# Hybrid time-resolved spectroscopic system for evaluating laser material using a table-top-sized, low-jitter, 3-MeV picosecond electron-beam source with a photocathode

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Hybrid time-resolved spectroscopy of laser media comparing electron-beam excitation and optically excited cases is performed using a newly developed, table-top-sized, low-jitter, 3-MeV picosecond electron-beam source with a photocathode. The properties of an electron-beam-pumped $Ce^{3+}$:$LiCaAlF_6$ (Ce:LiCAF) ultraviolet laser medium significant differ from those of an optically pumped medium. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1476384]

The history of ultrafast spectroscopy has almost dominantly been driven by the rapid progress of ultrafast lasers devices. Time-resolved radiolysis is a potentially powerful tool for optical material research and development, including gallium nitride[1] and zinc oxide[2] for a blue light-emitting diode and potential laser media that may be directly pumped by an electron-beam (e-beam). Some time-resolved radiolysis systems have already been developed with linear accelerators.[3–5] However, most of these systems are not laboratory sized, and they deliver electron beams with electron energy too high for solid-state spectroscopy. Such restrictions have significantly slowed the progress of ultrafast radiolysis and the expansion of these variations. In this letter, we describe a truly table-top-sized, hybrid fluorescence spectroscopy system with a low-jitter, picosecond, 3-MeV e-beam from photocathode and picosecond optical pulses. This system has shown that the fluorescence spectra and relaxation time constant properties of an e-beam pumped Ce:$LiCaAlF_6$ (Ce:LiCAF) ultraviolet laser medium to significantly differ from those of an optically pumped medium.

The experimental setup is illustrated in Fig. 1, where the total size of the system is approximately 2 m×3 m×1.5 m. This is much smaller than the previous linear accelerator with a photocathode for high-energy particle physics.[3] Details of the developed accelerator for material spectroscopy are described next. The photocathode rf gun was irradiated with 0.25-mJ, 262-nm, 5-ps, 10-Hz optical pulses from a Nd:$YLiF_4$ (Nd:YLF) regenerative amplifier seeded by a jitter-suppressed 79.33-MHz, mode-locked Nd:YLF oscillator synchronously operated with a 2.856-GHz, 6-MW klystron to accelerate the extracted 1-nC photoelectron beam. For the stable operation of the klystron, the acceleration energy was fixed to 3 MeV. The e-beam emittance was compensated by a solenoid magnet. The e-beam was irradiated onto the sample after being passed through titanium foil. A portion of the pumping optical pulse irradiated the same sample to compare the excitation scheme. The pulse duration of e-beam was evaluated by the Cherenkov radiation was measured by the streak camera to be 15 ps [full width at half maximum (FWHM)], as shown in Fig. 2. The optical pulse at 262 nm for exciting the photocathode and sample was measured to be 5 ps (FWHM) using the same streak camera in a single-shot mode. The temporal resolution of the total system including timing jitter is less than 30 ps (FWHM) even in the accumulation mode of the streak camera for over 5 min.

A well-developed tunable ultraviolet laser medium, Ce:LiCAF,[6,7] was used as a sample to examine the possibility of direct e-beam pumping.[8] Evaluating direct e-beam pumping of Ce:LiCAF is very important for fully utilizing recently grown large gain crystals (over 7 cm in diameter) for future chirped-pulse amplification laser system operating in deep

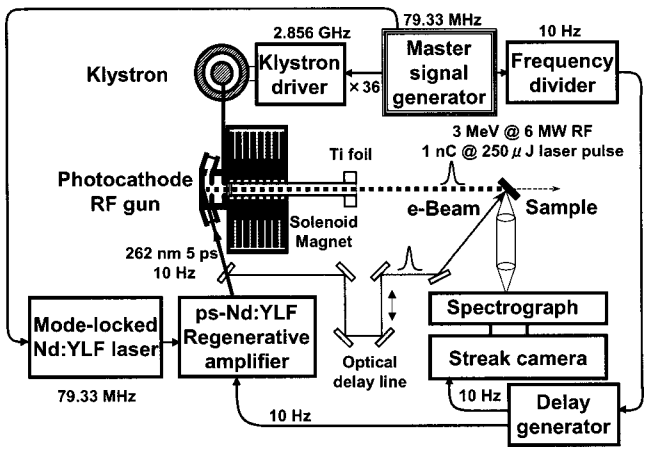


FIG. 1. Experimental setup. The photocathode rf gun was irradiated with 262-nm optical pulses from a Nd:YLF regenerative system synchronously operated with a 2.856-GHz, 6-MW klystron to accelerate the extracted 1-nC photoelectron beam. The e-beam was irradiated onto the sample after being passed through titanium foil. A portion of the pumping optical pulse irradiated the same sample to compare the excitation scheme. A streak camera was equipped with a 30-cm spectrograph to measure the fluorescence spectrum and fluorescence lifetime.

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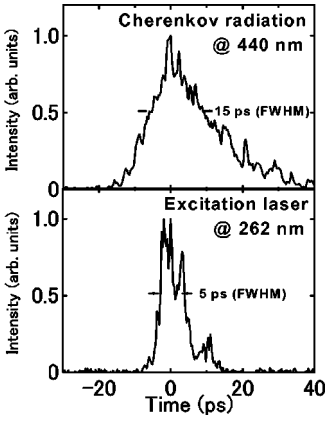


FIG. 2. Time profiles of Cherenkov radiation induced by the electron pulsed beam and the 262 nm laser pulse for excitation of photocathode or sample, measured by a spectrograph and a streak camera.

ultraviolet at multiterawatt level.[9] This is particularly important since the current bottleneck of such a laser system is the availability of 266-nm optical pulse energy for pumping.[10] Supporting this possibility of pumping, the excitation channel of Ce ion through the band gap absorption around 112 nm was found from photoexcitation spectrum measurement using synchrotron orbital radiation apparatus.[10] To evaluate the e-beam pumped properties of this material, a streak camera with a 30-cm spectrograph was installed to measure the fluorescence spectrum and fluorescence lifetime. The whole system is approximately the same size as a commercially available table-top terawatt laser system. The time-resolved fluorescence spectrum with e-beam pumping is shown in Fig. 3. Unlike the optically pumped case, broadband, short-duration Cherenkov radiation clearly overlaps the initial portion of the fluorescence from cerium ions. This is sufficiently short for the characterization of the various laser media. Compared with the optically pumped case in the same setup with a 262-nm picosecond optical pulse split from the photocathode excitation laser, the longer wavelength side of the spectrum is significantly enhanced, as shown in Fig. 4. Corresponding to this change, the fluorescence lifetime of this medium became slightly longer (44.2 ns) compared to the optically pumped case of 33.7 ns, as shown in Fig. 5. This longer lifetime could be attributed to the relaxation process

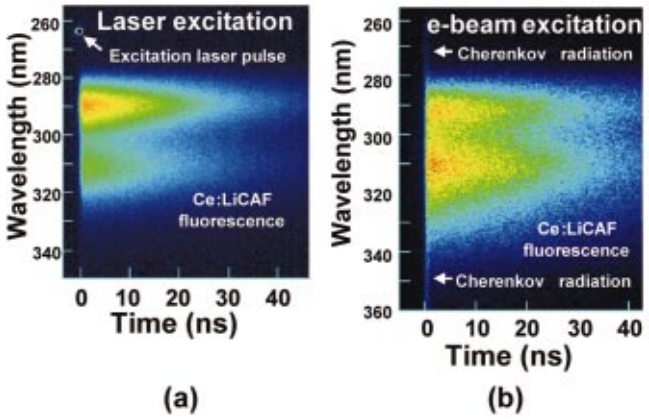


FIG. 3. (Color) Streak camera images of the fluorescence from Ce:LiCAF excited (a) by 262-nm pulse and (b) by electron pulse. In the case of electron pulse excitation, a broadband, short-duration Cherenkov radiation is clearly seen.

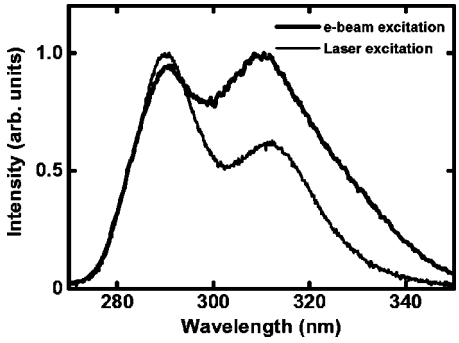


FIG. 4. E-beam and optically pumped fluorescence spectra. Compared with the optically pumped case in the same setup with a 262-nm optical pulse split from the photocathode excitation laser, the longer wavelength side of the spectrum is significantly enhanced.

of excited electrons from the host crystal to the active Ce ions. For a full explanation of the relaxation process, more detailed time-resolved spectroscopic experiments including temperature dependence and excitation e-beam energy dependence will be required additional to the time-resolved gain spectrum measurement. However, such hybrid time-resolved spectroscopy will significantly enhance electro-optical material developments.

In conclusion, a table-top-sized hybrid fluorescence spectroscopy system with a low-jitter, picosecond, 3-MeV e-beam from a photocathode and picosecond optical pulses was demonstrated. This system has shown that the properties of an e-beam pumped Ce:LiCAF: ultraviolet laser medium significantly differ from those of an optically pumped medium. The experimental results suggest that additional potential advantages of e-beam pumping of Ce:LiCAF for an ultrashort pulse amplifier can be derived from its modified fluorescence spectral shape and longer lifetime.[11]

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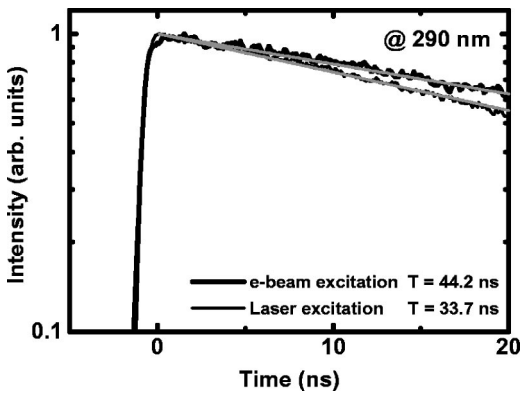


FIG. 5. Fluorescence decay curves of Ce:LiCAF excited by e-beam and by 262 nm pulse measured around 290 nm. The solid lines indicate the convolution curves with fixed lifetimes, 44.2 ns for e-beam and 33.7 ns for 262 nm. The lifetime from e-beam excitation was slightly longer than that from laser excitation.

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