Polarization control of high-harmonic generation via the spin-orbit interaction

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We observe the generation of high harmonics in the plane perpendicular to the driving laser polarization and show that these are driven by the spin-orbit interaction. Using R-Matrix with time-dependence theory, we demonstrate that for certain initial states either circularly- or linearly-polarized harmonics arise via well-known selection rules between atomic states controlled by the spin-orbit interaction. Finally, we elucidate the connection between the observed harmonics and the phase of the intial state.

Much progress in attosecond physics has been the result of advances in High Harmonic Generation (HHG) techniques [1]. Enhanced understanding and control of HHG has enabled ultrashort light sources [2], and myriad metrological techniques—under the umbrella of ‘high-harmonic spectroscopy’—for the observation and manipulation of ultrafast electron dynamics [3].

The HHG mechanism is well described by the ‘three-step model’: the tunnel ionization, acceleration and recombination of a valence electron driven by a strong laser field resulting in the emission of photons of odd, integer multiples of the driving laser photon energy [4]. The power of HHG as a spectroscopic tool is that the attosecond-timescale dynamics of the target atom are encoded in the spectrum of emitted light, and the rapid evolution of both experimental techniques and underlying theory has resulted in a parade of impressively sensitive and sophisticated measurements [5–9].

With this trend in mind, we may turn our attention beyond the non-relativistic picture of electron dynamics, and seek to address the role of the spin-orbit (SO) interaction in HHG. The SO interaction has two major effects on the dynamics: (i) a splitting of the atomic states and (ii) a coupling of these magnetic sublevels. In principle, high-harmonic spectroscopic techniques should reveal all the electronic processes which occur on the attosecond time-scale but, to date, signatures of processes driven by the SO interaction have proved elusive. The only effect reported in the literature is a small shift in the HHG cut-off energy, caused by the the SO splitting of the valence hole in krypton [10]. More recent studies have investigated the time-dependence of the SO interaction in strong-field processes other than HHG [11, 12]. Given the drive to manipulate electron dynamics on the one hand, and the range of techniques already available in high-harmonic spectroscopy on the other, there is a clear motivation to address SO in HHG.

From a theoretical standpoint, the calculation of harmonic spectra is straightforward: the harmonic spectrum is determined entirely from the time-dependent expectation value of the dipole operator $\mathbf{D}$

$$d(t) = \langle \Psi(t) | \mathbf{D} | \Psi(t) \rangle,$$

where $\Psi(t)$ is the wavefunction. However, determining this quantity is only as straightforward as the complexity of the atomic-structure permits, and to include the SO interaction requires a highly sophisticated theoretical approach. In this work we employ the R-matrix with time-dependence (RMT) [13–19] method to demonstrate that the SO interaction leads to non-zero values of the $x$ and $y$ components of $d(t)$ when the driving laser pulse is polarized in the $z$ direction. This means that the presence of the SO interaction may be indicated by the polarization of the HHG.

In order to simplify the atomic structure considerations, we employ a system with a single, $^1S$ threshold: $C^+$, and we adopt, initially, the $LS$-coupling framework for our discussion. We will show that a $C^+$ ion, initially in some specific superposition of states, will produce high-harmonics of a polarization orthogonal to that of the driving field. Furthermore, we can show unambiguously that these harmonics arise due to the SO interaction.

In Fig. 1, we demonstrate the mechanism for these SO-induced harmonics for an example ground state ($M_L = -1$) $C^+$ ion in a superposition of spin up and spin down. Those states which are coupled by the $y$ component of $\mathbf{D}$ must obey the selection rules $\Delta M_L = \pm 1$ and $\Delta M_S = 0$. However, the $z$-polarized driving field cannot change either the $M_L$ or $M_S$ of the system, and thus the relevant states are not accessible. Thus $d(t)$ only contains a component in the $z$ direction.

The action of the SO-interaction, however, is to transfer population from the $M_L = -1$ spin up states into the $M_L = 0$ spin down states. These latter states are coupled by $\mathbf{D}_y$ to the $M_L = -1$ spin down states as the dipole selection rules are satisfied. As a result, $d_y(t)$ is non-zero, and harmonics are emitted polarized in the $y$ direction.

The generation of $y$-polarized harmonics can also be explained from the $JK$-coupling perspective. This particular initial state is a superposition of the $J = (3/2)^o$ (with contributions from both $M_J = 1/2$ and $M_J = 3/2$)

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and $J = (1/2)^{\circ} (M_{J} = 1/2)$ states. This means that after a single transition (for example from $J = (3/2)^{\circ}$ to $J = (1/2)^{\circ}$) the $\Delta M_{J} = \pm 1$ selection rule for $D_{y}$ is immediately satisfied, and will remain so as the $z$-polarized driving pulse will not change $M_{J}$. Without the SO-interaction, the radial aspect the wavefunction corresponding to each $M_{J}$ value will propagate in such a way that $D_{y}$ in the positive and negative directions will cancel. As such, unless the SO-interaction induces differences in the radial aspect of the wavefunction, $d_{y}(t)$ will still be zero, and harmonic emission will be confined to $z$-polarization.

To test this hypothesis, we use RMT to solve the Schrödinger equation incorporating the SO interaction

$$\frac{d}{dt} \Psi(X, t) = -i \left[ \hat{H}_{A} + \hat{H}_{SO} + \mathbf{E}(t) \cdot \mathbf{D} \right] \Psi(X, t),$$

(2)

where $\hat{H}_{A}$ is the time-independent, non-relativistic atomic Hamiltonian, $\hat{H}_{SO}$ is the SO term from the Breit-Pauli equation, and $\mathbf{E}(t)$ is the time-dependent electric field strength of the driving laser pulse, polarized in the $z$ direction. From our chosen initial wavefunction, we then obtain the wavefunction at subsequent times solving Eq. (2) iteratively. At each time step we calculate the expectation value of $\mathbf{D}$ using Eq. (1). We then obtain the harmonic spectrum of the radiation emitted by the atom polarized in the $x$, $y$, and $z$ directions from the Fourier transforms of the $x$, $y$, and $z$ components of $\mathbf{d}(t)$ respectively.

For these calculations we consider a $C^{+}$ ion in two initial superpositions, described in Tabs. I and II. The ion description is built up by coupling an electron to bound orbitals for a $C^{2+}$ target state [20]. These continuum orbitals are constructed from a basis of 50 B-Splines of 9th order. For all results, the laser pulse is an 8 cycle 800nm pulse (3 cycles ramp on, 2 cycles at peak intensity, 3 cycles ramp off) of peak intensity $10^{14}$ W/cm$^{2}$. The field is linearly polarized in the $z$ direction and propagates in the $x$ direction.

Figure 2 shows the HHG spectrum calculated with and without the SO-interaction. The initial state is as described in Tab. I, a superposition of $M_{L} = -1$ spin up and $M_{L} = 1$ spin down- where we have adopted an initial phase difference of $\theta = 0$. As expected, omitting the SO-interaction yields harmonics polarized only in the $z$-direction, whereas including the SO-interaction gives harmonics with both $y$ and $z$ polarization.

Further calculations reveal that the polarization of the harmonic spectrum in the $x/y$ plane depends on the phase-difference, $\theta$, in the initial-state superposition. Specifically, we find that the $x$ and $y$ components of the

| $L$ | $M_{L}$ | $M_{S}$ | Weight |
|-----|---------|---------|--------|
| 1   | $-1$    | $\frac{1}{2}$ | $\sqrt{\frac{2}{3}}$ |
| 1   | $1$     | $-\frac{1}{2}$ | $\sqrt{\frac{1}{3}}$ |
| 2   | $\frac{1}{2}$ | $\frac{1}{2}$ | $\sqrt{\frac{1}{6}}$ |

TABLE I. The initial-state composition used for figures 2 and 3 in both LS (left) and JK (right) coupling. $\theta$ is the phase difference in the initial superposition.
harmonic spectrum combine to produce a linearly polarized harmonic spectrum of angle $\theta$ off the $y$ axis. Figure 3 shows the results from an initial state with a phase difference of $\theta = \pi/10$ within the superposition. Both $x$ and $y$ polarized harmonics are produced, although $y$ polarized harmonics are still of higher intensity. Moreover, the phases of the $x$ and $y$ polarized harmonics are identical throughout the spectrum, indicating that the harmonics are again linearly polarized. The angle from the $y$-axis is calculated from the intensity of the individual $x$ and $y$ polarized harmonics, is found to be equal to the phase difference in the initial superposition, $\theta$. This relationship between $\theta$ and the angle of polarization in the $x$-$y$ plane was found to hold true across angles in the range $\theta = 0 \ldots \pi/2$.

We now consider the superposition described in table II, where the $C^+$ ion is in the $M_L = -1$ state with a superposition of spin up and spin down. This means that after the effect of the SO interaction there will be a $y$-polarized dipole between the $M_L = -2$ and $M_L = -1$ spin up states, and another between the $M_L = -1$ and $M_L = 0$ spin down states. Again, we add an initial phase factor $e^{i\theta}$ into the spin-down component.

Figure 4 shows the $x$, $y$ and $z$ components of the HHG spectrum. As before, both $y$ and $z$ polarized harmonics are produced, but unlike the previous initial superposition, here $x$-polarized harmonics are also present.

Figure 5 shows that the $x$ and $y$ harmonic spectra have an identical amplitude throughout most of the energy range. The phase difference between the $x$ and $y$ polarized harmonics persists at a half-integer multiple of $\pi$, indicating circularly polarized harmonics (as all harmonics are of the same intensity). We note that there is no clear pattern in the phase relationship in the $x/z$ or $y/z$ planes. Finally, we find that for initial states of $\theta \neq 0$, there is no significant change in the character of the harmonic spectra.

There has been much interest recently in extending HHG to the generation of elliptical and circular harmonics. This is typically attempted using two-color counterrotating circular pulses [21], or pulses of varying linear polarization [22]. Here it is an atomic mechanism—the SO interaction—rather than the character of the
laser field, that provides a means of generating multidimensional harmonics. Although we have restricted our results to \( \text{C}^+ \), these results should persist in general. In those systems with a doublet ground state consisting of a single \( p \)-hole, such as noble gas ions, we would expect \( y \)-polarized HHG via the same mechanism described in Fig. 1.

In this work we have considered only the single atom response to the driving field, and as such we have included a description of the resulting \( x \) polarized spectra. However, as the driving field propagation is in the \( x \) direction, observing the \( x \) polarized harmonic spectra will be challenging. Nonetheless, these results show interesting observations are possible using only the \( z \) - and \( y \)-polarized harmonic spectra. For example, we could use the intensity of the emitted \( y \)-polarized harmonics as a measurement of the phase difference between two states in a superposition. This phase difference will most often have arisen as a result of the methods used to create the initial superposition, however when the phase difference is unknown this could be a useful experimental tool.

The question remains of how to produce such initial states. A linearly polarized pulse will preferentially eject an electron of \( m_\ell = 0 \). It might be suggested therefore, that a short pulse might be used to eject such an electron from a noble gas atom to form a target of a superposition of \( M_L = 0 \) spin up and spin down. We have suggested a different approach in [23] where the \( m_\ell \) of the ejected electron can be controlled by dipole selection rules when exciting an individual electron to the only energetically accessible s state. This method has the advantage of having been demonstrated theoretically using ultrashort laser pulses, providing greater of control over the purity of the initial superposition.

In conclusion, we have demonstrated that given an appropriate initial state, the presence of the spin-orbit interaction will cause harmonic spectra to be generated of a polarization orthogonal to that of the driving field. The specific superposition chosen may lead to linear or circular polarization in the \( x/y \) plane. As such, these results provide a clear route to observe spin-orbit effects through HHG processes.

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