Supplementary Material for:
"High harmonic spectra computed using time-dependent Kohn-Sham theory with Gaussian orbitals and a complex absorbing potential"

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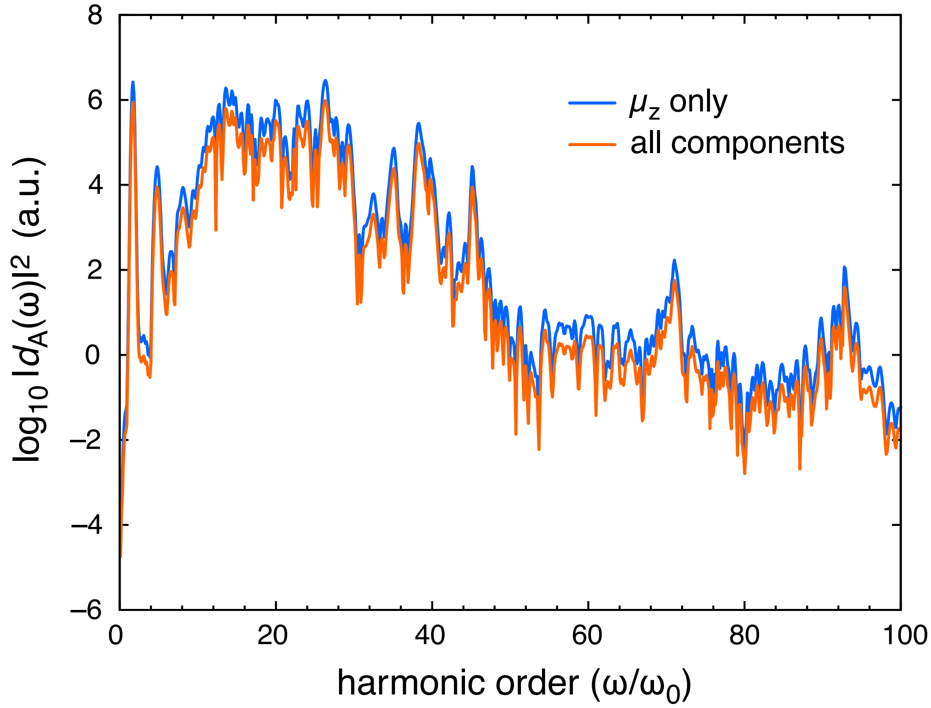

Figure S1: HHG spectra for H_2 computed at the TDHF/5-aTZ level. An impulsive field is applied, with parameters as in Fig. 1, and the simulations are propagated for 1,500 a.u. in time with no CAP. The Fourier transform used to obtain $|d_{\mathrm{A}}(\omega)|^2$ is computed either from $\mu_z(t)$ only (for a field that is polarized along the z axis) or else using all three components of the fluctuating dipole moment. The spectrum computed using μ_z only is the same as the TDHF spectrum in Fig. 4.

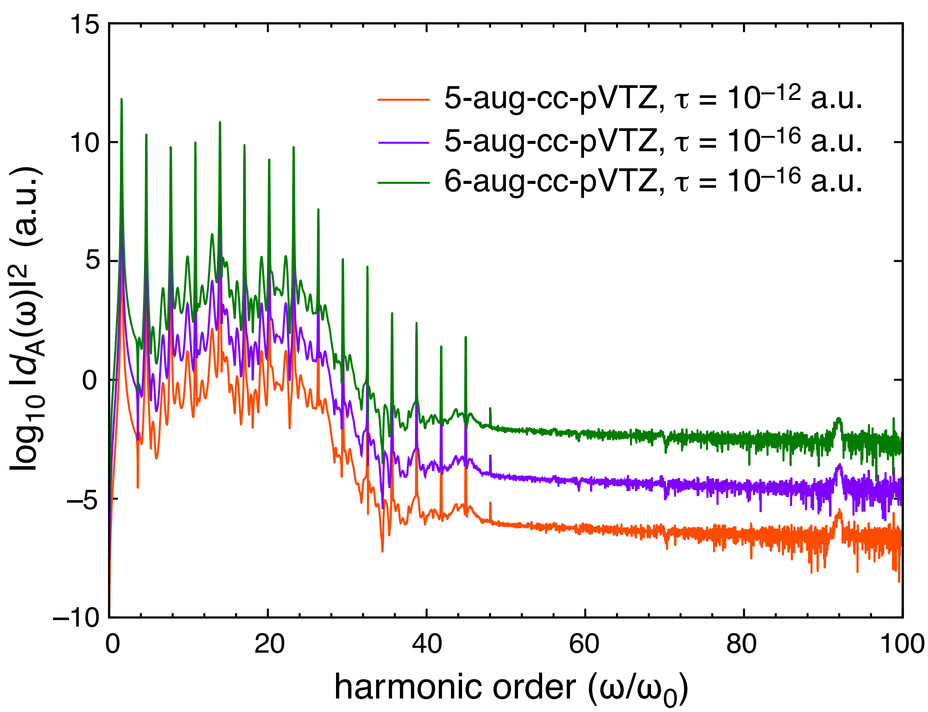

Figure S2: Convergence tests for TDHF/n-aTZ spectra for H_2, computed using a continuous wave field (see Fig. 1) and 5,000 a.u. of time propagation. CAP parameters are $r_0 = 9.524$ bohr and $\eta = 4.0$ Ha/bohr^2, as in Fig. 9a. Three spectra are shown, using either $n = 5$ or $n = 6$ diffuse shells and two different screening thresholds, τ. For legibility the three spectra have been offset vertically by successive factors of 100, as they are otherwise indistinguishable for the 60th harmonic.

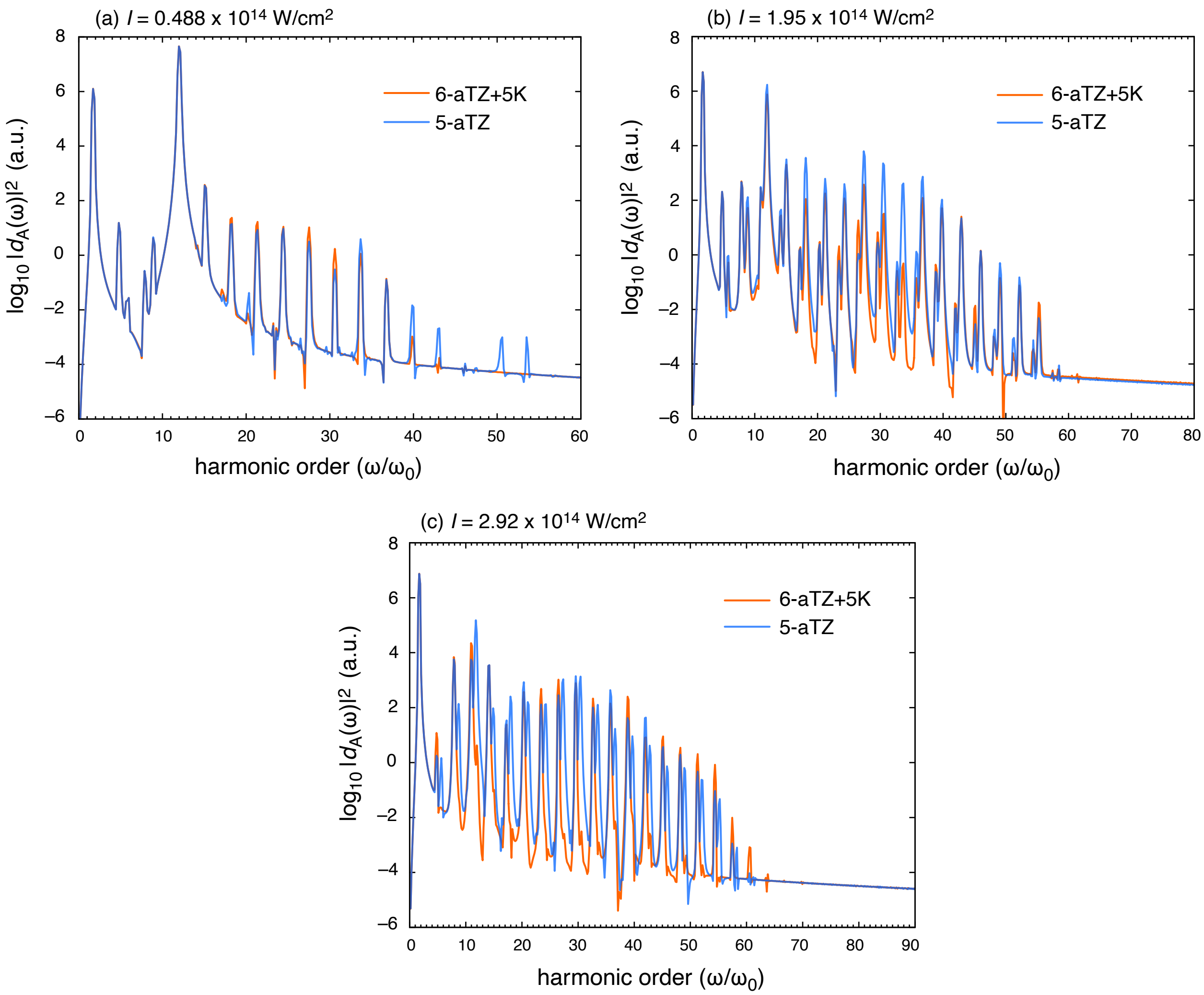

Figure S3: Convergence tests for HHG spectra of H_2^+ computed at the TDHF level in two different basis sets. These spectra were computed using a continuous-wave field with $\hbar\omega_0 = 1.55$ eV and $\lambda = 800$ nm, with three different laser intensities I as indicated. The TDHF equation was propagated for 2,000 a.u. time time using $\Delta t = 0.1$ a.u. but only the final 1,000 a.u. of data were used to compute the spectrum, analogous to the windowing procedure used in Fig. 11b. All simulations used a CAP with parameters $r_0 = 9.524\ a_0$ and $\eta = 4.0\ E_\mathrm{h}/a_0^2$. The 5-aTZ simulations used a threshold $\tau = 10^{-12}$ a.u. while for the 6-aTZ+5K simulations this was tightened to $\tau = 10^{-16}$ a.u., which we find to be necessary in the case of 6-aTZ in order to obtain numerically stable results.

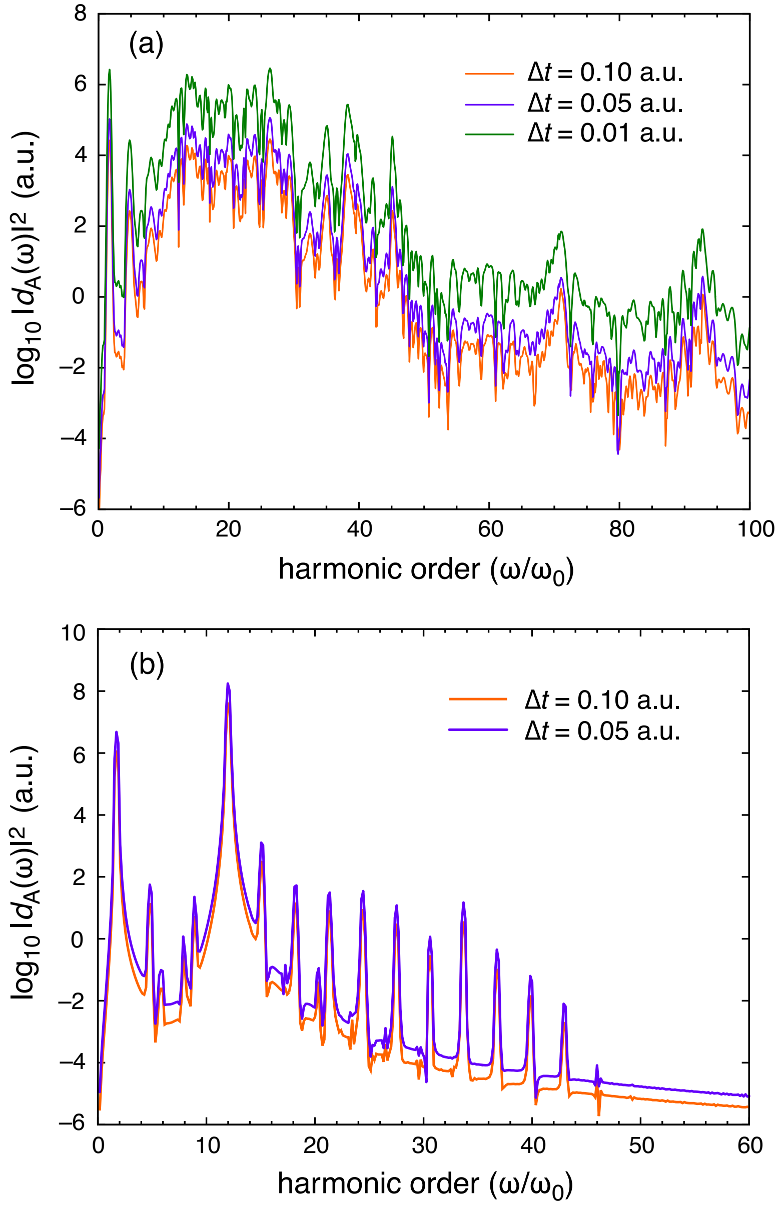

Figure S4: Tests of convergence for HHG spectra computed at the TDHF/5-aTZ level using different values of Δt. (a) HHG spectra for H_2, computed using an impulsive field corresponding to $I = 0.877 \times 10^{14}$ W/cm^2 (Fig. 1). All three simulations are propagated for 1,500 a.u. in time with no CAP, and the spectrum computed using $\Delta t = 0.10$ a.u. is the same as the TDHF spectrum in Fig. 4. Differences in intensity arise from the better resolution of the Fourier transform when Δt is smaller, but all of the peak structure is the same for $\Delta t = 0.10$ a.u. as it is for $\Delta t = 0.01$ a.u.. (b) HHG spectra for H_2^+ computed using a CAP with parameters $r_0 = 9.524\ a_0$ and $\eta = 4.0\ E_\mathrm{h}/a_0^2$. Both simulations used a continuous-wave field corresponding to $I = 0.488 \times 10^{14}$ W/cm^2, propagated for 5,000 a.u. in time. Spectra shown here are computed using only the final 1,000 a.u. of time propagation, analogous to the procedure used in Fig. 11e.

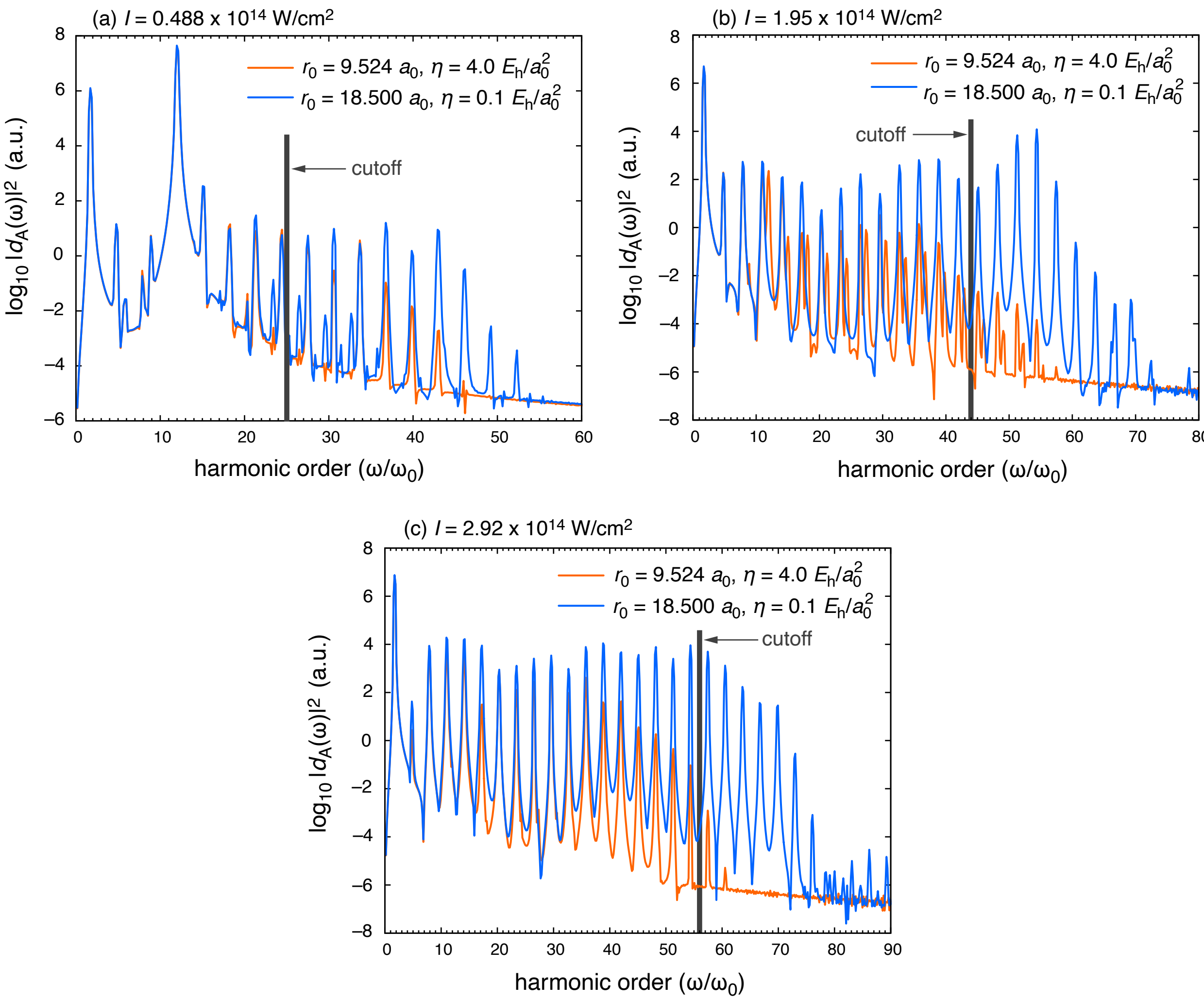

Figure S5: HHG spectra of H_2^+ computed at the TDHF/5-aTZ level using two different CAPs (with parameters as indicated) and three different laser field intensities, I. These are the same spectra as in Fig. 8 but presented here without the comparison to previous literature, for clarity. All simulations use a continuous-wave field and were propagated for 5,000 a.u. in time using $\Delta t = 0.1$ a.u.. The spectra were then computed using the final 1,000 a.u. of data only, analogous to the procedure used in Fig. 11e. (This was done in Fig. 8 as well.) Cutoff energies are indicated, estimated using Eq. (7).

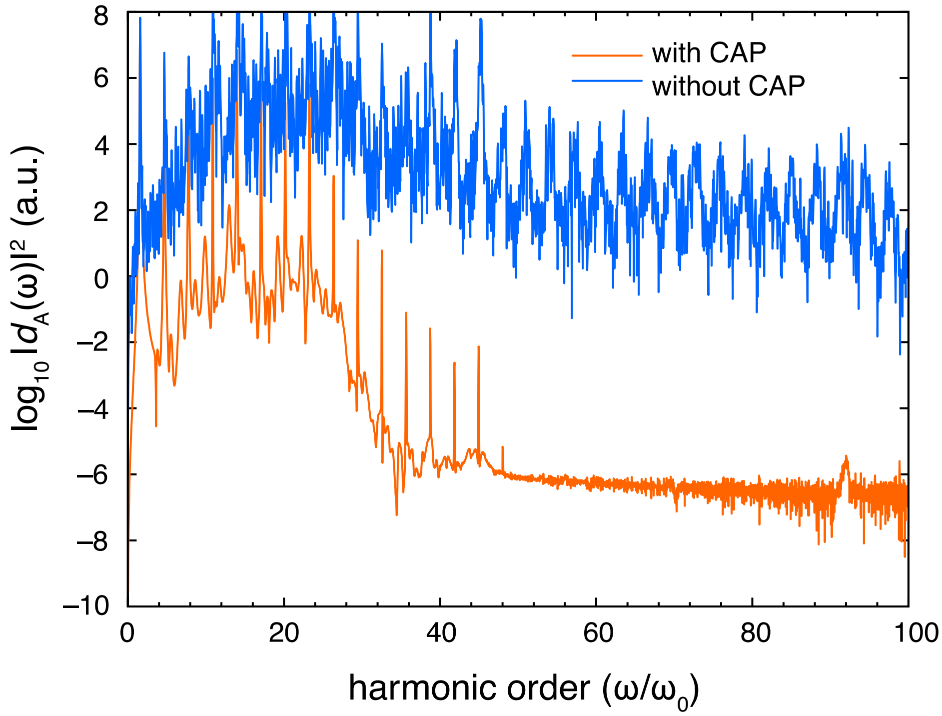

Figure S6: HHG spectra for H_2 computed at the TDHF/5-aTZ level using a continuous-wave field with parameters as in Fig. 1, propagated for 5,000 a.u. of time either with or without a CAP. Results with a CAP are the same as in Fig. 9a and use CAP parameters $r_0 = 9.524\ a_0$ and $\eta = 4.0\ E_\mathrm{h}/a_0^2$.

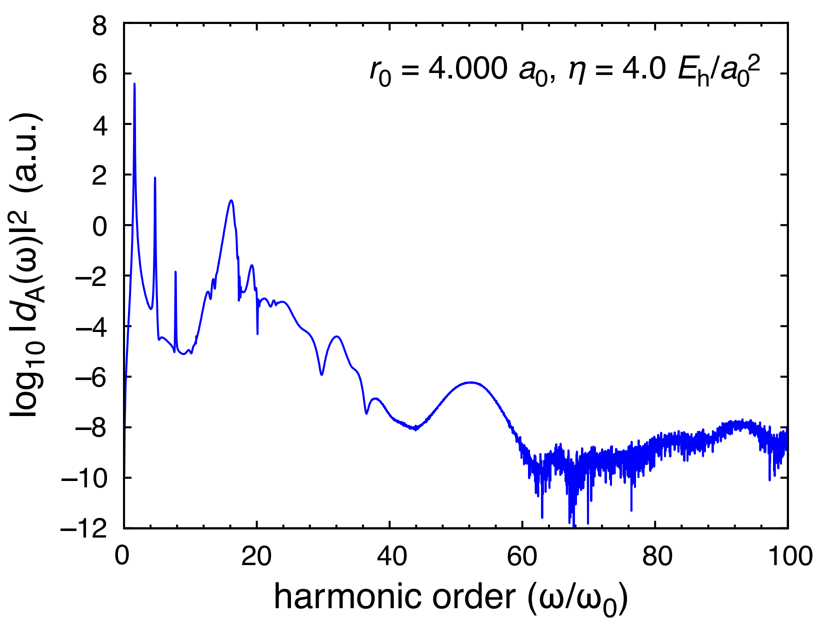

Figure S7: Continuous-wave HHG spectrum for H_2 computed at the TDHF/5-aTZ level using CAP parameters $r_0 = 4.0\ a_0$ and $\eta = 4.0\ E_\mathrm{h}/a_0^2$, based on 5,000 a.u. of time propagation. This is the same setup as in Fig. 9 but with a much smaller value of r_0 used here.

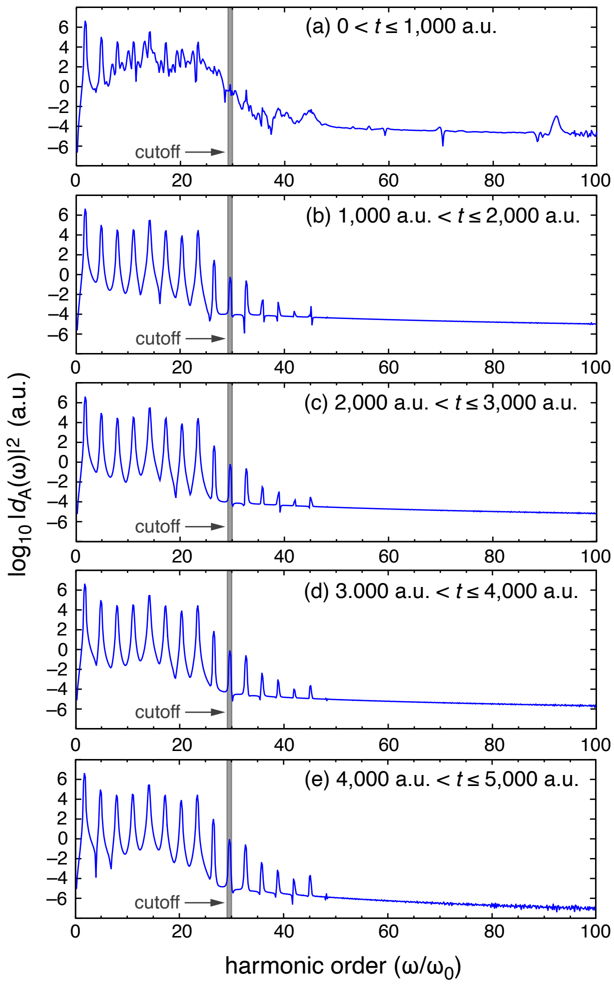

Figure S8: HHG spectra for H_2 computed at the TDHF/5-aTZ level from a continuous-wave simulation with CAP parameters $r_0 = 9.524\ a_0$ and $\eta = 4.0\ E_\mathrm{h}/a_0^2$, as in Fig. 9a. The total simulation time of 5,000 a.u. has been divided into consecutive segments and the spectra in panels (a)–(e) are obtained from the Fourier transform of the $\mu_z(t)$ data within just one segment, as indicated. The logarithmic intensity scale is the same in each panel and the cutoff predicted by Eq. (7) is indicated. The same windowing procedure is considered in Fig. 11 but using different CAP parameters.