Investigation of the transit-time broadening in optical nanofiber based spectroscopy

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Abstract: Optical nanofiber is a widely adopted platform for highly efficient light-matter interaction by virtue of its exposed evanescent field with high light intensity. However, the strongly constrained mode field with the wavelength-scale size makes the light-matter interaction time limited in consideration of the random thermal motion of warm molecules, which results in considerable transit-time dephasing and thus line broadening. Here we report a systematic study of the transit-time effect associated with the optical nanofibers. Both simulation and experiment for nanofibers exposed in acetylene demonstrate the considerable transit-time broadened linewidth in the low-pressure range.

1.   Introduction

Adiabatically tapering a piece of single-mode silica fiber so that the preciously core-cladding guided optical mode is smoothly converted into cladding-air guided mode for convenient light-matter interaction has been a conventional technique in the fields of atomic physics, nonlinear optics and optical sensing. The waist region of the fiber taper is typically of sub-micron diameter and is thus also called as optical nanofiber. When the wavelength of a guided light beam is larger than the diameter of nanofiber, there will be a large portion of evanescent field extending into the air. The tightly confined optical field with a sub-micron mode radius results in high light intensity even under moderate input power [1], which makes the nanofiber a versatile platform for efficient light-matter interaction with up to centimeters-long interaction length, far beyond the diffraction limit in a free-space scheme. Nanofibers coupled with an ensemble of neutral atoms have found a range of applications, including laser trapping [2], light storage [3], all-optical switch [4] and fluorescence probe [5], all with high efficiencies. While the thermal motion is suppressed by cooling the atoms, it is inevitable for experiments with warm atom/molecules. The process of molecules' traversing through the tightly confined evanescent field or colliding with the surface of the nanofiber will destroy their coherence with the light beam and result in additional transit-time broadening, as illustrated in Fig. 1. This transit-time dephasing effect associated with optical nanofibers has been previously observed in experiments of electromagnetically induced transparency (EIT) [6], two-photon absorption [7] and saturation absorption spectroscopy [8,9]. In addition, high performance photoacoustic [10], photothermal [11] and stimulated Raman spectroscopy [12] based on nanofiber for trace molecule detection have also been reported recently. However, there has not been a systematic study of the transit-time broadening effect associated with nanofibers.

Here, we investigate numerically and experimentally the additional line broadening effect for warm molecules interacting with the evanescent field of an optical nanofiber. The simulation is based on the density matrix method for a two-level system. Due to the transit-time effect, a unique lineshape which is quite different from the Gaussian or Lorentzian function is demonstrated. Experiments of absorption spectroscopy for acetylene with nanofibers of different diameters are performed and the results match well with the theory. It is concluded that the transit-time
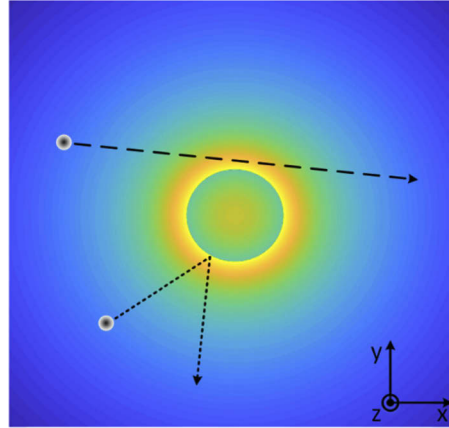
Fig. 1. Illustration of the transit-time dephasing of nanofiber based light matter-interaction. Both the processes of traversing through the mode field (the dashed trace) and collision with the wall of the nanofiber (the dotted trace) will cause decoherence between the molecule and the light beam. The in-plane (i.e., x-y plane) molecular movement results in transit-time broadening while the out-of-plane movement (z-direction) causes Doppler broadening.

broadening is of the same order but less than the Doppler Effect, and its magnitude highly depends on the diameter of the nanofiber.

2. Theoretical model

The model is based on the density matrix method for a molecular two-level system in the linear absorption range and thus the effect of power broadening is absent. The molecules are assumed to occupy the ground state initially and evolve following the density matrix equation [13]:

$$i\hbar \left(\frac{d}{dt} + \vec{v} \cdot \nabla \right) \rho = [\hat{H}_0 + \hat{V}, \rho] + i\hbar R(\rho - \rho_0),$$

(1)

where, $\vec{v}$ is the molecular velocity, $\hat{H}_0$ is the unperturbed molecular Hamiltonian, $\hat{V}$ is the perturbation due to the optical field, R is the relaxation rate, ρ_0 is the unperturbed diagonal density matrix. It is the first order perturbation of the off-diagonal elements $\rho_{ab}^{(1)}$ that is responsible for the linear absorption, which, under the rotating wave approximation, is expressed as:

$$\left(v_x \frac{d}{dx} + v_y \frac{d}{dy} \right) \rho_{ab}^{(1)} = i(\omega_p - \omega_{ab} - k_z v_z)\rho_{ab}^{(1)} - \gamma_{ab}\rho_{ab}^{(1)} - (2i\hbar)^{-1}\vec{\mu}_{ab}\vec{E}n_{ab}F(\vec{v}),$$

(2)

where, ω_p and ω_{ab} are the angular frequency of the probe beam and the transition respectively, γ_{ab} is the dephasing rate of the optical dipole proportional to the pressure, $\vec{\mu}_{ab}$ is the molecular transition dipole, $\vec{E}$ is the optical field of the fundamental mode of nanofiber, n_{ab} is the differential population between the upper and lower levels of transition, $F(\vec{v})$ is the Maxwell-Boltzmann velocity distribution function. The attenuation coefficient for the guided probe beam along a nanofiber in the atmosphere of light absorbing molecules will satisfy:

$$\alpha(\omega_p) = \frac{N\omega_p Im \left(\int_r^\infty \vec{\mu}_{ab}\vec{E} \cdot \bar{\rho}_{ab}^{(1)} ds \right)}{\frac{1}{2} \int \vec{E} \times \vec{H}^* ds},$$

(3)

where, N is the molecular density, r is the radius of the nanofiber, $\bar{\rho}_{ab}^{(1)}$ is the integrated $\rho_{ab}^{(1)}$ over the whole velocity distribution as $\bar{\rho}_{ab}^{(1)} = \int \rho_{ab}^{(1)} d^3 v$. Although the simulations will be performed with the example of acetylene, the results can be generalized to other atomic or molecular species. The pure transit-time dephasing can be obtained by freezing the z-directional molecular movement, i.e., setting v_z in Eq. (2) being zero, which is responsible for the Doppler Effect. Different transverse velocity group $v_{tr} = \sqrt{v_x^2 + v_y^2}$ will has different distribution of $\rho_{ab}^{(1)}$ and thus different contribution to the total absorption.
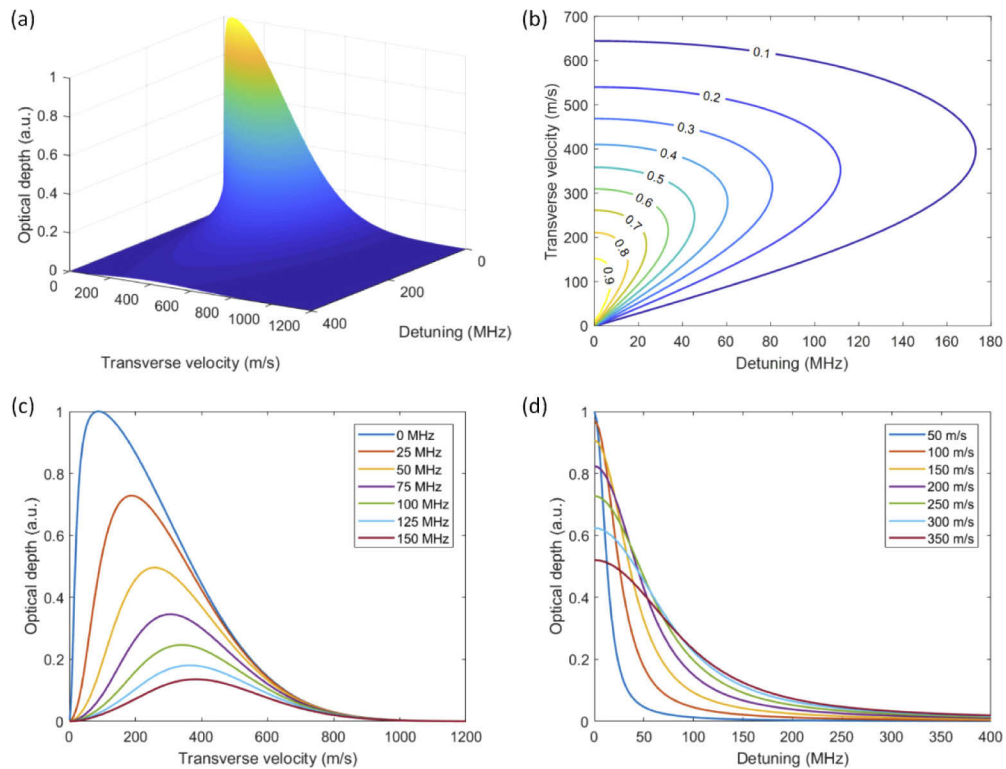
3. Simulations

The simulations are performed with Python based on the finite difference method. The $\vec{E}$ in Eq. (2) is the evanescent field of a circularly polarized fundamental mode of the nanofiber [14]. Then both the differential operation and the coefficients of terms containing $\rho_{ab}^{(1)}$ in Eq. (2) are transformed into matrixes. The unknown $\rho_{ab}^{(1)}$ can then be obtained by solving the linear matrix equations. The acetylene's P(13) ro-vibrational transition in $v_1 + v_3$ band with wavenumber of 6524 cm^{-1} [15] is adopted as the example of simulation. In the investigation of the transit-time and Doppler broadening, the pressure of pure acetylene is fixed to 10^{-5} bar, under which condition the inter-molecular collisional broadening is negligible.

For a 700-nm-diameter silica nanofiber, the peak normalized optical depth from different transverse velocity group under varied detuning is shown in Fig. 2(a) with its contour plot presented in Fig. 2(b). The simulation reveals the detailed interaction between the evanescent field and the sub-groups of molecules with different transverse velocity. It can be found that the slow molecules have the major contribution to the total absorption, as observed by Hald *et al.* in their experiment of saturation absorption spectroscopy in hollow-core photonics bandgap fiber [16]. In addition, it is the molecules with larger transverse velocity that contribute more to the absorption at larger detuning from the line center, as indicated in Figs. 2(c) and 2(d). The lineshape due to the pure transit-time effect for the whole ensemble of molecules can be obtained by integrating the optical depth over each velocity sub-groups. For this 700 nm-diameter nanofiber, the pure transit-time originated lineshape is shown as the purple line in Fig. 3(a), which is quite different from the commonly seen Lorentzian or Gaussian function. A similar simulated lineshape using the Monte Carlo approach was reported by Griesser *et al.* in their study of spectrum broadening of single-photon transition in the evanescent field of an exposed-core fiber [17]. As they approximated the evanescent field of the exposed-core fiber with a 1-dimensional exponential function, an analytical expression for the Doppler-free lineshape was obtained. However, due to the totally numerical procedure in our 2-dimensional nanofiber case, an analytical expression may not be obtained.

The simulation results of the pure transit-time effect for different diameters of nanofiber are shown in Fig. 3. It can be seen that the thinner the diameter of the nanofiber, the less the linewidth due to the transit-time effect. This is quite reasonable as the evanescent filed expands with reduced diameter of the nanofiber, which results in longer time for the molecule to interact with the optical field as well as less chance to collide with the wall of the nanofiber to cause decoherence. The simulated lineshape due to the pure transit-time dephasing match well with the ones observed by Hendrickson *et al.* in their experiment of nanofiber based two-photon absorption spectroscopy, where the resonant sharp peaks were attributed to the transit-time effect by the authors [7].

The Doppler Effect can be taken into consideration by setting different values of v_z and integrating the absorption over the whole z-directional velocity distribution. The simulation results are shown in Fig. 4. Different from the free space system, where the lineshapes are Gaussian in the low-pressure range due to the dominant Doppler Effect, the collisional broadening free lineshapes in the cases of nanofiber deviate from Gaussian function due to the considerable

Fig. 2. Simulation results of the pure transit-time broadening for a 700 nm diameter nanofiber. (a) Peak normalized optical depth for different transverse velocity and detuning; (b) Slices of (a) with different relative optical depth ranging from 0.1 to 0.9; (c) Slices of (a) with different detuning ranging from 0 to 150 MHz; (d) Slices of (a) with different transverse velocity ranging from 50 to 350 m/s.
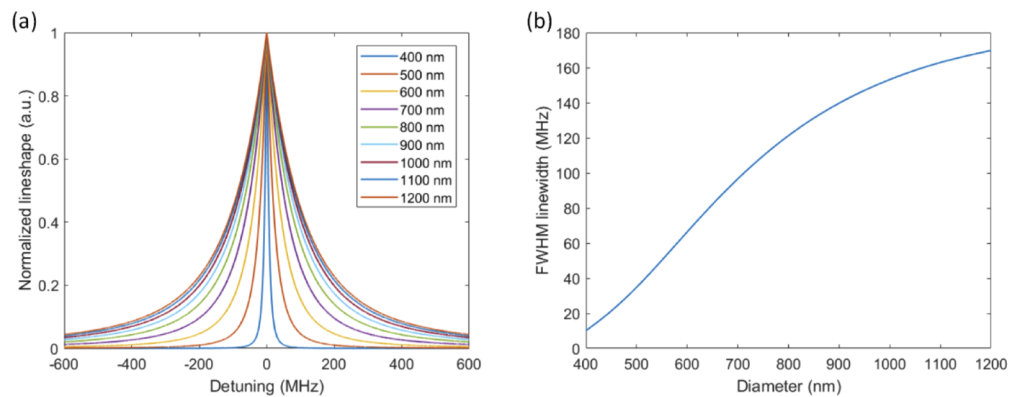
Fig. 3. Simulation results of (a) lineshape and (b) Full-width-half-maximum (FWHM) linewidth of pure transit-time broadening for different diameter of nanofibers.

transit-time effects. The magnitude of the pure transit-time broadened linewidth is about 30% of the Doppler linewidth for nanofiber with diameter near 1 micron. Although the transit-time effect can be relieved for reduced diameter, too thin nanofibers are generally not adopted due to the excessively expanded mode field and thus rapidly dropping intensity, which does not appeal to

the nanofiber based nonlinear light-molecule interaction such as photoacoustic or photothermal spectroscopy, stimulated Raman scattering and EIT. So for the cases where nanofiber operates in low-pressure condition, the transit-time effect may not be omitted.
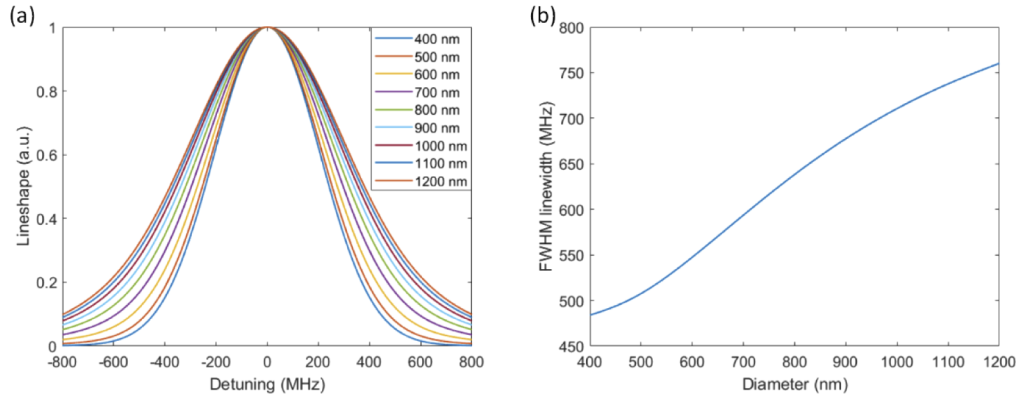
Fig. 4. Simulation results of (a) lineshape and (b) FWHM linewidth of collisional broadening free cases for different diameter of nanofibers.

4. Experiments

To verify the theory, experiments of nanofiber-based absorption spectroscopy are conducted. The nanofibers are taper-drawn from the standard single mode fiber (Corning SMF-28) by the flame-brushing method and have transmission loss less than 0.3 dB [18]. Immediately after the fabrication, the sample is sealed into an aluminum gas cell to prevent the dust in the air from degrading the optical transmission. Pure acetylene is inflated into the gas cell through the dust filter, which is used for elimination of any potential contamination, as shown in Fig. 5. The gas pressure is reduced by a mechanical vacuum pump and monitored by a pressure gage.
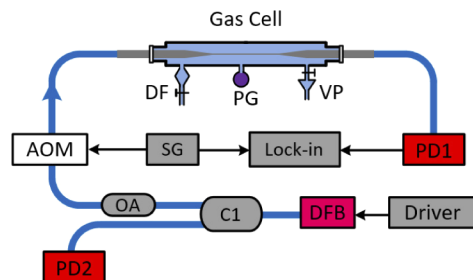

Fig. 5. Experimental setup of nanofiber-based absorption spectroscopy. DFB: distributed feedback laser (Eudyna FLD5F15CX); C1: 1:1 fiber-optic coupler; OA: Optical attenuator (GO4FIBER GVOA-1550); AOM: acousto-optic modulator (G&H Fiber-Q); SG: signal generator (RIGOL DG4000); Lock-in: Lock-in amplifier (Stanford Research Systems SR830); DF: dust filter (TAIYO SFF-08); PG: pressure gauge (ZHVAC ZJ-1P); VP: vacuum pump (EDWARDS E2M1.5).

The probe source is a distributed feedback (DFB) laser whose wavelength is ramped through the P(13) absorption line in $\nu_1 + \nu_3$ vibrational band of acetylene centering at 1532.83 nm. The probe beam is intensity modulated by an acousto-optic modulator at 30 kHz to improve the signal-to-noise ratio. The power of probe beam is then measured by a photodetector (PD1, Nirvana-2017). An optical attenuator is used to reduce the probe power level to within 1 µW

so that the power broadening effect, which would be appreciable for milli-watt power level, is surely excluded [8]. The spectrum-dependent output power of the DFB is monitored by a second photodetector (PD2, New Focus-2011) and compensated for the signal demodulated by the lock-in amplifier.

The experimentally measured full-width-half-maximum (FWHM) linewidth of acetylene for three pieces of nanofiber with waist diameter of 1020, 770 and 580 nm at different pressure are shown in Fig. 6. For comparison, the absorption linewidth is also measured in free space setup, which is made up of a pair of fiber collimators with light beam diameter of 0.5 mm. In the higher-pressure range, the linewidth is inter-molecular collisional broadening dominant with a slop of 88.5 MHz/kPa. In the lower pressure range, the linewidth for the free space case is Doppler limited (480 MHz for acetylene) [19]. However, for the nanofiber cases, the linewidth has a considerable contribution from the transit-time broadening effect, which results in a deviation from the Doppler limit. The thinner the diameter of the nanofiber, the larger the expansion of the evanescent field and thus the less the transit-time broadening due to the transversal molecular movement. So, with reduced diameter the transit-time broadening is relieved. It should also be mentioned that the Doppler linewidth for thicker nanofiber is also increased due to the larger propagation constant [20]. Compared with the free space one, the Doppler linewidth, which originates from the z-directional molecular movement, of the three pieces of nanofiber are 18%, 9% and 2% enlarged. Subtracting the residual collisional broadening of 17 MHz at 0.2 kPa, the collisional broadening free linewidth for these three pieces of nanofibers are 750, 648 and 568 MHz, which are respectively 4.6, 3.8 and 5.5% larger than the theoretical value as given in Fig. 4(b). The deviation should be caused by the residual absorption from the transition region of the taper, which has larger transit-time broadening compared with the waist region.
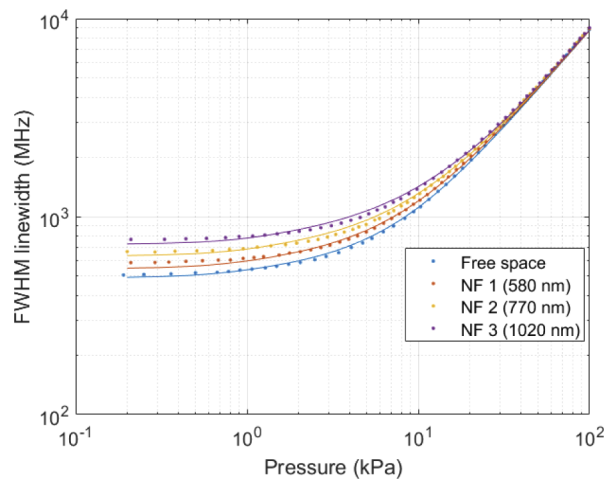
Fig. 6. Experimentally measured (dotted) and simulated (lines) linewidth of P(13) line of pure acetylene under different pressure for cases of nanofibers and free space. NF1: 580 nm, NF2: 770 nm, NF3 1020 nm.

5. Summary

We have studied the transit-time broadening effect in nanofiber-based absorption spectroscopy. The transit-time effect originates from the fast transversal motion of the molecules out of the tightly confined evanescent field associated with the nanofiber as well as the collision with the surface of the nanofiber. The magnitude of the pure transit-time broadened linewidth is about 30% of the common Doppler linewidth for a nanofiber with diameter about 1 micron and is

reduced for thinner nanofibers. The experimentally verified theory can work as guidelines for nanofiber based high-precision spectroscopy and nonlinear light-matter interaction, where the transit-time effect may not be neglected.

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Disclosures

The authors declare no conflicts of interest.

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