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High-order harmonic generation from confined Rydberg atoms

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Synopsis We report results from our simulations of High Harmonic Generation (HHG) from a confined atom in a Rydberg state. We find that for the $n = 2$ excited state of H the cut-off of the harmonic spectrum is substantially extended compared to that for a free atom at the expense of the harmonic yield. This effect is dependent on the radius of the confining shell for a given n. We also observe that the confined spectrum exhibits cusps similar to those seen in the HHG spectra from ground state atoms in the presence of Cooper minima.

Dynamically rich nature of the high-order harmonic generation (HHG) process lends itself to a variety of ways to extend the harmonic cut-off frequency. In this work, we investigate HHG from a Rydberg atom confined in an attractive shell as HHG from Rydberg states have already shown interesting physics and potential for extending the harmonic cut-off frequency [1, 2].

We solve the time-dependent Schrödinger equation within a one-dimensional s-wave model, where we use a $800n^3$ nm laser pulse with 4 cycles at FWHM. The laser intensity is $(3.5/n^8) \times 10^{14}$ W/cm^2. The cage is modeled as a -8.22 eV deep spherical shell potential with 5.8 $\mathring{A}$ inner radius and 7.69 $\mathring{A}$ outer radius. These parameters correspond to a C_{60} cage [3].

We compare the spectra for a confined atom with that for a free H atom to see how the modified properties of the caged atom affect its HHG spectrum. The results are shown in Fig. 1. We perform three calculations: first we calculate the HHG spectra for free (pink) and confined (red) atoms in the $n = 2$ state using the same laser frequency ω_0 and intensity I. Because part of the 2s state lies inside the attractive cage potential, its ionization potential I_p is increased relative to the free atom. This results in a decreased tunneling rate which drastically reduces the HHG yield. Surprisingly, although the cut-off frequency at $3.17U_p$ does not depend on I_p, it is also shifted by almost 30 harmonic orders. Here U_p is the ponderomotive potential. In order to compare the caged spectrum with that for a free atom with the same level of yield, we perform a third calculation for a free H atom where we now fix the Keldysh parameter γ at 0.57 (blue dotted). In this case, we fix I and the increase ω_0 to match the γ for the caged spectrum (red). Increased ω_0 results in decreased U_p, which reduces the cut-off by ∼150 harmonic orders. We estimate that ∼120 out of these 150 orders are associated with the increase in ω_0.

We also performed the same set of calculations for the 1s and the 4s states. However, we did not see any dramatic shift in the cut-off frequency. For $n = 2$, the last peak of the bound state wave function lies inside the cage, suggesting that the cage radius relative to the spatial extent of the initial state wave function is important. The cusps seen in the spectrum of the caged H in Fig. 1 (red) resemble those seen in HHG spectra from ground state atoms due to Cooper minima when photoionization is present in addition to tunneling. We investigate the mechanism behind these cusps through extensive calculations of electron flux through the C_{60} surface.

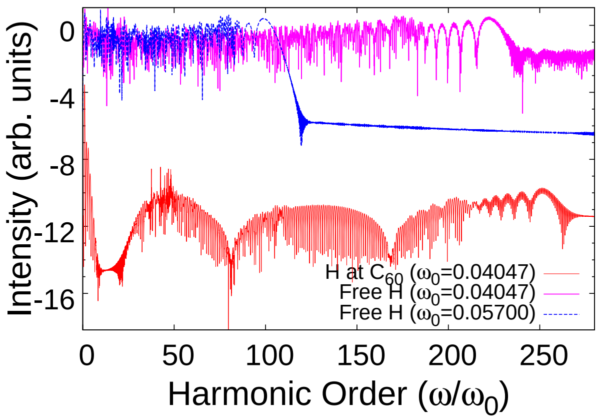

Figure 1. HHG spectra for free and confined H.

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