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Tunable picosecond infrared pulses generated by stimulated electronic Raman scattering of a mode-locked Ti:Sapphire laser in potassium vapor

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Abstract. A down-conversion to the mid-infrared region by using Stimulated Electronic Raman Scattering (SERS) in potassium vapor is described. The pump radiation is a frequency-doubled regeneratively amplified Ti:Sapphire laser with a pulse duration of 2 ps, pulse energy of 0.2 mJ, and repetition rate of 10 Hz. With the pumping frequency tuned around the potassium 4*s*–5*p* transition, nearly transform-limited infrared radiation tunable between 2.2 and 3.4 μm has been generated with a peak infrared energy of 12 μJ, corresponding to a quantum efficiency of 17%, and with a pulse duration of 2 ps. The present tuning range could be extended by extending the tuning range of the pump laser. In comparison, intense infrared radiation of 90 μJ energy but with a very narrow tunability around 2.9 μm has also been generated by SERS in barium vapor.

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Ultrashort high-power laser pulses tunable in the infrared region are a prerequisite for studies of fast-reaction dynamics on a surface and in condensed phase by means of ultrafast spectroscopy. Because of a lack of available tunable laser mediums in the infrared region, most tunable sources are based on downward frequency conversion of visible or near-IR lasers. Various approaches have been developed for the generation of short and intense pulses in the infrared region [1–12]. Recently, a considerable progress has been reported in extending the wavelengths and in increasing the pulse energy in the infrared region by using parametric generation or difference-frequency mixing in nonlinear crystals based on mode-locked Nd:YAG laser [4–6] and Ti:Sapphire laser [7, 8]. The mode-locked Ti:Sapphire lasers and amplifiers provide stable intense ultrashort pulses with wavelengths between 720 and 950 nm. However, when an ultrashort pulse from the mode-locked Ti:Sapphire laser is employed as a pump source in nonlinear crystals, the output infrared pulse energy has been restricted to a rather low level around 10 nJ [8], mainly due to the limited damage threshold of the nonlinear optical crystal. There have been no reports, to our knowledge, of producing intense infrared ultrashort pulses beyond 2 μm with a pulse energy of several μJ from a Ti:Sapphire laser. In this paper, we report a generation of infrared pulses tunable from 2.2 to 3.4 μm, mainly limited by the tuning range of the pump laser, with a maximum pulse energy of 12 μJ using a frequency conversion of a frequency-doubled picosecond mode-locked Ti:Sapphire laser by Stimulated Electronic Raman Scattering (SERS) in potassium vapor.

A number of recent studies have demonstrated the potential use of SERS in a metal vapor to generate infrared radiation [9–12]. Picosecond infrared pulses tunable between 3.3 and 8.4 μm [10], and between 3.3 and 4.3 μm [12] have been generated by SERS in an atomic cesium vapor pumped by a tunable picosecond dye laser. Nanasecond infrared pulses tunable from 2.8 to 16 μm have also been obtained by SERS of the cesium 6*s*–5*d* transition by using a dye laser [13].

The wide tuning range of a Ti:Sapphire laser makes it promising to be used as a pump source for generation of tunable infrared radiation by SERS. In alkali and alkaline-earth atoms, however, there are few appropriate atomic transitions for effective SERS pumped by the fundamental of Ti:Sapphire laser to generate frequencies in infrared region. The only exception is barium atom. Unfortunately, barium atom cannot be a good SERS medium for a widely tunable infrared generation, since it has a high conversion efficiency only for a very narrow range around ∼2.9 μm [14]. If the Ti:Sapphire laser is frequency doubled, all alkaline and alkaline-earth atoms could work as effective SERS media. Thus, we investigate SERS for the frequency-doubled Ti:Sapphire laser in potassium vapor, since the 4*s*–5*s* SERS wavelength falls in the 2–5.4 μm region, as has been demonstrated by using a four-wave parametric conversion [15].

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In order to get high pulse energy in the infrared region, pulses of about 10 nJ from the mode-locked Ti:Sapphire laser have been amplified by using a home-made regenerative amplifier up to 0.5 mJ with a pulse duration of 2 ps. A frequency-doubled YAG laser pumped regenerative amplifier for a picosecond mode-locked Ti:Sapphire laser is described in Sect. 1. The SERS experiments in potassium vapor are discussed in Sect. 2.

1 Picosecond regenerative amplifier

The setup is shown in Fig. 1. Our system uses the technique of chirped-pulse amplification [2] to amplify the output of a mode-locked Ti:Sapphire laser. A mode-locked Ti:Sapphire laser (Spectra-Physics, Tsunami) produces 2 ps transform-limited pulses with a repetition rate of 82 MHz, tunability of 750–840 nm and output power of 0.5 W. After passing through a Faraday isolator (OFR, IO-5-NIR), the pulse width of these pulses is stretched to 100 ps using a four-pass, grating-lens pulse stretcher, the grating being a gold-coated holographic diffraction grating (2100 lines/mm) and the lens being an achromatic lens with a focal length of 700 mm. The pulse stretching is necessary in order to prevent damages of optical components in the regenerative amplifier. The stretched pulse width is monitored with a commercial scanning autocorrelator (Spectra-Physics, Model 409).

The polarization of the stretched pulses is horizontal to make full use of the grating efficiency. It is rotated to vertical by using a pair of mirrors, before the pulses being guided to the regenerative-amplifier cavity. The cavity is ring-type, with a round-trip time of 12 ns consisted of two concave dielectric mirrors ($R = 10$ m) and two flat dielectric mirrors. In the cavity, a combination of a Pockels cell (Cleveland Crystals, QX-1020) and two thin-film polarizers are used to feed the stretched pulses into the cavity and to extract the maximum amplified pulse from the cavity by applying successively two half-wave voltages to the Pockels cell. The polarizers are set at an incidence angle of 72° reflecting 75% of *s*-polarized light and transmitting nearly 99% of *p*-polarized light over the wavelength region from 780 to 830 nm. The half-wave voltage pulse applied to the Pockels cell is synchronized to a mode-locked laser pulse to feed just one pulse into the cavity. The second harmonics of a *Q*-switched Nd:YAG laser (Spectra-Physics, GCR-12), with 30 mJ pulse energy and 7 ns pulse duration, is focused to a beam diameter of 2 mm on a 20-mm-long Brewster-cut Ti:Sapphire crystal (0.15% doped). The repetition rate of the regenerative amplification is 10 Hz.

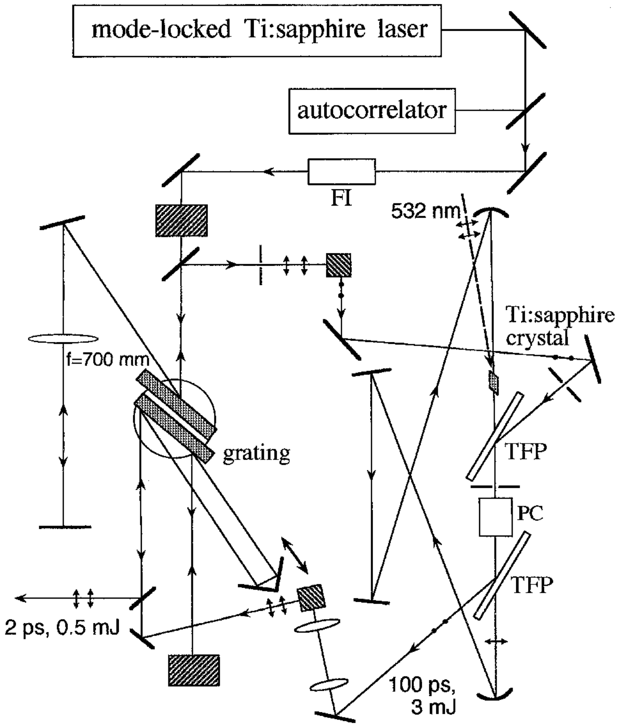

Fig. 1. Schematic diagram of the experimental setup of a regenerative-amplifier system for a mode-locked Ti:Sapphire laser. *FI*, Faraday isolator; *TFP*, thin film polarizer; *PC*, Pockels cell

The amplified pulse inside the cavity reaches the maximum in about 1 μs behind the pumping pulse. Then, the second half-wave voltage pulse is applied to the Pockels cell and, consequently, the maximum-amplified pulse is extracted from the cavity. The extracted pulse from the regenerative amplifier has an energy of ≈ 3 mJ.

The vertical polarization of the extracted amplified pulses is rotated again to horizontal by using a pair of mirrors. The pulse width of 100 ps is compressed back to about 2 ps with a four-pass grating pulse compressor with the grating identical to that used in the pulse stretcher. The output energy from the compressor has been measured to be 0.5 mJ between 780 and 830 nm and is used as a pump source in the frequency conversion to the infrared by SERS in potassium vapor.

2 SERS in potassium vapor

We used a heat-pipe oven similar to that used in [12] to get atomic vapor. The heat-pipe oven is made of a stainless-steel tube of 3.2 cm in diameter and heated over the central 25 cm region. Wicks consisting of four to five rolls of 100 mesh stainless steel are placed in the tube. In order to avoid contamination of CaF_2 windows with metal vapor, argon gas is introduced as buffer gas, and water cooling is also installed around the window section on both ends. Potassium metal was heated to 480–640°C corresponding to the atomic vapor pressures of 20–210 torr [16].

In potassium atom, the 4*s* ground state is connected to the 5*p* level at 404.5 nm ($24720\,\mathrm{cm}^{-1}$), and has the Raman-shift energy of $21027\,\mathrm{cm}^{-1}$ for the 4*s*–5*s* transition as shown in Fig. 2. To pump SERS for infrared generation in potassium vapor, the Ti:Sapphire laser must be frequency-doubled. With a type-I LiB_3O_5 crystal (35° cut, $5 \times 3 \times 8$ mm), 0.5 mJ pulse energy from the Ti:Sapphire laser was converted to Second-Harmonic (SH) light up to 200 μJ pulse energy. The SH light passed through the potassium vapor with a beam diameter of 0.5–1 mm at the center of the oven. Infrared pulse energies were measured by a pyroelectric detector (Molectron,

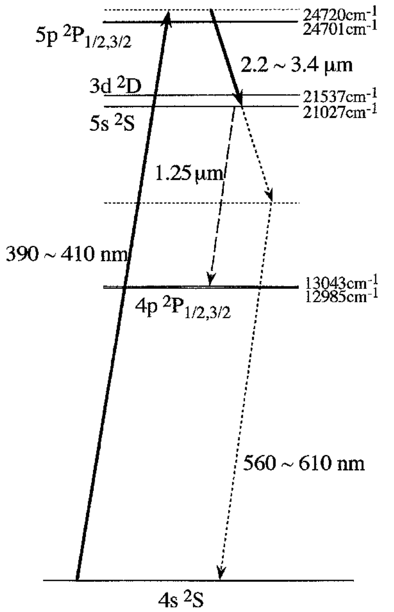

Fig. 2. Partial energy levels of atomic potassium. *Solid arrows* show the SERS transition, *dotted arrows* the additional four-wave mixing process and *dashed line* the emission at a fixed frequency

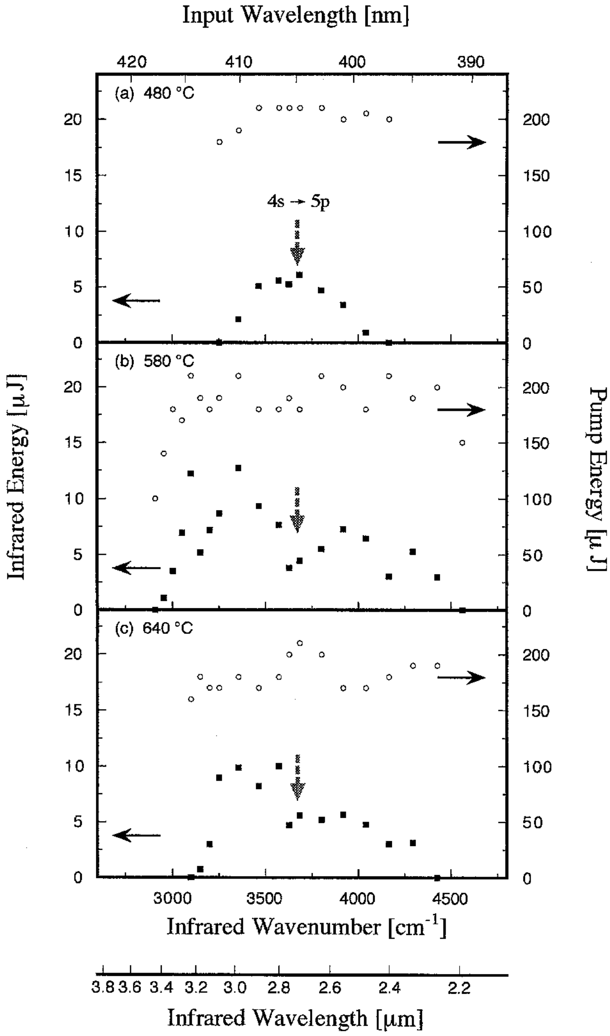

Fig. 3a–c. Infrared tuning curves obtained by SERS in potassium vapor at (**a**) 480°C, (**b**) 580°C and (**c**) 640°C, corresponding to (**a**) 20 torr, (**b**) 95 torr and (**c**) 210 torr, respectively, with an argon buffer-gas pressure of 100 torr. *Solid squares* and *open circles* show the infrared output and the pump pulse energy, respectively. *Dotted arrow* indicates the wavelength of the $4s$–$5p$ transition

J3–05) with Ge filters while the spectra of the IR output were measured with a 25 cm monochromator.

Tuning the pumping wavelength around the $4s$–$5p$ transition, the Stokes output was observed in wide wavelength and temperature ranges in potassium. The lowest thresholds for Stokes generation occurred with a buffer-gas pressure 60–120 torr. The output energies of infrared SERS, as well as the actual pump energies, as functions of Stokes IR wavelength are shown in Fig. 3 for oven temperatures of (a) 480°C, (b) 580°C and (c) 640°C, corresponding to potassium pressures of 20 torr, 95 torr and 210 torr [16], respectively, with a constant argon pressure of 100 torr filled at room temperature and with the input laser-beam diameter of 0.5 mm. In the experiment, we observed a fixed-frequency emission at 1.25 μm corresponding to the $5s$–$4p$ transition. This emission was blocked by Ge filters in the SERS energy measurements.

The SERS output was observable only for rather limited wavelength range at 480°C (Fig. 3a). The IR tuning range was maximum around the oven temperature of 580°C [Fig. 3(b)], where the SERS output was observed over the whole tuning range of our regeneratively amplified Ti:Sapphire laser (780–830 nm). The IR tuning range was measured to be 2.2–3.4 μm with a peak energy of 12 μJ at 3 μm, corresponding to a quantum efficiency of 17%. From the theoretical treatment of the transient SERS [17], the SERS threshold is increased when the pump is tuned further from resonance with the $4s$–$5p$ transitions. This effect, together with the contribution from the decrease of the pump energy, causes the gradual decrease of IR output towards the both ends of the tuning profile. When the oven temperature further increased to 640°C (Fig. 3c), the SERS efficiency decreased, which was especially prominent in the long-wavelength side of the tuning range. This is because the dimer-absorption losses are increased at higher pressure, as has been known for SERS in cesium [9, 10, 12, 13]. The diffuse absorption bands of potassium dimer have already been known in the wavelength range studied, with a stronger absorption at wavelengths longer than ~405 nm [15, 18, 19]. In a separate experiment to measure the dependence of the infrared energy on the oven temperature, the influence of dimer absorption on SERS at the pump wavelength of 413 nm was observed at temperatures as low as 550°C, which agrees with a previous report [18]. The results in Fig. 3c are thus consistent with the fact that the K_2 absorption decreases the pump radiation at a high vapor pressure and consequently the IR output is reduced abruptly at longer wavelengths. The optimum condition for SERS observed around an oven temperature of 580°C, corresponding to a potassium pressure of 95 torr, as shown by Fig. 3b, was thus attained as a result of a tradeoff between the increased gain due to the increased atomic density and the increased loss due to the dimer absorption.

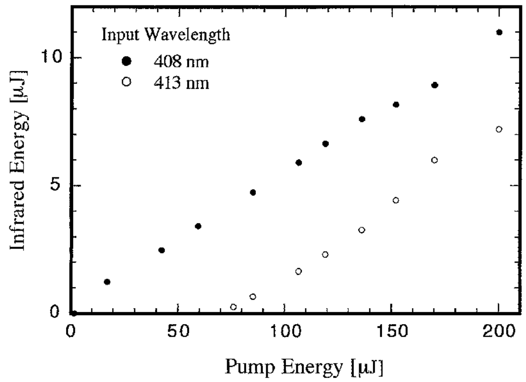

Fig. 4. IR output energy vs pump input energy for a beam diameter of 0.8 mm at input wavelengths of 408 nm (*closed circles*) and 413 nm (*open circles*) with an Ar buffer-gas pressure of 100 torr

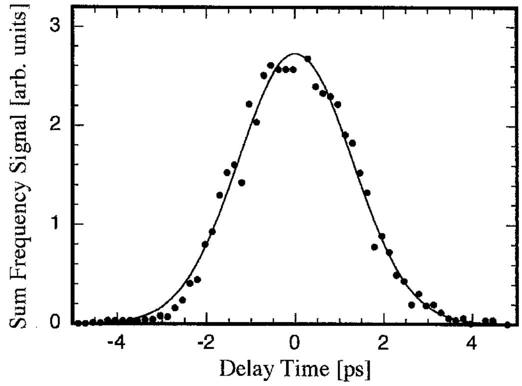

Fig. 5. A typical cross-correlation trace of the Stokes pulse at 2.87 µm mixed with the fundamental pulse of Ti:Sapphire laser at 816 nm in a thin MgO:LiNbO$_3$ crystal. *Closed circles* are experimental points and *solid line* is the Gaussian fit giving a FWHM width of 3 ps. With the Ti:Sapphire pulse width of 2.2 ps, the IR pulse width is derived to be 2 ps

We measured the SERS output energy as a function of the input energy at two input wavelengths at an oven temperature of 580°C; at 408 nm where the peak IR output was observed, and at 413 nm where the SERS threshold was strongly influenced by the dimer absorption. The results for a pump-beam diameter of 0.8 mm are shown in Fig. 4. Higher SERS threshold was obtained with a pump at 413 nm than at 408 nm. Figure 4 reveals that, beyond a threshold energy for SERS, the IR energy increased linearly with the pump energy up to our maximum pump energy available. When the pump beam was focused to a smaller spot, the SERS threshold always decreased. Reducing the pump-beam diameter, however, results in an increased nonlinear loss, which has been discussed to be caused by multiphoton ionization of atom and not by atomic ground-state depletion [9, 10]. We also observed a reduced conversion efficiency when the 200 µJ pump energy at 408 nm was focused to 0.2 mm diameter. Thus, a weakly focused pump beam was used to generate IR pulses shown in Fig. 3. For wavelength in both ends of the tuning profile, however, tight focusing of the pump radiation gave an increased IR output. Thus, the best conversion efficiency depends on the vapor pressure, the wavelength and the power density of the pump radiation. Tuning to longer wavelengths clearly requires reductions in the potassium-dimer concentration. This may be accomplished by employing a super-heated heat-pipe oven to reduce the dimer concentration by thermal dissociation as was demonstrated for cesium [10, 12].

To determine the pulse duration of the Raman Stokes, we used the cross-correlation method. In this method, the Raman-Stokes pulse was mixed with the pulse of the Ti:Sapphire fundamental to produce SFG signal at 0.59–0.67 µm in a thin MgO:LiNbO$_3$ crystal [20]. The SFG intensity varies with the delay between the two pulses and can be monitored using a SI photodiode detector after suitable bandpass filters. Figure 5 is a typical result showing a nearly Gaussian correlation trace with FWHM width of 3.0 ps. Since the auto-correlation measurement has given an FWHM width of 2.2 ps for the fundamental of Ti:Sapphire laser at 816 nm (bandwidth = 7 cm^{-1}), the IR pulse width (FWHM) at 2.87 µm was derived to be 2 ps. The corresponding IR bandwidth measured by the monochromator was 9 cm^{-1}. Therefore the time–bandwidth product of the SERS Stokes is 0.54, a value close to the Fourier-transform limit of 0.44 for a Gaussian pulse. Since the IR bandwidth of 9 cm^{-1} is close to the bandwidth of the pump radiation, the spectral broadening due to the optical Stark effect [21] is negligible in our experimental condition of weak focusing.

In addition to the $4s \rightarrow 5s$ Raman transition, we observed yellow radiation at around 560–610 nm. The yellow light is produced by a four-wave parametric mixing process as has been reported in [12] and diagrammed in Fig. 2. Our experiment confirms that the wavelength of the yellow light (ω_y) satisfied $\omega_y = \omega_L - 2 \times \omega_{\mathscr{R}}$, where ω_L is the input frequency and $\omega_{\mathscr{R}}$ is Stokes frequency. The yellow light was weak, but was visible to our eyes whenever the Stokes emission occurred. The Raman transition from $4s$ to $5d$ is in principle allowed, yet we did not observe simultaneously the infrared radiation related to this transition in accordance with the previous report [15].

For comparison and confirmation, we also worked on SERS in barium vapor, the only candidate among alkali and alkaline-earth atoms that can be pumped effectively by the fundamental of Ti:Sapphire laser. The pumping pulses of 0.5 mJ with a diameter of 2.5 mm were introduced into the oven at 980°C, corresponding to 0.6 torr barium-vapor pressure [22]. We observed SERS output at 2.92 µm ($^1S_0$–$^3D_2$) and at 2.78 µm ($^1S_0$–$^3D_1$), when the pumping wavelength was tuned to around the ^3P intermediate levels at 791.1 nm, in accordance with the previous report using nanosecond pulses [14]. We obtained a pulse energy as large as 90 µJ with a peak quantum efficiency of 33% at 2.92 µm at 100 torr argon-gas pressure. The tuning range, however, was limited to ~10 cm^{-1}, not appreciably larger than the transform limit of 2 ps pulse width, and was not increased by changing the buffer-gas pressure. The small tuning range is due to the forbidden transition employed connecting the singlet ground state to the triplet exited state.

In summary, we have demonstrated an efficient generation of nearly transform-limited picosecond IR pulses of several µJ energy and tunable over 2.2–3.4 µm from a regeneratively amplified mode-locked Ti:Sapphire laser by SERS in potassium vapor. The experimental arrangement as described could be improved to extend the tuning range and to increase the IR output energy in a number of ways: reduction of dimer concentration by employing a super-heated heat-pipe oven [10, 13], extension of the tuning range of the pump laser, and increase of the pump energy by a further amplification of regeneratively amplified pulses. Such improvements could readily be carried out.

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