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Effect of polarization spectroscopy in multistep excitation and ionization of atoms on ionization efficiency

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Abstract

In this paper, the seventeen real motion equation of atoms were shown for the first time in considering the polarization choices of laser lights. The importance of polarization spectroscopy in the determination of ionization yields of multi-step excitation and ionization of atoms (MSEIA) was shown.

Key words　polarization spectroscopy,　ionization efficiency

The interaction of multilevel atoms with laser lights has been studied for tens of years[1,2], but the polarization spectroscopy in multistep excitation and ionization of atoms (MSEIA) is rarely concerned. So far only a few papers were devoted to this topic[3,4,5,6]. In fact, polarization choices of laser lights are of significant importance in MSEIA as well as in laser cooling and trapping of atoms[7]. For example, in terms of the "bottle neck effect", which means that the selected good quantum number J of each level decreases step by step, for instance for gadolinium $J=2\rightarrow2\rightarrow1\rightarrow0$[4], the odd and even isotopes may be separated in products. By making use of different polarization match of laser lights and complex motion equations of atoms, the value of good quantum number of autoionization level J may be determined[6].

We suppose an optimized ionization efficiency, which is very important in laser isotope separation, may be reached easily by using real motion equations of atoms and comparing different polarization chices.

We consider a three colour three photon spectral path, which means a four-level atomic system is involved, and we use L,S,E,C,O,R,T,V,W,X to denote the following polarization match:

	light 1	light2	light 3		light 1	light 2	light 3
L	π	π	π	S	σ^+	σ^+	σ^+
E	σ^-	σ^+	σ^+	C	σ^+	σ^-	σ^+
O	σ^+	σ^+	σ^-	R	π	σ^+	σ^+
T	π	σ^+	σ^-	V	π	σ^-	σ^-
W	π	σ^-	σ^+	X	σ^-	σ^-	σ^-

The seventeen real motion equations of four-level atoms derived form Schrodinger equation are

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$$
\begin{aligned}
&\dot{\rho}_{11} = \Omega_1^m/2R_{12} \qquad \dot{\rho}_{22} = -\,\Omega_1^m/2R_{12} + \Omega_2^m/2R_{23} - \gamma_2\rho_{22} \\
&\dot{\rho}_{33} = -\,\Omega_2^m/2R_{23} + \Omega_3^m/2R_{34} - \gamma_3\rho_{33} \\
&\dot{\rho}_{44} = -\,\Omega_3^m/2R_{34} - \gamma_4\rho_{44} \\
&-\dot{R}_{12} = -\,(\Delta_1 - \Delta_2)Q_{12} - \Omega_1^m(\rho_{22} - \rho_{11}) + \Omega_2^m/2 + \gamma_2/2R_{12} \\
&\dot{Q}_{12} = -\,(\Delta_1 - \Delta_2)R_{12} + \Omega_2^m/2R_{13} - \gamma_2/2Q_{12} \\
&-\dot{R}_{13} = -\,(\Delta_1 - \Delta_3)Q_{13} - \Omega_1^m/2Q_{23} + \Omega_2^m/2Q_{12} + \Omega_3^m/2Q_{14} + \gamma_3/2R_{13} \\
&\dot{Q}_{13} = -\,(\Delta_1 - \Delta_3)R_{13} - \Omega_1^m/2R_{23} + \Omega_2^m/2R_{12} + \Omega_3^m/2R_{14} - \gamma_3/2Q_{13} \\
&-\dot{R}_{14} = -\,(\Delta_1 - \Delta_4)Q_{14} - \Omega_1^m/2Q_{24} + \Omega_3^m/2Q_{13} + \gamma_4/2R_{14} \\
&\dot{Q}_{14} = -\,(\Delta_1 - \Delta_4)R_{14} - \Omega_1^m/2R_{24} + \Omega_3^m/2R_{13} - \gamma_4/2Q_{14} \\
&-\dot{R}_{23} = -\,(\Delta_2 - \Delta_3)Q_{23} - \Omega_1^m/2Q_{13} + \Omega_3^m/2Q_{24} + (\gamma_2 + \gamma_3)/2R_{23} - \Omega_2^m(\rho_{33} - \rho_{22}) \\
&\dot{Q}_{23} = -\,(\Delta_2 - \Delta_3)R_{23} - \Omega_1^m/2R_{13} + \Omega_3^m/2R_{24} - (\gamma_2 + \gamma_3)/2Q_{23} \\
&-\dot{R}_{24} = -\,(\Delta_2 - \Delta_4)Q_{24} - \Omega_1^m/2Q_{14} - \Omega_2^m/2Q_{34} + \Omega_3^m/2Q_{23} + (\gamma_2 + \gamma_4)/2R_{24} \\
&\dot{Q}_{24} = -\,(\Delta_2 - \Delta_4)R_{24} - \Omega_1^m/2R_{14} - \Omega_2^m/2R_{34} + \Omega_3^m/2R_{23} - (\gamma_2 + \gamma_4)/2Q_{24} \\
&-\dot{R}_{34} = -\,(\Delta_3 - \Delta_4)Q_{34} - \Omega_2^m/2Q_{24} + (\gamma_3 + \gamma_4)/2R_{34} - \Omega_3^m(\rho_{44} - \rho_{33}) \\
&\dot{Q}_{34} = -\,(\Delta_3 - \Delta_4)R_{34} - \Omega_2^m/2R_{24} - (\gamma_3 + \gamma_4)/2Q_{34} \\
&\dot{\rho}_i^m = \gamma_4 \cdot \rho_{44} \qquad\qquad \rho_i = \frac{1}{2J_1 + 1}\sum_{m=-J_1}^{J_1} \rho_i^m
\end{aligned}
\tag{1}
$$

where $\Omega_n^m = \dfrac{2}{\hbar(-1)^{J_{n+1}-m_{n+1}}}\begin{pmatrix} J_{n+1} & 1 & J_n \\ -m_{n+1} & q_n & m_n \end{pmatrix} d_{n,n+1}E_n$, $m_{n+1} = m_n + q_n$ and the polarization properties of the laser lights are included in the 3-J symbol which may be calculated by using Wigner-Eckart theorem. Ω_n^m is the Rabi frequency, $d_{n,n+1}$ is the dipole moment, E_n is the amplitude of nth laser light ($n = 1, 2, 3$), $\Delta_1 = 0$ and Δ_i is the detunning by which an individual transition line is out of resonance with appropriate laser light, ρ_{ii} is the population probability of atoms in level i and ρ_i is the ionized atoms. The R_{ij} and O_{ij} are the several transition-operator amplitudes, in-phase and in-quadrature, respectively, with the three laser light. $q_n = -1, 0, +1$ denotes the polarization properties of laser lights. γ_2 and γ_3 dentoe the spontaneous radiation rates of level 2 and 3 respectively, and γ_4 denotes the autoionization rate of level 4. Assuming $\Omega_n^0 = (d_{n,n+1}E_n)/2\hbar$, we can estimate its value in terms of reference[5,8] when the bandwidths of laser lights and the Doppler broadening of atomic beam are omitted. In equation (1), the ac Stark shifts of each level have been omitted[8]. The special choices of the polarization guarantee the magnetic components of each level decouple from each other even though there is no magnetic field applied. We select the typical spectral routes of U^{238} atom three-step photoionization,

$$(0\ \mathrm{cm}^{-1},\ J = 6) \rightarrow (17362\ \mathrm{cm}^{-1},\ J = 6) \rightarrow (34659\ \mathrm{cm}^{-1}, J = 7) \rightarrow (50422\ \mathrm{cm}^{-1},\ J = 6,7,8)$$

$$(620\ \mathrm{cm}^{-1}, J = 5) \rightarrow (16505\ \mathrm{cm}^{-1}, J = 6) \rightarrow (33119\ \mathrm{cm}^{-1}, J = 7) \rightarrow (J = 6,7,8)$$

$$(999\ \mathrm{cm}^{-1},\ J = 5) \rightarrow (18084\ \mathrm{cm}^{-1},\ J = 5) \rightarrow (34719\ \mathrm{cm}^{-1}, J = 4) \rightarrow (J = 3,4,5)$$

Numerical computation results are shown in Fig. 1(a), Fig. 1(b) and Fig. 1(c) and the experimental result is shown in Fig. 1 in comparison with the theoretical result. Our experimental results are similar to those in reference[6] except that of the polarization match L. This differnce may result from different experimental conditions and from experimental error. It is obvious that the polarization choices of lasers are closely related to the ionization yields. The polarization match should be considered in the general design of laser isotope separation.

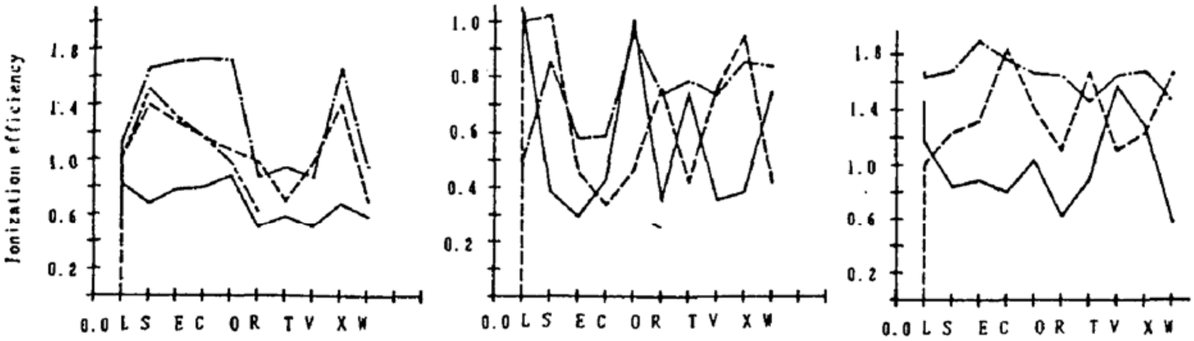

Fig. 1 The normalized ionization efficiency versus different polarization match

(a) U^{238} ($\Omega_1^0 = 3 \times 10^9 S^{-1}$, $\Omega_2^0 = \Omega_3^0 = \gamma_4 = 7 \times 10^9 S^{-1}$; $\Delta_1 = 0$, $\Delta_4 = 2\Delta_3 = 4\Delta_2 = 0.4$ GHz, $\gamma_2 = 2.5 \times 10^6 S^{-1}$, $\gamma_3 = 7.0 \times 10^6 S^{-1}$, $\tau = 20$ns

(b) U^{238} ($\Omega_1^0 = \Omega_2^0 = \Omega_3^0 = \gamma_4 = 7 \times 10^9 S^{-1}$, $\Delta_1 = 0$, $\Delta_4 = 2\Delta_3 = 4\Delta_2 = 0.4$ GHz, $\gamma_2 = 7.0 \times 10^6 S^{-1}$, $\gamma_3 = 2.5 \times 10^7 S^{-1}$, $\tau = 30$ns

(c) G_a^{160}(The conditions are the same as those of (b))

Reference

[1] WU Huang, XU Pinfang, YU Peizeng, Analysis of ionization yields under multilongitudinal-mode laser radiation fields, *Chinese J. Atom. and Molec. Phy.* ,1991,**8**(4):2020～2026

[2] M. V. Smirnov, Coherent effect in a three-level gas medium on exposure to a strong bichromatic pumping field and a weak probe wave, *J. Opt. Soc. Am. B* . ,1992,**9**(12):2171～2178

[3] WU Huang, YAN Donhai, LUO Wanxiang *et al* ., Fluorescence spectra of uranium induced by a linearly polarized laser light in a large magnetic field, *Chinese J. Laser (E. E)* ,1993,**2**,accepted and to be published

[4] R. C. Stern, J. A. Paisner, Atomic vapor laser isotope separation, *Lawrence Livermore National Laboratory*, *UCRL*-93584 (Nov. ,1985),9

[5] P. T. Greenland, Resonant ionization polarization spectroscopy of complex atoms, *J. Phys. B* ,1988,**21**(22):4117～4129

[6] P. T. Greenland, D. N. Travis, D. J. H. Wort, Resonant ionization polarization spectroscopy in unanium, *J. Phys. B* ,1990,**23**(17):2945～2956

[7] C. J. Foot, Laser cooling and trapping of atoms, *Contemporary Phys.* ,1991,**32**(6):369～381

[8] WU Huang, PAN Wenjie, XU Xiangdong *et al* ., Investigation on power broadening of uranium transition, *Chinese J. Laser (E. E)* ,1992,**1**(2):141～147