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Optical gas delay-line for ultrafast process measurements

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Abstract

A novel gas delay-line technique is introduced. A two-beam interference method is used to calibrate the delay-line. Time-delayed degenerate four-wave mixing experiments are performed by it at several gassing velocities. The gas delay-line exhibits well precision, stability and repetition.

1. Introduction

Ultrafast optical dephasing has been studied in many systems over the past two decades and has greatly increased our understanding of the dynamical processes in various systems [1–3]. The relaxation time constants are typically derived from time-delayed degenerate four-wave mixing (DFWM) measurements using ultrafast laser or broadband incoherent light. An optical mechanical delay-line (MDL) is usually used in the DFWM experiments. The delay-line must be hyper-precision-machined to warrant the stability of the optical path. It is often expensive when the time delay is of order femtosecond increment. Another kind of time-delayed method is using piezoelectric actuator [4]. In this letter, we introduced a novel gas delay-line (GDL) technique and the DFWM measurements were performed by it.

2. Experiment

The GDL was devised as following: gas was contained in a sealed tube connected with a gas chamber and a pressure sensor. There were quartz glass windows in both sides of the tube, so the light beam could transmit through the gas tube. The thickness of the windows was 0.5 cm and the diameter was 1.5 cm. The optical path length could be easily changed with the increase or decrease of the pressure of the gas. In our experiments, nitrogen gas of maxim pressure 10 bar was filled into the 30 cm-length tube, the range of time delay was about 2000 fs, and the precision of the sensor was 0.1 mV corresponding to 4.7 mbar.

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To determinate the relation between the pressure and the delay time, precise calibration of the GDL was carried out by a two-beam of laser interference method, one of the beams was delayed by the GDL. In order to determinate dispersion coefficient, the experiment was finished by He–Ne and Ar^+ ion lasers simultaneously.

3. Results and discussion

For a two-beam interference experiment, the detected intensity can be expressed as $I = I_0[1 + \gamma\cos(\omega t + \delta)]$, where γ is a constant, ω is the frequency of the light, and δ is the initial phase. The period of the intensity oscillation is just equal to light period, for He–Ne and Ar^+ ion (488 nm) laser the values are 2.109 and 1.627 fs, respectively. We filled gas into the chamber, then recorded the intensity oscillations of one of the interference strips, while the outlet of the gas chamber was open. Since the outgassing process was not with a constant velocity, we also recorded the gas pressure at the same time. The calibration results were shown in Fig. 1. The decrease of the gas pressure was well fitted by an exponential decay form. This was true because the outgassing velocity was proportional to the pressure difference between the chamber and atmosphere. Figs. 1(a) and (b) show a part of the outgassing curves and the intensity oscillations where the x-axis was corrected by the curve in Fig. 1(a). The slopes of the delay time versus the gas pressure were shown in Fig. 1(c) for the whole range of the pressure. The calibration results show well linear relation, the pressure changing 4.7 mbar corresponded to the time delays of 1.016 ± 0.004 and 1.026 ± 0.004 fs, for He–Ne and Ar^+ ion, respectively. Using dispersion formula $n = n_0 + B/\lambda^2$, we have the dispersion coefficient $B = 11.8$ nm^2.

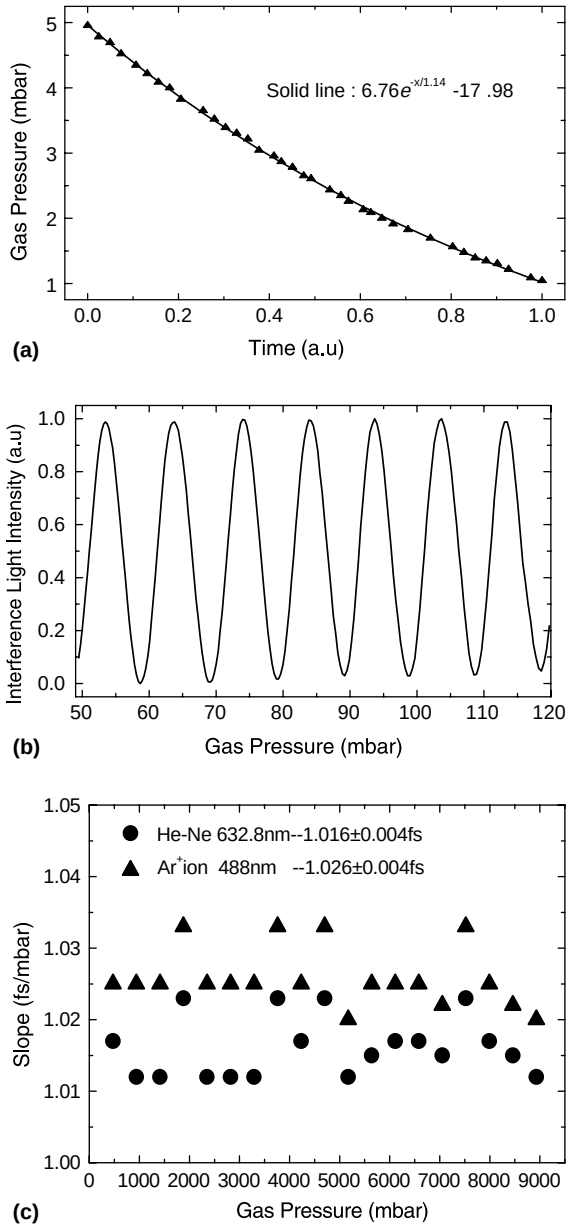

Fig. 1. Calibration curves of gas delay line: (a) Gas pressure versus time. (b) Shift of interference strips. (c) Calibration results for He–Ne and Ar+ ion lasers.

4. Conclusion

In fact, the outgassing process was a continuous one. One can see from Fig. 1(b), we recorded a well cosine-form intensity changing. So, if one slows down the outgassing velocity, and uses a more precise gas sensor, a GDL can easily resolve several tens attoseconds, even a few attoseconds.

In the following experiment, a three-beam time-delayed DFWM method with incoherent light was used to measure the population relaxation of the

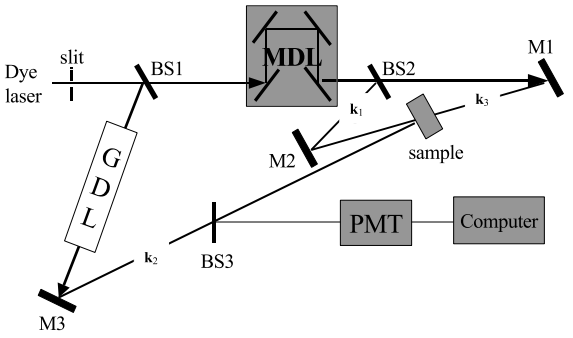

Fig. 2. Experimental setup for DFWM using gas delay-line.

sample (see Fig. 2). In order to avoid the degradation of the time resolution resulting from the thickness of the sample, the phase-conjugation configuration was used. The light beam $\mathbf{k}_2$ was retarded by the GDL, $\mathbf{k}_1$ and $\mathbf{k}_3$ were both retarded by a conventional mechanical delay-line. From beam $\mathbf{k}_1$, beams $\mathbf{k}_2$ and $\mathbf{k}_3$ were retarded by the time delays of τ and τ', respectively. A frequency-doubled Q-switched Nd:YAG laser pumped the laser dye Rhodamine 6G to generate a beam of amplified spontaneous emission, which had a well Gaussian line shape with half width at half maximum (HWHM) of 70 cm^{-1} corresponding to the correlation time $\tau_c = 337$ fs. Since testing the GDL was our main purpose, we chose the filter glass as a sample. Its population decay time T_1 is in the order of magnitude of ns, which is much larger than τ_c. When $\tau' \gg \tau_c$, for the light with a Gaussian autocorrelation function the signal can be expressed as: [5]

$$I(\tau) \propto 1 + \frac{0.8T_1}{\tau_c}\exp[-2(\tau/\tau_c)^2] + \frac{1.4\tau_c}{T_1}\exp[-2\tau/T_1] \quad (1)$$

for the positive delay time. The last term (called "tail" term) disappears when the delay time is negative. At our experimental condition, the tail term was about eight orders smaller than the second term. So, in fact, we just measured the autocorrelation peak of the incoherent light. To test the stability and repetition, we performed the experiments at several gassing velocities. In the experiment, the center wavelength of the light was 575 nm. Considering the dispersion the time delay was 1.019 fs when the pressure of GDL changed 4.7 mbar. The experimental DFWM signal was well fitted by the Gaussian autocorrelation function. The comparison of the HWHMs and peak positions at various gas outflow velocities was shown in Table 1. As mentioned above, the gas pressure versus time could be expressed as $\exp(-V_g t)$, where V_g is the outgassing velocity. The results show well repetition.

Table 1
HWHMs and peak positions of DFWM signals at various outgassing velocities

V_g (a.u.)	HWHM (fs)	Peak position (mbar)
0.6	220	5179.4
1	230	5193.5
1.5	235	5188.8

In our experiment, incoherent light was used. Time resolution of the measurement is limited by the coherent time of the light, so a nanosecond pulse even a continuous light may be used in picosecond or femtosecond measurements, as long as the coherent time of the light is short enough. In this case, group velocity dispersion in the GDL does not affect the time resolution.

On the other hand, if ultrafast light pulse is applied in the DFWM measurement, group velocity dispersion in the GDL will broaden the pulse width. In order to maintain the time resolution, a conventional compensation set-up should be put into the $\mathbf{k}_2$ beam behind the GDL.

In conclusion, a novel gas delay-line technique is introduced. By incoherent light time-delayed degenerate four-wave mixing experiments, the gas delay-line exhibits well precision, stability and repetition at several gassing velocities. It can be used in the measurement of ultrafast processes.

Acknowledgements

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