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Experimental Nonlinear Interference Comb Spectroscopy

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Abstract—A means of nonlinear interferential comb spectroscopy with increased sensitivity is propsed. Radiation from the comb-generator of a femtosecond laser is focused at the center of a cell with vapors of rubidium atoms, mounted in an arm of a Michelson interferometer. This technique is an improvement of Rozhdestvenski hooks method, with digital registration by the detectors on a CCD array being substituted for the photographic registration of interferograms, resulting in increased interferometric sensitivity (holographic interferometry). A primary spectrogram is processed in digital form on a dual-beam interferometer equipped with a phase modulator to improve sensitivity with the possibility of an a posteriori increase in interferometric sensitivity. Dispersive signals appear in the interferogram spectrogram on two-photon absorption lines when the pumping radiation is focused at the center of a cell with Rb vapors. Nonlinear processes of coherent radiation due to nonlinear interference effects are studied. Numerical simulations are performed that confirm the proposed theoretical model.

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This work presents the results from experimental studies of the susceptibility spectrum and atomic polarization by means of nonlinear comb spectroscopy. The object of study is vapors of Rb atoms (a natural mixture of isotopes). A sequence of femtosecond pulses from the comb generator of a broad-band femtosecond laser is used to analyze the spectral behavior of the refractive index near lines of resonance. The laser illuminates a Michelson interferometer; the cell filled with the gas is mounted in one of the arms of the interferometer. A spectrogram—the interference pattern at the exit slit of the spectrograph—is recorded with a CCD array. The digital picture is then processed by the means of holographic spectroscopy. The spectrographic interferogram reflects the course of anomalous dispersion at relatively low pump intensities. Adjusting the bands of the interferogram to Rozhdestvenski hooks allows us to obtain information on the oscillator forces. Accumulation of signals on the order of several tens of thousands of frames over several hundred microseconds is possible. The contrast of the interference pattern does not in this case change upon an increase in exposure, testifying to the stability of the phase of optical oscillations (i.e., the frequency stability of the comb generator). The duration of a laser pulse is ~150 fs; the pulse repetition rate is ~80 MHz. Figure 1 shows an interferogram with Rozhdestvenski hooks at moderate laser radiation intensities without radiation focusing in the cell. Times and spectra were also measured. A numerical simulation was performed in the density matrix formalism. The simulation confirmed the validity of the procedure proposed for generating coherent radiation with slight (close to diffraction) divergence upon the transition at a wavelength of 420 nm when pumped at a wavelength of 780 nm.

Figure 2a shows an interferogram at low Rb vapor density; Fig. 2b, an interferogram with doubled sensitivity [1]. This technique can be used to study an oscillator in highly excited states, along with nonlinear effects in atomic and molecular media under the impact of strong optical fields. As is well known, saturation of absorption is observed in nonlinear effects and the signal attenuates. It is therefore necessary to improve the interferometric sensitivity of the hook method. The primary spectrographic interferogram is stored in digital form and supplied to a LETO reflective SLM (Spatial Light Modulator; resolution,

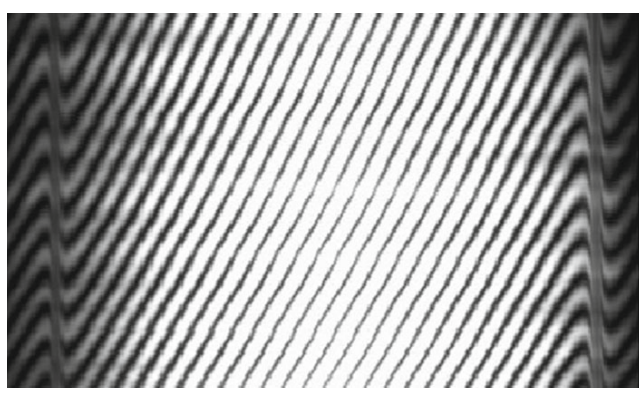

Fig. 1. Interferogram of Rozhdestvenski hooks near the resonant doublet of an Rb atom, taken with illumination of the Michelson interferometer by femtosecond laser comb-generator radiation.

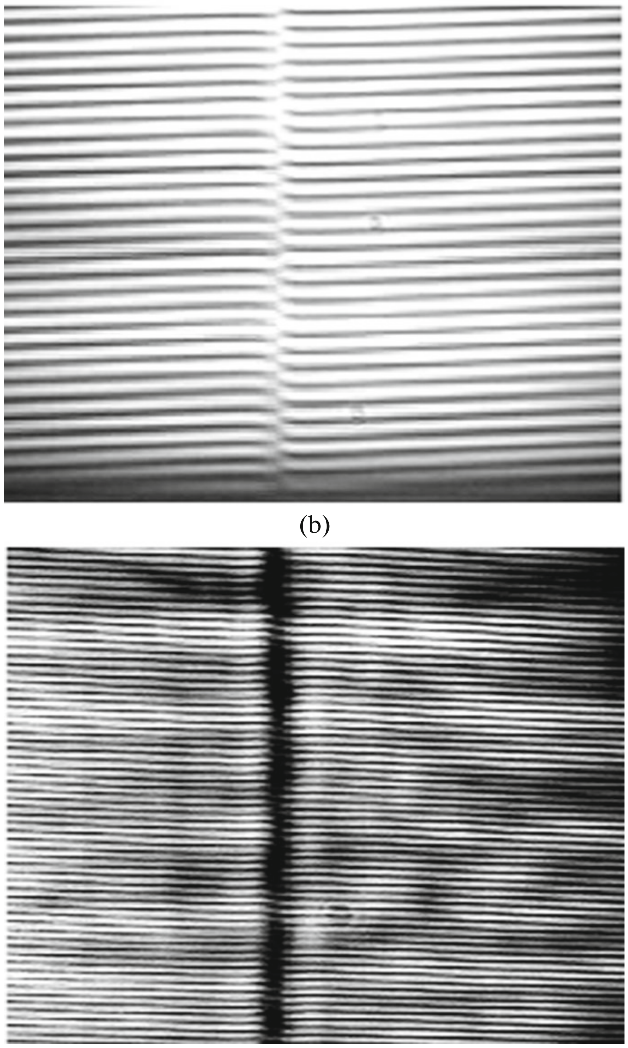

Fig. 2. (a) Initial spectrographic interferogram near the absorption line of Rb atoms (wavelength, 780 nm); (b) secondary interferogram with increased interferometric sensitivity.

1920 × 1080 pixels) for processing. The modulator is then placed in the hologram recovery unit [2], and a secondary interferogram is recorded with doubled sensitivity of the interference measurements. The sensitivity can be increased by 2^n times with subsequent rewriting of the interferograms (where n is the number of rewritings).

In addition to measuring time and spectra, the proposed technique was used to study nonlinear effects. Upon focusing the comb generator radiation at the center of the cell, we observed such nonlinear effects as radiation amplification upon the resonant $5^2S_{1/2} - 5^2P_{3/2}$ transition (780 nm), the $5^2P_{3/2} - 5^2D_{5/2}$ transition (776 nm), the resonant $5^2S_{1/2} - 5^2P_{1/2}$ transition (795 nm), and the $5^2S_{1/2} - 6^2P_{3/2}$ transition (420 nm). The radiation at the last transition was of particular interest. First, the divergence of this radiation was only 2–3 mrad; its intensity at 420 nm was proportional to the squared pump intensity at the resonance transition wavelength of 780 nm, and to the squared concentration of atoms at ground level. All of these properties of coherent radiation during the conversion of pump radiation from the near-IR to the blue region of the spectrum indicate strong nonlinear interaction associated with the nonlinear effects of the interference of atomic states produced in the atomic system [3]. Times and spectra were also measured, confirming the validity the proposed model.

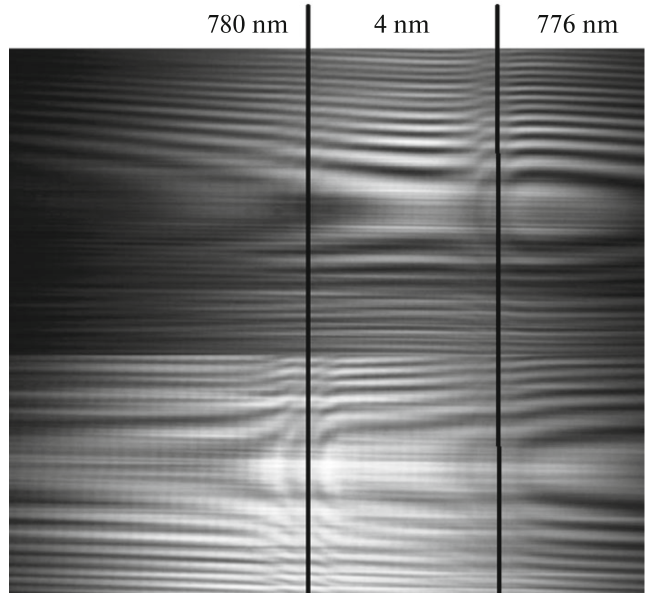

Fig. 3. Spectrographic interferogram of the variance in the refractive index of the atomic medium when the pump radiation was focused at the center of a cell with Rb vapors. Two-photon absorption of the pump radiation was observed at the wavelengths of the cascade transition, the resonance transition at 780 nm, and the transition at 776 nm.

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