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Major Goals: In recent years, there have been concerted efforts to develop high-power diode-pumped alkali vapor lasers (DPAL) for weapons applications. These hybrid gas phase / solid-state laser systems offer possibilities for constructing high-powered lasers with excellent beam quality. However, efforts to scale DPAL's have encountered two technical challenges that stem from the chemically aggressive nature of alkali metal vapors.
The first problem is photo-induced chemical damage of the windows that confine the gain medium. The second is reaction of the alkali metal with the spin-orbit relaxation agent that sustains the population inversion (typically, methane or ethane). This produces particulate matter in the gain medium ("laser snow"), and consumes both the hydrocarbon and the metal.
Rare-gas atoms promoted to metastable electronically excited states have spectroscopic properties that are closely similar to those of the alkali metals. They are readily generated using low-power electrical discharges. The rare gases offer the advantages that they are unreactive, can be used at ambient temperatures, and can provide a range of laser wavelengths that have excellent atmospheric transmission characteristics. Spin-orbit relaxation can be accomplished using rare gas/ rare gas collisions. Consequently, it will be possible to construct closed-cycle lasers that have long-term stability (comparable to that of He/Ne, Ar+ and Kr+ lasers).
Lasing of optically pumped Ar*, Kr* and Xe* has been demonstrated previously by the lead PI. The proposed program is focused on the development of optically pumped rare gas lasers. The primary objectives of the program are:
1. Determinations of the optimal gas mixtures, pressures and discharge conditions for efficient lasing using pulsed excitation.
2. High-pressure discharge development.
3. Measurements of metastable transport, energy transfer and quenching rate constants.
4. CW rare gas laser systems demonstrations at multiple wavelengths.
This sequence of tasks is designed to build the knowledge base that will guide the path to demonstrations of high-powered, diode pumped systems. The information obtained will be of fundamental value, suitable for publication in first-tier scientific journals. These projects will also provide excellent training opportunities for graduate students and post-doctoral fellows.

Accomplishments: The primary achievements have been demonstrations of continuous wave (CW) diode pumped Ar* and Xe* lasers. This progress was enabled through the development of electrical discharges that can sustain high concentrations of metastable excited rare gas atoms (denoted as Rg* in the following, where Rg=Ne, Ar, Kr or Xe) at high pressures. It has been determined that metastable number densities of 1e12 /cm^3 or greater are required for efficient lasing. This density of excited atoms must be sustained in the presence of 0.3 – 1.5 atm of Helium buffer gas. During this program we examined thee different types of discharge for metastable production.

These were high-frequency repetitively pulsed, dielectric barrier and micro-array discharges. In parallel with the experimental program we constructed computational models to guide the laser development effort and evaluate the potential for scaling to high powers.

(i) Repetitive pulsed discharges
A significant advance was achieved using high-frequency repetitively pulsed discharges. Conditions were found where Ar* densities in excess of 1e13 /cm^3 could be continuously maintained in the presence of 1atm He. This was accomplished using 80 ns duration, 1.5 kV pulses at a repetition frequency of 200 kHz. Optical pumping of this discharge, using the output from a tunable diode laser, yielded lasing on the Ar* 912.3 nm line (2p10-1s5) with an output power of 7.9 W and an optical power conversion efficiency of 30%. The efficiency was limited by the poor spatial mode matching between the pump beam and the output laser beam (a conversion efficiency of 55% had been observed in a micro-discharge array system pumped by a Titanium- Sapphire laser at lower powers). The performance characteristics of the Ar* laser were consistent with theoretical models and close to those of a typical DPAL system, indicating that scaling to kilowatt powers and beyond is feasible. For the pulsed discharge, metastable production steadily increased as the discharge voltage was increased. The best laser performance observed was at the limit of voltage and the minimum pulse duration that could be obtained with the available equipment.
Several computational models had been developed to analyze CW systems using steady-state approximations for the discharge excitation, optical pumping and lasing processes. However, our experiments demonstrated that repetitively pulsed discharges have advantages for producing larger volume, high pressure discharges. Consequently, we developed dynamic simulations of a CW laser that uses pulsed-discharge production of Ar metastables. Time-dependent equations were solved for both the discharge and lasing process. Two models were investigated. The first considered the conditions within the lasing medium to be spatially uniform (zero-dimensional model). The second allowed for spatial variations along the lasing axis (one-dimensional model). The models were evaluated by simulating the performance characteristics of an experimentally demonstrated system that provides time-averaged output energies in the range of 3-4 W. Time-dependent species densities, laser power and longitudinal spatial distributions were analyzed.
The group at Emory collaborated with researchers at the US Army Space and Missile Defense Command and the University of Alabama, Huntsville on the development of a diode pumped Xe laser. The pump source was a line-narrowed, temperature-tuned diode laser that operated at wavelengths close to 904.5 nm. In preliminary experiments an output power of 1.3 W was achieved. This experiment was the first demonstration of a CW diode pumped Xe* laser.

(ii) Dielectric barrier discharges
Computational modeling of Rg/He discharge plasmas shows that relatively high ratios of the electric field (E) to the particle number density (N) are needed for efficient Rg* production. Optimal values are in the range of E/N=8-20 V cm^2. This is difficult to achieve using a conventional DC discharge in a gas at 1 atm pressure. However, dielectric barrier discharges (DBD) can often provide high E/N values at high pressures. Consequently, we examined DBD technology for Rg* laser applications. The primary objective was to test the notion that a DBD could be used to generate high concentrations of Ar(1s5) or Xe(1s5) metastables (>1e12 /cm^3) in the presence of 200 – 760 Torr of He. Furthermore, the intention was to establish conditions for continuous or quasi-continuous (modulated) production. Experiments were conducted using Al2O3 coated Al electrodes, driven by a 20 kHz square-wave voltage source. Discharges in Ar:He mixtures yielded time-averaged Ar(1s5) number densities above 1e12 /cm^3 for a discharge gap of 2 mm at a He pressure of 700 Torr. Time resolved data showed that Ar(1s5) production was quite strongly modulated, with most of the excitation occurring during one half-cycle of the voltage wave. The Ar (1s5) number densities were sufficient for the construction of a quasi-continuous optically pumped Ar laser. DBD experiments with Xe:He mixtures also yielded promising results. Time-averaged Xe(1s5) number densities above 2x10^12 /cm^3 were observed over the He pressure range from 200 to 700 Torr. Metastable production near the electrode surfaces was sensitive to the polarity of the electrode, but in the center the modulation of [Xe(1s5)] was not strong. We conclude that the construction of an optically pumped Xe laser that uses a DBD to generate the metastables is feasible. Existing energy transfer rate constant data for Xe(2pi)+He collisions indicate that relatively high He pressures will be needed for efficient laser operation. The rate constants for Xe(2pi)+Ar energy transfer are more favorable, and preliminary kinetic modeling suggests that Xe:Ar:He mixtures may lase more efficiently. Measurements of Xe(1s5) number densities in DBD excited Xe:Ar:He mixtures show that the addition of Ar slightly reduces the production of the Xe metastables, but the densities are still high enough to support lasing.

(iii) Micro-array discharges
Research at Physical Sciences Inc. (PSI) and Tufts University has examined the application of linear-array microwave micro-discharges to generate metastable argon atoms and optically pumped gain in flowing Ar/He gas

mixtures. Ar(1s5) metastables were generated by a micro-discharge array consisting of 15 micro-strip resonators, with the microplasma produced in a small gap between the ends of the strips and the ground plane. Dilute Ar/He gas mixtures were passed along the length of the board and across the discharge gap (25 or 100 μm wide). The micro-discharge was driven by a variable-frequency microwave power train including a 28 VDC supply, a frequency generator, and a broadband 30 W power amplifier. The array board was operated at a frequency near ~900 MHz. Research at PSI demonstrated high gain (>1 cm^-1) and continuous-wave lasing at atmospheric pressure with an optically pumped micro-strip resonator array microplasma, as well as ~10x scaling of the active gain volume at sub-atmospheric pressures using an overlapping dual-array discharge-flow configuration. A detailed series of Ar(1s5) number density measurements and modeling studies, led by collaborators at Tufts University, observed greatly increased metastable number densities at total gas pressures of 100-300 Torr. Subsequent experiments at 300 Torr were conducted at PSI using a Ti:S pump laser, focused to a ~66 μm waist, together with tunable diode laser absorption and gain spectroscopy as well as imaging laser-induced fluorescence spectroscopy. These experiments confirmed discharge-generated Ar(1s5) number densities approaching 1e13 /cm^3 and open-loop optically pumped gain as high as 2.5 cm^-1, more than twice that observed at atmospheric pressure. Diode pumping of a microplasma array discharge laser was demonstrated in a collaborative experiment involving the Emory University and PSI groups.

Training Opportunities: This project has provided excellent training and professional advancement for one post doctoral fellow and three graduate students at Emory University. Mr. Carl Sanderson, a graduate student from the University of Alabama, Huntsville has also benefitted from participation in this research.

Results Dissemination: Publications

J. Han, M. C. Heaven, P. J. Moran, G. A. Pitz, E. M. Guild, C. R. Sanderson, and B. Hokr, "Demonstration of a CW diode-pumped Ar metastable laser operating at 4 W," Optics Letters 42, 4627 (2017)
https://doi.org/10.1364/OL.42.004627

P. A. Mikheyev, J. Han, A. Clark, C. R. Sanderson, and M. C. Heaven, "Production of Ar and Xe metastables in rare gas mixtures in a dielectric barrier discharge," Journal of Physics D: Applied Physics 50, 485203 (2017)
https://doi.org/10.1088/1361-6463/aa91bf

P. A. Mikheyev, J. Han, A. Clark, C. Sanderson, and M. C. Heaven, "Production of Ar metastables in a dielectric barrier discharge," XXI International Symposium on High Power Laser Systems and Applications 10254, 102540X-102540X-102546 (2017)
https://doi.org/10.1117/12.2256172

A. V. Demyanov, I. V. Kochetov, P. A. Mikheyev, V. N. Azyazov and M. C. Heaven, "Kinetic analysis of rare gas metastable production and optically pumped Xe lasers," Journal of Physics D: Applied Physics, 51 045201 (2018)
https://doi.org/10.1088/1361-6463/aa9e40

J. Han, C. R. Sanderson, B. Hokr, C. W. Ballmann, A. B. Clark, M. C. Heaven, "Optically pumped rare gas lasers," XXII International Symposium on High Power Laser Systems and Applications, 1104202 (2019)
https://doi.org/10.1117/12.2522346

P. A. Mikheyev, J. Han, M. C. Heaven, "Lasing in optically pumped Ar:He mixture excited in a dielectric barrier discharge," XXII International Symposium on High Power Laser Systems and Applications, 1104206 (2019)
https://doi.org/10.1117/12.2519438

P. Sun, D. Zuo. P. A. Mikheyev, J. Han, M. C. Heaven, "Time-dependent simulations of a CW pumped, pulsed DC discharge Ar metastable laser system," Optics Express, 27, 22289 (2019)
https://doi.org/10.1364/OE.27.022289

C. R. Sanderson, C. W. Ballman, J. Han, A. B. Clark, B. H. Hokr, K. G. Xu, M. C. Heaven,
"Demonstration of a quasi-CW diode-pumped metastable xenon laser," Optics Express, 27, 36011(2019)
https://doi.org/10.1364/OE.27.036011

A. A. Pershin, A. R. Ghildina, A. M. Mebel, V. N. Azyazov, P. A. Mikheyev, M. C. Heaven,
"Computational investigation of energy transfer and line broadening for Ar*+He collisions"
J. Chem. Phys. 151, 224306 (2019)
https://doi.org/10.1063/1.5133043

Hyunjun Kim and Jeffrey Hopwood, "Scalable Microplasma Array for Argon Metastable Lasing Medium," J. Appl. Phys. 126, 163301 (2019).
https://doi.org/10.1063/1.5119511

A.R. Hoskinson, J. Gregorio, J. Hopwood, K. Galbally-Kinney, S.J. Davis, and W.T. Rawlins, "Argon metastable production in argon-helium microplasmas," J. Appl. Phys. 119, no. 23, p. 233301 (2016).
DOI: 10.1063/1.4954077

A.R. Hoskinson, J. Gregorio, J. Hopwood, K. Galbally-Kinney, S.J. Davis, and W.T. Rawlins, "Spatially-resolved modeling and measurements of metastable argon atoms in argon-helium microplasmas," J. Appl. Phys. 121, 153302 (2017).
DOI: 10.1063/1.4981922

Presentations

W.T. Rawlins, A.R. Hoskinson, K.L. Galbally-Kinney, S.J. Davis, J. Han, and M.C. Heaven, "Diode-Pumped Rare-Gas Microplasma Laser," 20th Annual Directed Energy Symposium, Paper 18-AHPL-005, Oxnard CA, February

2018.

W.T. Rawlins, A.R. Hoskinson, K.L. Galbally-Kinney, S.J. Davis, J.A. Hopwood, J. Han, and M.C. Heaven, “Kinetics of Optically Pumped Argon Metastables in an Ar/He Microplasma,” AFOSR 2018 Molecular Dynamics/Theoretical Chemistry Program Review, poster presentation, May 2018.

M. C. Heaven, J. Han, W. T. Rawlins, S. J. Davis, J. Hopwood, “Optically-Pumped Rare Gas Lasers,” SPIE Conference on Security and Defense, Estrel Congress Center, Berlin, Germany, September 12, 2018

M. C. Heaven, J. Han, W. T. Rawlins, S. J. Davis, J. Hopwood, “Development and Scaling of Diode-Pumped Rare Gas Lasers,” XXII International Symposium on High Power Laser Systems and Applications, Frascati, Italy, October 10, 2018

W.T. Rawlins, K.L. Galbally-Kinney, S.J. Davis, A.R. Hoskinson, and J.A. Hopwood, “Laser excitation dynamics of argon metastables generated in atmospheric pressure flows by microwave frequency microplasma arrays,” Proc. SPIE 9729-10, High Energy/Average Power Lasers and Intense Beam Applications IX, San Francisco CA, 2016.

A.R. Hoskinson, J. Gregorio, J. Hopwood, K. Galbally-Kinney, S.J. Davis, and W.T. Rawlins, “Spatially-resolved modeling and measurements of metastable argon atoms in argon-helium microplasmas,” J. Appl. Phys. 121, 153302 (2017).

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: As an integral part of this program, we have worked with DoD scientists from the US Army Space and Missile Defense Command, University of Alabama (Huntsville), the Air Force Research Laboratory (Kirtland AFB) and the Air Force Institute of Technology.

The research that resulted in the 4W Ar* laser involved Dr. G. Pitz and Mr. P. J. Moran from AFRL, and Dr. B. Hokr and Mr. C. Sanderson from SMDC/UAH. The AFRL group provided the diode laser used in this experiment and made multiple trips to Emory to participate in the measurements. Dr. Hokr and Mr. Sanderson brought their tunable diode laser diagnostic system to Emory, allowing for critical measurements of metastable number densities and transition lineshapes. Our collaborations with Dr. Hokr, Dr. Ballman and Mr. Sanderson also resulted in the successful demonstration of the first optically pumped Xe* laser at UAH.

We are in regular communication with the research group of Prof. G. Perram at the Department of Physics, AFIT. These interactions are mostly focused on computational modeling of Rg* lasers, but Prof. Perram also develops Rg* laser systems in his laboratory. During the current reporting period we loaned laser mirrors to Prof. Perram for laser demonstration experiments.

Physical Sciences Inc. is an industrial partner in this multi-institutional research program. There has been regular communication with the PSI scientists and they have participated in experiments at Emory.

PARTICIPANTS:

Participant Type: PD/PI
Participant: Michael Charles Heaven
Person Months Worked: 3.00
Funding Support:
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International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI
Participant: Jeffrey Hopwood

Person Months Worked: 1.00 **Funding Support:**
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International Collaboration:
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Other Collaborators:

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Project Contribution:
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International Travel:
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National Academy Member: N
Other Collaborators:

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Project Contribution:
International Collaboration:
International Travel:
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Other Collaborators:

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International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

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International Collaboration:
International Travel:
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Project Contribution:
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International Travel:
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Other Collaborators:

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Project Contribution:
International Collaboration:
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National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI
Participant: Wilson Terry Rawlins
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
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Participant Type: Co-Investigator
Participant: Alan R Hoskinson
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Keywords: Optically pumped laser, rare gas metastables
Abstract: In this work, we present the first demonstration of a quasi-continuous-wave diodepumped metastable xenon laser at atmospheric pressures. Lasing in metastable noble gas species has received increased attention in the last few years as a possible high-power laser source. This demonstration shows that metastable xenon has a sufficiently broad absorption spectrum to be pumped with a broad-bandwidth diode laser. This implies that a high-power metastable xenon gas laser should be achievable using high-power pump diodes.
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Authors: Pavel A.Mikheyev, Jiande Han, Michael C.Heaven
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Authors: Michael C. Heaven, Jiande Han, Pengfei Sun, Duluo Zuo, Pavel A. Mikheyev
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Authors: W.T. Rawlins, K.L. Galbally-Kinney, S.J. Davis, A.R. Hoskinson, and J.A. Hopwood
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Nothing to report in the uploaded pdf (see accomplishments)