Femtosecond Lasers for Intracavity Molecular Spectroscopy

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Spectroscopy and gas sensing at low concentrations is of interest for science and a number of applications including medicine, industry, environment monitoring, etc. The absorption lines of many gases lie within the mid-infrared region including the atmospheric window between 2 – 5 μm, with overtones in the near infrared domain of 1 – 2 μm. In these regions there exist ultrabroadband solid-state oscillators that allow using the method of transient intracavity laser spectroscopy (ICLAS) [1-3]. The ultrahigh sensitivity of this method in detection of the weak narrowband absorbers results from the extremely large effective absorption length, which is achieved by many round-trips of the broadband laser field, before it collapses to a narrow laser line.

In this work we analyse a quasi-steady-state realization of the broadband intracavity spectroscopy, which is possible with a mode-locked femtosecond oscillator emitting an ultrabroad spectrum affected by the narrowband absorbers (e.g. gases). We demonstrate both numerically and experimentally that the resulting spectrum is different from the usual transient ICLAS [3] and the extra-cavity femtosecond absorption spectroscopy [4], because the soliton spectrum is affected mainly by the intracavity dispersion of the oscillator. The results of simulations based on the nonlinear cubic-quintic complex Ginzburg-Landau model of a femtosecond oscillator [5] are presented in Fig. 1. Although the integral absorption is very small, the effect is strong due to the large value of the associated dispersion, which is rapidly increasing with the narrowing of the absorption line.

Fig. 2 shows a comparison of the intra- (red line) and extra-cavity absorption experiments of acetylene using the same evacuated femtosecond Cr:ZnSe laser. The intra-cavity spectrum is purely dispersion-related, in contrast to the normal absorption spectrum recorded outside of the cavity. Note that the dispersion signal has comparable amplitude for same absorption length, despite 17 times lower C\textsubscript{2}H\textsubscript{2} concentration and broader line.

\textbf{Fig. 1} Simulated spectral shape of the 100 fs soliton without (black dashed curve) and with (solid black curve) Lorentz absorption line (40 GHz linewidth, 10% intracavity absorption at the line centrum). Blue curve is the ratio of the spectra, red curve is the phase response of the Lorentzian absorption line. Soliton width is 100 fs (3.2 THz).

\textbf{Fig. 2} Experimental spectral of acetylene absorption inside the 75-cm laser cavity (red line) and in a 80-cm extra-cavity absorption cell (gray line, [4]). The left graph shows the expanded portion of the whole spectrum. The additional periodic modulation on the left graph comes from the unwedged plate used as a laser chamber output window.

The dispersion profile of the absorption line can be reconstructed from the soliton spectrum with high accuracy, because the relative signal (blue curve in Fig. 1) reproduces the phase response of the line (red curve). Besides the narrowband feature, the absorption line also causes a red-shift of the soliton central frequency. In the time domain, the absorption line results in (1) group delay of the soliton, and (2) appearance of the exponentially decaying pedestal before the soliton with the decay time equal to the inverse absorption linewidth.

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References