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Spin-orbit dynamics in atomic krypton observed through decay of autoionising states

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Synopsis We study the spin-orbit interaction within excited states of krypton by extending R-Matrix with time-dependence theory to include spin-orbit effects. We study an attosecond pump-probe scheme where we produce an autoionising state with the probe pulse. We observe dynamics through the subsequent decay of the autoionising state, from which we find a time-dependent signal when spin-orbit interactions are included. We examine the effect of the relative polarisation of the pulses, enabling an interpretation of the dynamics in terms of the quantum number m_{ℓ} of the core of the excited krypton atom.

In recent years, it has become possible to study spin-orbit dynamics experimentally from a time-dependent perspective (e.g. [1, 2]). To meet the need for corresponding theoretical methods, we have extended R-Matrix with Timedependence (RMT) theory to include spin-orbit interaction. Over the past decade RMT has demonstrated its use in the ab initio study of ultrafast electron dynamics in atoms, and has recently been extended to describe dynamics in arbitrarily polarised light fields [3].

We extend RMT by enabling an interface with the RMatrixI package [4] such that spinorbit effects are present in the resulting electron dynamics. To demonstrate these effects, we model a pump-probe setup on atomic krypton. The pump pulse (6 cycles, 10 eV) excites the $4s^24p^55s$ ¹P state, after which the spin-orbit interaction will change the atom from the ${}^{1}P$ state to the ${}^{3}P$ state and back again. After a time delay, the probe pulse (6 cycles, 15 eV) will further excite the atom to the $4s4p^{6}5s$ autoionising state. This state will subsequently decay to $4s^24p^5k\ell$, which we observe through detection of the ejected electron.

Figure 1 shows the autoionisation signal from this model as a function of the time-delay between the two pulses. When the spin-orbit interaction is included in the model, we observe oscillations in the autoionisation signal with respect to the time delay between the two pulses. When the pulses are cross-polarised, we see the same oscillation, but out of phase.



Figure 1. Signal from autoionisation of atomic krypton caused by a 10 eV pump pulse and a 15 eV probe pulse separated by a time-delay.

We will present an interpretation of this phase difference in terms of the spin-orbit induced dynamics of an electron between m_{ℓ} states, and the differing selection rules of the crosspolarised and parallel polarised pulses. We will also present a discussion of the interference effects present in Fig. 1 for both parallel-polarised cases, but absent in the cross-polarised case. Again, this difference can be attributed to the dipole-selection rules for the cross- and parallelpolarised pulses. This result may have actionable implications for improving the signal-to-noise ratio in experiments of this type.

References

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