

論文の内容の要旨

論文題目 Laser based investigations in calcium isotope selective
excitation
(レーザーを利用したカルシウム同位体選択的励起の検討)

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Calcium is an important element for many reasons. Most famously it is a major constituent of human and animal bones, which gives it a valuable role in medicine and biomedical sciences. In regard to nuclear engineering, calcium is chemically a large proportion of the concrete that forms the biological shield around nuclear reactors. During the lifetime of a typical reactor, the majority abundance isotope Ca-40 (see Table 1.) undergoes neutron activation in the high neutron flux environment to form Ca-41. Ca-41 is a radioisotope with unusual decay pathway: only electron capture followed by low energy (< 3.6 keV) X-Ray or Auger electron release; and long half-life of 99.4 ka. Consequently, methods in quantitative determination of Ca-41 content for waste stream assignment (clearance level: 100 Bq/g \Rightarrow Ca-41/Ca-40 selectivity of $2E7$) is limited to Accelerator Mass Spectrometry (AMS) and Liquid Scintillation Counting (LSC), which both have significant practical drawbacks in cost and facility availability (AMS), and time investment/experimental difficulty (LSC). Other important uses for Ca-41 are as a biomedical tracer for bone analysis; and cosmogenic investigations (requiring selectivities approaching $1E14$). In this context, a method of high selectivity analysis for Ca-41, that could be condensed to a

benchtop system, while achieving comparable or better precision and isotope selectivity to the aforementioned methods is desirable.

In this thesis I have taken three approaches to investigate calcium isotopes. First, the higher lying Rydberg (1P1 and 3F1) levels are investigated to calculate their isotope shifts, and where possible, the hyperfine structure of Ca-43. Motivation for this experimentation lies in that this data for these Rydberg levels have not been reported before. In addition, higher lying Rydberg levels of the odd-isotopes are known to have energies affected by fine structure and hyperfine mixing, therefore further investigation of these levels could contribute to further understanding. In the context of selectivity, the experimental technique uses a triple-resonance excitation scheme, highly selective in ionising only the targeted isotope.

The second approach is an in-depth study on the optical polarisation rule effect on odd-even isotope ionisation efficiency. Through quantum mechanical treatments of stimulated absorption, by placing the exciting electron in an $l=1$, $J=1$, $m_J=0$ state through a linearly polarised laser, selecting the next excitation step to be $J=0$, $m_J=0$, then for a laser with polarisation linearly orthogonal to the first, the transition is forbidden by $\Delta m_J \neq 0$ selection rules.

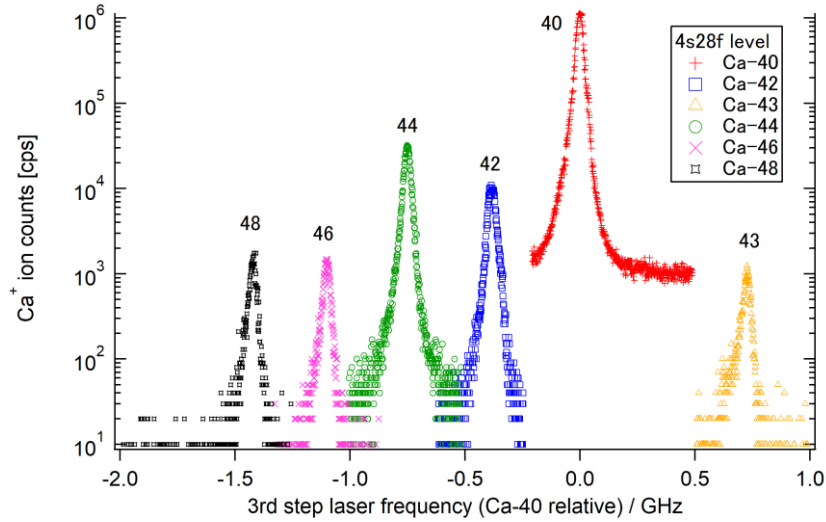


Figure 1: Example of experimental spectroscopic result, showing the resonance frequencies of all stable Ca isotopes (relative to Ca-40).

An additional aspect of this experiment is the investigation of a transition scheme that has not been reported on before. For reference, the ‘traditional’ scheme well reported on in the past uses the $[4s2\ 1S0 \rightarrow 4s4p\ 1P1 \bigcirc \rightarrow 4s4d\ 1D2 \rightarrow 4sn p\ 1P1 \text{ or } 4sn f\ 1F3]$ scheme, with laser wavelengths $[422.7\text{nm}$,

732.7nm, \sim 845nm] for each respective transition. In order for the optical selection rules to apply, the ‘new’ transition scheme is as follows: [4s2 1S0 \rightarrow 4s4p 1P1 \rightarrow 4s5s 1S0 \rightarrow 4snp 1P1] with laser wavelengths: [422.7nm, 1034.7nm, \sim 630nm]. All laser emission is provided by homemade external cavity diode lasers, with frequency stabilised against a He-Ne stabilised Fabry-Perot interferometer. The control system also allows for fine frequency control, which enables the frequency to be scanned for spectroscopy to be carried out.

With the results obtained here forming a new context on selectivities achievable through laser resonance excitation and selection rule manipulations, additional motivation to investigate the extremely high selectivities obtainable with the technique of ion trapping was developed.

Experimentally, for the Rydberg level spectroscopy, an all-ECDL laser system. Ions produced in the atom-laser interaction were mass selected and counted by a quadrupole mass spectrometer (QMS) system. The spectroscopy was performed by setting the first and second step lasers to the resonant frequencies (confirmed by ion signal maximisation) and scanning the third step laser over a range of 3 GHz. After each Ca isotope peak was observed, the first and second laser frequencies were switched to the resonant frequency of the next isotope of interest. In this way, the resonant ion peaks of each isotope are recorded, with the relative frequency difference between the Ca-40 peak and the other isotopes providing the isotope shift information - see Figure 1.

With ECDL replacement to access the 1034.7nm and \sim 630nm transitions, half-wave plates were introduced into the first and second step beams to control the linear polarisation orientation. The Ca-40 signal was first maximised to confirm the transition frequencies were set correctly, as this scheme had not been investigated before. First, variation of the Ca-40 ionisation efficiency was observed as a function of the second step laser polarisation orientation. Clear extinction of the ion signal was confirmed when the orientation coincided with the orthogonal condition with respect to the first step laser. Secondly, non-complete extinction of the Ca-43 ionisation efficiency was also accordingly confirmed, see Figure 2. However, due to the low abundance of Ca-43 in the materials, as well as relatively low ionisation efficiency for this scheme (due to high laser powers being unavailable), the highest achievable selectivity for this process is limited to $\sim 10^9$. Should higher laser powers be available at the required wavelengths, we expect the maximum selectivity measurable to be higher. As a comparison, the value achieved here is appropriately sufficient for Ca-41 concrete

clearance level detection.

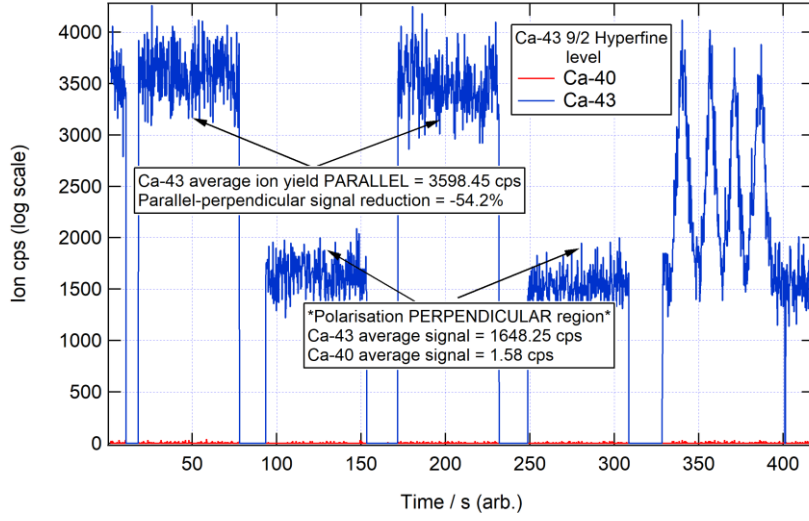


Figure 2: Effect of polarisation on the ionisation efficiency of Ca-43. Isotope shift suppressed Ca-40 signal included for comparison.

In this context, investigations into the use of ion trapping odd-isotopes of Ca were carried out. Experience in this laboratory of ion trapping Ca-43 is abundant, however ion trapping of Ca-41 has not been achieved or reported on in literature before, providing motivation for attempting so. In this thesis, a report is made of the activities carried out to reach this objective, including current experimental parameters.

To conclude, this thesis introduces the motivations for investigating calcium isotopes, then discusses the approaches used. Namely: triple-resonance laser selective ionization for Rydberg level isotope shift data; triple-resonance laser ionization combined with optical polarization selection rule manipulation for odd-Ca isotope selective ionization as a method for trace isotope detection of Ca-41; and also ion trapping of odd-Ca isotopes for ion-by-ion selectivity determination. Results at all these steps are fully discussed, with the newly reported Rydberg level isotopes shift values, the selectivity attained with the polarization effect, and ion trap development progress major achievements. In the future, higher power laser investigations of the polarization selection effect would be beneficial, to overcome the small ion count issue. In addition, reporting of the first successful Ca-41 ion trap, with associated spectroscopic data is an area of interest.