories, which introduce additional line broadening [1].

Electric field amplitude

$$E = Re A(y,z) E_0 \exp\left(-i\omega_L t + i k_L z\right),$$
$$A(y,z) = \exp\left(-i\left[\varphi(z) + (k_L/2q)y^2\right]\right),$$
$$\varphi(z) = \frac{1}{2}\left(\arctan\left(\frac{\lambda_L z}{\pi w_0^2}\right) - i \ln\sqrt{1+\frac{\lambda_L z}{\pi w_0^2}}\right),$$
$$q = i\pi w_0^2/\lambda_L + z.$$

(1)

$$A(z,y) = \exp\left(-\frac{y^2}{w^2(z)}\right)\exp\left[-i\frac{k_L y^2}{2R(z)} - i\varphi(z)\right],$$
$$w^2(z) = w_0^2\left[1 + \left(\lambda_L z/\pi w_0^2\right)^2\right],$$
$$R(z) = z\left[1 + \left(\pi w_0^2/\lambda_L z\right)^2\right].$$

(2)

Raby frequency: $\Omega_z(t) = \Omega_0 A(t v_f, z),$

Analysis of a two level system

Schroedinger equation in RWA and bare states representation for an open system without cascade transitions:

$$\begin{cases} \frac{d}{dt}c_f = -i\left(\Delta - i\Gamma_f/2\right)c_f - i\Omega_z(t)/2 \cdot c_g \\ \frac{d}{dt}c_g = -i\Omega_z^*(t)/2 \cdot c_f, \end{cases}$$

$$J_z = \Gamma_f \int_{-\infty}^{\infty} dt |c_f(t)|^2,$$

Weak excitation limit: $c_g \approx 1$

The integration yields Voigt profile resulting from the convolution between Gaussian and Lorentz functions. Then Excitation spectrum P(Δ):

$$P_z(\Delta) = \frac{\tau_{tr}^2 \Gamma_f \Omega_0^2}{8} \sqrt{\frac{1+\frac{4z^2}{w_0^4 k_L^2}}{1+\frac{2z}{w_0^2 k_L}}} \int_{-\infty}^{\infty} \frac{d\omega \exp\left(\frac{-\tau_{tr}^2 \omega^2}{8}\right)}{\Gamma_f^2 + 4\left(\omega - \Delta\right)^2}, \qquad (3)$$

FWHM of the Voigt profile can be accurately approximated by the expression:

$$\Delta\omega_V = 0.5346\Gamma_f + \sqrt{0.2166\Gamma_f^2 + 22.18/\tau_{tr}^2}.$$

Effect of the intermediate level

Given a sufficiently large detuning of S and P laser fields from the respective one-photon resonances, the level e is virtual and it can be disregarded in the Schroedinger equation and the density matrix equations of motion describing the dynamics of the three-level system g − e − f. Such step is justified by the adiabatic elimination principle [3], when the amplitude C_e of the virtual level can be exactly expressed via the remaining two levels. Population of level e is negligible. Under such conditions 3 level system can be replaced by 2 level system with Ω_{eff}:

$$\Omega_{eff} \simeq \frac{\Omega_S \Omega_P}{2\Delta_P} \simeq -\frac{\Omega_S \Omega_P}{2\Delta_S}.$$

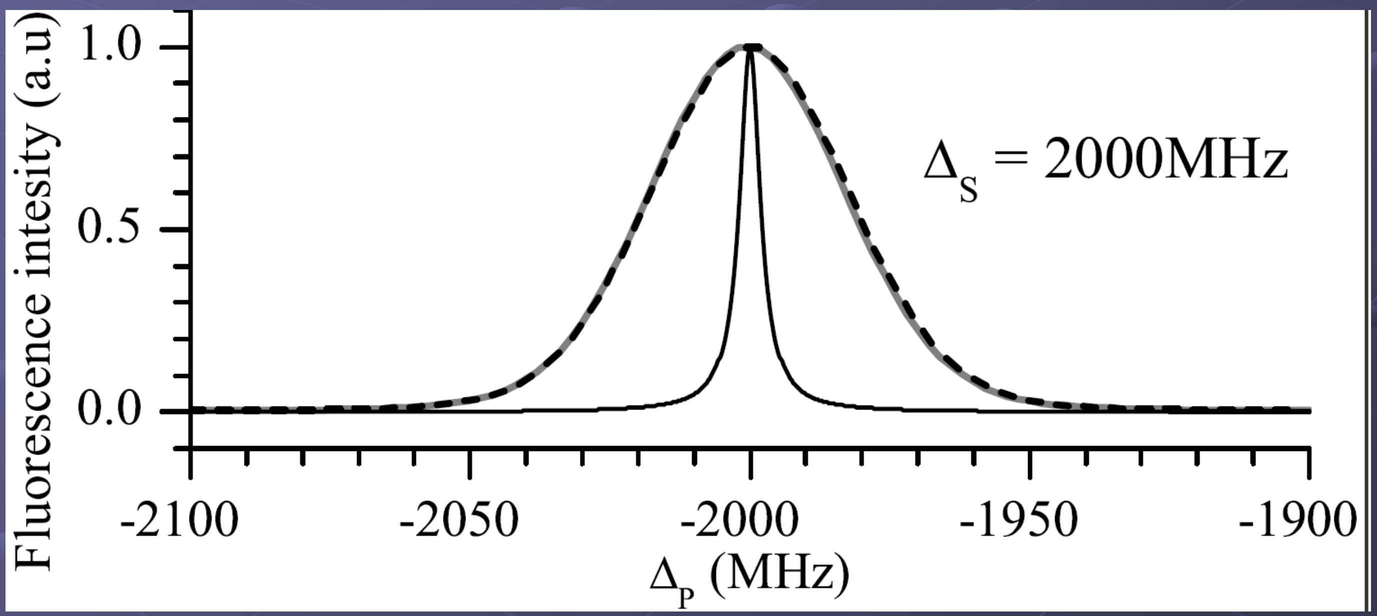
$$\Omega_z^{(eff)}(t) = \Omega_{eff} A_P(z, tv_f) A_S(z, tv_f)$$

This means that the effective Rabi frequency follows essentially the amplitude of the P laser pulse $A_P(z, tv_f)$. For our experiment this means that transit time with respect to the effective Rabi frequency corresponds to 19ns.

Numerical simulation of full sublevel system

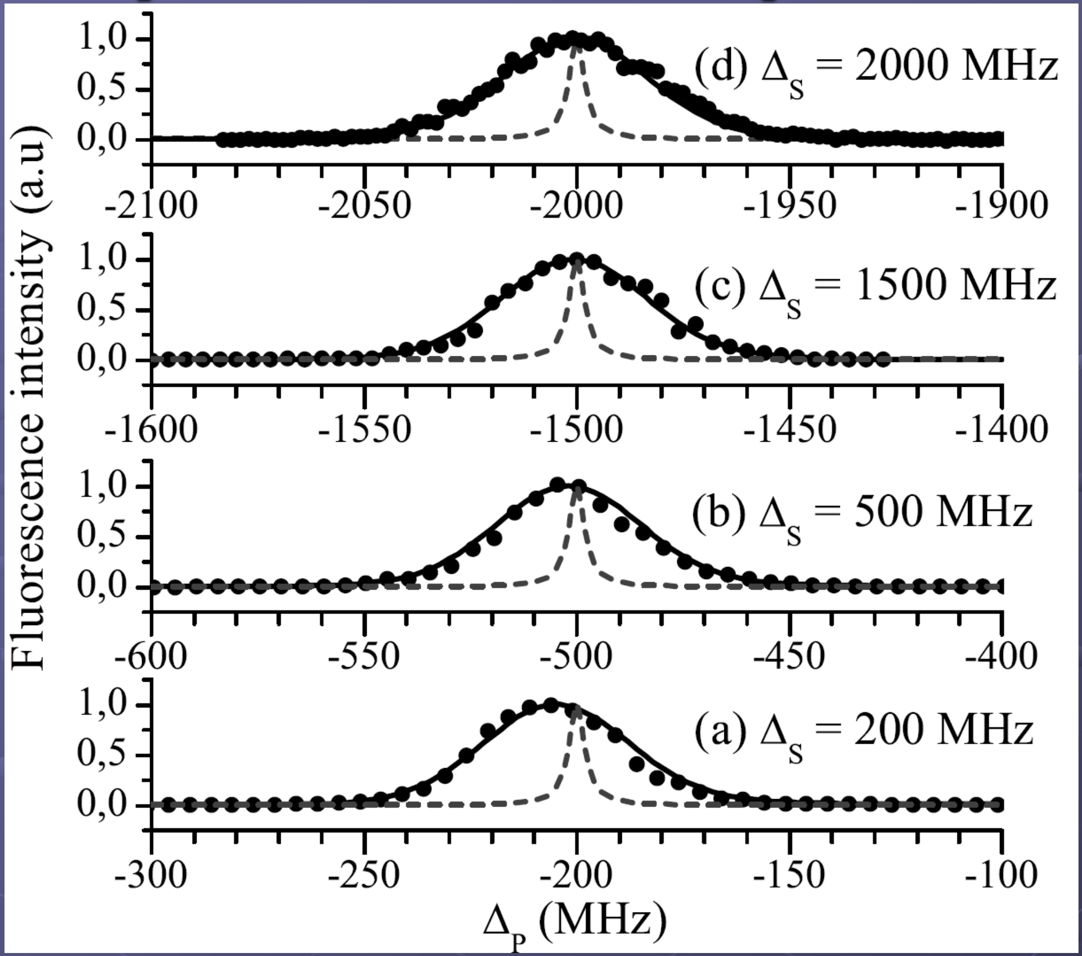
We employ the Split Propagation Technique [4,5] to solve numerically the density matrix equations. The observed fluorescence signal S_f from level f is proportional to the integrated over time total population of level f:

$$S_f \sim \sum_{m_J} \int_{-\infty}^{\infty} dt \rho^f_{m_J,m_J}(t).$$



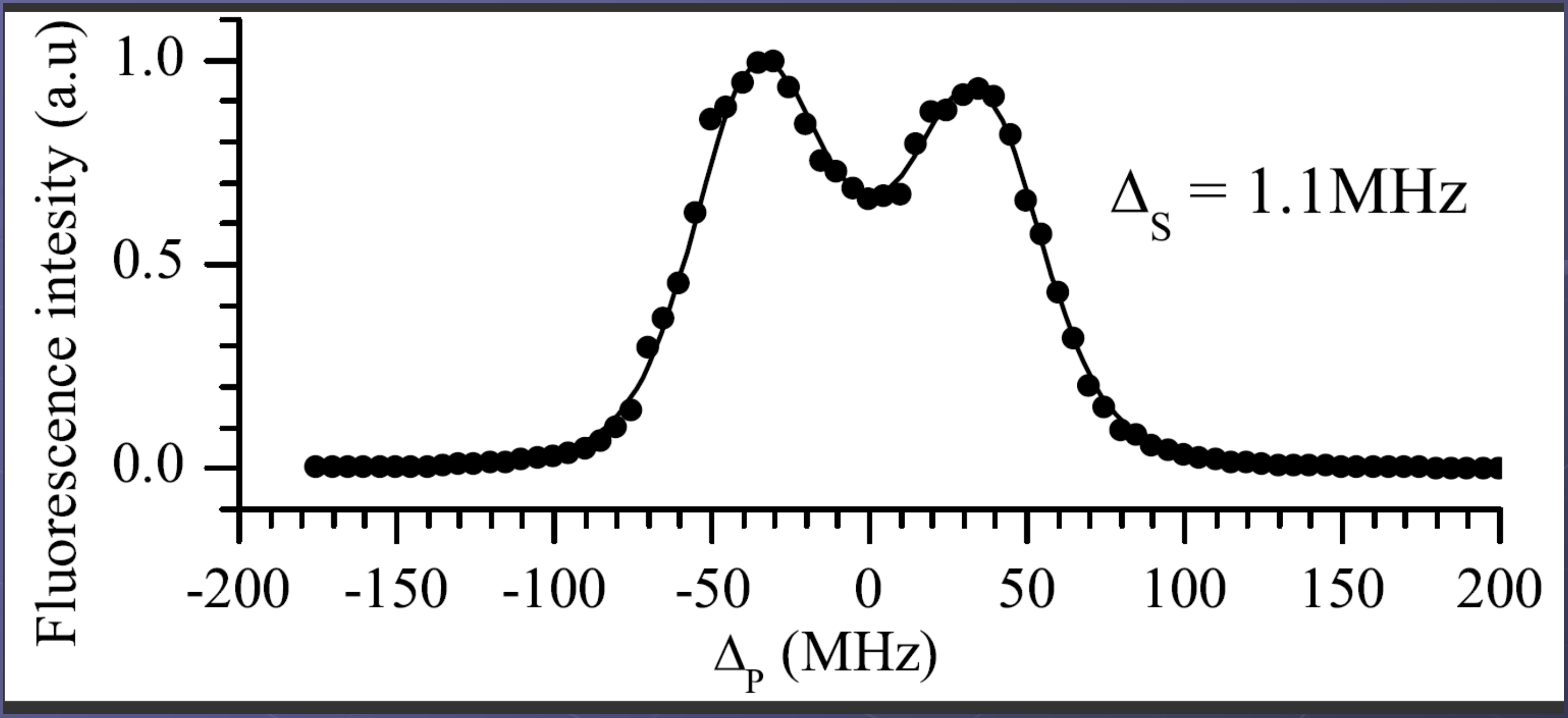
Comparison of different line broadening models for detuning Δ_S = 2000MHz: grey solid line is the result of accurate numerical simulations for the 3 level system of taking into account the full magnetic sublevel structure; dashed line corresponds to two-level Voigt profile (3); solid black line is the Lorenz profile that takes into account only the natural broadening of level f.

Comparision to experiment



Excitation spectrum of level f for different detunings ΔS of the S laser field. Note, that the P laser detuning ΔP differs from the two-photon detuning Δ = ΔP + ΔS at fixed ΔS. Dots are the experimental data points while solid lines show the results of numerical simulations For comparison, the dashed line shows the expected natural linewidth.

Comparision to experiment (rezonance)



The excitation profile for final f state in the case of small S laser detuning. Solid line exhibits numerical results while dots corresponds to experimental data.

Conclusions

We have analysed the effects of limited transit time on line broadening in the excitation spectra for a typical experimental situation of a beam of particles crossing focused Gaussian laser beams. We used a two-photon excitation in a three-level system of Na2 molecules.

- counter-propagating laser beams (to exclude Doppler broadening).
- only one parameter – the waist size of the laser beam - determines the transit time broadening.
- In the case of weak excitation limit if natural lifetime is comparable to transit time the resultant excitation line can be described by a Voigt-like profile.
- thermal velocity distribution of molecules in the beam can be disregarded for the typical supersonic beam conditions.
- We have demonstrated that for sufficiently large one photon detunings and sufficiently low effective Rabi frequencies the effects of transit time in two-photon excitation can be well described using a simple two-level model. We have also defined the range of parameters within which the two-level model can be applied.

References

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