Multiphoton single ionization of two-electron systems in intense laser fields

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Multiphoton single-ionization of a two-electron system (He and He-like ions) in intense circularly polarized laser field is reported in a relativistic field theoretic method. Coulomb-correction factor is introduced to estimate the effect of the Coulomb field of the residual ion on the rate. Antisymmetric wave function is considered both in the initial and in the final state. The spin-specific currents are calculated. Angular asymmetry in current generation is noted with the change in the spin direction of the ionized electron. Coulomb-corrected relativistic-result for total rate is compared with the Coulomb-corrected nonrelativistic KFR rate. At the high-intensity regime nonrelativistic rate overestimates the relativistic rate. Formation of orthopositronium and parapositronium from positronium negative ion by multiphoton ionization is discussed.

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I. INTRODUCTION

Multiphoton ionization of a two-electron system mainly He is looked into in this work for single-ionization in the intense laser field in a relativistic field theoretic method, with special emphasis on spin-current production which has many applications in spintronics and magnetoresistances. Field theory and Feynman diagrams are used to calculate single and double ionization of He in a perturbative-approach [1]. In the case of ionization in intense laser field nonperturbative approach is sufficient. In strong laser field the magnetic component of the field plays important role in the spin of the electron (free or bound). A characteristic aspect of the strong laser field is the coupling of the magnetic field with the spin degrees of freedom. Spin-dependent ionization current is obtained earlier using circularly polarized light shining multiphoton on hydrogen atom in paper [2] and single photon on helium atoms in paper [3]. In this paper, while we restrict ourselves to the straight forward relativistic generalization of strong field S-matrix approach (SFA) with regard to radiation field to obtain the rate, we multiply the rate by Coulombcorrection factor [4] of the residual ion analogous to the nonrelativistic Coulomb-corrected Keldysh-Faisal-Reiss (KFR) rate [5]. In principle the photo-electron is dressed by laser field [6] (Volkov wave function) and the Coulomb field of the residual ion. To estimate the effect of this additional Coulomb field, we multiply the relativistic-rate by the square of the Coulomb-correction factor. In the time reversed S matrix, the system is initially prepared in a state free of transition carrying field. The SFA method allows for the rigorous introduction of arbitrary temporal as well as spatial shape of the laser pulse. The pulse is relatively long but goes to zero at asymptotic time is automatically satisfied [7]. The energy of the photo-electron should be much larger than the binding energy.

A frequency-dependent asymmetry between the spin-up (u) and spin-down (d) currents that varies according to the direction of electron emission is found. Hopefully the rates

of spin-flips in the ionization process and possibly also the spin-asymmetry from target atoms could be measured in "second generation experiments." Analysis of such multiphoton ionization experiments would require knowledge of the spin-dependence of the ionization rates. No such spin specific ionization rate of He in intense field appeared to have been obtained so far. In this paper, we consider antisymmetric wave functions in the initial state as well as in the final state. Initially the two bound-electrons are in the ground spin-singlet $(S_i=0)$ state. It is difficult to solve ground-state helium wave function in a relativistic way. For simplicity, we have made an effective "Active-single-electron" (ASE) hypothesis. At high-laser intensity He in the ground state is taken as the antisymmetric product of two hydrogen-type Dirac-electrons in the spin-singlet state with [3] effective charge $Z_{eff}=27/16$. In this paper, unlike Ref. [3], we have considered the two electrons in the final state (one bound and one free) to be either in the spin-singlet $(S_f=0)$ or in the spin-triplet $(S_f=1)$ state. Inclusion of antisymmetry in the initial and in the final state gives rise to the two types of multiphoton ionization processes. (a) Direct process: photon interacting with one of the bound electrons ionizes it (with spin direction unchanged or flipped), the other electron remains bound in the same spin state. (b) shake-off process: where one of the bound electrons interacting with photon, remains bound with its spin flipped, while the other electron due to the change in the internal potential is kicked off without change of spin. Transition to the *singlet state* (S_f) =0, M_f =0) involves only *direct process* while that to the triplet-state involves both the spin-flip direct process and the shake-off process.

Explicit analytical formulae for the relativistic rate are derived for multiphoton ionization of a two-electron atom/ ion by circularly polarized light, and results of numerical calculations for multiphoton ionization rate after Coulomb-correction are presented. Atomic units are considered $\hbar = |\mathbf{e}| = \mathbf{m} = \mathbf{a}_0 = 1$, $\mathbf{c} = \alpha^{-1}$.

II. THEORY

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Using SFA analogous to the nonrelativistic KFR approach [5] the leading term of *S* matrix is

$$\begin{split} (S_{s_1s_2 \to s_1's_2'})_{S_f,M_f} &= -i \int_{-\infty}^{\infty} \langle \bar{\psi}_f^{(s_1',s_2')}(r_1,r_2) | \gamma^{\mu}(1) A_{\mu}(r_1) \\ &+ \gamma^{\nu}(2) A_{\nu}(r_2) | \psi_{1s^2}^{(s_1,s_2)}(r_1,r_2) \rangle d^3r_1 d^3r_2 dr_0 \end{split}$$
(1)

 $s_1 \neq s_2 = u$, *d* are the electron-spins in the initial ground state of He atom. s'_1 , s'_2 are the corresponding spins in the final state.

Transition rate per unit time

$$\Gamma_{s_1, s_2 \to s_1', s_2'} = \int |(S_{s_1, s_2 \to s_1', s_2'})_{S_f \cap M_f}|^2 \frac{d^3 p}{(2\pi)^3}.$$
 (1a)

The angular differential rate for transition from the groundsinglet state to the final states S_f , M_f on ionization is given by

$$\left\lfloor \frac{d(\Gamma)_{S_f,M_f}}{d\Omega} \right\rfloor = |(S_{s_1,S_2 \to s_1',s_2'})_{S_f,M_f}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}.$$
 (1b)

The parenthesis in $\gamma^{\mu}(i)$ in (1) indicates photon interacts with electron at $r_i = (r_0, \vec{r_i})$ and $p = (p_0, \vec{p})$ is the four-momentum of the ionized electron, $d\Omega = \sin \theta d\theta d\phi$.

The vector potential is given by

$$A(r_{1}) = \frac{eA_{0}}{2c} [\{\hat{\varepsilon}e^{ik \cdot r_{1}} + \hat{\varepsilon}^{*}e^{-ik \cdot r_{1}}\}].$$
 (2)

The polarization vectors for circularly polarized photons are, respectively,

$$\hat{\varepsilon} = \frac{1}{\sqrt{2}}(1, i, 0)$$
 and $\hat{\varepsilon}^* = \frac{1}{\sqrt{2}}(1, -i, 0)$,

 $k = (k_0, \vec{k})$ is the four-momentum of photon, $k_0 = \omega/c$,

where $\hat{\varepsilon} \cdot \hat{k} = 0$, $\vec{k} = (0, 0, k_z)$. Let us denote the spin-up electron as "*u*" and spin down-spin electron as "*d*." The initial ground state of He atom is considered as an antisymmetric combination of the two Dirac-hydrogen wave functions with [3] effective charge $Z_{eff} = 27/16$.

$$\psi_{1s^2}^{(s_1,s_2)}(r_1,r_2) = \left[\psi_{1s}(r_1)\psi_{1s}(r_2)\right] \frac{1}{\sqrt{2}} \{\left[u(1)d(2)\right] - u(2)d(1)\}.$$
(3)

The two electrons in the final state $\psi_f^{(s_1',s_2')}(r_1,r_2)$ may be either in the singlet state

 $S_f = s'_1 + s'_2 = 0$, $M_f = 0$ or in the triplet state $S_f = s'_1 + s'_2 = 1$, $M_f = 1, 0, -1$.

The final singlet state is

$$\psi_{f}^{(s_{1}',s_{2}')}(r_{1},r_{2}) = \frac{1}{\sqrt{2}} [\psi_{p}(r_{1})\psi_{1s}^{He^{+}}(r_{2}) + \psi_{p}(r_{2})\psi_{1s}^{He^{+}}(r_{1})] \\ \times \frac{1}{\sqrt{2}} \{ [u(1)d(2)] - u(2)d(1) \}.$$
(4)

The final triplet-states corresponding to the three magneticquantum states are as below

(i)
$$S_f = 1$$
, $M_f = 0$,
 $\psi_f^{(s_1',s_2')}(r_1,r_2) = \frac{1}{\sqrt{2}} [\psi_p(r_1)\psi_{1s}^{He^+}(r_2) - \psi_p(r_2)\psi_{1s}^{He^+}(r_1)]$
 $\times \frac{1}{\sqrt{2}} \{ [u(1)d(2)] + u(2)d(1) \}$ (5)
(ii) $S_f = 1$, $M_f = 1$,
 $\psi_f^{(s_1',s_2')}(r_1,r_2) = \frac{1}{\sqrt{2}} [\psi_p(r_1)\psi_{1s}^{He^+}(r_2) - \psi_p(r_2)\psi_{1s}^{He^+}(r_1)]$

$$\times \{ [u(1)u(2)] \}$$
 (6)

(iii)
$$S_f = 1$$
, $M_f = -1$,
 $S_1^{s_1^{\prime}, S_2^{\prime}}(r_1, r_2) = \frac{1}{\sqrt{2}} [\psi_p(r_1) \psi_{1s}^{He^+}(r_2) - \psi_p(r_2) \psi_{1s}^{He^+}(r_1)]$
 $\times \{ [d(1)d(2)] \}.$ (7)

Dirac-Volkov solution [6] for photo-electron in intense laser field $\psi_p(r)u_s$ for ionized electron is given by

 $\psi_{f}^{(}$

$$\psi_p(r_1)u_s = \sum_{n>n_0}^{\infty} N_{p_0} \exp\{i(p \cdot r_1 + \lambda_p k \cdot r_1)\}$$
$$\times \exp(-ink \cdot r_1) \phi_n^s(p), \tag{8}$$

where n_0 is the minimum photon number required for ionization.

$$\phi_{n}^{s}(p) = \left[J_{n}(a_{p}, b_{p}, \chi) + \frac{eA_{0}/2}{2k \cdot p} \{ k \notin J_{n-1}(a_{p}, b_{p}, \chi) + k \notin^{*} J_{n+1}(a_{p}, b_{p}, \chi) \} \right] u_{s}(p)$$

$$= f_{n} u_{s}(p)$$
(8a)

 $u_s(p)$ is the Dirac-spinor for free electron having fourmomentum p and spin s. The photon four-momentum is k and $N_{p_0} = \sqrt{\frac{c}{p_0}}$, χ_s is the Pauli spinor.

$$u_{s}(p) = \begin{pmatrix} m_{1}\chi_{s} \\ m_{2}\sigma \cdot \hat{p}\chi_{s} \end{pmatrix}$$

$$m_{1} = \sqrt{\frac{p_{0} + c}{2c}}, \quad m_{2} = -\sqrt{\frac{p_{0} - c}{2c}}$$

$$k \cdot p = k_{0}p_{0} - \vec{k} \cdot \vec{p}, \quad p_{0} = (\sqrt{c^{2} + \vec{p}^{2}})$$

$$J_{n}(a_{p}, b_{p}, \chi) = \sum_{m=-\infty}^{\infty} J_{n+2m}(a_{p})J_{m}(b_{p})e^{(2im+in)\chi}$$
(8b)

are the generalized Bessel function [2] of three arguments with

$$a_p = \frac{eA_0 |\vec{\varepsilon} \cdot \vec{p}|}{c \kappa \cdot p}, \quad b_p = \frac{e^2 A_0^2}{8c^2 \kappa \cdot p} \cos \xi,$$
$$\chi = \tan^{-1} [\tan \phi \tan(\xi/2)]$$
$$\lambda_p = \frac{e^2 A_0^2}{4c^2 \kappa \cdot p}.$$

For circularly polarized light $\xi = \pi/2$, eventually $\chi = \phi$ and $b_p = 0$

Since $J_m(0)=0$ for $m \neq 0$ and $J_0(0)=1$, $J_n(a_p, 0, \chi) = J_n(a_p)e^{in\phi}$

$$f_n = \left[J_n(a_p) + \frac{eA_0/2}{2k \cdot p} \{ k \notin J_{n-1}(a_p) + k \notin^* J_{n+1}(a_p) \} \right] e^{in\phi}.$$
(9)

In the final state the Dirac hydrogen-type wave function of He^+_{1s} is

 $\psi_{1s}^{He^+(s'_2)}(r')\omega_{s'_2}$ and the same as in [3] with z=2. We calculate below the transition matrix elements from the *singlet* ground state to the *singlet* and to the three *triplet states*. Let us denote the spin-up electron as " u_b or u_f " (the subscripts "*b* and *f*" indicate bound and free electron, respectively) and the spin-down electron as " d_b or d_f ."

A. Singlet to singlet transition

Using (2)-(4) in (1) the transition matrix from singlet to singlet state

$$(S_{s_1s_2 \to s_1's_2'})_{0,0} = \left\lfloor \frac{1}{\sqrt{2}} (S_{Dir}^{u_b, d_b \to u_f, d_b}) + \frac{1}{\sqrt{2}} (S_{Dir}^{d_b, u_b \to d_f, u_b}) \right\rfloor.$$
(10)

The general matrix element $S_{Dir}^{s_1,s_2 \rightarrow s'_1,s'_2}$ to be mentioned as direct term is given by

$$(S_{Dir}^{s_1, s_2 \to s_1', s_2'}) = \int (I_1^{s_1 \to s_1'}) (I_2^{s_2 \to s_2'}) dr_0, \qquad (10a)$$

where

$$(I_1^{s_1 \to s_1'}) = \int \bar{u}_{s_1'}(p) \psi_p^+(r_1) \gamma(1) A(r_1) \psi_{1s}(r_1) \omega_{s_1} d^3 r_1 \qquad (10b)$$

$$(I_2^{s_2 \to s_2'}) = \int d^3 r_2 \bar{\omega}_{s_2'} (\psi_{1s}^{He^+}(r_2))^+ \psi_{1s}(r_2) \omega_{s_2} \qquad (10c)$$

 $(I_1^{s_1 \to s'_1})$ gives the **bound-free** transition of electron from spin s_1 to s'_1 by photon interaction. $(I_2^{s_2 \to s'_2})$ gives the **bound-bound** transition of electron from spin s_2 to s'_2 . It is to note that in singlet to singlet transition there is no spin-flip after interaction, i.e., $u_b \to u_f$ and $d_b \to d_f$. Substituting from (8) and Dirac-type hydrogen wave function [2] for $\psi_{1s}(r)$ in Eq. (10b) we get

$$I_1^{s_1 \to s_1'} = L_1 \sum_{n \ge n_0} T_n^{s_1 \to s_1'} \exp i\{-l_{B1} + p_0 + (\lambda_p - n)k_0\}r_0 \quad (10d)$$

$$T_{n}^{s_{1} \to s_{1}'} = [\bar{u}_{p}^{s_{1}'} \mathcal{B}_{n}^{*} \mathcal{V}(q) \omega_{s_{1}}], \qquad (10e)$$

where $L_1 = N_{p_0} N_{1S} C_0(q) \frac{A_0}{2c}$.

The momentum transfer $\vec{q} = \vec{p} + (\lambda_p - n)\vec{k} = q\hat{q}$ and from [2]

$$k \cdot p = k_0 p_0 - \vec{k} \cdot \vec{p}, \quad p_0 = (\sqrt{c^2 + \vec{p}^2})$$

 l_{1B} =four-momentum of each of the bound electron in the ground state of He.

and l_{2B} is the four-momentum of the bound electron in the ground state of He⁺

$$l_{1B} = \sqrt{c^2 - p_{1B}^2} \text{ and } l_{2B} = \sqrt{c^2 - p_{2B}^2}$$

$$V(q) = [1, \beta_1'g(q)\hat{q}]$$

$$(q) = \frac{4\pi}{q} \frac{\Gamma(\gamma_1' + 1)}{(p_{1B}^2 + q^2)^{\frac{\gamma_1' + 1}{2}}} \sin\left[(\gamma_1' + 1)\tan^{-1}\left(\frac{q}{p_{1B}}\right)\right]$$

$$g(q) = \left\{\frac{p_{1B}}{q} - \frac{\gamma_1' + 1}{\gamma_1'}\left[1 + \left(\frac{p_{1B}}{q}\right)^2\right]^{1/2} \\ \times \frac{\sin[\gamma_1' \tan^{-1}(q/p_{1B})]}{\sin[(\gamma_1' + 1)\tan^{-1}(q/p_{1B})]}\right\}$$

$$N_{1s} = (2p_{1B})^{(\gamma_1' + 1/2)}\left[\frac{1 + \gamma_1'}{8\pi\Gamma(1 + 2\gamma_1')}\right]^{1/2},$$

$$N_{1s}' = (2p_{2B})^{(\gamma_2' + 1/2)}\left[\frac{1 + \gamma_2'}{8\pi\Gamma(1 + 2\gamma_2')}\right]^{1/2}$$

$$p_{1B} = z_1 = 27/16, \quad p_{2B} = z_2 = 2$$

$$B_{n0} = \frac{eA_0/2}{2\vec{k} \cdot \vec{p}}k_0\{2J_n + \cos\xi(J_{n-2} + J_{n+2})\}$$

$$\vec{B}_n = \lfloor (J_{n+1}\hat{\varepsilon}^* + J_{n-1}\hat{\varepsilon}) + kB_{n0} \rfloor$$

Similarly we get for (10c) as in [3]

$$(I_2^{s_2 \to s'_2}) = L_2 \delta_{s'_2 s_2} \exp i(l_{B2} - l_{B1})r_0, \qquad (10f)$$

where

 C_0

$$L_2 = J(1 - \beta_1'\beta_2'), \quad J = N_{1s}'N_{1s} \frac{4\pi\Gamma(\gamma_1' + \gamma_2' + 1)}{(|p_{1B}| + |p_{2B}|)^{\gamma_1' + \gamma_2'}}$$

 $(I_2^{s_2 \rightarrow s'_2})$ exists if $s'_2 = s_2$ and we write

$$I_2^{a_b \to a_b} = I_2^{a_b \to a_b} = I_2 = L_2 \exp i(l_{B2} - l_{B1})r_0$$

Finally integrating (10a) over r_0

$$S_{Dir}^{s_1,s_2 \to s_1',s_2'} = \int (I_1^{s_1 \to s_1'})(I_2^{s_2 \to s_2'})dr_0$$

= $L_1 L_2 \sum_{n \ge n_0} T_n^{s_1 \to s_1'} \delta[l_B + p_0 + (\lambda_p - n)k_0]$
 $l_B = l_{2B} - 2l_{1B}$ (10g)

Defining binding energy of each of the electrons in the initial and in the final states as ε_1 and ε_2 , respectively, and the kinetic energy as ε_{kin} , where

$$\varepsilon_1 = c(c - l_{1B}), \quad \varepsilon_2 = c(c - l_{2B}), \text{ and } \varepsilon_{kin} = c(p_0 - c).$$

Since photon frequency $\omega = ck_0$,

we can write

$$l_B + p_0 + (\lambda_p - n)k_0 = [\varepsilon_B + \varepsilon_{kin} + (\lambda_p - n)\omega]/c.$$

Delta function in (10g) gives

$$[2\varepsilon_1 - \varepsilon_2 + \varepsilon_{kin} + (\lambda_p - n)\omega] = 0.$$

Hence, energy conservation relation

$$\varepsilon_B + \varepsilon_{kin} + \lambda_p \omega = n\omega, \qquad (10h)$$

where $\varepsilon_B = 2\varepsilon_1 - \varepsilon_2$ is the ionization potential.

The terms of $T_n^{s_1 \rightarrow s_1'}$ (10e) for spin-symmetric transitions are

$$\begin{aligned} I_n^{u_b \to u_f} &= B_{0n}^*(m_1 + m_2 S_1 \hat{p} \cdot \hat{q}) - B_n^* \cdot (m_1 S_1 \hat{q} + m_2 \hat{p}) \\ &+ i \{ B_n^* \times (-m_1 S_1 \hat{q} + m_2 \hat{p}) \}_z \end{aligned}$$
(10h1)

and

$$T_n^{d_b \to d_f} = B_{0n}^*(m_1 + m_2 S_1 \hat{p} \cdot \hat{q}) - B_n^* \cdot (m_1 S_1 \hat{q} + m_2 \hat{p}) - i\{B_n^* \times (-m_1 S_1 \hat{q} + m_2 \hat{p})\}_z$$
(10h2)

 $T_{n}^{s_{1} \rightarrow s_{1}'}$ for spin-flip terms

$$\begin{aligned} T_n^{u_b \to d_f} &= B_{0n}^* m_2 S_1 [i(\hat{p} \times \hat{q})_x - (\hat{p} \times \hat{q})_y] \\ &+ i [B_n^* \times (-m_1 S_1 \hat{q} + m_2 \hat{p})]_x \\ &- i \{B_n^* \times (-m_1 S_1 \hat{q} + m_2 \hat{p})\}_y \end{aligned}$$
(10h3)

and

$$\begin{split} T_n^{d_b \to u_f} &= B_{0n}^* m_2 S_1 [i(\hat{p} \times \hat{q})_x + (\hat{p} \times \hat{q})_y] \\ &+ i [B_n^* \times (-m_1 S_1 \hat{q} + m_2 \hat{p})]_x \\ &+ i \{B_n^* \times (-m_1 S_1 \hat{q} + m_2 \hat{p})\}_y, \end{split} \tag{10h4}$$

where $S_1 = \beta'_1 g(q)$.

B. Singlet to triplet transition

Singlet to triplet $(S_f=1)$ transition matrix $(S_{s_1s_2 \rightarrow s_1's_2'})_{S_f \land M_f}$ consists of three matrices $(S_{s_1s_2 \rightarrow s_1's_2'})_{1,0}$, $(S_{s_1s_2 \rightarrow s_1's_2'})_{1,1}$, and $(S_{s_1s_2 \rightarrow s_1's_2'})_{1,-1}$, corresponding

to the three magnetic quantum numbers $M_f=0,1,-1$. Using (2), (3), and (5) in (1) it can be shown that

$$(S_{s_1s_2 \to s_1's_2'})_{1,0} = \left[\frac{1}{\sqrt{2}}(S_{Dir}^{u_b,d_b \to u_f,d_b}) - \frac{1}{\sqrt{2}}(S_{Dir}^{d_b,u_b \to d_f,u_b})\right].$$
(11)

Using (2), (3), and (6) in (1) we get

$$(S_{s_1s_2 \to s_1's_2'})_{1,1} = \lfloor (S_{Dir}^{d_b, u_b \to u_f, u_b}) - (S_{shk}^{u_b, d_b \to u_f, u_b}) \rfloor.$$
(12)

Using the Eqs. (2), (3), and (7) in (1) we get

$$(S_{s_1s_2 \to s_1's_2'})_{1,-1} = [(S_{Dir}^{u_b, d_b \to d_f, d_b}) - (S_{shk}^{d_b, u_b \to d_f, d_b})].$$
(13)

 $S_{Dir}^{d_b,u_b \to u_f,u_b}$ and $S_{Dir}^{u_b,d_b \to d_f,d_b}$ in (12)and (13) are the *spin-flip* direct-terms (10g). These are to be obtained using (10h3) and (10h4)

The general matrix element for shake-off term $(S_{shk}^{s_1,s_2 \to s_1',s_2'})$ is

$$(S_{shk}^{s_1,s_2 \to s_1',s_2'}) = \int dr_0 (I_3^{s_1 \to s_1'}) I_4^{s_2 \to s_2'},$$
(14)

where

$$(I_{3}^{s_{1} \to s_{1}'}) = \int d^{3}r_{1} [\bar{\omega}_{s_{1}'} [\psi_{1s}^{He^{+}}(r_{1})]^{\dagger} \gamma(1) A(r_{1}) \psi_{1s}(r_{1}) \omega_{s_{1}}]$$
(14a)

$$I_4^{s_2 \to s_2'} = \int d^3 r_2 \{ \bar{u}_{s_2'}(p) [\psi_p(r_2)]^{\dagger} \psi_{1s}(r_2) \omega_{s_2} \}.$$
 (14b)

 $(I_3^{s_1 \rightarrow s_1'})$ gives the *bound-bound* transition of electron from spin s_1 to s'_1 by direct photon interaction. $I_4^{s_2 \rightarrow s'_2}$ gives the *bound-free shake-off* transition of electron from spin s_2 to s'_2 .

$$(I_{3}^{s_{1} \to s_{1}'}) = N_{1s}' N_{1s} \frac{A_{0}}{2c} \{C_{0}'(k)g'(k)\} [-\sqrt{2}(\beta_{1}' + \beta_{2}')] \\ \times \exp[i(l_{B2} - l_{B1})r_{0}], \qquad (14a1)$$

where $C_0(k) \rightarrow C'_0(k)$ and $g(k) \rightarrow g'(k)$ as $\gamma'_1 \rightarrow (\gamma'_1 + \gamma'_2 - 1)$ and $p_{1B} \rightarrow (p_{1B} + p_{2B})$ in equations be-low (10e).

We find $(I_3^{s_1 \rightarrow s_1'})$ correspond to the spin-flip terms in bound-bound transition by photon interaction. Hence, the non-null terms are

$$I_{3}^{d_{b} \to u_{b}} = I_{3}^{u_{b} \to d_{b}} = I_{3} = L_{3} \exp[i(l_{B2} - l_{B1})r_{0}] \quad (14a2)$$
$$L_{3} = N_{1s}' N_{1s} \frac{A_{0}}{2c} \{C_{0}'(k)g'(k)\} [-\sqrt{2}(\beta_{1}' + \beta_{2}')].$$

Using Eqs. (8), (9), and (14b) becomes

$$I_4^{s_2 \to s_2'} = L_4 \sum_{n \ge n_0} T_n'^{s_2 \to s_2'} \exp i[-l_{1B} + p_0 + (\lambda_p - n)k_0]r_0.$$
(14c)

Integrating over r_0 Eq. (14) becomes

$$(S_{shk}^{s_1,s_2 \to s_1',s_2'}) = L_3 L_4 \sum_{n \ge n_0} (T_n')^{s_2 \to s_2'} \delta[l_B + p_0 + (\lambda_p - n)k_0],$$
(14d)

where

$$T_{n}^{\prime s_{2} \to s_{2}^{\prime}} = \bar{u}_{s_{2}^{\prime}}(p) f_{n}^{+} \mathcal{V}(q) \omega_{s_{2}}$$
(14e)

and $V = [1, \beta'_1 g(q) \hat{q}], L_4 = N_{1s} N_{p_0} C_0(q).$



FIG. 1. Representative diagram showing the geometry of interaction (see text)

The spin-specific matrices corresponding (14e) are

$$(T'_{n})^{u_{b} \to u_{f}} = \left[J_{n}^{*}(a)\Delta + \frac{A_{0}/2}{2\vec{k} \cdot \vec{p}} \{ J_{n-1}^{*}(a)\Delta_{2} + J_{n+1}^{*}(a)\Delta_{1} \} \right]$$
$$(T'_{n})^{d_{b} \to d_{f}} = \left[J_{n}^{*}(a)\Delta + \frac{A_{0}/2}{2\vec{k} \cdot \vec{p}} \{ J_{n-1}^{*}(a)\Delta_{1} + J_{n+1}^{*}(a)\Delta_{2} \} \right],$$

where

$$\begin{split} \Delta &= \left(m_1 - \frac{m_2 \beta_1' g(q)}{|q|} [|p| + (\lambda - n)|k| \cos \theta] \right) e^{-in\phi} \\ \Delta_1 &= 2\beta_1' g(q) k_0 \bigg[(m_1 - m_2 \cos \theta) \frac{|\vec{p}|}{|\vec{q}|} \sin \theta \bigg] \frac{A_0/2}{2k \cdot p} e^{-in\phi} \\ \Delta_2 &= \bigg[-2m_2 k_0 \sin \theta \bigg[1 - \frac{\beta_1' g(q)}{|\vec{q}|} \{ |\vec{p}| \cos \theta + (\lambda - n) k_0 \} \bigg] \bigg] \frac{A_0/2}{2k \cdot p} e^{-in\phi}. \end{split}$$

III. CALCULATION OF IONIZATION RATES

In this calculation photon propagation and electron spin are taken along the z-direction (Fig. 1). Using above equations we shall calculate below the matrix elements for transition from the singlet to singlet state and the singlet to triplet state by +ve helicity beams. With change of helicity of the beam the electric and the magnetic field vectors which are in the plane of polarization (xy plane) change their directions

A. Singlet to singlet

The angular differential transition rate from singlet to singlet state is obtained using (10) and (10g) in (1a).

$$\frac{d(\Gamma)_{0,0}}{d\Omega} = |(S_{s_1 s_2 \to s_1' s_2'})_{0,0}|^2 \frac{cp_0 |\vec{p}|}{(2\pi)^2}$$
(15)

$$= \left(\frac{d(\Gamma)_{0,0}}{d\Omega}\right)_{Dir}^{u_b \to u_f} + \left(\frac{d(\Gamma)_{0,0}}{d\Omega}\right)_{Dir}^{d_b \to d_f}.$$
 (16)

Since from Eqs. (10a)–(10f) and (10)

$$(S_{s_1s_2 \to s_1's_2'})_{0,0} = \frac{1}{\sqrt{2}} L_1 L_2 \sum_{n \ge n_0} (T_n^{u_b \to u_f} + T_n^{d_b \to d_f}) \\ \times \delta [l_B + p_0 + (\lambda_p - n)k_0]$$

we have

$$\left(\frac{d(\Gamma)_{0,0}}{d\Omega}\right)_{Dir}^{s \to s'} = \sum_{n \ge n_0} \left(\frac{1}{\sqrt{2}}L_1 L_2\right)^2 |T_n^{s \to s'}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}.$$
(16a)

B. Singlet to triplet state

The transition rate from singlet state to triplet $S_f=1$, M_f =0

is written as $(\Gamma)_{1,0}$.

$$\frac{d(\Gamma)_{1,0}}{d\Omega} = |(S_{s_1s_2 \to s_1's_2'})_{1,0}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}$$
$$= \left(\frac{d(\Gamma)_{1,0}}{d\Omega}\right)_{Dirt}^{u_b \to u_f} + \left(\frac{d(\Gamma)_{1,0}}{d\Omega}\right)_{Dir}^{d_b \to d_f}.$$
 (17)

From the Eq. (11)

$$(S_{s_1s_2 \to s_1's_2'})_{1,0} = \frac{1}{\sqrt{2}} L_1 L_2 \sum_{n \ge n_0} (T_n^{u_b \to u_f} - T_n^{d_b \to d_f}) \\ \times \delta[l_B + p_0 + (\lambda_p - n)k_0].$$
(17a)

There is only a phase difference of π between the 2nd term of $(S_{s_1s_2 \to s'_1s'_2})_{0,0}$ and $(S_{s_1s_2 \to s'_1s'_2})_{1,0}$. Hence from (16) and (17)

$$\frac{d(\Gamma)_{1,0}}{d\Omega} = \frac{d(\Gamma)_{0,0}}{d\Omega}.$$

The transition rate $(\Gamma)_{1,1}$ from the singlet-state to the tripletstate $S_f = 1$, $M_f = 1$ is written as

$$\frac{d(\Gamma)_{1,1}}{d\Omega} = |(S_{s_1s_2 \to s_1's_2'})_{s_f=1,M_f=1}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2} = \left(\frac{d(\Gamma)_{1,1}}{d\Omega}\right)_{\text{direct-flip}}^{d_b \to u_f} + \left(\frac{d(\Gamma)_{1,1}}{d\Omega}\right)_{\text{shake-off}}^{u_b \to u_f}.$$
 (18)

On using (10g) and (14d) in (12)

$$(S_{s_{1}s_{2} \to s_{1}'s_{2}'})_{1,1} = 2 \frac{1}{\sqrt{2}} (S_{Dir}^{d_{b},u_{b} \to u_{f},u_{b}}) - 2 \frac{1}{\sqrt{2}} (S_{shk}^{u_{b},d_{b} \to u_{f},u_{b}})$$
$$= 2 \frac{1}{\sqrt{2}} \bigg[L_{1}L_{2} \sum_{n \geq n_{0}} (T_{n}^{d_{b} \to u_{f}}) - L_{3}L_{4} \sum_{n \geq n_{0}} (T_{n}')^{u_{b} \to u_{f}} \bigg]$$
$$\times \delta [l_{B} + p_{0} + (\lambda_{p} - n)k_{0}].$$
(18a)

Hence in (18)

$$\left[\frac{d(\Gamma)_{1,1}}{d\Omega}\right]_{\text{direct-flip}}^{d_b \to u_f} = \sum_{n \ge n_0} \left(2\frac{1}{\sqrt{2}}L_1L_2\right)^2 |T_n^{d_b \to u_f}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}$$
(18b)

and

$$\left[\frac{d(\Gamma)_{1,1}}{d\Omega}\right]_{\text{shake-off}}^{u_b \to u_f} = \sum_{n \ge n_0} \left(2\frac{1}{\sqrt{2}}L_3L_4\right)^2 |(T'_n)^{u_b \to u_f}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}.$$
(18c)

Here Eqs. (18b) and (18c) produce spin-up currents (u_f) . The first term of (18) gives spin-up current (u_f) by spin-flip of the bound electron of spin (d_b) by direct interaction, the other electron remains in the same spin state (u_b) . The 2nd term of (18) gives shake-off current $u_b \rightarrow u_f$ while the bound electron (d_b) suffers spin-flip to (u_b) by direct interaction.

The angular differential transition rate to $S_f=1$, $M_f=-1$ is written as.

$$\frac{d(\Gamma)_{1,-1}}{d\Omega} = |(S_{s_1s_2 \to s_1's_2'})_{1,-1}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2} \\
= \left[\frac{d(\Gamma)_{1,-1}}{d\Omega}\right]_{\text{direct-flip}}^{u_b \to d_f} + \left[\frac{d(\Gamma)_{1,-1}}{d\Omega}\right]_{\text{shake-off}}^{d_b \to d_f}.$$
(19)

On using (10g) and (14d) in (13)

$$(S_{s_{1}s_{2} \to s_{1}'s_{2}'})_{1,-1} = 2 \frac{1}{\sqrt{2}} (S_{Dir}^{u_{b},d_{b} \to d_{f},d_{b}}) - 2 \frac{1}{\sqrt{2}} (S_{shk}^{d_{b},u_{b} \to d_{f},d_{b}})$$
$$= 2 \frac{1}{\sqrt{2}} \bigg[L_{1}L_{2} \sum_{n \ge n_{0}} (T_{n}^{u_{b} \to d_{f}})$$
$$- L_{3}L_{4} \sum_{n \ge n_{0}} (T_{n}')^{d_{b} \to d_{f}} \bigg]$$
$$\times \delta [I_{B} + p_{0} + (\lambda_{p} - n)k_{0}].$$
(19a)

Hence

$$\left[\frac{d(\Gamma)_{1,-1}}{d\Omega}\right]_{\text{direct-flip}}^{u_b \to d_f} = \sum_{n \ge n_0} \left(2\frac{1}{\sqrt{2}}L_1L_2\right)^2 |T_n^{u_b \to d_f}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}$$
(19b)

and

$$\left[\frac{d(\Gamma)_{1,-1}}{d\Omega}\right]_{\text{shake-off}}^{d_b \to d_f} = \sum_{n \ge n_0} \left(2\frac{1}{\sqrt{2}}L_3L_4\right)^2 |(T'_n)^{d_b \to d_f}|^2 \frac{cp_0|\vec{p}|}{(2\pi)^2}.$$
(19c)

Here both the terms of (19) produce spin-down currents (d_f) . The 1st term contributes spin-flip current by direct interaction and the 2nd term contributes shake-off current (d_f) the bound electron (u_b) suffers spin-flip to (d_b) by direct interaction.

IV. RESULTS AND DISCUSSIONS

Total angular differential rate $\frac{dW}{d\Omega}$ is obtained by taking average over the initial spin states and sum over the final spin states and multiplying the rate thus obtained by the square of the Coulomb-correction factor (CC).



FIG. 2. Total ionization rate in atomic unit (a.u.) vs intensity at different frequencies. Present theory: 2.02 eV (continuous line with filled circles), 2.5 eV (broken line with open circle), 3 eV (line with filled triangle). Coulomb corrected KFR theory [4]: 2.02 eV (continuous line).

$$\frac{dW}{d\Omega} = C^2(Z, \varepsilon_B, F) \left\{ \frac{1}{4} \frac{d(\Gamma)_{0,0}}{d\Omega} + \frac{3}{4} \left[\frac{d(\Gamma)_{1,0}}{d\Omega} + \frac{d(\Gamma)_{1,1}}{d\Omega} + \frac{d(\Gamma)_{1,-1}}{d\Omega} \right] \right\}.$$
(20)

The Coulomb-correction factor [4]

$$C(Z, \varepsilon_B, F) = \left[\frac{(2\varepsilon_B)^{3/2}}{F}\right]^{Z/\sqrt{2\varepsilon_B}}$$

where $\varepsilon_B = \varepsilon_2 - 2\varepsilon_1$ is the ionization potential (10h), $F = A_0 \omega/c$ is the peak field strength

and $Z=z_1$ (=27/16) is the core charge of the atom.

The first term of (20) is the contribution from singlet to singlet transition and the square bracket in (20) is the contribution from singlet to triplet transition. The total transition rate is obtained by integrating (20) over solid angle Ω .

In Fig. 2 the present result for the total transition rate Γ is shown for frequencies 2.02, 2.5, and 3 eV. At any intensity, the rate increases with frequency. At each frequency present rate attains peak value depending on intensity. The peaks shift toward higher intensity as frequency increases. Drop of the rate curve after attaining the peak shows start of the stabilization with the increase in intensity [7]. Total Rate at each intensity is obtained by summing the rates over photon order $n \ge n_0$. The minimum photon number n_0 depends on intensity and frequency. In the present case we take $n=n_0$ +n'. Any increase in the value of n' above 50 does not change the total rate any further. As intensity increases there is a competition between kinetic energy of the electron and the quiver energy leading to stabilization. The nonrelativistic Coulomb-corrected KFR result [4] for frequency 2.02 eV



FIG. 3. Total ionization rate vs. intensity at 20.2 eV beam frequency.

(614 nm) is also shown. Up to the intensity 10^{15} W cm⁻² present Coulomb-corrected relativistic result at 2.02 eV compares well with the nonrelativistic Coulomb-corrected KFR result. As intensity increases the nonrelativistic result overestimates the relativistic result. Figure 3 gives the total ionization rate at soft x-ray wavelength 61.4nm (20.2 eV) where stabilization starts after 10^{16} W/cm².

From (16) to (18) the Coulomb-corrected total up-current and the total down-current are, respectively,

$$\begin{bmatrix} \underline{d(W)} \\ \underline{d\Omega} \end{bmatrix}^{u_p} = C^2(Z, \varepsilon_B, F) \left\{ \frac{1}{4} \begin{bmatrix} \underline{d(\Gamma)}_{0,0} \\ \underline{d\Omega} \end{bmatrix}^{u_b \to u_f}_{Dir} + \frac{3}{4} \begin{bmatrix} \underline{d(\Gamma)}_{1,0} \\ \underline{d\Omega} \end{bmatrix}^{u_b \to u_f}_{Dir} + \frac{3}{4} \begin{bmatrix} \underline{d(\Gamma)}_{1,1} \\ \underline{d\Omega} \end{bmatrix} \right\}$$
(21)

and

$$\begin{bmatrix} \underline{d(W)} \\ d\Omega \end{bmatrix}^{down} = C^2(Z, \varepsilon_B, F) \left\{ \frac{1}{4} \begin{bmatrix} \underline{d(\Gamma)}_{0,0} \\ d\Omega \end{bmatrix}_{Dir}^{d_b \to d_f} + \frac{3}{4} \begin{bmatrix} \underline{d(\Gamma)}_{1,0} \\ d\Omega \end{bmatrix}_{Dir}^{d_b \to d_f} + \frac{3}{4} \begin{bmatrix} \underline{d(\Gamma)}_{1,-1} \\ d\Omega \end{bmatrix} \right\}.$$
(22)

Angular spin-asymmetry parameter $\langle A \rangle$ between spin-up current and spin-down current is defined as the ratio

$$\langle A \rangle = \frac{\left(\frac{dW}{d\Omega}\right)^{up} - \left(\frac{dW}{d\Omega}\right)^{down}}{\left(\frac{dW}{d\Omega}\right)^{up} + \left(\frac{dW}{d\Omega}\right)^{down}}.$$
 (23)

Spin-flip currents are obtained from the singlet to triplet transitions with $M_f=1,-1$ and are given respectively from Eqs. (18) and (19) and are defined as



FIG. 4. Asymmetry parameter $\langle A \rangle$ vs angle (deg) of electron emission from the incident beam direction for photon frequency 2.02 eV. Outer curve is for laser intensity 10^{13} W/cm² and the inner curve is for laser intensity 10^{14} W/cm².

$$\left[\frac{d(W)}{d\Omega}\right]^{d_b \to u_f} = C^2(Z, \varepsilon_B, F) \left[\frac{d(\Gamma)_{1, -1}}{d\Omega}\right]^{d_b \to u_f}_{\text{direct-flip}}$$
(24)

and

$$\left[\frac{d(W)}{d\Omega}\right]^{u_b \to d_f} = C^2(Z, \varepsilon_B, F) \left[\frac{d(\Gamma)_{1,-1}}{d\Omega}\right]^{u_b \to d_f}_{\text{direct-flip}}.$$
 (25)

We define spin-flip asymmetry parameter "A" as the ratio

$$A = \frac{\left[\frac{d(W)}{d\Omega}\right]^{d_b \to u_f} - \left[\frac{d(W)}{d\Omega}\right]^{u_b \to d_f}}{\left(\frac{dW}{d\Omega}\right)^{u_p} + \left(\frac{dW}{d\Omega}\right)^{down}}$$
(26)

Angular dependents of $\langle A \rangle$ and "A" are shown in the Figs. 4–7. The Fig. 4 gives the angular asymmetry parameter $\langle A \rangle$ at 10¹³ and 10¹⁴ W/cm². The absolute value of $\langle A \rangle$ at any angle increases with intensity as in [2]. Similar increase in the absolute value of spin-flip parameter A with intensity is found in Fig. 5. At soft x-ray wavelength spin-asymmetry parameter and spin-flip parameter are shown in Fig. 6 and Fig. 7, respectively, for 10¹⁶, 10¹⁷, and 10¹⁸ W/cm². It is to note that asymmetry decreases as frequency increases. The absolute value of asymmetry obtained at 20.2 eV frequency (70–110 deg) are quite large of the order of 20% in magnitude and they are well within the current resolution of spinanalyzers in the laboratory. The total angular differential rate (20) from direct interaction and shake-off interaction is shown in Fig. 8.

The spin-symmetric currents $u_b \rightarrow u_f$ and $d_b \rightarrow d_f$ from the direct-interaction gives the total current $u_b \rightarrow u_f$ from (16a) and (18b)



FIG. 5. Flip-parameter "A" vs angle (deg) of electron emission from incident beam direction for photon frequency 2.02 eV. Outer curve is for the intensity 10^{14} W/cm² and the inner curve is for the intensity 10^{15} W/cm².

$$\left[\frac{d(W)}{d\Omega}\right]^{u_b \to u_f} = C^2(Z, \varepsilon_B, F) \left\{ \frac{1}{4} \left[\frac{d(\Gamma)_{0,0}}{d\Omega}\right]^{u_b \to u_f}_{Dir} + \frac{3}{4} \left[\frac{d(\Gamma)_{1,0}}{d\Omega}\right]^{u_b \to u_f}_{Dir} \right\}$$
(27)

and the total current $d_b \rightarrow d_f$ from (16) and (19b)

$$\begin{bmatrix} \underline{d(W)} \\ d\Omega \end{bmatrix}^{d_b \to d_f} = C^2(Z, \varepsilon_B, F) \left\{ \frac{1}{4} \begin{bmatrix} \underline{d(\Gamma)}_{0,0} \\ d\Omega \end{bmatrix}^{d_b \to d_f}_{Dir} + \frac{3}{4} \begin{bmatrix} \underline{d(\Gamma)}_{1,0} \\ d\Omega \end{bmatrix}^{d_b \to d_f}_{Dir} \right\}.$$
(28)

The currents from direct interaction and shake-off interaction are shown (Fig. 8).



FIG. 6. Same as in Fig. 4 for photon frequency 20.2 eV. The upper, middle and the lower curves are for intensities 10^{16} , 10^{17} , and 10^{18} W/cm², respectively.



FIG. 7. Same as in Fig. 5 for photon frequency 20.2 eV. The upper, middle and the lower curves are at intensities 10^{16} , 10^{17} , and 10^{18} W/cm², respectively.

The currents (27) and (28) from the direct interaction are identical (Fig. 8) but less than the shake-off currents from (18c) and (19c), which are also of the same magnitude. The flip-current $d_b \rightarrow u_f$ from (18) is almost 10 order less than the flip-current $u_b \rightarrow d_f$ from (19). This explains why the values of $\langle A \rangle$ and A are negative but of equal magnitude (see Figs. 4, 5 and Figs. 6, 7). However flip-currents have peaks (Fig. 8) above and below the plane of polarization and minima on the plane of polarization, while $u_b \rightarrow u_f$ and $d_b \rightarrow d_f$ currents have maxima on the plane of polarization.

In the present case, there is no spin-orbit interaction arising from the derivative of the atomic potential, either in the initial state (ground *s* state) or in the final state (Dirac plane-wave Volkov state). In intense laser the high-magnetic field $\vec{B} = \frac{\vec{k} \times \vec{E}}{|\vec{k}|}$ in the laboratory interacts with the magnetic moment (= $-\frac{1}{4c^2}\sigma$ a.u.) of the bound electron and may cause



FIG. 8. Differential spin rates (per sec) vs angle in degree of the electron emission from the beam direction for beam frequency 2.02 eV at intensity 1.4×10^{15} W/cm² for direct and shake-off interactions (see text). Direct interaction: $u_b \rightarrow u_f$ line with open circles; $d_b \rightarrow d_f$ line with filled-squares; $u_b \rightarrow d_f$ (spin-flip) line with open square; $d_b \rightarrow u_f$ (spin-flip) line with filled-circles. Shake-off interaction: $u_b \rightarrow u_f$ line with open triangles; $d_b \rightarrow d_f$ line with open diamond. Sum of spin rates (direct and shake-off) broken line with cross.

spin-flip of the bound electron (14a1). On ionization the electric field in the laboratory is Lorentz transformed [8], in the moving frame of the emitted electron, into a (motional) magnetic field component $\vec{B'} \cong \vec{E} \times \vec{p}/c$ a.u. Coupling of $\vec{B'}$ with the magnetic moment may provide further torque to the spinning electron causing spin-flip even if the retardation effect on the electron is switched off $(\vec{k}=\vec{0})$, as is evident from the expressions $T_n^{u_b \rightarrow d_f}$ and $T_n^{d_b \rightarrow u_f}$ of (10e), further, the weak components of the Dirac wave-functions in the free and the bound states contain factors such as m_2 and $\beta'_1g(q)$, respectively. These factors are largely responsible for spin currents and necessary flip.

V. CONCLUSION

We have calculated spin-dependent multiphoton ionization current from the ground state of He by intense circularly polarized laser. Considering the effect of the Coulomb field of the ion on the relativistic rate good agreement is obtained with the KFR rate in low intensity regime. As intensity increases nonrelativistic KFR rate overestimates the relativistic rate. Spin-flip current $u_b \rightarrow d_f$ is higher than the spinsymmetric currents $u_b \rightarrow u_f$ or $d_b \rightarrow d_f$ except around the polarization plane. The spin-direction of in spin-specific current can be altered from outside by changing the helicity of the beam. Hence, asymmetry $\langle A \rangle$ will change its sign on changing the helicity of the beam from left circular polarization to right circular polarization. At any frequency stabilization against ionization starts with the increase in intensity causing drop in the ionization curve. The peak value of the rate decreases with frequency while the position of the peak shifts toward the higher intensity side.

An interesting application of this formalism is in calculating the probability of formation of orthopositronium (otho-Ps) and parapositronium (para-Ps) from positronium negative ion (Ps⁻) [9]. Most stable (Ps⁻) forms in the singletstate. On multiphoton ionization the final state consists of a continuum electron of spin u or d and positronium para-Ps or otho-Ps. Para-Ps thus formed can be quenched into two photons in presence of a spectator electron. The formalism is used in the case of ionization by high-energy single photon [3] derived from circularly polarized laser taking only the plane wave part of the Volkov solution. In the present case of multiphoton ionization we have considered full-Volkov solution for the electron and the number of photons $(n \ge 1)$ involved in the process depend on the intensity and the frequency of the laser field. Hence importance of this field theoretic formalism lies in calculating the probability of spin-polarized current from the two-electron atom or ion by the polarized laser.

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