On the search for quantum interference in multiphoton ionization of atoms and molecules

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Introduction

Quantum interference (QI) in ionization can be thought of as a temporal analogue of a Young’s double slit experiment, with a pair of pulses separated by a delay \( \tau \) representing the two slits. We refer to this pair of pulses as a twin-peaked pulse (TPP). Each peak of the TPP produces an excited electron wave packet. Interference of these wave packets causes a periodic oscillation in the energy structure of the photoelectrons. Optical interference (OI) due to the overlap of the peaks’ electric fields will also play a role. In Ref. [13], QI is observed in a single-photon ionization, but the oscillation periods of OI and QI in this experiment are equal.

Quantum interference in bound state transitions

The advantage of multiphoton excitation, studied in Ref. [12], over single-photon transitions is that QI oscillations with a period \( \tau \) is the optical period for an \( n \)-photon transition. This allows for a differentiation between QI and OI. In the case of Ref. [12], the transition was to a pair of bound states, in which case the total electron yield (a population in the upper bound state) does vary as \( \tau \) changes. The authors conclude that there have been no experimental reports that have studied QI in multiphoton ionization with TPPs. In such an experiment, an energy-resolved electron measurement is necessary.

Experimental Setup

The TPPs are made with a Michelson interferometer. One arm is on a translation stage so that \( \tau \) can be directly changed.

Our velocity map imager [5] is used to record electron distributions on the phosphor screen with a 800-1000 CCD camera. Typical exposure time is 10 s.

Results - \( \tau \simeq 250 \) fs

- 1800 images, 10 second exposure for each image.
- Small amount of optical interference, total TPP energy was nearly constant at 15.5 \( \mu \)J.
- The TPP spectrum was recorded for each image.
- By fitting the spectrum in \( \Delta \phi \) relative phase \( \phi \) could be recovered (shift of interference fringes).
- Accuracy in \( \Delta \phi \) was 0.5 radians.
- Images were binned according to their recovered phase and summed together to get composite images.

Results - \( \tau \simeq 0 \) fs

- 3000 images, 10 second exposure for each image.
- Spectrum recorded for each image.
- At \( \tau = 0 \) fs, \( \Delta \phi \) cannot be recovered from shift in interference fringes because there is only one single fringe.
- Instead, bin images based on the measured TPP energy, which will vary due to OI.
- TPP energy is proportional to \( \int \sin \Delta \phi (\Omega) d\Omega \).
- \( \Delta \phi \) is not a single-valued function of the TPP energy!

Because the intensity and relative phase of a single-electron wave packet are identical to the intensity and relative phase of the threshold electron (integrated aperture).

References


Acknowledgements

This work was supported by the National Science Foundation under grant number PHY-0550852.

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