Spin Signatures in Intense Laser-Ion Interaction

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Spin-orbit coupling of a multiply charged ion is investigated in an intense laser field with a non-negligible magnetic field component. The Lorentz force induces an enhanced angular motion of the bound electron, especially in the vicinity of the nucleus and, consequently, an orbital angular momentum and spin-orbit coupling significantly larger than without the presence of the intense laser field. This gives rise to clear deviations in the electronic wave packet motion and to a strongly increased splitting of resonant lines in the corresponding radiation spectrum.

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High power laser systems have become available in recent years in an intensity regime where the interaction with electrons and atoms has entered the relativistic regime [1]. Free electrons in such intense laser fields were predicted long ago to propagate with velocities near the speed of light c in a plane spanned by the laser polarization and propagation directions [2]. Recently, experimentalists were able to observe the transition from Thompson to Compton [3] and nonlinear Thompson [4] scattering of such fast laser accelerated electrons. In extremely powerful laser fields the generation of electro-positron pairs [5] and neutrons [6] was observed and, most recently, laser induced nuclear fusion for [7] up to the forefront of presently achievable intensities [8]. In the high harmonic radiation spectrum, which has meanwhile entered the soft x-ray regime beyond the water window [9], Doppler shifts of the harmonics were predicted due to the relativistic mass shift [10]. Similar effects in the radiation spectrum were shown to arise from the magnetic field component of an intense laser field, in particular if modified by an additional external magnetic field [11]. The magnetic field of the laser field, even if oscillatory, may be larger than any static magnetic field prepared in a laboratory up to date. Its influence on the spin degree of freedom of the laser driven bound electron was investigated numerically via the Pauli [12] and Dirac equation [13] with clear evidence of spin flipping due to the laser field, however no substantial influences on the electron motion and radiation were put forward. Small quantitative deviations due to the spin were predicted in an analytic approximate treatment of the scattering of a laser driven electron at a nucleus [14].

In this Letter, we investigate numerically the effect of the spin degree of freedom on bound electron dynamics and radiation in an intense laser field. Under the irradiation of the laser pulse, the bound electron obtains a large velocity in its polarization direction and then due to the magnetic field and the Lorentz push a partially angular motion with considerable orbital angular momentum $\mathbf{L}$ with respect to the origin set by the nucleus. We show that the resulting enhanced spin-orbit coupling gives rise to observable effects in the electron dynamics and radiation. In particular, we note a significant splitting of the nonsymmetric bound states due to this additional interaction which leads to well-separated doublets and four-line structure in the radiation spectra. We furthermore note that the relativistic interaction leads to a shift of the spectral line with respect to results arising from the Pauli equation. In general terms we understand those as indications that the influence of the spin and other relativistic effects are both principally observable in experiments and non-negligible in theoretical treatments for laser intensities well below those assumed before.

We are interested in the weakly relativistic regime of optical laser intensities of up to 10^{17} W cm^{-2}, which have been implemented in several laboratories worldwide and which still allow for laser-bound electron dynamics. For those parameters we are justified to employ the Foldy-Wouthuysen expansion [15] of the Dirac equation for weakly relativistic velocities in $1/c$, where $c \simeq 137.036$ is the light velocity in atomic units. The first correction term to the Schrödinger equation gives rise to the additional term in the Pauli equation. The second order terms include the spin orbit coupling of interest in this paper as well as the leading relativistic mass shift term and *Zitterbewegung*. Under the weakly relativistic approximation, we neglect terms of the order of $O(1/c^3)$ and have checked that the leading terms here are negligible for the set of parameters employed in this paper. The main advantage of using this equation in comparison to the full Dirac equation [13] is the possible isolation of the influence of each physical mechanism arising, as in particular spin orbit coupling.

For the circumstances with laser parameters described above, the Hamiltonian of a bound electron in a strong laser field can be written (in atomic units) as

$$\begin{aligned}
\mathbf{H} &= \mathbf{H}_0 + \mathbf{H}_p + \mathbf{H}_{\mathrm{kin}} + \mathbf{H}_D + \mathbf{H}_{\mathrm{so}}\,, \\
\mathbf{H_0} &= [\mathbf{p} + \mathbf{A}(z,t)/c]^2/2 + V(x,z)\,, \\
\mathbf{H}_P &= \sigma \mathbf{B}(z,t)/2c\,, \\
\mathbf{H}_{\mathrm{kin}} &= -\mathbf{p}^4/8c^2, \\
\mathbf{H}_D &= \mathrm{div}\mathbf{E}'(x,z,t)/8c^2, \\
\mathbf{H}_{\mathrm{so}} &= i\sigma\,\mathrm{curl}\mathbf{E}'/8c^2 + \sigma\mathbf{E}' \times \mathbf{p}/4c^2.
\end{aligned} \tag{1}$$

Here $\mathbf{H}_0$ denotes the standard nonrelativistic Hamiltonian in Schrödinger form, where $\mathbf{p}$ is the momentum operator and $\mathbf{A}(z,t)$ is the time t dependent vector potential of the laser field $E(z,t)$, which is linearly polarized along the x axis and propagates in the z direction. For the vector potential we include the magnetic field component and do not apply the dipole approximation, urging us to perform a two-dimensional numerical integration in the x-z plane. We consider atoms in the single active electron approximation which are preionized by of the order of 10 electrons, and thus are easily available today via lasers [16] or with highest accuracy via shooting the atoms through thin foils [17]. Those are well described by the soft-core potential [18] to model the Coulomb field experienced by the active electron of a multiple-charged ion, i.e., $V(x,z) = -k/\sqrt{s + x^2 + z^2}$. The parameters k and s are functions of the effective number of positive charges Z as sensed by the electron. s compensates for the effect of possible inner electrons and reduced distances of the electronic wave packet to the ionic core in two- rather three-dimensional calculations. k is adapted such that we obtain the correct ionization energy for the system of interest with effective charge of the ionic core Z and charge of the ion $Z - 1$. The static field of the ionic core is expressed by the gradient of the potential $-\nabla V(x,z)$, and $\mathbf{E}'(x,z,t)$ stands for the sum of this field plus the laser field $\mathbf{E}(z,t)$. The following term $\mathbf{H}_P$ in Eq. (1) indicates the coupling of the laser magnetic field $\mathbf{B}$ to the electronic spin as described by the Pauli matrix σ. The sum $\mathbf{H}_0 + \mathbf{H}_P$ leads to the Hamiltonian in the well-known Pauli equation. Further in Eq. (1) $\mathbf{H}_{\mathrm{kin}}$ denotes the leading term for the relativistic mass increase, and $\mathbf{H}_D$ is the well-known Darwin term. Finally, the term of most interest here in the Hamiltonian is $\mathbf{H}_{\mathrm{so}}$ and stands for the spin-orbit coupling.

Considering our central potential $V(x,z)$ the first term of $\mathbf{H}_{\mathrm{so}}$ in Eq. (1) disappears because $\nabla \times [-\nabla V(x,z)] = 0$ and the contribution due to the laser field is of the order of $1/c^3$. Thus, the spin-orbit coupling term becomes

$$\mathbf{H}_{\mathrm{so}} = \sigma\mathbf{E}' \times \mathbf{p}/4c^2 = \sigma\mathbf{E} \times \mathbf{p}/4c^2 + f(x,z)\sigma\mathbf{L}\,, \tag{2}$$

with $f(x,z) = -k(s + x^2 + z^2)^{-3/2}/4c^2$ and where $\mathbf{L} = \mathbf{r} \times \mathbf{p} = (0, zp_x - xp_z, 0)$ is the orbital angular momentum, of which only the component along the y direction is nonzero. The origin of spin-orbit coupling can alternatively be viewed also as being due to the interaction between the magnetic moment of the electron and the magnetic field $\mathbf{B}'$ due to the motion of the positively charged core as sensed by the electron in its own rest frame.

From the numerical point of view, we first solve for the eigenstates of the bound electron in the ionic core potential, where our system of interest with $s = 1$ and $k = 93.5$ corresponds to an ion with ground state energy -84.5 a.u. and charge $Z - 1 = 12$, and is thus a good model for the one electron ion Al^{+12} and for Ga^{+12} with a single active $4s$ electron. We then use the split-operator algorithm [19] to investigate the evolution of the system under the irradiation of an intense laser pulse, which consists of a 5-cycle linear turn-on and 100-cycle duration with constant amplitude. The laser parameters involve the wavelength 527 nm of a frequency doubled Nd:glass laser and a peak intensity of 7×10^{16} W/cm^2. This intensity was chosen in the gap of 3×10^{16} to 10^{17} W/cm^2, when the spin splitting started to be visible and ionization began to become significant. Details, including numerical aspects, on highly charged ions in strong laser fields and general spin dynamics will be the subject of future works [20].

In order to isolate the effects due to more intense laser fields and spin-orbit coupling we compare effects arising from describing the dynamics with the complete Hamiltonian $\mathbf{H}$ in Eq. (1), including all second order corrections $\mathbf{H}_{\mathrm{kin}} + \mathbf{H}_{\mathrm{so}} + \mathbf{H}_D$ with, respectively, the situation in which just the first order terms in the Pauli Hamiltonian $\mathbf{H}_0 + \mathbf{H}_P$ or the complete Hamiltonian but $\mathbf{H}_{\mathrm{so}}$ have been taken into account.

We begin to discuss our results by considering the center-of-mass motion of the electronic wave packet along the propagation direction (z axis) in Fig. 1. The solid and dashed lines represent the situations where spin-orbit coupling has not and has been included, respectively. Here the spin-orbit interaction is shown to induce a closer attachment of the electronic wave packet towards the ionic core. From evaluating the partial derivative in propagation

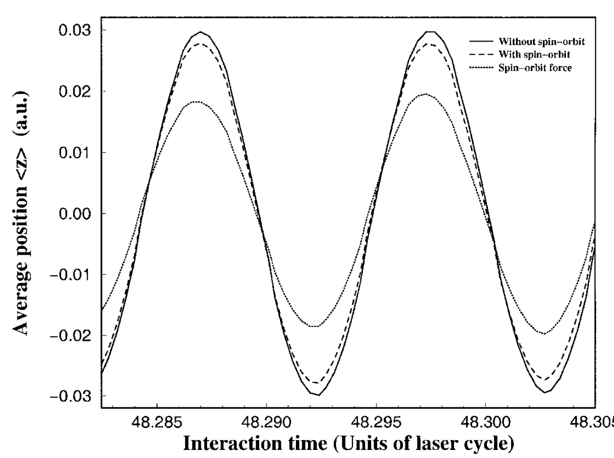

FIG. 1. The center-of-mass position $\langle z \rangle$ of the electronic wave packet as a function of the interaction time via the application of the complete second order equation including spin-orbit coupling (dashed line) and that only without spin-orbit coupling (solid line). The laser parameters involve a wavelength of 527 nm, an intensity of 7×10^{16} W/cm^2, a 5-cycle linear turn-on, and a 100-cycle duration with constant amplitude. The parameters for the ionic core are $s = 1$, $k = 93.5$. We note that the spin-orbit coupling implies an additional force towards the ionic core (proportional to approximately $\langle \sigma_y p_x \rangle$, dotted line).

direction of $\mathbf{H}_{\mathrm{so}}$ in Eq. (2) we note that the leading term of this force is proportional to $\langle \sigma_y p_x \rangle$. This is plotted as a dotted line in Fig. 1 and, remembering the π phase shift of force and corresponding spatial evolution, we associate the additional attractive force with $\langle \sigma_y p_x \rangle$. For the evolution in x direction, p_x needs to be replaced above by the much smaller p_z in the weakly relativistic regime, and is thus far less affected by the spin orbit force.

In Fig. 2 we address the spin polarization itself and have displayed the expectation value of the electronic wave function in "spin-down" configuration as a function of the interaction time. We compare results from the full second order Hamiltonian $\mathbf{H}$ including spin-orbit coupling with those where spin-orbit coupling $\mathbf{H}_{\mathrm{so}}$ has been ignored. Both situations involve an oscillation with twice the laser frequency. With spin-orbit coupling, however, the total oscillation amplitude is higher because of a second oscillation due to the magnetic field of the frame of reference transformed nucleus $\mathbf{B}'$. Finally, the figure shows an effective polarization due to spin-orbit coupling in the turn-on phase, while without spin-orbit coupling the electron periodically returns to the initial polarization in complete spin-up configuration.

The most significant qualitative features appear in the radiation spectrum in terms of strongly laser-enhanced line splitting and shifting. In Fig. 3 we have displayed the radiation spectrum of light emitted perpendicular to the plane spanned by the laser polarization and propagation direction and being polarized in the x direction. The upper row describes the situation governed by the Pauli Hamiltonian, while the lower involves the full second order Hamiltonian $\mathbf{H}$ in Eq. (1). Figures 3(i-a), 3(i-b) and 3(i-c) show the spectral segments corresponding to the resonances of the first excited state $|1e\rangle$ to the ground state $|g\rangle$, the third excited state $|3e\rangle$ to the ground state $|g\rangle$, and the third excited state $|3e\rangle$ to the first excited state $|1e\rangle$. Comparing the upper and lower row we note shifts and splittings of the spectral components into a doublet in (a) and (b) and a four-line structure in (c). We have carried out the same runs comparing the spectra arising from the full Hamiltonian $\mathbf{H}$ in Eq. (1) with the one where just the contribution due to spin-orbit coupling $\mathbf{H}_{\mathrm{so}}$ is missing. We find that the splitting disappears in the latter case; however, the shifting remains. We therefore associate this shift with the relativistic mass shift governed by $\mathbf{H}_{\mathrm{kin}}$ and consider it as relativistic correction to the Stark shift. We found that the Darwin term due to $\mathbf{H}_D$ has no notable

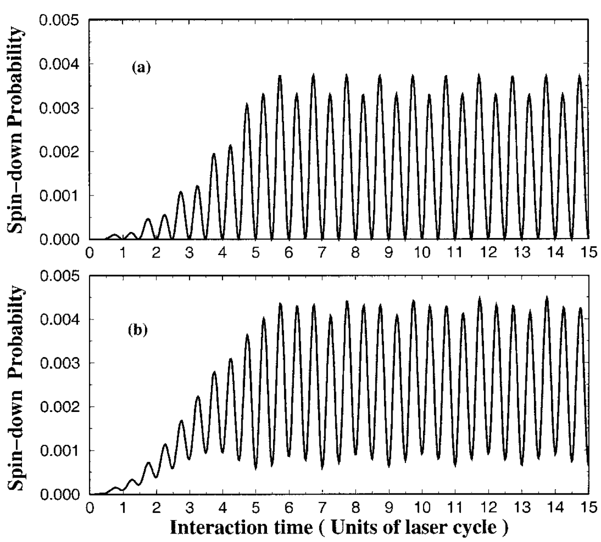

FIG. 2. The dynamics of the spin degree of freedom for the situations without (a) and with (b) spin-orbit interaction as viewed from the population of the wave function with spin-down polarization. The initial electron is spin-up polarized. The intense laser-enhanced spin-orbit coupling leads to an effective spin polarization in the turn-on phase and an additional oscillation due to magnetic field from the nucleus in the rest frame of the electron. The interaction parameters are identical to those employed in Fig. 1.

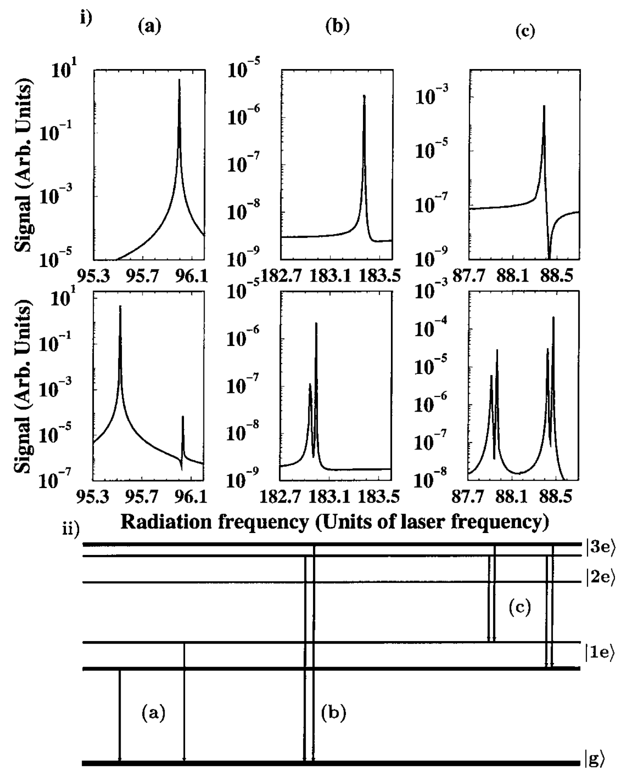

FIG. 3. (i). Radiation spectrum of the laser driven ion close to its lowest resonances. The first row corresponds to the Pauli modeled system, and the second row is for the case where spin-orbit coupling including the relativistic mass shift and Zitterbewegung is taken into account. (a), (b), and (c) are associated, respectively, with transitions from $|1e\rangle$ to $|g\rangle$, $|3e\rangle$ to $|g\rangle$, and $|3e\rangle$ to $|1e\rangle$ [see also (ii)]. The spectral lines split into doublets [(a) and (b)] and four-line structure (c) configurations due to the spin-orbit interaction. A line shift arising from the mass shift H_{kin} is also observable. All parameters are as in Fig. 1. (ii). The schematic diagram of state splitting induced by the intense laser enhanced spin-orbit interaction. We note that nonsymmetric states split as opposed to symmetric states. Transitions (a), (b), and (c) are associated with the corresponding spectral lines in (i).

effects in this situation. We confirmed that the spectral features displayed are generally well separated for ions with different charge states, should a possible experiment not allow for a pure sample of the ion of choice.

We explain the doublets and four-line structure with the splitting of the antisymmetrical excited states $|1e\rangle$ and $|3e\rangle$ due to the additional spin-orbit interaction as depicted in Fig. 3(ii), while the symmetrical states, possibly s states, remain unchanged. The splitting becomes larger with increasing laser intensity or charge of the ionic core. All transitions give rise to spectral features because the common selection rules do not apply in the parameter regime beyond the dipole approximation as investigated here. The bound states in Fig. 3(ii) drawn with thick lines indicate which of the split states is more populated, explaining the relative heights of the spectral lines in Fig. 3(i). Extending the frequency range of Fig. 3(i), we note additional smaller lines displaced by integer multiples of the laser frequency ω_L.

We note that the amount of the splitting is $\frac{\Delta E}{\omega_L} \simeq 0.51$ for the state $|1e\rangle$ and 0.05 for the state $|3e\rangle$ (here, $\omega_L = 0.0866$ a.u. is the applied laser frequency) so that the enhanced spin-orbit splitting should be easily measurable in experiments. Comparing those values with the amount of spin-orbit splitting without the presence of the laser field, we have evaluated numerically via the same techniques $\frac{\Delta E_0}{\omega_L} \simeq 0.046$ for $|1e\rangle$ and 0.005 for the state $|3e\rangle$. Thus, we find that the total enhancement factor of the spin-orbit splitting due to the intense laser field for our set of parameters amounts to approximately $10-12$. We note that those values should increase considerably for more intense laser fields and for higher charged ions. However, we also emphasize that for less charged ions, in particular hydrogen, spin-orbit coupling has still little significance. In terms of applications, the amount of spin-orbit splitting and time dependent spin polarization may also be used to obtain information about the laser intensity and pulse shape or the ion charge.

In conclusion, we have shown that the spin degree of freedom of an electronic wave packet has a significant influence on its own dynamics when more deeply bound electrons of an atomic system begin to be involved in the interaction with a very intense laser field. In the laser propagation direction it induces a force towards the nucleus and in the radiation spectra splittings arise easily an order of magnitude larger than with the presence of the nucleus alone.

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