A cw rhodamine 800 dye laser passively mode-locked with neocyanine

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There is considerable interest in the development of femtosecond lasers operating in the near infra-red around 800 nm. This spectral region is important for the time-resolved spectroscopy of semiconductor materials, for the possibility of frequency doubling to the blue spectral region and for amplification of femtosecond pulses to terawatt and even petawatt peak powers in vibronic solid state lasers such as titanium-doped sapphire and alexandrite. Mode-locked dye lasers have provided a convenient means to generate ultrashort pulses in this region, first using synchronous mode-locking of dye lasers to generate picosecond pulses and then employing hybrid [e.g. 1–3] or passive [e.g. 4–9] mode-locking to reach the femtosecond regime.

The infra-red dyes whose absorption profiles match the wavelengths of the most common pump lasers in the green (e.g. styryl dyes) are unfortunately unsuitable for passive mode-locking because of their relatively short upper state lifetimes. The dyes rhodamine 700 and rhodamine 800 can be passively mode-locked but they must be pumped either using a krypton ion laser in the deep red [4,6,8,9] or using the technique of energy transfer with a composite gain medium [5,7]. The former approach has worked well but only the performance at particular wavelengths was discussed. Ref. [7] described a tunable rhodamine 800 dye laser, passively mode-locked with neocyanine 1(1-bis-4-(1-ethylquinolinyl)-1-(1-ethylquinolinyl)pentamethine cyanine diiodide) in a linear cavity, which employed an energy transfer from pyridine 1 to facilitate pumping with an argon ion laser. The relative inefficiency of this energy transfer limited the minimum pulse duration to 260 fs and the tuning range to 783–815 nm. In this letter we report on an extension to this work in which the rhodamine 800 was excited directly using a krypton ion laser.

The improves efficiency of the laser amplifier permitted a collapsing-pulse ring configuration with six cavity mirrors to be constructed which was similar to that commonly employed in standard CPM lasers and which is shown in fig. 1. Owing to the relatively low dispersion of quartz at 800 nm, the Brewster-angled prisms were made of flint glass. All the cavity mirrors supported narrow-band, single-stack dielectric coatings which were of 100% reflectivity except mirror M3, which was of 98% reflectivity. Mirrors M2 and M4, which comprised the active folded section, were of 75 mm radius of curvature and the passive folding mirrors, M3, and M5, were of 35 mm. A 50 mm radius of curvature mirror, M6, coupled the pump light (the 647 nm line of Spectra-Physics krypton ion laser) into the amplifier dye jet which was of a 200 µm thickness. The cavity round trip time was 6 ns. Tuning of the laser was accomplished by translating an aperture, A, located at the midpoint of the prism sequence where there was a spatial distribution of the laser frequency spectrum. The amplifier was a 2×10⁻⁴ M solution of rhodamine 800 in a 1:9 mixture of propylene carbonate:ethylene glycol. The saturable absorber, neocyanine 1 in-
ylene glycol, varied between $7 \times 10^{-4}$ and $10^{-3}$ M and flowed through a 100 µm jet stream. Fig. 2 shows the absorption profile and structural formula of neo-cyanine. The shaded area represents the spectral region over which the laser was mode-locked.

Pulse durations were measured using the standard collinear second harmonic generation autocorrelation technique with lithium iodate as the frequency doubling crystal. Sub-500 femtosecond pulses were generated from 779 nm to 820 nm with the shortest pulses of ~110 fs duration being observed at ~800 nm. Fig. 3 shows the autocorrelation trace of these pulses. The optimised laser typically produced ~10 mW average output power when pumped ~2 W above threshold (~1 W) and routinely operated with only one pulse per cavity round trip (in each direction) although multi-pulsing regimes were observed. The stability of the laser followed that of the krypton ion pump.

**Conclusions.** A rhodamine 800 CPM ring dye laser has been demonstrated using neo-cyanine as the saturable absorber. Sub-500 femtosecond pulse generation has been obtained from 779 nm to 820 nm. This
system is an improvement on the energy transfer laser of ref. [7] and should prove to be a useful source for time-resolved spectroscopy and for exploiting the new vibronic laser amplifiers in the near infra-red.

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