1/50 of the peak, only a single pulse is present for each cavity transit. As the pulse grows in intensity, the characteristic broadening and spectral structure of self-phase modulation appear. Finally, the spectrum reaches a maximum width of 80 Å with random structure that changes from one pulse to the next. Neither extensive frequency sweep nor subpicosecond structure can be supported by the early 3-Å width. The minimum pulse duration possible for this spectral width is 6 ps. It is during and after the period of self-phase-modulation spectral broadening that the mode-locked pulse attains the characteristics observed in autocorrelation and compression measurements. In the analysis of the pulse development we examine the propagation of a 6-ps Gaussian pulse with transverse distribution of the fundamental mode.

We used a computer calculation, including saturable absorption, self-phase modulation, self-focusing, dispersion, and amplification, to follow the development. As the pulse grows the nonlinear index of refraction causes the more intense portions to experience longer optical path lengths. The leading edge of the pulse expands and the trailing edge compresses, producing a positive frequency sweep. At this point usual transverse intensity distributions will self-focus, with resultant high losses and power limitations. If there is limited self-focusing, normal dispersion will cause different frequency components to interfere, finally producing a burst of random fluctuations. Again a high-loss situation results, because self-phase modulation of short-duration fluctuations will broaden the spectrum beyond the fluorescence bandwidth. We find that the maximum energy for a 6-ps pulse is roughly 10 mJ. Although these calculations are only qualitative, they clearly show the importance of self-phase modulation and self-focusing in operation of high-power pulsed mode-locked lasers.

R.9 Mode-Locking Characteristics of Substructured and Nonlinearly Broadened Picosecond Laser Pulses, D. M. Kim, F. K. Tittel, and T. A. Rabson, *Department of Electrical Engineering, Rice University, Houston, Tex. 77001.*

The purpose of this paper is to describe analytically and in a self-consistent way the saturated dye-induced nonlinear frequency chirping in the linearly swept Gaussian pulses and show explicitly its effects. The significant modifications caused by the nonlinear dispersion, namely, the substructures in the pulse envelope and the nonlinear pulse broadening by a substantial amount, will be discussed analytically. In addition, this substructured pulse will be followed in time domain inside the resonant cavity and its stability will be presented. This kind of direct time-domain analysis was initiated in this field by Cutler[1] and was generalized by Kuizenga and Siegman[2] who described the essential features of FM and AM mode-locked lasers, including the effect of a linear frequency chirping. However, as Svelto's numerical anlysis indicates,[3] the nonlinear phase dispersion plays an important role in mode locking, and it is thus necessary to incorporate this term in the theory of mode locking.

We consider a Gaussian pulse (with bandwidth $\Delta\Omega$) having linear α as well as nonlinear β phase dispersions. Assuming that $\beta < \alpha$, we obtain the exact expression of the pulse envelope as

$$\mathcal{E}(\tau) = \mathcal{E}_0 \sum_n (j^n/n!)(\beta/4\Gamma^2)^n e^{-\tau^2/2\Gamma} \cdot H_{4n}[\tau/(2\Gamma)^{1/2}].$$

Here H_m denotes the mth-order Hermite polynomial. $\tau = \Delta\Omega t$ is a dimensionless scaled time parameter and the magnitude of the complex quantity $\Gamma = 1 + j\alpha$ represents the scaled time duration of linearly chirped pulses. The nonlinear term gives rise to a special subset of Hermite polynomials. Each term in the sum is a harmonic oscillator eigenfunction, aside from an additional weighting factor $\exp -(\tau^2/4\Gamma)$. Since the nth-state eigenfunction has n nodal points and the pulse envelope consists of the smooth Gaussian ground state as well as eigenfunctions of higher states, the envelope will develop substructures. More significantly, the probability distributions of higher excited states have considerably wider half-widths than that of the ground state. Thus the time duration of nonlinearly frequency chirped pulses is longer than the linear time duration by a considerable amount. Finally, the pulse envelope is symmetric in time for the case considered here, in which the first nonlinear effect comes in the fourth Taylor expansion term of the phase. The more general case of nonzero third term will be shown to lead to the same conclusions, except that the time dependence of the pulse envelope becomes asymmetric. The above general features will be shown to be born out in graphs where we will draw the intensity time profile of the lasers for various values of the ratio between β and α. For a small ratio the time dependence of the intensity remains peaked at the center with a slightly increased half-width from the linearly chirped case. As one raises the ratio and sweeps through a certain threshold value, the substructures begin to appear, while the gradual increase in time duration has now reached approximately twice the value of the linearly chirped pulse. With a further increase in the ratio, the substructures will become quite pronounced and complicated, while the overall half-width in time increases still gradually, as more and more higher order Hermite polynomials are added into the envelope. Since the nonlinear phase dispersion causes a substantial broadening in pulse shape, it is necessary to exercise care in interpreting the discrepancy existing between the bandwidth-limited time duration and the measured ones. The existence of this nonlinear term can perhaps be detected in the following type of measurements. 1) The third or higher order intensity correlation measurements: if the substructures are distinct enough, the trace can have sidebands around the central peak. 2) The two-dimensional two-photon fluorescence slope measurements as performed by Treacy.[4] According to his results, the slope seems to depart from a straight line especially at the edge. This can be attributed to the nonlinear phase dispersion.

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R.10 Effects of Fluctuations on Higher Order Correlation Measurements of Mode-Locked Laser Pulses, T. J. Hylden, D. M. Kim, T. A. Rabson, P. L. Shah, and F. K. Tittel, *Department of Electrical Engineering, Rice University, Houston, Tex. 77001.*

This paper will consider the effects of fluctuations in the mode spectrum of a mode-locked laser pulse on the two-photon fluorescence (TPF) display produced when the fundamental pulse interacts with its nth harmonic in a suitable fluorescent material. The primary result of the paper is that the signal portion of the TPF display more closely approximates the temporal characteristic of the original mode-locked pulse as one goes to higher orders in the correlation function. However, for a given experimental setup with given fluctuations in a fundamental pulse, it is also shown that increasing the order of the correlation function tends to obscure the signal-to-noise ratio of the TPF trace. In effect, therefore, one can get no more information from a higher order TPF trace after a certain order has been reached.

The analysis of higher order correlations with fluctuations is based upon the Fourier series representation of the field where each complex mode amplitude is considered as an independent stochastic process. The distribution function describing each mode is described by its variance, and in this way the amplitude and phase fluctuations of the field are taken into account. The fundamental laser pulse is assumed to be, on the average, describable by a Gaussian time-dependence envelope. We then consider various types of conventional and higher order TPF measurements.

We begin by deriving an analytic expression, the conventional TPF trace:

$$\mathcal{F}(\tau) = \mathcal{F}_0[1 + 2e^{-1/2(\Delta\Omega\tau)^2} + 8\sqrt{2}\, f(1 + \tfrac{1}{2}e^{-1/4(\Delta\Omega\tau)^2})].$$

This is the fluorescence trace of an input pulse assumed to be of the form $\mathcal{E}_0 e^{-(\Delta\Omega\tau)^2} \cos \omega_0 t$ with fractional fluctuations of its component modes in Fourier space described by the parameter f. The first term results in a constant background term and is a single-pass fluorescence contribution of the fundamental beam in the dye. The second term is the signal and is seen to approximate the input pulse, though its width is larger by a factor of $\sqrt{2}$. The third and fourth terms arise because of the fluctuations present in the input beam. The third term is a constant intensity background term which degrades the contrast ratio $\mathcal{F}(\tau = 0)/\mathcal{F}(\tau = \infty)$ [which is 3:1 for $f = 0$ and 1.5:1 for $f \to \infty$], and the fourth term alters the time dependence of $\mathcal{F}(\tau)$, further obscuring the original signal.

We next consider a different type of experimental situation in which the fundamental laser pulse is mixed with its second harmonic. We show that the TPF produced by two such pulses is proportional to the third-order intensity correlation function of the field and gives a fluorescence trace that more closely approximates the input pulse. For an experimental situation in which only a fundamental pulse propagates in one direction in the fluorescent dye, and only its second-harmonic propagates in the opposite direction, one can eliminate the constant-intensity background terms in the fluorescent trace. The result is

$$\mathcal{F}(\tau) = \mathcal{F}_0\left[\frac{1}{\sqrt{3}} e^{-2/3(\Delta\Omega\tau)^2} + 2\sqrt{2}\, f + 8f'\right].$$

f is a parameter resulting from the fluctuations in the amplitudes and phases of the fundamental beam modes and f' is a fluctuation in the second-harmonic pulse, which arises from the action of the nonlinear crystal on the fundamental. Note that for zero fluctuations, this gives an ideally infinite contrast ratio. Also, it is clear that the time dependence of $\mathcal{F}$ more closely approaches that of the input.

The generalization of the above equation to the interaction of the fundamental with its nth harmonic yields

$$\mathcal{F}(\tau) = \left\{\frac{1}{\sqrt{n+1}} e^{-[n/(n+1)](\Delta\Omega\tau)^2} + 4\left(2f_n + \frac{f}{\sqrt{n}}\right)\right\}.$$

Clearly, as n gets larger the signal portion of the signal approaches exactly the shape of the input pulse. The analysis also yields that $f_n \approx nf$. In other words, the fluctuations in the fundamental are, in a sense, amplified by the harmonic generation process. The result of this is that, for a given fluctuation, going to higher order correlation measurements of the field eventually reaches a point where the signal-to-noise contrast ratio has become so degraded by the fluctuations that the signal portion of $\mathcal{F}(\tau)$ is completely obscured.

R.11 Direct Observation of Picosecond Structure in the Amplified Output of a Pockes Cell Q-Switched Nd:Glass Oscillator,[1] L. W. Coleman, R. L. Carman, R. L. Schriever, and J. E. Swain, *Lawrence Livermore Laboratory, Livermore, Calif. 94550.*

There have been many previous reports of the observation of picosecond structure in the output of wide-bandwidth normally Q-switched lasers.[2,3] Typically, investigators used the two-photon absorption fluorescence technique (TPF) for observing the effect.

We report on recent studies at the output of the oscillator and will compare it with the output observed at various amplifier stages in the LLL "long path" Nd:glass disk laser system. These measurements have revealed the existence and preservation of considerable picosecond time structure in the pulse. The principal diagnostic tool used for these measurements was the LLL ultrafast streak camera.

The oscillator is a Pockel cell Q-switched Nd:glass system, employing a second Pockel cell for cavity dumping. Outside the cavity, a second pair of Pockel cells is then used to clip the cavity-dumped pulse down to ~2-ns duration. The spectral width of the ~2-ns burst is ~50 cm^{-1}. The rod amplifier chain then consists of seven 1/2-in diameter by 9-in long amplifiers followed by three 1-in diameter and 20-in long amplifiers, where each amplifier is placed one (low intensity) Rayleigh range from the proceeding rod. Less than 1 mJ out of the pulse clipped oscillator produces 10 J out of the last 1-in amplifier. The pulse is then fed into the final 9 pass 5.5-in disk laser amplifier.

The streak camera used had a streak rate of 25 ps/mm with a 70-mm display capability. The measured resolution is ~10 ps corresponding to a detectivity of ~5 ps. Camera sychronization is accomplished by an optically triggered avalanche transistor circuit (requiring microjoules to trigger) that is used to directly generate both the necessary gating and deflection voltages.

The streaked slit photographs of the oscillator output show amplitude modulation with a period of ~200-ps spacing superimposed on a regular uniform ~20-ps modulation for the entire duration of the ~2-ns pulse. Simultaneous spectra, TPF, and streaked slit photographs made where possible in the amplifier chain show that the shortest pulse (bandwidth limited) could be <0.5 ps but was only measured to be ≈2.5 ps by TPF.

The streak photographs taken on spectroscopic film are highly adaptable to optical Fourier analysis. The reproducibility and regularity of the picosecond structure can be readily established using the spatial transform of the observed temporal structure. In addition, the changes in the temporal structure, as the pulse is being amplified, can be readily evaluated again with this technique. Such an analysis has indicated that little change in the temporal structure is observed through the rod amplifier stages. Since no amplifier saturation has occurred, this implies that no other nonlinear effects are important in the driver chain.

Both the TPF and Fourier transform techniques verify the existence of a 20-ps periodic structure. The TPF shows secondary maximum at ~10 and ~30 ps which is also in agreement with the spatial Fourier transforms of the streak photographs.

The effects of amplifier saturation and possible self-phase modulation in the disk laser are now being investigated. Also, we will discuss the changes in system performance after installation of a time–bandwidth product ~2-ns duration pulse generator.

[1] This work was supported by the U. S. Atomic Energy Commission.

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SESSION S

Thursday, May 11, 1972 8:30 A.M.–12:00

Spectroscopy III

S.1 Infrared-Microwave Double Resonance in Molecules (*Invited*), K. Shimoda, *Department of Physics, University of Tokyo, Tokyo, Japan, and Institute of Physical and Chemical Research, Wako, Japan.*

Although the infrared gaseous laser is tunable only within a narrow range, there exist some molecular three-level systems in which one of the transition frequencies is very close to the laser frequency while the other is in a conventional microwave region. Infrared-microwave double resonance in molecules has recently been studied by using the infrared lasers.

Powerful CO_2 lasers were employed in observing double resonance effect in CH_3Br by Ronn *et al.*[1] and in CH_3Cl by Frenkel *et al.*[2] Infrared transitions in these experiments were not resolved, however. Double-resonance signals in the well-resolved transitions have subsequently been observed in $^{14}NH_3$ and in $^{15}NH_3$ by Shimizu and Oka by using an N_2O laser,[3] and in H_2CO by Takami and Shimoda by using a He–Xe laser.[4]

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