

Electron spin-polarized alkaline-earth ions produced by multiphoton ionization

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(Received 2 January 2002; accepted 13 May 2002)

We propose two realistic schemes for producing electron spin-polarized alkaline-earth ions. One of the schemes is based on one-photon resonant two-photon ionization via fine structure manifolds of triplet states, while the other is based on one-photon resonant one-photon ionization. For both schemes we find that the ionization must take place from a *triplet state* to produce spin-polarized photoions. Photoions produced from a singlet state are always unpolarized regardless of the laser polarization and a magnetic sublevel from which ionization takes place. We carry out specific analysis for both schemes with Sr atoms, and find 85%–90% and 66% polarizations for the former and the latter schemes, respectively, if the wavelengths and polarizations of lasers are judiciously chosen. In particular, if all the produced photoions are in the ground *s* state, we find an exact one-to-one correspondence between the spin-polarization of *photoions* and ejected *photoelectrons*.

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I. INTRODUCTION

During the last few decades much effort has been made for the development of a spin-polarized electron source.^{1–5} Although polarized electrons are typically produced by photoionization of a gaseous or solid target by circularly polarized radiation, their spin cannot be directly polarized through electric dipole interaction, since it works only on the orbital angular momentum of electrons. That is, it is the spin–orbit interaction that plays an essential role to transfer the polarization of orbital angular momentum to the spin before or upon the photoionization processes. As is well known, the most practical and efficient way to create spin-polarized electrons is to irradiate a GaAs crystal by circularly polarized radiation, by which more than 80% spin polarization has been achieved. One of the main problems of this method as a polarized electron source is that the GaAs crystal can be seriously damaged if an intense laser is used to obtain spin-polarized photoelectrons. In order to overcome this problem, we have recently proposed a new scheme utilizing multiphoton ionization of Xe.⁶ Obviously the scheme proposed in Ref. 6 is damage-free since it employs a gas as a target, and the theoretical results show that more than 90% polarization can be achieved when the laser wavelength is properly chosen. Needless to say, as a polarized-electron source, the use of rare gas rather than alkali-metal^{7,8} or alkaline-earth vapor is by far preferable from a technical point of view, since rare gas is much easier to handle and higher electron current can be obtained simply by increasing the gas pressure.

When we turn our eye to the residual *ions* after photo-

ionization, there is no report which examines a possible scheme for creating electron spin-polarized ions. Recalling that the ground state of singly ionized alkali-metal ions and rare gas ions have a closed p^6 or an open p^5 configuration, respectively, one would immediately notice that these ions are not suitable for the purpose of studying spin-dependent effects. Ions with simple electronic structures such as alkaline-earth ions are much more preferable. Now the question we would like to address is how to produce electron spin-polarized alkali-earth ions. Although one may naively think that the photoion is necessarily polarized if the ejected photoelectron is polarized, it is not *a priori* obvious, since the photoelectron takes away some spin as well as orbital angular momentum. Therefore the degree of spin-polarization of photoions needs to be quantitatively examined for specific cases.

Electron spin-polarized ions can be used to probe various spin-dependent dynamics at a surface^{9,10} and in gas phase collisions, which are of great importance in chemical physics. For the study of spin-dependence of atomic collisions,^{11–15} spin-polarized alkali-metal atoms prepared by optical pumping have often been used as targets. The use of spin-polarized alkaline-earth ions instead of alkali-metal atoms would add another dimension to understanding spin-dependent collision dynamics. As for the ion–molecule reactive collisions, it has been found that the reactive cross sections strongly depend on the spin–orbit states of ions, which are selectively prepared by multi–single-photon ionization together with coincidence techniques.^{16–18} Thus, a method to create spin-polarized ions with *thermal energy* must be established for more detailed study. An obvious but not necessarily the simplest scheme for producing electron spin-polarized alkaline-earth ions is to ionize neutral atoms

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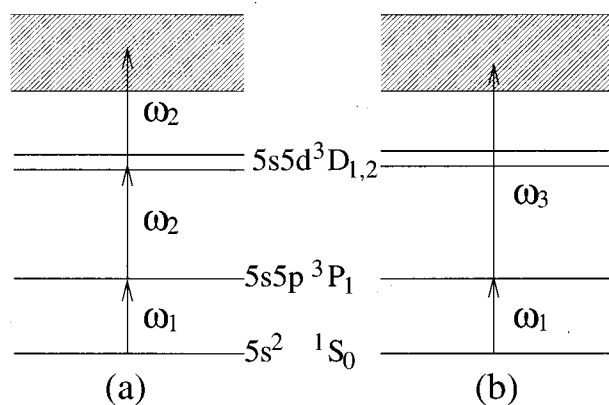


FIG. 1. Level schemes for producing electron spin-polarized alkaline-earth (Sr) ions.

first, and then polarize the ions using optical pumping, as reported by Reihl *et al.*,¹⁹ where they have obtained Sr⁺ beam of a few keV energy with more than 90% polarization. In chemical physics, however, important inelastic and reactive collisions take place at a *thermal energy* of a few eV. An ion beam with thermal energy can be easily produced by making use of photoionization of an effusive neutral atomic beam. Therefore, the question is how to create electron spin-polarized ions through photoionization.

The purpose of this paper is to propose and examine two realistic schemes for producing electron spin-polarized alkaline-earth ions via multiphoton ionization of an effusive neutral atomic beam. One of the schemes is based on one-photon resonant two-photon ionization via fine structure manifolds of triplet states, while the other is based on one-photon resonant one-photon ionization. Which schemes would be more preferable for real applications depends on how much spin polarization and ion yield are needed, and therefore, both schemes have to be compared. We would like to emphasize that the proposed schemes are rather simple, and do not require sophisticated equipment. Furthermore, the photoions produced by our schemes have thermal kinetic energy, which is in a completely different energy range reported in Ref. 19. The electron configuration of the ground state of a neutral alkaline-earth atom is given by $s^2 1S_0$, which means that the total spin quantum number is zero. As we show later in this paper, *photoions from a singlet state cannot be spin-polarized regardless of the polarization of the laser used, and it is essential that the photoionization takes place from a triplet state.* This is because spin-orbit interaction does not occur for a singlet state, as the vector model for spin-orbit interaction suggests.

II. ANALYSIS

In this section we present our analysis of a possible production of electron spin-polarized alkaline-earth ions in the ground $s^2 1S_0$ state. Specifically we examine two schemes for Sr depicted in Figs. 1(a) and 1(b), which we shall call scheme A and scheme B from now on. In both schemes, it is essential to ionize an alkaline-earth atom in the *singlet* ground state via a *triplet* excited state. This implies that a singlet-triplet transition must be introduced in one way or

another before ionization. Recall that a singlet-triplet transition is spin-forbidden in the pure LS coupling. In reality, none of the real atomic states is described by a pure LS coupling, and therefore such a spin-forbidden transition is possible, regardless of its weakness, due to the configuration mixing among states with different spin states. In other words, it is neither orbital angular momentum nor spin angular momentum but total angular momentum that conserves momentum. It should be clear by now that even a state that is labeled as a singlet state cannot be 100% singlet, and always contains small fraction of triplet component and vice versa. For efficient ionization, it is best to induce a singlet-triplet transition from the singlet ground state to the first excited triplet state, since, although it is already much weaker than the singlet-singlet and triplet-triplet transitions, this is the strongest singlet-triplet transition. That is the reason why we employ a singlet-triplet transition in both schemes as a first step toward ionization. According to our estimate, the necessary intensity to saturate the transition $|5s^2 1S_0\rangle \rightarrow |5s5p^3 P_1\rangle$ would be about 0.2 W/cm² for a CW laser (with a laser beam diameter of 1 mm), and 10³ W/cm² for a 10 ns pulsed laser, respectively. Therefore, we expect that $\sim 50\%$ of the population in the ground state can be easily brought up to $|5s5p^3 P_1\rangle$.

In scheme A [Fig. 1(a)], after the first excitation to the triplet state $|5s5p^3 P_1\rangle$, we utilize two-path interference via near-resonant fine structure states, $|5s5d^3 D_{1,2}\rangle$, from which the ionization takes place by absorbing one additional photon. The key of this scheme is to induce quantum-mechanical interference, by the proper choice of laser detuning from the near-resonant fine structure states, in such a way that the alkaline-earth ions in one of the ground-state magnetic sublevels $5s^2 1S_{1/2} m_j = \pm 1/2$ are favorably produced. As we show later on in this paper, there is an exact one-to-one correspondence between the degree of polarization of produced ions and photoelectrons, as one might naively imagine, if all photoions are in the ionic ground $2S_{1/2}$ state.

In scheme B [Fig. 1(b)], we directly ionize atoms in $|5s5p^3 P_1\rangle$. We should note that in both schemes A and B, a particular magnetic sublevel of $|5s5p^3 P_1 M_j\rangle$ ($M_j = 0, \pm 1$) can be selectively excited by the proper choice of the polarization of laser ω_1 . Our main objective of this work is to examine which scheme can produce more polarized ions. Since the different laser polarization and magnetic sublevels involved in the process lead to different spin polarization of photoions and photoelectrons, we must specify both laser polarization and involved magnetic sublevel(s) in both schemes for the quantitative analysis.

An obvious advantage of scheme B over scheme A is that only one photon is needed from $|5s5p^3 P_1\rangle$ to ionize atoms, while in scheme A two photons are required via near-resonant $|5s5d^3 D_{1,2}\rangle$ intermediate states. This makes a huge difference in terms of ionization efficiency, since one-photon ionization cross sections are many orders of magnitude larger than two-photon ionization cross sections. Of course, small ionization cross sections can be always compensated by utilizing intense lasers, and this is not a very serious problem. That is, it is a matter of how much laser intensity we want to pay for how much degree of spin polarization. At this mo-

TABLE I. Deviation from the Lande's interval rule for the lowest $nsnp\ ^3P$ and $nsnd\ ^3D$ states of Ca, Sr, and Ba.

	Ca ($n=4$)	Sr ($n=5$)	Ba ($n=6$)	Pure LS
$nsnp\ ^3P$	2.04	2.11	2.37	2
$nsnd\ ^3D$	1.51	1.53	1.22	1.5

ment, which scheme produces more spin-polarized ions is not *a priori* obvious, and needs to be quantitatively examined.

A. Validity of the use of LS coupling scheme

If we may employ the *LS coupling description with a single term*, the analysis will be greatly simplified, leading to analytical expressions for spin polarization. The validity of such an approximation, however, needs to be examined before proceeding further, since, strictly speaking, none of the pure coupling schemes represents a real atomic structure. For that purpose, we first check the Lande's interval rule, which is given by²⁰

$$E_{\gamma LSJ} - E_{\gamma LSJ-1} \propto J, \quad (1)$$

where L , S , and J represent quantum numbers for the orbital, spin, and total angular momenta, respectively. γ represents all quantum numbers other than L , S , and J . Small deviation of the *experimental* fine structure splittings from the Lande's interval rule suggests that the LS coupling description is valid. Now we examine the Lande's interval rule for the lowest $nsnp\ ^3P$ and $nsnd\ ^3D$ of Ca, Sr, and Ba using experimental energy values. The results are listed in Table I together with the values for the pure LS coupling. Naturally the deviation from the Lande's interval rule becomes larger for the heavier elements. As a second check, we calculate the configuration mixing for those states using a Cowan's atomic structure code.²⁰ For Ca, the wave functions are found to be expanded as

$$|4s4p\ ^3P_1\rangle' \approx 0.994|4s4p\ ^3P_1\rangle, \quad (2)$$

$$|4s4d\ ^3D_1\rangle' \approx 0.96|4s4d\ ^3D_1\rangle + 0.25|3d5s\ ^3D_1\rangle, \quad (3)$$

$$|4s4d\ ^3D_2\rangle' \approx 0.96|4s4d\ ^3D_2\rangle + 0.24|3d5s\ ^3D_2\rangle. \quad (4)$$

For Sr, we obtain

$$|5s5p\ ^3P_1\rangle' \approx 0.99|5s5p\ ^3P_1\rangle + 0.12|4d5p\ ^3P_1\rangle, \quad (5)$$

$$|5s5d\ ^3D_1\rangle' \approx 0.98|5s5d\ ^3D_1\rangle + 0.18|4d6s\ ^3D_1\rangle, \quad (6)$$

$$|5s5d\ ^3D_2\rangle' \approx 0.98|5s5d\ ^3D_2\rangle + 0.18|4d6s\ ^3D_2\rangle, \quad (7)$$

and for Ba

$$|6s6p\ ^3P_1\rangle' \approx 0.99|6s6p\ ^3P_1\rangle - 0.14|5d6p\ ^3P_1\rangle, \quad (8)$$

$$|6s6d\ ^3D_1\rangle' \approx 0.82|6s6d\ ^3D_1\rangle + 0.55|5d7s\ ^3D_1\rangle - 0.13|5d6d\ ^3D_1\rangle, \quad (9)$$

$$|6s6d\ ^3D_2\rangle' \approx 0.84|6s6d\ ^3D_2\rangle + 0.49|5d7s\ ^3D_2\rangle - 0.12|5d6d\ ^3D_2\rangle - 0.15|6s6d\ ^1D_2\rangle, \quad (10)$$

where the primes of the kets on the left-hand sides of the above expressions indicate that they are the leading terms, while the kets on the right-hand sides represent the pure LS states. As one expects, the configuration mixing for Ba is stronger than those for Sr and Ca. However, it is a little surprising that the mixing for Ca $4s4d$ turns out to be larger than that for Sr $5s5d$. This is due to the presence of $3d4p$ and $4p^2$ states lying nearby, which causes strong configuration mixing with Ca $4s4d$.

The outcomes of these two checks indicate that the states labeled as Sr $|5s5p\ ^3P_1\rangle$ and $|5s5d\ ^3D_{1,2}\rangle$ are well represented by the *single LS state*. Before moving onto the quantitative description of the systems, we note that the initial state is considered to be $|5s5p\ ^3P_1\rangle$ rather than $|5s^2\ ^1S_0\rangle$ in the following sections. The reason is that, due to the symmetry, the transition strength from $|5s^2\ ^1S_0\rangle$ to $|5s5p\ ^3P_1\ M_J\rangle$ is exactly the same for all $M_J=0, \pm 1$. Therefore the first excitation process does not have to be taken into account when we calculate the degree of spin polarization.

B. Scheme A: two-path interference via fine structure manifolds

First we examine scheme A which is two-photon ionization from $|5s5p\ ^3P_1\rangle$. To start with, we assume that the initial state is given as $\Lambda_1 = |(n_1 l_1\ n_2 l_2)\ L\ S\ J\ M_J\rangle$, where n_i and l_i are a principal quantum number and orbital angular momentum of the i th ($i=1,2$) electron with L, S, J, M_J being the quantum numbers for the total orbital angular momentum, total spin, total angular momentum, and its projection onto the quantization axis, respectively. Now the single-photon excitation amplitude to the state represented by $\Lambda_2 = |(n'_1 l'_1\ n'_2 l'_2)\ L'\ S'\ J'\ M'_J\rangle$ is given by²⁰

$$\begin{aligned} A(\Lambda_1, \Lambda_2) &= \langle (n'_1 l'_1\ n'_2 l'_2)\ L'\ S'\ J'\ M'_J | r_q | (n_1 l_1\ n_2 l_2)\ L\ S\ J\ M_J \rangle \\ &= \delta_{SS'} (-1)^{J'-M'_J+L'+S'+J+1} \sqrt{(2J+1)(2J'+1)} \\ &\quad \times \begin{pmatrix} J' & 1 & J \\ -M'_J & q & M_J \end{pmatrix} \begin{Bmatrix} L' & S' & J' \\ J & 1 & L \end{Bmatrix} \\ &\quad \times \langle (n'_1 l'_1\ n'_2 l'_2)\ L'\ S' \| r \| (n_1 l_1\ n_2 l_2)\ L\ S \rangle, \quad (11) \end{aligned}$$

where q is 0, 1, and -1 for linearly, right-, and left-circularly polarized light, respectively. Throughout this paper the z axis is taken along the wavevector (polarization vector) of the photon for circularly (linearly) polarized light. Similarly, the single-photon partial ionization amplitude from the state Λ_2 to the continuum state $\Psi = |\mathbf{k}; m''_s j''_1 m''_{j_1}\rangle$, i.e., the continuum with an ionic core $|(n''_1 l''_1 s''_1) j''_1 m''_{j_1}\rangle$ and photoelectron with wave vector \mathbf{k} and spin m''_s , can be derived, using a partial wave expansion, as

$$\begin{aligned}
M(\Lambda_2, \Psi)[m''_{s_2}, j''_1, m''_{j_1}] = & \sum_{\chi} a''_{l_2} m''_{l_2} \delta_{m''_{s_2} m''_{s_2}} \delta_{j''_1 m''_{j_1}} (-1)^{-m''_{l_2} + L' - S' + M'_J - l'_2 + m'_L - s'_2 + M'_S + m'_{j_1}} \\
& \times \sqrt{(2l'_2 + 1)(2l''_2 + 1)(2L' + 1)(2S' + 1)(2J' + 1)(2j'_1 + 1)} \begin{pmatrix} l''_2 & 1 & l'_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l''_2 & 1 & l'_2 \\ -m''_{l_2} & q & m'_{l_2} \end{pmatrix} \\
& \times \begin{pmatrix} L' & S' & J' \\ M'_L & M'_S & -M'_J \end{pmatrix} \begin{pmatrix} l'_1 & l'_2 & L' \\ m'_{l_1} & m'_{l_2} & -M'_L \end{pmatrix} \begin{pmatrix} s'_1 & s'_2 & S' \\ m'_{s_1} & m'_{s_2} & -M'_S \end{pmatrix} \begin{pmatrix} l'_1 & s'_1 & j'_1 \\ m'_{l_1} & m'_{s_1} & -m'_{j_1} \end{pmatrix} R_{n'_2 l'_2}^{kl''},
\end{aligned} \quad (12)$$

where $\chi = \{l'_2, m'_{l_2}, M_L, M_S, m_{l_1}, m_{l_2}, m_{s_1}, m_{s_2}, j_1, m_{j_1}\}$, and the coefficient $a''_{l_2} m''_{l_2}$ is defined as, $a''_{l_2} m''_{l_2} = 4\pi i^{-l''_2} \times \exp(-i\delta''_2) Y_{l''_2 m''_{l_2}}$ with δ''_2 being the phase shift. $R_{n'_2 l'_2}^{kl''}$ represents a radial matrix element from $|n'_1 l'_1 n'_2 l'_2\rangle$ to $|n'_1 l'_1 k l''_2\rangle$, which is essentially a single-electron transition from $n'_2 l'_2$ to kl''_2 . When the ionization takes place after two-photon absorption through the process $A(\Lambda_1, \Lambda_2)$ and $M(\Lambda_2, \Psi)$, the photoelectron yield $P_e[m''_{s_2}]$ with spin-up(-down) ($m''_{s_2} = \pm 1/2$), irrespective of the ionic state $|(n''_1 l''_1 s''_1) j''_1 m''_{j_1}\rangle$, can be computed using the effective two-photon matrix element, as

$$P_e[m''_{s_2}] = \sum_{\text{core}} \int d\Omega_{\mathbf{k}} \left| \sum_{\Lambda_2} \frac{A(\Lambda_1, \Lambda_2) M(\Lambda_2, \Psi)}{\Delta_{\Lambda_2}} \right|^2, \quad (13)$$

where Δ_{Λ_2} is the detuning of laser ω_2 from an intermediate state Λ_2 . $\int d\Omega_{\mathbf{k}}$ indicates integration over all solid angle for the ejected photoelectron, and the sum is taken over all possible ionic core states. Similarly, the ion yield $P_{\text{ion}}[(n''_1 l''_1 s''_1) j''_1 m''_{j_1}]$ into the ionic state $|(n''_1 l''_1 s''_1) j''_1 m''_{j_1}\rangle$ can be computed as

$$\begin{aligned}
P_{\text{ion}}[(n''_1 l''_1 s''_1) j''_1 m''_{j_1}] = & \sum_{m''_{s_2} = \pm 1/2} \int d\Omega_{\mathbf{k}} \\
& \times \left| \sum_{\Lambda_2} \frac{A(\Lambda_1, \Lambda_2) M(\Lambda_2, \Psi)}{\Delta_{\Lambda_2}} \right|^2. \quad (14)
\end{aligned}$$

When we apply these formulas to the scheme depicted in Fig. 1(a), $\Lambda_1 = |5s5p^3 P_1 M_J\rangle$, $\Lambda_2 = |5s5d^3 D_{1,2} M'_J\rangle$, and $\Psi = |k; m''_{s_2} 5s_{1/2} m''_{j_1}\rangle$.

C. Scheme B: selective excitation of a particular magnetic sublevel

Now we examine scheme B. This scheme is nothing but one-photon ionization from a selected state by laser ω_1 , which is $|5s5p^3 P_1\rangle$ in this case. As for the bound-free matrix elements, we can again use the formula given by Eqs. (12). The ion yield $P_{\text{ion}}[(n''_1 l''_1 s''_1) j''_1 m''_{j_1}]$ can be computed using

$$P_{\text{ion}}[(n''_1 l''_1 s''_1) j''_1 m''_{j_1}] = \sum_{m''_{s_2} = \pm 1/2} \int d\Omega_{\mathbf{k}} |M(\Lambda_2, \Psi)|^2, \quad (15)$$

with $\Lambda_2 = |5s5p^3 P_1 M'_J\rangle$ and $\Psi = |k; m''_{s_2} 5s_{1/2} m''_{j_1}\rangle$.

III. RESULTS AND DISCUSSION

In this section we present quantitative results for schemes A and B. Under the pure LS coupling with a single term whose validity has been confirmed for our specific case in Sec. II A, computation of the angular part in Eqs. (11) and (12) is straightforward. As for the pertinent radial matrix elements, we employ a single-channel quantum defect theory, since the excitations from $\Lambda_1 = |5s5p^3 P_1\rangle$ to $\Lambda_2 = |5s5d^3 D_{1,2}\rangle$, and $\Lambda_2 = |5s5d^3 D_{1,2}\rangle$ to $\Psi = |5skl\rangle$ are essentially a single-electron transition, i.e., $5p$ to $5d$, and $5d$ to kp or kf where k being the wave vector of the photoelectron, with the $5s$ electron as a spectator. In this work we assume that the bound-free radial matrix elements R_{5d}^{kl} are the same for both $|5s5d^3 D_1\rangle$ and $|5s5d^3 D_2\rangle$. We should note that the use of the single-electron transitions approximation and the neglect of the fine-structure-dependence of pertinent radial matrix elements may cause small inaccuracy in terms of the *absolute* ionization cross sections. That should not be a serious problem, however, for our specific study, since what is of interest is a degree of spin polarization: Recall that the degree of spin polarization is a *relative ratio* of ionization cross sections into different channels with specific polarizations.

In the following subsections, we introduce the notations $P_{M_J}^\uparrow$ and $P_{M_J}^\downarrow$ ($M_J = \pm 1, 0$) to represent the ionization from the state $|5s5p^3 P_1 M_J\rangle$ into the ionic ground-state $\text{Sr}^+ 5s^2 S_{1/2}$ with spin-up/-down, respectively. Using this notation, the total polarization can be calculated from the relation,

$$P = \frac{\sum_{M_J=0, \pm 1} a_{M_J} (P_{M_J}^\uparrow - P_{M_J}^\downarrow)}{\sum_{M_J=0, \pm 1} a_{M_J} (P_{M_J}^\uparrow + P_{M_J}^\downarrow)}, \quad (16)$$

where a_{M_J} ($M_J = 0, \pm 1$) are the weighting factors for the population in each magnetic sublevel $|5s5p^3 P_1 M_J\rangle$.

Before going into the details of our results, we would like to make a brief remark on ionization via a singlet state. By carrying out calculations for ionization from $|5s5p^1 P_1\rangle$ and $|5s5d^1 D_2\rangle$ with all possible magnetic sublevels, we have numerically confirmed that the *spin polarization for ions as well as electrons is always zero regardless of the laser polarization and magnetic sublevels chosen, if the ionization takes place from a singlet state*. This is simply because a singlet state does not have spin-orbit interactions, i.e., $\mathbf{L} \cdot \mathbf{S} = 0$ since $\mathbf{S} = \mathbf{0}$.

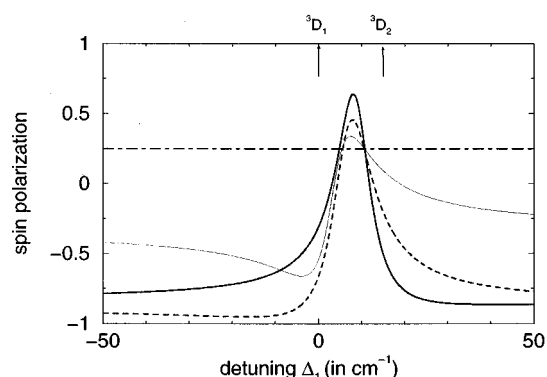


FIG. 2. Variation of the degree of spin-polarization of photoions for scheme A as a function of detuning Δ_1 by right-circularly polarized laser ω_2 . Ionization takes place from $5s5p\ ^3P_1$ (a) $M_J = -1$ (solid), (b) $M_J = 0$ (dashed), (c) $M_J = 1$ (dotted-dashed), and (d) all M_J (thin solid).

A. Scheme A

By making use of the formula presented in Eq. (14), it is straightforward to obtain the expressions for scheme A. For completeness, we consider two possibilities for the polarization of lasers ω_1 and ω_2 , i.e., right-circularly or linearly polarized. The results for left-circularly polarized laser can be immediately obtained by just changing signs of the obtained spin polarizations of ions for right-circularly polarized laser, and therefore we do not need to present the expressions here.

1. Ionization by right-circularly polarized laser ω_2

First we assume that the laser ω_2 is right-circularly polarized. Expressions for the amount of produced $\text{Sr}^+ 5s$ ions from each of the magnetic sublevels $|5s5p\ ^3P_1 M_J\rangle$ ($M_J = 0, \pm 1$) are given in the Appendix A1 as functions of Δ_1 and Δ_2 . Note that Δ_1 and Δ_2 are the detunings of laser ω_2 from $|5s5d\ ^3D_1\rangle$ and $|5s5d\ ^3D_2\rangle$, respectively, with the relation $\Delta_2 = \Delta_1 - 15.1$ (in units of cm^{-1}) for our specific scheme in Fig. 1(a).

Regarding the expressions for the amount of *photoelectrons* with spin-up (-down), we find that they are exactly the same with those of *ions* in $\text{Sr}^+ 5s\ ^2S_{1/2}$ with spin-up (-down). This means that *there is a one-to-one correspondence between the degree of spin polarization of photoion and photoelectron*, as one might naively imagine. This is because all ions are in the ionic ground state $5s\ ^2S_{1/2}$, which does not have orbital angular momentum. If some of the produced ions were in the excited 2P or 2D state, etc., which has orbital angular momentum, there would be no clear correspondence between them. Using Eqs. (A1)–(A6) together with Eq. (16), we can calculate the variation of the degree of polarization as a function of Δ_1 . As for a_{M_J} ($M_J = 0, \pm 1$) in Eq. (16), we consider four cases. The results for $(a_{-1}, a_0, a_1) = (1, 0, 0), (0, 1, 0), (0, 0, 1)$, and $(1/3, 1/3, 1/3)$, which can be easily realized by making use of a right-circularly, linearly, and left-circularly polarized laser, and unpolarized laser for ω_1 , are shown in Fig. 2. When the laser ω_2 is far off-resonance, i.e., $|\Delta_1| \gg 1$, the degree of polarization is rather high, and it is about -92% at $\Delta_1 = -50\ \text{cm}^{-1}$ for ionization from $|5s5p\ ^3P_1 M_J = 0\rangle$ (dashed line) by right-circularly polarized light. This might

look strange at first glance. This is so, because we have calculated ionization from a particular fine structure state $|5s5p\ ^3P_1\rangle$: If it were *nonresonant* multiphoton ionization directly from the ground state, the spin polarization would have been zero. For ionization from $|5s5p\ ^3P_1 M_J = -1\rangle$ (solid line), the spin polarization changes its sign when Δ_1 is varied from negative to positive values, and it reaches a maximum positive polarization of 65% at $\Delta_1 = 8\ \text{cm}^{-1}$ due to the interference effect, and then decreases again. A similar tendency can be seen for the case starting from $|5s5p\ ^3P_1 M_J = 0\rangle$ (dashed line). For ionization from $|5s5p\ ^3P_1 M_J = 1\rangle$ (dotted-dashed), however, it is only $|5s5d\ ^3D_2\rangle$ that is involved as an intermediate state due to the selection rule $\Delta M_J = +1$ for excitation by right-circularly polarized radiation. No two-path interference takes place. Therefore the curve for two-photon ionization from $|5s5p\ ^3P_1 M_J = 1\rangle$ (dotted-dashed) is completely flat. When the initial state is an isotropic mixture of all $|5s5p\ ^3P_1 M_J\rangle$ ($M_J = 0, \pm 1$) (thin solid), the variation of the curve is rather small, which is obviously due to the contribution from $|5s5p\ ^3P_1 M_J = 1\rangle$. As a final remark on Fig. 2, we note that the best choices of detunings would be $\Delta_1 = -5$ and $0\ \text{cm}^{-1}$, starting from $M_J = 0$, which leads to -90% and -65% polarizations, respectively. This is sort of a compromise between the degree of spin polarization and the amount of expected ion yield. The use of interference effect represented by the polarization peak at $\Delta_1 = 8\ \text{cm}^{-1}$ for $M_J = -1$, turned out not to be very useful, since the same degree of polarization with opposite sign, 65% , can be obtained at perfect resonance without two-path interference, i.e., $\Delta_1 = 0$ for $M_J = 0$, which necessarily guarantees huge enhancement in ionization yield.

2. Ionization by linearly polarized laser ω_2

We can carry out similar calculations for the case in which the laser ω_2 is linearly polarized. Expressions for the amount of produced Sr ions from each of the magnetic sublevels $|5s5p\ ^3P_1 M_J\rangle$ ($M_J = 0, \pm 1$) are given in the Appendix A2.

Again, using Eqs. (A7)–(A12), we calculate the variation of the degree of polarization as a function of detuning Δ_1 . The results for $(a_{-1}, a_0, a_1) = (1, 0, 0), (0, 1, 0), (0, 0, 1)$, and $(1/3, 1/3, 1/3)$ in Eq. (16) are shown in Fig. 3. Because of the symmetry, the curves for $|5s5p\ ^3P_1 M_J = -1\rangle$ (solid line) and $|5s5p\ ^3P_1 M_J = 1\rangle$ (dotted-dashed) are upside-down. At far off-resonance, $|\Delta_1| \gg 1$, the polarization is about $\pm 65\%$ for both cases. Moreover, in both cases, the spin polarization changes its sign, due to the two-path interference via intermediate $|5s5d\ ^3D_1\rangle$ and $|5s5d\ ^3D_2\rangle$, as the detuning Δ_1 is varied from negative to positive values and vice versa. For $|5s5p\ ^3P_1 M_J = -1\rangle$ and $|5s5p\ ^3P_1 M_J = 1\rangle$, the maximum–minimum spin polarization of $\pm 85\%$ is obtained at $\Delta_1 = 5\ \text{cm}^{-1}$ where the laser ω_2 is tuned between $|5s5d\ ^3D_1\rangle$ and $|5s5d\ ^3D_2\rangle$ where two-path interference is significant. As for ionization from $|5s5p\ ^3P_1 M_J = 0\rangle$ (dashed line), the spin polarization is zero at any Δ_1 , which is obviously because of the symmetry. It is interesting to point out that the maximum spin polarization of 85% is achieved through two-path interference if the laser ω_2 is lin-

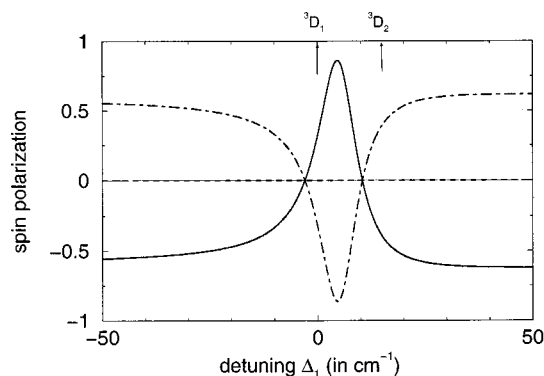


FIG. 3. Variation of the degree of spin-polarization of photoions as a function of detuning Δ_1 by linearly polarized laser ω_2 . Ionization takes from from $5s5p\ ^3P_1$ (a) $M_J = -1$ (solid), (b) $M_J = 0$ (dashed), (c) $M_J = 1$ (dotted-dashed), and (d) all M_J [thin solid, which is identical to graph (b)].

early polarized. In contrast, two-path interference does not help so much if the laser ω_2 is right-circularly polarized, as we have already seen in Fig. 2.

Note, however, that the degree of polarization is completely zero with linear polarization for laser ω_2 , if the ionization takes place from an isotropic $|5s5p\ ^3P_1\rangle$ (thin solid line in Fig. 3), while the spin polarization is -67% at $\Delta_1 = -3.3\text{ cm}^{-1}$ (thin solid line in Fig. 2) with right-circular polarization for laser ω_2 .

B. Scheme B

We perform similar calculations for scheme B [Fig. 1(b)] using Eqs. (12) and (16). In particular, for the evaluation of bound-free radial matrix elements, we have assumed that the photon energy for laser ω_3 is about $32\,470\text{ cm}^{-1}$, corresponding to the wavelength of an excimer (XeCl) laser. For completeness, we again consider two possibilities for the polarization of lasers ω_1 and ω_3 , i.e., right-circularly or linearly polarized. The results for the case with left-circularly polarized laser can be immediately obtained by making use of the prescription which is explained just before Sec. III A.

1. Ionization by right-circularly polarized laser ω_3

First we consider the case of right-circular polarization for laser ω_3 . Expressions for the amount of produced Sr ions from each of the magnetic sublevels $|5s5p\ ^3P_1\ M_J\rangle$ ($M_J = 0, \pm 1$) are given in the Appendix B 1. Using Eqs. (B1)–(B6) together with Eq. (16), we can calculate the degree of polarization. The result is given in the first line of Table II.

2. Ionization by linearly polarized laser ω_3

Second we consider the case of linear polarization for laser ω_3 . Expressions for the amount of produced Sr ions

from each of the magnetic sublevels $|5s5p\ ^3P_1\ M_J\rangle$ ($M_J = 0, \pm 1$) are given in the Appendix B 2. Using Eqs. (B7)–(B12) together with Eq. (16), we can calculate the degree of spin polarization. The result is given in the second line of Table II. Comparing this result with that for right-circular polarization, the achievable degrees of spin polarization (in absolute value) are almost the same: Spin polarization is -66% for ionization from $|5s5p\ ^3P_1\ M_J = -1\rangle$ by right-circularly polarized laser ω_3 , while it is $\mp 60\%$ for ionization from $|5s5p\ ^3P_1\ M_J = \mp 1\rangle$ by linearly polarized laser ω_3 .

IV. CONCLUSION

In conclusion, we have quantitatively studied two realistic schemes for creating electron spin-polarized alkaline-earth ions via multiphoton ionization of neutral atoms, and compared both results. In order to obtain polarized ions, it is essential that the ionization takes place from a triplet state, which we have chosen as $sp\ ^3P_1$ for both schemes.

The first scheme (scheme A) utilizes one-photon resonant two-photon ionization via fine structure manifolds. The ionization path is described by $s^2\ ^1S_0 \rightarrow sp\ ^3P_1 \rightarrow (sd\ ^3D_{1,2}) \rightarrow \text{ion}$, where two-path interference plays an important role to obtain high degree of spin polarization with near-resonant enhancement of ionization. We have found that the more polarized ions can be obtained if the laser frequencies and the laser polarizations are properly chosen. In particular, due to the two-path interference, 85% spin polarization turned out to be possible for Sr if the ionization takes place from $|5s5p\ ^3P_1\ M_J = -1\rangle$ with a linearly polarized laser. With a right-circularly polarized laser, about -90% polarization is possible with a reasonably small detuning, but this is not due to the interference.

The second scheme (scheme B) is based on one-photon resonant one-photon ionization. The ionization path is described by $s^2\ ^1S_0 \rightarrow sp\ ^3P_1 \rightarrow \text{ion}$. The achievable degree of polarization for Sr^+ has been found to be no more than 66%. However, much more ionization yield can be expected for the second scheme compared with the first one, simply because it is one-photon ionization from the resonant $sp\ ^3P_1$ state. We should keep in mind that a one-photon ionization cross section is typically of the order of $10^{-18} - 10^{-16}\text{ cm}^2$, while two-photon ionization cross section is of the order of $10^{-52} - 10^{-45}\text{ cm}^4\text{ s}$. Clearly the first scheme is more favorable for the purpose of obtaining more polarized ion at the expense of ionization yield, which can be always compensated only if intense lasers are available. Therefore, both schemes have advantages and disadvantages. Which scheme would be more suitable depends on how the polarized ions are used afterwards for various spin-dependent experiments. In any case, neither requires a sophisticated experimental setup, which is perhaps the largest advantage of the proposed schemes.

Before closing, we would like to note that, although our quantitative study has been carried out specifically for Sr, very similar results are expected for other alkaline-earth atoms such as Ca and Ba. After all, it is an angular part of matrix elements rather than a radial part that plays an essential role in producing electron spin-polarized ions. As for the

TABLE II. Spin-polarization (in %) of photoions for scheme B by linearly/right-circularly polarized laser ω_3 .

	$M_J = -1$	$M_J = 0$	$M_J = 1$	Isotropic $5s5p\ ^3P_1$
right-circular	-66	-60	+33	-21
linear	-60	0	+60	0

radial part, it is the *ratio* of radial matrix elements and not the absolute values that affects the degree of polarization. Therefore, we estimate that the difference of polarization for other alkaline-earth atoms would be no more than $\pm 10\%$.

ACKNOWLEDGMENTS

The work by T.N. was supported by the Grant-in-Aid for scientific research from the Ministry of Education and Science of Japan, and the Grant for Basic Science from Sumitomo Foundation.

APPENDIX A: SCHEME A

1. Right-circular polarization for laser ω_2

If the laser ω_2 is right-circularly polarized, expressions for the amount of ionization are derived, using Eq. (14) together with Eqs. (11) and (12), as

$$P_{-1}^{\uparrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{-0.2450R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.3162R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1604R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} - \frac{0.2070R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1155R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} \right|^2 + \left| \frac{0.1852R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} \right|^2 \right], \quad (\text{A1})$$

$$P_{-1}^{\downarrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{0.1155R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} \right|^2 + \left| \frac{0.1852R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} \right|^2 + \left| \frac{0.2928R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.3780R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A2})$$

$$P_0^{\uparrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{-0.0817R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \sqrt{3} \frac{0.1826R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1309R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} - \sqrt{3} \frac{0.2928R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{-0.2070R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \sqrt{3} \frac{0.1543R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A3})$$

$$P_0^{\downarrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{-0.2070R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \sqrt{3} \frac{0.1543R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.5071R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} - \sqrt{3} \frac{0.3780R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A4})$$

$$P_{+1}^{\uparrow} = 4\pi^2 |A_1|^2 \left[\left| \sqrt{3} \frac{0.3087R_{5d2}^{kf}}{2\sqrt{5}\Delta_2} \right|^2 + \left| \sqrt{3} \frac{0.3780R_{5d2}^{kf}}{2\sqrt{5}\Delta_2} \right|^2 \right], \quad (\text{A5})$$

$$P_{+1}^{\downarrow} = 4\pi^2 |A_1|^2 \left| \sqrt{3} \frac{0.3780R_{5d2}^{kf}}{2\sqrt{5}\Delta_1} \right|^2, \quad (\text{A6})$$

where Δ_1 and Δ_2 are the laser detunings from states $|5s5d^3D_1\rangle$ and $|5s5d^3D_2\rangle$, respectively, with the relation $\Delta_2 = \Delta_1 - 15.1$ (in units of cm^{-1}). A_1 is the reduced matrix element defined as $A_1 = \langle 5s5d^3D || r || 5s5p^3P \rangle$, namely the radial part of $A(\Lambda_1, \Lambda_2)$.

2. Linear polarization for laser ω_2

Similarly, if the laser ω_2 is linearly polarized, expressions for the amount of ionization are derived as

$$P_{-1}^{\uparrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{0.2928R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} - \frac{0.3779R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1732R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.2236R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1852R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.2390R_{5d1}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A7})$$

$$P_{-1}^{\downarrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{0.1732R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.2236R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1852R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.2390R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1633R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.6324R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1604R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.6211R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A8})$$

$$P_0^{\uparrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{0.3162R_{5d2}^{kp}}{\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.3381R_{5d2}^{kf}}{\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A9})$$

$$P_0^{\downarrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{-0.2070R_{5d2}^{kf}}{\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.3381R_{5d2}^{kf}}{\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A10})$$

$$P_1^{\uparrow} = 4\pi^2 |A_1|^2 \left[\left| \frac{0.1633R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.6324R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1604R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.6211R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1732R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.2236R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 + \left| \frac{0.1852R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.2390R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right], \quad (\text{A11})$$

$$\begin{aligned}
P_1^\perp = 4\pi^2 |A_1|^2 & \left[\left| \frac{0.1732R_{5d1}^{kp}}{2\sqrt{6}\Delta_1} + \frac{0.2236R_{5d2}^{kp}}{2\sqrt{10}\Delta_2} \right|^2 \right. \\
& + \left| \frac{0.1852R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} + \frac{0.2390R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \\
& \left. + \left| \frac{0.2928R_{5d1}^{kf}}{2\sqrt{6}\Delta_1} \right|^2 + \left| \frac{0.3779R_{5d2}^{kf}}{2\sqrt{10}\Delta_2} \right|^2 \right]. \quad (A12)
\end{aligned}$$

APPENDIX B: SCHEME B

1. Right-circular polarization for laser ω_3

If the laser ω_3 is right-circularly polarized, expressions for the amount of ionization are derived, using Eqs. (14) and (12), as

$$P_{-1}^\uparrow = 0.0835|R_{5p}^{ks}|^2 + 0.0166|R_{5p}^{kd}|^2, \quad (B1)$$

$$P_{-1}^\perp = 0.0835|R_{5p}^{ks}|^2 + 0.1165|R_{5p}^{kd}|^2, \quad (B2)$$

$$P_0^\uparrow = 0.1665|R_{5p}^{ks}|^2 + 0.0335|R_{5p}^{kd}|^2, \quad (B3)$$

$$P_0^\perp = 0.1998|R_{5p}^{kd}|^2, \quad (B4)$$

$$P_{+1}^\uparrow = 0.1997|R_{5p}^{kd}|^2, \quad (B5)$$

$$P_{+1}^\perp = 0.0999|R_{5p}^{kd}|^2. \quad (B6)$$

2. Linear polarization for laser ω_3

Similarly, if the laser ω_3 is linearly polarized, expressions for the amount of ionization are derived as

$$P_{-1}^\perp = 0.0502|R_{5p}^{kd}|^2, \quad (B7)$$

$$P_{-1}^\perp = 0.1665|R_{5p}^{ks}|^2 + 0.1834|R_{5p}^{kd}|^2, \quad (B8)$$

$$P_0^\uparrow = 0.0999|R_{5p}^{kd}|^2, \quad (B9)$$

$$P_0^\perp = 0.0999|R_{5p}^{kd}|^2, \quad (B10)$$

$$P_{+1}^\uparrow = 0.1665|R_{5p}^{ks}|^2 + 0.1834|R_{5p}^{kd}|^2, \quad (B11)$$

$$P_{+1}^\perp = 0.0502|R_{5p}^{kd}|^2. \quad (B12)$$

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