All-optical Atom Trap Trace Analysis – Potential use of ^{85}Kr in Safeguards activities

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Abstract:
Sensitive measurement techniques for the detection of anthropogenic tracers demand measurement resolutions down to single atoms, as it has been demonstrated by the first atom trap trace analysis experiments. However, technical limitations had lowered the sample throughput to about 200 per year per machine. We have developed an all-optical apparatus which allows both higher sample throughput and smaller sample sizes than current techniques.
Krypton-85 as an anthropogenically produced isotope is an ideal tracer for nuclear activities since the only relevant source term is fission. An increased ^{85}Kr concentration in an air sample indicates a (Kr-85)-plume passing by during sampling. In practice, however, its applicability may be limited by the global and regional background concentrations caused by the emissions of nuclear fuel reprocessing plants.
The potential of ^{85}Kr monitoring for safeguards applications has been discussed extensively. Among these is the short range detection of elevated concentrations of ^{85}Kr in the vicinity of reprocessing plants. Our ATTA technique needs sample sizes of about 1 l of air only and thus for the first time will allow simple environmental sampling of ^{85}Kr with high spatial and temporal resolution. The design of such a study including local sampling and tracer transport modelling in proximity to a reprocessing plant is outlined. In addition, such a study could also be used for validating near-field atmospheric dispersion models if the ^{85}Kr source term is known. The potential of environmental analyses of ^{85}Kr during an IAEA short-notice access is discussed. It is shown that it crucially depends on the emission dynamics after shut-down of fuel dissolution which demands further studies.

Introduction

Since 2000 the fulminate development in the fields of laser cooling and trapping demonstrated the potential for single atom detection of neutral atoms. Even the cooling and trapping of noble gas atoms, krypton and argon, have been published down to the single atom regime. The potential applicability to measure the concentration of the rare radioactive krypton isotopes has been shown by the Argonne National Laboratory (Du, 2003) for the first time. This apparatus is based on a three dimensional magneto-optical trap, which is loaded by a Zeeman-slower. For cooling and trapping, krypton has to be excited to a metastable state, as there do not exist light sources (123.6 nm) to cool and trap krypton from the ground state. Alternatively, state-of-the-art experiments use a radio-frequency driven plasma to excite krypton with a probability of 10^{-4} to a metastable state, from which krypton can be cooled and trapped with laser light (811.5 nm).
With an Atom Trap Trace Analysis (ATTA-) apparatus, the determination of the ^{85}Kr content within current air-samples of about 1 - 10 litres could be done within 2 hours. The measurement follows a cleaning procedure of 36 hours to remove krypton implanted into the walls of the apparatus. By the excitation through electrons not only metastable krypton but also ionized krypton is produced, which is accelerated by the radio-frequency field. These fast ions can be implanted into the walls and degas during the measurement as long as the plasma is driven. To reduce this implanted amount of krypton, a xenon plasma is sustained for 36 hours. However, a slight degassing of krypton arisen from pre-measurements has to be accounted by the determination of the real concentration of the rare radioactive krypton isotopes. This effect limits the throughput per machine to about 200 per year (Lu, 2010).

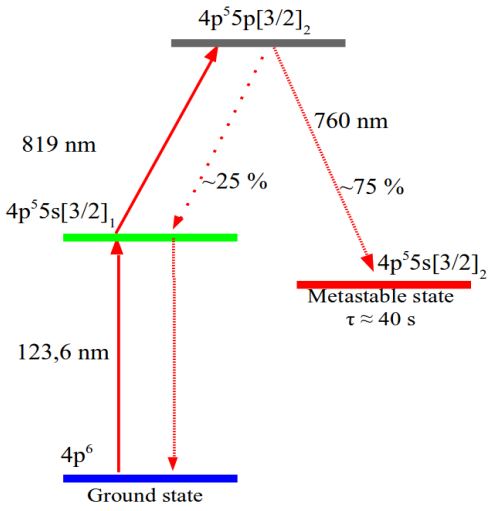

Figure 1 Optical excitation scheme to transfer krypton to the metastable state

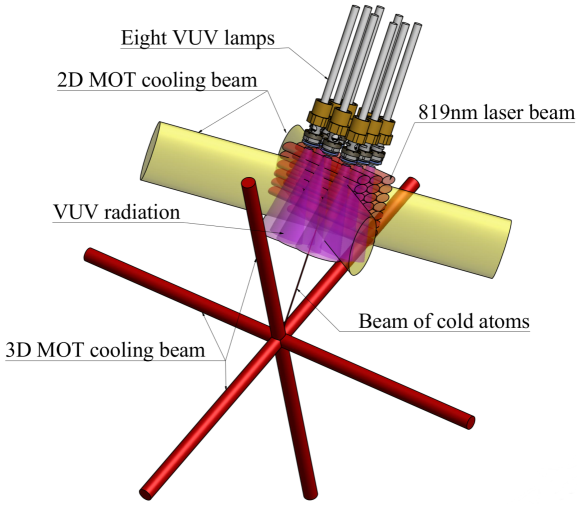

Figure 2 Illustration of the beam guidance used in the setup at the University of Hamburg.

All-optical production of metastable krypton

In 2002 first experiments to replace the production of the metastable krypton through collisions by an all-optical excitation scheme were executed. This scheme includes the absorption of two resonant photons (123.6 nm and 819 nm) followed by a spontaneous decay (760 nm) to the metastable state (see figure 1). The technical issue was to get a light source for the 123.6 nm radiation with a long lifetime as the common lamps had lifetimes of about 10 hours only (Young, 2002). In 2011 a design of a discharge lamp with a lifetime of more than 500 hours was published, which furthermore was attachable to an ultra-high vacuum chamber. With this item, an all-optical ATTA-apparatus, which overcomes the problems of cross-contamination, seemed to be available (Daerr, 2011).

Cooling and trapping of single krypton isotopes

The cooling and trapping mechanism within a magneto-optical trap is based on doppler-cooling. Here, an aligned absorption of red detuned photons, with propagation direction towards a metastable krypton atom slows the atom by the absorption process; the following disexcitation process will be affected spontaneously, so that every direction of the emission has the same probability. The momentum over several spontaneous emissions will add to zero. In addition to that, the amount of slowed down atoms will increase with every aligned absorption process.

In combination with an inhomogeneous magnetic field, atoms can be trapped at the place where the magnetic field has the value B = 0 T. These trapped atoms scatter continuously photons out of the laser beams. This fluorescence signal will be used to identify the presence of atoms. A magneto-optical trap is isotropically sensitive as the force caused by the scattering of photons is too small to cool down unwanted isotopes. Therefore, other isotopes will fall down through the trap inter alia by gravity.

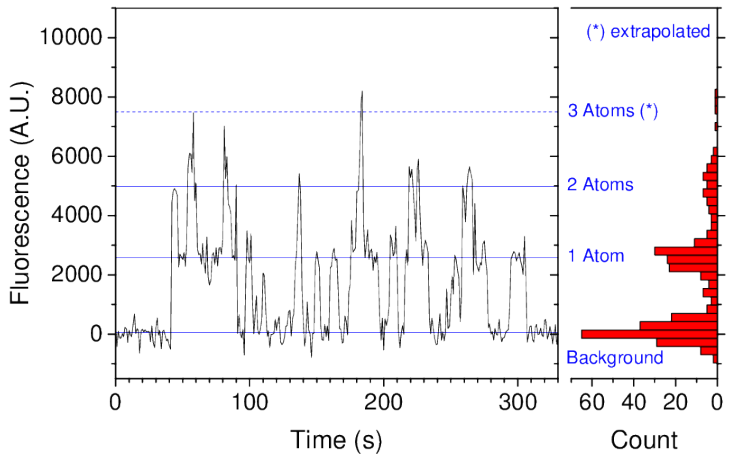

Figure 3 Fluorescence of few trapped 83Kr atoms to demonstrate single atom detection capability.

Single atom detection and analysis

Magneto-optical traps offer the possibility to trap and detect single atoms. The determination of the accurate number of trapped atoms is limited to about 1000 atoms (Hume, 2013). For the rare krypton isotopes only a few atoms are expected to be trapped within a magneto-optical trap. As parameters of the apparatus can change during measurement, a method which compares two isotopes is preferred. The amounts of ^{85}Kr and ^{81}Kr have the same ratio in air samples; therefore the sum of counted atoms of these isotopes in combination with the constant concentration of ^{81}Kr can be used to calculate the concentration of ^{85}Kr (Du, 2003).

All-optical apparatus to cool and trap neutral krypton atoms

The time for measuring a single sample consists of two parts: the time to count single atoms until the statistical error is acceptable and the time to clean the ultra-high vacuum chamber from residual sample gas. In state-of-the-art experiments, the second quota is much larger than the first. As a consequence the throughput depends on the cleaning (Lu, 2010). In all-optical machines the cleaning procedure is done by vacuum pumps only. In practice, the cleaning takes a few minutes until the pressure is lowered by some orders of magnitude. Therefore the limiting factor reduces to the time needed for measuring.

An all-optical apparatus based on a VUV-lamp has been built up at the University of Hamburg (Kohler, 2014). It consists of a 2D-3D magneto-optical trap setup (see figure 2); in the two dimensional trap metastable krypton is all-optically produced and a beam of atoms is formed by doppler-cooling simultaneously as illustrated in figure 1. With this setup ^{84}Kr and ^{83}Kr have been cooled and trapped, starting with atom clouds with thousands of atoms down to the single atom regime (see figure 3). This demonstrates that an all-optical ATTA-apparatus paves the way to overcome the technical problems of state-of-the-art setups.

Applications for rare krypton tracer isotopes

There is a wide field of applications in earth sciences and non-proliferation. As a dating tracer, both isotopes ^{85}Kr and ^{81}Kr can be used. ^{85}Kr has a half-life of about 10.76 years and can be used for dating young groundwater sources and their refreshment rates. On the other side, ^{81}Kr with a long half-life of about 229,000 years is useful to date old groundwater (Sturchio, 2004) or ice cores.

Moreover, the use of ^{85}Kr as an anthropogenic fission product for safeguard issues was outlined in a technical meeting of the IAEA (STR-351). The use of this isotope to detect reprocessing activities in on-area inspections was attested. In the case of a “short notice access” which is implemented by the additional protocol, air samples could identify ongoing reprocessing. However, this approach requires detailed knowledge of the dynamics of the ^{85}Kr emissions during reprocessing to get knowledge of the source terms during and after dissolution of spent fuel. This information may be crucial, since an emergency shutdown will be a plausible technical response to mask a clandestine reprocessing activity after a noticed visit through IAEA inspectors. Therefore a cooperation with operators of reprocessing plants to get access to ^{85}Kr atmospheric emission monitoring data with high time resolution should be intended and arranged.

On the other hand, the concentration of ^{85}Kr in weekly samples taken by the University of Hamburg and characterized by the Bundesamt für Strahlenschutz show high fluctuations. There are weekly samples which have more than double of the background concentration of ^{85}Kr and the only reasonable explanation is that the plume of a reprocessing facility passed Hamburg. For a definite identification of the reprocessing facility responsible for the higher ^{85}Kr content in the air the time resolution is too poor. As one example figure 4 demonstrates that the output of trajectories given by HYSPLIT could tag the emission to both civilian reprocessing plants in Europe. The implementation of a higher sampling rate in the order of three hours will show up the possibility to better identify the time when the plume reaches a specific sampling point; furthermore the characteristics of a plume can be determined and ideally a single trajectory to identify the source area will result. In addition, measurements of ^{85}Kr with high temporal resolution could be used to validate mesoscale and regional atmospheric transport models.

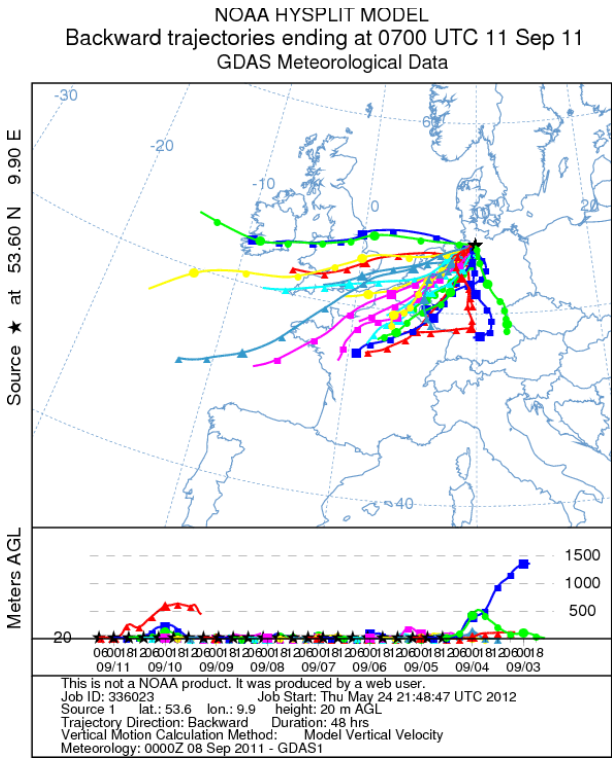

Figure 4 Analysis of a weekly sample. The trajectories "identify" La Hague as well as Sellafield as possible emitter.

Conclusions

There is a wide field of applications for radioactive noble gas tracers. In particular, applications of the atom trap trace analysis (ATTA) detection method of rare krypton isotopes are promising. As an ideal environmental tracer the use for groundwater dating is outlined. Applicability in potential future safeguard regimes is discussed. An all-optical Atom Trap Trace Analysis apparatus will allow research to reveal clandestine reprocessing activities. On the one hand, the precise dynamics of the krypton emissions during reprocessing and after an emergency shut-down have to be measured. As a result the implementation of this method in the safeguard regime seems feasible. On the other hand, a better temporal identification of atmospheric ^{85}Kr plumes and their characteristic on large scales will verify experimentally whether this method is suitable to identify emitters.

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