

## Raman self-conversion of femtosecond laser pulses and generation of single-cycle radiation

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Stimulated Raman scattering of light in the regime of laser pulse durations shorter than the period of Raman-active vibrational mode  $2\pi/\Omega_R$  is investigated. Using full equations for the field and the medium polarization, we obtain an insight into the stage of frequency conversion where the slowly varying amplitude approximation is inapplicable. It is found that the laser pulse experiences a continuous downshift of its spectrum, resulting in the formation of an intense single-cycle electromagnetic pulse. The role of the competing processes is analyzed. [S1050-2947(98)51207-3]

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Since the discovery of stimulated Raman scattering (SRS) this fundamental nonlinear optical phenomenon has been studied in great detail both for the case of quasistationary interaction [1] and for laser pulses shorter than the vibrational relaxation time [2]. In the last few years the interaction of intense femtosecond laser pulses with matter has been the focus of attention because of the many novel nonlinear-optical phenomena that have been observed. Appropriate methods of theoretical description have been developed [3].

The purpose of this Rapid Communication is to show that the SRS with pulses of femtosecond duration differs qualitatively from the well-known (and commonly accepted) physical picture of SRS. We also demonstrate that the use of full equations for the pulse field, instead of equations for the field slowly varying amplitudes and phases (SVAPs), plays a key role in the adequate treatment of the processes underlying femtosecond-pulse SRS. The problem is the following: Typical values of the vibrational period  $T_R = 2\pi/\Omega_R$  corresponding to the Stokes frequency shift  $\Omega_R$  in many Raman-scattering media are of the order of  $10^{-13}$ – $10^{-14}$  s. This means that for laser pulses in the femtosecond region the condition

$$\tau_p < T_R = 2\pi/\Omega_R \quad (1)$$

can be easily met, implying a qualitatively different regime of SRS. Indeed, in traditional SRS with nano- and picosecond pulses, the presence of a seeding at the Stokes frequency (normally, of spontaneous noise) is necessary for the SRS process to develop. The spectrum of a femtosecond pulse with a duration (1) initially contains frequency components satisfying the conditions of Raman-type resonance (see Fig. 1). Therefore only one ultrashort pulse is required to initiate the SRS process. The importance of this specific (impulsive) mechanism of medium excitation for spectroscopy of elementary processes in matter was first discussed in [4], where femtosecond-pulse excitation of coherent optical phonons in liquids was demonstrated. Here, we point out that the dynamics of a laser pulse under the condition (1) is rather unusual. In contrast to the nano- and picosecond-pulse SRS [1,2], the SRS process with pulses (1) has no intensity threshold, and one could expect the SRS process to occur until a major part of the pump femtosecond-pulse energy is transformed into vibrational excitation (that would lead to generation of spectral components far from the input pulse

frequency). Since the process appears as a continuous self-conversion of the pulse spectrum, the use of the SVAP method would imply an analysis of nonlinear interaction of an infinite number of Stokes and anti-Stokes Fourier components of the field. Thus, the problem of femtosecond-pulse SRS cannot be resolved in terms of the traditional theory of SRS [1] based on the SVAP approximation and a first-principles approach is necessary. Although some aspects of SRS with femtosecond pulses have already been discussed in the literature [4–6], the analysis was limited by the assumption of a relatively small (compared to the pulse carrier frequency) change of the pulse spectrum, and by neglecting such competing effects as medium dispersion and electronic nonlinearity, which are often dominant on the femtosecond time scale [7].

Starting from general assumptions about linear and nonlinear properties of a SRS medium, we investigate the SRS process in condition (1) beyond the scope of the SVAP approximation. On the basis of numerical solution of the equations of pulse dynamics we show that the well-known picture of cascade generation of discrete Stokes (anti-Stokes) frequency components transforms, on the femtosecond-time scale, into a continuous self-conversion of laser pulse spectrum towards lower frequencies. The ultimate form of the generated laser field represents a single-cycle electromagnetic pulse. The nature of the self-conversion mechanism differs significantly from that of the soliton self-frequency shift in fibers [8,9], where the effect is quasistationary in nature and therefore depends on the structure and width of the Raman line. The discussed coherent mechanism of SRS

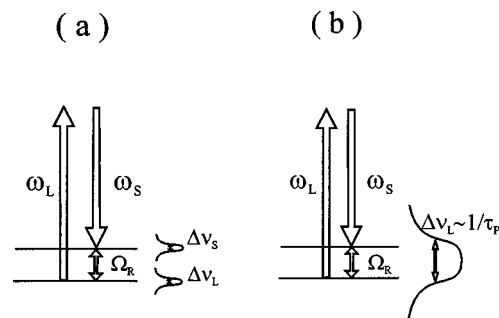


FIG. 1. Schematic diagram of the SRS process: (a) long excitation pulses (traditional version of SRS); (b) ultrashort excitation pulse.

is not sensitive to these factors but depends essentially on all the laser-molecule interaction history on the time scale of one molecular vibration.

To analyze this extreme regime of SRS we start from the following physical model. Consider a linearly polarized laser pulse  $\mathcal{E}(z, t) = (2\pi)^{-2} \int dk \int d\omega E_{\omega k} \exp i(\omega t - kz)$  propagating in the  $z$  direction in an isotropic medium. The pulse evolution is described by the wave equation

$$\frac{\partial^2 \mathcal{E}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 P}{\partial t^2}, \quad (2)$$

where  $P$  is the field-induced medium polarization, which can be written in the following form:

$$\begin{aligned} P(t) = & \int_0^\infty \theta^{(1)}(\tau) \mathcal{E}(t-\tau) d\tau + \int_0^\infty d\tau \int_0^\infty d\tau' \int_0^\infty d\tau'' \theta^{(3)} \\ & \times (\tau, \tau', \tau'') \mathcal{E}(t-\tau) \mathcal{E}(t-\tau-\tau') \mathcal{E}(t-\tau-\tau'-\tau'') \\ & + P_{\text{Ram}}. \end{aligned} \quad (3)$$

The first term ( $P_L$ ) on the right-hand side of Eq. (3) describes the linear part of the medium response, the second one ( $P_{\text{electr}}$ ) is due to the third-order processes in the medium except for the Raman process; the response functions  $\theta^{(1)}$  and  $\theta^{(3)}$  lead, in the frequency domain, to the medium first-order ( $\chi^{(1)}$ ) and third-order ( $\chi^{(3)}$ ) polarizability. The last term  $P_{\text{Ram}} = N(\partial\alpha/\partial Q)Q\mathcal{E}$  accounts for the Raman contribution of molecules of an effective concentration  $N$ . This form of  $P_{\text{Ram}}$  implies that the field spectral components are far from electronic transitions. In this case the expectation value  $Q$  of the normal mode operator of the molecule obeys the equation

$$\frac{\partial^2 Q}{\partial t^2} + \frac{2}{T_2} \frac{\partial Q}{\partial t} + \Omega_R^2 Q = \frac{1}{2M} \frac{\partial \alpha}{\partial Q} \mathcal{E}^2. \quad (4)$$

The model equation (first introduced in [1]) represents a linear oscillator with a reduced mass  $M$ , a natural frequency  $\Omega_R$ , and a relaxation time  $T_2$ , driven by the force quadratic in the field  $\mathcal{E}$ . For simplicity we consider isotropic Raman transitions, so that the tensor  $(\partial\alpha/\partial Q)$ , which stands for the change of the molecule linear polarizability with respect to the vibrational coordinate  $Q$ , is reduced to a scalar quantity  $(\partial\alpha/\partial Q)$  [2]. If the excited vibrational level population is much less than unity, we arrive at Eq. (4). We assume that the molecule is not active in infrared (IR) absorption and its electronic frequencies  $\omega_{mn} = (E_m - E_n)/\hbar$  (lying typically in the vacuum ultraviolet region) are sufficiently high compared to the pulse characteristic frequency  $\omega$ . In this case the linear part ( $P_L$ ) of the response (3) can be approximated by

$$P_L(t) = \chi(0) \mathcal{E} - \frac{1}{2} \chi_{\omega\omega}(0) \frac{\partial^2 \mathcal{E}}{\partial t^2}, \quad (5)$$

where  $\chi(0)$  is the medium linear polarizability  $\chi(\omega)$  in the limit  $\omega \rightarrow 0$ , and  $\chi_{\omega\omega}(0) = (\partial^2 \chi / \partial \omega^2)_{\omega=0}$ . The second term in Eq. (5) describes the positive medium dispersion. The small ratio  $(\omega/\omega_{mn})$  also implies that the nonlinear part of

the electronic response ( $P_{\text{electr}}$ ) follows adiabatically the value of the pulse field. This allows the following approximation:

$$P_{\text{electr}}(t) = \chi_e^{(3)}(0) \mathcal{E}^3, \quad (6)$$

where  $\chi_e^{(3)}(0)$  denotes the third-order electronic susceptibility in the low-frequency limit.

First we obtain a qualitative insight into the laser-molecule interaction under condition (1) and consider the SRS process in the limit  $\tau_p \ll 2\pi/\Omega_R$ . In this case terms  $\Omega_R^2 Q$  and  $(1/T_2) \partial Q/\partial t$  in the material Eq. (4) can be neglected in comparison to  $\partial^2 Q/\partial t^2$ . Assuming that the laser pulse appears at the moment  $t=0$  and is nonzero within the time interval  $0 < t < \tau_p$ , we find that during the laser-molecule interaction ( $0 < t < \tau_p$ ) the vibrational coordinate changes as

$$Q(t) \approx \frac{1}{2M} \frac{\partial \alpha}{\partial Q} \int_0^t dt' \int_0^{t'} dt'' \mathcal{E}^2(t'') \quad (7a)$$

and shows a very slow response to the field. After the interaction, e.g., for  $t \gg \tau_p$ , the nuclear coordinate exhibits freely damping oscillations

$$Q(t) \approx \frac{2\pi}{Mc\Omega_R} \frac{\partial \alpha}{\partial Q} W \exp(-t/T_2) \sin(\tilde{\Omega}_R t), \quad (7b)$$

where  $W = (cn/4\pi) \int_{-\infty}^\infty \mathcal{E}^2 dt$  is the pulse energy,  $n = [1 + 4\pi\chi(0)]^{1/2}$  and  $\tilde{\Omega}_R^2 = \Omega_R^2 - (1/T_2)^2$ . The displacement of the nuclear coordinate (7a) during the laser pulse action is seen to be much smaller [ $\sim (\tau_p/T_R)$ ] than the amplitude of subsequent free oscillations (7b). In fact, the molecule acquires some initial velocity of vibrational motion, but its coordinate has no time to change.

For a quasimonochromatic field of a frequency  $\omega \gg \Omega_R$ , the initial stage of short pulse ( $\tau_p \ll T_R$ ) evolution can be treated as a self-phase modulation process. By passing in Eqs. (2)–(6) to the ‘‘slow’’ amplitude  $E = |E| \exp(i\varphi)$  of the field and using Eq. (7a), we find that the centrum of the

spectrum  $\varpi = \int_{-\infty}^\infty (\omega + \dot{\varphi}) |E|^2 d\tau / \int_{-\infty}^\infty |E|^2 d\tau$  moves towards lower frequencies:

$$d\varpi/dz = -\beta\omega W. \quad (8)$$

The inertial character of the SRS process is manifested by the fact that the shift (8) is determined by the pulse energy  $W$  rather than its intensity. In Eq. (8) the energy  $W$  is expressed in units of the laser energy  $w_0 = [2\hbar\Omega_R M c^2 n^2]^{1/2} [2\pi(\partial\alpha/\partial Q)]^{-1}$  required to transmit to the molecule the energy of one vibrational quantum  $\hbar\Omega_R$ . The reciprocal  $Z_{\text{SRS}} = \beta^{-1}$  of the constant  $\beta = 2\pi N(\partial\alpha/\partial Q)[\hbar\Omega_R/Mc^2 n^2]^{1/2}$  can be treated as the Raman self-conversion length.

Consider now the pulse evolution at propagation distances where the change of the pulse spectrum and the energy is significant. It is convenient to introduce the parameter,

$$\gamma = \chi_e^{(3)} \left/ \frac{N(\partial\alpha/\partial Q)^2}{M\Omega_R^2} \right., \quad (9)$$

which characterizes the ratio of the third-order electronic contribution to the impulsive Raman contribution. The value of parameter  $\gamma$  can be measured experimentally [7,9] and

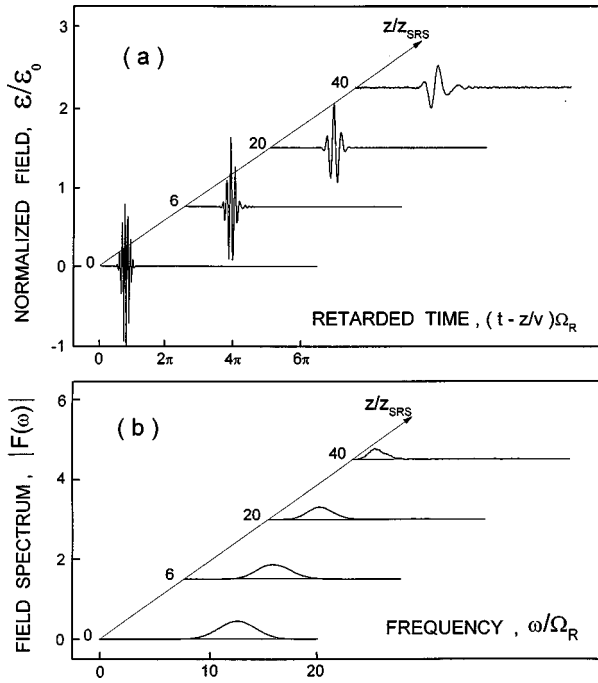


FIG. 2. Evolution of a femtosecond laser pulse in a SRS medium (the case of small electronic contribution,  $\gamma=0.25$ ): the laser field temporal distribution (a) and spectrum (b) at different propagation lengths  $Z$ . The field is normalized by  $\mathcal{E}_0=(w_0\Omega_R/2\pi c)^{1/2}$ ; the distance by  $Z_{\text{SRS}}=\beta^{-1}$ . The input pulse parameters are  $\tau_p\omega=24$ ,  $\tau_p\Omega_R=1.5$ , and  $W(0)=0.1$ . For a SRS medium with  $\Omega_R\sim 10^3\text{ cm}^{-1}$ ,  $(\partial\alpha/\partial Q)\sim 10^{-15}\text{ cm}^2$ ,  $M\sim 10^{-22}\text{ g}$ , and  $N=10^{21}\text{ cm}^{-3}$ , the normalization units are  $\mathcal{E}=5\times 10^7\text{ V/cm}$  and  $Z_{\text{SRS}}=0.1Z_D=0.2\text{ cm}$ .

estimated from the value of Raman gain. In different materials  $\gamma$  varies in a wide range: in optical fibers  $\gamma\approx 4$  [9]; in molecular gases  $\gamma\sim 1$  [7]; for some liquids (e.g.,  $\text{CS}_2$ ) the relative contribution of the third-order electronic response is estimated to be only  $\gamma\sim 0.1$  [10].

In Figs. 2(a,b) are shown the results of the numerical solution of the full system of equations (2)–(6) for the pulse electric field  $\mathcal{E}$  in the case of a dominating SRS process ( $\gamma=0.25$ ). As can be seen, the pulse evolution is characterized by the monotonic downshift of the center of the pulse spectrum and by the energy depletion due to medium excitation. Interestingly, at propagation distances where the pulse central frequency becomes as low as the pulse spectral width ( $\Delta\omega_p\sim 1/\tau_p$ ), the laser field represents in fact a single-cycle electromagnetic pulse. Therefore, the SRS process under the condition (1) provides a mechanism of laser field “rectification.” For the rectification process to be efficient condition (1) has to be held over all the interaction distance. This means that the characteristic rectification length  $Z_0$  has to be smaller than the group-velocity dispersion length  $Z_D$ . Surprisingly, however, in the discussed situation of a normally dispersive SRS medium, the dispersion in combination with the Raman nonlinearity has a stabilizing effect on the pulse shape [Fig. 2(a)]. This is due to the fact that the SRS process under condition (1) is inertial in nature; therefore the pulse instant frequency decreases from the pulse leading to its trailing edge. In a positively dispersive medium the lower-

frequency components travel faster than the higher-frequency components, and this results in an additional pulse temporal compression, allowing effective interaction over the propagation lengths that are substantially longer than the dispersion spreading length  $Z_D$ .

Let us estimate the efficiency of laser energy transformation into the single-cycle pulse. By inserting solution (7a) into wave equation (2), one can find that the pulse energy  $W$  changes as

$$dW/dz = -\beta W^2. \quad (10)$$

If we now turn to Eq. (8) and consider the pulse frequency  $\omega$  and the energy  $W$  on the right-hand side of Eq. (8) to be functions of the propagation length  $z$  with  $\omega(z)\approx\varpi(z)$  and  $W(z)$  determined by Eq. (10), we readily find that the pulse “effective  $\varpi(z)$  frequency” is always proportional to the pulse energy  $W(z)$ :

$$\varpi(z)/\varpi(0) = W(z)/W(0) = 1/[1 + \beta W(0)z], \quad (11)$$

where  $\varpi(0)$  is the input laser frequency  $\omega$ . From Eq. (11) it follows that the efficiency of the laser energy transformation into the single-cycle pulse with  $\varpi\sim 1/\tau_p$  is of the order of  $\eta\sim(\tau_p\omega)^{-1}$  and for  $\tau_p\sim 10^{-14}\text{ s}$  and  $\omega\sim 10^{15}\text{ s}^{-1}$  can be as high as several percent. The remarkable fact that during the interaction the photon number  $W(z)/h\varpi(z)$  is kept constant suggests that in the discussed interaction regime a successive Stokes “processing” of the field photons occurs.

In the case of  $\gamma\geq 1$  the pulse dynamics differs significantly, because of the influence of Kerr self-phase modulation and the contribution of the third-order sum-frequency generation process. We have found, however, that an effective down-conversion process and a single-cycle pulse generation can be realized for a femtosecond pulse traveling with a certain time delay after the first (pump) pulse and interacting with the vibrational mode coherently excited by the pump pulse. The initial stage of the process can be analyzed using the SVAP method. Let an intense pump pulse with the complex amplitude  $E_1=|E_1|\exp(i\varphi_1)$  and frequency  $\omega_1$  be followed, with a delay  $\Delta\tau>\tau_p$ , by an injection pulse  $E_2=|E_2|\exp(i\varphi_2)$  of frequency  $\omega_2$ . Assuming that the injection pulse only slightly disturbs the molecular free vibrational motion (7b) produced by the pump pulse and  $T_2\gg T_R$ , for the change of the injection pulse central frequency  $\varpi_2$  we have

$$\frac{d\varpi_2}{dz} = -2\beta\omega_2 W_1 \exp(-\Delta\tau/T_2) \cos(\Omega_R\Delta\tau). \quad (12)$$

From Eq. (12) it follows that the injection pulse experiences a continuous shift of its central frequency proportional to the pump pulse energy  $W_1$ . Depending on the value of the delay time  $\Delta\tau$ , the shift can be both negative (down-conversion) and positive signed (up-conversion). Particularly, the efficiency of injection pulse down-conversion reaches its maximum if the delay time is a multiple of the molecular oscillation period; e.g.,  $\Delta\tau=2\pi\Omega_R^{-1}k$  ( $k=1,2,3,\dots$ ). The corresponding value of the injection pulse downshift is seen to differ from that of the pump pulse (8) by the factor  $2(\omega_2/\omega_1)$  on the order of 1. Therefore, for a sufficiently weak injection pulse  $E_2$ , the fast electronic contribution  $\sim(E_2)^3$  will be small compared to the contribution  $\sim E_2 Q$  due to SRS on the coherent vibrations  $Q$  excited by the in-

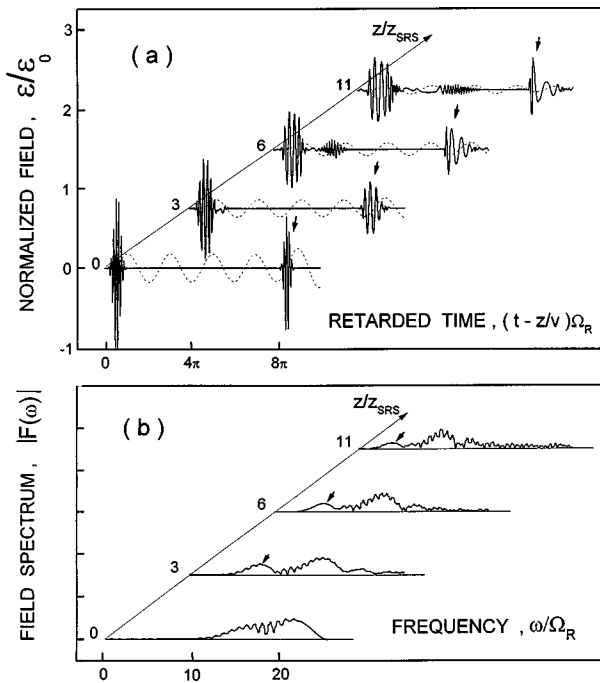


FIG. 3. Dynamics of SRS in the delayed-pulse propagation regime (the case of large electronic contribution,  $\gamma=2$ ). The field temporal distribution (a) and spectrum (b) at different propagation lengths  $Z$ . The dashed line shows the temporal dynamics of the normal coordinate  $Q$ . The injection pulse field and spectrum are shown with arrows. The input pulse parameters are  $\tau_{p1}\omega_1=28$ ,  $\tau_{p1}\Omega_R=1.5$ ,  $\tau_{p2}\omega_2=21$ ,  $\tau_{p2}\Omega_R=1.3$ ,  $W_1(0)=0.2$ , and  $W_2(0)=0.05$ .

tense pump pulse  $\mathcal{E}_1$ . These qualitative arguments are supported completely by our numerical results [Figs. 3(a,b)], which demonstrate the propagation dynamics of an intense pump and a weaker injection pulse for the case of  $\gamma=2$  and a delay time  $\Delta\tau=3T_R$ . The evolution of the two pulses in the SRS medium differs dramatically. While the pump pulse in the SRS medium shows a significant superbroadening of its spectrum both to the red and to the blue, the second pulse interacting with the impulsively excited molecular vibrations experiences predominantly a spectral shift towards lower frequencies that leads to a generation of a single-cycle IR pulse.

Let us estimate the laser pulse parameters required for experimental observation of the discussed effects in a high-pressure gas of homonuclear diatomic molecules. From Eq.

(11) it follows that the length  $Z_0$  of laser frequency rectification down to the single-cycle pulse [with  $\varpi(Z_0)\sim 1/\tau_p$ ] is about  $Z_0\sim(\omega\tau_p)/[\beta W(0)]$ . Assuming molecular frequency  $\Omega_R\sim 1000\text{ cm}^{-1}$  ( $T_R\approx 30\text{ fs}$ ), effective mass  $M\sim 10^{-22}\text{ g}$ , polarizability  $(\partial\alpha/\partial Q)\sim 10^{-15}\text{ cm}^2$ , for a laser pulse with  $\tau_p=10\text{ fs}$ , photon energy  $\hbar\omega=1\text{ eV}$ , and fluence  $I=10\text{ TW/cm}^2$ , the characteristic length of single-cycle pulse formation can be found to be  $Z_0\approx 100\text{ cm/atm}$ . The value of group-velocity dispersion of the IR laser pulse in a gas of molecules with a typical energy of electronic transition  $I_e\sim 10\text{ eV}$  can be roughly estimated by  $k_{\omega\omega}\sim 0.1\text{ fs}^2/\text{cm}$  per 1 atm pressure. Thus, for a pressure  $p=30\text{ atm}$  the single-cycle pulse is formed at a propagation distance  $Z_0\approx 3\text{ cm}$ , which is still short compared to the pulse dispersion length  $Z_D\approx 30\text{ cm}$ . The field strength in the generated single-cycle pulse is about  $\mathcal{E}\sim 10^7\text{ V/cm}$ . These estimations show that the femtosecond-pulse SRS is interesting from the viewpoint of generation of intense IR electromagnetic transients. Unlike the existing THz technique based on far infrared (FIR) generation in quadratic crystals [11], the discussed frequency conversion mechanism requires no phase matching at all, because there is a continuous self-transformation of the laser pulse spectrum rather than a generation of discrete frequencies. We also note that some evidence for the predicted features of the femtosecond-pulse SRS was observed in our recent experiment [12].

In conclusion, we have shown that SRS with pulses shorter than the period of vibrational motion leads to a physical picture qualitatively different from the traditional one: the stimulated Raman effect appears as a continuous self-transformation of the pulse spectrum towards lower frequencies. The ultimate wave form of the generated field represents a FIR pulse containing only one oscillation period of the electromagnetic wave. The specifics of SRS has been revealed by treating the Raman process beyond the scope of the SVAP approximation. Because of the universality of Raman-type nonlinearity, the predicted features of SRS should be observable in a wide class of nonlinear materials, including gases, liquids, solids, and plasmas. The discussed mechanism may also be interesting as an alternative approach to the generation of femtosecond-pulse tunable IR radiation down to single-cycle electromagnetic transients.

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