

High resolution ablation fluorescence spectroscopy for remote isotopic analysis

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Laser ablation has been applied in many fields such as material science and nuclear industry. In the Fukushima decommissioning, the use of laser induced breakdown spectrometry (LIBS) is desired for remote analysis of highly radioactive fuel debris. However, it is feared that LIBS may not provide sufficient spectroscopic resolution for obtaining isotopic compositions of the debris samples. In this study, we attempted high-resolution spectroscopy using an ablation atomic source by irradiating two resonant laser beams in opposite directions onto a laser plasma cooled in a reduced gas environment and by observing the resonant fluorescence from the highly excited state produced by the two-step resonant excitation of atoms.

Laser plasma was generated by irradiating a 532-nm (10-Hz) Nd:YAG laser beam vertically onto a compressed cement pellet on a rotating sample stage in a vacuum chamber. The plasma was irradiated by two external cavity diode lasers at $\lambda_1=422\text{nm}$ and $\lambda_2=732\text{nm}$, which are necessary for the two-step excitation of Ca atoms, at about 2mm above the sample surface. The 2nd-step laser wavelength was scanned continuously, and when the ⁴⁰Ca fluorescence peak was passed, the 1st-step laser wavelength was switched to the ⁴²Ca and ⁴⁴Ca resonance wavelengths in sequence to obtain a high-resolution fluorescence spectrum of Ca isotopes (Fig.1) [1]. The linewidth of Ca observed with Doppler-limited fluorescence spectroscopy is about 2 GHz, and the Doppler-free technique has narrowed the linewidth to about 70 MHz. This has demonstrated the possibility of remote isotope analysis even for nuclides with small isotope shifts, which have been difficult to measure.

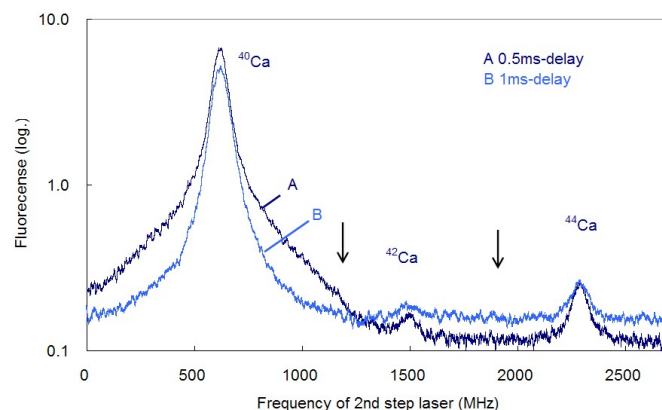


Fig1 Isotope-selective fluorescence spectrum of Ca

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References:

[1] M. Miyabe, M. Kato, S. Hasegawa, "Doppler-free ablation fluorescence spectroscopy of Ca for high-resolution remote isotopic analysis", J. Anal. At. Spectrom, 38 (2023) 347-358