34th JANNAF Combustion Subcommittee Meeting, MURI Technology,
CPIA Publication 662, Vol. II, pp. 307-314, October 1997

HIGH-RESOLUTION X-RAY DIAGNOSIS OF ROCKET COMBUSTION WITH LASER-PLASMA SOURCES

Martin Richardson, Chris DePriest, Steve Grantham & David Torres
LPL, CREOL, University of Central Florida
4000 Central Florida Blvd., Orlando, FL 32816-2700
Tel # 407-823-6819, Fax # 407-823-3570
Email: mcr@creol.ucf.edu

ABSTRACT

Recent progress in high intensity lasers has lead to the development of a new class of ultra-bright sub-picosecond point (micron-size) hard x-ray sources. Used for x-ray radiographic imaging, these sources offer a means to analyze transient phenomena in dense media with high time and space resolution. We are applying this new technology to analysis of the combustion of metal particles in solid propellant rocket burners. In this highly transient environment, knowledge of the particle size and velocity can provide an important understanding of the combustion dynamics. Although optical techniques can be used to analyze the burn-rates of individual particles, this x-ray approach will permit the analysis of particle combustion in the optically opaque conditions of a rocket burner.

A laser-plasma hard x-ray facility has been constructed and is currently being characterized that will permit the radiographic analysis of transient phenomena in dense media. It is based on a high energy (> 1J), 100 femtosecond solid state laser system that is capable of producing ultrashort laser plasmas at intensities in the 10^{19} W/cm^2 range. Initial results from the system are described and its application to the analysis of particle combustion will be discussed.

INTRODUCTION

Solid propellants routinely use aluminum particles as a fuel ingredient to increase the specific impulse of rocket motors[1]. They also to reduce aerodynamic instabilities in the combustion. The aluminum particles are typically 20 – 100 microns in size. The macroscopic burning behavior of aluminum combustion has been well characterized. However the detailed dynamics of the aluminum particles in the combustion region are not firmly understood. In particular, the role of a protective AlO_3 layer that is formed around the particle is thought to inhibit ignition and prolong burn times. The melting and boiling points of AlO_3 are significantly higher than those for Al. Shorter ignition times are crucial to achieving complete utilization of the high energy density of aluminum by ensuring that the particles remain longer in the combustion region and that most of the combustion products enter the gas phase. A number of detailed studies of the combustion of Al have been made in recent years[2-5]. Roberts et.al. [4] studied the ignition and combustion in high pressure O_2 in shock tube experiments, while other investigations have studied the combustion of Al particles in atmospheric pressures of air, water vapor and CO_2 mixtures [2,3]. Ishihara and Brewster[5] studied the combustion behavior of aluminized composite propellants.

Another approach to understanding the combustion of Al is through single particle studies. Experiments in air with laser-ignited single Al particles are currently being made by Yetter et.al. [6], using planar laser-induce fluorescence techniques which allow for detailed diagnosis of the chemical species and temperature profiles around the burning particle. These follow earlier spectroscopic studies of Al in the combustion region [7,8]. None of these approaches so far has allowed study of the microscopic behavior of the individual particles in the combustion region. Although optical scattering techniques have been used to study the Al particles [9], with new experiments with ultrashort laser pulses planned[10], no technique has been deployed so far that is capable of examining the physical shape and structure of the individual Al particles while they are in the combustion region. Such a capability would provide much useful data on the ignition and combustion of the particles. It would also help understand the process and the effects of particle agglomeration in the combustion region. Previous studies [11,12] have demonstrated the impact of agglomeration on the combustion process with aluminized propellants.

In this paper we describe a new approach to particle analysis in turbulent media. This approach depends upon the use of a new, extremely bright source of x-rays, and high resolution x-ray imaging to resolve the individual particles in the combustion region. X-ray diagnostic techniques have previously been used to study combustion using conventional discharge tube x-ray sources, however the source size and brightness and the emission duration were inadequate to make time resolved studies of individual particles in the combustion region. The approach we employ utilizes new capabilities arising from the study of dense plasmas produced by high intensity ultrashort laser pulses. This provides a synchronizable, picosecond-duration, ~10 micron-size point source of hard x-rays. This is an ideal x-ray source for point projection imaging of transient phenomena in dense media.

ADVANTAGES OF X-RAY ANALYSIS

The use of x-rays for probing and characterizing high temperature combustion environments has several advantages over optical probing techniques. These advantages stem from the properties of the burning materials in the x-ray region, and from the wavelength of the probing emission. The simplest configuration for probing the combustion region of a high temperature burner is shown schematically in Fig. 1. This is a straightforward point-projection imaging configuration in which the source is positioned some distance from the object, and a magnified image can be recorded at the image plane, located some distance on the far side of the object.

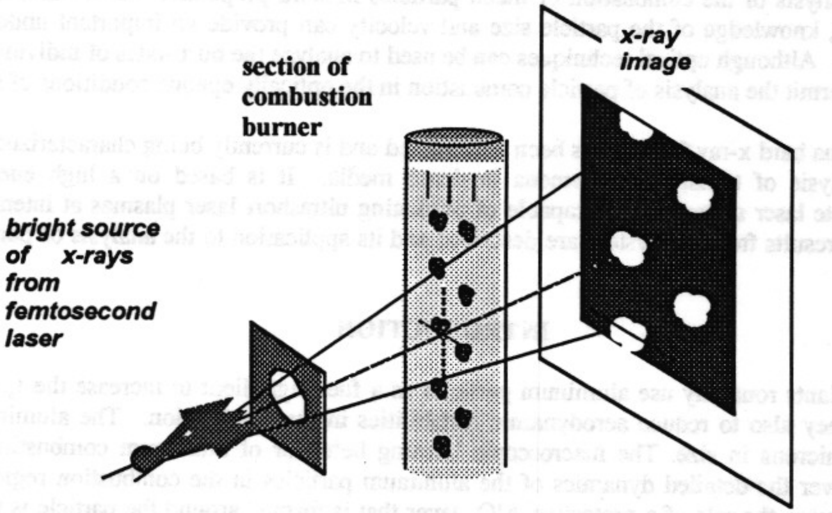

Fig.1. X-ray probing of the combustion region of solid propellants.

Because the wavelength of x-ray radiation, in the 0.1 – 1.0 nm range, is much smaller than most particles sizes in the combustion region, x-ray probing will be easier, since the level of scattering will be reduced*. This is one factor that leads to improved image contrast in a back-illuminated (or radiographed) image. Secondly, short wavelength radiation is less-sensitive to turbulent features within the combustion region, thereby improving the resolution of small solid, or liquid, particles. Thirdly, the absorption of x-rays by most of the materials commonly used in combustion is strongly wavelength-dependent in this wavelength range, corresponding to the absorption edges of their inner shell electrons. Consequently, judicious choice of the x-ray wavelength used in probing can provide high contrast of the x-ray image, and can also be used in mapping individual elements in the combustion region. Lastly, with x-ray probing the wavelength of the radiation is well removed from the peak of the Planckian spectral distribution of the light emitted by the combustion products. This distribution is centered in the visible region. Thus with selective x-ray filtering, image acquisition in the x-ray region is not hampered by the background self-emission.

The use of a laser-plasma x-ray source has several additional advantages for x-ray probing of combustion. This type of x-ray source is small, typically < 100 μm, and precisely located in space. The x-ray source that we have developed for this purpose is < 10 μm in physical extent. Thus in the simple projection imaging regime, in which the image resolution is directly related to the source size, laser plasma x-ray sources have distinct advantages over other x-ray sources. A second advantage arises for the intrinsic brightness of laser plasma x-ray sources. Consider a source like that shown in Fig.1. that is produced by a 1 J laser, situated 10 cm from a combustion burner of 1 cm spatial extent. Assuming a conversion efficiency, conservatively, of 0.01% into useful x-rays in the 1.0 nm wavelength region, the source will generate ~ 10^{12} useful photons that emit uniformly into a 2π region. Thus the photon density in the combustion region is ~ 10^{10} ph/cm^2. Assuming a desired pixel size (resolution) in this region of 10 μm, the number of photons per resolution element is ~ 10^4 ph/pxl. With the use of single-photon counting array detectors, this flux is ample to record distinct images of individual particles, even taking into account the absorption of the required x-ray windows etc.

A final advantage of laser plasma x-ray sources for this type of diagnosis stems from the duration of the x-ray emission. Most laser plasma x-ray sources have a duration of the order of 10 ns or less. Thus single shot images of 10 μm features can be recorded without motional blurring for particle velocities up to 10^5 cm/s. Particle velocities in combustion usually have velocities much less than this. Conventional discharge x-ray sources having emission durations in the microsecond region would not be free of this limitation.

A PICOSECOND POINT SOURCE OF HARD X-RAYS

Recently, development of ultrashort pulse optical lasers and high bandwidth solid-state amplifiers has resulted in relatively compact systems capable of multiterawatt power level [13]. Plasmas formed by focussing these laser pulses onto solid targets were discovered to radiate prompt hard x-rays at extremely high brightness [14,15].

The significance of these new hard x-ray laser-plasma sources is that they combine three unique characteristics. They are small in size (5 μm is reasonable), they are very bright (of order 10^{19} photons/mm^2•s•mrad^2 into 0.01% bandwidth at 5 keV, see Fig. 2), and the x-rays are hard (out to MeV, with reasonable fluences for single-shot imagery at 10 keV). Each of these attributes is substantially different from the radiation emitted by plasmas generated using longer-pulse high energy lasers such as YAG and Nd:glass lasers, because the laser-matter interaction physics is very different for very short pulses. Long pulses produce plasmas with outflowing electron density profiles having shallow gradients; the dominant absorption mechanism is inverse bremsstrahlung, and the energy is thermalized by electron collisions. Laser focal spot sizes are generally large (> 50 μm) with these systems, so the thermal energy is distributed among many ions. The result is, except for the very largest lasers, plasma electron temperatures of only hundreds of eV, and x-ray spectra falling off exponentially in intensity above 1 keV (following the blackbody curve). In contrast, short-pulse lasers interact with plasmas having very steep electron density fronts, whose 1/e scale lengths are comparable to an optical wavelength.

---

* separately we are examining the potential of x-ray scattering approaches to combustion diagnosis. There may be advantages in sensitivity to examining the characteristics of nanoscale particles with x-ray scattering techniques compared to optical approaches.

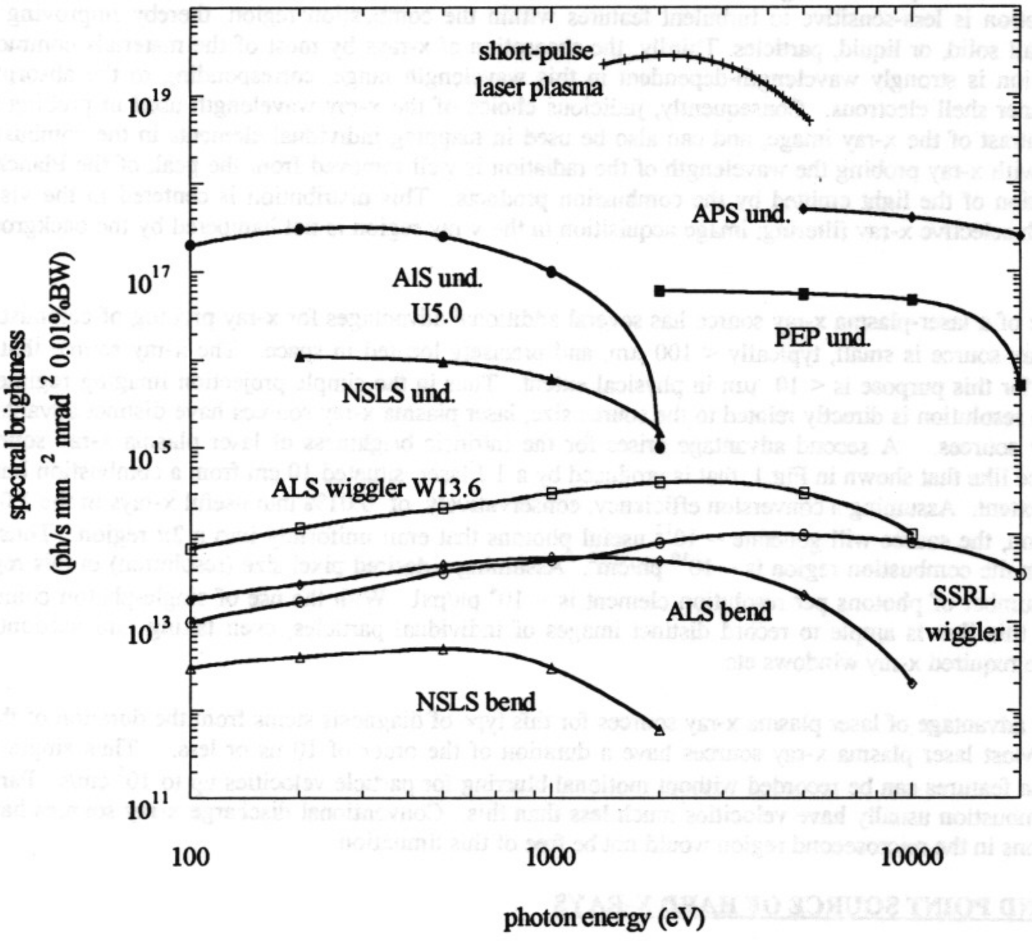

Figure 2. Some of the world's brightest x-ray sources. A comparison of the brightness figures between short-pulse laser plasma sources (data from Ref. 14, assuming blackbody) and large synchrotron facilities (ALS: Advanced Light Source, ALPS: Advanced Photon Source, PEP and SSRL: Stanford Synchrotron Radiation Laboratory, NSLS: National Synchrotron Light Source).

This changes the dominant absorption mechanism to resonance absorption, generating much higher energy electrons. As well, the optical pulse has an electric field gradient which is very steep, and this gives rise to a "ponderomotive potential" which accelerates the electrons into the target surface. The effect is like a high-current (10^{16} A/cm^2) electron beam at 10 keV or higher energy bombarding a tiny spot on the target. The bulk of the x-ray emission consists of high-energy continuum bremsstrahlung. So we have in these sources a unique microscopic "flashbulb" of penetrating radiation. An obvious use of these sources is therefore to take "snapshots" of the interiors of objects.

A new ultrashort laser facility has recently been commissioned at CREOL[16]. This laser system produces pulses of energy ~ 1 J in pulse durations of ~ 100 fs. This has now been configured with an integral target chamber that provides for the intense laser pulses to be focused precisely (with an accuracy of several microns) on to mass-limited targets. The arrangement of this target chamber and some of the primary x-ray diagnostics is shown in Fig.3. The laser pulse is focused onto the target with a highly reflecting off-axis parabolic mirror to focal dimensions of less than 10 μm.

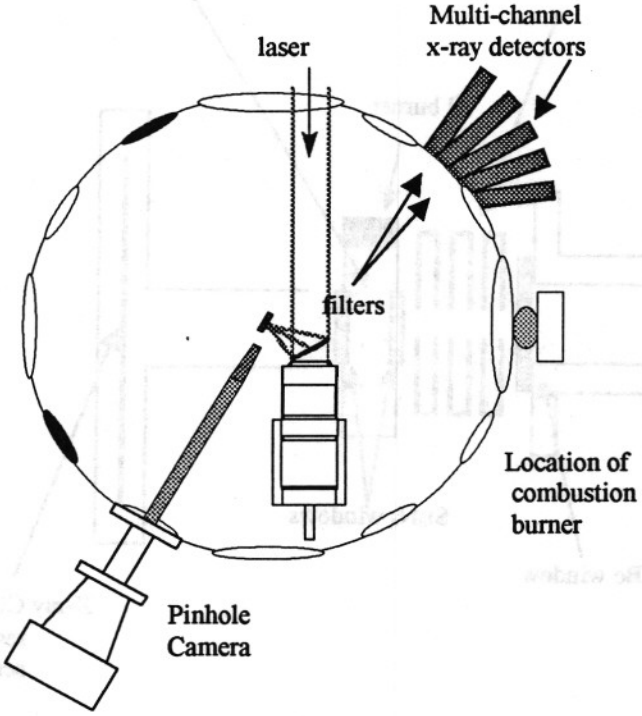

Fig.3. Laser plasma configuration of the picosecond x-ray source.

The spectral characteristics of the emission will be characterized by several different x-ray diagnostics. A calibrated multi-channel filtered x-ray detector spectrometer comprising both PIN x-ray diodes and scintillator-photomultiplier combinations will measure the x-ray emission in the 1 – 100 keV region. Crystal x-ray spectrographs and flat-field grazing incidence spectrographs will monitor the x-ray line emission in the lower energy (100 eV – 10 keV) region. A pinhole camera with a sensitive x-ray CCD has been constructed to image the plasma, and therefore x-ray source size.

The general configuration of the initial experiments on x-ray radiography of a combustion burner is also shown in Fig.3. The burner will be located close to one of the x-ray exit ports of the target chamber. A sensitive x-ray CCD will be used to capture the projection image.

INITIAL RADIOGRAPHIC MEASUREMENTS OF SMALL ALUMINUM PARTICLES

Experiments are now planned to make x-ray probe measurements with a high temperature combustion burner. The configuration of this burner, adjacent to an x-ray port on the target chamber is shown in Fig. 4. Images of the particles will be recorded with a sensitive x-ray CCD.

Initial experiments have been carried out to demonstrate the potential of this diagnostic approach to combustion. These experiments were made with single, cold Al particles. In these experiments, the burner was replaced with a thin beryllium substitute on which several Al particles were located. The aluminum particles were coarsely sieved to select particles with sizes in the range 80 – 120 microns.

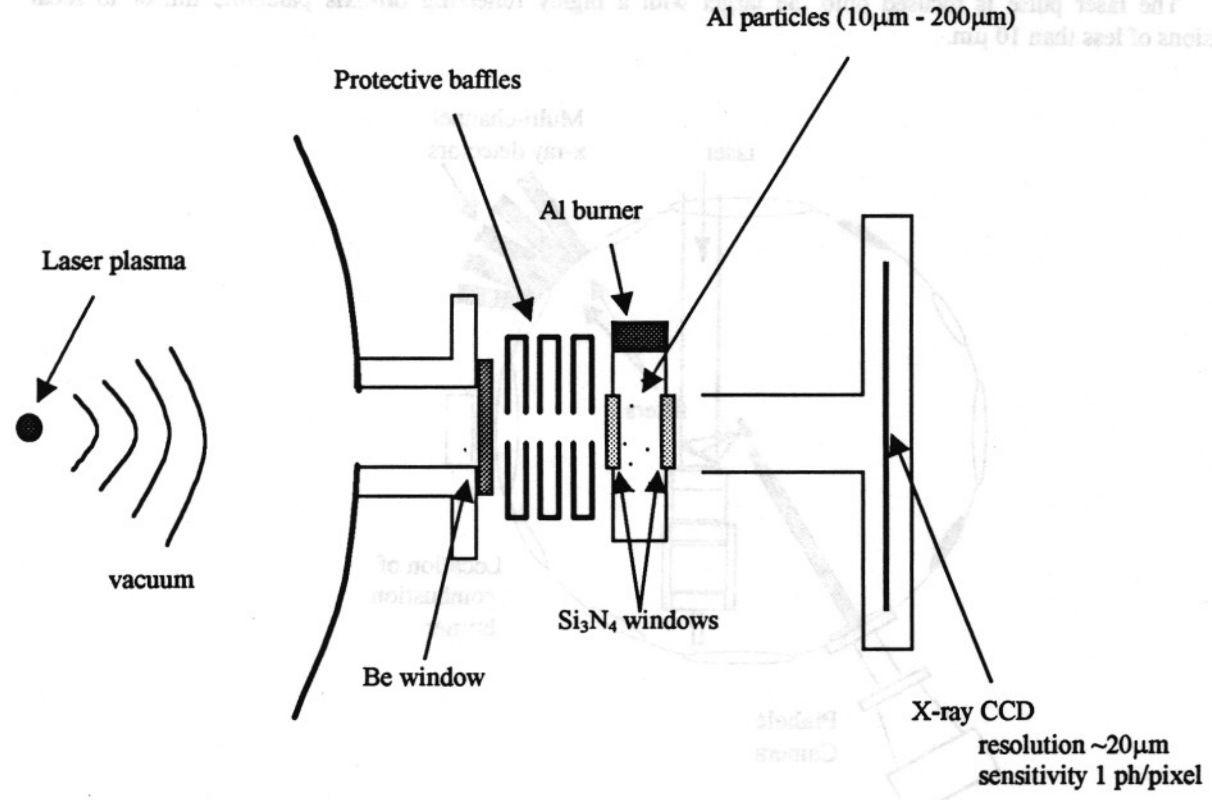

Fig. 4. Initial setup for imaging Al particles with picosecond projection x-ray imaging

The laser system was implemented under modest operating conditions with a bulk aluminum laser plasma target. A measured energy of 200 mJ after the last amplifier provided ~170 mJ on target, following typical reflective losses from windows in the beam path. The aluminum particles were placed on a 7.62 μm thick Be filter situated ~55 cm from the laser plasma. The distance from the Be filter to the CCD plane was ~8.6 cm, producing a modest magnification of ~1.1.

The first results of these tests are shown in Figure 5. As is evident, the aluminum particles were imaged with good contrast (measured to be ~10:1) even in single-shot experiments. In addition, initial image analyses estimate an achievable resolution of less than a single pixel size (20 μm). Improvements in contrast will undoubtedly increase this resolution.

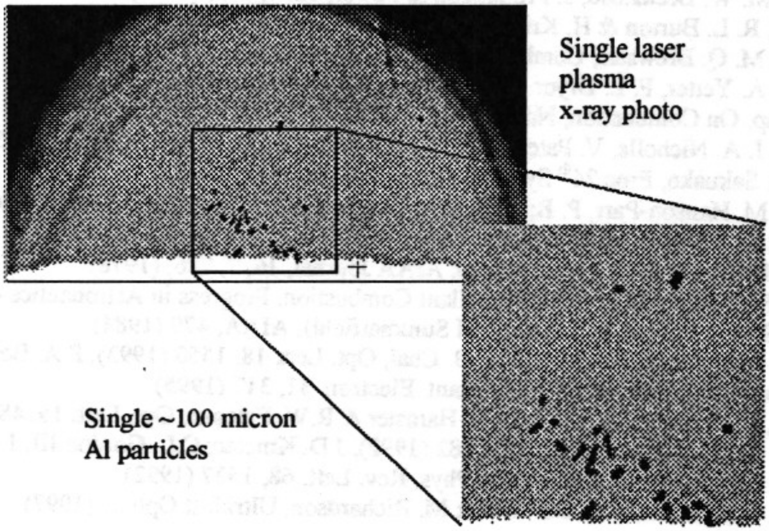

Fig.5. Single shot, picosecond x-ray images of single aluminum particles

FUTURE INVESTIGATIONS

The success of these initial experiments defines a clear level of parameter space in which our approach to x-ray diagnosis of a high temperature combustion burner will provide meaningful results. Together with our colleagues at the University of Illinois, Urbana-Champaign, (H. Krier, Rob Burton and Quinn Brewster), we are now assembling a burner design with which this x-ray radiographic diagnostic can be used to study the particle burn rate by measuring the size of the aluminum particles at various locations in the combustion region. In addition, this technique will allow a detailed analysis of the process of agglomeration.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the support and guidance of Herman Krier in the pursuit of this work. Our group is new to combustion science. The help and support of the UIUC group is warmly recognized, as are discussions with R. Yetter, M. Beckstead, R. Glick and others in the community. This work is part of the UIUC MURI administered by BMDO (contract #N00014-95-1-1339) by L. Caveny and R. Miller of ONR. In particular, Dick Miller recognized the benefits that this new diagnostic can bring to combustion science.

REFERENCES

1. E. W. Price, K. J. Kraeutle, J. L. Prentice, T. L. Boggs, J. E. Crump & D. E. Zurn, " Behavior of aluminum particles in solid propellant combustion" NWC TP 6120 1982
2. E. L Drezin, Combustion & Flame 105, 541, 1996
3. S.E. Olsen & M. W. Beckstead, J. Propulsion & Power, 12, 4, 662, (1996)
4. T. A. Roberts, R. L. Burton & H. Krier, Combustion & Flame, 92, 125, (1993)
5. A. Ishihara & M. Q. Brewster, Combustion Science & Technology, 87, 275, (1992)
6. P. Bucher, R. A. Yetter, F. L. Dryer , T. P. Parr, D. M. Hanson-Parr, & E. P. Vicenci, Proc 26th Symp. On Combustion, Naples Italy, (1996)
7. J. F. Driscoll, J. A. Nicholls, V. Patel, B. K. Shahili & T. C. Liu, AIAA 24, 856, (1986)
8. S. Yuasa & S. Sakuako, Proc 24th Symp. On Combustion, 1871, 1992
9. T. P. Parr, D. M. Hanson-Parr, P. Bucher, R. A. Yetter & F. L. Dryer, 32 JANNAF Meeting (1995)
10. A. Dogariu & M. Richardson (this proceedings)
11. A. Gany, L. H. Caveny & M. Summerfield, AIAA Journal, 16, 7, 736, (1978)
12. E. W. Price, Fundamentals of Solid Propellant Combustion. Progress in Astronautics & Aeronautics Series. 90, (ed. K.K.Kuo & M Summerfield), AIAA, 479 (1984)
13. P. Beaud, M. Richardson, E.J. Miesak & B. Chai, Opt. Lett. 18, 1550 (1993), P.A. Beaud, M. Richardson & E.J. Miesak, IEEE J. Quant. Electron. 31, 317 (1995)
14. S. P. Gordon, T. Donnelly, A. Sullivan H. Hamster & R.W. Falcone, Opt. Lett. 19, 484 (1994)
15. J.D. Kmetec, IEEE J. Quant. Elect. 28, 2382 (1992), J.D. Kmetec, C.L. Gordon III, J.J. Macklin, B.E. Lemoff, G.S. Brown & S.E. Harris, Phys. Rev. Lett. 68, 1527 (1992)
16. S. Grantham, J. Eichenholz, C. DePriest & M. Richardson, Ultrafast Optics, (1997)