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University of California - Berkeley
Sponsored Projects Office
2150 Shattuck Avenue, Suite 300
Berkeley, CA 94704 -5940

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RPPR Final Report
as of 06-Jan-2023

Agency Code: 21XD

Proposal Number: 74634PEII **Agreement Number: W911NF-19-1-0017**

INVESTIGATOR(S):

Name: Dan Stamper-Kurn
Email: dmsk@berkeley.edu
Phone Number: 5106429618
Principal: Y

Organization: **University of California - Berkeley**
Address: Sponsored Projects Office, Berkeley, CA 947045940
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Report Date: 04-Dec-2019 Date Received: 05-Jan-2023
Final Report for Period Beginning 05-Dec-2018 and Ending 04-Sep-2019
Title: A metastable beam of titanium atoms for laser cooling
Begin Performance Period: 05-Dec-2018 **End Performance Period:** 04-Sep-2019
Report Term: 0-Other
Submitted By: Dan Stamper-Kurn Email: dmsk@berkeley.edu
Phone: (510) 642-9618
Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 1 **STEM Participants:** 2

Major Goals: The goal of this project was to identify a pathway for applying laser cooling to titanium atoms as a means of producing an ultracold atomic gas of titanium.

Accomplishments: See PDF document.

Training Opportunities: Training was provided to three scientists:

1. Scott Eustice, graduate student
2. Kayleigh Cassella, postdoctocal scholar
3. Andrew Neely, undergraduate researcher

All three received direct training in experimental skills from the PI in the laboratory, and also in broader scientific skills, such as data analysis, interpretation, and scientific communication, through frequent communication and meetings.

Scott Eustice is still in graduate school, continuing to work toward laser cooling atomic titanium. He is expected to complete his PhD within 1-2 years.

Dr. Cassella is now a quantum engineer working at Atom Computing, developing a neutral-atom quantum computer.

Andrew Neely is now a graduate student at Yale University.

Results Dissemination: The work supported by this STIR grant has been disseminated through two peer-reviewed publications:

Scott Eustice, Kayleigh Cassella, and Dan M. Stamper-Kurn, “Laser Cooling of Transition Metal Atoms,” Physical Review A 102, 053327 (2020).

Andrew O. Neely, Kayleigh Cassella, Scott Eustice, and Dan M. Stamper-Kurn. “Isotope Shifts in the Metastable a5F and Excited y5G° Terms of Atomic Titanium,” Physical Review A 103, 032818 (2021).

Our work was also described in numerous oral/poster presentations given by project personnel.

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: Dan Moses Stamper-Kurn
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Kayleigh Cassella
Person Months Worked: 9.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Scott Eustice
Person Months Worked: 9.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Andrew Neely
Person Months Worked: 9.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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Volume: 102 Issue: 5 First Page #: 053327
Date Submitted: 1/5/23 12:00AM Date Published: 11/1/20 7:00AM
Publication Location:
Article Title: Laser cooling of transition-metal atoms
Authors: Scott Eustice, Kayleigh Cassella, Dan Stamper-Kurn
Keywords: Spin-orbit coupling, Synthetic gauge fields, Atoms, Exotic atoms & molecules, Ultracold gases, Polarizability, Atom & ion cooling
Abstract: We propose the application of laser cooling to a number of transition-metal atoms, allowing numerous bosonic and fermionic atomic gases to be cooled to ultralow temperatures. The nonzero electron orbital angular momentum of these atoms implies that strongly atom-state-dependent light-atom interactions occur even for light that is far-detuned from atomic transitions. At the same time, many transition-metal atoms have small magnetic dipole moments in their low-energy states, reducing the rate of dipolar-relaxation collisions. Altogether, these features provide compelling opportunities for future ultracold-atom research. Focusing on the case of atomic titanium, we identify the metastable a 5F5 state as supporting a J to J+1 optical transition with properties similar to the D2 transition of alkali-metal atoms, and suited for laser cooling. The high total angular momentum and electron spin of this state suppresses leakage out of the nearly closed optical transition to a branching ratio esti
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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review A
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Volume: 103 Issue: 3 First Page #: 032818
Date Submitted: 1/5/23 12:00AM Date Published: 3/1/21 8:00AM
Publication Location:
Article Title: Isotope shifts in the metastable
Authors: Andrew O. Neely, Kayleigh Cassella, Scott Eustice, Dan M. Stamper-Kurn
Keywords: Electron correlation calculations for atoms & ions, Electronic structure of atoms & molecules, Electronic transitions, Atoms, Exotic atoms & molecules, Optical spectroscopy
Abstract: We measure and analyze the isotope shifts in the multiplet of transitions between the metastable a5F and excited y5G terms of neutral titanium by probing a titanium vapor in a hollow cathode lamp using saturated absorption spectroscopy. We resolve the five J to J+1 and the four J to J transitions within the multiplet for each of the three I=0 stable isotopes (46, 48, 50 Ti). The isotope shifts on these transitions allow us to determine the isotope-dependent variation in the fine-structure splitting of the a5F and y5G levels themselves. Combined with existing knowledge of the nuclear charge radii of titanium nuclei, we derive the specific mass and field shifts, which arise from correlated electronic motion and electronic density at the nucleus respectively, and further observe a strong J-dependent variation in each. Our results yield insight into the electronic and nuclear structure of transition metal atoms like titanium and also characterize optical transitions that may allow for opti
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Acknowledged Federal Support: **Y**

Partners

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I certify that the information in the report is complete and accurate:
Signature: Dan Stamper-Kurn
Signature Date: 1/5/23   5:20PM

Final report
ARO STIR grant W911NF1910017

Dan Stamper-Kurn, PI

Accomplishments

This STIR grant was absolutely essential in allowing us to kickstart our project to laser-cool transition metal atoms, beginning with atomic titanium.

The granting period was devoted to three major activities:

1) The first step was to flesh out our ideas and present a complete proposal for laser cooling transition-metal atoms. Our team, composed of a young graduate student (Scott Eustice), postdoctoral researcher (Kayleigh Cassella), and the PI (Dan Stamper-Kurn), analyzed the level structures and spectra of several dozen transition-metal atoms, relying on data in the NIST Spectral Database. We identified a pattern that allows for laser cooling on a nearly closed (cycling) dipole-allowed optical transition connecting a lower-energy state with angular momentum J to a high-energy level state with angular momentum $J+1$. These conditions allow this optical transition to be used for standard methods of laser cooling as have been applied successfully to alkali atoms, such as Zeeman slowing, magneto-optical trapping, and polarization-gradient cooling.

The ”Rosetta stone” that revealed this pattern is atomic titanium, which is also our first choice for experimentally realizing laser cooling in transition-metal atoms. In titanium, there exists a metastable low-energy term in which the valence electrons arrange so that the valence s orbital is singly occupied. The maximum J state within this term supports a cycling optical transition which the valence electron is driven as $n\, s_{1/2} \rightarrow n\, p_{3/2}$, with n being the valence principal quantum number. The transition is essentially that of an alkali atom, with similar near-unity oscillator strength and similar 10-MHz-range linewidth. The maximum-J condition of the lower-energy state thins out the atomic spectrum so that there are no dipole-allowed transitions from the excited state other than the laser-cooling transition itself. We estimated that leakage out of the laser cooling transition would be no greater than around10^{-5}. A more refined estimate, based on advanced coupled-channel calculations performed in collaboration with Marianna Safronova at the University of Delaware, confirm and, indeed, improve our estimate, with leakage now being more at the level of 10^{-7}.

Having identified this laser cooling pathway, we found that 10 other elements similarly support laser cooling, either from metastable states or, in one case (Ru), from the ground state. Our findings were presented in a publication [Eustice et al., Physical Review A **102**, 053327 (2020)] that was highlighted as an Editor’s Suggestion.

2) We built up much of the experimental hardware needed for laser cooling atomic titanium. This hardware includes a frequency-doubled titanium-sapphire laser, operating at a fundamental wavelength of 996 nm that, then, when doubled, produces several 100 mW of light at the 489 nm wavelength of the laser cooling transition. We have also built up laser sources to drive several other transitions in titanium, including transitions out of the $a\ ^3F$ ground term (wavelength 398 nm for imaging) and out of low-J states of the metastable $a\ ^5F$ term (wavelength 451 nm for optical pumping). We built several vacuum chambers in which to operate a crucible-based atomic titanium source, perform Zeeman

slowing and magneto-optical trapping, and, later, to study atomic ablation into a buffer-gas source and the operation of a direct titanium sublimation source.

Our experimental work hit some snags. The biggest snag was the failure to produce a titanium atomic beam by use of a high-temperature crucible oven. We initially thought this approach to be the most promising, easiest, and most straightforward: all we needed to do was purchase a commercial high-temperature crucible and then operate it above the sublimation temperature (around 1500 C) of titanium. Unfortunately, this approach failed. We found that, while the oven did produce a titanium atomic beam initially, this beam was extinguished within just a few hours. We eventually determined that this failure was caused by the formation of a thin impermeable crust enveloping the titanium metal within the crucible. Titanium is chemically reactive, particularly at high temperature. Further, at high temperature, various components within the crucible begin either to outgas or simply to leech through the titanium. As a result, exposed high-temperature titanium soon becomes encrusted by a titanium nitride or titanium carbide surface; nitrogen comes from disintegration of electrical insulators within the crucible heater at high temperature, and carbon comes from the graphite crucible itself.

As a result, we have begun exploring other ways of producing atomic beams of titanium. One approach involves laser-ablation of titanium into a room-temperature argon buffer gas. This approach works, and, as a significant side benefit, directly produces a titanium beam that is rich in the metastable-state atoms needed for laser cooling. A second approach involves operating a commercial titanium sublimation pump (easy!). We have confirmed, by laser spectroscopy, that a sublimation pump produces a steady beam of atomic titanium. This beam, sublimating at a temperature around 1600 C, contains only a trace population of atoms in the metastable laser-cooling state, whose energy above the ground state corresponds to an equivalent temperature of 9000 C. We are currently exploring pathways for optically pumping this ground-state population in the laser cooling state.

3) We also began exploring the spectrum of titanium by performing spectroscopy on titanium within a hollow-cathode lamp. The aim here was to identify the laser-cooling transition, at the wavelength of 489 nm, and also to measure isotope shifts so that we can apply laser cooling selectively to any of the five stable isotopes of titanium. This effort was successful – we soon observed strong optical resonances for the nuclear-spin-zero isotopes of titanium (^{46,48,50}Ti), and some signal of the hyperfine-split resonances of the half-integer nuclear-spin isotopes (^{47,49}Ti).

We then undertook to explore the isotope shifts of titanium in greater detail. Our frequency-doubled titanium:sapphire laser allowed us to scan through several resonance lines. Among these is a multiplet of lines connecting the different fine-structure states of the lower-energy ($a\ ^5F$) and higher-energy ($y\ ^5G$) terms connected by the laser-cooling transition. Analyzing these isotope shifts, we produced what we consider to be the cleanest data revealing isotopic shifts in fine structure. One byproduct of these measurements is a clean measurement of the fine-structure variations in the electronic density at the nucleus for titanium atoms. Such measurements provide a stringent test for advanced coupled-cluster calculations of the titanium atom's electronic structure, such as are being performed by Marianna Safronova and her team at the University of Delaware.

These isotope-shift measurements were performed largely by a young undergraduate student, Andrew Neely, together with the rest of our team (Scott Eustice, Kayleigh Cassella and Dan Stamper-Kurn). The work is reported in a recent publication [Neely et al., Physical Review A **103**, 032818 (2021)].