Generation of Coherent Pulses of 60 Optical Cycles Through Synchronization of the Relaxation Oscillations of a Mode-Locked Dye Laser

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Coherent interaction experiments on single photon transitions have so far been restricted to dilute vapors [1] (collision times in the nanosecond range), or "frozen media" [2] (like ruby cooled down to liquid He temperature, for which the phase relaxation time of some transitions can exceed 100 ns[3]). There is a need for a stable, highly coherent source of pulses of about 0.1 ps to apply these diagnostic techniques to complex molecules at room temperature. We report the successful development of such a source, yielding bandwidth limited pulses of 0.12 ps duration with 300W peak power.

In the design of ultrashort mode locked dye lasers, an essential consideration is the absence of intracavity dispersion in the wavelength range of interest. To this end, the cavity is reduced to a bare minimum: no other intracavity element than the dye jet, which contains a solution of Rh6G, DODCI and malachite green in ethylene glycol is used [4]. The laser output mirror is a "third order reflector" with a flat reflectivity in the range 590 to 620 nm which defines the spectral range of operation. The dye mixture and mirror reflectance that give the shortest pulses are critical and interrelated. For instance, at a longer wavelength (using a mirror with a reflectance from 605 to 640 nm) the optimum dye mixture contains a higher concentration of DODCI.

When pumped close to threshold by a CW argon ion laser, the laser emits a train of mode-locked pulses. If the cavity does not exceed 60 cm, the pulses are spaced by the cavity roundtrip time, forming trains which contain pulses as short as 0.16 psec. The shortest pulses are emitted in bursts of a few μsec duration with an average periodicity of 20 μsec. These bursts, which are detected as peaks in the second harmonic signal, coincide with relaxation oscillations in the laser intensity, as shown in Fig.1. For larger pump powers or even at thresh-

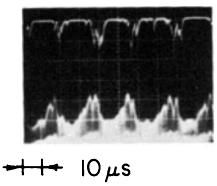

Fig.1 The laser intensity relaxation oscillations which coincide with bursts of short pulses

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hold for cavity lengths exceeding 60 cm, a continuous train of pulses is generated at twice the cavity round trip rate (two pulses per cavity round trip time). Pulse repetition rates of 600 MHz or more can be observed. For the stable generation of the shortest pulses, two stability problems have to be addressed:

a) long term stability: the laser has to operate a few mW above the threshold (1.5W) which evolves with dye temperature, age, and cavity losses.

b) short term stability: the randomness in the time of arrival of the short pulse bursts limits the use of this source, in particular, in combination with amplifier chains which have to be timed accurately.

Both long and short term stability problems have been corrected, the former by the use of active feedback, the latter through the use of a pulsed modulation in the argon laser pump.

Long Term Stabilization

Unlike synchronously mode-locked systems, we measure that optimum short pulse generation coincides with a maximum in the average second harmonic of the dye laser output. Long term stabilization is attained by adjusting the argon laser intensity, using a voltage responsive remote control, to maintain the second harmonic of the laser output at its peak. This is done by making phase sensitive detection of a low frequency modulation (10 Hz) introduced into the second harmonic through the argon laser remote control. The in-phase component of that 10 Hz oscillation is measured with a lock-in amplifier and fed back into the argon laser remote control. The feedback loop seeks stable equilibrium at any local maximum of the second harmonic output. Hence the laser will stabilize at the shortest pulse in the single pulse or the two pulse per cavity roundtrip time mode, depending on the initial pump power setting.

Short Term Stabilization

The ultrashort pulse operation of the dye laser appears to be related to the relaxation oscillation observed at a frequency of 50 kHz. By pulse modulating the argon laser intensity at that same frequency we are able to synchronize these relaxation oscillations, thus removing the randomness in the short pulse production. A low voltage modulator is used to periodically reduce the pump power intensity below threshold, for a few microseconds, with an adjustable repetition rate. Fig.2 shows the evolution with time of the envelope of the synchronized pulse train, as measured by an avalanche photodiode. It should be noted that the particular shape (in the microsecond time scale) of the pulse envelope as well as the duration of the pulses along the train depend strongly on the exact frequency of the dips in the pump power.