Coherent effects in a self-mode-locked Ti:sapphire laser

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The spectral and temporal characteristics of ≈10-fs pulses from a self-mode-locked Ti:sapphire laser were found to exhibit large deviations from the predictions of current theories of mode locking. A simple model that takes into account coherent coupling between the circulating pulse and the gain medium gives results in good agreement with experimental observations.

The technique of self-mode locking[1] is now routinely exploited to generate sub-100-fs pulses from solid-state lasers in the near infrared. Optimization of a solitonlike interplay between negative group-delay dispersion (GDD) and self-phase modulation (SPM) and the minimization of high order dispersive perturbations[2] have resulted in sub-20-fs pulse generation from Ti:sapphire lasers.[3–6] In particular, minimization of the cubic phase distortion from intracavity prisms[7–9] by use of an operating regime close to the zero-cubic-dispersion wavelength of fused silica (≈850 nm) generated exceptionally broad mode-locked spectra with FWHM bandwidths as broad as 150 nm and pulse durations of ~10 fs.[9] A mode-locked spectrum typical of this operating regime is shown as the solid curve in Fig. 1(a), with the corresponding fringe-resolved autocorrelation trace shown in Fig. 1(b). The prominent double-peaked structure of the optical spectrum, the modulation in the envelope of the autocorrelation, and the calculated time–bandwidth product of >0.6 (Ref. 9) represent a striking deviation from the expected near-sech^2 mode-locked pulses. In this Letter we show that these experimental results can be understood with a simple qualitative model based on coherent pulse propagation effects.

Existing models of passively mode-locked lasers[10–12] use a rate-equation approach, which is justified if the pulse bandwidth is small compared with the transition bandwidth. As the two become comparable, however, the coherent interaction between the circulating pulse and the laser gain medium can lead to effects that are not predicted by conventional mode-locking theories. In the mode-locked argon laser, coherent pulse propagation causes an oscillatory ringing structure on the pulse envelope as a result of the coherent Rabi oscillation of the atomic polarization in the gain medium.[13,14] This coherent interaction is observed in a high-Q configuration of the mode-locked argon laser in which the interaction Rabi frequency Ω_R can be comparable with or larger than the transition linewidth.[15] In contrast, mode-locked solid-state lasers cannot meet this condition for strong coherent coupling because their broad bandwidth and small transition dipole moment imply that Ω_R is much smaller than the homogeneous linewidth. In the 10-fs Ti:sapphire oscillator described in Ref. 9 the peak Rabi frequency is less than 10 THz. Since this value is small compared with the homogeneous linewidth of Ti:sapphire (≈100 THz) there is little dynamic gain saturation during the interaction between the mode-locked pulse and the gain medium, and strong-signal coherent coupling is not expected.

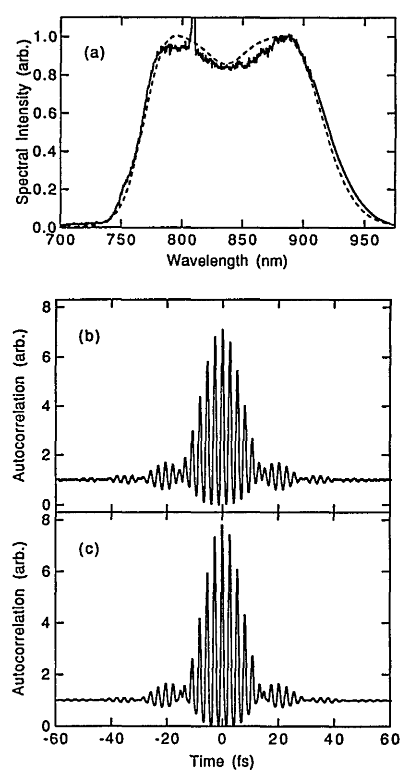

Fig. 1. (a) Mode-locked spectrum and (b) corresponding fringe-resolved autocorrelation of a self-mode-locked Ti:sapphire laser operating in the 10–fs regime. The narrow spike in the spectrum relates to a low-intensity background with negligible energy content. (c) Calculated fit to (b), assuming a pulse envelope with coherent ringing, with the dashed curve in (a) representing the corresponding calculated spectrum.

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However, because the pulse width is comparable with the dephasing time T_2, the atomic polarization cannot instantly respond to the envelope of the electric field, and the delayed polarization response can still cause deviations from rate-equation long-pulse behavior.

Although a rigorous treatment of the effect of this delayed coherent polarization would also need to include both GDD and SPM, here we examine the possibility of coherent pulse propagation with a simple qualitative analysis. The equations describing the coherent interaction between an incident light pulse and a homogeneously broadened two-level atomic system are

$$\frac{\partial P(z,t)}{\partial t} + \left[\frac{1}{T_2} + i(\omega_0 - \omega_a)\right]P(z,t) = i\frac{\mu^2 \Delta N}{\hbar}E(z,t), \quad (1)$$

$$\frac{\partial E(z,t)}{\partial z} = -i\frac{\omega_a}{2\epsilon_0 cn}P(z,t), \quad (2)$$

where $E(z,t)$ and $P(z,t)$ are the complex electric-field and resonant atomic polarization envelopes propagating along the z axis, t denotes the local time, ω_a and ω_0 represent the center frequencies of the laser transition and the mode-locked spectrum, respectively, and μ is the transition dipole moment. All other symbols have their usual meanings. The population inversion ΔN can be regarded as constant owing to the small dynamic gain saturation in the Ti:sapphire laser. We can estimate the effect of the delayed atomic response on a short pulse traversing the gain medium by considering an incident pulse $E_{\text{in}}(t)$ that is significantly shorter than T_2. Assuming a small perturbation (low-loss cavity), we can solve differential equations (1) and (2) to give the change in the pulse shape on passage through the laser medium $E_{\text{out}}(t) - E_{\text{in}}(t)$ as approximately

$$\Delta E(t) \sim \exp(-t/T_2)\exp[-i(\omega_0 - \omega_a)t]. \quad (3)$$

Note that, for a pulse comparable with or longer than T_2, $\Delta E(t)$ can be shown to include an additional component that follows approximately the incident pulse shape. The frequency of the oscillatory perturbation in relation (3) is determined by the detuning $|\omega_0 - \omega_a|$, and the amplitude is damped on a time scale T_2. In a mode-locked laser configuration, however, the perturbation could accumulate over many round trips and could result in a ringing behavior on the trailing edge of the circulating pulse. It should be stressed that this ringing is caused merely by the delayed coherent response of the atomic polarization rather than by a Rabi-type flopping of the population inversion, as is the case in the argon laser.[14–16]

We model the electric-field envelope of the pulse with coherent ringing as a sequence of four Gaussian pulses with identical pulse durations and temporal separations but decaying amplitudes of alternating signs:

$$E(t) = E_0\{\exp(-\beta t^2) - \alpha_1 \exp[-\beta(t-T)^2] + \alpha_2 \exp[-\beta(t-2T)^2] - \alpha_3 \exp[-\beta(t-3T)^2]\}. \quad (4)$$

Here $\beta = 2\ln 2/\tau_0{}^2$, where τ_0 is the pulse duration (intensity FWHM). The relative amplitudes of the subsidiary pulses are denoted α_1, α_2, and α_3, respectively, and T is the temporal delay between subsequent pulses. With this model a fit to the autocorrelation data in Fig. 1(b) yields optimum values for the pulse parameters of $\tau_0 = 11.2$ fs, $\alpha_1 = 0.32$, $\alpha_2 = 0.073$, $\alpha_3 = 0.018$, and $T = 16$ fs. The resultant electric-field and intensity envelopes are shown in Fig. 2. According to our simple model, the separation of the subsequent pulses is given by $\pi/|\omega_0 - \omega_a| \approx 18$ fs, in good agreement with the experimental result [Fig. 1(b)].

For comparison with the measured data, the calculated autocorrelation function is depicted in Fig. 1(c), and the calculated spectrum is shown as the dashed curve in Fig. 1(a). The similarity between these calculated results and those obtained experimentally is striking. In particular, the calculated interferometric autocorrelation reproduces the π phase change in the fringe structure observed experimentally, and these phase changes are now interpreted physically as corresponding to points where the amplitude of the pulse envelope changes sign. The theoretical fit to the measured data is not unique, of course, and it would also be possible to obtain the same fit by use of an equal-amplitude set of leading pulses. It is difficult, however, to identify plausible physical processes in the self-mode-locked Ti:sapphire laser that would generate such a perturbation.

One physical mechanism that could potentially account for the observed pulse characteristics is third-order dispersion, because it is well known that such dispersion can lead to an oscillatory structure near one of the pulse edges.[17] As is shown in Fig. 3(a), however, as negative GDD is reduced in the laser cavity, third-order dispersion would simultaneously make the spectrum increasingly asymmetric, shift its center wavelength, and give rise to the emergence of a dispersive resonance.[4,18] It can be seen from Fig. 3(b) that neither of these characteristics of third-order dispersion is observed with the laser

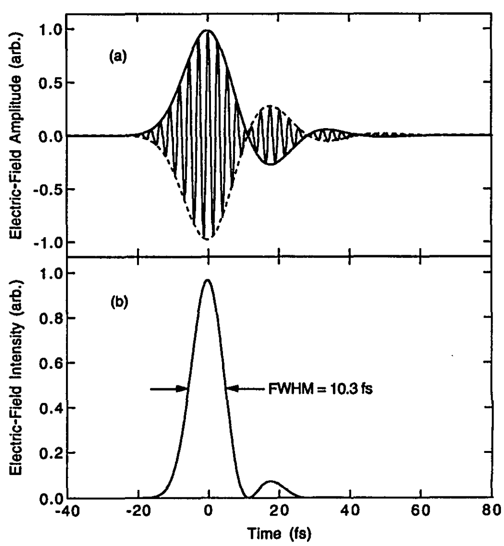

Fig. 2. (a) Pulse envelope and (b) intensity, illustrating the coherent ringing.

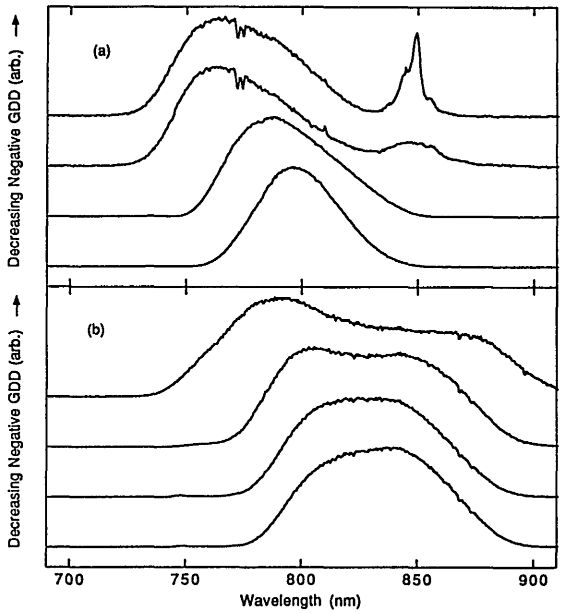

Fig. 3. Evolution of the mode-locked spectrum in the Ti:sapphire laser near (a) 780 nm, where cubic dispersion is the dominant perturbation, and (b) 840 nm, where cubic dispersion is negligible, for decreasing negative GDD in the cavity. All data are corrected for detector response.

tuned to ≈840 nm; hence third-order dispersion can be ruled out as the responsible effect, in agreement with the results of cavity third-order dispersion calculations.[9,19] It can also be seen that a negative fourth-order dispersion characteristic of a prism-controlled oscillator[9] does not modify qualitatively the phase dispersion of a cavity having negative GDD, and hence it cannot account for the observed spectral evolution in Fig. 3(b) either.

The separated action of SPM and GDD[2] and possibly the finite response time of the Kerr nonlinearity[17] are other sources of perturbations to the circulating pulse, but neither of these effects fully explains the experimental results. We also note that, although there is some apparent similarity between the results reported here and the recent observation of self-spectral splitting in the self-mode-locked Ti:sapphire laser,[20] a more detailed examination of the results indicates that the two phenomena appear to be unrelated. In particular, self-spectral splitting is associated with a distinct laser operating regime and is accompanied by the complete splitting of the mode-locked spectrum. Neither of these observations was noted with our laser configuration. To avoid the large time–bandwidth products associated with coherent ringing, one must operate the laser near the peak of the transition so that the off-resonant delayed coherent polarization does not introduce a significant perturbation into the circulating pulse. Correspondingly, one must be careful when evaluating pulse durations from autocorrelation functions obtained in the presence of coherent effects. Although a more detailed quantitative model that includes SPM and GDD, as well as coherent coupling in the gain medium, will lead to more insight into the physics of femtosecond laser oscillators, our preliminary investigations indicate that coherent pulse effects may play an important role.

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