Spin Asymmetry in an Intense-Field Ionization Process

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A relativistic analysis of the rate of spin flips in ionization of an ensemble of Dirac H atoms subjected to intense circularly polarized laser fields is made. A remarkable intensity-dependent asymmetry between the spin up and spin down electron currents is found. It is nonzero even when the retardation effect, hence the magnetic component of the field, as well as the spin-orbit interaction responsible for the well-known Fano effect, is negligible. Transformation properties of the amplitudes show that the sign of asymmetry can be controlled by changing the helicity of the laser photons from outside.

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The advent of very intense laser sources at near-infrared (Ti:sapphire laser, e.g., [1]) and shorter than optical wavelengths (free-electron laser, e.g., [2]) has stimulated relativistic investigations of laser-atom interaction dynamics in intense fields within the framework of the Dirac theory. Thus, for example, reinvestigations of spinor-electron wave-packet motion (e.g., [3]), Klein paradox [4], bound-state spin oscillation [5], Mott scattering (e.g., [6–8]), and Möller-scattering [9] have revealed new features of relativistic dynamics in intense fields.

Ionization in strong fields has been investigated within the framework of Dirac theory in the past but mainly for the spin unresolved currents (e.g., [10,11]). Little is known about spin resolved electron currents from ionization of an ensemble of unpolarized ground-state atoms or about the spin-flip ionization probability in intense fields. In this Letter we report on the result of the analysis of spin-flip probabilities and the up spin and down spin electron currents from ionization of an ensemble of Dirac H atoms subjected to intense circularly polarized laser radiation. Explicit analytical formulas are derived, and results of numerical calculations are presented using the ground-state Dirac wave function, and the Dirac-Volkov wave function with no spin-orbit interaction in the final state [12]. The spin-orbit interaction in the ground state is identically zero. The results reveal an intensity-dependent asymmetry between the up- and down-spin currents in any direction from the laser propagation axis. The asymmetry is shown to be nonzero even when the retardation effect and the spin-orbit interaction, which is responsible for the well-known Fano effect [13], are negligible.

Since spin is essentially relativistic in nature, therefore, for the present purpose we shall describe its interaction with the radiation field using a straightforward relativistic generalization (e.g., [10]) of the intense-field S-matrix method, which has been used very fruitfully in its nonrelativistic version (so-called Keldysh-Faisal-Reiss theory [14]) for atomic and molecular ionization (e.g., [15]) in the past. The leading term of the resulting S-matrix series for the transition amplitude for ionization from a bound state $\psi_{1s}^{(s)}(\vec{r}, t)$ of spin s [16] into the Dirac-Volkov continuum states $\psi_{\vec{p}}^{(s')}(\vec{r}, t)$ of spin s' [17], where $(s, s') =$ u (up) or d (down), is given by (in a.u.: $\hbar = |e| = m = a_0 = 1, c = \alpha^{-1}$),

$$S_{s \to s'} = -i \int_{-\infty}^{\infty} \langle \bar{\psi}_{\vec{p}}^{(s')}(t) | \gamma^{\mu} A_{\mu} | \psi_{1s}^{(s)}(t) \rangle \, dt. \qquad (1)$$

Explicit analytical expressions of the spin-specific probabilities of ionization per unit time, $\frac{dW_{s \to s'}}{d\Omega}$, in the direction (θ, ϕ) in an element of solid angle $d\Omega = \sin\theta d\theta d\phi$, have been derived recently [18] by evaluating Eq. (1), in the general case of an elliptically polarized electromagnetic field given by the vector potential $\vec{A} = A_0[\vec{\epsilon}_x \cos(\frac{\xi}{2}) \cos(\omega t - \vec{\kappa} \cdot \vec{r}) - \vec{\epsilon}_y \sin(\frac{\xi}{2}) \sin(\omega t - \vec{\kappa} \cdot \vec{r})]$. The spin-specific ionization rates of interest are

$$\frac{dW_{s \to s'}}{d\Omega} = \sum_{n \ge n_0} \left[\frac{A_0}{2c} N_{p_0} N_{1s} c_0(q) \right]^2 |t_{s \to s'}^{(n)}|^2 \frac{c p_0 |\vec{p}|}{(2\pi)^2}. \qquad (2)$$

The number of absorbed photons n is determined by the energy-momentum conservation relation, $n\omega = \epsilon_B + \epsilon_{\rm kin} + \lambda_p \omega$ where $\epsilon_B = c(c - \sqrt{c^2 - p_B^2})$ is the binding energy (ionization potential) and $\epsilon_{\rm kin} = c(\sqrt{c^2 + p^2} - c)$ is the kinetic energy. The spin-specific reduced $t^{(n)}$-matrix elements are

$$t_{u \to u}^{(n)} = B_n^{0*}[m_1 + m_2 g(q) \hat{p} \cdot \hat{q}] + \vec{B}_n^* \cdot [m_2 \hat{p} + m_1 g(q) \hat{q}] - i(\vec{B}_n^* \times [m_2 \hat{p} - m_1 g(q) \hat{q}])_z, \qquad (3)$$

$$t_{u \to d}^{(n)} = m_2 g(q) B_n^{0*}[i(\hat{p} \times \hat{q})_x - (\hat{p} \times \hat{q})_y] - i(\vec{B}_n^* \times [m_2 \hat{p} - m_1 g(q) \hat{q}])_x + (\vec{B}_n^* \times [m_2 \hat{p} - m_1 g(q) \hat{q}])_y, \qquad (4)$$

$$t_{d \to u}^{(n)} = m_2 g(q) B_n^{0*}[i(\hat{p} \times \hat{q})_x + (\hat{p} \times \hat{q})_y] - i(\vec{B}_n^* \times [m_2 \hat{p} - m_1 g(q) \hat{q}])_x - (\vec{B}_n^* \times [m_2 \hat{p} - m_1 g(q) \hat{q}])_y, \qquad (5)$$

$$t_{d \to d}^{(n)} = B_n^{0*}[m_1 + m_2 g(q) \hat{p} \cdot \hat{q}] + \vec{B}_n^* \cdot [m_2 \hat{p} + m_1 g(q) \hat{q}] + i(\vec{B}_n^* \times [m_2 \hat{p} - m_1 g(q) \hat{q}])_z. \qquad (6)$$

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where,

$$B_n^0 = \frac{A_0\kappa_0}{4c\kappa\cdot p}[2J_n + \cos\xi(J_{n+2} + J_{n-2})],$$
$$\vec{B}_n = \vec{\epsilon}(\xi)J_{n-1} + \vec{\epsilon}^*(\xi)J_{n+1} + \hat{\kappa}B_n^0,$$
$$\kappa\cdot p = \kappa_0 k_0 - \vec{\kappa}\cdot\vec{p}, \qquad \kappa = (\kappa_0, \vec{\kappa}), \qquad \kappa_0 = \omega/c.$$

$J_n \equiv J_n(a, b, \chi) = \sum_m J_{n+2m}(a)J_m(b)e^{i(n+2m)\chi}$ are generalized Bessel functions of three arguments with $a = \frac{A_0|\vec{\epsilon}(\xi)\cdot\vec{p}|}{c\kappa\cdot p}$, $b = \frac{A_0^2}{8c^2\kappa\cdot p}\cos\xi$, $\chi = \tan^{-1}[\tan\phi_p\tan(\xi/2)]$.

$$c_0(q) = \frac{4\pi}{q}\frac{\Gamma(\gamma'+1)}{(p_B^2+q^2)^{[(\gamma'+1)/2]}}\sin\left[(\gamma'+1)\tan^{-1}\left(\frac{q}{p_B}\right)\right], \tag{7}$$

$$g(q) = \beta'\left[\frac{p_B}{q} - \frac{\gamma'+1}{\gamma'}\times\sqrt{1+\left(\frac{p_B}{q}\right)^2}\frac{\sin[\gamma'\tan^{-1}(q/p_B)]}{\sin[(\gamma'+1)\tan^{-1}(q/p_B)]}\right]. \tag{8}$$

The other parameters are "field-dressed" electron momentum $\vec{q} \equiv q\hat{q} = \vec{p} + (\lambda_p - n)\vec{\kappa}$, $\lambda_p = \frac{A_0^2}{4c^2\kappa\cdot p}$; $\vec{\epsilon}(\xi) = [\vec{\epsilon}_x\cos(\frac{\xi}{2}) + i\vec{\epsilon}_y\sin(\frac{\xi}{2})]$, with $\xi[0, \pm\pi/2]$; $p_0 = \sqrt{c^2+\vec{p}^2} = (n-\lambda_p)\kappa_0 + \sqrt{c^2-p_B^2}$, and the ground-state momentum $p_B = Z$; $\gamma' = \sqrt{1-(Z\alpha)^2}$, $\beta' = (1-\gamma')/(Z\alpha)$, $m_1 = \sqrt{(p_0+c)/(2c)}$, $m_2 = -\sqrt{(p_0-c)/(2c)}$, and the normalization constants for the ground-state and the Volkov states, $N_{1s} = (2p_B)^{\gamma'+(1/2)}(\frac{1+\gamma'}{8\pi\Gamma(1+2\gamma')})^{1/2}$, and $N_{p_0} = \sqrt{\frac{c}{p_0}}$. The usual spin *unresolved* ionization rate for an unpolarized target atom, if desired, is easily obtained by simply adding the four spin-specific rates given above and dividing by 2 (for the average with respect to the two degenerate initial spin states),

$$\frac{d\Gamma^{(+)}}{d\Omega} = \frac{1}{2}\sum_{n\geq n_0}\left[\frac{A_0}{2c}N_{p_0}N_{1s}c_0(q)\right]^2 cp_0\frac{|\vec{p}|}{(2\pi)^2}\sum_{(s,s')=u,d}|t_{s\to s'}^{(n)}|^2. \tag{9}$$

We note that for linear polarization the above formulas [Eqs. (2)–(6)] hold with $\xi = 0$ and $J_n = J_n(a(\xi=0), b(\xi=0))$ (e.g., p. 12 of [19]); similarly, they hold for circular polarization with $\xi = \pm\pi/2$ and $J_n = J_n(a)e^{\pm in\phi_p}$, with $a = a(\xi = \pm\pi/2)$.

For the present purpose we shall restrict the calculations below to the case of a right circularly polarized electromagnetic field ($\xi = +\pi/2$). We choose the field propagation direction (z axis) as the quantization axis, with the spin "up" state defined to be along the positive z direction. The spin-up and the spin-down electron currents can now be obtained from Eqs. (2)–(6) as

$$\frac{dW^{\rm up}}{d\Omega} = \frac{1}{2}\left(\frac{dW_{u\to u}}{d\Omega} + \frac{dW_{d\to u}}{d\Omega}\right), \tag{10}$$

$$\frac{dW^{\rm down}}{d\Omega} = \frac{1}{2}\left(\frac{dW_{d\to d}}{d\Omega} + \frac{dW_{u\to d}}{d\Omega}\right). \tag{11}$$

Any asymmetry in the two currents is best characterized by the ensemble averaged asymmetry parameter $\langle A\rangle$ associated with the unpolarized target atoms, defined by

$$\langle A\rangle = \left(\frac{dW^{\rm up}}{d\Omega} - \frac{dW^{\rm down}}{d\Omega}\right)\Big/\left(\frac{dW^{\rm up}}{d\Omega} + \frac{dW^{\rm down}}{d\Omega}\right). \tag{12}$$

In Fig. 1 we show the result of calculations for the asymmetry parameter $\langle A\rangle$ as a function of the polar angle of the emitted electrons, for $\omega = 1.55$ eV at two laser intensities, $I = 10^{16}$ W/cm^2 (outer curve) and $I = 10^{17}$ W/cm^2 (inner curve). Remarkably, unlike the asymmetry parameter for the Fano effect [13], the two curves reveal a strong dependence of $\langle A\rangle$ on field intensity at all angles. The absolute size of $\langle A\rangle$ is larger for the higher intensity at all angles. The "peak values" are seen to occur not on the plane of polarization ($\theta = 90°$) but at a somewhat smaller angle from it. It is also seen to move farther away from the polarization plane with increasing intensity [20]. This behavior is because of the change of the electron momentum in an intense field caused by the combined effect of retardation and field intensity. This can be seen from the expression of the "dressed" momentum $\vec{q}$ of the electron in the field [see below Eq. (8)] where the extra term that adds to $\vec{p}$ depends on $n\vec{\kappa}$ and on intensity via $(A_0/c)^2 = I/\omega^2$ (a.u.) in $\lambda_p\vec{\kappa}$. In Fig. 1 for both intensities $\langle A\rangle$ is mostly as large as $O(10^{-3})$ in magnitude but negative. Clearly, the negative sign indicates a dominance of the spin-down electron current over the spin-up current at all angles. To understand the dominance we examine the corresponding spin-flip asymmetry parameter A. It is defined as the difference at a given

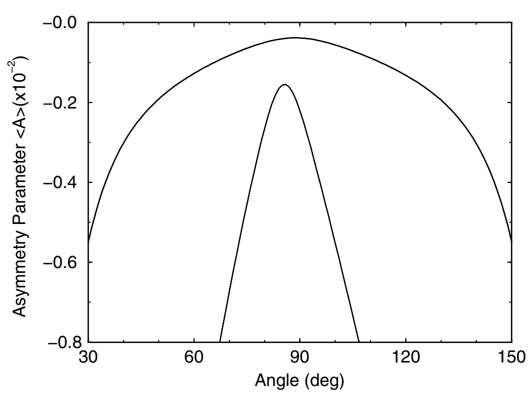

FIG. 1. Intensity-dependent ensemble averaged asymmetry parameter $\langle A\rangle$ [see Eq. (12) for definition] vs electron emission angle; $\omega = 1.55$ eV, $I = 10^{16}$ W/cm^2 (outer curve), and $I = 10^{17}$ W/cm^2 (inner curve).

angle between the spin-flip rates in the opposite directions scaled by the total rate at the same angle,

$$A = \left(\frac{dW_{u\to d}}{d\Omega} - \frac{dW_{d\to u}}{d\Omega}\right) \Big/ \left(\frac{dW^{\rm up}}{d\Omega} + \frac{dW^{\rm down}}{d\Omega}\right). \quad (13)$$

The calculated results are shown in Fig. 2. At both the intensities A is positive implying clearly that the $u \to d$ flip rate is greater than the $d \to u$ flip rate. We note that the angular dependences of A and $\langle A \rangle$ in Figs. 1 and 2 are rather similar (but for the opposite sign). Also the magnitude of the asymmetry A is comparable to that of $\langle A \rangle$. Thus the dominance of the $u \to d$ spin-flip rate itself over the $d \to u$ rate (for the present choice of the field polarization) leads to the negative values of $\langle A \rangle$ despite the 50/50 weighting of the two initial spin states in the latter case [21]. These characteristics of the spin asymmetry observed above holds also in other frequency and intensity domains. Thus, in Figs. 3 and 4 we show the corresponding results for the ensemble averaged asymmetry $\langle A \rangle$ and the spin-flip asymmetry A for a vacuum ultraviolet (VUV) frequency, $\omega = 20$ eV, at an intensity $I = 10^{20}$ W/cm^2. The general behavior of both $\langle A \rangle$ and A is similar to that in Figs. 1 and 2 for $\omega = 1.55$ eV. We note, however, that the actual magnitude of the asymmetries in this case is larger $O(10^{-2})$ than in the previous cases.

Perhaps the simplest way to examine the necessity or otherwise of the retardation effect for the spin-flip process is to put the light propagation vector $\vec{\kappa}$ identically equal to zero in the transition matrix elements $t^{(n)}_{u\to d}$ and $t^{(n)}_{d\to u}$ [Eqs. (4) and (5)]. Clearly the argument a of the Bessel function $J_n(a)$ defined above in the limit of zero retardation ($\vec{\kappa} = 0$) remains finite and the field-dressed momentum $\vec{q}$ simply reduces to the free momentum $\vec{p}$. Hence the spin-flip amplitudes do not vanish in the limit of zero retardation or if the magnetic component of the incident laser field is neglected. Note also that in the present case there is no spin-orbit interaction in the initial state (ground s state) or in the final state as approximated by the plane-wave Dirac-Volkov continuum state. So one may rightly enquire: What is the mechanism for the finite spin-flip transition probability in intense fields in the absence of retardation and the spin-orbit interaction? We first note that Eqs. (4) and (5) for the spin-flip amplitudes depend on the parameters m_2 and $g(q)$ both of which arise from the "weak" components of the Dirac spinor of the free electron and the ground state of the Dirac H atom, respectively. Hence they certainly go beyond the usual Pauli mechanism of coupling of the external magnetic field to the spin (magnetic moment) of the electron. In fact, further examination of the two equations shows that the nonvanishing coupling occurs through the vector product of the polarization $\vec{\epsilon}$ and the electron momentum $\vec{p}$. This, along with the outer factor $A_0/c \equiv E_0/\omega$ [cf. Eq. (2)], where E_0 is the electric field amplitude of the laser field in the laboratory, shows that the coupling depends on factors of the form $\vec{E}_0 \times \vec{p}/c \approx \vec{B}'$ (a.u.) where $\vec{B}'$ is, in fact, an effective magnetic field seen by the electron in its own frame of reference. It arises from the Lorentz transformation (e.g., [22]) of the electric field E_0 of the laser in the laboratory, into an effective (or "motional") magnetic field in the rest frame of the electron moving with a momentum $\vec{p}$ in the laboratory. Therefore the dominant mechanism that leads to the spin-flip transition in *intense* laser fields is the coupling of the motional magnetic field $\vec{B}'$ with the spin $\vec{\sigma}$ or magnetic moment $= -\frac{1}{4c^2}\vec{\sigma}$ (a.u.) of the electron. This is rather analogous but *not* identical to the Lorentz transformation of the electric field associated with the static *atomic* potential $V(r)$ into an effective magnetic field and the resulting spin-orbit interaction responsible for the well-known Fano effect [13] observed in the perturbative domain of intensity [12].

It is worth noting from the point of view of controlling the relative dominance of the up $\to$ down or down $\to$ up

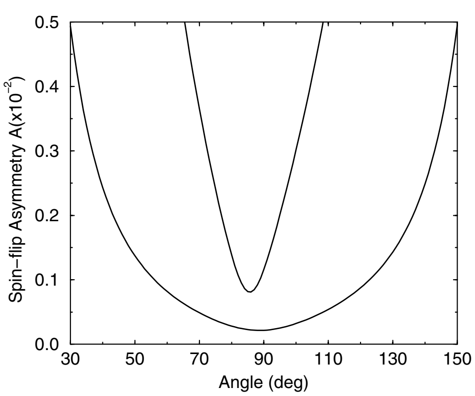

FIG. 2. Intensity-dependent spin-flip asymmetry parameter A [see Eq. (13) for definition] vs electron emission angle; $\omega = 1.55$ eV, $I = 10^{16}$ W/cm^2 (outer curve), and $I = 10^{17}$ W/cm^2 (inner curve).

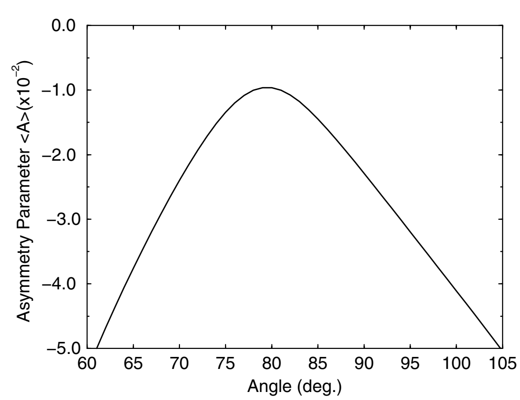

FIG. 3. Ensemble averaged asymmetry parameter $\langle A \rangle$ vs electron emission angle; $\omega = 20$ eV, $I = 10^{20}$ W/cm^2.

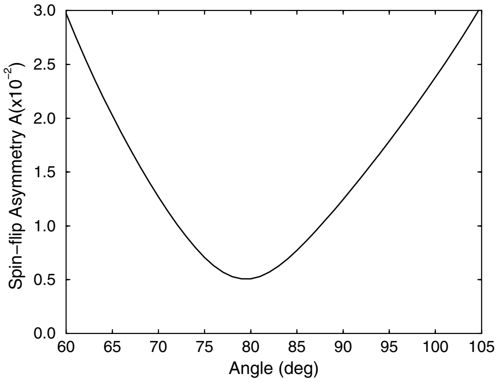

FIG. 4. Spin-flip asymmetry parameter A vs electron emission angle; $\omega = 20$ eV, $I = 10^{20}$ W/cm^2.

spin-flip probability (and hence of the spin-down or spin-up electron currents) that their magnitudes can be reversed from outside, for example, by changing the *helicity* of the incident light. This can be seen by replacing the right circular polarization vector $\vec{\epsilon}(\xi = +\pi/2)$ by the left circular polarization vector $\vec{\epsilon}(\xi = -\pi/2)$ in Eqs. (3)–(6) and observing that the following transformations of the amplitudes hold: $t^{(n)}_{u\to u} \to t^{(n)*}_{d\to d}$, $t^{(n)}_{d\to d} \to t^{(n)*}_{u\to u}$, $t^{(n)}_{u\to d} \to -t^{(n)*}_{d\to u}$, and $t^{(n)}_{d\to u} \to -t^{(n)*}_{u\to d}$. Hence, the spin-flip rates would exchange their magnitudes and the asymmetries A and $\langle A \rangle$ would change their signs on changing the helicity of the photons from the right circular to the left circular polarization.

Before concluding, we may recall that the magnitude of the asymmetry parameters $\langle A \rangle$ and A, in the cases explicitly considered above, are of the orders of 10^{-3} for the near-infrared wavelength and 10^{-2} for the VUV wavelength. These values lie well above the threshold efficiency $\approx 2.4 \times 10^{-4}$ of currently available spin analyzers in the laboratory (e.g., [23,24]).

To summarize, we have analyzed the spin response in ionization of an ensemble of spin-unpolarized Dirac H atoms subjected to intense circularly polarized laser radiations at near-infrared and VUV frequencies. An intensity-dependent asymmetry between the up-spin and down-spin electron currents is found to exist, which survives even when the retardation effect and the spin-orbit interaction responsible for the well-known Fano effect are negligible. The effect is expected to be observable for any atom (e.g., H or alkali atoms) when subjected to intense laser radiation of currently available wavelengths.

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