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An ultrafast light-intensity modulator, based on stimulated Raman scattering, is described. The intensity of a continuous wave laser is fully modulated at 17 THz using hydrogen in a high-finesse cavity. The modulation frequency is determined by the molecular constant of the Raman medium, i.e., the Raman shift frequency. The modulation frequency can be changed in the tetrahertz range by replacing the Raman medium. Due to the accurate modulation frequency and the high beam coherence, this device is amenable to a variety of applications such as in basic science and also in advanced industrial technology. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2174091]

Various types of optical devices have been used to modulate the intensity of light. For example, a laser beam can be modulated at ~100 MHz by means of an acousto-optic modulator, in which a transient grating effect is formed in a glass by an acoustic wave and diffracts the input light, thus generating a modulated beam. On the other hand, an electro-optic modulator (EOM) changes the polarization of a beam by the application of a high electric potential to a crystal, e.g., potassium dihydrogen phosphate, and generates a modulated beam to ~1 GHz, when the beam is passed through a polarizer. A state-of-the-art quasi-velocity-matched electro-optic modulator with periodic domain inversion can generate a train of pulses modulated at 16.25 GHz with a sideband spectral width of 2 THz.[1]

When an intense laser pulse is focused into a Raman medium such as molecular hydrogen, stimulated Raman emission occurs. Numerous vibrational/rotational emission lines can be simultaneously generated from the ultraviolet to the near-infrared by subsequent four-wave Raman mixing.[2] It has been proposed and also verified experimentally that an ultrashort optical pulse can be generated by phase locking the emission lines that are produced in this process.[3–5] Ultrashort optical pulses have already been generated using Raman emissions.[6,7] When a cw laser is used as a pump source, it is difficult to exceed the threshold for the generation of stimulated Raman scattering. This problem can be solved by utilizing a high-finesse laser cavity, to confine the laser energy in the resonator.[8,9] It has been suggested that highly-repetitive optical pulses, e.g., 17 THz for *ortho*-hydrogen, can be generated by phase locking more than three emission lines.[10]

In this letter, we report on the development of an ultrafast molecular-optic modulator (MOM) based on stimulated Raman scattering. This technique allows the intensity modulation of a cw laser beam at 17 THz, obtained as the result of the optical beat of the fundamental and rotational Raman emissions. The modulation frequency is determined by the rotational Raman shift frequency of *ortho*-hydrogen, i.e., 587 cm^{-1}, providing a sinusoidal optical wave with an interval of 57 fs. We experimentally confirmed light-intensity modulation at 17 THz by constructing a sensitive autocorrelator, with a time resolution of several femtoseconds.

The experimental apparatus used in this study is shown in Fig. 1. A cw Ti:sapphire laser (Coherent, MBR-110, 792 nm, 450 mW, $\Delta\nu < 100$ kHz) is pumped by a Nd:YVO_4 laser (Coherent, Verdi, 532 nm, 5 W). The laser beam is passed through an optical isolator (Optics for Research Co, Ltd., IO-5-NIR-HP), to prevent feedback of the reflected beam. The diameter of the beam is reduced by a factor of 2 by means of a pair of lenses and is passed through a half wave plate, an EOM (New Focus, model 4001) modulated at 12 MHz, a half wave plate, a polarization beam splitter, in series. These optics can be used to stabilize the cavity length of a Raman resonator by means of Pound-Drever-Hall method.[11] A quarter wave plate is employed to rotate the beam polarization to generate a rotational Raman emission. The beam is expanded and weakly focused into the high-finesse Raman cavity. The pressure of hydrogen in the Raman cell was adjusted to 10 atm. The cavity length (8 cm) can be changed in increments of 5 μm using a piezoactuator to match the longitudinal mode with the wavelength of the laser. The reflectivity of the mirrors used for the high-finesse Raman cavity is 99.982% at 792 nm (fundamental beam) and 99.972% at 831 nm (Stokes beam). The beam diameter and the transverse mode are monitored using a beam profiler (OPHIR, BeamStar-V). The spectrum of the output beam is measured by means of a spectrometer (Ocean Optics, USB-2000). The long-term stability of the fundamental or Stokes beam was recorded by replacing the spectrometer with a monochromator (Jasco, CT-10) equipped with a photodiode. An ultrafast, fringe-resolved autocorrelator, based on two-photon absorption of a photocathode in a photomultiplier (e.g., 1P28),[12] was constructed in this laboratory and used in the experiments described herein.

Figure 2 shows a photograph of the MOM device (17 cm long, 6.5 cm wide, 6.5 cm high) constructed in this study. The fundamental beam of the Ti:sapphire laser can be transmitted by the spatial mode matching of the propagating beam with a high-finesse cavity and by adjusting the cavity length, so as to fit the longitudinal mode of the resonator with the wavelength of the fundamental beam.[8,9] Figure 3 shows a spectrum of the output beam from the high-finesse cavity filled with hydrogen. The intensity of the Stokes beam has become nearly equal to that of the fundamental beam. The

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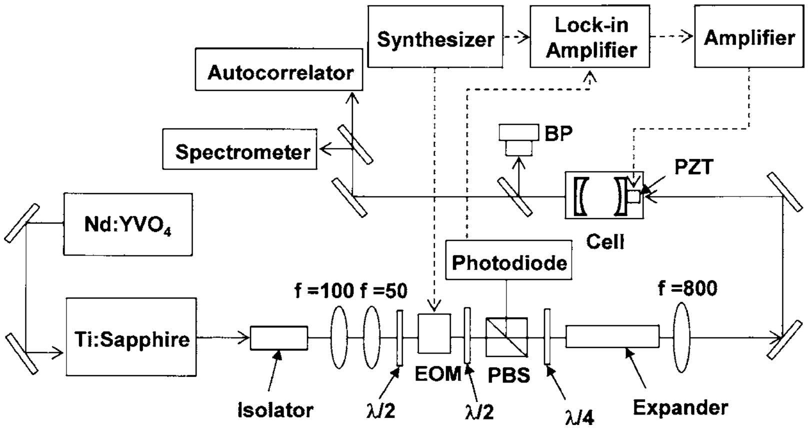

FIG. 1. Experimental apparatus for ultrafast molecular-optic modulation. EOM—electro-optic modulator; PBS—polarization beam splitter; PZT—piezotranslator; and BP—beam profiler.

laser power of the transmitted beam was ~10 mW, suggesting the laser power for the fundamental and Stokes beams to be 5 mW, respectively. Then, the efficiency of conversion to the Stokes emission in the high-finesse cavity was calculated to be ~2%. The continuous generation of Stokes emission (not spikes) was confirmed by measuring the intensity using a photodiode after passing through a monochromator. In this experiment, the light intensity was stable and the transverse mode was maintained at TEM_{00} for over 60 min by means of a manual control of the piezotranslator (PZT) actuator without any automatic feedback control for stabilization of the high-finesse cavity.

In order to measure the intensity modulation of the beam, we constructed an autocorrelator with a femtosecond resolution and confirmed that it was sufficiently sensitive (minimum intensity, 4.5×10^3 W/cm^2) to permit the pulse to be measured in the milliwatt range, based on the *Z*-scan method.[13] This autocorrelator based on a Michelson interferometer allowed the measurement of the pulse width in a spectral range of 770–830 nm and in a time domain of 1–600 fs. Figure 4 shows a fringe-resolved autocorrelation trace of the output beam from the MOM. The fringe spacing, as calculated from the wavelength of the output beam, was 2.7 fs, the time interval between the peaks being 57 fs. Therefore, the modulation frequency is calculated to be 17 THz (=1/57 fs). To our knowledge, this is the fastest optical device for intensity modulation of a beam. The Stokes emission has nearly the same intensity as the fundamental emission, and the output beam is fully modulated as a beat signal. When the laser wavelength was tuned, a similar result was obtained by changing the cavity length with a piezoactuator. The fundamental and Stokes beams are completely overlapped, collinear with respect to each other, since these beams are completely mode matched with a high-finesse cavity. The linewidth of the Stokes beam is reported to be ~8 kHz in Ref. 14. The finesse of the cavity is four times smaller and the laser power of the transmitted Stokes beam is two times larger in this study, relative to those reported in the paper. This suggests that the linewidth of the Stokes beam (the Schawlow-Towns linewidth that is reciprocally proportional to the laser power and to the square of the cavity finesse) is eight times broader, and providing a linewidth of ~64 kHz. The coherent time was then calculated to be ~0.016 ms and ultrastable beam modulation over a long distance (~5 km). The modulation frequency (17 THz) is passively determined by the rotational Raman shift frequency of 587 cm^{-1} for *ortho*-hydrogen, which is accurately determined as the molecular constant of the Raman medium. The bandwidth of the Raman gain is ~1 GHz, and the frequency of the Raman emission would be precisely locked at the center of the gain curve. Therefore, the modulation frequency generated by this MOM device could be used as a modulation frequency standard in the tetrahertz (THz) range. It is possible to utilize vibrational Raman scattering, in which the Raman shift frequency is 4155 cm^{-1}, using a linearly polarized fundamental beam. In this case, the modulation frequency would be increased to 125 THz. In order to change the modulation frequency, the *ortho*-hydrogen of the Raman medium can be replaced with other Raman-active molecules such as *para*-H_2, D_2, N_2, O_2, CH_4, SF_6, etc. As a result, such a MOM device can be widely utilized for frequency modulation of the laser beam in the THz range. Thus, present technology, i.e., intensity modulation of a laser beam in the period of molecular rotation or vibration, may have the potential for use in a variety of applications. For example, a phase shift arising from a coherent process could be accurately measured in the 0.1–50 fs range. On the other hand, an electron beam modulated at 17 THz can be emitted from a surface into a vacuum by focusing the modulated beam onto a semiconductor, e.g., a photocathode, such as is used in a photomultiplier (e.g., 1P28). Such a coherently modulated

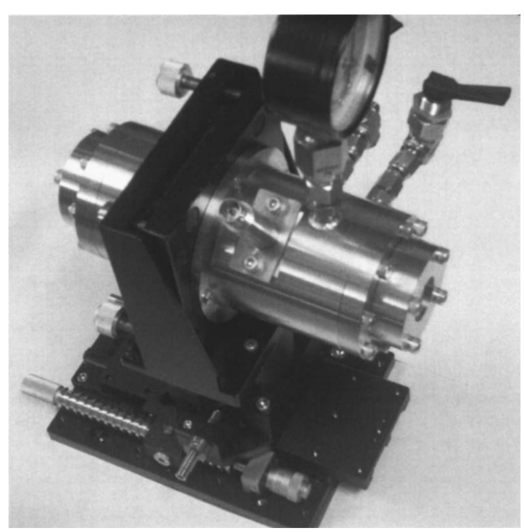

FIG. 2. Photograph of the MOM. A pair of mirrors is installed inside the stainless steel vessel. Hydrogen gas was introduced from the valve and was maintained at 10 atm throughout the experiment. The pressure was monitored using a gauge located at the top of the device. A high electric potential was applied to the piezoactuator through a high-voltage connector.

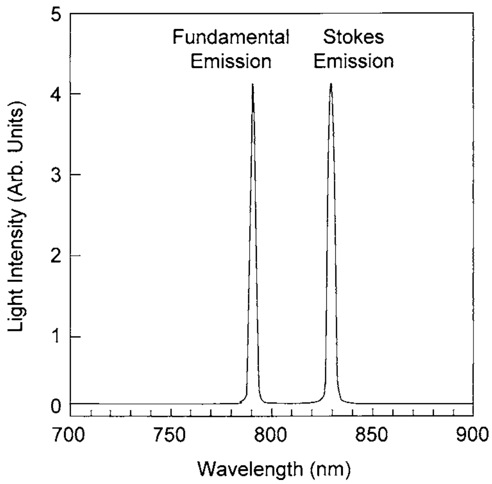

FIG. 3. Spectrum of the output beam from the high-finesse Raman cavity. The spectrum was measured by means of a multichannel spectrometer. Anti-Stokes emission was observed at 758 nm, but it was very small and could not be identified in the present scale. The signal peak for the Stokes emission is slightly saturated and then is broadened in the spectrum.

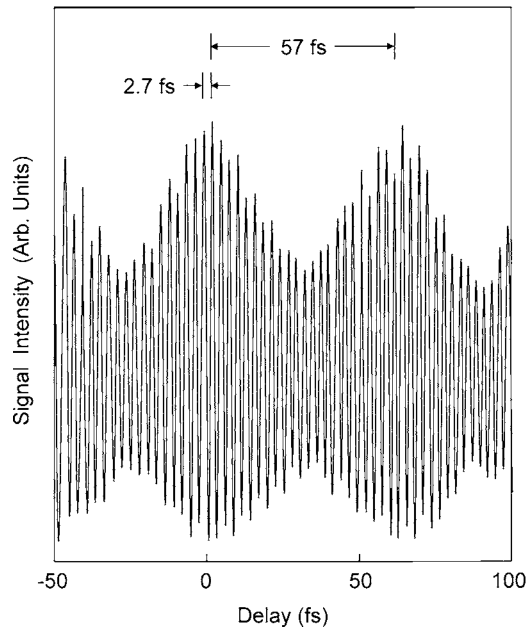

FIG. 4. Fringe-resolved autocorrelation trace of the output beam from the high-finesse Raman cavity. The spacing between the fringes is 2.7 fs, and the time period between the peaks of the sinusoidal waves is calculated to be 57 fs.

electron beam may be employed for scanning electron-beam lithography to produce an optical device consisting of periodic microstructures, like a grating. For example, such an electron beam would allow the fabrication of a substructure with a sinusoidal wave form of more than 2.5×10^8 (=0.016 ms/57 fs) times, repetitively, without any defect, the spacing of which would be absolutely determined by the molecular constant within an error of 1/17 000 (Raman gain linewidth/Raman shift frequency=1 GHz/17 THz). This corresponds to the fabrication of a 1-μm-size substructure in an area of 2.5×1 cm^2 (=1 μm$^2\times2.5\times10^8$) without any defects. The spectral linewidth of the output beam reaches the 10–100 Hz level by the isolation of vibrations experienced by the high-finesse Raman cavity.[14] This suggests that the linewidth of the Raman emission would be decreased further by a factor of ~100–1000 (~100–1000 Hz) by reducing vibrations present on the optical table. Therefore, the dimension of the device can be multiplied by a factor of 100–1000, thus allowing the fabrication of a grating-like device with a size of $\sim1\times1$ m^2. This MOM device represents a basic science tool and would also have uses in advanced industrial technology.

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