observed even with aluminized mirrors overcoated with MgF₂ with a reflectivity of only 81 and 84% (i.e., 35% round-trip cavity loss). This type of ion laser can serve as a convenient probe for noble-gas excimer and dimer laser gain or loss. KrF can be probed using the Ne IV line at 247.3 nm, KrBr probed using the Ne IV line at 206.5 nm, ArF probed using the Kr IV line at 195.0 nm, and both Ne or ArCl probed using the Kr IV line at 175.6 nm. Ion laser transitions near 200 nm have already found biological use as illumination sources in photoelectron emission microscopy. Additional applications for this simple source of vacuum ultraviolet laser emission are apparent, such as in photochemistry and kinetics.


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Bandwidth-limited picosecond pulses from a neodymium-phosphate glass oscillator

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Direct measurements using a picosecond streak camera have shown that transform-limited pulses having durations ∼5 ps are produced throughout the pulse train of a mode-locked neodymium-phosphate glass laser operating in the TEM₀ₐ mode. Under the conditions of higher-power multimode operation, bandwidth-limited pulses are generated in the initial part of the train only. Spectral and temporal broadening due to intensity-dependent nonlinear effects are observed as the higher-power mode-locked trains evolve.

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Over the past few years, the spectral and temporal properties of mode-locked Nd: silicate glass lasers have been studied extensively. Early measurements of pulse widths were based primarily on the two-photon fluorescence (TPF) technique, where durations were averaged over entire pulse trains. Later, studies of single pulses using both the TPF technique and direct streak camera measurements showed that transform-limited pulses, as short as 3 ps, were produced only at the beginning of the mode-locked train. As the trains evolved, spectral and temporal broadening occurred with observed pulse durations in excess of 10 psec.

We have carried out a similar appraisal of the mode-locking of Nd: phosphate glass which has lower values of n₂ and d(n²)/dT than silicate glass. The 166.5-mm Brewster-angle Nd: phosphate glass rod (3.3% doped with Hoya LHG-5) was used in the same pumping geometry as previously described. The cavity configuration consisted of a 2-mm radius-of-curvature concave mirror and a plane-wedged mirror of 100 and 70% reflectivity, respectively, placed ∼ 95 cm apart. The mode-locking dye used was Eastman Kodak 9860 in 1, 2-dichloroethane (∼2 × 10⁻⁴ M) with a low light level transmission ∼80%. It was contained in an adjustable-width dye cell, normally set at 100 μm, such that it was in optical contact with the 100% reflector. The dye cell window was made from red filter glass to prevent photodecomposition of the dye by stray light. A 2-mm-diam aperture was placed in the cavity ∼10 cm from the output reflector for single transverse mode operation. The electrical energy discharged through the flashlight for operation in TEM₀ₐ mode was ∼300 J, and the laser was fired at 1-min intervals.

Completely mode-locked outputs were obtained with a success rate ∼80% in the TEM₀ₐ mode and ∼90% when the laser was operated multimode. Figure 1(a) shows a typical oscillogram for a TEM₀ₐ train of pulses lasing at 1053 nm, where the average pulse energy is typically ∼30 μJ.

The spectral characteristics of the output were recorded on Kodak I-Z infrared sensitive photographic plates using a Monospek 1000 1-m spectograph. Fig-
Figure 1(b) is a microdensitometer trace of the spectrum associated with the entire pulse train. The FWHM of 0.28 mm (Δν = 7.6 × 10^{10} Hz) for this profile shows that there is no apparent frequency broadening due to self-phase-modulation throughout the pulse train at these power levels.

A Photochron I streak camera with an S-1 photocathode was used to measure the duration of individual pulses selected at various locations in the mode-locked pulse trains by a Pockels cell switch as described earlier. The instrumental time resolution of this camera at 1060 nm is 2.5 psec.

Near the beginning of the TEM_{00} pulse envelope (~150 nsec before the peak maximum) the recorded pulse widths were in the range 3–8 psec, with the peak of the distribution occurring at ~6 psec. The corresponding spectral widths were within the range 0.25–0.6 nm. Generally the time-bandwidth product ΔνΔt was 0.4–0.6, indicating that the pulses were usually transform limited. Figure 2(a) shows a microdensitometer trace of the streaked images of a 5.6-psec pulse and Fig. 2(b) shows the corresponding spectral profile of a group of five pulses switched at the beginning of the pulse train, for which ΔνΔt = 0.44.

Pulses selected from the peak region of the train had durations in the range 3–8 psec (see Fig. 3). For the typical temporal and spectral profiles shown in Figs. 2(c) and 2(d), respectively, ΔνΔt = 0.46, which indicates that the pulses are still transform limited. Near the end of the train (~200 nsec after the peak) the recorded pulses retained bandwidth-limited durations, typically between 3 and 8 psec and with the maximum of the distribution ~5 psec. From the profiles in Figs. 2(e) and 2(f), ΔνΔt = 0.44.

When operated multimode, however, pulse evolution both temporally and spectrally becomes similar to that for silicate glass. The mode-locked trains were longer, ~1 μsec, and the average recorded single pulse energy...
was ~0.7 mJ for a total flashlamp input energy of ~230 J. The integrated spectral output of the entire pulse train [Fig. 4(c)] clearly exhibited the distinctive features of frequency broadening by self-phase-modulation. In this case streak camera measurements of pulses selected from the early stages of the mode-locked train indicated transform-limited durations in the range 4–9 psec. Figures 4(a) and 4(b) respectively show typical corresponding temporal and spectral records of pulses selected approximately 300 nsec before the peak of the pulse train where $\Delta \nu \Delta t = 0.45$. Temporal and spectral broadening and pulse breakup became evident as the mode-locked trains evolved. Towards the end of the train, measured pulse widths were in the range 4–16 psec and bandwidths of 4.8 nm ± 1.1 nm ($\Delta \nu = 13.0 \pm 3.0 \times 10^{11}$ Hz) were comparable to those reported for the silicate glass laser.6

From these direct experimental results we conclude that transform-limited pulses can be produced throughout the entire mode-locked train of a Nd:phosphate laser, provided the power density in the active medium is limited to ~150 MW cm$^{-2}$, such that no significant self-phase-modulation or self-focusing effects arise.12 This would be consistent with the generation by Kuroda et al.15 of 1-mJ bandwidth-limited 5-psec pulses that had been selected near the beginning of a TEM$_{00}$ train. From streak camera measurements made at the second harmonic frequency Fleming et al.15 have reported that no transform-limited pulses were generated by their LHG-5 laser. This is in marked contrast to our results, because their average pulse energy of ~1 mJ and power density are similar to that of our system in multimode operation where the initial pulses in the train are bandwidth limited. Therefore it is probable that their switching technique only permitted pulses in the latter stages of the pulse train to be examined, where broadening had already taken place.

In summary, the mode-locked Nd:phosphate glass oscillator operated in the TEM$_{00}$ mode at relatively low power levels provides a convenient source of high spatial, temporal, and spectral quality picosecond pulses. In addition the repetition rate capability of other phosphate glasses, e.g., LHG-7,14 offers a potentially excellent pulsed optical source for applications in picosecond spectroscopy.

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