Selective photoionization of the ytterbium atom by coherent two-photon excitation

Hyunmin Park and Jongmin Lee
Laboratory for Quantum Optics, Korea Atomic Energy Research Institute, P.O. Box 105, Yusong, Taejon 305-600, Korea

Jai-Hyung Lee and Joon-Sung Chang
Department of Physics, Seoul National University, Seoul 151-742, Korea
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Highly selective photoionization of the ytterbium atom by coherent two-photon excitation was demonstrated experimentally. The Yb atom was excited by two single-mode lasers and ionized by a time-delayed broadband laser. The isotope ratio of the ions was analyzed by a time-of-flight mass spectrometer. The ion yield and the selectivity were recorded as a function of detuning of the first exciting laser. The maximum of ion yield was observed when the laser was detuned from resonance. The selectivity of ^{168}Yb was increased higher than 20 000 when the laser was blue-detuned to the most efficient position. Also, numerical analysis was performed to explain the experimental results.

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I. INTRODUCTION

The selective photoionization of an atom by a stepwise excitation has been of interest for the last few decades and a number of experimental results as well as theoretical ones on this topic have been reported [1–4]. The usual scheme of the selective photoionization consists of the excitation of a particular isotope from the ground state to an upper excited state by narrow-band lasers and the subsequent ionization of the excited atom by another laser. The isotope selectivity in the multistep excitation is obtained by multiplication of the selectivity of each excitation step which is mainly determined by isotope shift and bandwidth of the laser.

It was predicted by Diels [5] that an isotope can be excited efficiently and selectively by using a π pulse [6] in each excitation step. Nontarget isotopes have small transition amplitudes due to isotope shift when the pulse area of the laser for the target isotope is π. Although there are a number of reports concerning the efficient population transfer in a three-level system [7–10], few papers deal with selectivity and efficiency together. Recently, Choe *et al.* [11] suggested a principle of the selective and effective excitation of a three-level medium by extending Diels' idea. They showed analytically that with special detunings for a given set of the atom-field interaction all the atomic population can be inverted into the third level. The detunings which give the maximum population transfer were found to be near two-photon resonance and depend on the energies and the pulse durations of lasers as well as the dipole moments of the atomic transitions. Further, they showed that since the bandwidth of the excitation process is very narrow, the high selectivity of the excitation is expected. From their studies, it was known that the excitation process of a three-level medium with this optimal detunings enhances the efficiency of photoionization of target isotopes in the medium with high selectivity.

In this paper we experimentally investigate the principle in Ref. [11] by observing the ion yield and the selectivity of ^{168}Yb versus the frequencies of two excitation lasers near two-photon resonance. ^{168}Yb has the natural abundance of 0.135% among seven Yb isotopes. In addition, numerical analysis by density-matrix equations is presented to show the possibility of efficient population transfer and high selectivity of ^{168}Yb by coherent two-photon excitation.

II. NUMERICAL ANALYSIS

Figure 1 shows an ionization scheme [12] of ^{168}Yb, which was used in our experiment. The ground state $|0\rangle$ is coherently coupled to the second excited state $|2\rangle$ through the intermediate state $|1\rangle$ by two single-mode lasers. The excited Yb atom is ionized through an autoionization state $|i\rangle$ by an ionizing laser with sufficient time delay. The population transfer from $|0\rangle$ to $|2\rangle$ is probed by a time-delayed ionizing laser.

The analytical solutions for the maximum population transfer in Ref. [11] considers the ideal case of monochromatic laser and square temporal profile. Actually, the spectrum from a single-mode laser has some phase diffusion bandwidth [13] and the realistic temporal profile of the laser

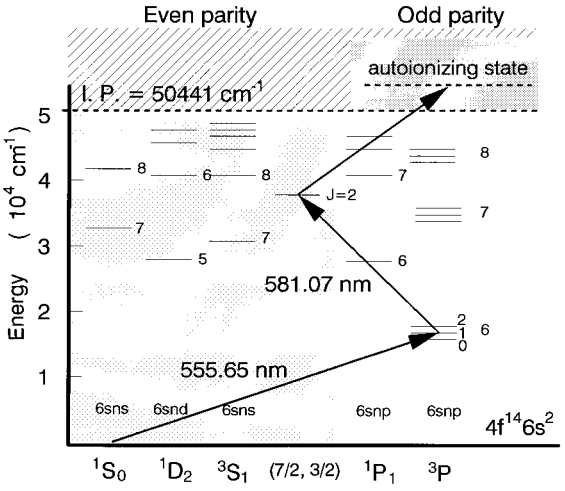

FIG. 1. Energy-level diagram and photoionization scheme of ytterbium. I.P. denotes the ionization potential.

is Gaussian-shaped, so that it is difficult to obtain analytical expressions about the population transfer. Generally, the atomic population dynamics is described by the following density-matrix equations [14]:

$$\frac{d}{dt}\rho_{01}=\left[-i\Delta_a-\left(\frac{1}{2\tau_1}+2b_a\frac{\beta_a^2}{\Delta_a^2+\beta_a^2}\right)\right]\rho_{01}-i\frac{\Omega_b}{2}\rho_{02}+i\frac{\Omega_a}{2}(\rho_{11}-\rho_{00}), \quad (1)$$

$$\frac{d}{dt}\rho_{12}=\left[-i\Delta_b-\left(\frac{1}{2\tau_1}+\frac{1}{2\tau_2}\right)+2b_b\frac{\beta_b^2}{\Delta_b^2+\beta_b^2}\right]\rho_{12}+i\frac{\Omega_a}{2}\rho_{02}+i\frac{\Omega_b}{2}(\rho_{22}-\rho_{11}), \quad (2)$$

$$\frac{d}{dt}\rho_{02}=\left[-i(\Delta_a+\Delta_b)-\frac{1}{2\tau_2}+2b_a\frac{\beta_a^2}{\Delta_a^2+\beta_a^2}+2b_b\frac{\beta_b^2}{\Delta_b^2+\beta_b^2}\right]\rho_{02}-i\frac{\Omega_b}{2}\rho_{01}+i\frac{\Omega_a}{2}\rho_{12}, \quad (3)$$

$$\frac{d}{dt}\rho_{00}=\frac{1}{\tau_1}\rho_{11}-\frac{i}{2}(\Omega_a\rho_{01}-\text{c.c.}), \quad (4)$$

$$\frac{d}{dt}\rho_{11}=-\frac{1}{\tau_1}\rho_{11}+\frac{1}{\tau_2}\rho_{22}+\frac{i}{2}(\Omega_a\rho_{01}-\text{c.c.})-\frac{i}{2}(\Omega_b\rho_{12}-\text{c.c.}), \quad (5)$$

$$\frac{d}{dt}\rho_{22}=-\frac{1}{\tau_2}\rho_{22}+\frac{i}{2}(\Omega_b\rho_{12}-\text{c.c.}). \quad (6)$$

The Rabi frequencies Ω_a and Ω_b are defined as $\Omega_{a,b}=\mu_{a,b}E_{a,b}/\hbar$, where $E_{a,b}$ are electric-field amplitudes of lasers and $\mu_{a,b}$ are the electric dipole moments of the first and the second transitions. The detuning frequencies Δ_a and Δ_b are defined as $\Delta_a=\omega_a-\omega_{01}$ and $\Delta_b=\omega_b-\omega_{12}$, where ω_{01} and ω_{02} are the resonant frequencies of each transition and ω_a and ω_b are the laser frequencies. $1/\tau_1$ and $1/\tau_2$ are the spontaneous emission rates.

Equations (1)–(3) describe the time evolution of the atomic coherence, ρ_{01}, ρ_{12}, and ρ_{02}. Additional damping terms in these equations describe the destruction of the atomic coherence due to the phase fluctuation of the lasers. The factor of $2b_a$ and $2b_b$ arise from the bandwidth of the laser. The cutoff parameters β_a and β_b stem from the non-Lorentzian nature of the laser line shape [13]. For $\Delta\ll\beta$, these damping terms reduce to $2b_a$ and $2b_b$ while for $\Delta\gg\beta$, the bandwidth-dependent terms disappear. It means that a laser with a spectrum falling off faster than a Lorentzian appears monochromatic to the atom for large detunings [13]. Equations (4)–(6) describe the time evolution of the populations of $|0\rangle$, $|1\rangle$, and $|2\rangle$ states, respectively.

The first and the second laser pulses are supposed to have Gaussian temporal profiles with full width at half maximum (FWHM) of 5 ns and the energy densities of 50 and 120 μJ/cm^2, respectively, equal to experimental conditions. The electric dipole moments are known to be $\mu_a=2.7\times10^{-30}$ C m and $\mu_b=4.3\times10^{-31}$ C m. The peak values of the Rabi frequencies, Ω_a and Ω_b are 10.5 and 1.08 rad/nsec in this condition. $1/\tau_1$ and $1/\tau_2$ are neglected because the pulse duration is very short compared with the lifetime of each level. The phase diffusion bandwidths $2b_a$ and $2b_b$ were set to 80 MHz in order to incorporate the laser bandwidth effect.

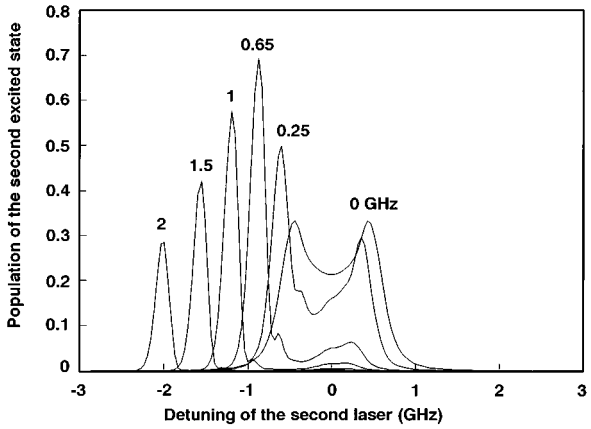

FIG. 2. Numerical results for the population of the second excited state. The detuning of the first laser was fixed at 0, 0.25, 0.65, 1, 1.5, and 2 GHz while the detuning of the second laser was scanned.

Figure 2 shows calculated results for the population of state $|2\rangle$ when the first laser is kept at a detuned frequency from the resonance and the second laser frequency is scanned. When the first laser is tuned to the resonance, the Autler-Townes splitting is observed. As the detuning of the first laser increases, the peak value of the population of state $|2\rangle$ increases and reaches a maximum when the detuning is 0.65 GHz. Further increase of the detuning decreases the population. At the detuning of 0.65 GHz, the bandwidth of excitation process is about ~200 MHz, which is exceptionally narrow and even narrower than the laser bandwidth. Therefore, the isotopes which have larger isotope shift than the bandwidth have very small excitation probabilities and the isotope selectivity of ^{168}Yb much higher than the case of two-step excitation is expected. The result shows that the largest excitation efficiency of 70% is achieved by the detuning of the laser from resonance and that the amount of detuning is determined by the intensities of the lasers. The optimal detuning and the excitation efficiency predicted from analytical solutions of Ref. [11] are 0.4 GHz and 100%, respectively. There are some differences because the analytical solutions were obtained for the case of monochromatic and square laser profile.

III. EXPERIMENTAL SETUP

The experimental setup for the selective ionization of ^{168}Yb is schematically shown in Fig. 3. As shown in the figure, the setup consists of a laser system, an apparatus for ion mass analysis, and a signal processing system. For the coherent excitation we used two single-mode dye lasers (Lumonics, HyperDYE-SLM) pumped by a frequency-doubled Nd:YAG (neodymium-doped yttrium-aluminum-garnet) laser (Lumonics, HY-750), giving a Gaussian temporal profile of 5 ns pulsewidth and 300 MHz linewidth. Ionization was made

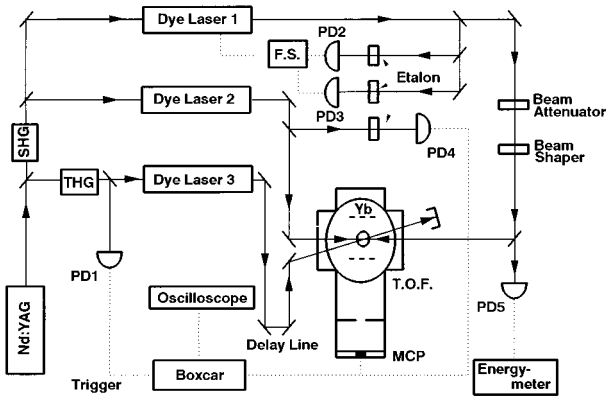

FIG. 3. Schematic diagram of the experimental setup. (SHG: second-harmonic generation; THG: third-harmonic generation; F.S.: frequency stabilizer; PD: photodetectors; MCP: microchannel plate; T.O.F.: time-of-flight mass spectrometer).

by a broadband dye laser (Lumonics, HD-500) pumped by the frequency-tripled Nd:YAG laser. The pulsewidth and the linewidth of the dye laser were 5 ns and 1.5 GHz, respectively. The frequency of the first excitation laser was stabilized within 100 MHz for a few hours by a feedback system which consisted of a dual photodiode, a solid étalon [free spectral range (FSR): 7 GHz], and a piezoelectric transducer (PZT) attached to the oscillator mirror of the laser [15]. The second single-mode laser was scanned in the range of several tens of GHz without mode hopping. The frequencies of each laser were calibrated and monitored with an accuracy of 100 MHz by recording the fringe from a Fabry-Perot etalon (FSR: 7 GHz). In the experiment, the first dye laser was tuned close to the $6s^2\ {}^1S_0-6s6p\ {}^3P_1$ resonance transition of ^{168}Yb at 555.648 nm. The second dye laser was scanned across the $6s6p\ {}^3P_1-4f^{13}6s^26p\ (\frac{7}{2},\frac{3}{2})_2$ transition of ^{168}Yb at 581.068 nm. The ionizing laser was tuned to the wavelength of 582.782 nm, corresponding to an autoionization transition.

After spatial filtering by a lens and an aperture, two excitation lasers were arranged to counterpropagate simultaneously through the Yb atomic beam with uniform intensity distribution. Two lasers were linearly polarized along the same direction. The broadband photoionization laser was made to co-propagate with the first laser with time delay of about 10 ns, i.e., long compared with the pulse duration, but short compared with the lifetime of the second excited level. Hence there was no pulse overlap. The energies of each laser pulses were controlled by two linear polarizer and monitored by a calibrated energy meter.

The natural solid Yb atom placed in a tantalum boat was heated resistively in a vacuum chamber. The generated Yb atomic beam was collimated by two slits and directed perpendicular to laser beam propagation direction to reduce Doppler broadening to 30 MHz. The Yb atoms at interaction zone were ionized between the acceleration plates of a time-of-flight mass spectrometer (TOFMS). Yb ions were detected at the end of the flight tube by a microchannel plate (MCP). The ion signal was integrated by a boxcar averager (Stanford, SR250) and the mass spectrum was recorded by a digitizing oscilloscope (Hewlett Packard, 54512B). Finally, they were transferred to a personal computer for analysis.

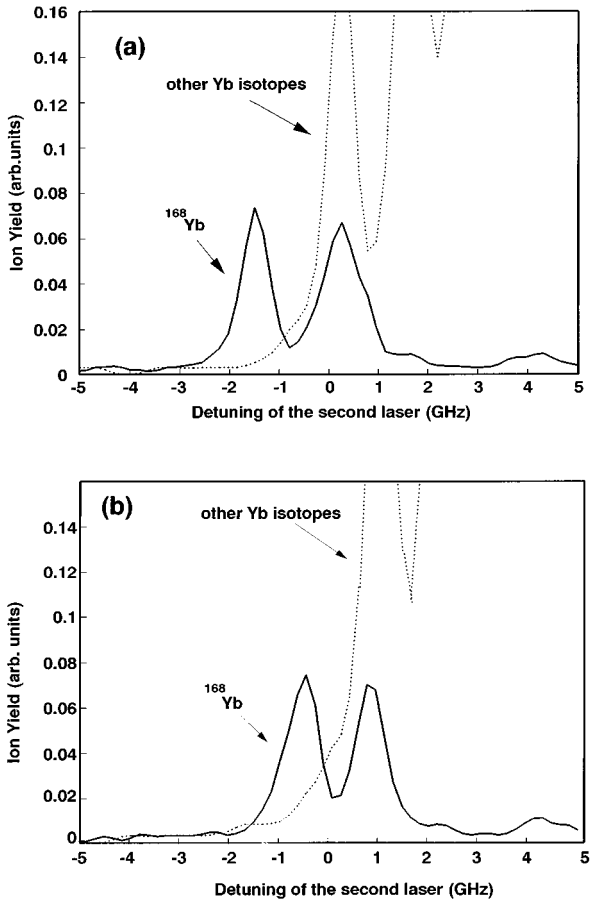

FIG. 4. Experimental ion yields of ^{168}Yb and other Yb isotopes as a function of the detuning of the second laser when the detuning of the first laser was fixed (a) at 0.65 GHz and (b) at 0 GHz.

IV. RESULTS AND DISCUSSION

Figures 4(a) and 4(b) show the ion yields of ^{168}Yb and other Yb isotopes when the detuning of the second laser (Δ_b) was varied and the detuning of the first excitation laser (Δ_a) was fixed at 0.65 and 0 GHz, respectively. For these experiments, the energy densities of the first and the second laser were fixed at 50 and 120 μJcm^2, respectively. The ion yields of ^{168}Yb and other Yb isotopes were taken at the same time while the second laser was scanned.

Figure 4(a) shows that at $\Delta_b=-1.5$ GHz the ion yield of ^{168}Yb is maximum and the selectivity of ^{168}Yb is very high. The results show obviously the increase of the selectivity and the ion yield by coherent two-photon excitation, as expected in numerical analysis. But the width ($\sim$500 MHz) of the excitation at the peak is larger than numerical calculation ($\sim$200 MHz), which is originated from the frequency and the energy fluctuation of the laser. In addition, the mass spectrum of Yb recorded by a digitizing oscilloscope at $\Delta_a=0.65$ GHz and $\Delta_b=-1.5$ GHz is shown in Fig. 5. The abundance of ^{168}Yb, 0.135% in nature, becomes nearly 100% after coherent excitation, which corresponds to selectivity higher than 20 000. The selectivity is defined as follows [16]:

$$\text{Selectivity}\equiv\frac{X^f_{168}}{1-X^f_{168}}\Big/\frac{X^i_{168}}{1-X^i_{168}},$$

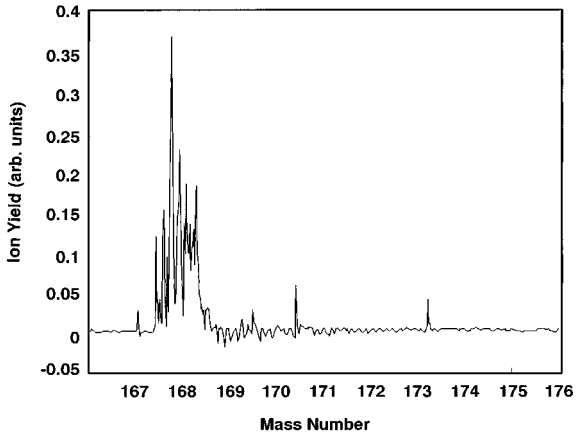

FIG. 5. Mass spectrum of Yb ions detected by MCP and oscilloscope when the ion yield of ^{168}Yb was maximum in Fig. 4(a).

where X^f_{168} is the abundance of ^{168}Yb after selective excitation and X^i_{168} is the natural abundance of ^{168}Yb 0.001 35.

The selectivity is enhanced more than 300 times compared with our previous experiment of two-step excitation [17]. Meanwhile, there is another peak at $\Delta_b=0$ GHz in Fig. 4(a). This can be interpreted as due to the optical Autler-Townes effect, which is caused by the simultaneous excitation of two lasers [18]. This Autler-Townes effect appears obviously in Fig. 4(b) when the first laser is tuned to the resonance. The amount of splitting between two symmetric peaks in the figure agrees well with the result by the numerical calculation including the laser bandwidth effect.

Comparing Figs. 4(a) and 4(b), we can see that the selectivity of ^{168}Yb in the case of $\Delta_a=0.65$ GHz is much higher than that in case of $\Delta_a=0$ GHz. This represents that the selectivity is much higher when the frequency of the first laser is optimally detuned from resonance rather than on-resonance. As mentioned in numerical analysis, this increase of the selectivity and the ion yield of ^{168}Yb at a detuned frequency is caused by the efficient population transfer by the coherent excitation.

Figure 6 shows the experimental results for the selectivity and the ion yield of ^{168}Yb when the detuning of the first laser was varied. The energies of two lasers are the same as before. The ion yield of ^{168}Yb has a maximum at $\Delta_a=0.65$ GHz and has a small dip at resonance. As the detuning moves from the red to the blue side the selectivity of ^{168}Yb increases very steeply. This was originated from absorption spectra of Yb isotopes. The absorption spectra of Yb isotopes for the first and the second excitation step are shown in Fig. 7. The isotope shift of ^{168}Yb from ^{171}Yb in the second transition is -1.6 GHz, while the isotope shift of ^{168}Yb from ^{171}Yb in the first transition is -0.15 GHz. When the frequency of the first excitation laser is red-detuned from the resonance frequency of ^{168}Yb, the second excitation laser must be blue-detuned to get the two-photon transition. Then the coherent excitation spectrum of ^{168}Yb is partially overlapped to the two-step excitation spectrum of ^{171}Yb. Therefore, the selectivity of ^{168}Yb in case of red detuning is lower than that in case of blue detuning.

The dotted line and the solid line in Fig. 6 represent the numerical results of the selectivity and the ion yield of ^{168}Yb, respectively. Reasonable agreement between experimental

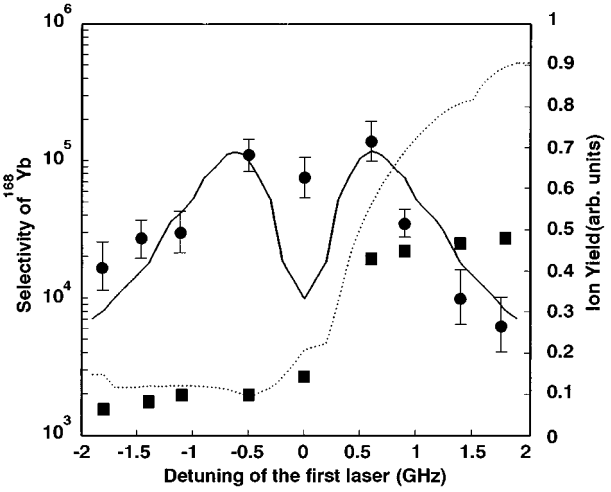

FIG. 6. Experimental and numerical results for the selectivity and ion yield of ^{168}Yb as a function of the detuning of the first laser. —: ion yield (numerical), ●: ion yield (experimental), ·····: selectivity (numerical), ■: selectivity (experimental).

and numerical results is obtained qualitatively, although there are some quantitative differences in the blue-detuned region. In this region it is quite difficult to measure the selectivity because extremely low ion signals from the other isotopes are mixed with noise. In the future we hope that the discrepancy would be removed by increasing the sensitivity of MCP at low signal level.

V. CONCLUSION

An experiment concerning the selective excitation of ^{168}Yb with two single-mode lasers and the subsequent ionization by the other broadband laser was conducted. Also, experimental results were compared with numerical ones.

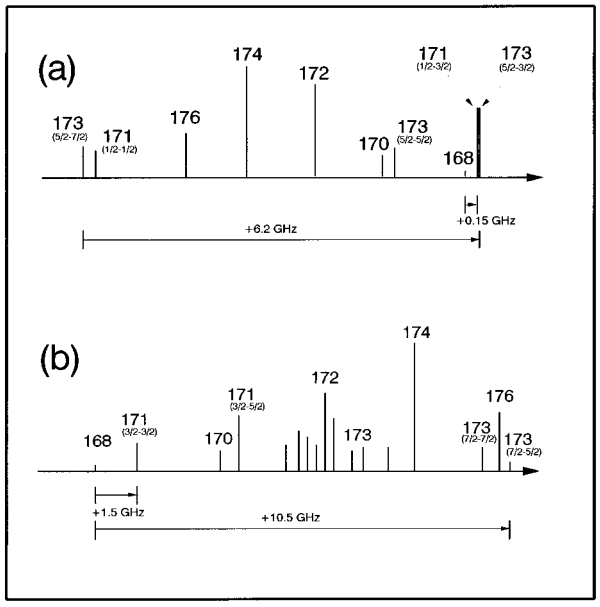

FIG. 7. The absorption spectra of ytterbium (a) for the first transition line: $6s^2\ ^1S_0-6s6p\ ^3P_1$ and (b) for the second transition line: $6s6p\ ^3P_1-4f^{13}6s^26p\ (\frac{7}{2},\frac{3}{2})_2$.

When the first exciting laser was blue-detuned from the resonance of ^{168}Yb by 0.65 GHz, the ion yield became maximum and the selectivity of ^{168}Yb higher than 20 000 was obtained under the condition of this experiment. We can conclude that utilization of coherent two-photon excitation for high ionization selectivity and large ionization yield is most efficient if the detuning of each laser can be optimized from the resonance line according to the laser energies and isotope shift structures.

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