Quantum interference of high-order harmonics from mixed gases

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Abstract.

We present a theoretical study about the interference of the harmonics generated by a mixture of two gases, He-Ne. Our model is based on the electron quantum paths, a discrete number of electron trajectories, and continuum-bound transitions. A laser with intensity around $10^{14} W/cm2$ that interacts with a mixture of gases, He-Ne, produces an interference that is destructive at the low-order harmonics and oscillates between constructive and destructive near to cutoff. This destructive interference at high-order harmonics may be used to explore other transitions, which are currently hidden. At low-order harmonic frequencies, our numerical results are in very good agreement with experimental data. At higher-order harmonics, where there are no experimental data, comparison is with a Schrodinger solver.
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1.- Theoretical model

- Based on Lewenstein model[1]. the electron movement:
  $d(t_r) = i\int_{t_0}^{t_r} dt_i \int dp \langle \Psi_0|r|v(p,t_r)\rangle\langle v(p,t_i)|E(t_i)r|\Psi_0\rangle e^{-iS(p,t_i,t_r)}$
- Dipole transition : $< v|r|\Psi_0 >$, depends on the ionization energy Ip
- Quantum paths are a small set of electron trajectories that are enough to describe the full physical interaction.
- The quantum paths are specified by the triada (t_r, t_i, p_s) obtained as solution of three equations:
  - $\nabla_p S(p_s, t_i, t_r) = p_s(t_r - t_i) - \int_{t_i}^{t_r} A(t)dt = 0$
  - $\frac{\partial\Theta(p_s,t_i,t_r)}{\partial t_i} = \frac{[p_s - A(t_i)]^2}{2} + I_p = 0$
  - $\frac{\partial\Theta(p_s,t_i,t_r)}{\partial t_r} = \frac{[p_s - A(t_r)]^2}{2} + I_p - \omega = 0$
- The solutions (t_r, t_i, p_s) permit transform the integral into a sumatory which fourier transform is: $d(\omega) = \sum_q |x_q(\omega)| exp[i\Phi_q(\omega)]$

2.- Quantum paths

- There are two electron trajectories per laser subcycle: short and long, according to the recombination and ionization time t_i, t_r .
- The short and long paths converge into only one trajectory near the cut-off.
- The long path always has higher intensity than the short one, this difference is most important at low frequency. Fig B.
- The electron can recombine in the first visit to parent ion, or on the following, although with lower intensity. Fig B.

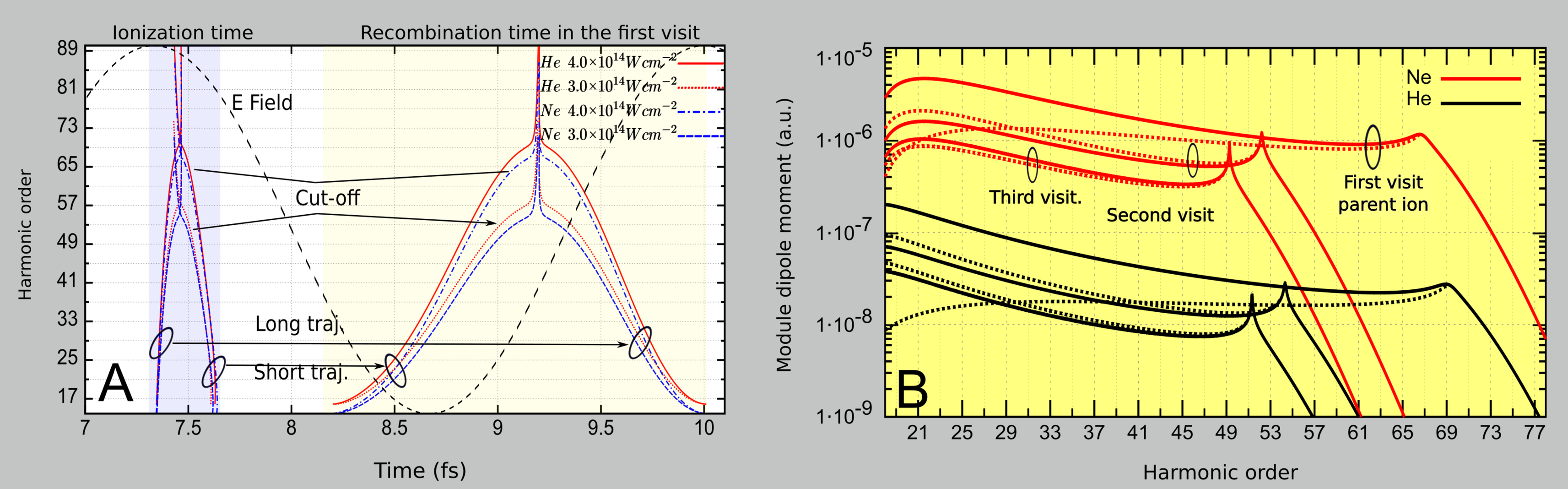

Figure A: This figure shows the value of the harmonic produced when the electron is set free at t_i and when it is recombined at t_r. The t_i and t_r, from long and short paths, produce the same frequency. The figure contains the times from two gases He and Ne for two intensities Figure B: Dipole modulus, in atomic units, when the electron recombines in same cycle i.e., the first parent visit, in the second parent visit, and when the electron recombines in the third parent visit.

3.- Mixture of gases

- When a laser, with enough intensity $\sim 10^{14} W/cm^2$, interacts with a gas, produces harmonics.
- The phase of each harmonic depends on a combination of the ionization energy, kinetic electron energy and atomic structure
- When there are two gases mixed, every harmonic is the result of an interference process :
  $|d_{He}(\omega)+d_{Ne}(\omega)|^2 = |d_{He}(\omega)|^2+|d_{Ne}(\omega)|^2+2\Re e[d_{He}(w)d_{Ne}(w)]$
- Intensity depends on the density ρ of each gas:
  $I(\omega) = \rho_{He}^2|d_{He}(\omega)|^2|(1+\frac{r_{Ne}}{1-r_{Ne}}|\frac{d_{Ne}(\omega)}{d_{He}(\omega)}|e^{i\Delta\Phi(\omega)})|^2$
  $\Delta\Phi(\omega) \equiv \Phi_{He}(\omega) - \Phi_{Ne}(\omega)$
  $r_i = \rho_i/\rho_{total}$ with $\rho_{total} = \rho_{He} + \rho_{Ne}$.
- The adjustment of the ratio between gases, allow us to get some spectrum equalization over some of the harmonics.

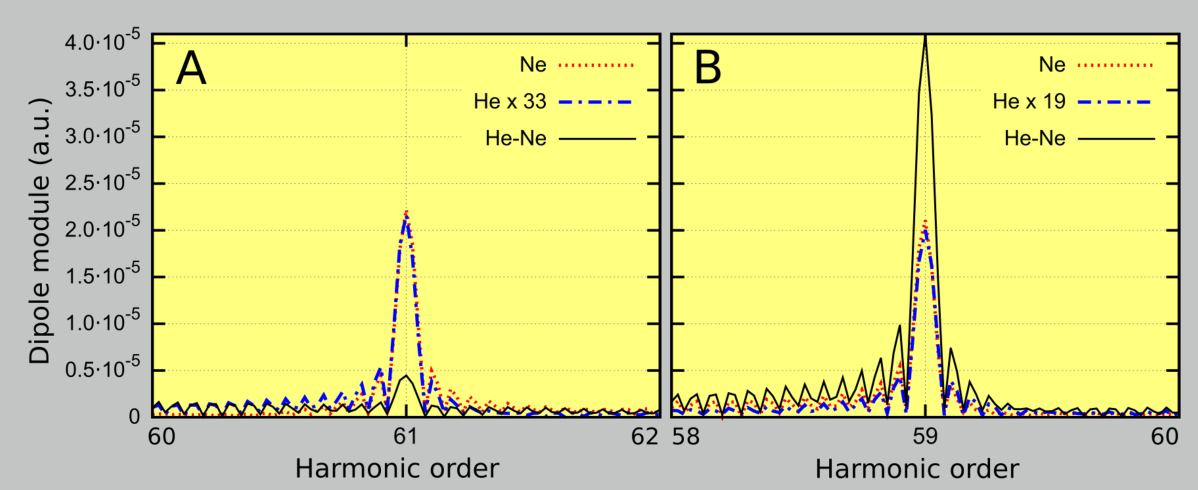

Figure A: The solid black line is the spectrum obtained with a mix of gases in which the number of He atoms is 33 times higher than the number of Ne atoms for the intensity $4.0 \times 10^{14} W/cm2$. The 61st harmonic appears practically annihilated
Figure B: Constructive interference for the 59th harmonic

4.- He-Ne Comparison with experimental data.

- The model based on quantum paths has a good agreement at low-order harmonics where the long paths are predominant.
- At high-order harmonics, where there is not experimenta data, calculations are compared with an Schrodinger solver Qprop[3].
- Only the quantum paths from the first visit to parent ion contribute to high-order harmonics.
- The model has not a good prediction in the middle range.

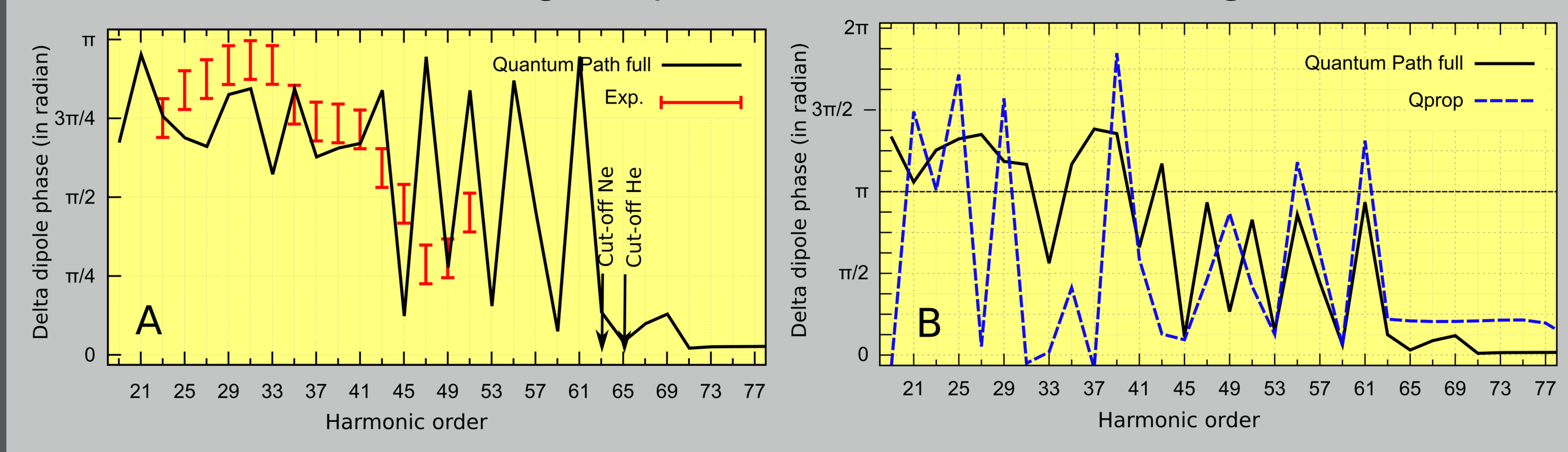

Figure A: Relative phase $\Delta\phi = \Delta\phi_{He} - \Delta\phi_{Ne}$ with a laser intensity of I $= 4.0 \times 10^{14} W/cm2$ and $\lambda = 800nm$. Comparison result from Quantum paths and experimental data from Kanai et al. [2].
Figure B: For the higher-order harmonics where there are no experimental data, we use a reference to a Schrodinger solver (Qprop) [3].

5.- Quantum path. Simplification

- The short and long paths from the first visit to parent produce an oscillation which increases its the amplitud upto the cutoff.
- The interference bewteen long and short paths explains the strong oscilations from 45th harmonics onwards. Fig A.
- The long trajectory indicates the evolution of the interference. Fig B.

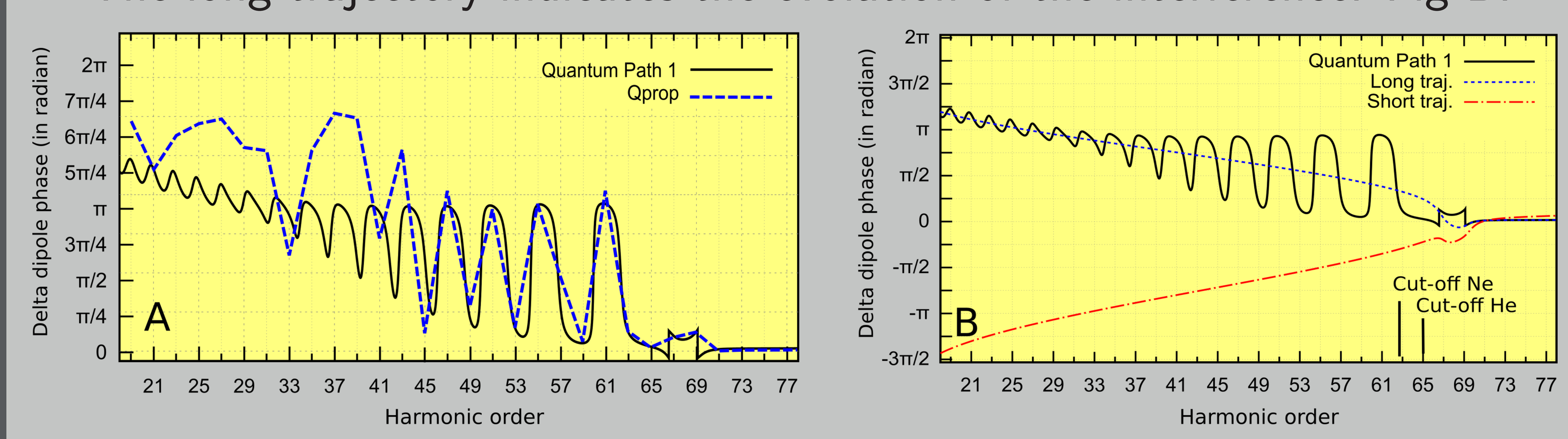

Figure A: Comparison between calculation with a Schrodinger solver and the quantum paths.
Figure B: Black line is the interference between long and short paths from every atom He and Ne.

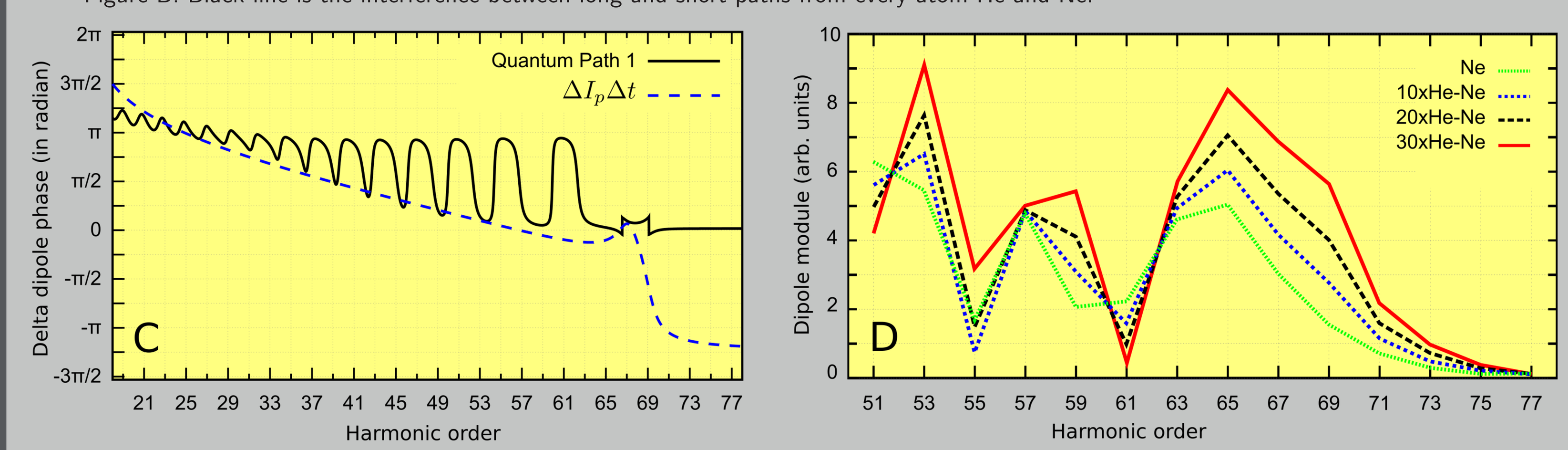

Figure C: Comparison between $\Delta I_p \Delta t$ and calculations from Quantum paths in the first visit.
Figure D: Interference due to the mixture He-Ne for several density ratios (He/Ne): 0, 10, 20, and 30. Some harmonics have constructive interference compared with the case of only neon, and others have destructive interference.

7.-Conclusions

- The model based on quantum paths allows one to explain the constructive and destructive interference between harmonics generated with a mixture of two noble gases like He-Ne.
- The ratio between the densities of each gas can be used for tuning the intensity and the phase of harmonics.
- Finally, the selection of the dipole matrix for Ne which takes into account the parity of the electron, with mathematical expressions different from those commonly used, was crucial in reproducing the experimental and numerical results from a Schrodinger solver.

8.- References

1 Kanai et al. PhysRevLett.98.153904. (2007).
2 Lewenstein. PhysRevA.49.2117. (1994).
3 D. Bauer and P. Koval, Comput. Phys. Commun. 174, 396 (2006).
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