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14. ABSTRACT The overall goal in this research proposal was to probe the limits of acoustic cavitation and sonoluminescence to develop a fundamental understanding of the nature of acoustic cavitation at its most extreme. We specifically explored the consequences of the shockwaves internal to the collapsing bubble produced during cavitation. This convergent shockwave generates a high temperature plasma inside the bubble, which is optically opaque and only from whose outer surface may one see emission (i.e., the plasma line emission observed during SBSL in ionic					
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Report Title

Final Report on Probing the Limits of Cavitation, Sonoluminescence, and Mechanoluminescence

ABSTRACT

The overall goal in this research proposal was to probe the limits of acoustic cavitation and sonoluminescence to develop a fundamental understanding of the nature of acoustic cavitation at its most extreme. We specifically explored the consequences of the shockwaves internal to the collapsing bubble produced during cavitation. This convergent shockwave generates a high temperature plasma inside the bubble, which is optically opaque and only from whose outer surface may one see emission (i.e., the plasma line emission observed during SBSL in ionic liquids (e.g., concentrated sulfuric acid). We completed time-resolved SBSL spectroscopic studies and have demonstrated the rise and fall of internal temperature during the formation of a plasma inside a collapsing bubble on a sub-ns timeframe. In addition, we made unsuccessful attempts to find evidence for the formation of X-rays created during cavitation using nanoscale scintillators to down-convert X-rays to visible light emitters.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

"Conditions created during acoustic cavitation" Acoustics 2012, Hong Kong. Invited speaker.

"Time resolved spectroscopy of single bubble sonoluminescence" International Symposium on Nonlinear Acoustics, Tokyo, 2012. Keynote lecturer.

Number of Presentations: 1.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

2011 Fellow, Guggenheim Memorial Foundation.
2010 Fellow, American Chemical Society.
2010 3ème Cycle Lectureship, Switzerland.

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Hangxun Xu	0.50	
Brad Zeiger	0.50	
FTE Equivalent:	1.00	
Total Number:	2	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Darya Radziuk	0.50
FTE Equivalent:	0.50
Total Number:	1

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Kenneth S. Suslick	0.11	
FTE Equivalent:	0.11	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

- The number of undergraduates funded by this agreement who graduated during this period: 0.00
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- The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00
- Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00
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- The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00
- The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 0.00

Names of Personnel receiving masters degrees

<u>NAME</u>
Total Number:

Names of personnel receiving PHDs

<u>NAME</u>
Hangxun Xu
Total Number:
1

Names of other research staff

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
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Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

The conditions formed inside an acoustically-driven, cavitating bubble during collapse are extreme, reaching temperatures on the order of 15,000 K and pressures of over a thousand atmospheres. These conditions have been extensively investigated by spectroscopic observation of light emitted from excited atoms or molecules inside the bubble, most often using a noble gas as an atomic thermometer. Sonochemistry permits access to extreme conditions and consequently the production of novel materials, and knowing the conditions inside the bubble is necessary in order to understand and predict the gas-phase chemistry. Unfortunately, typical experiments divulge only time-averaged information and consequently the actual conditions achieved inside the bubble – the peak temperatures – are largely unexplored. These measurements collect all the light produced over the life of the bubble, using either atomic emission lines or the broad continuum that is characteristic of sonoluminescence emission, to extract information about bubble conditions.

Although there has been some debate over the nature of the continuum emission, there is strong experimental evidence that under typical single-bubble sonoluminescence (SBSL) conditions an optically opaque plasma forms [1–6]. Because the conditions measured for sonoluminescence are not extreme enough to create sufficient ion density to achieve optical opacity (i.e., the mean free path of photons produced inside the plasma is expected to be greater than the bubble radius) it is inferred that the plasma has a relatively cool shell surrounding a much hotter plasma core [6], or Debye shielding reduces the effective ionization potential of gas atoms in the bubble [1,2], or some combination of the two. Driving at very high frequency to reduce the size of the bubble results in Bremsstrahlung-type emission [7], and long-lived bubbles driven at infrasonic frequencies have been reported to transition from a dilute plasma early in the bubble's collapse and an opaque, blackbody emitter during peak conditions and back to a dilute plasma in the final stages of its rebound [2].

There is, however, a recent report that is difficult to reconcile with the usual understanding of sonochemistry as a consequence of compressional heating: time resolved spectra indicate that the bubble heats up, with spectral changes that are entirely inconsistent with the Stefan-Boltzmann law (and consequently incompatible with a blackbody emitter). Nothing approaching temporal symmetry, as one might expect to occur during bubble re-expansion (i.e., spectra at early times being qualitatively similar to spectra at late times), was observed [8].

In this work, sonoluminescence was produced at 21 kHz in a 250 mL Erlenmeyer flask with a quartz window affixed to the bottom by an acid-resistant epoxy (EE-10, ARCOR Epoxy Inc.). The acoustic field was applied via a piezoelectric lead zirconate titanate ceramic bonded to the bottom of the resonator with Loctite Quick Set™ epoxy. The ceramic had a large diameter (1" OD) to permit light collection through its center.

Light emission from SBSL in sulfuric acid is long-lived (typically several nanoseconds at low ultrasonic frequencies) relative to emission from water (as fast as 50 picoseconds [10]) and total light emission in sulfuric acid is hundreds or thousands of times more intense than in water. These long emission times and high intensities make it an attractive medium to study. In this work 85 wt% sulfuric acid solutions were degassed under vacuum and regassed with 50 Torr of noble gas.

Time-resolved spectra were acquired with an Oriel MS-257 spectrograph attached to a Hamamatsu C4334-01 streak camera detector. Fig. 1 diagrams the light collection apparatus. A delay generator is triggered by the response of a photomultiplier tube to a sonoluminescence pulse. A delay corresponding to one acoustic period is created followed a trigger sent to the streak camera for collection. A second delay of roughly one thousand acoustic cycles before the next collection allows the CCD detector to reset and ensures that each frame collected represents only a single bubble event. Spectra were acquired with 300 picosecond resolution.

Because bubbles in sulfuric acid are spatially unstable only a small number (~10%) of frames contained bubble emission, and due to the low intensity of a single emission event each usable frame consisted of only a handful of photons, necessitating the collection of tens of thousands of frames in order to produce a spectrum. These frames were summed about their centers of mass, as is typical for correcting streak camera data for signals with imperfect synchronicity. A more detailed description of this method as applied to SBSL can be found elsewhere [9].

The time evolution of xenon emission is seen in Fig. 2. Emission spectra are fit well by a blackbody distribution and obey the Stefan-Boltzmann law, which governs the relative intensities at different temperatures. While all spectra presented are fit to Planck's law, they do not represent the entire emission event. Fig. 3 compares the temperature evolution derived from the blackbody fits shown in Fig. 2 to the complete wavelength-integrated emission profile. There is insufficient light at the edges of the bubble emission to resolve the spectral shape and we are consequently unable to comment on whether it is reasonable to describe the bubble as a blackbody emitter (and by extension, an opaque plasma) for its entire life. The blackbody fit assumes the plasma emissivity and emission radius are both effectively constant over the resolvable time range.

One criticism of blackbody emission as a possible explanation for the continuum emission in SBSL is that the pulse width of the UV region of the spectrum as measured by time-correlated photon counting is identical [11]. The resolvable emission region offers a small temperature range and a correspondingly modest change in spectral shape. There is no indication from these data that one would expect to see a difference in pulse widths in the two regions probed in [11] due to their extremely low spectral resolution (derived from a single long-pass filter). A dilute plasma during initial heating and final cooling stages as proposed by Putterman [2] with an optically opaque blackbody providing most of the light emission and dominating the time-averaged spectrum provides a coherent and consistent empirical description of the sonoluminescence process. The low thermal conductivity of xenon relative to other noble gases results in long emission times and high temperatures, resulting in the generation of a sufficient number of photons for streak camera collection. Neon and argon were both also tried, but proved too dim. Krypton, however, is a bright enough emitter for time-resolved measurements.

Krypton emission, Fig. 4, is qualitatively the same as xenon emission. In both cases total emission intensity rises and falls smoothly with no dramatic change in spectral shape and a blackbody model of the emission during peak output times still fits

well.

A comparison of Kr and Xe temperature dynamics under similar conditions is shown in Fig. 5. It is not surprising that the xenon emission has a longer duration than the krypton emission. The lower thermal conductivity of the heavier gas means that the bubble will become effectively adiabatic earlier in its collapse and lose adiabaticity later in its rebound. The xenon emission is brighter in aggregate as a result of being longer-lived, but has lower peak temperatures than the krypton bubbles, perhaps as a result of its lower ionization potential consuming much of the available energy [12].

Although peak emission temperatures are hotter than time-averaged temperatures the differences are not stunning. Fig. 6 shows a comparison between peak temperatures and time-averaged temperatures (obtained by summing all time-resolved spectra and fitting to a blackbody). This is consistent with expectations; total emission intensity for a blackbody has a quartic relationship with temperature and therefore emission at times when the bubble is hottest is expected to dominate the time-averaged spectrum.

The insignificant change in bubble conditions over the range measured is consistent with previous observations. Higher pressures could not be probed because bubbles became too unstable to collect with the streak camera.

Summary:

The conditions formed inside an acoustically-driven, cavitating bubble during collapse are extreme, reaching temperatures on the order of 15,000 K and pressures of over a thousand atmospheres. These conditions have been extensively investigated by spectroscopic observation of light emitted from excited atoms or molecules inside the bubble, most often using a noble gas as an atomic thermometer. Sonochemistry permits access to extreme conditions and consequently the production of novel materials, and knowing the conditions inside the bubble is necessary in order to understand and predict the gas-phase chemistry. Unfortunately, typical experiments divulge only time-averaged information and consequently the actual conditions achieved inside the bubble – the peak temperatures – are largely unexplored. The time evolution of xenon emission has now been observed using a streak camera on spectrally resolved emission. The emission spectra are fit well by a blackbody distribution and obey the Stefan-Boltzmann law, which governs the relative intensities at different temperatures. Light emission from SBSL in sulfuric acid is long-lived (typically several nanoseconds at low ultrasonic frequencies) relative to emission from water and total light emission in sulfuric acid is hundreds or thousands of times more intense than in water. These long emission times and high intensities make it an attractive medium to study. In this work 85 wt% sulfuric acid solutions were degassed under vacuum and regassed with 50 Torr of noble gas. The emission profile from SBSL in sulfuric acid under Xe or Kr show a smooth increase in temperature to a maximum of ~14,000 K with a symmetrically smooth decrease over a 6 ns time period.

- [1] S. Khalid, B. Kappus, K. Weninger, and S. Putterman, *Physical Review Letters* 108, 1-5 (2012).
- [2] B. Kappus, S. Khalid, A. Chakravarty, and S. Putterman, *Physical Review Letters* 106, 234302 (2011).
- [3] D. J. Flannigan and K. S. Suslick, *Nature Physics* 6, 598-601 (2010).
- [4] D. J. Flannigan and K. S. Suslick, *The Journal of Physical Chemistry. A* 110, 9315-8 (2006).
- [5] D. Flannigan and K. Suslick, *Physical Review Letters* 95, 1-4 (2005).
- [6] D. J. Flannigan and K. S. Suslick, *Nature* 434, 52-55 (2005).
- [7] C. Camara, S. Putterman, and E. Kirilov, *Physical Review Letters* 92, 1-4 (2004).
- [8] W. Chen, W. Huang, Y. Liang, X. Gao, and W. Cui, *Physical Review E* 78, 1-4 (2008).
- [9] R. Pecha, B. Gompf, G. Nick, Z. Wang, and W. Eisenmenger, *Physical Review Letters* 81, 717-720 (1998).
- [10] M. Moran and D. Sweider, *Physical Review Letters* 80, 4987-4990 (1998).
- [11] B. Gompf, R. Günther, G. Nick, R. Pecha, and W. Eisenmenger, *Physical Review Letters* 79, 1405-1408 (1997).
- [12] C. F. Xavier and R. A. Clemente, *Journal of the Physical Society of Japan* 70, 387-393 (2001).

Technology Transfer

The overall goal in this research proposal was to probe the limits of acoustic cavitation and sonoluminescence to develop a fundamental understanding of the nature of acoustic cavitation at its most extreme. We specifically explored the consequences of the shockwaves internal to the collapsing bubble produced during cavitation. This convergent shockwave generates a high temperature plasma inside the bubble, which is optically opaque and only from whose outer surface may one see emission (i.e., the plasma line emission observed during SBSL in ionic liquids (e.g., concentrated sulfuric acid)). We completed time-resolved SBSL spectroscopic studies and have demonstrated the rise and fall of internal temperature during the formation of a plasma inside a collapsing bubble on a sub-ns timeframe. In addition, we made unsuccessful attempts to find evidence for the formation of X-rays created during cavitation using nanoscale scintillators to down-convert X-rays to visible light emitters.

Time Resolved Spectroscopy of Single Bubble Sonoluminescence

Abstract:

The conditions formed inside an acoustically-driven, cavitating bubble during collapse are extreme, reaching temperatures on the order of 15,000 K and pressures of over a thousand atmospheres. These conditions have been extensively investigated by spectroscopic observation of light emitted from excited atoms or molecules inside the bubble, most often using a noble gas as an atomic thermometer. Sonochemistry permits access to extreme conditions and consequently the production of novel materials, and knowing the conditions inside the bubble is necessary in order to understand and predict the gas-phase chemistry. Unfortunately, typical experiments divulge only time-averaged information and consequently the actual conditions achieved inside the bubble – the peak temperatures – are largely unexplored. The time evolution of xenon emission has now been observed using a streak camera on spectrally resolved emission. The emission spectra are fit well by a blackbody distribution and obey the Stefan-Boltzmann law, which governs the relative intensities at different temperatures. Light emission from SBSL in sulfuric acid is long-lived (typically several nanoseconds at low ultrasonic frequencies) relative to emission from water and total light emission in sulfuric acid is hundreds or thousands of times more intense than in water. These long emission times and high intensities make it an attractive medium to study. In this work 85 wt% sulfuric acid solutions were degassed under vacuum and regassed with 50 Torr of noble gas. The emission profile from SBSL in sulfuric acid under Xe or Kr show a smooth increase in temperature to a maximum of $\sim 14,000$ K with a symmetrically smooth decrease over a 6 ns time period.

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the effective ionization potential of gas atoms in the bubble [1,2], or some combination of the two. Driving at very high frequency to reduce the size of the bubble results in Bremstrahlung-type emission [7], and long-lived bubbles driven at infrasonic frequencies have been reported to transition from a dilute plasma early in the bubble's collapse and an opaque, blackbody emitter during peak conditions and back to a dilute plasma in the final stages of its rebound [2].

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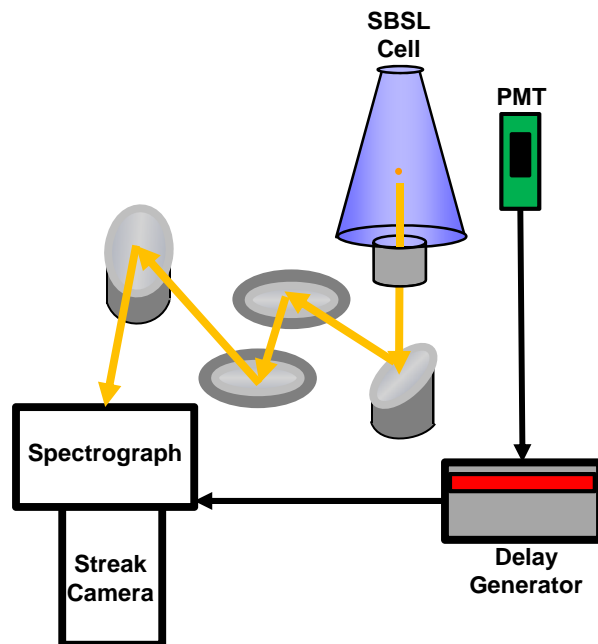


FIG. 1 (color online). Experimental set-up for time-resolved measurements. Bubble is trapped in 250 mL Erlenmeyer flask with 85 wt% sulfuric acid solution regassed with Kr or Xe. Light is collected through a quartz window by a collimating parabolic off-axis mirror, steered to a focusing off-axis mirror, and directed into the spectrograph. The streak camera is triggered by a delay generator which is in turn triggered by the PMT response to a sonoluminescence event.

Because bubbles in sulfuric acid are spatially unstable only a small number ($\sim 10\%$) of frames contained bubble emission, and due to the low intensity of a single emission event each usable frame consisted of only a handful of photons, necessitating the collection of tens of thousands of frames in order to produce a spectrum. These frames were summed about their centers of mass, as is typical for correcting streak camera data for signals with imperfect synchronicity. A more detailed description of this method as applied to SBSL can be found elsewhere [9].

The time evolution of xenon emission is seen in Fig. 2. Emission spectra are fit well by a blackbody distribution and obey the Stefan-Boltzmann law, which governs the relative intensities at different temperatures. While all spectra presented are fit to Planck's law, they do not represent the entire emission event. Fig. 3 compares the temperature evolution derived from the blackbody fits shown in Fig. 2 to the complete wavelength-integrated emission profile. There is insufficient light at the edges of the bubble emission to resolve the spectral shape and we are consequently unable to comment on whether it is reasonable to describe the bubble as a blackbody emitter (and by extension, an opaque plasma) for its entire life. The blackbody fit assumes the plasma emissivity and emission radius are both effectively constant over the resolvable time range.

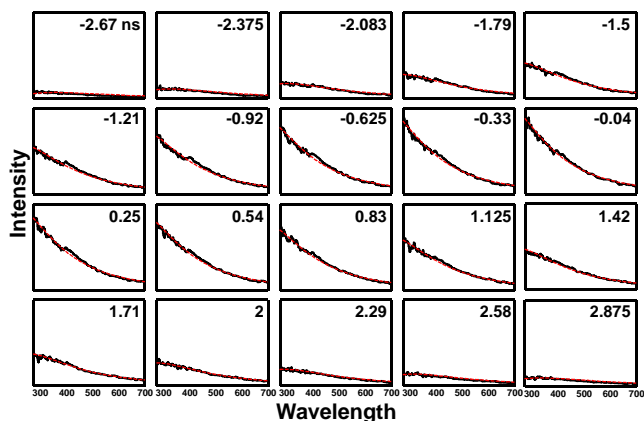


FIG. 2 (color online). Time-resolved emission spectra from single bubble in 85 wt% sulfuric acid regassed with 50 Torr Xe and driven at 1.9 atm. Solid black line is experimental data, dashed red line represents blackbody fit.

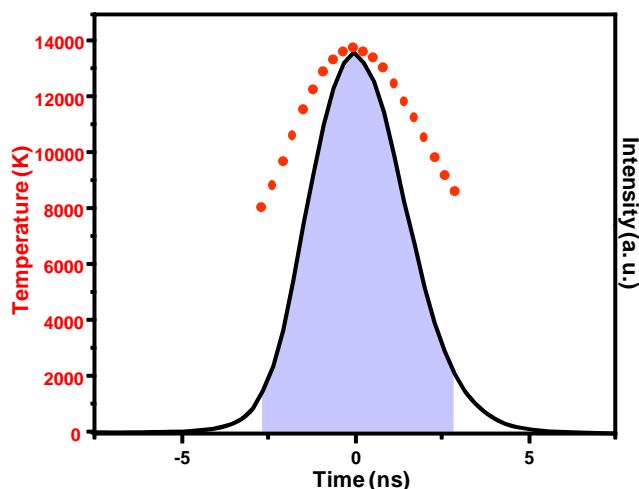


FIG. 3 (color online). Temperatures derived from blackbody fits to time resolved spectra of Xe emission at 1.9 atm (red circles corresponding to left axis) and total light output as a function of time (solid line, right axis). Shaded region indicates time block with sufficient intensity to reliably fit to a blackbody.

One criticism of blackbody emission as a possible explanation for the continuum emission in SBSL is that the pulse width of the UV region of the spectrum as measured by time-correlated photon counting is identical [11]. The resolvable emission region offers a small temperature range and a correspondingly modest change in spectral shape. There is no indication from these data that one would expect to see a difference in pulse widths in the two regions probed in [11] due to their extremely low spectral resolution (derived from a single long-pass filter). A dilute plasma during initial heating and final cooling stages as proposed by Putterman [2] with an optically opaque blackbody providing most of the light emission and dominating the time-averaged spectrum provides a coherent and consistent empirical description of the sonoluminescence process.

The low thermal conductivity of xenon relative to other noble gases results in long emission times and high temperatures, resulting in the generation of a sufficient number of photons for streak camera collection. Neon and argon were both also tried, but proved too dim. Krypton, however, is a bright enough emitter for time-resolved measurements.

Krypton emission, Fig. 4, is qualitatively the same as xenon emission. In both cases total emission intensity rises and falls smoothly with no dramatic change in spectral shape and a blackbody model of the emission during peak output times still fits well.

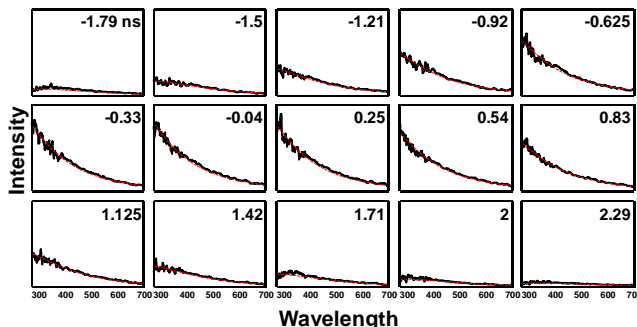


FIG. 4 (color online). Time-resolved emission spectra from single bubble in 85 wt% sulfuric acid regassed with 50 Torr Kr and driven at 2.3 atm. Solid black line is experimental data, dashed red line represents blackbody fit.

A comparison of Kr and Xe temperature dynamics under similar conditions is shown in Fig. 5. It is not surprising that the xenon emission has a longer duration than the krypton emission. The lower thermal conductivity of the heavier gas means that the bubble will become effectively adiabatic earlier in its collapse and lose adiabaticity later in its rebound. The xenon emission is brighter in aggregate as a result of being longer-lived, but has lower peak temperatures than the krypton bubbles, perhaps as a result of its lower ionization potential consuming much of the available energy [12].

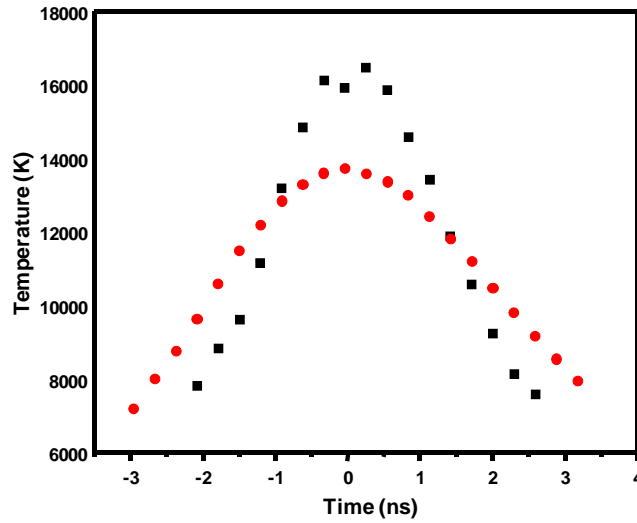


Fig. 5 (color online). Temperature evolution of krypton (black squares) and xenon (red circles) at ~ 2 atm.

Although peak emission temperatures are hotter than time-averaged temperatures the differences are not stunning. Fig. 6 shows a comparison between peak temperatures and time-averaged temperatures (obtained by summing all time-resolved spectra and fitting to a blackbody). This is consistent with expectations; total emission intensity for a blackbody has a quartic relationship with temperature and therefore emission at times when the bubble is hottest is expected to dominate the time-averaged spectrum.

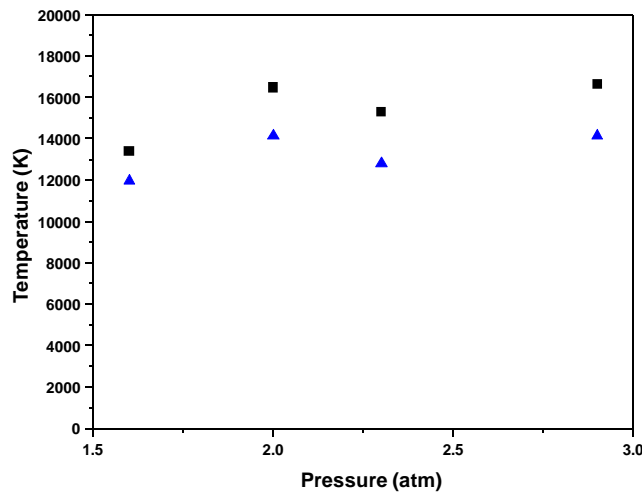


Fig. 6 (color online). Peak (black squares) versus average (blue triangles) temperatures for krypton bubbles under different driving pressures.

The insignificant change in bubble conditions over the range measured is consistent with previous observations. Higher pressures could not be probed because bubbles became too unstable to collect with the streak camera.

Conclusions.

The time evolution of xenon emission has now been observed using a streak camera on spectrally resolved emission. The emission spectra are fit well by a blackbody distribution and obey the Stefan-Boltzmann law, which governs the relative intensities at different temperatures. Light emission from SBSL in sulfuric acid is long-lived (typically several nanoseconds at low ultrasonic frequencies) relative to emission from water and total light emission in sulfuric acid is hundreds or thousands of times more intense than in water. These long emission times and high intensities make it an attractive medium to study. In this work 85 wt% sulfuric acid solutions were degassed under vacuum and regassed with 50 Torr of noble gas. The emission profile from SBSL in sulfuric acid under Xe or Kr show a smooth increase in temperature to a maximum of $\sim 14,000$ K with a symmetrically smooth decrease over a 6 ns time period.

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