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Efficient ionization of atomic tin assisted by N-type multiphoton Raman resonances

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ABSTRACT

We report on the experimental results for laser ionization spectroscopy of atomic tin using a four-color three-step laser ionization scheme in conjunction with an atomic beam apparatus. Typically, the laser ionization efficiency is remarkably limited for certain elements whose thermal population is distributed over its higher lying components of the fine structure at high temperature (e.g. about 2000 K). We demonstrate that the laser ionization efficiency can be increased by simultaneous excitation from the $^3P_0$ and $^3P_1$ fine-structure components of the $5s^25p^2$ ground-state of atomic tin. This was achieved using two lasers, operating at 283 nm and 301 nm. Furthermore, four-level N-type mutiphoton Raman resonances in this four-color ionization scheme can be utilized to enhance the ionization efficiency allowing an additional ionization process from the third ground state $5s^25p^2\ ^3P_2$.

1. Introduction

Resonant laser ionization nowadays has become the most popular as well as the standard ionization technique for production of isobaric-pure Radioactive Ion Beams (RIBs) and for various applications in laser spectroscopy at radioactive ion beam facilities [1–3]. The exceptionally high chemical selectivity of the laser ion source comes from the fact that the atom of each element has its own intrinsic energy level structure. The total efficiency of the laser ion source is mainly determined by the efficiency of the laser ionization process. Thus, the developing new efficient ionization schemes for each element with different atomic energy level structure is of particular importance to increase the ionization efficiency.

In certain elements the ground state is split into several states due to a pronounced fine structure. When the splitting exceeds the bandwidth of the first step laser the thermal distribution over the different states should be taken into account. The occupation η_i of the individual levels can be calculated with a Boltzmann distribution [4]: $\eta_i \sim g_i \exp(-E_i/k_B T)$ with the statistical weights $g_i = 2J_i + 1$ of the individual states where E_i and T are the excitation energy and the absolute temperature of the ionizer, respectively. At a high temperature, e.g. $T \approx 2000$ K, the thermal distribution in an ion source spreads the population over these fine-structure ground levels. Thus, only a fraction of the atoms can be excited in the first excitation step and the laser ionization efficiency is reduced correspondingly.

The efficiency can be increased by just providing more laser beams to excite the atoms in the other low-lying levels simultaneously [5]. For Al atomic system, the light at 308.22 nm and 309.27 nm is used to excite simultaneously the $2P^\circ_{1/2}$ and $2P^\circ_{3/2}$ states in the experiments, e.g., in Refs. [6,7]. This is a routinely used approach for hot-cavity RILIS application, such as at ISOLDE, for the elements Ga, In, Ni, Sn, etc., depending on their energy level structure and the availability of the laser lights for the resonant excitation. For the element Sn, for instance, three resonant transitions at 286 nm [8], 301 nm [5,9,10], and 317 nm [11] are required to excite the atoms in the ground states $5s^25p^2\ ^3P_0$,$^3P_1$, and $^3P_2$ to the first excited state $5s^25p6s\ ^3P^\circ_1$, respectively, for the first excitation step in the three-step laser ionization scheme (see Fig. 1). So far, the overall ionization efficiencies of up to 22% for Sn have been reported based on a hot-cavity type laser ion source using a three-step ($\{\omega_1|\omega_3|\omega_4\}$) ionization scheme [8].

In this work, we have conducted a laser spectroscopic study of the efficient ionization of Sn atom in the four-color laser ionization scheme as shown in Fig. 1(a). In this scheme, two transitions as the first excitation step in a three step resonant ionization scheme for Sn were used together to excite the thermal populations of the two grounds states $^3P_0$ ($\lambda_1 = 286$ nm) and $^3P_1$ ($\lambda_2 = 301$ nm) to improve the ionization efficiency. The two lower ground levels $^3P_0$ and $^3P_1$ are almost evenly populated at around 2200 K as shown in Fig. 2. Thus, in this four-color

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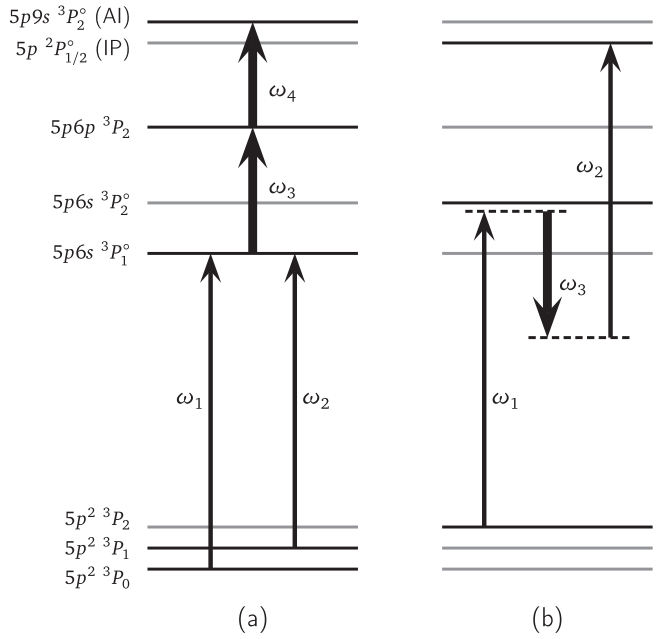

Fig. 1. Laser ionization schemes for Sn used in the experiment. The energy of each level is listed in Table 1. (a) Four-color laser ionization of Sn consists of two three-step laser ionization processes with ω_1-ω_3-ω_4 and ω_2-ω_3-ω_4 photoionization. (b) The N-type ionization scheme with four states can be characterized by two weak UV beams (ω_1 and ω_2) and one strong IR beam (ω_3) resulting in the photoionization from the third ground state $^3P_2$ to the ionization potential (IP).

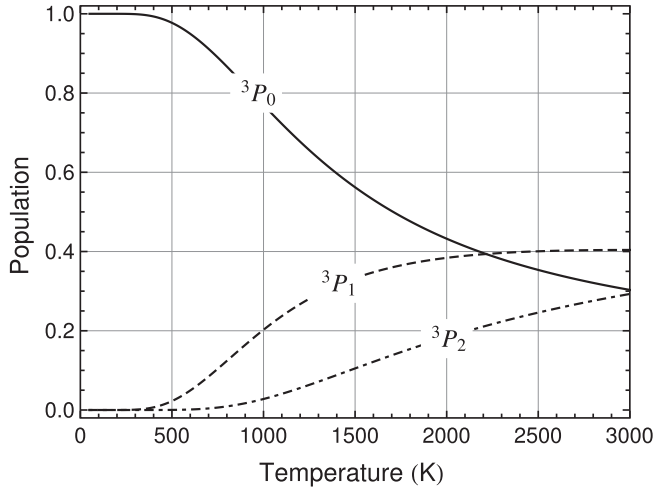

Fig. 2. Population ratios of the ground states ($^3P_0$ (solid line), $^3P_1$ (dashed line), and $^3P_2$ (dot-dashed line)) of Sn as a function of temperature.

laser ionization scheme, one can expect a significant improvement over the laser ionization efficiency at high temperature. Furthermore, we experimentally demonstrate the effect of a three-photon ionization in a four-level scheme that involves the three-photon Raman transitions via a virtual level as shown in Fig. 1(b), which additionally allows the ionization of the atoms in the third ground state $^3P_2$.

2. Experimental set-up

A highly selective laser ion source based on Ti:Sapphire (Ti:Sa) lasers has been developed in the ISOL facility of the Rare Isotope Science Project (RISP)/IBS. In order to facilitate the work on the development of ion sources and atomic ionization schemes, an off-line ion source test facility has been established. This laboratory is capable of providing extensive atomic spectroscopic data to study various laser ionization

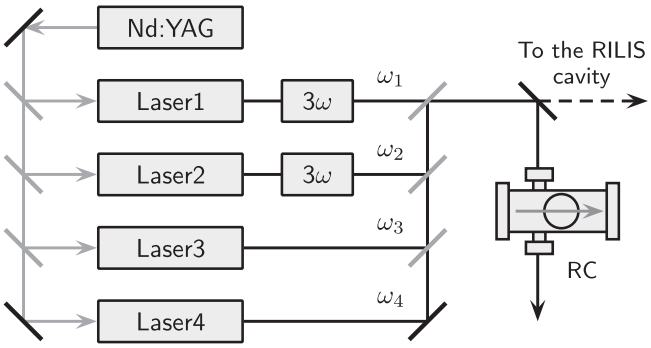

Fig. 3. Schematic diagram of the laser system along with a reference cell (RC). The laser beam direction is perpendicular to the direction of the atomic beam. The gray arrow in the RC indicates the atomic beam produced from an oven.

schemes, which is required for the improvement in ionization efficiency of the laser ion source.

2.1. Laser system

A tunable laser system consisting of four Ti:Sa lasers pumped by a high repetition rate Q-switched Nd:YAG laser has been set up for application in the laser ion source development for RISP (Fig. 3). The pump laser is a frequency-doubled Nd:YAG laser from Lee Laser, which operates at 10 kHz repetition rate with a maximum of 100 W average output power at 532 nm.

The Ti:Sa laser system can generate narrow-band (~5 GHz) tunable laser radiation in the 700–1000 nm (fundamental) and 350–500 nm (frequency doubling) spectral regions. With third harmonic generation capabilities, the beams near UV region (λ_1 = 286 nm and λ_2 = 301 nm) were generated for the first step excitation transition. With an output power of approximately 3 W in the fundamental, ~30 mW in the third harmonic was available for the spectroscopy. A small fraction of the Ti:Sa laser beam was transported to a fiber coupler to allow simultaneous monitoring of timing and wavelength. All laser beams were spatially overlapped and transported to the reference cell or the hot-cavity in the test facility.

2.2. Reference cell setup for laser spectroscopy

A compact atomic beam reference cell (RC) [12] designed by Kron at Mainz University was used in this research. An electrically heated graphite oven can achieve temperatures up to 1700 K which were measured with a pyrometer. Measurements were performed in a crossed-beam geometry as illustrated in Fig. 3. A graphite oven placed in the RC was filled with metallic tin powder and heated electrothermally by a current of ~ 14 A to a temperature of about 1500 K, where the pressure in the RC was 10^{-6} mbar. All laser beams were overlapped and sent through the RC perpendicular to the atomic beam. A pinhole with a radius of 1 mm was placed just before the RC and the laser power of each laser was measured right after the RC. The atoms passing through an aperture were photoionized shortly after by the resonant laser radiation in a transversal geometry and the resonantly ionized atoms were then guided by ion-optical elements into the secondary electron multiplier (SEM).

The Sn laser excitation scheme involves a three-step resonant excitation from the ground states to the auto-ionizing state (AI). As represented in Fig. 2, the thermal populations of Sn are calculated to be 64%, 29%, and 7% for the $^3P_0$,$^3P_1$, and $^3P_2$ states, respectively, assuming a pure Boltzmann distribution within the oven at about 1500 K. The laser light at 286 nm (ω_1) excites the atoms in the lowest ground state ($^3P_0$) and the laser light at 301 nm (ω_2) was added to increase the ionization efficiency by exciting the atoms in the upper ground state ($^3P_1$) as shown in Fig. 1(a). Resonant photoionization was done with two