

Novel technique for low-jitter dual-laser synchronization in a thin film deposition system

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The need for precise laser pulse synchronization in a dual-laser ablation system to optimize the quality of the deposited thin films has been previously demonstrated. We present, in this article, a novel technique for the synchronization of an excimer and a CO₂ laser with synchronization having a temporal fluctuation (jitter) of less than ± 14 ns. This is several times better than the best precision of temporal synchronization possible using traditional electronic techniques and is crucial for the application of dual-laser ablation in the manufacturing of thin films. Evidence for reproducibility in the ablation of targets using this system is presented by analyzing the initial stages of the ablated plasma using a time-gated charge coupled device imaging system. © 2001 American Institute of Physics. [DOI: 10.1063/1.1367359]

I. INTRODUCTION

We have previously established the need for precise laser pulse synchronization in a dual-laser ablation system to optimize the quality of the deposited thin films in an earlier article.¹ The synchronization of two laser pulses on the ultrafast time scale is important in a variety of other applications including various pump-probe experiments,²⁻⁵ double-resonance spectroscopy,^{6,7} nonlinear optics,⁸⁻¹⁰ and laser welding.¹¹⁻¹³ If the second laser pulse is derived from the first, i.e., if it is identical in wavelength and pulse duration, and the delays involved are on the nanosecond time scale or less, the use of beam splitting and synchronization by optical delay of the second pulse upon introducing a larger optical path length with respect to the first is sufficient to allow precise temporal synchronization. The same technique can be used if the second laser is the second, third, or multiple harmonic of the first laser pulse simply by introducing a nonlinear frequency conversion mechanism in the path of the second laser (such as second harmonic generation for frequency doubling, etc.). However, when the requisite time delays are larger than tens of nanoseconds, such a synchronization technique becomes progressively impractical with increasing delays that require corresponding unfeasibly long optical delay lengths.

Also, if the two laser pulses are derived from different sources, the issue of temporal synchronization becomes more complicated. Such a situation could occur either when the frequencies are nonharmonically related or are not easily attainable by frequency up-conversion techniques. In such cases, the conventional technique is to use an electronic pulse generator as the master oscillator for two separate lasers and to introduce variable delays for the two lasers by using electronic pulse delay generators. Although appropriate in some circumstances, these conventional electronic

techniques are not applicable when precise synchronization of two lasers with a temporal jitter of less than ± 25 ns is required. This situation is encountered in the practical application of a dual-laser ablation system discovered in our laboratory for large-area, defect-free growth of a variety of multicomponent thin films.¹⁴

This dual-laser system and its applicability to high-quality film growth is described elsewhere.¹ Briefly, this laser ablation system consists of an infrared CO₂ laser ($\lambda = 10.6 \mu\text{m}$) and an ultraviolet KrF excimer laser ($\lambda = 248$ nm) having pulse widths of approximately 150 and 20 ns, respectively. This pair of laser pulses is temporally synchronized and spatially overlapped on the target to be ablated for thin film deposition. Previous experiments have demonstrated that precise synchronization of these two laser pulses to within tens of nanoseconds is crucially important for the particulate-free deposition of thin films.¹⁵ The mechanism responsible for such an effect has also been discussed. We report, in this article, on the implementation of a novel optical synchronization technique for the two lasers in the dual-laser ablation system. This system is an important practical example of the synchronization of two laser pulses derived from independent laser sources that require delays greater than several tens of nanoseconds with reliably low shot-to-shot temporal fluctuations.

II. IDENTIFICATION OF THE SOURCE OF INTERPULSE JITTER

The first step in the reduction of the temporal jitter between the CO₂ and KrF lasers in the dual-laser ablation system is the determination of the primary source of the fluctuations. This can be done by isolating the individual contributions of each laser to the collective interpulse jitter.

A. Measurement of interpulse temporal fluctuations

An obvious approach for the synchronization of the KrF and CO₂ lasers in the dual-laser ablation system is to use the

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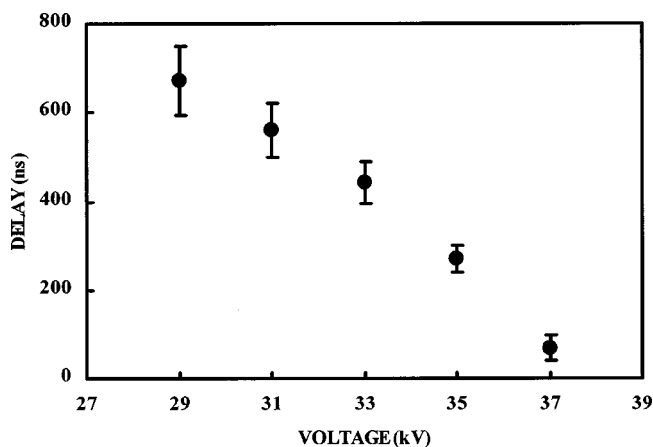


FIG. 1. Delay between the excimer laser pulse and CO₂ laser pulse as a function of the CO₂ laser discharge high voltage while using electronic interpulse synchronization.

electronic method discussed in Sec. I. The resultant ranges of temporal fluctuations in interpulse delay for 50 successive laser shots are shown in Fig. 1, as a function of the high voltage of the CO₂ laser discharge. Fluctuations as high as 155 ns at low values of the discharge voltage are somewhat alleviated at the higher laser voltages where the minimum observable fluctuation at a CO₂ discharge voltage of 37 kV is still at an unacceptably high value of approximately 57 ns. Thus, electronic synchronization does not provide a reliable method for the practical application of film deposition using the dual-laser ablation technique in which the KrF laser has to be positioned precisely on the 100 ns rising edge of the CO₂ laser.

A series of measurements was made to locate the source of the largest contribution to the jitter in the whole system. Measurements of temporal delay were made between the optical flash from the CO₂ discharge and the CO₂ laser pulse, the electronic trigger pulse for the excimer laser and the output excimer laser pulse, the trigger pulse for the CO₂ laser and the CO₂ laser pulse, and the CO₂ and excimer laser pulses. All of these experiments were performed at the highest CO₂ laser discharge voltage (without arcing) of 37 kV, as this yielded the lowest fluctuations (see Fig. 1). After 50 measurements were taken, the jitter was determined by taking the standard deviation of the delay times. Figure 2 demonstrates that the greatest variation is within the CO₂ laser. The fluctuation between the trigger pulse and the excimer laser pulse is approximately 15 ns while the corresponding value between the CO₂ and excimer laser pulses is ~325 ns, which is 20 times greater than the internal jitter of the excimer laser. It can be concluded that the largest source of jitter is between the trigger from the pulse generator to the CO₂ laser pulse. This is not surprising since the CO₂ laser is a gain-switched laser with delays involved in both the triggering of the high voltage discharge as well as the buildup of gain in the laser cavity. Remediation of the fluctuation problem therefore requires a new method for pulse synchronization.

There are two factors of importance in precise control of the synchronization of the two laser pulses in the dual-laser ablation system. First, in order to place the excimer laser on

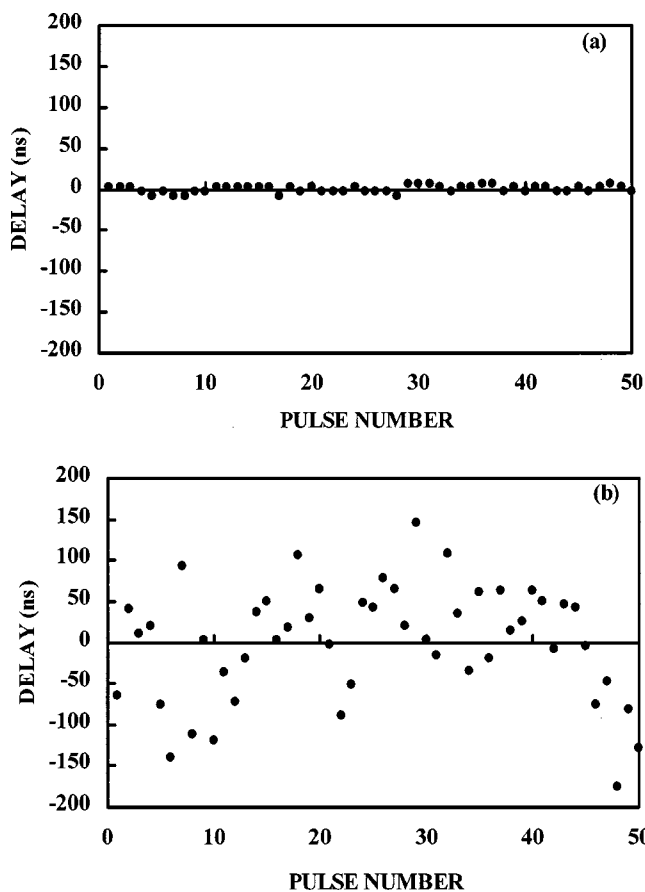


FIG. 2. Individual delays for 50 shots between (a) the pulse generator and the excimer laser pulse and (b) the CO₂ laser pulse and excimer laser pulse. The CO₂ laser discharge voltage was 37 kV.

the rising edge of the CO₂ laser pulse (a condition necessary for control of film quality¹), the excimer laser (which has low jitter) could not be triggered by the CO₂ laser output since there was a 1250 ns delay between the trigger to the excimer laser and the emission of the laser pulse. Second, any technique relying on electronic synchronization of the trigger to the two lasers would be ineffective due to the large jitter inherent in the CO₂ laser, as illustrated in Fig. 2. In tandem, these two factors required a trigger source from the CO₂ laser that minimized the fluctuations with respect to the output CO₂ laser pulse while serving as a trigger that could be electronically delayed to trigger the excimer laser. This trigger source would also need to precede the emission of the CO₂ laser pulse by greater than 1250 ns to allow suitable temporal positioning of the excimer laser pulse with respect to the CO₂ laser.

We used the optical flash from the CO₂ laser high voltage discharge to provide this trigger source. This eliminated the fluctuations associated between the trigger to the thyatron and the generation of the high voltage excitation of the CO₂ laser gas mixture. In addition to using the optical flash, a variety of CO₂ laser parameters had to be optimized, both to reduce the temporal fluctuations as well as to maintain the minimum delay between the optical flash and the emission of the CO₂ laser pulse, to obtain the required KrF-CO₂ laser interpulse synchronization. The optimization of these laser

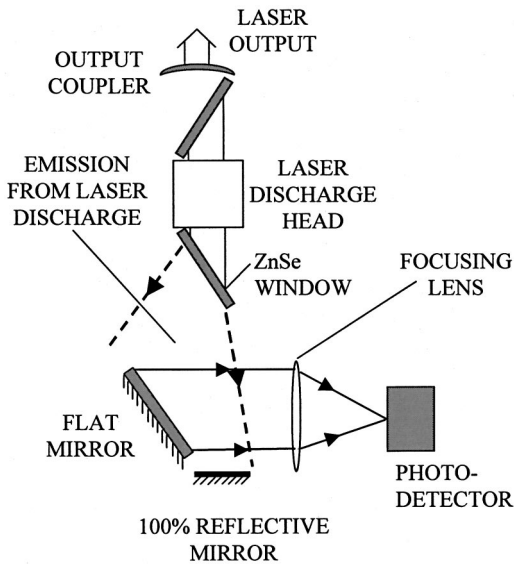


FIG. 3. Schematic diagram of the arrangement for optimizing the internal delay of the CO₂ laser.

parameters is discussed in Sec. II B, followed by details of the optical triggering scheme in Sec. II C.

B. Effect of CO₂ laser parameters

In initial experiments to study the effect of the various CO₂ laser parameters on the jitter of the system we used measurements of the delays between the discharge flash (from the region between the electrodes) and the outgoing CO₂ laser pulses. These delays (internal delays) were measured by focusing a portion of the light of the discharge flash onto a photodetector. Figure 3 shows the initial layout used to obtain these delay measurements. In order to access light from the internal flash, the laser cavity was opened up and extended. A flat mirror was placed just outside the path of the laser cavity to intercept and reflect a portion of the light beam from the flash caused by the high voltage discharge. Subsequently, an 8 in. focal length lens was used to focus this light onto a photodetector. A pyroelectric detector was used to monitor the CO₂ laser pulse and the delay between the flash and the laser pulse was measured on a 400 MHz Tektronix TDS 380 digital oscilloscope.

1. Effect of laser mirrors

The laser output and internal photon buildup in the cavity can be affected by the reflectivities and radii of curvature of the output coupler (OC) and the rear mirror of the laser cavity. Since both these parameters affect the single-pass gain, they impact both the laser pulse energy and the timing and jitter of the emitted pulse. Our experiments with a variety of output couplers and rear mirrors resulted in low pulse energy and high jitter, or high pulse energy and low jitter, or even high pulse energy and high internal jitter. A balance must be found so that the laser pulse has both an acceptable pulse energy and low jitter.

The buildup in the cavity is related to the circulating intensity. The net intensity in the cavity for N roundtrips (taking time T for one roundtrip) is

$$I(T) = I_0 \times R_1 R_2 (R_3 \dots) \exp[2\alpha_m p_m - 2\alpha_0 p] \\ = I_0 \exp[\delta_m - \delta_c], \quad (1)$$

where the R_i 's are the mirror reflectivities and I_0 is the initial intensity from the noise level. Larger than 2 mirror reflectivities are encountered when additional turning mirrors are introduced into the cavity. α_m , α_0 , p_m , and p are the laser/molecular gain coefficient, scattering loss, roundtrip length through the medium, and roundtrip path length of the cavity, respectively. The δ_m and δ_c variables are the gain coefficients due to the laser atoms and the total roundtrip power loss due to the cavity, respectively.¹⁶ Equation (1) demonstrates the effect of mirror reflectivity on the buildup of photons in the cavity. If the rear mirror (R_2) has a reflectivity close to unity, Eq. (1) is dependent only on the reflectivity of the output coupler. As the output coupler's reflectance is increased, the $I(T)$ will increase, leading to a delay in the emission of the CO₂ laser pulse and a decrease in the peak intensity emitted. A longer duration pulse with larger internal jitter will result.

The radii of curvature of the laser mirrors will affect the beam waist (ω_0) and the laser spot size (ω_i). The following equations, Eqs. (2) and (3), are related to the effect of the radii on these two parameters.¹⁷ In these equations, L is the length of the laser cavity.

$$\omega_0^2 = \frac{\lambda L}{\pi} \\ \times \sqrt{\frac{[1 - (1 - L/R_1)(1 - L/R_2)](1 - L/R_1)(1 - L/R_2)}{2 - L/R_1 - L/R_2 - 2(1 - L/R_1)(1 - L/R_2)}}, \quad (2)$$

$$\omega_i^2 = \frac{\lambda L}{\pi(1 - L/R_i)} \sqrt{\frac{(1 - L/R_1)(1 - L/R_2)}{1 - (1 - L/R_1)(1 - L/R_2)}}. \quad (3)$$

In Eq. (3), i is 1 and 2 for the output coupler and the rear mirror, respectively. For a flat rear mirror R_2 , with an infinite radius of curvature, Eqs. (2) and (3) simplify to

$$\omega_0^2 = \frac{\lambda L}{\pi} \sqrt{1 - L/R_1}, \quad (4)$$

$$\omega_2^2 = \frac{\lambda L}{\pi} \sqrt{\frac{1 - L/R_1}{L/R_1}} = \frac{\lambda L}{\pi} \sqrt{\frac{R_1}{L} - 1}, \quad (5)$$

With a small spot size, ω_2 , on the rear mirror, a smaller portion of the gain medium is being utilized. This can lead to gain saturation and inconsistent pulse energy with associated temporal jitter. With a larger spot size, more of the gain medium is used, allowing reproducible buildup of the laser pulse.

Although we did not continuously vary the reflectivities and radii of curvature of the laser mirrors, experimentation with a variety of available possibilities confirmed the considerations previously discussed. All of these measurements were performed under conditions of maximum pumping at a 37 kV discharge voltage. For example, using an output coupler with a reflectivity of 65% and a radius of curvature of 44 m, the pulse energy of the resulting TEM₀₀ laser output was

400 mJ. However, the jitter was too large at ± 81 ns. Upon changing the output coupler to one with a reflectivity of 86% and a 10 m radius of curvature, the laser pulse energy dropped to 250 mJ, but the internal delay still fluctuated at an unacceptably high value of ± 53 ns. Increasing the reflectivity of the output coupler to 96% while maintaining the 10 m radius of curvature produced the same laser pulse energy of 250 mJ with a reduction of the jitter to ± 8 ns. However, the internal delay between the flash and the pulse was 980 ns, too short for the synchronization of our dual-laser ablation system in view of the 1250 ns internal delay of the KrF laser. The general trend of larger jitter associated with longer delays in the emission of the laser pulse was consistently observed.

In our case, another important parameter was the placement of the discharge head within the laser cavity. In all the previous measurements reported above, the discharge head was located close to the rear mirror. Therefore, the spot size of the intracavity oscillating laser beam was close to its minimum value, leading to possible gain saturation and larger fluctuations in the temporal delays of the laser output. This can easily be remedied by moving the discharge head or the laser mirrors and results in a relative shift of the active medium closer to the curved mirror. Because of space considerations, we simply switched the radii of curvature of our laser mirrors while keeping their positions and that of the discharge head invariant.

The final combination used was a curved rear mirror with a 20 m radius of curvature and a flat 50% reflectivity output coupler. This resulted in a stable CO₂ laser pulse energy of 250 mJ and low jitter of ± 14 ns. Interestingly, the delay was large enough to place the excimer pulse 110 ns before the CO₂ laser pulse for synchronous triggering of the excimer laser with the optical flash.

2. Effect of cavity layout

As stated earlier, the internal delay of the CO₂ laser was required to be at least 1250 ns for dual-laser synchronization. One obvious approach is to change the length of the cavity—the longer the cavity, the longer the delay. This is indicated in the buildup time of the steady-state intensity in Eq. (6),

$$T_b \approx \frac{\tau_c}{r-1} \ln\left(\frac{I_{ss}}{I_0}\right), \quad (6)$$

where I_{ss} is the steady-state oscillation intensity, τ_c is the cavity decay lifetime, and r is the normalized inversion ratio given by¹⁶

$$r = \frac{\delta_{mo}}{\delta_c} = \frac{\alpha_{mo}}{\alpha_c} = \frac{2\alpha_{mo}p_m}{2\alpha_{op} + \ln(1/B_{tot})}, \quad (7)$$

where $R_{tot} = R_1 R_2 R_3 \dots$. The relationship between the roundtrip path p and the cavity buildup time is evident from Eqs. (6) and (7). The length of the cavity was therefore adjusted to meet the needs set by the excimer laser triggering delay.

The cavity was lengthened to incorporate extra delay for the appropriate dual-laser synchronization. In an attempt to reduce the overall length occupied by the laser cavity, changes from a linear configuration into an ‘‘L-shaped cav-

ity’’ (adding one extra mirror) and a ‘‘U-shaped cavity’’ (adding two extra mirrors) were explored. Although the delay in laser pulse emission was increased, the addition of extra mirrors served as additional loss mechanisms for the multipass CO₂ laser and contributed to lower pulse energies as well as unacceptable levels of jitter. Therefore, we restored the elongated straight line configuration with a cavity length of 4.4 m, to get the delay needed, with a constant pulse to pulse energy and an acceptable jitter.

3. Effect of gas mixture

Another parameter that affected the laser pulse energy and the jitter was the gas mixture in the cavity. To reduce the effect of the gases being depleted, the cavity was set up as a flowing system. This allowed the gases to be continuously replenished, thus reducing the buildup of the depleted gases and CO. By controlling the amount of each component (N₂, CO₂, and He) of the gas mixture individually, it was possible to optimize the laser output and jitter. Increasing the percentage of N₂ in the mixture can increase the pulse energy by pressure broadening.¹⁶ The increase in the N₂–CO₂ collisions will result in a longer pulse width and increased jitter. Slightly increasing the buffer gas, He, stabilized and increased the pulse energy. This was most likely due to the increase in the amount of He assisting in depletion of the lower CO₂ excited level to the ground state. The final gas mixture for stable laser operation without arcing at 37 kV was 20% N₂, 30% CO₂, and 50% He, which resulted in a pulse energy of 250 mJ in the TEM₀₀ mode. Higher pulse energies were obtainable by increasing the partial pressure of N₂, but resulted in increased jitter. The CO₂ laser output used in our experiments was at the strongest 10P(20)CO₂ laser transition, which corresponds to a wavelength of 10.59 μm .

4. Effect of discharge voltage

After optimizing the laser cavity configuration, length, and the gas mixture, we determined the effect of the high voltage of the CO₂ laser discharge on the jitter. The effect of the high voltage is to change the pumping rate by coupling more energy into the N₂, which isoenergetically transfers the energy to the upper CO₂ laser level.¹⁶ The time delay to inversion is given by

$$t_d = \frac{\tau \ln[2W_p \tau / (W_p \tau - 1)]}{W_p \tau + 1}, \quad (8)$$

where τ is the lifetime in the excited state and W_p is the pumping transition probability.¹⁶ The pumping transition probability can be related to the pumping rate by

$$\frac{dN_4}{dt} = W_p(N_1 - N_2) - N_4/\tau_4, \quad (9)$$

where the N_i 's are the population densities at specific energy levels of the four-level CO₂ laser system and τ_4 is the lifetime of the upper most energy level.¹⁶

With lower voltage and consequently lower W_p , less photons are emitted and a longer buildup time results. As the

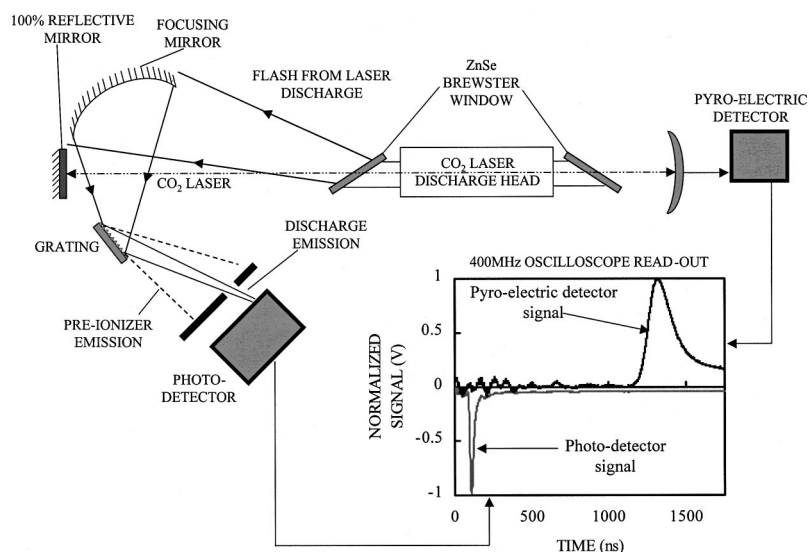


FIG. 4. Schematic diagram of the final optical triggering arrangement.

high voltage decreases, the internal delay increases, as seen in Fig. 1. There was also an increase in the delay between the optical flash from the high voltage discharge and the emission of the CO_2 laser pulse with a decrease in high voltage. This could be an advantage when setting the temporal delay between the CO_2 and excimer laser pulses, but the increase in jitter was too great. Therefore, the maximum high voltage without arcing should be used to minimize the jitter and the delay should be adjusted by modifying the length of the cavity linearly.

III. DESCRIPTION OF THE OPTICAL TRIGGERING SYSTEM

Since the variation in the internal delay of the CO_2 laser could be small (<15 ns) after the optimization discussed in Sec. II, and the delay between the flash of the CO_2 laser discharge and the outgoing laser pulse was sufficient (>1250 ns), an optical triggering scheme can be implemented for synchronizing the two lasers for the dual-laser ablation process.

There are two parts to the flash of the CO_2 discharge, a white continuum from the preionizers and a purple glow from the high voltage gas discharge. The purple glow was used as the optical trigger. A schematic representation of the optical triggering scheme is shown in Fig. 4. The flat mirror in Fig. 3 (used for optimization of the CO_2 laser parameters in Sec. II) was replaced by a 10 cm circular aperture concave mirror of 25 cm focal length for increased efficiency of light gathering. This light was then incident on a grating and focused onto a photodetector (as shown in Fig. 4). The photodetector signal was sent to a pulse-delay generator, which triggered the excimer laser.

The purpose of the grating was to isolate the purple glow of the high voltage discharge from the preionizer illumination. However, the grating results in an overall reduction in light intensity that reduces the photodetector signal, thereby leading to an associated increase in the observed interpulse jitter. This is easily overcome by using a suitable optical band-pass filter or a more sensitive detector such as a photomultiplier tube.

We have also implemented an alternative gratingless configuration in which the grating is replaced by a flat mirror. Since the preionizer emission is much weaker than the purple glow of the high voltage discharge, a less sensitive photodetector can be used to screen it out. This option is the least expensive approach to implementing the optical triggering technique. To ensure that the light from the preionizers was not being detected by the photodetector, the high voltage was reduced to a level where only light from the firing of the preionizers was visible. Under these conditions, when no purple glow was visible and no laser output was detected, there was no detectable optical signal from the photodetector. As the high voltage was raised and the main electrodes discharged, a photodetector signal was obtained. Once it was determined that only the purple flash from the main discharge was being detected, a series of experiments was performed to compare the electronic and optical triggering techniques.

IV. COMPARATIVE PERFORMANCE OF ELECTRONIC AND OPTICAL TRIGGERING

The delay between the excimer and CO_2 laser pulses was obtained at different CO_2 high voltages. As before, the standard deviation of the 50 delay measurements provided jitter for each high voltage. This was done for both the optical trigger and the previous method of electronic triggering. The results can be seen in Fig. 5. The lowest jitter of ± 13 ns was obtained for optical triggering at a high voltage of 37 kV and also resulted in the excimer laser pulse being detected before the CO_2 pulse. This allows the use of the pulse generator to temporally adjust the excimer pulse with respect to the CO_2 pulse. As the high voltage decreased, the delays increased along with the jitter for both triggering methods. However, the relative increase in jitter for the optical trigger was much smaller than that for the electronic trigger. As expected, there was also a decrease in the pulse energy with decreasing high voltage. Thus, for optimum performance, the CO_2 laser must be run at its maximum voltage to ensure both the largest pulse energy and the lowest jitter.

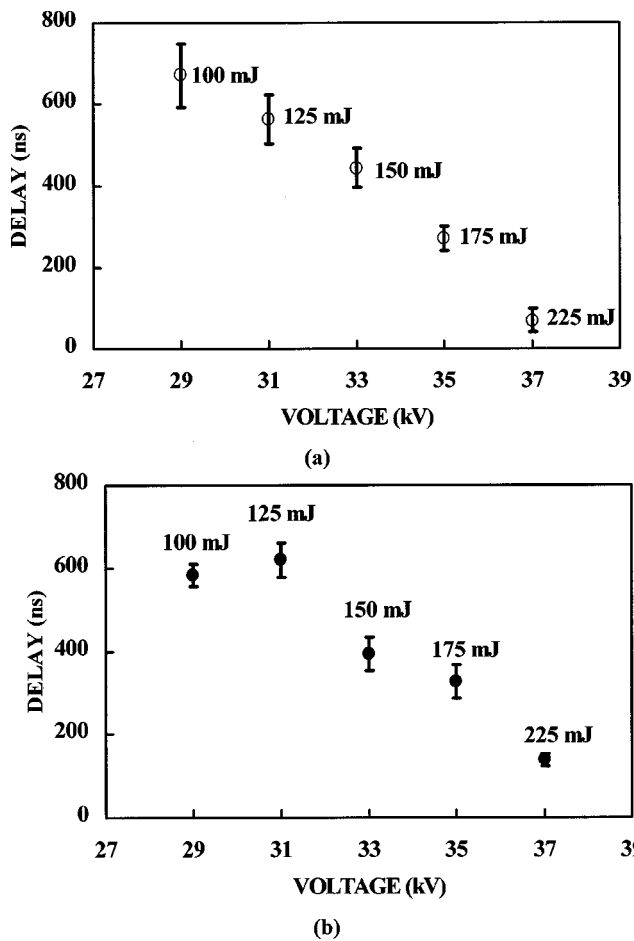


FIG. 5. Delay of the CO₂ laser pulse and the excimer laser pulse at different CO₂ laser high voltages under (a) electronic and (b) optical triggering. The output CO₂ laser pulse energy is indicated for each CO₂ discharge voltage.

The operational efficacy of the optical triggering technique for greater control over the temporal location of the synchronization of the two lasers can be assessed by imaging the early plasma from the dual-laser ablated plume. We used a ZnO target as a representative example. The KrF laser pulse was positioned at a delay of 35 ns with respect to the onset of the CO₂ laser pulse as shown in Fig. 6. This timing

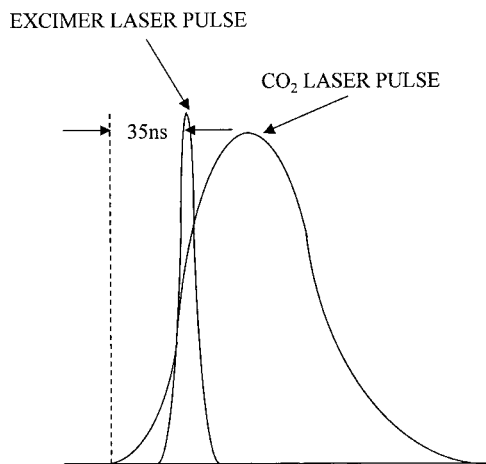


FIG. 6. Illustration of the timing between the excimer and CO₂ lasers for the ablation of ZnO.

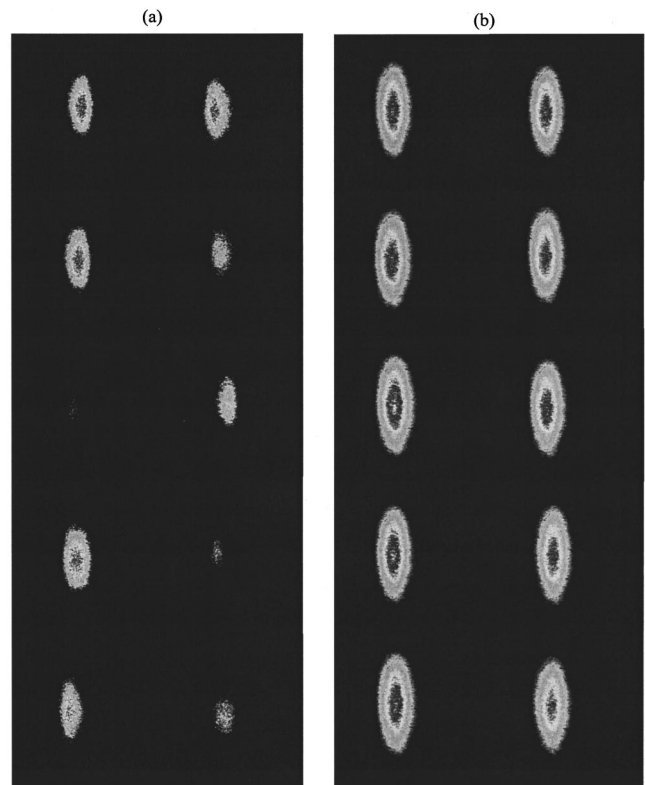


FIG. 7. Individual CCD images of the ZnO plume for 10 successive shots using (a) electronic and (b) optical triggering for the dual-laser synchronization.

coincided with separately published studies indicating surface melting of ZnO due to the CO₂ laser¹⁸ and particulate-free film deposition using dual-laser ablation at this interpulse delay.¹⁹ Charge coupled device (CCD) images of the first 150 ns of the ZnO plume immediately following dual-laser ablation under these conditions was obtained for 10 successive shots (for both optical and electronic triggering) to assess the reproducibility of the interpulse synchronization between the KrF and CO₂ lasers. These CCD images, shown in Fig. 7, demonstrate the reproducibility of the optical trigger compared to the electronic triggering. The optical trigger results show a consistently reproducible image of the plume that does not vary appreciably from shot to shot.

A plot of the full width at half maximum (FWHM) of the emission along the central propagation axis of the CCD image shows the reproducibility of the optical triggering as compared to electronic synchronization (Fig. 8). The FWHMs for the 10 shots vary by $\pm 50\%$ for the electronic trigger, a factor of 10 greater than the approximately $\pm 5\%$ variations for the optical trigger. This consistency using the optical triggering technique for the synchronization of the CO₂ and KrF laser pulses in dual-laser ablation implies less shot-to-shot variations in the laser-target interaction and consequently the possibility of better quality films. The use of this technique for reproducible particulate-free, thin film growth is currently under investigation and will be reported elsewhere.^{18,19}

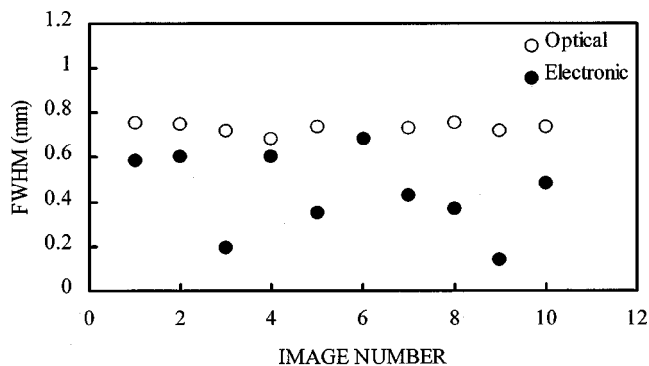


FIG. 8. FWHM of each of the 10 CCD images of the ZnO plume in Fig. 7 using electronic and optical triggering.

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