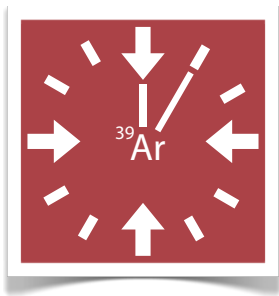


# Rare Noble Gas Isotopes: Dating Ice and Water



Sven Ebser

Kirchhoff-Institute for Physics, Group Oberthaler  
Heidelberg University

May 27, 2016



Werner  
Aeschbach-Hertig



Stefan  
Beyersdorfer



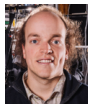
Emeline  
Mathouchanh



Arne  
Kersting



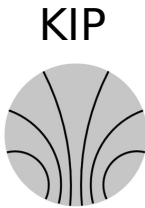
Florian  
Ritterbusch  
USTC



Sven  
Ebser



Lisa  
Ringena



Landreas  
Kamrad



Markus K.  
Oberthaler



Zhongyi  
Feng

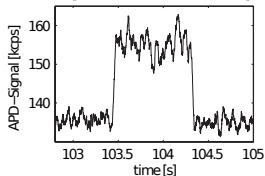
Water sampling



Argon extraction



Single  $^{39}\text{Ar}$  atom counting



# Rare Noble Gas Isotopes

The chemical symbol 'He' is displayed in a glowing orange-red neon font against a black background.

Studying exotic nuclear structure, testing QED

${}^6\text{He}$  and  ${}^8\text{He}$  or  ${}^3\text{He}$  and  ${}^4\text{He}$

The chemical symbol 'Ne' is displayed in a glowing red-orange neon font against a black background.

Searching for physics beyond the standard model

Rare Neon isotopes

The chemical symbol 'Ar' is displayed in a glowing purple neon font against a black background.

Dating of ice and water

${}^{39}\text{Ar}$

The chemical symbol 'Kr' is displayed in a glowing yellow-orange neon font against a black background.

Monitoring of the Nuclear Non-Proliferation Treaty

${}^{85}\text{Kr}$

The chemical symbol 'Xe' is displayed in a glowing blue neon font against a black background.

Krypton contamination in Xenon Dark Matter detectors

Kr contamination in Xe

# Rare Noble Gas Isotopes

He

Studying exotic nuclear structure, testing QED

${}^6\text{He}$  and  ${}^8\text{He}$  or  ${}^3\text{He}$  and  ${}^4\text{He}$

Ne

Searching for physics beyond the standard model

Rare Neon isotopes

Ar

Dating of ice and water

${}^{39}\text{Ar}$

Kr

Monitoring of the Nuclear Non-Proliferation Treaty

${}^{85}\text{Kr}$

Xe

Krypton contamination in Xenon Dark Matter detectors

Kr contamination in Xe



## Groundwater

- Water resource management
- Climate reconstruction

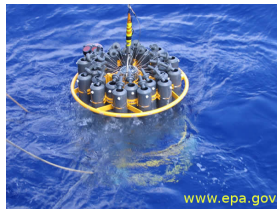


## Groundwater

- Water resource management
- Climate reconstruction

## Ocean

- Circulations (Global Conveyor Belt, North Atlantic Current, ...)
- Mixing processes (CO<sub>2</sub> uptake by oceans,...)



# Dating Water and Ice

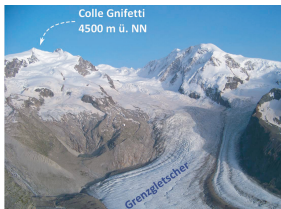
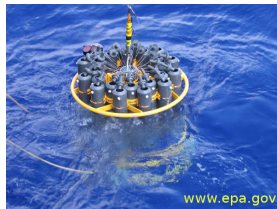


## Groundwater

- Water resource management
- Climate reconstruction

## Ocean

- Circulations (Global Conveyor Belt, North Atlantic Current, ...)
- Mixing processes (CO<sub>2</sub> uptake by oceans,...)



## Ice

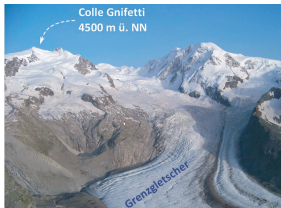
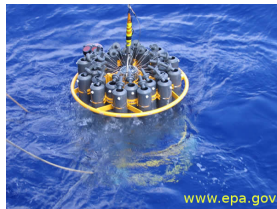
- Climate reconstruction
- Glaciological studies

decreasing sample size



## Ocean

- Circulations (Global Conveyor Belt, North Atlantic Current, ...)
- Mixing processes (CO<sub>2</sub> uptake by oceans,...)



## Groundwater

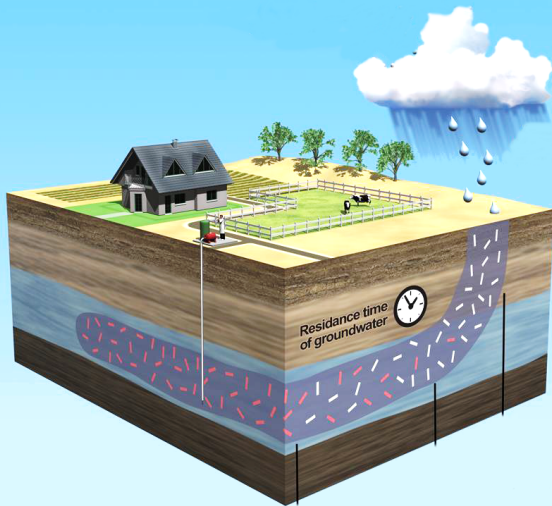
- Water resource management
- Climate reconstruction

## Ice

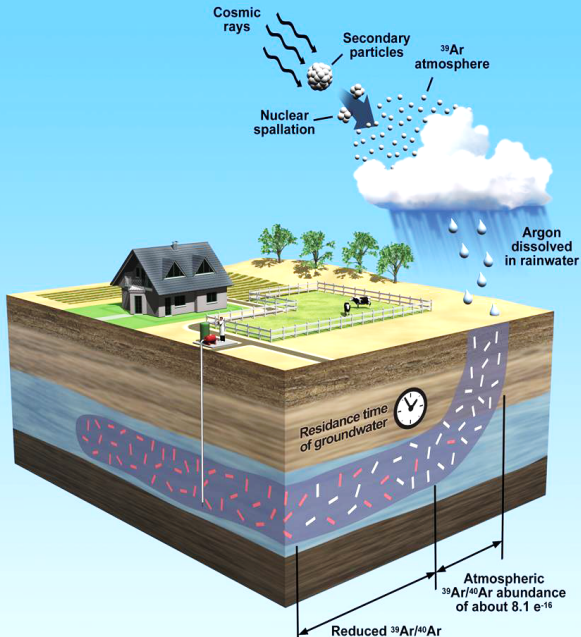
- Climate reconstruction
- Glaciological studies



# Radioisotope Dating

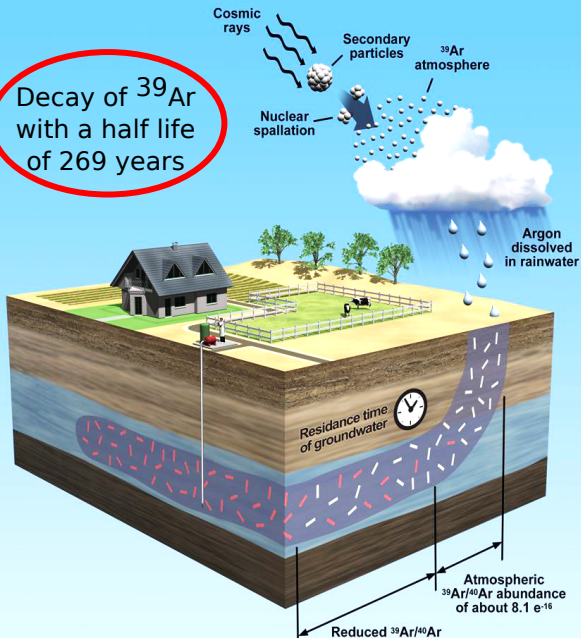


# Radioisotope Dating

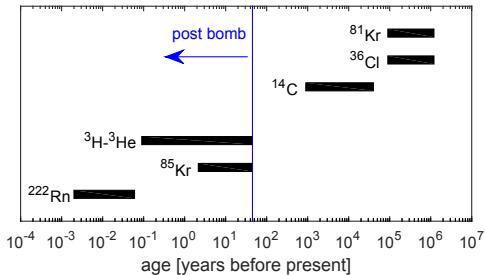


# Radioisotope Dating

Decay of  $^{39}\text{Ar}$   
with a half life  
of 269 years

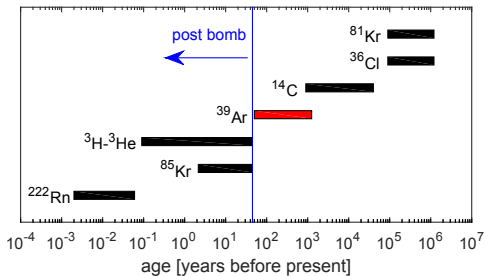


# Radioisotope Dating



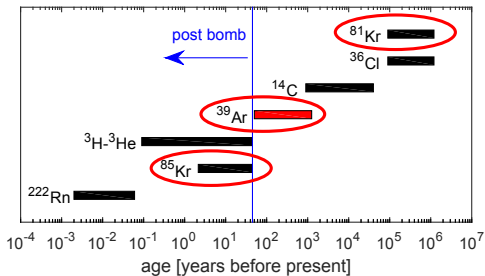
isotope	half life
$^{222}\text{Rn}$	3.824 d
$^3\text{H}$	12.32 a
$^{85}\text{Kr}$	10.76 a
$^{14}\text{C}$	5730 a
$^{81}\text{Kr}$	232 ka
$^{36}\text{Cl}$	301 ka

# Radioisotope Dating



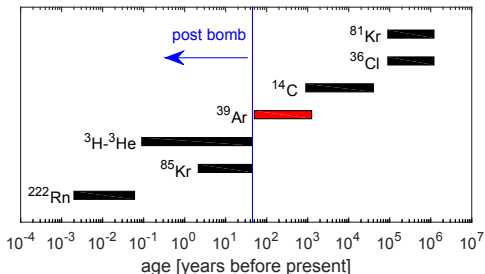
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$^{36}\text{Cl}$	301 ka
$^{39}\text{Ar}$	269 a

# Radioisotope Dating



- Conservative tracer  
not involved in chemical  
processes

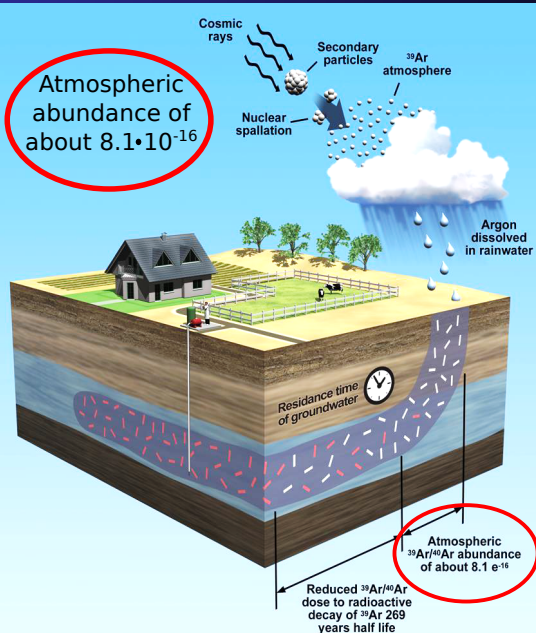
# Radioisotope Dating



isotope	half life
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$^{36}\text{Cl}$	301 ka
$^{39}\text{Ar}$	269 a

- Conservative tracer  
not involved in chemical processes
- Anthropogenic contribution  
< 5 %
- Variations in atmospheric  
 $^{39}\text{Ar}$  concentration during  
the last 1000 years < 7 %

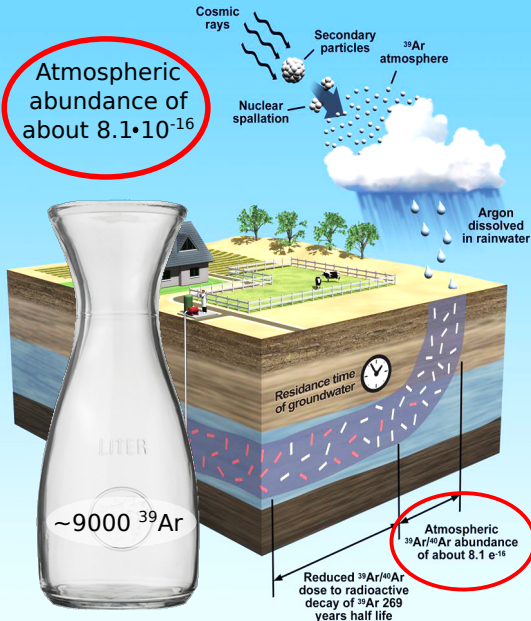
# Radioisotope Dating





# Radioisotope Dating

Atmospheric abundance of about  $8.1 \cdot 10^{-16}$



$^{39}\text{Ar} : ^{40}\text{Ar} = 1 : 1\,000\,000\,000\,000\,000$

Public debt of the USA in US-cents

$^{39}\text{Ar} : ^{40}\text{Ar} = 1 : 1\,000\,000\,000\,000\,000$

Public debt of the USA in US-cents



$^{39}\text{Ar} : ^{40}\text{Ar} = 4000$  times to the moon and there is one cent different!

- **Radioactivity:**

Low-Level-Counting (LLC):  $^{39}\text{Ar} \rightarrow ^{39}\text{K} + e^- + \bar{\nu}_e$   
Only large samples (1-3 tons of water)

- **Mass:**

Accelerator Mass Spectrometry (AMS):  
Possible, but difficult for noble gases

- **Atomic spectrum:**

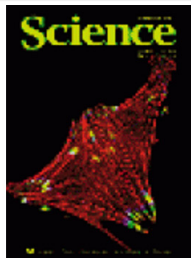
Atom Trap Trace Analysis (ATTA)  
Complement for rare and long-lived noble gases

## Ultrasensitive Isotope Trace Analyses with a Magneto-Optical Trap

C. Y. Chen,<sup>1</sup> Y. M. Li,<sup>1</sup> K. Bailey,<sup>1</sup> T. P. O'Connor,<sup>1</sup> L. Young,<sup>2</sup>

Z.-T. Lu<sup>1\*</sup>

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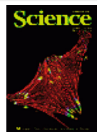


	<sup>85</sup> Kr	<sup>81</sup> Kr	<sup>39</sup> Ar
half life	10.76 a	232 ka	269 a
relative abundance	$2 \cdot 10^{-11}$	$6 \cdot 10^{-13}$	$8 \cdot 10^{-16}$

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Z.-T. Lu<sup>1\*</sup>

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## One million year old groundwater in the Sahara revealed by krypton-81 and chlorine-36

N. C. Sturchio,<sup>1</sup> X. Du,<sup>2,3</sup> R. Purtschert,<sup>4</sup> B. E. Lehmann,<sup>4</sup> M. Sultan,<sup>5</sup> L. J. Patterson,<sup>1</sup>  
Z.-T. Lu,<sup>2</sup> P. Müller,<sup>2</sup> T. Bigler,<sup>4</sup> K. Bailey,<sup>2</sup> T. P. O'Connor,<sup>2</sup> L. Young,<sup>6</sup> R. Lorenzo,<sup>4</sup>  
R. Becker,<sup>5</sup> Z. El Alfy,<sup>7</sup> B. El Kaliouby,<sup>8</sup> Y. Dawood,<sup>8</sup> and A. M. A. Abdallah<sup>8</sup>

Received 8 December 2003; revised 28 January 2004; accepted 12 February 2004; published 12 March 2004.

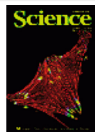


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R. Becker,<sup>3</sup> Z. El Alfy,<sup>7</sup> B. El Kaliouby,<sup>8</sup> Y. Dawood,<sup>8</sup> and A. M. A. Abdallah<sup>8</sup>

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PHYSICAL REVIEW LETTERS

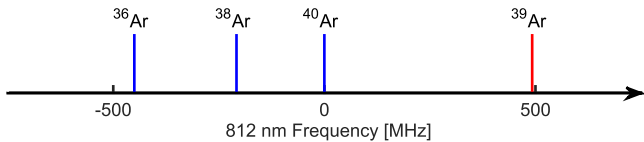
week ending  
11 MARCH 2011

## <sup>39</sup>Ar Detection at the 10<sup>-16</sup> Isotopic Abundance Level with Atom Trap Trace Analysis

W. Jiang,<sup>1</sup> W. Williams,<sup>1</sup> K. Bailey,<sup>1</sup> A. M. Davis,<sup>2,3</sup> S.-M. Hu,<sup>4</sup> Z.-T. Lu,<sup>1,2,5</sup> T. P. O'Connor,<sup>1</sup> R. Purtschert,<sup>6</sup>  
N. C. Sturchio,<sup>7</sup> Y. R. Sun,<sup>4</sup> and P. Mueller<sup>1</sup>

	<sup>85</sup> Kr	<sup>81</sup> Kr	<sup>39</sup> Ar
half life	10.76 a	232 ka	269 a
relative abundance	2 · 10 <sup>-11</sup>	6 · 10 <sup>-13</sup>	8 · 10 <sup>-16</sup>

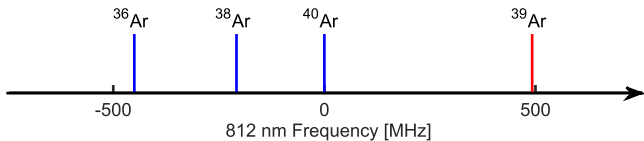
Isotope shifts:



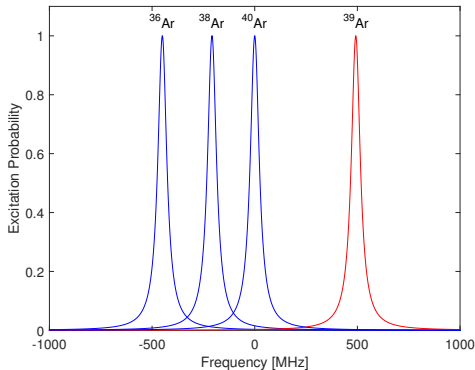


# Principle of ATTA

Isotope shifts:

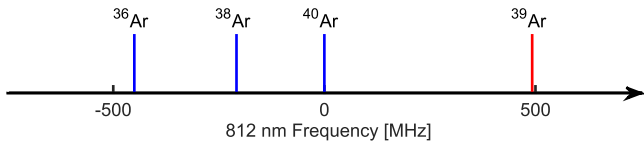


Transition rate:  $\sim 10^7$  Hz

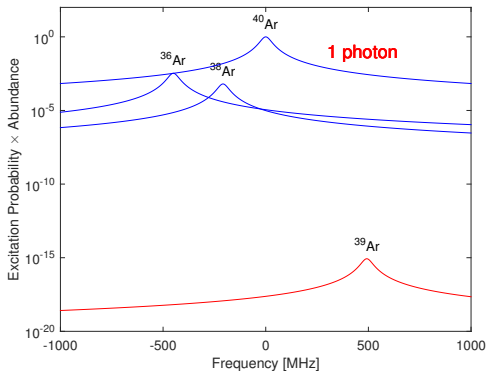


# Principle of ATTA

Isotope shifts:

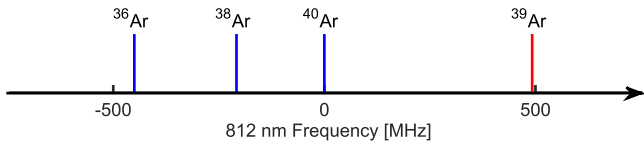


$(\text{Rate})^1 \times \text{Iso. abundance}$

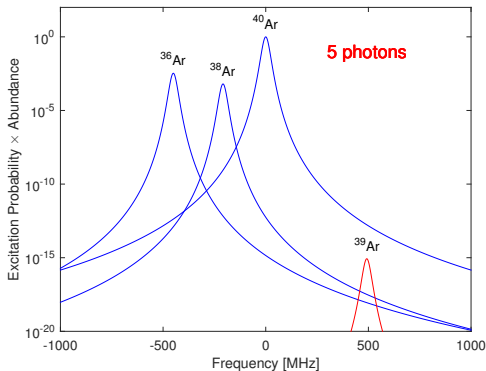


# Principle of ATTA

Isotope shifts:

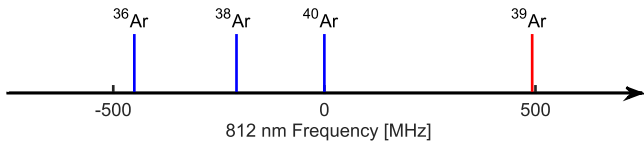


$$(\text{Rate})^5 \times \text{Iso. abundance}$$

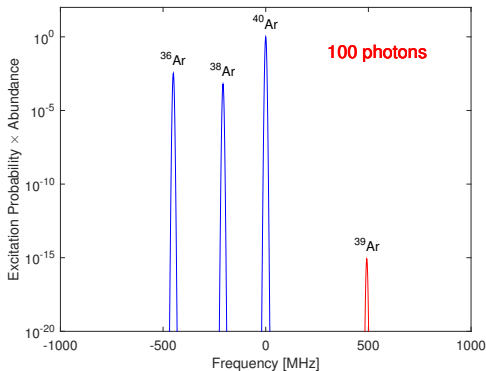


# Principle of ATTA

Isotope shifts:

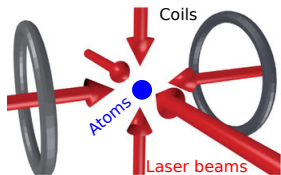
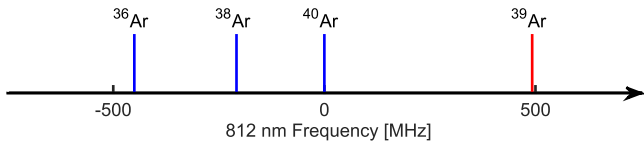


$$(\text{Rate})^{100} \times \text{Iso. abundance}$$

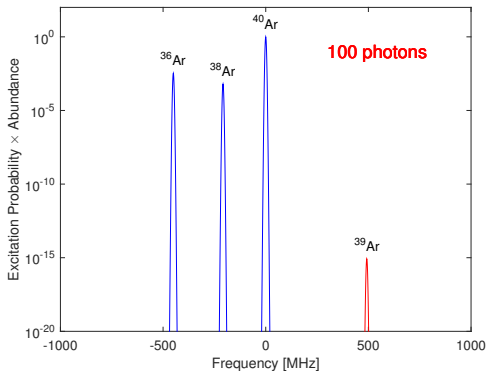


# Principle of ATTA

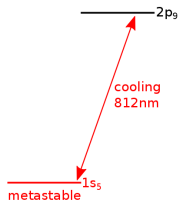
Isotope shifts:



Key characteristics:  
**Resonance & repetition**  
→ **Zero background**

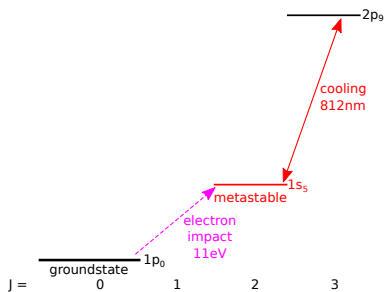
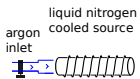


# $^{39}\text{Ar}$ -ATTA System

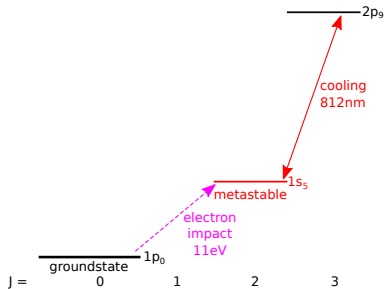
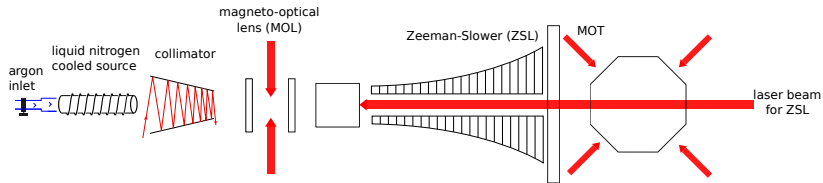


$J =$   $\overline{\text{groundstate}}$   $1p_0$   
0 1 2 3

# $^{39}\text{Ar}$ -ATTA System

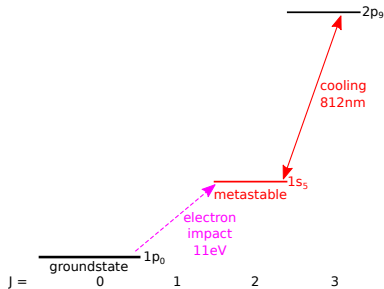
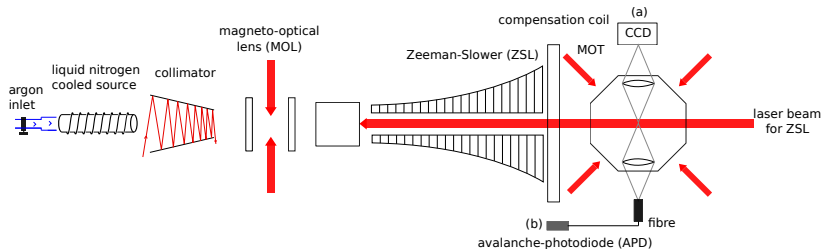


# $^{39}\text{Ar}$ -ATTA System

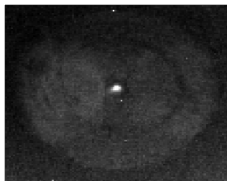




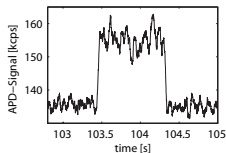
# $^{39}\text{Ar}$ -ATTA System



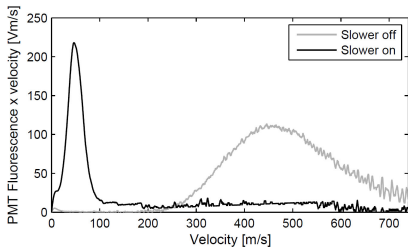
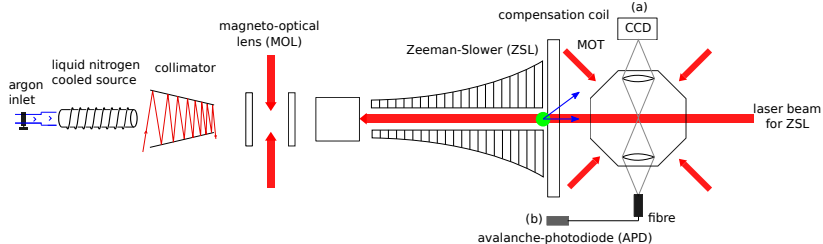
a)  $^{39}\text{Ar}$  atom on CCD



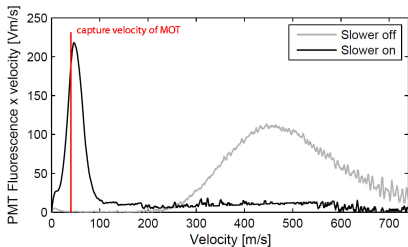
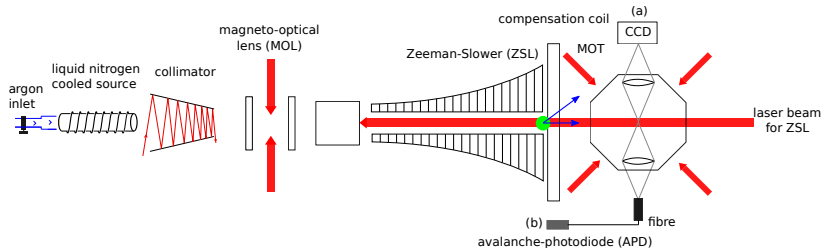
b)  $^{39}\text{Ar}$  atom on APD



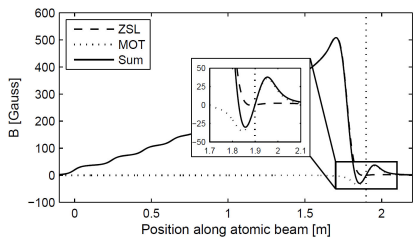
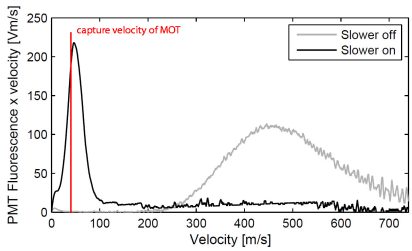
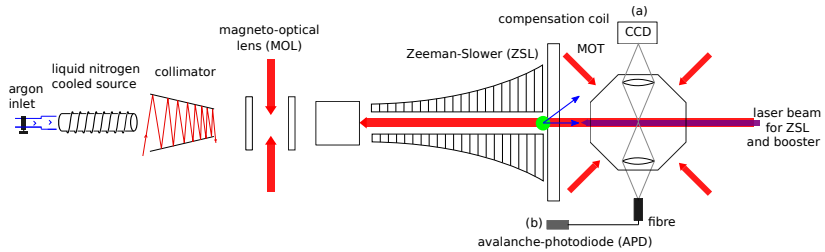
# <sup>39</sup>Ar-ATTA System



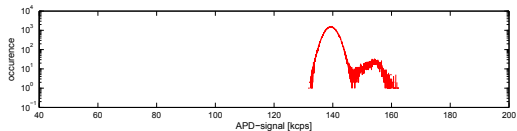
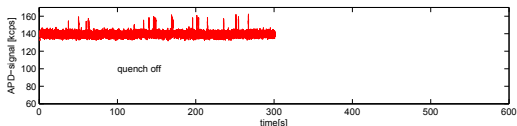
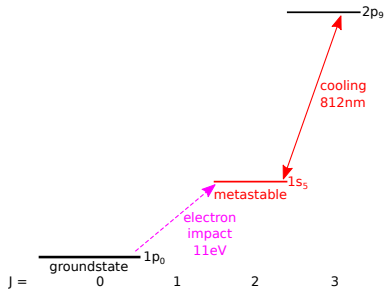
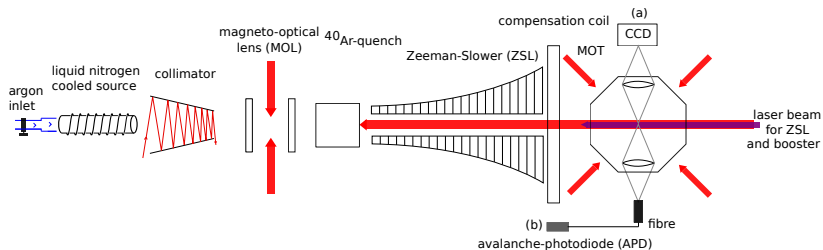
# <sup>39</sup>Ar-ATTA System



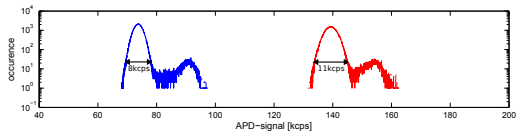
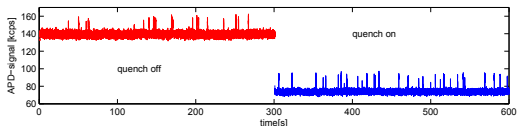
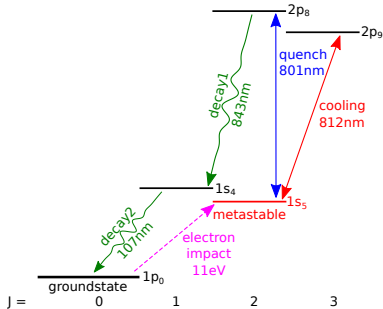
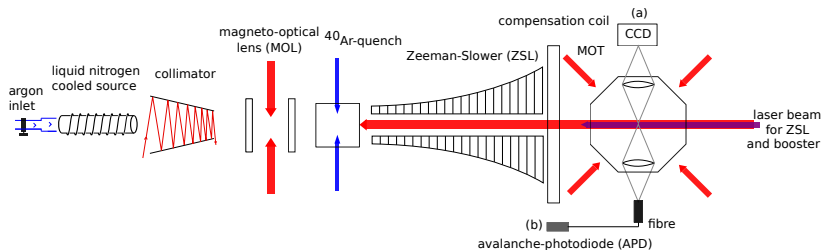
# <sup>39</sup>Ar-ATTA System



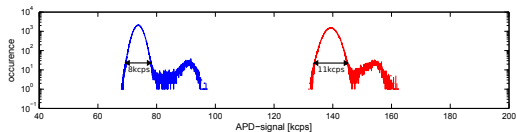
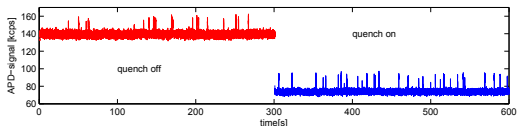
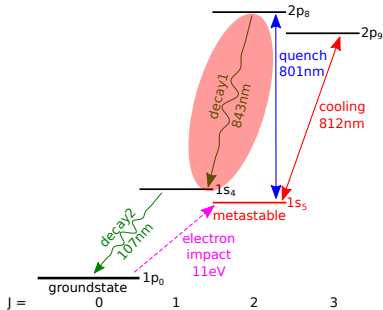
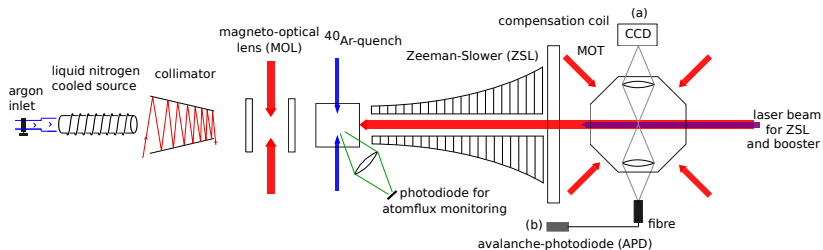
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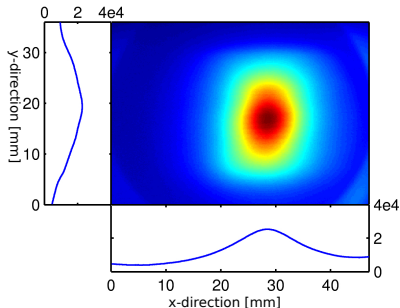
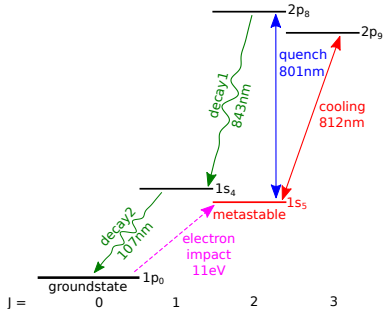
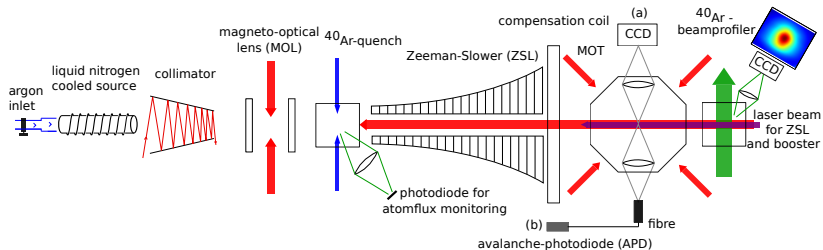
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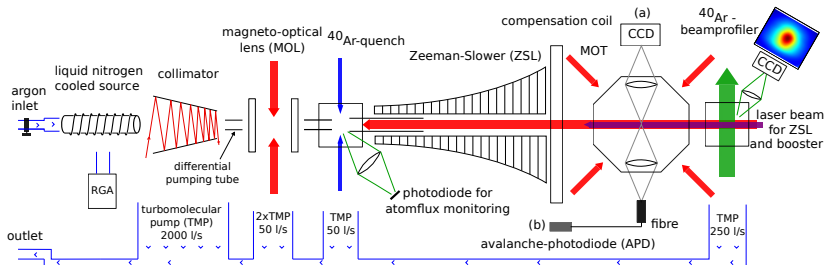


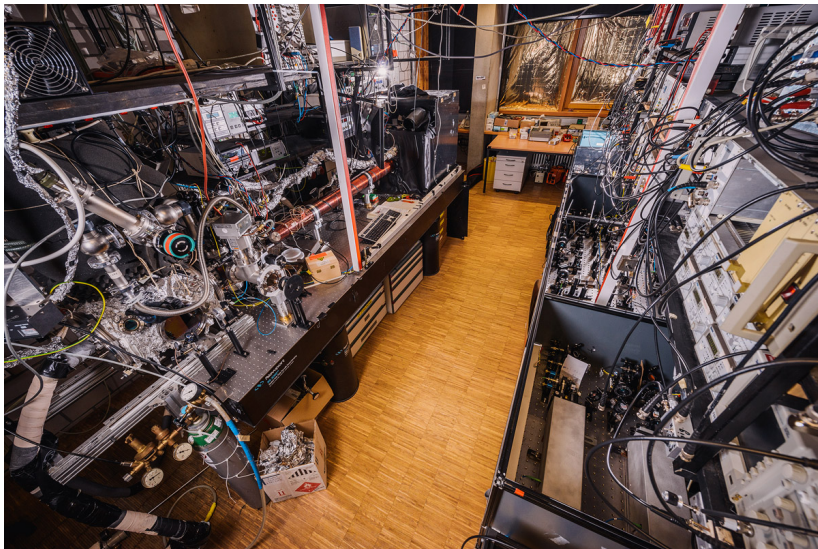
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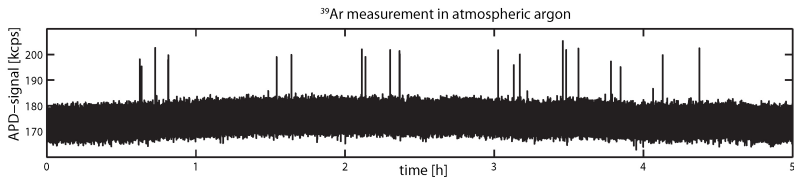
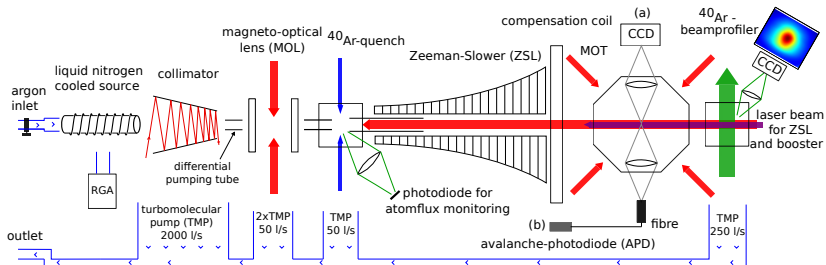


# <sup>39</sup>Ar-ATTA System





# $^{39}\text{Ar}$ -ATTA System



Atmospheric  $^{39}\text{Ar}$  count rate:  
3.6 atoms/h

# How to translate an $^{39}\text{Ar}$ count rate into a concentration?

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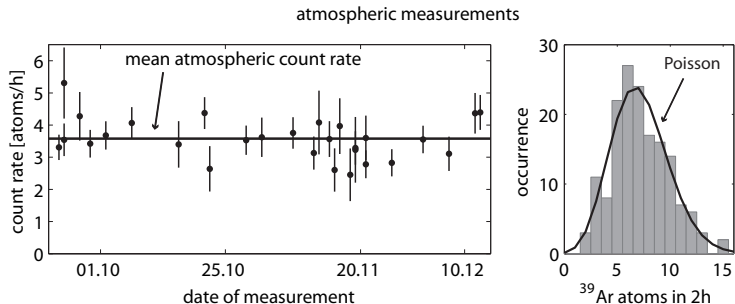
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F. Ritterbusch et al. (2014), Groundwater dating with Atom Trap Trace Analysis of  $^{39}\text{Ar}$ , Geophys. Res. Lett., 41



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 **AGU** PUBLICATIONS



## Geophysical Research Letters

### RESEARCH LETTER

10.1002/2014GL061120

#### Key Points:

- First dating of groundwater with Atom Trap Trace Analysis of Argon-39
- Argon-39-ATTA has the potential for Argon-39 analysis of small water and ice samples

### Groundwater dating with Atom Trap Trace Analysis of $^{39}\text{Ar}$

F. Ritterbusch<sup>1</sup>, S. Ebser<sup>1</sup>, J. Welte<sup>1</sup>, T. Reichel<sup>2</sup>, A. Kersting<sup>2</sup>, R. Purtschert<sup>3</sup>, W. Aeschbach-Hertig<sup>2</sup>, and M. K. Oberthaler<sup>1</sup>

<sup>1</sup>Kirchhoff-Institute for Physics, Heidelberg University, Heidelberg, Germany, <sup>2</sup>Institute of Environmental Physics, Heidelberg University, Heidelberg, Germany, <sup>3</sup>Climate and Environmental Physics, University of Bern, Bern, Switzerland

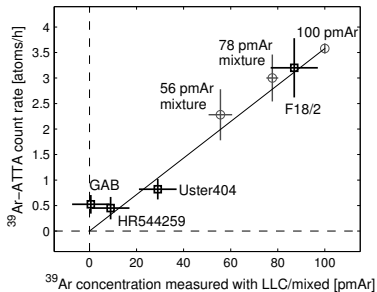
**Abstract** We report on the realization of Atom Trap Trace Analysis for  $^{39}\text{Ar}$  and its first application to dating of groundwater samples. The presented system achieves an atmospheric  $^{39}\text{Ar}$  count rate as high as

F. Ritterbusch et al. (2014), Groundwater dating with Atom Trap Trace Analysis of  $^{39}\text{Ar}$ , Geophys. Res. Lett., 41

# First Groundwatersamples: Results

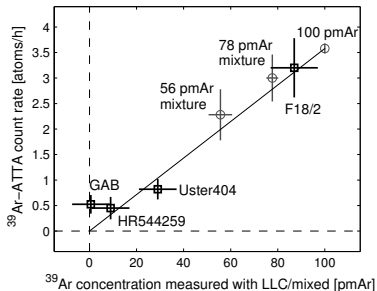


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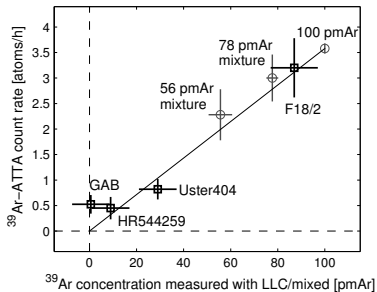
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	<i>F18/2</i>	<i>Uster404</i>	<i>HR544259</i>
NGT [°C]	7.8 ± 0.6	7.3 ± 0.5 <sup>a</sup>	4.3 ± 1.1
<sup>3</sup> H [TU]	7.40 ± 0.16	0.4 ± 0.1 <sup>a</sup>	0.6 ± 0.9
<sup>14</sup> C [pmC]	84.03 ± 0.24	22.1 ± 0.4 <sup>a</sup>	0.025 ± 0.020
<sup>39</sup> Ar age ATTA [a]	46 ± 71	574 ± 95	> 652

<sup>a</sup>values from *Beyerle et. al, 1998*

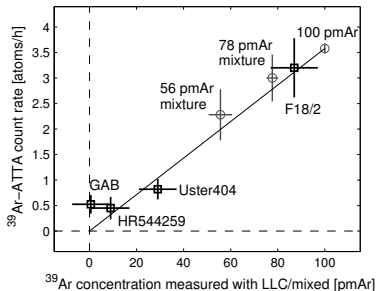
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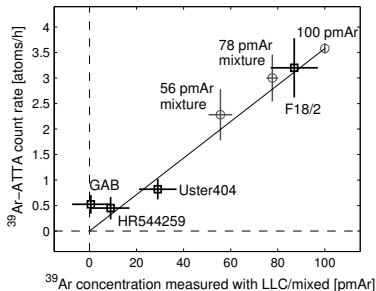
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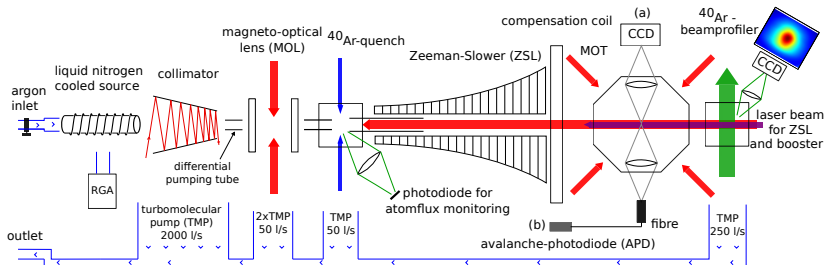
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→ Groundwater dating with  $^{39}\text{Ar}$ -ATTA demonstrated

F. Ritterbusch et al. (2014), Groundwater dating with Atom Trap Trace Analysis of  $^{39}\text{Ar}$ , *Geophys. Res. Lett.*, 41

# First Results with 4 ml Samples (Preliminary)



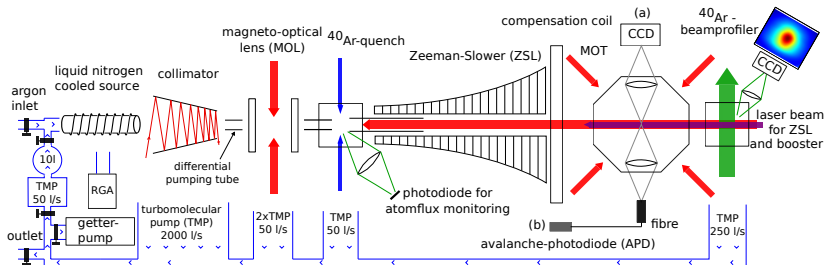
## Throughput configuration

Required sample size: 30 mL/h

0.5 -1 L STP argon  $\leftrightarrow$  1000 - 2500 kg water or 500 - 1000 kg ice



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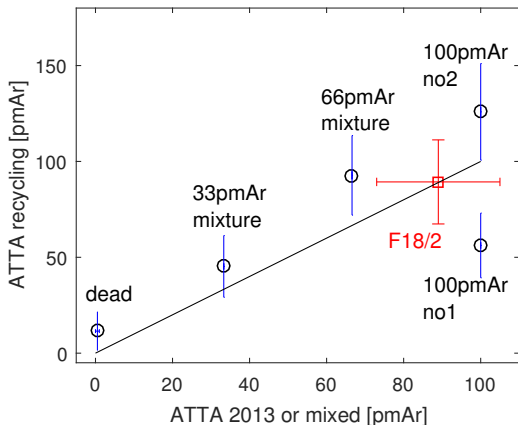
0.5 -1 L STP argon  $\leftrightarrow$  1000 - 2500 kg water or 500 - 1000 kg ice

## Recycling configuration

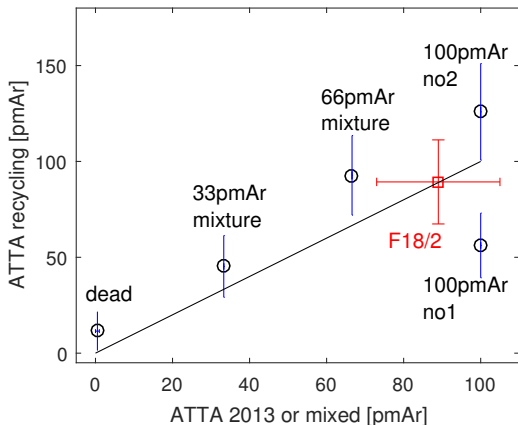
Required sample size due to contamination:

4 -10 mL STP argon  $\leftrightarrow$  10 - 25 L water or 4 - 10 kg ice

# First Results with 4 ml Samples (Preliminary)

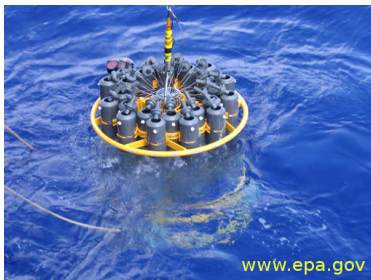


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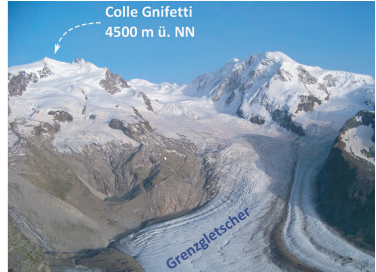


Reduction of the needed sample size by more than a factor 100!

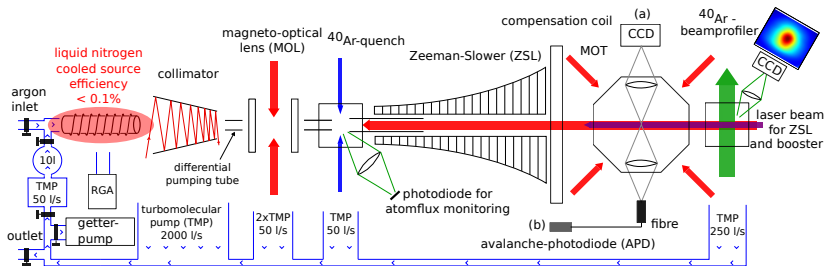
# In Progress: $^{39}\text{Ar}$ Dating of Glacier Ice and Ocean Water



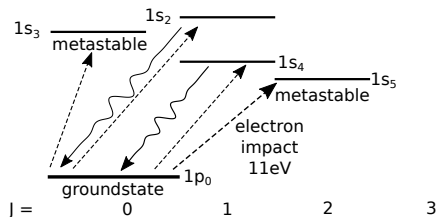
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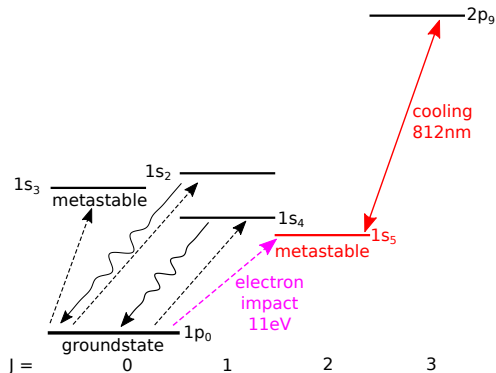
# Optical Pumping



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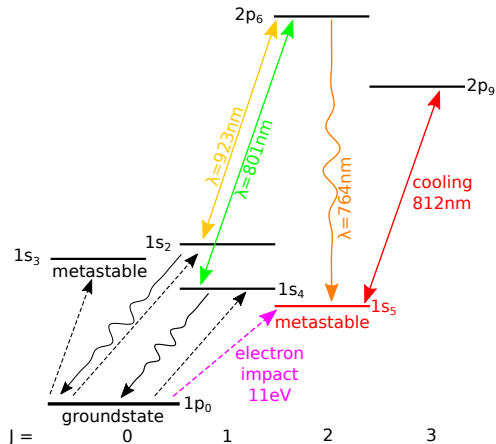


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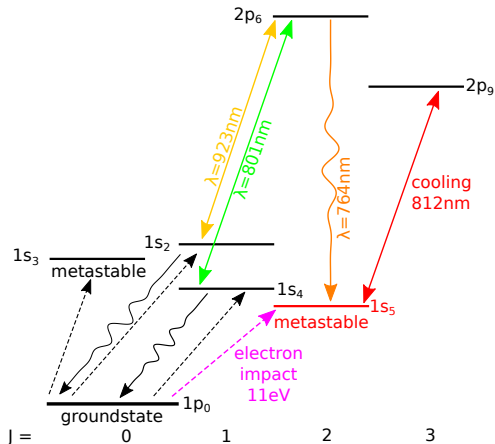




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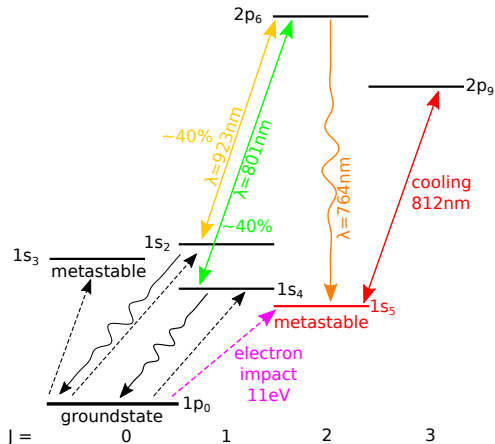
Anika  
Frölian



Maurus  
Hans

see: [www.matterwave.de](http://www.matterwave.de)

# Optical Pumping



Anika  
Frölian

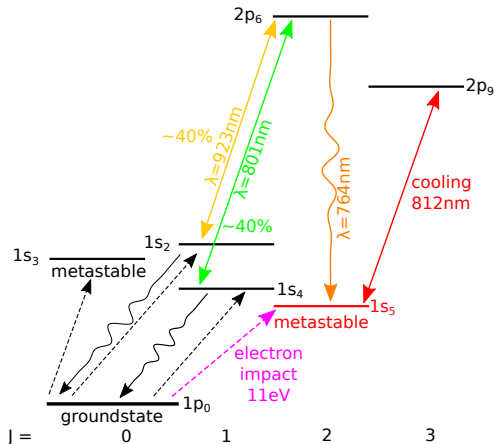


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In an  $^{40}\text{Ar}$  spectroscopy cell: 40% are gained by each transition, together 80%

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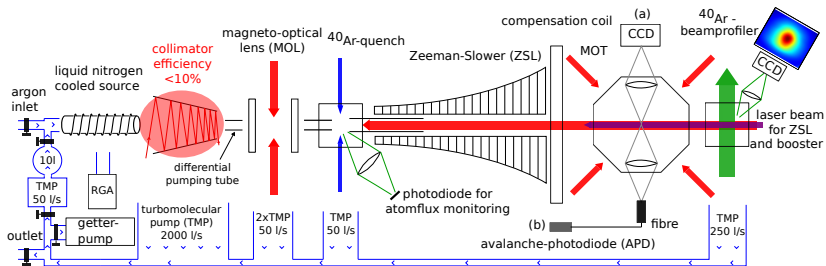
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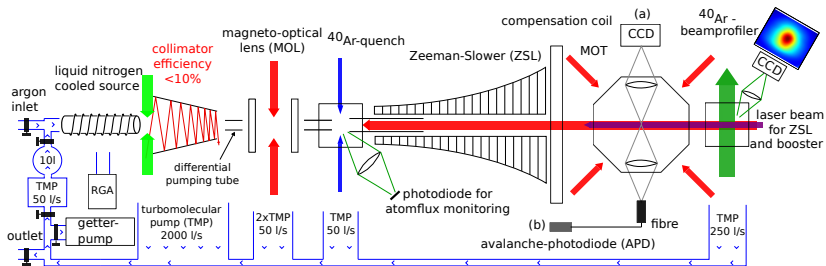
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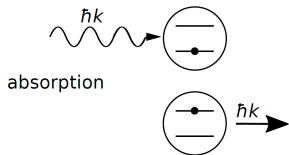


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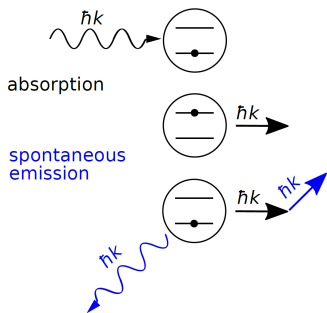
# Bichromatic Force: Basic Principle

Radiative force



# Bichromatic Force: Basic Principle

## Radiative force

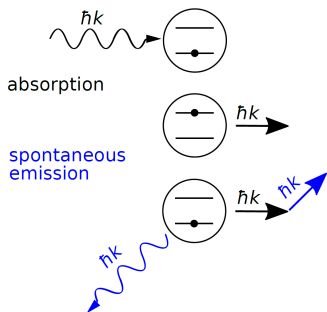


Force is limited:  $F_{rad} < \hbar k \frac{\gamma}{2}$

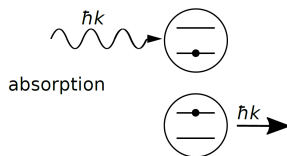


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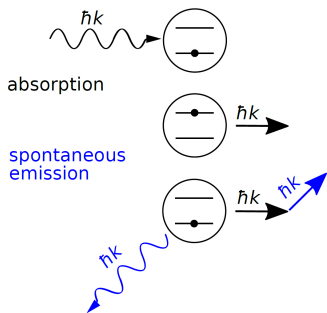
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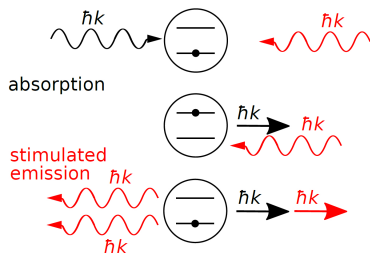
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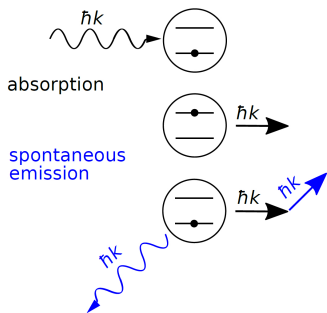
## Bichromatic force



Force is not limited by the spontaneous decay rate

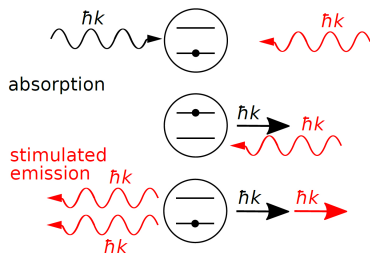
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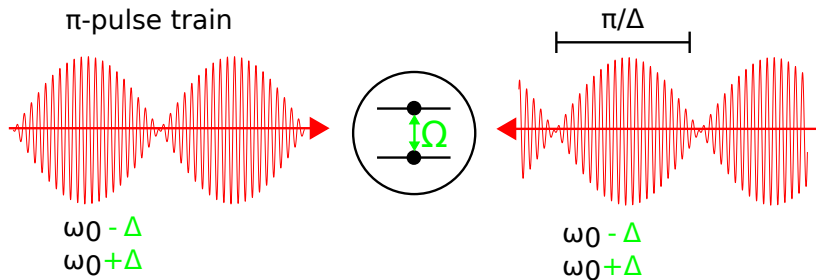
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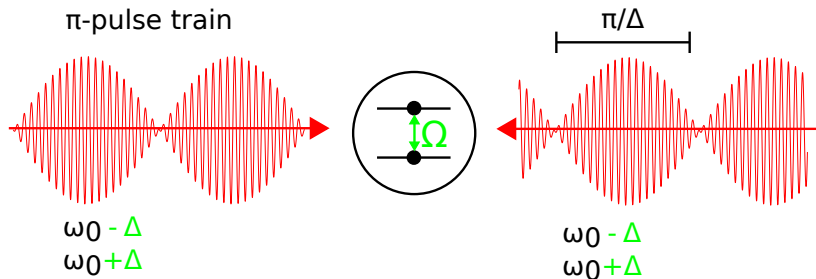
Force is not limited by the spontaneous decay rate

Calculation: Yatsenko (1991, 2004), Cs: J. Söding et. al. (1996),  
Rb: Williams/Metcalf (1999), He: Cashen/Metcalf (2001)

# Bichromatic Force: Realisation

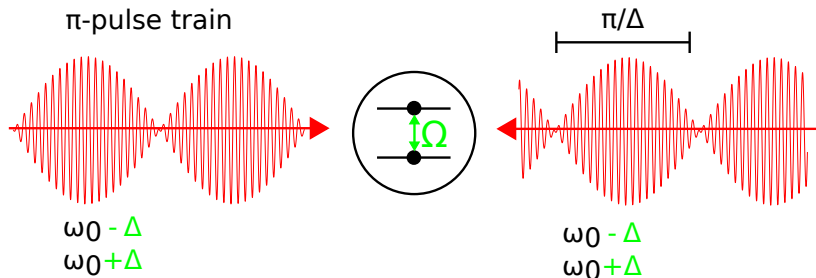


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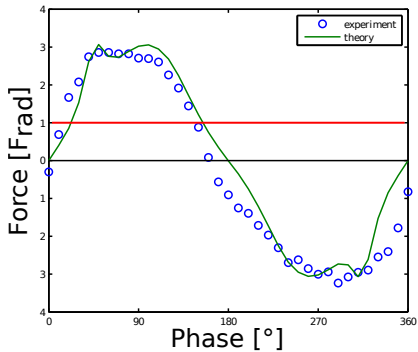
- Average force  $\bar{F} = \frac{\hbar k}{\pi} \Delta$  (only limited by power!)

# Bichromatic Force: Realisation



- Average force  $\bar{F} = \frac{\hbar k}{\pi} \Delta$  (only limited by power!)
- Optimum phase  $\phi = \frac{\pi}{2}$

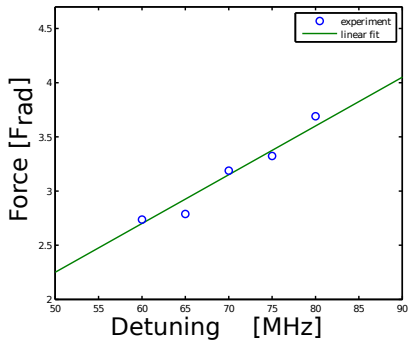
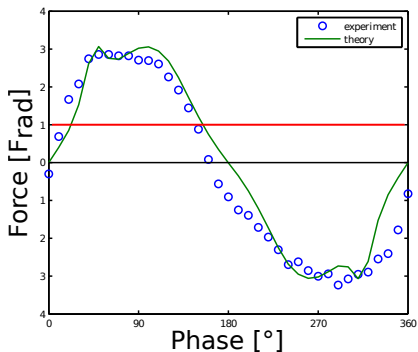
# Bichromatic Force: Experimental Results



Zhongyi  
Feng

see: [www.matterwave.de](http://www.matterwave.de)

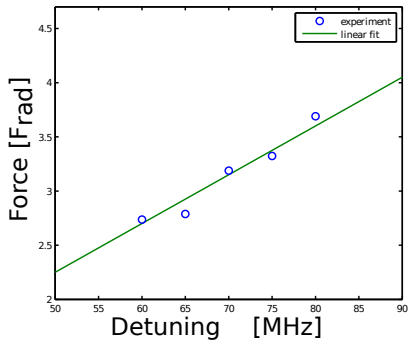
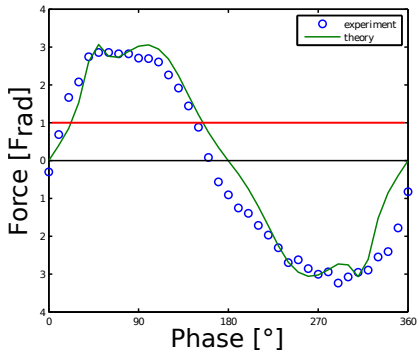
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$$\bar{F} = \frac{\hbar k}{\pi} \Delta$$

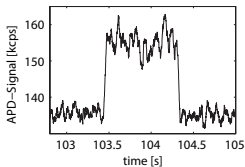


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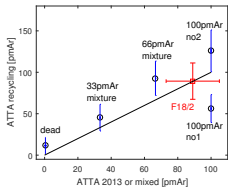


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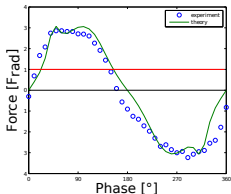
- For  $^{39}\text{Ar}$  the realisation of the repumper still has to be investigated



$^{39}\text{Ar}$  and Atom Trap Trace Analysis as an application of a rare noble gas



First demonstration of ATTA of  $^{39}\text{Ar}$  with groundwater (GRL 2014) and reduction of the sample size down to 4 ml



Study of new techniques for ATTA:

- Optical pumping
- Bichromatic force