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Towards Ultrasensitive Isotope Trace Analysis of ^{41}Ca

I.D. Moore[a], K. Bailey[a], Z.-T. Lu[a,*], P. Müller[a], T.P. O'Connor[a], L. Young[b]

[a] *Physics Division, Argonne National Laboratory, Argonne, IL 60439, USA*

[b] *Chemistry Division, Argonne National Laboratory, Argonne, IL 60439, USA*

Abstract

An Atom Trap Trace Analysis (ATTA) system based on the technique of laser manipulation of neutral atoms is being developed to count individual ^{41}Ca atoms present in natural samples with an isotopic abundance of 10^{-15}. Trapping of all stable calcium isotopes has been demonstrated and single-atom counting has been realized. For the most abundant isotope, ^{40}Ca (97% isotopic abundance), a magneto-optical trap (MOT) loading rate of 2×10^{10} atoms/s has been reached at the overall capture efficiency of 1×10^{-4}. System improvements could increase the efficiency by at least an order of magnitude.

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*Corresponding author. URL: www-mep.phy.anl.gov/atta/; fax: +1-630-252-3903

E-mail address: lu@anl.gov

1. Introduction

Atom Trap Trace Analysis (ATTA) developed by our group has been used to successfully count individual ^{85}Kr and ^{81}Kr atoms present in a natural krypton gas sample with isotopic abundances in the range of 10^{-11} and 6×10^{-13}, respectively [1]. Advantages of ATTA compared to other laser-based techniques have been discussed elsewhere [2]. The ability of ATTA to tolerate impure samples with the atom counts free from contamination from other isotopes, elements, or molecules promises to enhance the capabilities and expand the applications of ultrasensitive isotope trace analysis.

Trace analysis of ^{41}Ca has promising applications in both radiocalcium dating and in the biomedical field. With a half-life of 1.03×10^5 years, ^{41}Ca could be used to date bones ranging from 50 thousand to 1 million years old [3, 4]. This period is important in the study of early human development, and is beyond the reach of ^{14}C-dating. Early accelerator mass spectrometry (AMS) work indicated that ^{41}Ca/Ca in different bone samples vary in the range of 10^{-15}-10^{-14} [5]. If ATTA can achieve a detection sensitivity of 1×10^{-16} or lower, then a dedicated setup can be used to investigate in detail the feasibility of ^{41}Ca-dating.

One particularly interesting proposal that is currently under investigation with AMS and resonance ionization mass spectrometry (RIMS) is to use ^{41}Ca-tracing to diagnose osteoporosis, a disease commonly found in women after menopause [6, 7, 8]. Measuring the bone loss rate directly could be more sensitive than the current method of monitoring bone density with X-ray imaging. After a subject ingests a "^{41}Ca pill," urine samples containing ^{41}Ca with an isotopic abundance in the range of 10^{-13}-10^{-9} can then be measured on a regular basis as

a monitor of the bone metabolism. ATTA will be a complimentary method to these existing techniques, with an advantage over AMS due to a significant reduction of cost (greater than a factor of 10).

2. Experimental techniques

Our design (Fig. 1) consists of a well-collimated atomic beam, a 40 cm - long Zeeman slower and a magneto-optical trap (MOT) chamber. A metallic calcium sample is heated to 400°C to form a thermal atomic beam. The atoms are decelerated with the Zeeman slowing technique [9], and then loaded into the MOT [10]. This MOT operates on the $4\,^1S_0 \rightarrow 4\,^1P_1$ trapping transition (Fig. 2) at 422.7 nm (natural linewidth, $\Gamma_{nat}/2\pi = 34.6$ MHz). The appropriate laser light at 423 nm is produced by a frequency-doubled Ti:Sapphire ring laser, resulting in about 100 mW of 423 nm radiation. Active frequency stabilization is provided by fringe-offset-locking, relative to a commercial frequency-stabilized He-Ne laser [11].

In the trap, the vacuum is maintained at 1×10^{-8} Torr. A single atom scatters resonant photons at a rate of $5 \times 10^7 s^{-1}$, of which 2% are collected, and focussed onto a low-gain photodiode detector. The trap lifetime is measured by observing the fluorescence decay after the slowing beam is turned off. Atoms remain trapped for an average of 18 ms, a relatively short lifetime due to the fact that a weak decay channel exists for excited atoms in the $4\,^1P_1$ state to the $3\,^1D_2$ state (Fig. 2). By repumping the $3\,^1D_2$ atoms to the $5\,^1P_1$ state using a diode laser operating at a wavelength of 671.95 nm, the trap lifetime is lengthened by factors of between three and six, agreeing well with previous observations [12, 13]. In order to perform isotope shift measurements on the repumping line, the frequency of the trapping

light is locked to a selected isotope and the diode laser is scanned over the 672 nm transition.

This trap system can capture the abundant ^{40}Ca (isotopic abundance 96.9%) atoms at a rate of 2×10^{10} atoms/s and with a capture efficiency of 1×10^{-4}. With a 0.3mg sample of calcium and a 5% precision requirement for biomedical applications (isotopic abundance of ^{41}Ca/Ca $\sim 10^{-12}$) this trap system can count 400 atoms in six hours. To attain a 20% precision for ^{41}Ca-dating (isotopic abundance of ^{41}Ca/Ca $\sim 10^{-15}$) the trap could capture 25 atoms in 15 days, with a 20mg sample. Further improvement of our system by 1-2 orders of magnitude is realistic.

3. Results and discussion

We have trapped and measured the MOT lifetime of all stable calcium isotopes. Typical values obtained are 18 ms without repumping and 80ms with repumping. By assuming that the maximum loading rate is achieved for all isotopes with the same frequency detuning from resonance (-120 MHz), measurements of the trap fluorescence agree with the expected even isotopic abundance (Fig. 3) to an accuracy of 15%. Work is still in progress to improve this result in order to ensure a higher probability of detecting the fluorescence signal from a single ^{41}Ca atom.

An important step towards our goal of realizing ultrasensitive isotope trace analysis of ^{41}Ca is to determine the isotope shift of the $3\,^1D_2 \rightarrow 5\,^1P_1$ repumping transition between ^{40}Ca and ^{41}Ca. Preliminary isotope shift measurements have been made for all stable Ca isotopes. Further measurements under different trap conditions are necessary to improve our understanding of the trap systematics. Both the number density of atoms within the trap

and a variation in the magnetic field gradient over the trapped atom volume may attribute to systematic uncertainties in the isotope shift. Final results will be published elsewhere.

In order to analyze the rare isotope, ^{41}Ca, the ATTA system must be able to count single atoms. In 2001, we realized the single-atom-detection capability (Fig. 4) with this setup. The observed photon count rate from a single ^{40}Ca atom is 4 kHz, while the background rate is 5 kHz. With a trap lifetime of 100 ms, the signal-to-noise ratio of a single trapped atom is 18.

4. Outlook

An order-of-magnitude increase in the system efficiency is expected with the introduction of two-dimensional transverse cooling to reduce the atomic beam divergence and amplify the atom flux in the forward direction. Improvements in the single-atom detection capability will increase the signal-to-noise ratio, by reducing scattered background light from the trapping laser beams and increasing the transmission efficiency of the lens system. New oven designs are presently being developed to enable both medical samples and geological samples to be tested.

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References

[1] C.Y. Chen, Y.M. Li, K. Bailey, T.P. O'Connor, L. Young, Z.-T. Lu, Science 286 (1999) 1139.

[2] K. Bailey, C.Y. Chen, X. Du, Y.M. Li, Z.-T. Lu, T.P. O'Connor, L. Young, Nucl. Instr. and Meth. B 172 (2000) 224.

[3] W. Henning, W.A. Bell, P.J. Billquist, B.G. Glagola, W. Kutschera, Z. Liu, H.F. Lucas, M. Paul, K.E. Rehm, J.L. Yntema, Science 236 (1987)725.

[4] R.E. Taylor, Anal. Chem. 59 (1987) 317A.

[5] D. Fink, J. Klein, R. Middleton, Nucl. Instr. and Meth. B 52 (1990) 572.

[6] D. Elmore, M.H. Bhattacharyya, N. Sacco-Gibson, D.P. Peterson, Nucl. Instr. and Meth. B 52 (1990) 531.

[7] S.P.H.T. Freeman, J.C. King, N.E. Vieira, L.R. Woodhouse, A.L. Yergey, Nucl. Instr. and Meth. B 123 (1997) 266.

[8] P. Müller, B.A. Bushaw, K. Blaum, S. Diel, Ch. Geppert, A. Nähler, N. Trautmann, W. Nörterhäuser, K. Wendt, Fres. J. Anal. Chem. 370(5) (2001) 508.

[9] W.D. Phillips, H. Metcalf, Phys. Rev. Lett. 48 (1982) 596.

[10] E.L. Raab, M. Prentiss, A. Cable, S. Chu, D.E. Pritchard, Phys. Rev. Lett. 59 (1987) 2631.

[11] W.Z. Zhao, J.E. Simsarian, L.A. Orozco, G.D. Sprouse, Rev. Sci. Instrum. 69 (1998) 3737.

[12] C.W. Oates, F. Bondu, R.W. Fox, L. Hollberg, Eur. Phys. J. D 7 (1999) 449.

[13] J. Grünert, A. Hemmerich, Appl. Phys. B 73 (2001) 815.

Fig. 1. Schematic layout of the atom beamline. Total length of the apparatus is about 1.5 m. Transverse cooling will be installed later this year.

Fig. 2. Relevant energy levels of ^{40}Ca including the trapping transition at 422.7 nm and the repumping transition at 671.95 nm. The dashed line indicates atoms lost to a metastable state.

Fig. 3. Fluorescence of trapped stable calcium isotopes versus laser frequency. The small peak observed approximately 100 MHz to the right of ^{40}Ca is the beam fluorescence from this isotope. Fluorescence was measured with a low-gain photodiode detector.

Fig. 4. Signal of a single trapped ^{40}Ca atom. The signal-to-noise ratio is ~ 20.

Figure 2

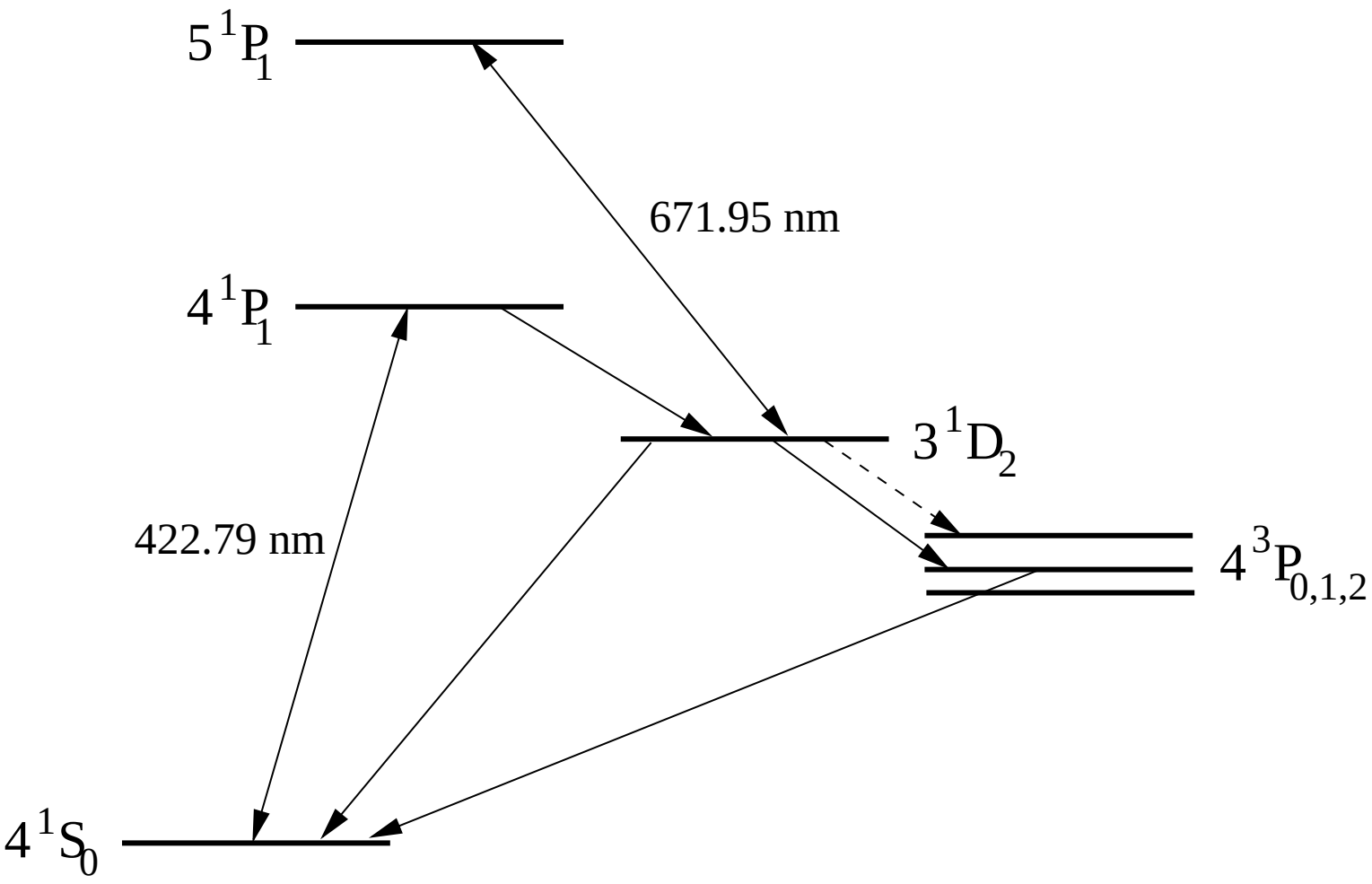


Figure 3

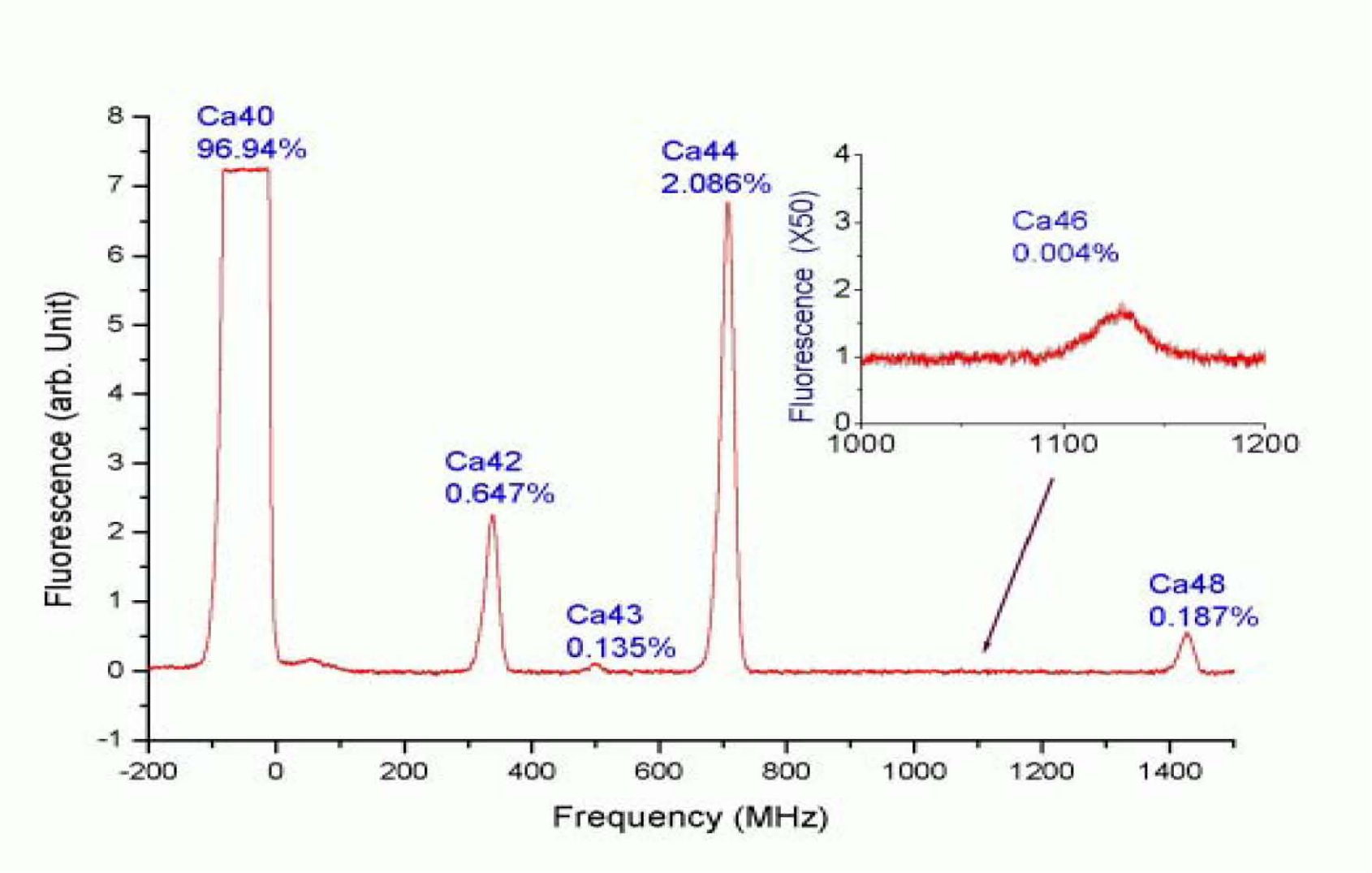
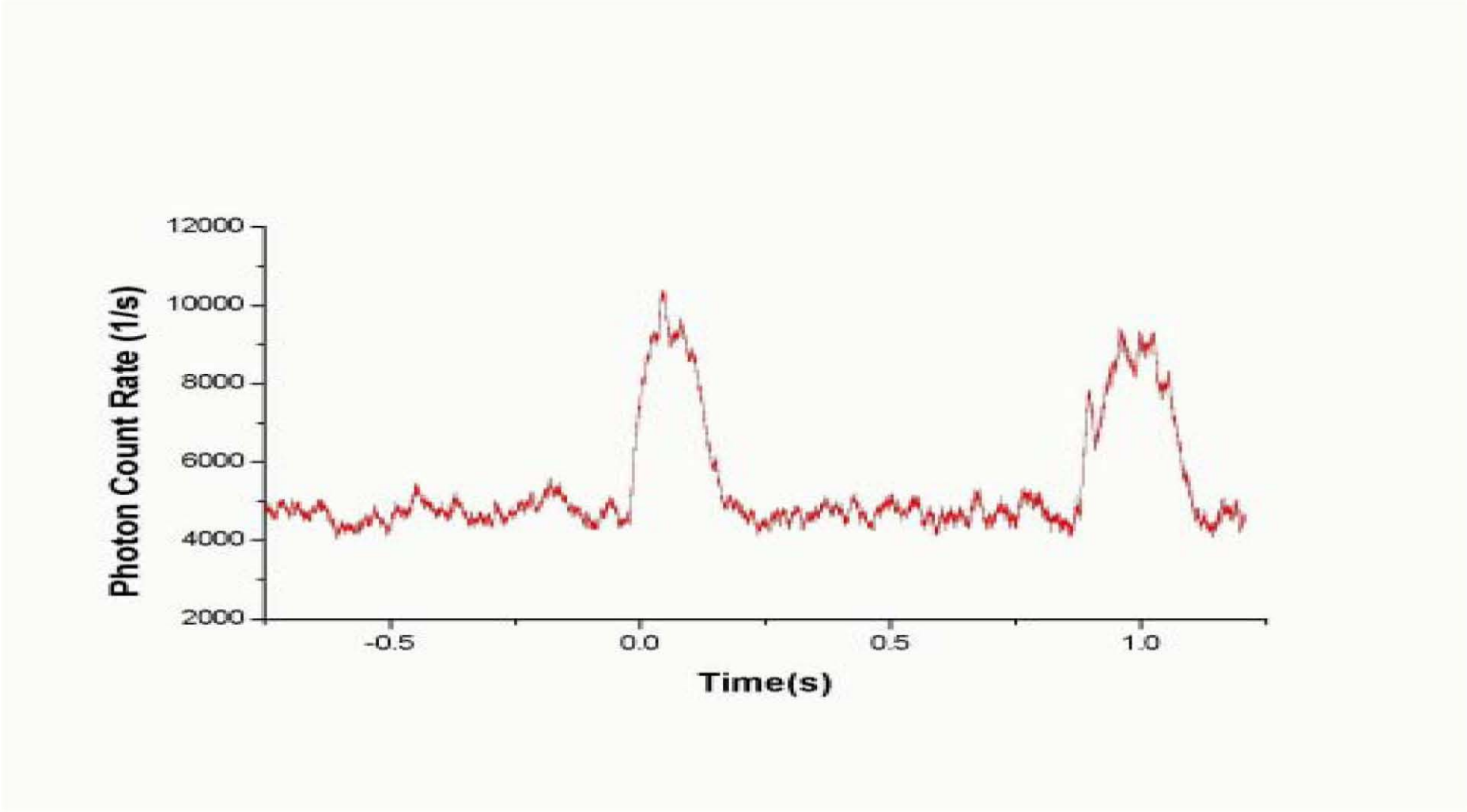

Ca40
96.94%
Ca42
0.647%
Ca43
0.135%
Ca44
2.086%
Ca46
0.004%
Ca48
0.187%
Fluorescence (arb. Unit)
Frequency (MHz)
Fluorescence (X50)

Figure 4


Photon Count Rate (1/s)
12000
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8000
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4000
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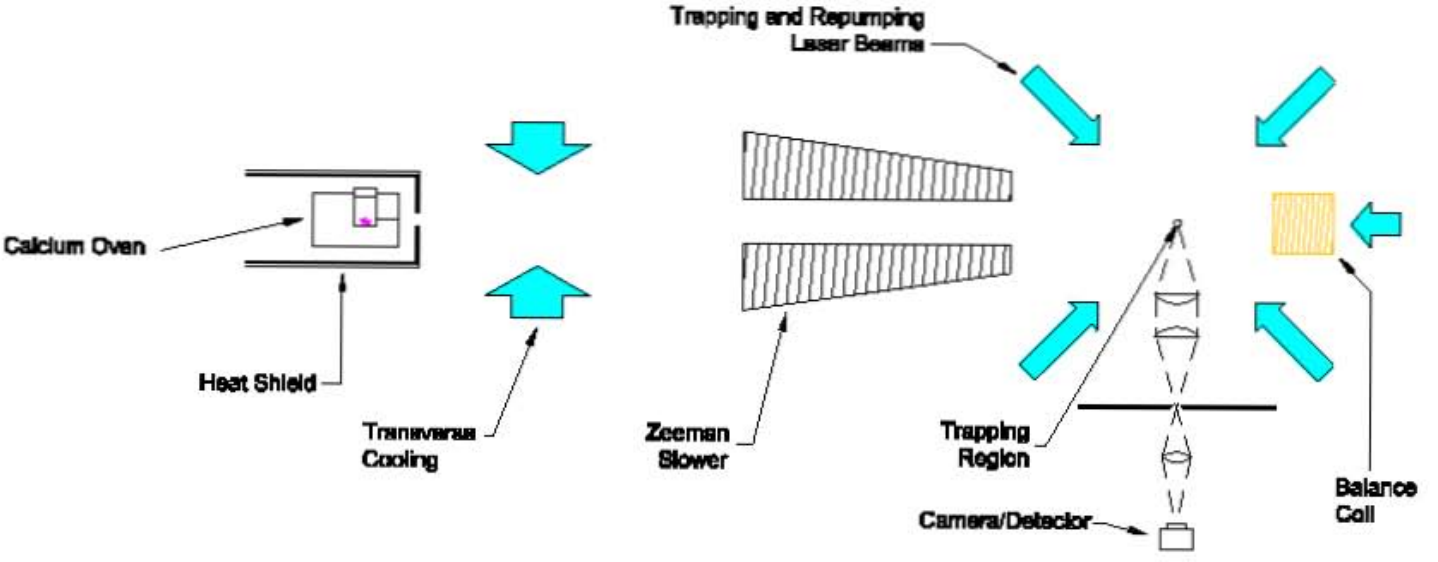
-0.5
0.0
0.5
1.0
Time(s)


Trapping and Repumping
Laser Beams
Calcium Oven
Heat Shield
Transverse
Cooling
Zeeman
Slower
Trapping
Region
Camera/Detector
Balance
Coil