BANDWIDTH LIMITED AMPLIFICATION OF 220 fs PULSES IN XeCl: THEORETICAL AND EXPERIMENTAL STUDY OF TEMPORAL AND SPECTRAL BEHAVIOR

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The spectral structure of the gain-profile of XeCl was found to strongly influence the spectral and temporal characteristics of amplified ultrashort light pulses. A simple model was used to numerically study both effects as a function of amplifier length and input pulse energy. The results are in good agreement with experiments. With these data the performance of a XeCl amplifier was optimized for shortest pulse duration with minimum distortion.

1. Introduction

In the preceding paper [1] Szatmári et al. reported on the generation of ultrashort light pulses at 308 nm and their subsequent amplification in a XeCl amplifier. The ultrashort pulses are generated at 616 nm in a distributed feedback dye laser (DFDL) and frequency doubled in a KDP crystal. These light pulses have a smooth spectrum with a width of ca. 50 cm^{-1} (see fig. 2 in [1]). The autocorrelation measurement of the DFDL light pulses at 616 nm yields a width of 500 fs corresponding to a pulse duration of 320 fs when a $sech^2$ pulse shape is assumed. Hence we have strong evidence that the seed pulses at 308 nm are transform limited with a pulse duration of 200–250 fs.

When amplification of these pulses in a XeCl amplifier was attempted, the spectrum of the amplified pulse displayed a modulation apparently reflecting the vibrational structure of the lasing transition in XeCl. At the same time side peaks appeared in the autocorrelation traces located at ± 1.3 ps from the central peak. Also, the central peak was considerably broadened. After a second pass through the XeCl amplifier the spectral as well as the temporal modulation strongly increased.

Through careful readjustment of the whole setup including exchange of all reflecting surfaces it was shown that the temporal modulation apparent from the autocorrelation is not due to a reflection but intrinsic to the amplifying process in XeCl. Hence there was strong evidence that the temporal modulation is directly related to the spectral modulation. On the first sight this leads to a very pessimistic prognosis concerning the possibility to amplify light pulses of less than 250 fs duration to high power levels with a XeCl amplifier.

In order to gain a better insight into the spectral and temporal evolution of ultrashort light pulses in an amplifier with structured gain profile we performed numerical calculations. The underlying simple model will be described in the following section. With this model we were able to reproduce the increasing temporal and spectral distortion of the light pulse with increasing length of the amplifier in good agreement with our observations.

Computer simulations with a fixed amplifier length and varying input pulse energy predicted a strong influence of the latter on the shape of the output light pulses, which was experimentally verified. With the combined experience from numerical simulations and experiments it was, finally, possible to find optimal operating conditions for the XeCl amplifier. Under these conditions amplified pulses of 5 mJ energy and 220 fs duration are obtained which show only slight modulation in their spectra and no sidebands in the autocorrelation traces. We believe that these pulses are close to the shortest pulses which can be achieved through amplification in XeCl.

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2. Numerical simulations

We simulated the pulse spectra and autocorrelation traces which can directly be compared to experimental results. As starting point we took the well known equation of Frantz and Nodvik [2]

$$W_o/W_s = \ln\{1 + [\exp(W_i/W_s) - 1] \exp(g_0 L)\} . \quad (1)$$

In this expression W_i is the input energy (per area) of the seed pulse fed into the amplifier, W_o is the corresponding energy of the output pulse, W_s is the saturation energy, and g_0 a constant gain coefficient. Eq. (1) has been derived with a rate-equation model neglecting the finite spectral width of the light pulse and the gain curve. Hence it is, strictly speaking, only applicable to homogeneously broadened amplifier media with phase-relaxation time T_2 much shorter then the duration of the light pulse to be amplified.

These conditions are not fulfilled for XeCl: Corkum and Taylor [3] have investigated the amplifier properties of XeCl with seed pulses of 2 ps duration and 10 cm^{-1} bandwidth. They could burn holes into the gain-profile which recovered with a time constant of 40 ps. Nevertheless they obtained a good fit of their data to eq. (1) with $W_s \approx 1$ mJ/cm^2. Recently Glownia et al. [4] performed a similar experiment with seed pulses of 350 fs duration and obtained $W_s = 2.1$ mJ/cm^2 and $g_0 = 0.13$ cm^{-1}. Corkum and Taylor used pulses of 1 Å bandwidth and were thus able to measure the gain $g(\nu)$ as a function of frequency. This gain profile shows four maxima reflecting the vibrational structure of the A→X transition in XeCl (see fig. 1).

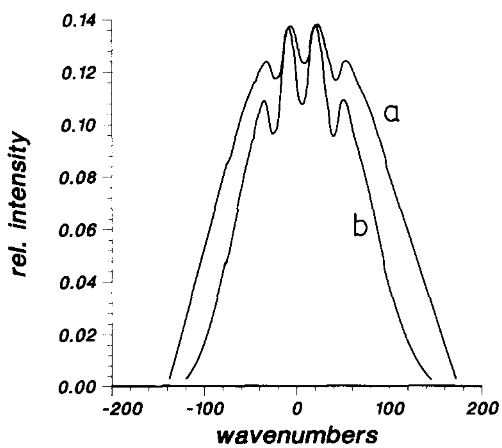

Fig. 1. Curve (a): uncorrected gain profile of XeCl adapted from the measurements of Corkum and Taylor [3]. Curve (b): corrected gain profile obtained from curve a through deconvolution with a gaussian with 10 cm^{-1} width. The wavenumber scale refers to a center frequency of 32468 cm^{-1}, i.e. 308 nm.

Our approach to the calculation of the spectral evolution of a light pulse during amplification is to use eq. (1) for each spectral component of the pulse and assume independent amplification of all spectral components. The latter assumption seems to be justified by the large inhomogeneous contribution to the bandwidth of the gain and the slow (40 ps) cross-relaxation.

Hence we replace the energies W with the corresponding power spectra $I(\nu)$ and use a frequency dependent gain coefficient $g(\nu)$:

$$I_o(\nu)/I_s = \ln\{1 + [\exp(I_i(\nu)/I_s) - 1] \times \exp(g(\nu)L)\} . \quad (2)$$

For low input intensities this reduces to

$$I_o(\nu) = I_i(\nu) \exp(g(\nu)L) , \quad (3)$$

i.e. the output spectrum is the input spectrum modulated by the small signal gain. This fact was already observed by Glownia et al. [4].

To calculate the autocorrelation curves from these spectra we use the fact that the electric field $E(\nu)$ in frequency domain and $E(t)$ in time domain are related by the Fourier transform:

$$E(\nu) = \int dt\, E(t) \exp(i2\pi\nu t) =: \mathscr{F}[E(t)] ,$$

$$E(t) = \int d\nu\, E(\nu) \exp(-2\pi i\nu t) =: \mathscr{F}^{-1}[E(\nu)] . \quad (4)$$

The power spectrum is $I(\nu) = E(\nu)\, E^*(\nu)$, and the temporal intensity profile of the pulse is $I(t) = E(t)\, E^*(t)$. Note that $I(t)$ and $I(\nu)$ are not related by the Fourier transform as in eq. (3), since all phase information is lost in taking the absolute square. Hence there is no unique way to calculate the (slow) autocorrelation function

$$A(t) = \int dt'\, I(t')\, I(t'+t) \quad (5)$$

from $I(\nu)$. Therefore, we must make an assumption concerning the phase in $E(\nu)$. We use the simplest approach $E(\nu) = \sqrt{I(\nu)}$.

This approximation is equivalent to the assumption that the amplification process introduces no phase shift between various frequency components, and that a transform limited pulse remains transform-limited throughout the amplification process. For the numerical evaluation of the autocorrelation function we employed the convolution theorem of the Fourier transform leading to

$$A(t)=\mathscr{F}^{-1}[J(\nu)\,J^*(-\nu)]\,, \tag{6}$$

with

$$J(\nu)=\mathscr{F}[I(t)]\,. \tag{7}$$

With a Fast-Fourier-Transform routine [5] these calculations could be performed within a few seconds on a personal computer.

3. Experimental

The experimental setup used for the generation of ultrashort light pulses at 308 nm was the same as that described in ref. [1]. The energy of the light pulses injected into the second amplifier pass was varied with attenuators. In this way the response of the XeCl-amplifier to short input pulses could be studied over a wide range of input-intensities approaching the saturation intensity. The first observations of the temporal and spectral pulse modulation mentioned in the introduction were obtained with a slightly different setup. There the arrangement to cut the ASE level consisted of two telescopes, resulting in much greater losses and hence reducing the input energy available for the second amplifier pass. The spectra of the output pulses were dispersed by a 3600 l/mm grating and measured with a diode array. Autocorrelation traces of the ultrashort pulses were measured through two photon resonant multiphoton ionization in triethylamine as in ref. [1].

4. Results and discussion

For all simulations an input pulse at 308 nm with sech^2-shape and 200 fs pulse width was assumed. The parameters varied were the length of the amplifier, L, and the ratio $I_i(\max)/I_s=Q$.

From the wavelength-dependent gain measurements of Corkum and Taylor [3] we approximated the smooth gain-profile shown in fig. 1a. The maximum of this gain profile was scaled to a value of $g_0=0.138\ \mathrm{cm}^{-1}$ corresponding to a small signal gain of $\exp(g_0L)\approx 1000$ observed with an effective amplifier length of $L=50$ cm.

This gain profile has been obtained with a resolution of 1 Å ($10\ \mathrm{cm}^{-1}$), which is not negligible compared to the separation of the maxima and minima in the gain profile. Hence this uncorrected gain curve considerably underestimates the difference in gain $\Delta g=g_{\max}-g_{\min}$ between the maxima and the minima in the gain curve. The simulations are, however, very sensitive to the value of Δg. Hence we used a corrected gain profile calculated through deconvolution of the uncorrected gain profile with a gaussian of 10 cm^{-1} width. This corrected gain profile is shown as curve (b) in fig. 1.

The simulated performance of the amplifier as a function of its length is shown in fig. 2. The input intensity was scaled to $Q=10^{-5}$. The spectrum and autocorrelation curve corresponding to this input pulse are displayed in the first row of fig. 2. The following rows show the corresponding characteristics of the pulse after passage through an amplifier of 33 cm, 100 cm, and 200 cm length. At 33 cm amplifier length the spectrum shows considerable modulation due to the different gain at different frequencies. At the same time two sidebands appear in the autocorrelation curve ca. ± 1 ps from the main peak. With increasing length of the amplifier the spectral modulation and the intensity of the sidebands in the autocorrelation curve increase strongly. This is evident from the third row in fig. 2, corresponding to $L=100$ cm. However, when the amplifier length is further increased, the spectrum of the pulse becomes broader with less modulation. At the same time the sidebands in the autocorrelation curve almost disappear.

This behavior can be easily understood on the basis of eq. (2). For input intensities much smaller than the saturation intensity eq. (2) can be rewritten as

$$I_o(\nu)/I_s=\ln\{1+I_i(\nu)/I_s\,\exp(g(\nu)L)\}\,. \tag{8}$$

For amplifier lengths small enough to satisfy

$$I_i(\nu)/I_s\,\exp(g(\nu)L)\ll 1\,, \tag{9}$$

this reduces to eq. (3). In this limit the modulation

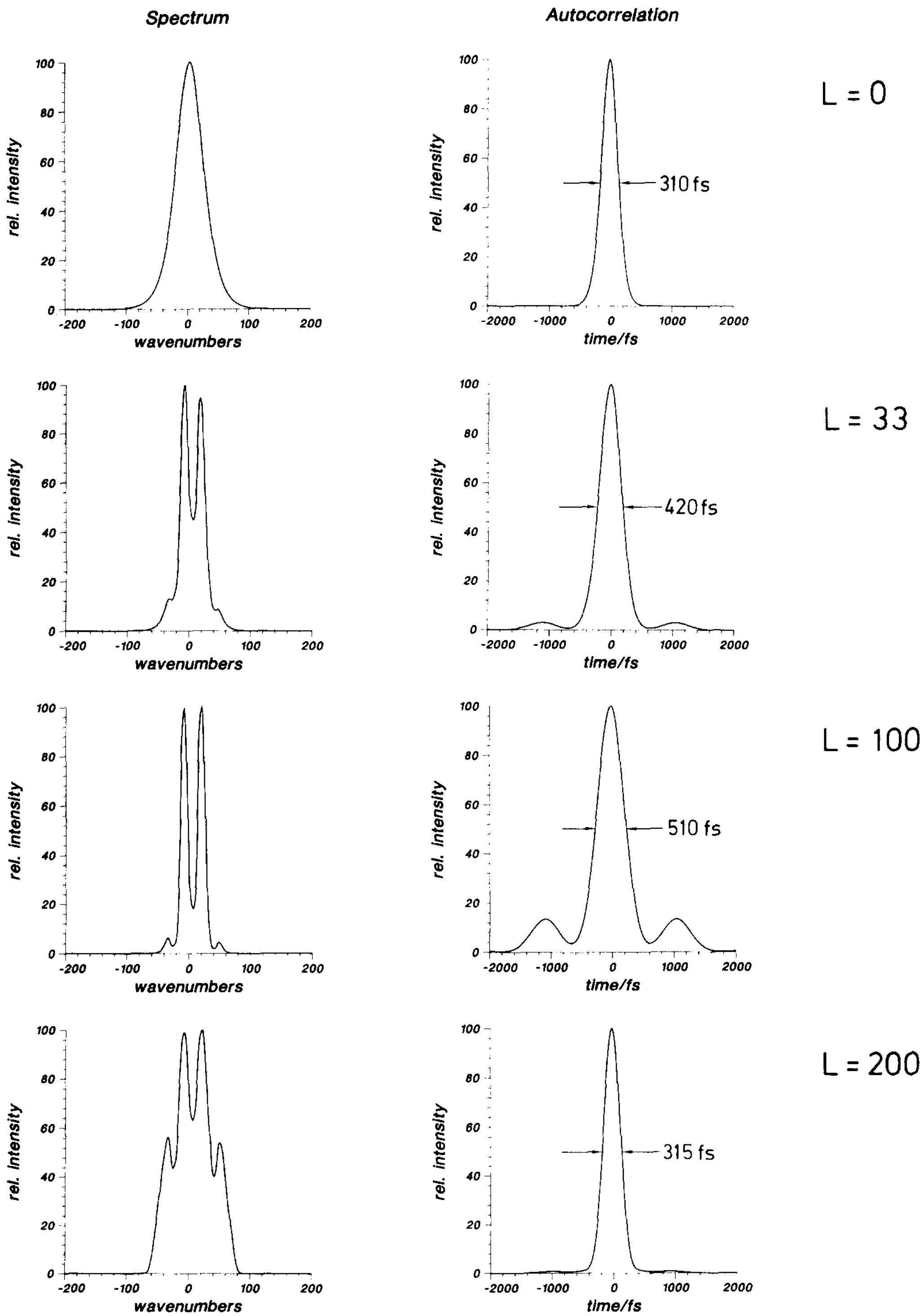

Fig. 2. Simulated amplifier performance as a function of length. Left column: power spectra of the light pulse, centered at 32468 cm^{-1} (308 nm). Right column: corresponding autocorrelation curves. The first row refers to the input pulse which was approximated by a $sech^2$-shape of 200 fs duration and with intensity of $I_{max}/I_s = 10^{-5}$. The following rows show the output pulse after passage through an amplifier of length 33 cm, 100 cm, and 200 cm, from top to bottom.

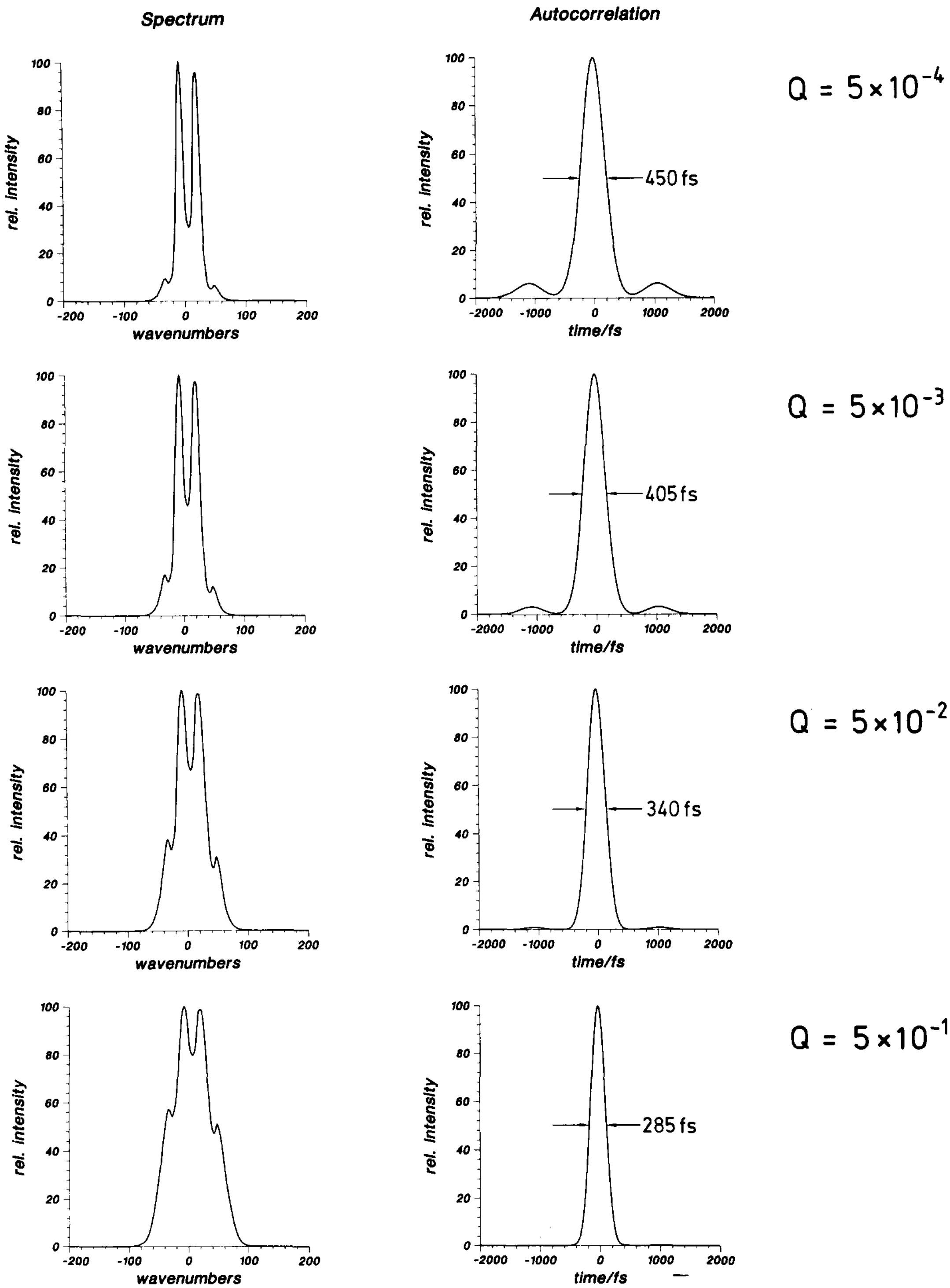

Fig. 3. Simulated amplifier performance as a function of input intensity for a fixed length of 50 cm. Left column: power spectra of the amplified light pulses. Right column: corresponding autocorrelation curves. The input pulse had sech^2-shape with a pulse width of 200 fs. The input intensity I_{max}/I_s from top to bottom is: 5×10^{-4}, 5×10^{-3}, 5×10^{-2}, 5×10^{-1}.

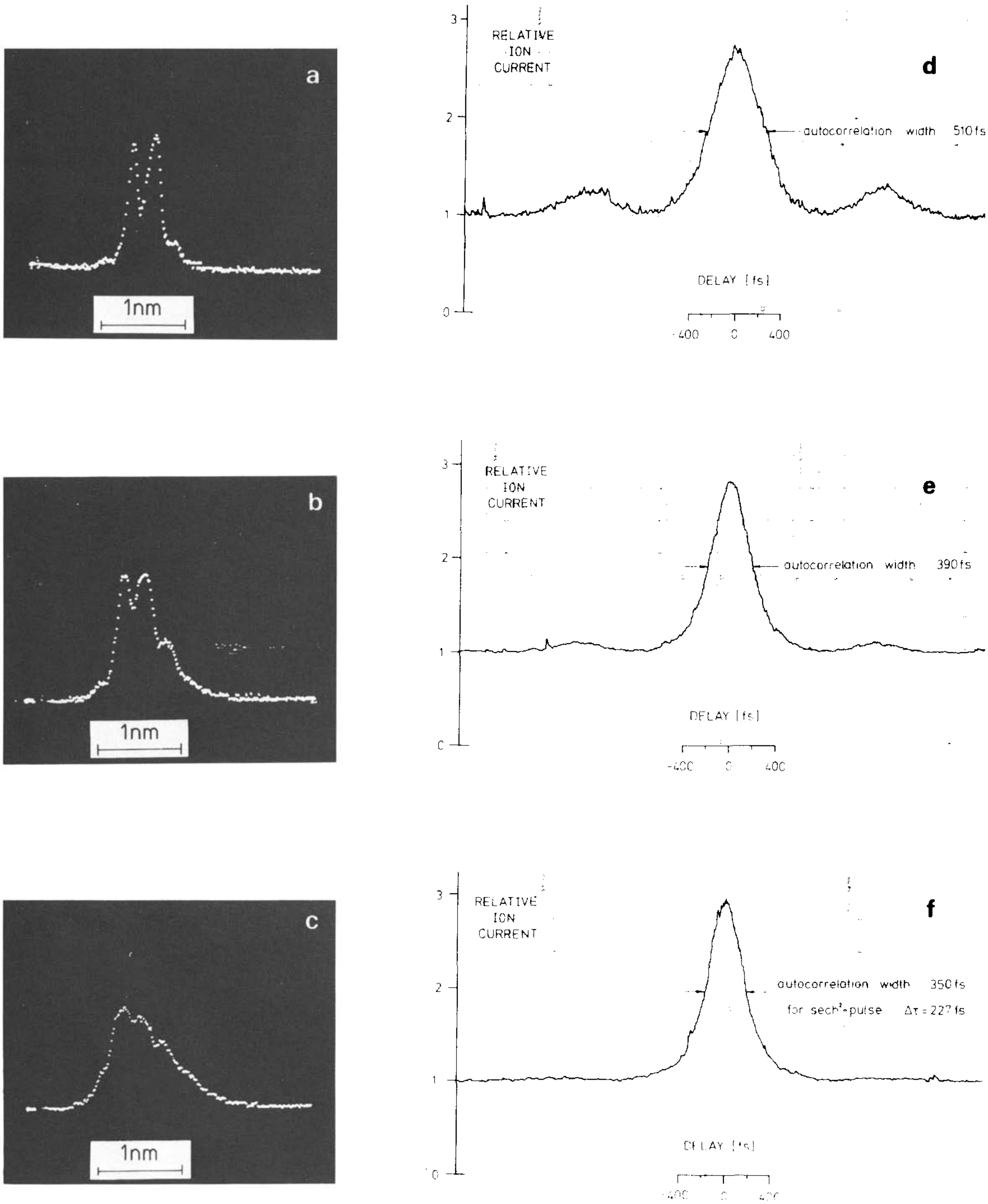

Fig. 4. Experimentally determined amplifier performance as a function of the input pulse energy. Left column: power spectra of the output pulse, right column: autocorrelation curves. The input energy from top to bottom is 1 μJ, 10 μJ, and 100 μJ.

of the spectrum at the frequencies ν_1 and ν_2 of maximal and minimal gain respectively, increases exponentially with the length of the amplifier:

$$\frac{I_o(\nu_1)}{I_o(\nu_2)}=\frac{I_i(\nu_1)}{I_i(\nu_2)}\exp(\Delta gL) . \qquad (10)$$

This behavior changes when saturation sets in. Then eq. (8) reduces to

$$I_o(\nu)/I_s=\ln(I_i(\nu)/I_s)+g(\nu)L . \qquad (11)$$

Hence for long amplifiers the pulse spectrum will approach the gain profile $g(\nu)$. The reason for this is the fact that the spectral components in the maxima of the gain curve will be saturated at shorter amplifier lengths than those at the minima, leading to stronger amplification of the valley of the spectrum with further amplification. We can estimate that this effect sets in a saturation length L_s for which

$$g_{max}L_s=-\ln(I_i(\nu_1)/I_s) . \qquad (12)$$

At this point the largest modulation of the spectrum will be obtained.

This largest modulation is proportional to $\exp(\Delta gL_s)$. Hence a shorter L_s will lead to much smaller modulation, and in order to achieve this, the input intensity must be increased. The input spectrum has its maximum at ν_2 (the minimum of the gain curve) and 70% of this value at ν_1 (the maximum of the gain curve). Hence for the pulse used in fig. 2 with $Q=10^{-5}$ we obtain $L_s=86$ cm and $\exp(\Delta gL_s)=12.1$. Increasing the input intensity to $Q=5\times10^{-4}$ reduces L_s to 58 cm and the maximum modulation to 5.3. Hence for a given length L of the amplifier the input energy must be high enough to yield $L_s<L$ in order to reduce the spectral and temporal distortion of the pulse.

The behavior of an amplifier with a fixed length of 50 cm to input pulses of increasing energy is simulated in fig. 3. The same pulse shape as in fig. 2 was used as input. The first row of fig. 3 corresponds to $Q=5\times10^{-4}$ and $L_s=58$ cm. Under this condition the maximum distortion in the spectrum and the autocorrelation are observed. The width of the central peak in the autocorrelation curve also indicates a lengthening of the pulse duration by 50%. The following rows show the results for $Q=5\times10^{-3}$, 5×10^{-2}, and 5×10^{-1} corresponding to $L_s=41$ cm, 24 cm, and 7.6 cm, respectively. With increasing input intensity the spectrum becomes broader and less modulated. The sidebands in the autocorelation curves decrease in intensity, and the central peak becomes narrower.

The situation simulated in fig. 3 was experimentally realized through attenuation of the input pulse injected into the second pass of the XeCl amplifier. We approximated the normalized input intensity Q with the ratio of the input energy to the saturation energy density. The latter was taken to be 2.1 mJ/cm^2 as measured by Glownia et al. [4] with subpicosecond pulses. (Preliminary experiments in our laboratory indicate that the saturation energy might be somewhat lower.) Fig. 4 shows the results obtained with injected pulse energies of 1 μJ, 10 μJ, and 100 μJ (unattenuated), corresponding to $Q=5\times10^{-4}$, 5×10^{-3}, and 5×10^{-2}, respectively. These experimental results agree very well with the simulations: With increasing input energy the distortion of the spectrum is reduced, the sidebands in the autocorrelation curve disappear, and the pulse duration is compressed.

This observation is strong evidence that the approximations in our simple model are justified. Since broadening of the spectrum and reduction of its modulation is accompanied by pulse-shortening it is also likely that the light pulse remains transform-limited during amplification.

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