Mechanisms of spectral shift in ultrashort-pulse laser oscillators

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A number of factors that influence spectral position of ultrashort pulses in mode-locked lasers have been identified: high-order dispersion, gain saturation, reabsorption from the ground state, and stimulated Raman scattering. Using the one-dimensional numerical model for the simulation of the laser cavity, we analyze the relative contributions of different factors to the spectral position of the mode-locked pulses using the example of the Cr:LiSGaF laser. In this case the Raman effect provides the largest self-frequency shift from the gain peak (up to 60 nm), followed by the gain saturation (≈25 nm), whereas the high-order dispersion contribution is insignificant (≈5 nm). The results of the simulation are in good agreement with experimental data, confirming that stimulated Raman scattering is the dominant mechanism that causes the pulse self-frequency shift. © 2001 Optical Society of America

1. INTRODUCTION

Progress in the ultrashort-pulse-generation technique allows the reaching of 14-fs pulse duration in Cr\(^{3+}\):LiSrGaF\(_6\) (Cr:LiSGaF)\(^1\) and Cr\(^{4+}\):MgSiO\(_4\) (Cr:forsterite)\(^2\), as well as 12 fs in Cr\(^{3+}\):LiSrAlF\(_6\) (Cr:LiSAF).\(^3\) All of these active media are attractive due to possibilities of sub-20-fs pulse generation directly from the diode-pumped laser. At the same time the lasers demonstrate a significant Stokes shift of the pulse spectrum at such short pulse widths.\(^1\)\(^-\)\(^5\) This shift decreases the accessible bandwidth due to the worse overlap of the gain and the pulse spectra, thus setting the limit to the efficiency and the pulse duration.\(^5\) Therefore investigation of the nature of the ultrashort-pulse spectrum transformation has not only academic but also practical significance.

A number of explanations for the ultrashort-pulse spectrum shift in mode-locked lasers have been suggested. For example, it was supposed that, due to the high-order dispersions, the spectral region of the negative group-delay dispersion, which is optimal for pulse formation, may be displaced.\(^6\) However, in the framework of the perturbation theory the third-order dispersion does not contribute to the pulse carrier frequency but results in the spectrum asymmetry and fragmentation.\(^7\) Only in the case of the non-steady-state operation is there a possibility for a strong dynamic frequency shift.\(^8\)

Another explanation of the frequency shift takes into account frequency filtering due to reabsorption in the gain medium.\(^3\) However, as it is shown below, this explanation cannot explain the dependence of the frequency shift on the pulse energy. Moreover, the latter dependence suggests that a nonlinear mechanism is involved in the frequency shift.

As it was shown in Ref. 9, stimulated Raman scattering in the active medium can produce the experimentally observed Stokes frequency shifts in the sub-50-fs domain. However, the analysis in Ref. 9 was based on the soliton-perturbation theory, which did not allow taking into account the effect of the high-order dispersion and such frequency-dependent dissipative lasing factors as the asymmetric gain band, spectral filtering due to the output mirror, reabsorption in the gain medium, and gain saturation.

In this study we performed numerical analysis of the spectral characteristics of the ultrashort pulses on the basis of a relatively simple model of Kerr-lens mode locking (KLM). The main advantages of our model are taking into consideration the high-order dispersion, the exact profiles of the loss and gain bands, the frequency-dependent reabsorption in the active medium, the gain saturation, and the fast absorber action of Kerr lensing. The modeling is performed in an example of a mirror-dispersion-controlled KLM Cr:LiSGaF laser, which demonstrates a significant Stokes shift and for which we had extensive spectroscopic and experimental data. The obtained results are in a good agreement with the experiment and allow us to estimate the contribution of the different factors to the spectral position of the ultrashort pulses.

The paper is organized as follows: first, a summary of the relevant measurements and experimental results is given, followed by the construction of the analytical and computational models. Then we present the results of our calculations and discuss the influence of each of the above-mentioned factors separately. Finally, we conclude with the results of simulation with all factors included, using distributed and discrete-element models.
2. EXPERIMENTAL OBSERVATIONS

The systematic Stokes shift of the ultrashort mode-locked pulse, to our knowledge, was first reported in 1997 in Cr:LiSGaF- and Cr:LiSAF-based KLM oscillators.\(^1\)\(^4\)\(^10\) Stimulated Raman scattering has been suggested as a possible mechanism that causes the shift. It has been found that (i) the shift could be observed in oscillators with different dispersion characteristics, (ii) the shift increases with the pulse energy, and (iii) the shift increases with pulse shortening (see, e.g., Refs. 1 and 5). At pulse durations <20 fs the peak of the pulse spectrum may shift as far as 50–70 nm into the infrared as compared with the cw wavelength or the mode-locked spectrum at long pulse durations (~840 nm in Cr:LiSGaF).\(^1\) Later, analogous shifts in Cr:LiSAF were reported by Uemura and Torizuka\(^3\) and R. Gäbel et al.\(^11\) Recently, a significant shift has also been observed in a Cr:forsterite laser.\(^2\)

![Schematic diagram of the laser oscillator](image1)

Fig. 1. General scheme of the KLM Cr:LiSGaF laser used in this paper. This scheme directly corresponds to the experiments in Refs. 1 and 4: HR, high reflector; CM, chirped mirror; OC, output coupler.

![Measured group-delay dispersion](image2)

Fig. 2. Measured group-delay dispersion of the active media (8 mm in double pass), output coupler, and chirped mirrors in dependence on the wavelength \(\lambda\).

![Dispersion curves](image3)

All the experiments mentioned used a common optical scheme, differing only in pump arrangements and dispersion-compensation techniques. The schematic diagram of the laser oscillator is shown in Fig. 1. This is a representative scheme for any X-shaped KLM laser because the different types of dispersion compensation can always be represented by lumped dispersion of a chirped mirror. For modeling purposes we used the parameters of the experiments, reported in Refs. 1 and 10: the Brewster-angled 4-mm-long LiSGaF crystal doped with 1.5% Cr and the beam diameter in a Cr:LiSGaF crystal of 40 \(\mu\)m \(\times\) 60 \(\mu\)m. The high reflectors (HRs) had negligible dispersion. The dispersion of the chirped mirrors (CMs) has been calculated from their original design and additionally measured by white-light interferometry. The intracavity dispersion was calculated with the dispersion data of Cr:LiSGaF\(^4\) the measured dispersion curves of the chirped mirrors, and the calculated dispersion of the output coupler (Fig. 2). We also used the experimental loss spectra (the dimensionless round-trip mirror transmission and the double-pass ground-state absorption in Cr:LiSGaF) as shown in Fig. 3.
The laser was pumped by 1.2–1.5 W from the Kr⁺-ion laser at 647 nm, having a TEM₀₀ beam, of which 0.9–1.1 W is absorbed in the active medium, generating 60–100 mW of average output power in the mode-locked regime. The resonator was slightly asymmetric, with distances between the curved mirrors (radii of curvature 100 mm) and the end mirrors being 88 and 109 cm, corresponding to the 72-MHz pulse-repetition rate. Mode-locking was achieved primarily by a hard aperture in the form of an adjustable slit close to the output coupler (Fig. 1). Figure 4 shows normalized output spectra at different pulse durations and energies, demonstrating the spectral shift (see Table 1). We observed similar spectral behavior also in Cr:LiSAF oscillators. However, for the sake of simplicity we provide experimental data and perform simulations using the data for Cr:LiSGaF only.

The Raman gain spectrum of the undoped LiSGaF crystal was measured according to the procedure described in Ref. 10, with the orientated crystalline quartz as a reference and taking into account the thermal phonon population factor. The spectrum (see Fig. 5) is obtained from the spontaneous Raman-scattering spectrum, recorded in the confocal backscattering geometry. Both the incident and the scattered light is polarized along the c axis, corresponding to the polarization of light in the laser. The LiSGaF crystal possesses the spatial symmetry group D²₃d(P31c) with two formula units in a unit cell, resulting in total of 32 optical phonon modes 3A₁₉ + 4A₂₉ + 4A₁₉ + 5A₂₉ + 8E₉ + 8E₈, of which 3A₁₉ + 8E₉ are Raman active. In the scattering geometry as described above, only three full-symmetric A₁₉ modes are visible (Fig. 5), with relevant parameters given in Table 2.

### Table 1. Pulse Durations and Pulse Energies for Fig. 4

<table>
<thead>
<tr>
<th>Curve in Fig. 4</th>
<th>tₚ (fs)</th>
<th>E (nJ)</th>
</tr>
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<tr>
<td>1</td>
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<td>75</td>
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<tr>
<td>2</td>
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<td>83</td>
</tr>
<tr>
<td>7</td>
<td>14</td>
<td>105</td>
</tr>
</tbody>
</table>

### Table 2. Raman Gain of Undoped LiSGaF

<table>
<thead>
<tr>
<th>Frequency (cm⁻¹)</th>
<th>Raman Gain gₛ (cm/GW)</th>
<th>Width (FWHM) (cm⁻¹)</th>
<th>T_j (ps)</th>
</tr>
</thead>
<tbody>
<tr>
<td>230</td>
<td>0.014 ± 0.005</td>
<td>9 ± 3</td>
<td>1.2 ± 0.4</td>
</tr>
<tr>
<td>349</td>
<td>0.021 ± 0.006</td>
<td>14 ± 2</td>
<td>0.7 ± 0.1</td>
</tr>
<tr>
<td>551</td>
<td>0.32 ± 0.05</td>
<td>12.5 ± 0.6</td>
<td>0.86 ± 0.05</td>
</tr>
</tbody>
</table>

### 3. MODEL

Different approaches exist for modeling of ultrashort-pulse generation in solid-state lasers, which are based either on soliton or fluctuation models. The soliton approach can be applied only to the distributed laser model (where the dispersion and the nonlinearity are implied to be evenly distributed over the round trip and to act simultaneously) but allows us to build a comparatively simple analytical description, thus promoting easy interpretation of results. To overcome the limitations of the soliton approach, we used numerical simulations according to the fluctuation model, allowing us to account for the high-order dispersion, the laser-field reabsorption, the complicated spectral profiles of the gain and the output-coupler transmission, and the induced Raman scattering within the active medium. For simplicity, we based our calculations on the distributed laser model, but the results were tested by simulations on the basis of the discrete-element scheme corresponding to Fig. 1. The field evolution was simulated numerically within the fixed-time window.
where $g$ is the modulation depth (KLM loss), set by the cavity arrangement, $\sigma$ is the inverse loss saturation intensity, and $\alpha$ is the field depending on the local time $t$ and the longitudinal coordinate $z$ (\(|\alpha|^2\) has the meaning of the field intensity). Parameters $\gamma$ and $\sigma$ are controlled by changing the cavity configuration, which is a common procedure for Kerr-lens mode-locking experiments.

Another fundamental factor in our model is the presence of the high-order dispersions due to the active medium, the dispersion compensator, the output coupler, and high-reflective mirrors. The corresponding experimental characteristics are shown in Fig. 2. For the numerical calculations the original data were represented by the polynomial approximation. The action of dispersion can thus be presented in the following form:

\[
a(z, t) = \int_{-\infty}^{\infty} a(z, t')G(t - t')dt',
\]

\[
G(t - t') = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp \left[ -i \sum_{j=2}^{8} \frac{1}{j!} D_j (\omega - \omega_0)^j \right]
- (t - t') \omega \delta \omega,
\]

where $\omega$ is the frequency and $G(t - t')$ is the Green's function depending on the dispersion coefficients $D_j$ up to the eighth order of $j$. The eighth-order approximation was necessary to describe correctly the oscillations on the dispersion curve of the chirped mirrors.

The next important factor in our model is the gain saturation, which can be described on the basis of a quasi-two-level scheme of the active-medium operation.\(^{14}\)

\[
\frac{\partial a}{\partial t} = \frac{I_p \sigma_{14}}{h\nu} (\alpha_{\max} - \alpha) - \frac{|a|^2 \sigma_{32}}{h\nu} \alpha - \frac{\alpha}{T_r},
\]

where $I_p$ is the pump intensity, $\nu_p$ and $\nu$ are the pump and the carrier frequencies, $\sigma_{14}$ and $\sigma_{32}$ are the loss and the gain cross sections, respectively, and $T_r = 85 \mu s$ is the gain relaxation time. If the pulse duration is much less then the cavity round-trip time $T_{cav} = 14$ ns, then this equation can be replaced by

\[
\frac{\partial a}{\partial z} = P(\alpha_{\max} - \alpha) - \frac{E_s}{E_s} \alpha - \frac{T_{cav}}{T_r} \alpha,
\]

where $P = (I_p \sigma_{14}/h\nu)T_{cav}$ is the dimensionless pump intensity and, $z$ is the dimensionless longitudinal coordinate, i.e., the number of the cavity round trips. $E_s = h\nu\sigma_{32}$ is the gain-saturation energy flux, and $E$ is the full-pulse energy flux.

Self-phase modulation in the active medium was represented by nonlinear transmission operator $\exp[-i\beta a(z, t)t^2]$, where $\beta = 2\pi n_2 x/\lambda n = 3.4$ cm$^2$/TW is the self-phase modulation coefficient. Here $n$ and $n_2$ are the linear and nonlinear coefficients of refraction, respectively, $\lambda$ is the central wavelength corresponding in our case to the gain-band maximum, and $x = 8$ mm is twice the length of the active crystal.

Finally, we consider the stimulated Raman scattering within the active medium. Following Ref. 9, where the Raman-scattering contribution was calculated analytically on the basis of the soliton model, we supplement the model with the following equations:

\[
\frac{\partial a_s}{\partial z} = i \sum_{j=1}^{3} Q_j a_p, \quad (5a)
\]

\[
\frac{\partial a_p}{\partial z} = i \sum_{j=1}^{3} Q_j a_s, \quad (5b)
\]

\[
\frac{\partial^2 Q_j}{\partial t^2} + \frac{2}{T_j} \frac{\partial Q_j}{\partial t} + \frac{\Omega_j^2 Q_j}{T_j} = \mu_j a_p a_s^*, \quad (5c)
\]

where $\xi$ is the longitudinal coordinate (pulse-propagation axis) inside the active medium, $a_{p,s}$ are the amplitudes of the pump and the Stokes components within the generation spectrum, and $\Omega_j$ are the phonon-resonance frequencies \((j = 1, 2, 3)\) corresponding to the three Raman-active phonon resonances in LiSGaF; see Fig. 5). $T_j$ are the inverse bandwidths of the Raman lines, and $\mu_j = g_j^p \Omega_j / T_j$ are the coupling parameters for the Raman gain coefficients $g_j^s$.

Solving the third equation of the system, we obtain the steady-state phonon amplitude for the fixed-pump and the Stokes components with the corresponding frequencies $\omega_p$ and $\omega_s$:

\[
Q_j = \frac{\mu_j a_p a_s^*}{\Omega_j^2 - \frac{2i(\omega_p - \omega_s)}{T_j} - (\omega_p - \omega_s)^2}
- \frac{\mu_j a_p a_s^*}{2\Omega_j(\Omega_j - (\omega_p - \omega_s)) - 2i\Omega_j}.
\]

The validity of the last approximate expression follows from the fact that the Raman lines are narrow in comparison with the pulse spectrum. Summing up the contributions of all spectral components of the pulse to the phonon amplitude, the equations for the Stokes and the pump fields in the frequency domain can be written as

\[
\frac{\partial a_s}{\partial \xi} = i a_s \sum_{j=1}^{3} \mu_j \sum_k |a_{p,k}|^2
- \frac{2i\Omega_j}{T_j}[\Omega_j - (\omega_{p,k} - \omega_s)] + \frac{2i\Omega_j}{T_j},
\]

(7a)
cies by the Raman frequencies of the main oscillation pulse, shifted to the lower frequencies results in the appearance and growth of spectral replicas. With this seed signal the stimulated Raman scattering results can be solved on the basis of numerical simulation in the Fourier domain and the split-step method for nonlinear propagation of the initial pulselike seed.

There are two main mechanisms of the generation of the initial seed at the Stokes frequency for Eqs. (8). The first one is the spontaneous Raman scattering with the increments of the scattered intensity growth, \[ \frac{\partial a_p}{\partial \xi} = \frac{\lambda}{4} \sum_{j=1}^{3} \sum_{k} \frac{|a_{s,k}|^2}{2\Omega_j \Omega_j - (\omega_p - \omega_{s,k}) - \frac{2i\Omega_j}{v c}}. \] (7b)

It should be noted that in these equations, \( \omega_p - \omega_s = \Omega_j \), and there is the pairwise interaction of the spectral components within the wide-enough generation spectrum.

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where \( k \) is the index of the field’s spectral (i.e., Fourier) component (in the simulations we considered 2\(^{13} \) components). Since the Raman lines are narrow, the field variation within these lines is negligible, i.e., \( a_{s,k} \) are constant and can be taken out of the second summation. Then the summation can be executed explicitly by transition to the integral, resulting in

\[ \frac{\partial a_s}{\partial \xi} = \frac{\lambda}{4} \sum_{j=1}^{3} \sum_{k} \frac{a_{s,k}^j |a_p|^2}{\Omega_j}, \] $$(8a)$$

\[ \frac{\partial a_p}{\partial \xi} = -\frac{\lambda}{4} \sum_{j=1}^{3} \sum_{k} a_{s,k}^j |a_p|^2. \] $$(8b)$$

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where \( n_s \) is the index of refraction at Stokes frequency, \( T \) is the temperature, and \( k_B \) is the Boltzmann’s constant. With this seed signal the stimulated Raman scattering results in the appearance and growth of spectral replicas of the main oscillation pulse, shifted to the lower frequencies by the Raman frequencies \( \Omega_j \).

A more significant source for the Stokes component’s amplification, however, is the broadband pulse field itself. When the pulse spectrum is wide enough to become comparable with the Raman frequency shift, the lower-frequency part of the spectrum can play a role of the Stokes-component seed with respect to the higher-frequency part of the spectrum. The stimulated Raman scattering then transfers the energy from the higher-frequency components to the lower-frequency ones, resulting in the continuous redshift of the pulse spectrum as a whole. As the field amplitude of the laser pulse significantly exceeds the spontaneous seed, the second mechanism strongly dominates the first one. However, we included both mechanisms in our model because their influence on the pulse spectrum is quite different.

Later in the paper it is convenient to normalize the time to the inverse gain bandwidth \( t_g = 2.25 \) fs and the intensity to \( \beta^{-1} \), resulting in the normalization of the field energy to \( (\beta t_g)^{-1} \). As already pointed out, we analyzed the model described above in two ways: on the basis of distributed and of discrete-element approaches. In the framework of the distributed model we did not consider the propagation through the individual laser element and supposed that the pulse envelope is formed by the overall net dispersion in the cavity. As result, we have a split-step scheme describing ultrashort-pulse propagation from \( z = 1 \) to \( z + 1 \) transits:

\[ a(z', t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} a(z, t') C(t - t') L(t' - t'') \times A(t'' - t''' - G(t''' - t'')) dt' dt'' dt''' - \infty, \] $$(10a)$$

\[ a(z + 1, t) = a(z', t) \exp \left[ \frac{-it |a(z', t)|^2}{1 + \sigma |a(z', t)|^2} \right], \] $$(10b)$$

\[ a(z + 1) = a(z) \exp \left[ -\tau \int_{-\infty}^{\infty} |a(z, t')|^2 dt' - \frac{T_{cav}/T_r - P}{T_{cav}/T_r} \right] \times \frac{P \sigma_{\text{max}}}{T_{cav}/T_r} \left[ 1 - \exp(-T_{cav}/T_r - P) \right] + \frac{T_{cav}/T_r}{P}, \] $$(10c)$$

where \( \tau = t_g/E_\beta = 0.00079 \) is the normalized saturation parameter. The Green’s functions \( A, L, \) and \( C \) describe spectral bands of gain, reabsorption, and output loss, respectively (note that the dispersion is already included in \( G \)):

\[ A(t - t') = \frac{1}{2\pi} \int_{-\infty}^{\infty} [1 + a(\omega)] \Phi_a(\omega) \exp[i \omega(t - t')] d\omega, \] $$(11a)$$

\[ L(t - t') = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Phi_r(\omega) \exp[i \omega(t - t')] d\omega, \] $$(11b)$$

\[ C(t - t') = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Phi_{out}(\omega) \exp[i \omega(t - t')] d\omega, \] $$(11c)$$

Here, \( \Phi_a, r, out \) are the form factors describing spectral profiles of the gain, reabsorption, and output-coupler bands, which resulted from the functional approximation of the experimental data (Fig. 3).

System (10) has to be solved by system (8). The result can be solved on the basis of numerical simulation in the Fourier domain and the split-step method for nonlinear propagation of the initial pulselike seed.

The discrete-element approach is based on the element-to-element simulation of the pulse propagation through the cavity on every round trip, following the laser scheme in Fig. 1. The nonlinear and spectral characteristics of each laser element are considered separately. Further refinement of the simulation is achieved by considering the pulse propagation through an active medium by splitting it into five slices and applying the split-step procedure to each slice consecutively.

Typical pulse spectra that result from the calculation are shown in Fig. 6. The corresponding intracavity pulse energies are used as subscripts to the individual spectra.
4. DISCUSSION

Our simulations are aimed at investigation of the influence of the different factors on spectral characteristics of the ultrashort pulses. Therefore to simplify the interpretation, we first consider the high-order dispersion action without Raman scattering and without reabsorption; then reabsorption and Raman scattering are taken into consideration without high-order dispersion action; and finally, the joint action of all factors is analyzed. To conclude, we also compare the obtained results with the calculation based on the discrete-element model.

A. High-Order Dispersion Action

As it was discussed in Ref. 7 a small contribution of the third-order dispersion to the characteristics of the Schrödinger soliton does not cause the frequency shift but does introduce additional field time delay on the cavity roundtrip. The soliton model also predicts generation of multiple side bands due to the third- and higher-order dispersion.16 However, the influence of the linear and nonlinear dissipative terms in Eqs. (10) can destroy the soliton character of the pulse; in particular, it can add the chirp. The latter results in the frequency shift of the pulse spectrum in the non-steady-state regime. Additionally, in the real-world laser systems the contribution of the high-order dispersions, as a rule, lies beyond the bounds of the perturbation-theory validity.

To analyze the possible influence of the higher-order dispersion on the pulse spectrum, we start with a typical net-dispersion curve corresponding to the experiment with the chirped mirrors [Fig. 7(a) solid curve]. By adding the nonlinear terms, the dispersion can be shifted [Fig. 7(a), dotted curve] and can undergo significant form distortion [Fig. 7(a), dashed curve]. The resulting output spectra obtained from the distributed model are shown in Fig. 7(b). It is generally assumed that there exists a certain spectral window of dispersion that is optimal for steady-state pulse generation. The contribution of the high-order dispersion terms may effectively shift this window and change its size, thus changing the spectral position of the pulse. According to the simulation, the net-dispersion shift to the longer wavelengths is indeed accompanied by the redshift of the pulse spectrum [transition from solid to dotted curves in Fig. 7(b)], whereas change of the dispersion profile generally causes pulse spectrum distortion [Fig. 7(b), dashed curve] and sideband generation.5,16

However, we cannot consider this shift as the cause of the experimentally observed effect because the dispersion shift has a linear nature, i.e., there is no obvious dependence of this shift on the field energy. This is demonstrated by Fig. 8, where the pulse-energy variation due to the pump variation changes the spectral profile but does not cause any noticeable frequency shift (compare solid and dash curves in this figure; the typical spectrum shift is \( \sim 5 \) nm, which is much less than the experimentally observed values). See also Table 3.

The obtained results demonstrate that the self-frequency shift cannot be caused by the nondissipative factors. As the pulse duration is too large for the nonlinear dispersion to play any significant role, we concentrate on spectrally dependent losses and the Raman effect.

![Fig. 6. Spectral profiles of the output laser pulses, obtained with the discrete model. The corresponding pulse energies are used as subscripts to each curve.](image1)

![Fig. 7. Dependence of (a) dispersion and (b) generation spectra on wavelength. \( P = 3.2 \times 10^{-4}, \sigma = 1, \gamma = 0.05, \) and pulse energy \( E = 20 \) nJ. Pulse durations \( t_p: \) 27 (solid curve), 38 (dash), and 36 fs (dot).](image2)
the presence of dissipative factors only, and in the absence of simulations on the basis of the model described above in gain maximum.

Besides that, the location of the pulse-energy growth. However, the magnitude of this coefficient decrease, the behavior of the net-gain maximum on the saturated steady-state gain coefficient \( g \). For the corresponding pulse parameters, see Table 3.

As mentioned above, the dependence of the frequency shift on the pulse energy implies the involvement of some nonlinear mechanism. Since in the real-world systems the gain band often does not coincide with the filtering band (output mirror in our case) and the reabsorption band, the spectral position of the net-gain maximum changes with the gain value. The latter is defined by the pump and by the pulse energy [see Eq. (10c)]: pulses with higher energy experience lower gain due to the multipass gain saturation. The dependence of the net-gain maximum on the saturated steady-state gain coefficient \( \sigma = 1 \) and \( \gamma = 0.05 \). For the corresponding pulse parameters, see Table 3.

B. Output Loss and Reabsorption in Gain Medium

As already mentioned above, the main contribution to the stimulated Raman process comes from the energy transfer from the blue part of the pulse spectrum (pump) to the red one (Stokes). The efficiency of the stimulated Raman scattering is therefore defined by the product of intensities at pump and Stokes frequencies. The separation between Stokes and pump components is fixed; it is equal to the Raman line frequency \( \Omega_j \). Therefore de-

Table 3. Normalized Pump Power, Pulse Duration, and Energy for Fig. 8

<table>
<thead>
<tr>
<th>Curve in Fig. 8</th>
<th>( P \times 10^4 )</th>
<th>( t_p ) (fs)</th>
<th>( E ) (nJ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>solid</td>
<td>3.2</td>
<td>27</td>
<td>20</td>
</tr>
<tr>
<td>dash</td>
<td>2</td>
<td>30</td>
<td>12</td>
</tr>
<tr>
<td>dot</td>
<td>3</td>
<td>25</td>
<td>16</td>
</tr>
</tbody>
</table>

C. Raman Scattering

As pointed out in Subsection 4.B, the net-gain shift model fails to provide correct description of the experimental data by a factor of 2. However, taking the stimulated Raman scattering into account allows us to obtain large frequency shifts, increasing with the pulse-energy growth. In Fig. 9 the curve \( IJK \) demonstrates the Raman-scattering action in the absence of the reabsorption effect. As the simulation demonstrates, the red components originate from the amplified Raman signal, which pulls the whole spectrum over the long-wavelength limit at the given pump power, defined by the spectral filtering. Note the pronounced thresholdlike character of the effect. For the small energies (point \( J \) ) the spectral shift is negligible, but the energy growth causes a very strong shift (60 nm in our case), in good agreement with experimental results\(^{1,4,5}\) and with analytical prediction.\(^9\) Since the gain saturation does not play an important role in this case, the dependence of the frequency shift on the gain coefficient is insignificant.

As already mentioned above, the main contribution to the stimulated Raman process comes from the energy transfer from the blue part of the pulse spectrum (pump) to the red one (Stokes). The efficiency of the stimulated Raman scattering is therefore defined by the product of intensities at pump and Stokes frequencies. The separation between Stokes and pump components is fixed; it is equal to the Raman line frequency \( \Omega_j \). Therefore de-
creasing the pulse spectrum width strongly suppresses the effect and reduces the self-frequency shift. Assuming that the pulse spectrum has an exponential fall-off to the blue and the red sides, we see that the dependence of the Raman shift on the pulse spectrum width should be asymptotically exponential at long pulse durations. This is also the result of the analytical theory in Ref. 9, where a sech² pulse shape has been assumed.

As the Raman effect strongly depends on the pulse intensity and the Raman gain, it should be especially pronounced in low-gain lasers working with low output coupling and high intracavity pulse energy, such as Cr:LiSGaF, Cr:LiSAF, and Cr:YAG. These materials also possess strong and broad Raman lines. The large power-dependent redshift in Cr:LiSGaF and Cr:LiSAF is well documented. In femtosecond Cr:YAG lasers, a femtosecond pulse spectrum is also always redshifted with respect to the cw wavelength in the same resonator. Recently, significant redshift has also been observed in a 14-fs Cr:forsterite laser.

D. Discrete-Element Model

Finally, we can compare the simulation results in the case of distributed and discrete-element models. In the case of the high-order dispersion action the transition to the distributed-element model does not significantly change the spectral characteristics of the pulse [the dot curve in Fig. 8(b)]. The long-wavelength shoulder of the spectrum in the case of the net dispersion corresponding to the dashed curve in Fig. 7(a) is somewhat stronger than in the distributed model. This is because this shoulder results from a local dispersion maximum due to the chirped mirror (CM₁ in Fig. 1), which is the closest to the output mirror. Additionally, the self-phase modulation contribution is found to be slightly higher in comparison with the distributed model. However, all the differences with the distributed model are minor. We found also that the transformation of the spectrum on passage through each of the laser elements is small, ensuring the validity of the distributed model. It should also be noted that the transition from the distributed to the discrete model slightly increases the contribution of the Raman scattering, resulting in the lower threshold energy.

E. Comparison with the Soliton Model

Although the soliton model correctly predicts almost negligible dependence of the spectrum position upon the higher-order dispersion, there is a certain difference between our present numerical simulations and the results of the analytical theory of the induced Raman effect in mode-locked oscillators. The latter theory, using a soliton-perturbation approach, predicts the following dependence of the wavelength shift $\Delta \lambda$ upon the pulse duration $\tau$ and energy flux $E$:

$$\Delta \lambda \propto \sum_j \frac{\beta_j}{F_j} G(\Omega_j \tau)(E \tau)L,$$

where $L$ is the crystal length. Within the soliton approximation the product $(E \tau)$ is constant $(E \tau \approx \beta_p |D|$, where $D$ is the group-velocity dispersion at the soliton center wavelength). The function $G(\Omega_j \tau)$ exponentially decays at long $\tau$ and linearly goes to zero at small $\tau$. For most laser materials with typical frequencies of strong Raman absorption.
lines lying between 300 and 800 cm⁻¹ the function \( G(\Omega, \tau) \) demonstrates a broad peak around 20 fs.⁵,¹⁰ Thus the soliton theory predicts a decrease of the Raman shift when the pulse duration becomes shorter than ~20 fs. However, both the experiment and the numerical simulations presented above do not confirm such a trend: the frequency shift does not decrease at short pulse durations but remains approximately constant at ~50–60 nm for pulse durations of ≤20 fs.¹,⁴ A larger shift is simply impossible due to the increased losses through the output coupler.

We believe that the reason for this discrepancy is that the soliton-perturbation approach necessarily loses its validity when the frequency shift becomes comparable with the spectral width of the pulse. In fact, in our case the shift is so strong that spectral intensity at 842 nm, the center wavelength of an unperturbed soliton, is already much less than that of the spectrum peak (see Figs. 4 and 6). Inapplicability of the soliton approach can also be seen because the condition \( (E\tau = \text{const}) \) at constant dispersion does not hold, both in the experiment (Table 1) and in simulation (Table 4). At the same time, soliton-perturbation theory⁹ remains applicable to the active medium with a weak Raman spectrum, like Ti:sapphire.¹⁰

We must thus conclude that the significant (comparable with the spectral pulsewidth) Raman self-frequency shift, once started, does not reduce back to the shorter pulse duration, as implied from the soliton model. In practice, this means that the reduction of the energy flux inside the active medium (e.g., defocusing the beam in the active medium or decreasing the intracavity pulse energy) and shortening of the active medium to reduce the interaction length remain the only effective ways to combat the Raman shift, if shorter pulses are to be generated in crystals with strong Raman spectra.

5. CONCLUSION

Using the numerical simulations performed in the framework of one-dimensional distributed and discrete-element models, we analyzed the spectral characteristics of a Kerr-lens mode-locked laser, using the Cr:LiSGaF as an example. The two main factors causing the ultrashort-pulse self-frequency shift have been established: the nonlinear shift of the net-gain band due to the gain saturation in the presence of reabsorption in the active medium, and the stimulated Raman scattering. The first effect is essential for comparatively small pulse energies and produces wavelength shifts up to 30 nm. The Raman scattering occurs as a result of pulse-energy growth and may cause the large (>50 nm) redshift. The contributions of the high-order dispersion and the gain-band asymmetry to the pulse central frequency is much smaller than that of the reabsorption and the stimulated Raman scattering, which is the main cause of the spectral redshift in a Kerr-lens mode-locked laser. The shift values obtained from the numerical simulations agree well with experimental data. For practical application the onset of the Raman self-frequency shift is a negative phenomenon, and it can only be suppressed by decreasing the pulse-energy flux inside the active medium or by decreasing the length of the latter.

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REFERENCES