LA-SUB--94-59

LASER PHYSICS RESEARCH
FOR KrF LASERS - Fluorescence
Suppression Measurements

9-XG9-9123Z1

Final Report

Submitted to
LOS ALAMOS NATIONAL LABORATORY

Submitted by
STI OPTRONICS, INC.

October 1991

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Section 1

EXECUTIVE SUMMARY

This final report describes the work performed for Los Alamos National Laboratory (LANL) to measure the wavelength-dependent KrF fluorescence suppression as a function of intracavity flux. To our knowledge, this is the first time the wavelength-dependent suppression has been measured for KrF.

The motivation for this work is the need to improve our understanding on the amount of energy extractable from the KrF upper state manifold. This information has direct impact on the ultimate efficiency of KrF and, therefore, on its utility as an ICF driver.

The data obtained during this experiment indicates that nonuniform suppression occurs over the KrF gain spectrum. Spectral features on the gain spectrum can be approximately associated with the lower vibrational levels of the B-state. Hence, the nonuniform suppression implies the vibrational levels are either not being extracted uniformly or they are not being repopulated uniformly. Earlier theoretical work by others predicts uniform extraction should be occurring for the first 4 ν -levels. Therefore, we believe the latter situation is probably occurring.

Analysis performed by Dr. Chris Patterson and Dr. David Hanson of LANL supports the explanation that nonsequential relaxation from the D-state to the B-state may be occurring. Using this assumption they are able to predict the nonuniform suppression observed during the experiment.

The next section of this report describes the experiment. Section 3 presents the experimental results. Conclusions are given in Sec. 4. In the appendices are details of the experiment calibration procedure, data reduction process, and calibration of the film used during the experiment.

Section 2

DESCRIPTION OF FLUORESCENCE SUPPRESSION EXPERIMENT

2.1 BACKGROUND

Efficient energy extraction from the KrF^* molecule requires efficient coupling of the lower vibrational levels in the KrF^* B-state into the laser photon field. Various modeling efforts^{(2-1),(2-2)} have predicted comparable coupling efficiencies for the $\nu = 0, 1, 2$, and 3 upper state levels. One way to examine this experimentally is to observe the amount of suppression of the KrF^* sidelight fluorescence emission under nonlasing and lasing conditions. This gives a direct indication of how much population is left in the upper state manifold. Previous sidelight suppression measurements^{(2-3),(2-4)} in KrF indicated that some population is left; however, these measurements were integrated over wavelength and did not give any information on how this population is distributed among the vibrational levels. The work performed during this program represents the first wavelength-dependent measurements of the KrF^* sidelight fluorescence suppression in an electron-beam (e-beam) pumped KrF laser (Tahoma).

Detecting the sidelight fluorescence emission requires careful control of the light collection procedure in order to avoid including in the measurement contributions from excited laser gas that is outside the optical volume of the laser resonator. In addition, since the intent of this work was to use a spectrometer to resolve the KrF spectrum, it was important to design a reasonably efficient means for collecting the incoherent fluorescence emission. This section describes the technique used to overcome these problems.

2.2 DESCRIPTION OF MEASUREMENT APPARATUS

Figure 2-1 is a schematic of the rather novel collection system used to direct the light into the spectrometer. It features a light collection tube inserted directly into the active volume. The end of the tube is sealed with a fused quartz window with the inside of the tube at ambient air pressure. An

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Figure 2-1. Schematic of Sidelight Fluorescence Measurement System. The e-beam pumps the active volume perpendicular to the plane of the paper.

optical block consisting of razor blades sandwiched together with their edges facing the quartz window is positioned ≈ 1 cm from the window. This optical block has a measured reflectivity of $\lesssim 0.6\%$ in 2π steradians and serves to accurately define the volume of excited gas imaged by the collection system while preventing unwanted light from reflecting off it and into the collection tube.

The e-beam direction is into the plane of the figure and has dimensions of approximately 4 cm x 70 cm at a magnetic guide field strength of 2 kgauss. The e-beam peak current density is 30 A/cm^2 at the foil (0.13 mm aluminized Kapton) at an erected Marx voltage of 375 kV. During these measurements the e-beam pulse length is 440 ns. Not shown in the figure is a lead e-beam stop that reflects some of the electrons back into the active volume and increases the volume averaged deposition to 335 kW/cm^3 . This is the same e-beam pumping arrangement used during the previous KrF laser measurements performed earlier for LANL.⁽²⁻⁵⁾

Two gas mixtures were investigated: Ar-diluent ($F_2/\text{Kr}/\text{Ar} = 2.8/142/859$ Torr) and Kr-diluent ($F_2/\text{Kr} = 3.5/661$ Torr). However, due to budget limitations, suppression data were only obtained for the Ar-diluent case.

An intracavity iris restricts the diameter of the optical volume to just fit between the collection tube and razor blade block. The rear mirror ($>98\%$ R, 10 m radius of curvature) is separated from the output coupler (flat) by 156 cm. By varying the output coupler reflectivity (2.5% R to 90% R), the intracavity flux can be varied. Since the collection tube is positioned only 4 cm from the end of the e-beam excitation region nearest the output coupler, the intracavity flux can be estimated by multiplying the output energy by the factor $(1 + R)/(1 - R)$, where R is the output coupler reflectivity.

Nonlasing data is obtained by inserting an opaque block between the iris and output coupler. During nonlasing shots, the ASE flux from the rear reflector is 55 kW/cm^2 . Its effect, although small, was accounted for during the data reduction process (see Appendix B).

Because the best available grating (1200 grooves/mm) for the 1-m spectrometer is blazed for 1 μm , maximum light throughput in the UV is found to occur at 4th order on the spectrometer with a resulting resolution of 2 $\text{\AA}$. Hence, the dispersion is high, which means that spectral features are well separated. This also means that blooming of images on the film from the brighter portions of the spectrum does not interfere with detection of the fainter spectra at shorter wavelengths (see comments of Reference 3-7 and Appendix C). However, the high dispersion does spread out the spectrum on the film thereby reducing its intensity.

The output from the spectrometer is recorded on Kodak T-Max film (ASA 400). The film development is pushed 3-stops in order to record the faint images from the higher vibrational levels. Even so, multiple exposures of the film (e.g. 5 shots) are necessary to ensure the $\nu = 3$ emission is well above the noise background. (More details regarding problems with using film are discussed in Appendix C.)

2.3 DESCRIPTION OF DIAGNOSTICS

Besides careful control of the collection process, accurate calibration of the detection system is needed, especially of the film. Details of the calibration procedure are described in Appendix A. Briefly, calibration of the film is performed by using the spontaneous emission from the end of the Tahoma laser to reflect off a scattering plate and uniformly expose a sheet of film covered with calibrated UV neutral density (ND) filters of various values. This simulated the exact short pulse length (440 ns) of the actual experiment and the UV wavelength. This procedure is also used to calibrate multiple shot exposed film and to verify that the pushing process during the film development does not affect the film performance.

A tunable pulsed dye laser doubled to the UV is also used as a calibration source. By measuring the amount of energy transmitted through the collection optics and spectrometer as a function of UV wavelength, the spectral transmission characteristics of all the optics is obtained. Based upon a D_2 lamp and the spectral transmission characteristics of the spectrometer, it

appears the spectral response of the film is fairly flat over the wavelength range of interest (241 nm - 251 nm).

Finally, a CCD camera viewing the film negative lying on a light table acts as a 2-D densitometer for analyzing the spectra. The linearity of this process is verified using the calibrated ND filters. The spectra are typically several millimeters wide, which permits averaging over several raster lines of the camera, thereby helping to reduce the noise in the data. However, high film densities corresponding to high light intensities can start approaching the black limit of the camera resulting in more noise in the data. (Decreasing the camera f-stop can help compensate for this at a cost of increased sensitivity to stray light.) Hence, the small fluctuations in the data presented in the next section are due to noise in the densitometry process and are probably not real spectral features.

Although our novel collection scheme eliminates the need to correct for unwanted light detection, it is known the fluorescence suppression does not occur instantaneously during lasing. There is a finite build-up time before the laser reaches threshold and a finite time required before the fluorescence decreases to its equilibrium level.^{(2-4),(2-6)} The film cannot resolve this temporal behavior and simply records it as additional light.

The procedure for determining the true suppression value from the data is described in detail in Appendix B. Summarizing it here, the temporal waveforms of the unsuppressed sidelight fluorescence (taken at a different port from the collection tube) and the laser output are measured by photodiodes and modeled as simple trapezoids. Lasing threshold occurs during the rising slope of the trapezoid and, to simplify the analysis, it is assumed equilibration is reached at the peak of the trapezoid. The suppressed fluorescence waveform will consist of a spike at the beginning, due to the build-up and equilibration time, and a plateau level lower than the unsuppressed waveform (see Refs. 2-4 and 2-6 for an examples). This plateau level, of course, represents the suppression which is lower at the higher ν levels. This implies a higher value for the effective saturation flux $I_{\text{sat}}^{\text{eff}}(\nu)$ for each higher ν level. Using this simple model for the suppressed waveform behavior, the standard saturation equation, and the raw suppression data, $I_{\text{sat}}^{\text{eff}}(\nu)$ is calculated for each of the ν

levels. We assume for this last step that each of the peaks in the fluorescence spectrum are due primarily to the corresponding ν level (i.e. the peak at 248.5 nm is $\nu = 0$, 245.3 nm is $\nu = 1$, 242.9 nm is $\nu = 2$, and 240.6 nm is $\nu = 3$). This is only approximately true since the peaks generally have contributions from several ν levels, especially the primary one at 248.5 nm.^{(2-1),(2-2)} With $I_{\text{sat}}^{\text{eff}}(\nu)$ known, the appropriate plateau level of the suppressed waveform can be determined, yielding the true suppression value.

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Section 3

EXPERIMENT RESULTS

3.1 UNSUPPRESSED (NONLASING) FLUORESCENCE SPECTRA

Figure 3-1 is a typical KrF* fluorescence spectrum for the Ar-diluent gas mixture. This spectrum is for a single laser shot and is plotted on a linear scale. Later, in Section 3.2 when more detailed comparisons are made between nonlasing and lasing data, the spectra are plotted on $\log_2$ scales to show details of the higher ν levels. The noise in the spectrum, as explained in Section 2.3, is due primarily to limitations in the video densitometer system. This is especially evident in the 248.5 nm peak where the highest film density is encountered. The dotted sections of the spectrum are where wavelength fiducial markers are located (see Figures 3-2 and 3-3 for examples).

One can improve on the noise problem by averaging over several spectra. Figures 3-2 and 3-3 are unsuppressed fluorescence spectra averaged over 3 shots for the Ar-diluent and Kr-diluent gas mixture cases, respectively. The spectra are very similar to each other; however, they are not normalized to one another therefore their absolute magnitudes cannot be compared.

One feature that is clear from these spectra is that the second spectral peak centered at 245.3 nm is approximately 10 times lower than the primary peak at 248.5 nm. The third peak centered at 242.9 nm is roughly 39 times lower than the primary peak. To compare this with other measurements, the literature was searched to find examples of fluorescence spectra for e-beam excited KrF lasers. There are many examples; however, most of these are for uncalibrated film. The few that calibrated their film, including this work, are listed in Table 3-1. Given are the ratios of the fluorescence spectrum peaks of 248.5 nm to 245.3 nm, and 248.5 nm to 242.9 nm. These ratios give an indication of the population distributions in the upper manifold under nonlasing conditions.

Figure 3-1. Typical Unsuppressed (Nonlasing) KrF^* Sidelight Fluorescence Spectrum for the Ar-Diluent Gas Mixture. Noise spikes on the spectrum are probably due to noise from the densitometer system.

Figure 3-2. Unsuppressed Sidelight Fluorescence Spectrum Averaged Over 3 Shots for the Ar-Diluent Gas Mixture.

Figure 3-3. Unsuppressed Sidelight Fluorescence Spectrum Averaged Over 3 Shots for the Kr-Diluent Gas Mixture.

Table 3-1

**COMPARISON OF UNSATURATED FLUORESCENCE SPECTRAL DATA
WITH OTHER WORKS**

	Ratio of Peaks in Spectrum		Width of 248.5 nm peak [FWHM] (nm)
	$\frac{248.5 \text{ nm}}{245.3 \text{ nm}}$	$\frac{248.5 \text{ nm}}{242.9 \text{ nm}}$	
This Work	10	39	2.4
Tisone, et al. ⁽¹⁾	9	-	2.5
Brau & Ewing ⁽²⁾	16	45	2.0
Murray & Powell ⁽³⁾	4	8	2.6
Gomez (LANL ASE data)	3	6	3.0
Tellinghuisen, et al. (model) ⁽⁴⁾	7	24	?
Patterson & Hanson (model)	4	7	2.4

(1) G.C. Tisone, et al., Opt. Comm. 15, 188 (1975).

(2) C.A. Brau and J.J. Ewing, J. Chem. Phys., 63, 4640 (1975).

(3) J.R. Murray and H.T. Powell, Appl. Phys. Lett., 29, 252 (1976).

(4) J. Tellinghuisen, et al., J. Chem. Phys. 65, 4473 (1976).

The ratios found in this work agree very well with Tisone, *et al.*,⁽³⁻¹⁾ and approximately so with Brau and Ewing;⁽³⁻²⁾ whereas there is a large disagreement with the data of Murray and Powell,⁽³⁻³⁾ and Gomez⁽³⁻⁴⁾ [the Gomez data is of the KrF* amplified spontaneous emission (ASE) spectrum]. Although the laser parameters are not the same between researchers, it is not clear this explains the differences. For example, the Tisone e-beam voltage and current were much higher than this work, and yet there appears to be good agreement. Problems with accurately calibrating the film is certainly a possibility (see Appendix C).

Also shown in Table 3-1 are the model predictions of Tellinghuisen, *et al.*,⁽³⁻⁵⁾ and Patterson and Hanson.⁽³⁻⁶⁾ The Tellinghuisen, *et al.* prediction for the 248.5 nm/245.3 nm ratio is in-between our result and the Murray and Powell result, but the prediction for the 248.5 nm/242.9 nm ratio is, relatively speaking, closer to our result. The predictions of Patterson and Hanson are in good agreement with the Murray and Powell, and the Gomez data.

Finally, in Table 3-1 are listed the measured and predicted spectral widths [full width at half maximum (FWHM)] for the 248.5 nm peak. Here there appears to be closer agreement among the experiments⁽³⁻⁷⁾ and the model predictions.

3.2 SPECTRAL SUPPRESSION DATA

Figure 3-4 shows an example of the nonlasing and lasing spectra for a 90% R output reflector corresponding to an intracavity flux of 4.9 MW/cm^2 . This flux is ≈ 4 times the saturation flux ($I_{\text{sat}} = 1.2 \text{ MW/cm}^2$) determined for this device from previous measurements⁽³⁻⁸⁾ and in agreement with others.⁽³⁻⁹⁾ Dotted sections of the spectra at approximately 242.0 and 246.8 nm are where wavelength fiducial marks were located (the absolute wavelength calibration was determined using a mercury lamp source). The dotted spike at 248.4 nm in the lasing spectra is a small amount of lasing light that leaks into the spectrometer. (Note, the lasing wavelength is slightly lower than the fluorescence peak at 248.5 nm.) Fortunately, this spike only slightly obscures the suppression data and it does provide a convenient verification of the

Figure 3-4. Unsuppressed and Suppressed Sidelight Fluorescence Spectra for the Ar-Diluent Mixture at an Intracavity Flux of 4.9 MW/cm^2 . The percentage amount of suppression is plotted in the lower curve.

absolute wavelength calibration. Our lasing wavelength agrees with Tisone, *et al.*,^{(3-1),(3-5)}

The ratio of the lasing to nonlasing data, which represents the suppression, is also plotted in Figure 3-4. At 248.5 nm the suppression is >75%, but the suppression then steadily decreases with shorter wavelength reaching a value of $\approx 40\%$ at 240.6 nm. This is surprising considering the good overlap of the theoretically predicted^{(3-6),(3-9)} Einstein A coefficients for the four lowest vibrational levels at 248.4 nm.

This same trend is observed at lower intracavity flux levels as shown in Figure 3-5. The lower group of data points is for an internal cavity flux of 1.7 MW/cm^2 . This set of data still displays this tendency for the suppression to decrease with decreasing wavelength; however, the slope is shallower. Although it may appear the two suppression curves cross at approximately 242 nm, the suppression data for the 1.7 MW/cm^2 case actually lie below the 4.9 MW/cm^2 data. This is consistent with less overall suppression occurring at lower cavity fluxes.

3.3 CAVITY FLUX DEPENDENT RESULTS

Figure 3-6 plots the peaks of the fluorescence spectra, corresponding roughly to the various ν levels, as a function of the intracavity flux. The dashed curve is from the standard saturation equation $[I_{\text{out}} \propto (1 + I/I_{\text{sat}})^{-1}]$ assuming $I_{\text{sat}} = 1.2 \text{ MW/cm}^2$. This value for I_{sat} was obtained from earlier extraction measurements on this same laser device. The agreement with the data for the 248.5 nm peak is quite good demonstrating the consistency of the fluorescence data with previous work.

The fluorescence peaks for the shorter wavelengths (i.e. higher ν levels) appear to plateau at higher values of saturation flux. It is possible to infer an "effective" I_{sat} value for each of these peaks. To do this requires a more complicated analysis than simply using the standard saturation equation. This is because of the aforementioned temporal dependence of the saturation behavior of the higher ν levels. This analysis is described in Appendix B from which average values for the effective I_{sat} for each of the fluorescence

Figure 3-5. Comparison of Suppression Amounts for Intracavity Fluxes of 4.9 and 1.7 MW/cm^2 for the Ar-Diluent Mixture.

Figure 3-6. Relative Magnitude of the Peaks in the Fluorescence Spectrum for the Ar-Diluent Mixture as a Function of Intracavity Flux. The dashed curve is a plot of the standard saturation equation assuming an I_{sat} value of 1.2 MW/cm^2 obtained from previous measurements.

peaks can be calculated. These are listed in Table B-1 and are repeated here. The calculated average I_{sat} value associated with the fluorescence peak at 248.5 nm is $1.3 \pm 0.1 \text{ MW/cm}^2$; at 245.3 nm is $2.5 \pm 0.7 \text{ MW/cm}^2$; at 242.9 nm is $4.4 \pm 2.1 \text{ MW/cm}^2$; and at 240.6 nm is $7.9 \pm 4.2 \text{ MW/cm}^2$. Note how the variance in the average value increases at the shorter wavelengths (higher ν values). This is because the calculated I_{sat} values appear to be more sensitive to the intracavity flux level at the shorter wavelengths (see Table B-1). This probably implies that the analysis technique used for deriving the effective I_{sat} may be losing its validity in this region of the spectrum. In any case, the value of I_{sat} at 248.5 nm of 1.3 MW/cm^2 is in good agreement with the 1.2 MW/cm^2 value determined from separate measurements.

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Section 4

CONCLUSIONS

Our sidelight fluorescence measurements indicate that extraction from the KrF^* upper state manifold is not as homogeneous as previously thought. Theoretical efforts must be performed to understand why this occurs and how it impacts the usage of KrF lasers for ICF applications.

Dr. Chris Patterson and Dr. David Hanson at LANL have developed one possible explanation. If there was nonsequential relaxation occurring from the D-state to the B-state, this would repopulate the KrF^* upper state levels nonuniformly. Unfortunately, we are not aware of any experimental data, other than our own, which supports this hypothesis. There is perhaps a more fundamental question of whether this can even happen in this type of molecular system. All of these questions are beyond the scope of this program.

It would be very informative to perform experiments specifically designed to examine the D-state behavior relative to the B-state. There has been little work done in examining the D-state, probably because no one thought its role in the KrF kinetics process was very important until now.

The effects of nonuniform energy extraction from the KrF upper state manifold on the performance of the laser can be best determined by including the results of this program in the existing KrF models. This information can then be used to estimate the effects on laser efficiency.

Appendix A

CALIBRATION OF FLUORESCENCE DETECTION SYSTEM

A.1 INTRODUCTION

The primary recording medium during this experiment is film. Film has advantages and disadvantages. It was selected to use during this work because of the very low fluorescence emission from the higher ν levels which require a very sensitive detector. Photodiodes, which are used to detect the wavelength-integrated fluorescence signal, do not have enough gain to detect the higher ν level emissions. Photomultipliers do have enough gain; however, past attempts to use a photomultiplier near the Tahoma laser have been marginally successful at best. This is because the relatively high magnetic guide field of the laser (2 kgauss) and the x-rays emitted from the device tend to disrupt the photomultiplier despite shielding the tube. Conventional vidicon and CCD cameras also do not have enough gain and/or are not sensitive to the UV light. Image intensified CCD cameras may work, although there still may be noise problems associated with operating near the laser (delicate electronic equipment such as oscilloscopes cannot be operated unshielded near the laser) and some type of phosphor may still be needed to convert the UV light to visible. In any event, an intensified CCD camera was not available during these experiments.

The major disadvantage of film is that it must be carefully calibrated. Complicating this calibration is the fact the film response changes depending not only on the intensity of the light, but also the pulse duration and wavelength of the light. Additional care must be taken when developing the film to ensure that the same process is used every time a different batch of negatives is developed. Although the development process in general does not alter the basic calibration of the film, it can affect other characteristics such as the film background grey level. The film response may also have a wavelength dependence; however, in this particular experiment it turns out to have a relatively flat response over the spectral range of interest.

Because the usefulness of the experimental data depends critically on an accurate calibration of the film and the rest of the fluorescence collection system, a good portion of this experiment concentrated on calibrating all the components and understanding their characteristics. This appendix describes in the detail the calibration procedure.

A.2 DETERMINATION OF FILM DENSITY RESPONSE

The film used during the entire experiment is Kodak T-Max (ASA 400). The negative is exposed to the UV light and developed using a 3-stop push during the process. Even when using this sensitive film, we found it necessary to push the film during development to reveal the very faint emissions from the $\nu = 3$ level. As explained earlier in this report, part of the reason the emission was very faint is because we operated the 1-m spectrometer at 4th order. We also found it necessary to use multiple exposed film (e.g. 5 shots) to ensure that the $\nu = 3$ signal was well out of the noise.

The ideal manner to calibrate the film is to use the exact same pulse duration and wavelength of the actual experiment. A discharge KrF laser produces the correct wavelength, but its typical pulse duration (10 - 20 ns) is over an order of magnitude shorter than the e-beam pumped fluorescence pulse during the experiment (440 ns). Hence, there can be a question regarding the accuracy of the calibration if a discharge KrF laser is used. We overcame this problem by using the ASE output from the Tahoma laser as the source of light to expose the film. This meant the pulse duration and wavelength were exactly the same as during the actual experiment.

With the output coupler removed on the Tahoma resonator and only the Max R rear mirror in place, a fairly uniform, but incoherent and broad beam of ASE light is emitted from the laser chamber window on the output side of the resonator. As mentioned in the main report, this ASE flux is not intense (55 kW/cm²), but it is more than enough to expose the film. The entire film negative (7.6 cm x 12.7 cm) is exposed to the ASE beam. To help ensure uniform illumination of the negative, the ASE beam is reflected off a scattering plate onto the negative. (The homogeneity was verified by measuring the uniformity of an entire negative exposed in this manner.) Covering the negative

is a holder in which 2.54 cm diameter calibrated UV neutral density (ND) filters of varying transmission values are held. This means a wide range of intensities can be obtained on the same negative without the need to normalize to the ASE output intensity. (The ND filters are calibrated separately using a separate UV laser source and energy ratiometer, and a Beckman spectrometer.)

Since the film during these calibration measurements is developed in the same manner as the actual experiment (i.e. with a 3-stop push), the development process is also accounted for during the calibration. Multiple exposed film is tested in this same manner and found to follow the expected dependency (e.g. 5 shots gives 5 times more intensity).

The results of this calibration are shown in Figure A-1, which gives the film density as a function of the normalized intensity. Also plotted in Figure A-1 are the calibration data (open circles) from C. Gomez of LANL who also used Kodak T-Max film. He used a commercial discharge KrF laser as his calibration source, hence the pulse length was very short. There is a large difference in the two calibration curves whose reason is unknown (see Appendix C). It may not be simply because of the difference in pulse lengths. Since we do not know the absolute intensity of Gomez's source, we must be careful when comparing the two sets of data.

A.3 WAVELENGTH-DEPENDENT TRANSMISSION CALIBRATION

As shown in Figure 2-1, the fluorescence emission from the sample volume is imaged through a number of different optics before reaching the spectrometer. Each of these components may have nonuniform transmission characteristics over the wavelength range examined during the experiment (240 - 251 nm). The spectrometer itself will in general have a wavelength-dependent transmission characteristic. Hence, it is important to calibrate the transmission characteristics of the entire optical system.

This calibration is performed using a tunable Nd:YAG pumped dye laser whose output is doubled into the UV and sent through the optical system. A laser energy ratiometer is used to compare the light input with the transmitted light output as a function of wavelength.

Figure A-1. Film Calibration Curves for Kodak T-Max film. The + symbols are STI data and the open circles are data from LANL.

Figure A-2 shows the transmission characteristics for the 1-m spectrometer (open circles) operated at 4th order, and the collection tube assembly (+ symbols) that includes the fused quartz input window and all the lenses (see Figure 2-1). Combining these results in Figure A-3 yields the net transmission characteristics of the entire fluorescence collection system up to but not including the film. Basically, the transmission decreases in a monotonic fashion with shorter wavelength. Hence, the raw data is corrected by multiplying the spectral data by the inverse of this transmission curve.

As mentioned earlier, the spectral response of the film over the KrF spectrum is fairly flat (<5%). This is inferred from tests exposing the film to the output of a D₂ lamp sent through the spectrometer. Since the D₂ lamp output intensity has nearly the inverse spectral characteristics as the spectrometer (see Figure A-2), in other words its output intensity increases with decreasing wavelength, one would expect a uniform film density with wavelength if the film response is flat. Indeed, this is what is found.

A.4 CALIBRATION OF DENSITOMETER SYSTEM

The film density is measured using a CCD camera to view the negative against a white light background. The output from the CCD camera is sent into PC computer system where a frame grabber board digitizes the image of the negative (256 bits resolution). By orienting the negative so that the spectral image lies parallel to the camera raster lines, it is possible to average the data over several raster lines covering the width of the spectral image. Software programs have been developed to automatically incorporate the calibration and correction factors after digitizing the negative.

Separate steps are taken to calibrate this densitometer system and to understand its characteristics. ND filters calibrated in the visible using the Beckman spectrometer are imaged by the CCD camera system. The results are shown in Fig. A-4. The slope is very close to unity indicating the camera response is linear over this range.

Other more subtle aspects of using the video densitometer system include operating with the room lights off and positioning the negatives on the same

Figure A-2. Transmission Characteristics for the 1-m Spectrometer (Open Circles) and the Collection Tube Assembly (+ Symbols).

Figure A-3. Net Transmission Characteristics of the Entire Fluorescence Collection System.

Figure A-4. Calibration Check of the CCD Camera Used in the STI Densitometer System.

location on the light table. These steps all help ensure the densitometry process is identical for each set of negatives.

A.5 MATHEMATICAL BASIS FOR FILM DENSITY INTERPRETATION

The actual process of determining the spectral density from the film density requires several steps in order to eliminate the background density inherent in the film negative. We assume that the background is uniform and is due to the development process (i.e. the 3-stop push process), and that it is not affected by the fluorescence emission. These assumptions are reasonable and were verified by testing unexposed film. This implies the fluorescence data has a simple background density level that can be subtracted out.

The densitometry process involves obtaining the spectral image on the negative yielding, $D_{\text{spec}}/D_{\text{table}}$, where D_{spec} is the digitized data of the spectrum and D_{table} is the light table intensity. (The exact value of this latter quantity is not important since it will be canceled out later.) Then,

$$-\log \left(\frac{D_{\text{spec}}}{D_{\text{table}}} \right) \propto \text{Spectral Density} + \text{Background Density}.$$

The digitizing measurement is performed on the same negative in a region away from the spectral image to obtain the background level density D_{back} . Then,

$$-\log \left(\frac{D_{\text{back}}}{D_{\text{table}}} \right) \propto \text{Background Density}.$$

Combining these two measurements yields the net spectral density,

$$\log \left(\frac{D_{\text{back}}}{D_{\text{spec}}} \right) = \text{Net Spectral Density}.$$

Appendix B

DATA REDUCTION PROCESS

B.1 INTRODUCTION

Reduction of the raw data to yield the net sidelight suppression requires a method for incorporating the calibration and transmission factors of the diagnostics and, more importantly, accounting for time-dependent behavior effects of the sidelight fluorescence that are not revealed in the time-integrated measurements. Specifically, it is well known that the sidelight fluorescence does not suppress immediately when the e-beam is fired. Instead during the beginning of the e-beam pulse when the laser is below lasing threshold, the sidelight fluorescence increases in a manner similar to its behavior during nonlasing shots. When the laser begins to lase the fluorescence starts to decrease and levels off at its suppressed level. This temporal behavior is shown schematically in Fig. B-1. At the termination of the e-beam pulse, lasing stops and the fluorescence begins to rise again due to residual pumping from the tail end of the e-beam pulse. Since the film integrates over time, the "extra" photons produced at the beginning and end of the e-beam pulse tend to make the net suppression appear less than it really is. Therefore, a way is needed to subtract these extra photons from the raw data to yield the true suppression. (Another way of stating the problem is that the time-dependent suppressed sidelight fluorescence profile could not be obtained at the same time as the wavelength-dependent data. To do this would have required using a detector such as a UV-sensitive, light-amplified streak camera instead of film.)

Our basic approach is to model the sidelight fluorescence and laser output temporal profiles as simple trapezoids and to estimate using the standard homogeneous saturation equation [i.e. $(1 + I/I_{\text{sat}})^{-1}$] the temporal behavior of the fluorescence. The saturation flux, I_{sat} , is solved from the raw data in a self-consistent manner that includes the temporal behavior depicted in Fig. B-1.

Figure B-1. Schematic Representation of the Temporal Behavior of the Suppressed Sidelight Fluorescence. The dashed curve is the unsuppressed (nonlasing) fluorescence, the dotted curve is the laser output, and the solid curve is the suppressed (lasing) fluorescence.

B.2 DESCRIPTION OF REDUCTION PROCESS

B.2.1 Transmission Correction Factors

The total transmission characteristics of the sidelight fluorescence optical collection system is given by,

$$T_{\text{tot}}(\lambda) = T_{\text{spec}}(\lambda) \cdot T_{\text{optics}}(\lambda) \cdot T_{\text{detect}}(\lambda) \cdot R_{\text{film}}(\lambda),$$

where $T_{\text{spec}}(\lambda)$, $T_{\text{optics}}(\lambda)$, and $T_{\text{detect}}(\lambda)$ are the transmission characteristics as a function of wavelength of the spectrometer, the collection optics (e.g. lens, coated mirrors), and the energy meter (used during the calibration measurements), respectively. $R_{\text{film}}(\lambda)$ is the wavelength-dependent film response.

The spectral response was measured for all the parameters. It was found that T_{detect} and R_{film} are fairly flat with wavelength. Hence, we set $T_{\text{detect}}(\lambda) \cdot R_{\text{film}}(\lambda) = 1$ for all λ . The spectral response of $T_{\text{spec}}(\lambda) \cdot T_{\text{optics}}(\lambda) \cdot T_{\text{detect}}(\lambda)$ was given earlier in Fig. A-3. Using a best fit polynomial routine applied to the response curve in Fig. A-3, we find that

$$T_{\text{tot}}(\lambda) = 14.6 - 0.148\lambda(\text{nm}) + 3.73\lambda^2(\text{nm}).$$

B.2.2 Temporal Behavior Correction

Figure B-2 is a schematic representation of the unsuppressed (nonlasing) sidelight fluorescence, $I^{\text{non}}(t, \lambda)$, and the laser output, $I^{\text{lase}}(t, \lambda)$, pulse shapes modeled as trapezoids, where,

$$I^{\text{non}}(t, \lambda) = I_0^{\text{non}}(\lambda) \cdot f(t),$$

$$I^{\text{lase}}(t, \lambda) = I_0^{\text{lase}}(\lambda) \cdot g(t),$$

and $f(t)$ and $g(t)$ are simple linear functions of time. Except for a dip in the central portion of the real pulse shapes caused by impedance ringing,

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Figure B-2. Schematic Representation of the Unsuppressed (Nonlasing) Sidelight Fluorescence and Laser Output Waveforms.

the trapezoid shapes are reasonable first-order approximations to the actual pulse shapes.

We next assume the KrF gain medium saturates homogenously so that,

$$\frac{\text{Suppressed Fluorescence}}{\text{Unsuppressed Fluorescence}} \propto \frac{g}{g_o} = \frac{1}{1 + \frac{I_{\text{cav}}(t)}{I_{\text{sat}}(\lambda)}} ,$$

where $I_{\text{cav}}(t)$ is the time-dependent intracavity flux at 248 nm, and $I_{\text{sat}}(\lambda)$ is assumed not to change with time.

Then the time-integrated intensity for the normalized waveform during lasing is given by,

$$S_{\text{lase}}(\lambda) = \int_{T_0}^{T_1} \frac{I_{\text{non}}^{\text{non}}(t, \lambda)}{I_o^{\text{non}}} dt + \int_{T_1}^{T_2} \frac{I_{\text{non}}^{\text{non}}(t, \lambda)}{I_o^{\text{non}}} \left[\frac{1}{1 + \frac{I_{\text{cav}}(t)}{I_{\text{sat}}(\lambda)}} \right] dt +$$

$$\int_{T_2}^{T_3} \dots + \int_{T_3}^{T_4} \dots + \int_{T_4}^{T_5} \dots + \int_{T_5}^{T_6} \dots + \int_{T_6}^{T_7} \frac{I_{\text{non}}^{\text{non}}(t, \lambda)}{I_o^{\text{non}}} dt . \quad [\text{B1}]$$

The corresponding normalized waveform during nonlasing is given by,

$$S_{\text{nonl}}(\lambda) = \int_{T_0}^{T_7} \frac{I_{\text{non}}^{\text{non}}(t, \lambda)}{I_o^{\text{non}}} dt . \quad [\text{B2}]$$

The intracavity flux $I_{\text{cav}}(t)$ as a function of time can be obtained from the lasing data,

$$I_{\text{cav}}(t) \approx \frac{E_{\text{out}}}{A\tau} \left[\frac{1+R}{1-R} \right] g(t) , \quad [\text{B3}]$$

where E_{out} is the time-integrated energy output measured during the experiment, A is the area of the cavity mode inside the laser [essentially defined by the spacing between the sidelight observation tube and razor-blade beam block (see Fig. 2-1)], τ is the temporal width of the trapezoid waveform equal to $[(T_6 - T_1) + (T_4 - T_3)]/2$, R is the output mirror reflectivity, and $g(t)$ describes the trapezoidal time behavior of the lasing waveform. For our situation, the plateau of the trapezoid is given by,

$$I_o^{\text{lase}} \approx \frac{E_{\text{out}}}{A\tau} \left[\frac{1+R}{1-R} \right] . \quad [\text{B4}]$$

Therefore,

$$I_{\text{cav}}(t) \approx I_o^{\text{lase}} \cdot g(t) . \quad [\text{B5}]$$

Hence, in Eqns. B1 and B2, all the parameters are known except for $I_{\text{sat}}(\lambda)$.

B.2.3 ASE Correction

The intensities $S_{\text{lase}}(\lambda)$ and $S_{\text{non}}(\lambda)$ used in Eqns. B1 and B2 are adjusted values, in which the raw intensity data is divided by the transmission factor $T_{\text{tot}}(\lambda)$ and an ASE correction factor $C_{\text{ASE}}(\lambda)$. This correction factor accounts for the small amount of flux that is present in the cavity even during nonlasing primarily because of the rear mirror (see Fig. 2-1) which is unblocked during nonlasing conditions. (Only the output coupler is blocked to prevent lasing during nonlasing conditions.) This flux was measured to be 55 kW/cm².

From data taken for both single and double pass ASE conditions, the value of $C_{\text{ASE}}(\lambda)$ is,

$$C_{\text{ASE}}(248.4 \text{ nm}) = 0.95 \text{ and } C_{\text{ASE}}(245.3 \text{ nm}) = 0.99.$$

B.3 TABULATED DATA RESULTS

Table B-1 lists as a function of the intracavity flux, the raw data suppression ratios (referenced to the peak value at 248.5 nm) at the designated wavelengths (corresponding roughly to the ν level), the corrected suppression ratios, the calculated values for I_{sat} , and the average value of I_{sat} over the range of intracavity fluxes tested.

Table B-1.

SUPPRESSION RATIO DATA

	$I_{\text{cav}2}$ (MW/cm ²)	Wavelength (nm)			
		248.5	245.3	242.9	240.6
Raw Data Ratios	4.9	0.30	0.46	0.63	0.76
	3.6	0.34	0.50	0.61	0.73
	1.7	0.53	0.60	0.65	0.75
Corrected Ratios	4.9	0.23	0.39	0.57	0.72
	3.6	0.27	0.43	0.55	0.68
	1.7	0.43	0.51	0.57	0.69
Calculated I_{sat} (MW/cm ²)	4.9	1.44	3.10	6.45	12.26
	3.6	1.30	2.70	4.36	7.73
	1.7	1.29	1.79	2.28	3.82
Average value of I_{sat} (MW/cm ²)		1.3	2.5	4.4	7.9
		±0.1	±0.7	±2.1	±4.2

Appendix C

KODAK T-MAX FILM CALIBRATION

C.1 BACKGROUND

A key issue during this program is the correct calibration of the film used to obtain the spectral data. As mentioned earlier, Kodak T-Max film (ASA 400) is used, which is the same film used at LANL to obtain their ASE spectral data (see Table 3-1). We performed our own calibration of the film independent of the calibration done by LANL. Comparing the calibration curves, we find there is a significant difference between our curve and the LANL curve regarding how the density of the film scales with exposure. This difference dramatically affects the shape of the spectral data, especially the ratio of the peaks of the spectrum. As shown in Table 3-1, the ratios measured by LANL are much smaller than the ones measured during this program.

In this appendix we discuss in more detail how we calibrated our film and how, as we understand, it is different from the method LANL used to calibrate their film. Unfortunately, there is not enough known to explain why the two calibration curves are so different. We offer some speculation on possible causes, but this issue cannot be resolved without further experimental tests.

C.2 POSSIBLE SOURCES OF ERROR IN CALIBRATION

It should be first mentioned that neither LANL or STI conducted densitometry calibration measurements on a common negative. Therefore, although we were using the same film from the same manufacturer, we never had a chance to calibrate the same piece of film. We must assume that the LANL densitometer was properly calibrated. We calibrated our densitometer separately from this experiment using calibrated ND filters whose transmission characteristics were experimentally verified (see Appendix A).

The second point is if we assume the densitometers at both facilities were operating correctly (i.e. the same transmission for a negative would be measured at both facilities), then issues concerning film or developer characteristics, the development process, or illumination dependent dynamics, such as the physics of photographic grain formation during very short exposure times, need to be addressed.

Assuming that none of the preceding factors are responsible for the disagreement, then the third possibility is analysis error. Specifically, we are referring to errors in reducing the data during the calibration process. Without going through and examining in great detail both the LANL and STI lab books and data, it is not possible to determine whether analysis error is to blame. Therefore, the remainder of this discussion will be limited to the second point.

C.3 ISSUES AND POTENTIAL PROBLEMS WHEN WORKING WITH FILM

There are a number of potential problems when working with film. For example, during the development process we encountered background darkening in some of the developed T-Max negatives which probably were due to using developer a few months beyond its 2-month shelf-life. The resultant net film density diminished, but it did not appear to significantly affect the relative intensity ratios obtained from the same negative. This problem with old developer was not investigated further because the questionable data was simply discarded and fresh developer was always used for the rest of the measurements. In addition, we carefully controlled the chemical temperatures and development times to ensure that the development process was identical every time a negative was developed.

As explained in Appendix A, component transmission characteristics were measured for the entire optical collection system up to the film. A narrowband, tunable, pulsed laser source and an energy meter were used for this measurement. The film spectral response was also checked using a cw D_2 light source and the spectrometer. The folded film response and the spectrometer transmission data from the pulsed laser measurements yielded the expected D_2 intensities ($\pm 10\%$) when checked at two points spanning ≈ 7

nm (241.5 nm and 248.5 nm). This indicates a flat film response with wavelength, which we believe was also concluded by LANL.

The response characteristics of film for short pulses of light may be an important issue; however, it is not clear how significant this effect may be. LANL calibrated their film using a short pulse (~ 10 ns) laser source; we calibrated our film using the same exposure time (440 ns) as used during the fluorescence suppression measurements. Photographic grain formation during very fast (10 ns) exposure times might impact density by reducing the grain size or the number of grains (reducing the exposed emulsion depth).

Figure C-1 plots film density versus $\log(I/I_0)$ for our pulsed (0.44 μ s) and quasi-cw (1-2 s) D_2 lamp calibration data, and the LANL calibration curve as transcribed from data they provided us. The vertical line represents the minimum intensity ($I/I_0 = 0.07$) that could be detected above the noise level for our experiment. The calibration discrepancy is clearly evident in the difference in slopes (relative film response) between our data and LANL's curve. Note that our pulsed and quasi-cw data yield the same relative response. This might imply that for pulse lengths ≥ 0.5 μ s the film response is similar. However, it does not necessarily imply that this trend still holds true for pulse lengths much shorter than ~ 0.5 μ s (i.e. 10 ns). There is insufficient data to make a conclusion one way or the other.

Density blooming occurs in films when attempting to record a faint intensity region that is close to a bright region, such as occurs when looking at the higher ν levels next to the $\nu = 0$ level in the KrF fluorescence spectrum. The fainter area appears brighter (i.e. denser) than it should be because of scattered light coming from the neighboring high intensity region. Significant blooming can occur even when the high intensity region is below saturation of the film.

We verified that blooming is negligible for relative intensity gradients of $\sim 13X/mm$ (at the film plane) within 1.5 mm of a region have a peak density of 1.1. Because we operated our 1-m spectrometer on 4th

T-MAX FILM CALIBRATION (CW AND PULSED)

Figure C-1. Kodak T-Max Film Density Versus Light Intensity. The open triangles are STI data using the Tahoma laser (440 ns pulse length). The solid circles are STI data using the cw D₂ lamp. The solid curve is LANL data using a short pulse (~10 ns) laser source.

order, the dispersion at our film plane is very large (0.15 nm/mm) resulting in <1X/mm maximum relative intensity gradient in the nonlasing KrF spectrum. Therefore, we were far from any blooming effects during our measurements.

We can perform a similar calculation for the LANL measurements, although we do not know for certain all the parameters for their conditions. Making a reasonable estimate for gain in the SAM (1%/cm) and assuming a conservative estimate for the dispersion of a small spectrometer operating at 1st order (1.5 nm/mm), an ASE calculation (discussed next) predicts that a relative intensity gradient of 11X/mm is possible on a LANL negative. However, we still lack short pulse exposure data at densities of ≥ 1.5 to know if this gradient is high enough to start encountering blooming problems.

We performed a simple ASE calculation to check the predicted g_0 for the SAM based on the LANL ASE spectrum shown in Fig. C-2 and using both ours and LANL's calibration curves. The calculation assumes that the calibration curves and ASE spectrum density are correct, and uses a SAM gain length of 100 cm. Assuming negligible gain loading, the ratio of the peak intensities at $\nu = 0$ and $\nu = 1$ is,

$$\frac{I_{\nu=0}}{I_{\nu=1}} = \frac{g'_0}{g'_1} \exp \left[g'_0 \left(1 - \frac{g'_1}{g'_0} \right) L \right], \quad [C1]$$

so that,

$$g'_0 = \ln \left(\frac{I_{\nu=0} g'_1}{I_{\nu=1} g'_0} \right) \left[\left(1 - \frac{g'_1}{g'_0} \right) L \right]^{-1}. \quad [C2]$$

The small signal effective gains at 248.4 nm and 245.3 nm are g'_0 and g'_1 , respectively. In our experiment, the gain length L is ~ 1 cm, so the measured intensity ratio is roughly the same as the relative gain ratio. From Table 3-1, the Patterson and Hanson model predicts a gain ratio for g'_0/g'_1 of 4. Using this value in Eq. C2 and the LANL intensity ratio of 2.9 (see Fig. C-2), we obtain an unreal value for g'_0 . If we assume $g'_0/g'_1 = 2$, then $g'_0 = 0.7\%/cm$.

Figure C-2. LANL KrF Amplified Spontaneous Emission (ASE) Spectrum of the Small Aperture Module (SAM). Courtesy of C. Gomez (LANL).

This low gain ratio necessary to achieve the expected gain, which might be related to an intensity compression occurring in the ASE spectrum, suggests a film exposure problem, e.g. blooming.

If we apply our calibration data instead to the LANL ASE spectrum, the intensity ratio then equals $2.9^{2.6}$ (the ratio of the calibration slopes is exponentiated) or 15.9. An effective gain ratio of 10 (using our own data given in Table 3-1) then yields $g'_0 = 0.5\%/cm$. This value for the gain is too low for the SAM. If we work backwards and ask ourselves what amount of density blooming would be necessary to have an intensity ratio of 15.9, an effective gain ratio of 10, and a gain of $1\%/cm$, we find it only takes 1.6X blooming to be occurring in the ASE spectrum. These preceding two calculations do not prove that blooming is occurring in the LANL data, but it does suggest that some intensity compression may be occurring in the LANL ASE spectrum and that blooming is one effect which can explain this in a self-consistent manner.

Further evidence supporting a possible blooming problem with the LANL data is that the width of the 248.5 nm peak is significantly wider than either measured or predicted by others (see Table 3-1).

C.4 ADDITIONAL EVIDENCE SUPPORTING OUR MEASUREMENT RESULTS

Additional data that supports our sidelight suppression results are shown in Fig. C-3, which is a ratio of two KrF fluorescence spectra centered at 246.8 nm. The first spectrum is an unattenuated single-shot exposure. The second spectrum is a 2-shot exposure using an ND 1.05 filter to attenuate the signal by 11.2X. The dashed horizontal line is the predicted ratio based upon the ND filter ($1/11.2 = 0.9$) and the open circle represents the wavelength region where the filter was calibrated. The sharp rise in the relative intensity ratio below 245 nm is caused by density that is too low for accurate analysis and, therefore, it delineates the lower limit of the measurement.

To first order, the analysis normalizes the calculated intensity to the number of shots. The entire reduction procedure is put to a stringent test here because density scales with the product of exposure time and exposure

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NONLASING SPECTRA RATIO USING OD 1.05

DOTS = Ratio

Mix: 14.5% Kr/Ar/F₂

Reflectors: NONE >98%

Output Fluence: 45 mJ/cm²

Cavity Flux: ≈55 kW/cm²

NOTES:

1. 1 shot exposure on Tmax film.
2. λ cal. set by Hg calibration and fiducial marker on film.
3. Output beam diameter = 1.8 cm
4. Spectrometer N.D. = 1.05

↓ Indicates fiducial marker
λ_{FD} = 248.775

Figure C-3. Ratio of Two Nonlasing Fluorescence Spectra Where One Spectrum Has Been Attenuated by 11.2X.

intensity. Both quantities are changed simultaneously in this test. (This test will not, however, reveal erroneous system transmission problems.) The error bar in the figure is the uncertainty associated with the ND filter. Conservative estimates for the uncertainty in the calculated intensity ratio are comparable to this error bar. The second spectrum was double-exposed to assure that the density at 245.3 nm ($\nu = 1$) is high enough for reliable analysis, and the ~10X attenuation was intentionally chosen to match our measured ratios between 248.5 nm and 245.3 nm (see Table 3-1). We see that the data is within the expected range, thereby adding further support to our calibration data.

The two spectra are plotted in Fig. C-4 without correcting for exposure time. The nominal maximum relative intensity encountered in the data is ≈ 5 . This corresponds to a maximum dynamic range of 70 for a single negative. (The usable upper density limit was not determined.) Hence, the data we collected should be well within the dynamic range of the film. [Additional nonlasing data was obtained operating at 1st order on the spectrometer (0.6 nm/mm at the film plane) and indicated a primary to secondary fluorescence peak intensity ratio of ~10 in agreement with the 4th order results.]

C.5 CONCLUSIONS

As stated at the beginning of this appendix, the only real conclusion that can be made is there is insufficient data available to explain why the film calibrations differ. Blooming may be able to explain the difference in the ratio of the fluorescence peaks, but it cannot explain the difference in the slopes of the calibration curves.

However, if we were forced to choose a reason, probably the most likely candidate is the short pulse (10 ns) exposure procedure LANL used to calibrate their film. It is conceivable that nonlinear effects in the film grain formation behavior could occur that would be sensitive to the peak power of the light source rather than the energy dosage. This also might be consistent with what appears to be a minimum intensity threshold seen in Fig. C-1 for the LANL calibration curve before the onset of any appreciable density formation. Our calibration data does not demonstrate any threshold-like behavior. Again,

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SPECTRA WITH AND W/O OD 1.05 FILTER

Figure C-4. Same Spectra Plotted in Fig. C-3 Demonstrating That the Data are Well Within the Dynamic Range of the Film.

this is only speculation on our part. Further tests of the film are required before this issue can be resolved.