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TITLE **VERY HIGH RESOLUTION SATURATION SPECTROSCOPY OF LUTETIUM ISOTOPES VIA CW SINGLE-FREQUENCY LASER RESONANCE IONIZATION MASS SPECTROMETRY**

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VERY HIGH RESOLUTION SATURATION SPECTROSCOPY OF LUTETIUM ISOTOPES VIA CW SINGLE-FREQUENCY LASER RESONANCE IONIZATION MASS SPECTROMETRY

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ABSTRACT

In this paper, we discuss the use of Resonance Ionization Mass Spectrometry (RIMS) to perform isotopically selective saturation spectroscopy of lutetium isotopes. Utilizing this technique, it is shown that accurate measurements of the relative frequencies of hyperfine (HF) components for different isotopes easily can be made without the need for an isotopically enriched sample. The precision with which the HF splitting constants can be determined is estimated to be ~5 times greater than in previous work.

INTRODUCTION

Considerable interest in the technique of RIMS has developed over recent years due to: 1) the selective manner in which isobaric interferences are discriminated against,^{1,2} and 2) the large dynamic range available for measuring isotopic ratios.³ Earlier work³ has demonstrated the ability to measure lutetium isotopic ratios down to the 0.4 ppm level on very small samples (~60 ng). In this previous work, the isotope ratio dynamic range was basically limited by low signal. Recently however, we have demonstrated that using a second, high-power, non-resonant laser for the ionisation step can dramatically increase ionisation efficiency.^{4,5} Here, the mass spectrometer resolution ultimately limits the dynamic range. In order to significantly increase the dynamic range even further, it may become necessary to perform isotopically selective resonance ionization. In the present work, Doppler-free saturation spectroscopy via RIMS is utilized for this goal, i.e., to obtain precise determination of the various HF components for the lutetium isotopes.

EXPERIMENTAL SECTION

Briefly, an ultraviolet Ar⁺ laser pumped ring dye laser operating with Stilbene 3 (~65 mW) was tuned to the one-photon transition of lutetium ($5d6s6p\ ^2D^0_{3/2} \leftarrow 5d6s^2\ ^2D_{3/2}$) at $\sim 22125\text{ cm}^{-1}$ (~452 nm). To increase the ionization efficiency,^{4,5} a second Ar⁺ laser tuned to the 457.9 nm line ionized the excited atom. Laser beams were propagated parallel to and ~2 mm above the sample filament, with typical beam diameters of ~100 μm . A spherical mirror was inserted to retroreflect the laser beams collinearly. A magnetic sector mass spectrometer, equipped with pulse counting electronics, was used for detection of the lutetium ions. Samples were prepared by depositing 1-2 μg of total lutetium onto a zone-refined rhenium filament, along with a similar amount of uranium oxide to provide a diffusive barrier. Spectra were taken at a filament temperature of ~1225° C.

RESULTS AND DISCUSSION

Saturation spectroscopy was discovered soon after the advent of the first gas laser. Bennet⁶ and Lamb⁷ recognized that the narrow resonances (Lamb dips) that appeared in the center of inhomogeneously broadened gain profiles interacting with counterpropagating laser beams resulted from "holes" burned into the Maxwell-Boltzmann velocity distribution.

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This phenomenon simultaneously provides a means for determining the center frequency of a transition and for removing the inhomogeneous line broadening.

Much saturation spectroscopy has been performed in a pump-probe scheme using a relatively high pressure static cell, with inherent difficulty in examining rare isotopes. In contrast, for the present experiments, the pump and probe are the same laser, and a low-pressure mass spectrometer is used as the detector. This removes possible pressure broadening effects and permits simple isotopically selective saturation spectroscopy of unenriched samples. Two features generally are observed: (1) dips which reflect line centers approaching their natural linewidth, and (2) crossover peaks which occur at the mean frequency of two hyperfine lines whose Doppler profiles overlap. The details of the exact processes occurring in these experiments will be discussed more fully in a forthcoming paper.⁸

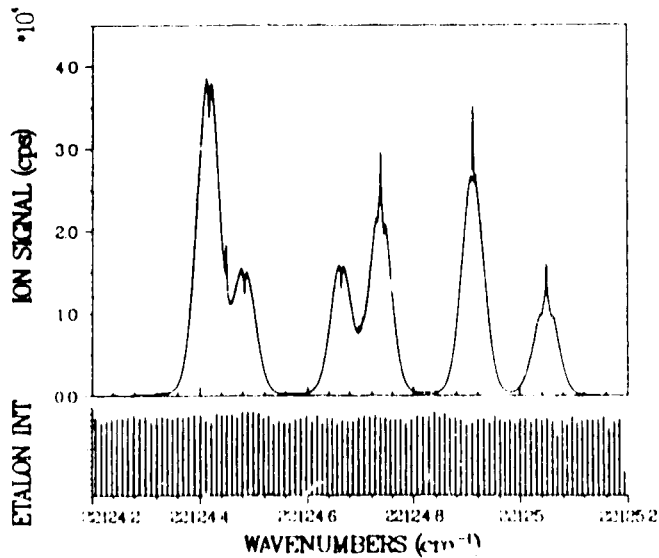


Figure 1. Saturation spectrum of $(5d6s6p\ ^2D_{3/2}^0 \leftrightarrow 5d6s^2\ ^2D_{3/2})$ transition of ^{175}Lu at $\sim 22125\text{ cm}^{-1}$.

A typical experimental spectrum for ^{175}Lu is shown in Figure 1. The dye and Ar^+ powers were $\sim 40\text{ W/cm}^2$ and $\sim 8.5\text{ kW/cm}^2$, respectively. Similar ^{176}Lu spectra were also observed but are not shown. The dips and peaks correspond to the line centers and crossovers for the atomic HF components for lutetium (see reference 8 for details). Excellent signal-to-noise (S/N) was observed for both ^{175}Lu and ^{176}Lu spectra. Figure 2 is an unsmoothed, expanded view of the first two bands in the ^{175}Lu spectrum and illustrates the S/N and resolution attained using this technique.

Included in the spectra, Figures 1 and 2, are the transmission peaks of a $\sim 300\text{ MHz}$ confocal etalon used for frequency calibration. For increased precision, the free spectral range of the etalon was determined to better than 2 ppm using the Los Alamos National Laboratory Fourier Transform Spectrometer (LANL FTS), greatly exceeding data requirements. Because of this precision and the near natural linewidth characterizing the dips and peaks, the hyperfine splitting constants for the ^{175}Lu excited states determined from the dip spectra (see Figures 1 and 2) are expected to be ~ 5 times more precise than previously determined.^{9,10} The exact determination of the HFS constants and their precision is presently underway and will be presented in detail in reference 8.

With a simple experimental modification, i.e., the inclusion of a "vibrating" mirror, removal of the Doppler pedestal is possible. Earlier studies in a standard gas cell¹¹ have shown that this can be accomplished by modulating the mirror and phase detecting the ionisation signal.

CONCLUSIONS

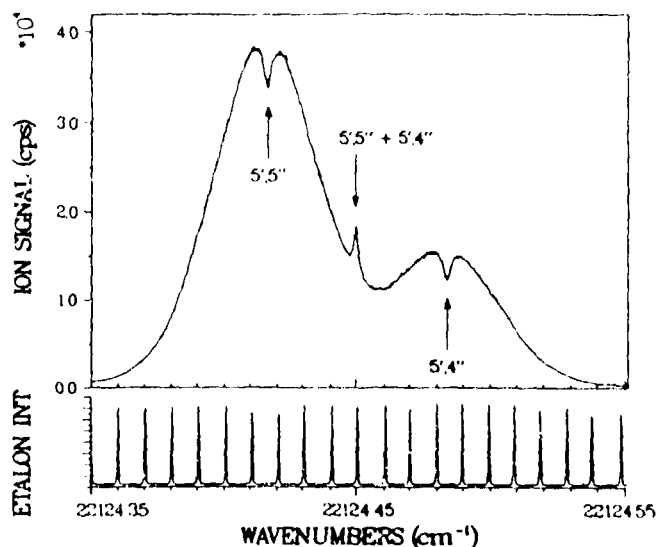


Figure 2. Expanded, unsmoothed spectrum of first two bands of ^{175}Lu shown in Figure 1. The arrows point out the dips and crossover for the indicated hyperfine transitions.

A new technique utilizing RIMS for obtaining very high resolution atomic spectra with isotopic selectivity was demonstrated. This technique allows the precise determination of HF splitting constants, limited only by the transition's natural line-width. In addition, it is also feasible with this technique to accurately determine atomic isotope shifts. The exact determination of HF component line positions provides data for isotopically selective ionization which, in turn, will increase RIMS' dynamic range. Future work includes the incorporation of a "vibrating" mirror and the study of rarer isotopes, i.e., ^{174}Lu , ^{173}Lu , ^{172}Lu , ^{171}Lu , and possibly, ^{170}Lu .

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