

Theoretical Studies of Interactions of Atoms, Molecules and Surfaces

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Program Scope:

We investigate the interaction of intense lasers and attosecond light pulses with atoms and molecules, including high-order harmonic generation, energy and momentum spectra of the electrons or ions, from the breakup of atoms or molecules, with the goal of interpreting and/or guiding experimental observations. References to our published papers or preprints (the full list is given at the end of this report) are given in bold letters in the text.

1. Alignment dependence of high-order harmonic generation (HHG) and two-center interference

Recent progress

In the last two years, a number of experimental groups have made measurements of high-order harmonic generation (HHG) from aligned molecules. The procedure is rather straightforward: A weak laser pulse is first used to partially align molecules, and another probe laser is later used to generate HHG at different time delays, especially at the short time intervals when the molecules undergo "rotational revival", i.e., when their angular distributions undergo rapid changes. The polarization directions of the two pulses can be either parallel or varied.

There is no reliable theoretical calculations of HHG from aligned molecules. In general, one expects that the HHG from molecules would depend on the symmetry of the molecular orbital of the outermost electron, or the highest occupied molecular orbital (HOMO). To test to what extent the recent experiments can be explained with the simple Lewenstein model, we generalized this model to molecular systems where the molecular wavefunctions were calculated by using the GAMESS code. Initially we have tested our calculations for N₂ and O₂ targets (see **A9**, **A10**) and we were able to show that the experimental results are well explained by our calculations. The calculations and experiments both show that the ionization rates and the HHG yields for N₂ peak in the forward direction, while for O₂ they tend to peak at large angles near 45°. To compare with experimental data, we also calculated the angular distributions of the molecules at different time delays, using laser parameters and gas temperature suggested by the experiments.

The situation is different for CO₂. The first experiment on CO₂ by Kanai et al [Nature, **435**, 470, 2005] observed that the ionization rates and the HHG yields are inverted in that when one is maximum the other is minimum, at least for some harmonics. Since the first step of HHG is the tunneling ionization, this inversion was interpreted as evidence of two-center interference in the recombination process. The latter was proposed by Lein et al [Phys. Rev. A **88**, 183903, 2002] earlier. They argued that the wave packet from the returning electron reaches the atomic centers at different time or different phase such that under proper condition, the harmonics from the two atomic centers can interfere destructively and a minimum in the HHG can be observed if the standard two-center interference condition is satisfied. Note that in this interference model, the position of the HHG minimum does not depend on the laser intensity. Similar experiment with somewhat different laser parameters was carried out by Vozzi *et al* [Phys. Rev. Lett. **95**, 153902, 2005]. Inversion was observed but it is at a different HHG order. In a private communication, the HHG from CO₂ is also being studied in Colorado. Inversion has been observed also but again at

different HHG orders. Thus the situation is quite confusing. What is the origin of the inversion in CO₂? Can the inversion be interpreted in terms of two-center interference?

We have made a theoretical investigation of HHG from CO₂ using the Lewenstein model, see **A4**. In the calculation, we accounted for the depletion effect -- i.e., when a molecule is ionized it does not contribute to the generation of HHG. We found inversion in our calculation, but the position of the HHG minimum depends on the laser intensity, and the inversion has nothing to do with the two-center interference. In other words, we found an alternative interpretation of the inversion of HHG and the result is intensity dependent.

Future plans:

All of our calculations were carried out in the length gauge for the dipole matrix element. Because of the gauge dependence of the calculated HHG and the approximate nature of the Lewenstein model, we cannot conclude precisely whether the inversion is indeed due to two-center interference or not. This project will be further pursued in the coming grant period.

2. Attosecond pulses probing atomic and molecular dynamics

Recent Progress:

We have carried out numerical calculations to illustrate that attosecond pulses can be used to probe the time dependence of the motion of atoms in a small molecule, as well as the correlated motion of two excited electrons. We assume that a vibrational wave packet of D₂⁺ is created at time t=0. The subsequent motion of this wave packet can be probed accurately by using an attosecond xuv pulse of duration of about 300 as to ionize D₂⁺. From the D⁺ energy one can recover the wave packet, revealing detailed spreading and interference of the vibrational wave packet. Analysis of the time evolution of such a wave packet would allow experimentalists to extract the amplitude and phase of each vibrational state in the wave packet. (**A3**). This is an extension of the experiment of Alnaser et al [Phys. Rev. A72, 030702(R), 2005] where an 8 fs laser pump is used to generate the wave packet and its time evolution is probed by another 8 fs pulse.

We have also investigated the time evolution of a wave packet made of a coherent sum of doubly excited states. The wave packet is then probed by using an attosecond pulse to doubly ionize the atoms. From the momentum distributions of the two ionized electrons partial information about the initial wave packet can be probed. A preliminary result of such a study was published in **A1**.

Future plans:

In the coming year we will make further calculations to probe electron-electron dynamics. We will first study the rovibrational motion of two excited electrons in the time domain. For the probe, an xuv attosecond pulse will be used to double ionize the atom. We will use products of Coulomb wavefunctions to represent the two ionized electrons. We will begin to develop a time-dependent two-electron code in hyperspherical coordinates which would allow us to perform calculations for pump-probe experiments with attosecond pulses which can be compared to actual experiments in the future. The code would also allow us to study nonlinear processes using xuv or soft-x-ray light pulses from high-order harmonic generation, as well as from x-ray free electron lasers.

3. Electron energy and momentum spectra of atoms and molecules induced by intense short laser pulses

Recent progress:

Motivated by the experimental observations of the electron momentum spectra carried out at Kansas State University and elsewhere, we initiated a new theoretical study along this same line last year. While ionization of atoms by intense lasers in the multiphoton ionization regime is considered to be well understood, the situation is different in the tunneling ionization region. In the latter, the electron distribution was expected to be rather smooth with a strong peak at the zero momentum. Experiments from the Frankfurt group, which were later confirmed by the experiments from Kansas State University, showed that there are still numerous structures in the electron energy and momentum distributions, even for laser intensities well into the tunneling ionization region.

We have calculated the electron momentum spectra from Ar atoms by a short intense laser pulse by directly solving the time-dependent Schrodinger equation (TDSE) and by using the strong field approximation (SFA). We found that the two theories give rather similar electron spectra, and the spectra show the familiar multiphoton peaks even in the tunneling ionization region. For the short pulses, there also exist many substructures which have been attributed to the rapidly changing ponderomotive potential in the pulse. In the two-dimensional momentum spectra, the major features from the two theories are the same, but for low energy electrons, we noticed substantial differences. The positions of the ATI peaks and the substructures are sensitive to the laser intensities but the structures for the low energy electrons are rather insensitive to it.

Future plans:

Up to now we have not tried to compare the calculated ATI spectra with experimental measurements. We intend to do so in the coming year. For the high laser intensities used in the experiments where the laser is highly focused, the intensity within the gas volume is not constant. In order to compare with experiments, electron momentum spectra have to be calculated over a large range of laser intensities with small steps and the total yield is then calculated. Major contribution to the electron spectra in this case does not come from the peak intensity, but rather from the lower intensities. Since depletion could be quite severe for high laser intensities, its effect has to be included as well. After these factors are considered we will be able to compare with experimental measurements. In the future we will also examine the ATI electrons from aligned molecules.

4. Other activities

In the last year we carried out a few other projects which do not fall under the three categories above but are within the general scope of this program. Among them, **A2** is a review article, **A6** and **B2** are invited papers from two conferences. The rest of them are one-time projects that may be pursued further only if needed.

Publications

A. Published papers

A1. C. D. Lin, X. M. Tong and Toru Morishita, "Direct experimental visualization of atomic and electronic dynamics with attosecond pulses", J. Phys. **B39**, S419 (2006)

A2. C. D. Lin and X. M. Tong, "Probing orbital symmetries and ionization dynamics of simple molecules with femtosecond laser pulses," in ADVANCES IN MULTI-PHOTON PROCESSES AND SPECTROSCOPY, VOL 17, Ed. S. H. Lin, A. A. Villaeys and Y. Fujimura, World Scientific, 2006.

A3. X. M. Tong and C. D. Lin, "Attosecond xuv pulses for complete mapping of the time-dependent wave packet of D_2^{++} ", Phys. Rev. **A73**, 042716 (2006).

- A4. A. T. Le, X. M. Tong and C. D. Lin, "Evidence of two-center interference in high-order harmonic generation from CO₂", Phys. Rev. A73, 041402(R), (2006).
- A5. M. Wickenhauser, X. M. Tong and C. D. Lin, "Laser induced substructures in above-threshold-ionization spectra from intense few-cycle laser pulses", Phys. Rev. A73, 011401(R) (2006).
- A6. C. D. Lin, X. M. Tong and Z. X. Zhao, "Effects of orbital symmetries on the ionization rates of aligned molecules by short intense laser pulses", J. of Mod. Optics, 53, 21 (2006).
- A7. A. T. Le, Toru Morishita, X. M. Tong and C. D. Lin, "Signature of chaos in high-lying doubly excited states of the helium atom," Phys. Rev. A72, 032511 (2005)
- A8. A. S. Alnaser, B. Ulrich, X. M. Tong, I. V. Litvinyuk, C. M. Maharjan, P. Ranitovic, T. Osipov, R. Ali, S. Ghimire, Z. Chang, C. D. Lin and C. L. Cocke, "Simultaneous real-time tracking of wave packets evolving on two different potential curves in H₂⁺ and D₂⁺" Phys. Rev. A72, 030702(R) (2005).
- A9. X. X. Zhou, X. M. Tong, Z. X. Zhao and C. D. Lin, "Alignment dependence of high-order harmonic generation from N₂ and O₂ molecules in intense laser fields", Phys. Rev. A72, 033412(R) (2005).
- A10. X. M. Tong and C. D. Lin, "Empirical formula for static field ionization rates of atoms and molecules by lasers in the barrier-suppression regime", J. Phys. B38, 2593 (2005).
- A11. Z. X. Zhao and C. D. Lin, "Theory of laser-assisted autoionization by attosecond light pulses", Phys. Rev. A71, 060702(R) (2005).
- A12. X. X. Zhou, X. M. Tong, Z. X. Zhao and C. D. Lin, "Role of molecular orbital symmetry on the alignment dependence of high-order harmonic generation with molecules", Phys. Rev. A71, 061801(R) (2005).
- A13. Z. X. Zhao, Z. H. Chang, X. M. Tong and C. D. Lin, "Circularly-polarized laser-assisted photoionization spectra of argon for attosecond pulse measurements", Optics Express 13, 1968 (2005).
- A14. X. M. Tong and C. D. Lin, "Double photoexcitation of He atoms by attosecond XUV pulses in the presence of intense few-cycle infrared lasers", Phys. Rev. A71, 033406 (2005).

B. Papers accepted for publications

- B1. X. M. Tong, Z. X. Zhao and C. D. Lin, Comment on "Correlation quantum dynamics between an electron and D₂⁺ molecules with attosecond resolution", Phys. Rev. Lett.
- B2. C. D. Lin and X. M. Tong, "Dependence of tunneling ionization and harmonic generation on the structure of molecules by short intense laser pulses", J. photochemistry and photobiology.
- B3. M. Wickenhauser, X. M. Tong, D. G. Arbo, J. Burgdorfer and C. D. Lin, "Signature of multiphoton and tunneling ionization in the electron momentum distributions of atoms by intense few-cycle laser pulses", Phys. Rev. A.