**Supplemental Data and Figures**

***Ab-initio* multi-scale simulation of high-harmonic generation in solids**

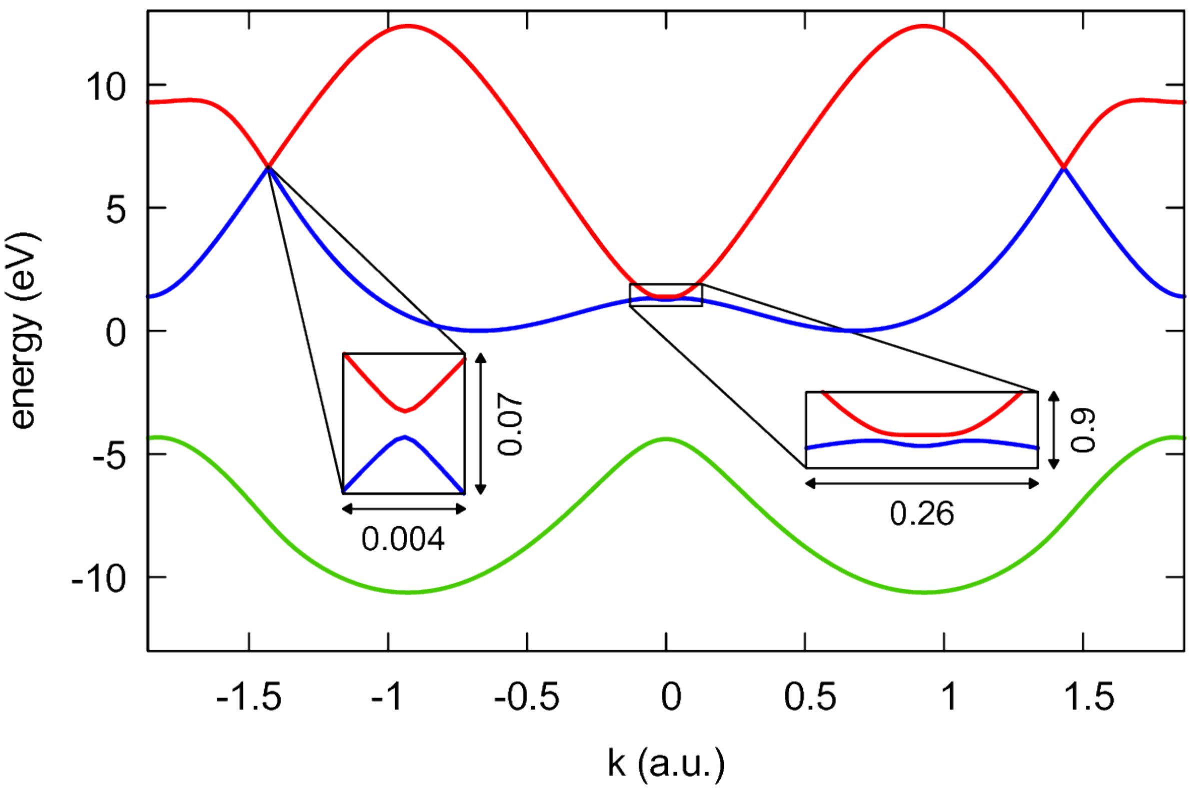


FIG. S1. (Color online) Cut through the Brillouin zone of diamond (calculated from DFT using LDA) along a line parallel to Γ−X [which is at $\vec{k}_\perp = (0, 0)$] with $\vec{k}_\perp = (0.03, 0.03)$ displaying narrow avoided crossings. The bands at the avoided crossing on the left (left inset with a total $\Delta k = 0.004$ a.u.) are separated by $\Delta E = 13$ meV. Agreement between TDDFT and SBE results crucially depends on such detailed representation of the band structure.

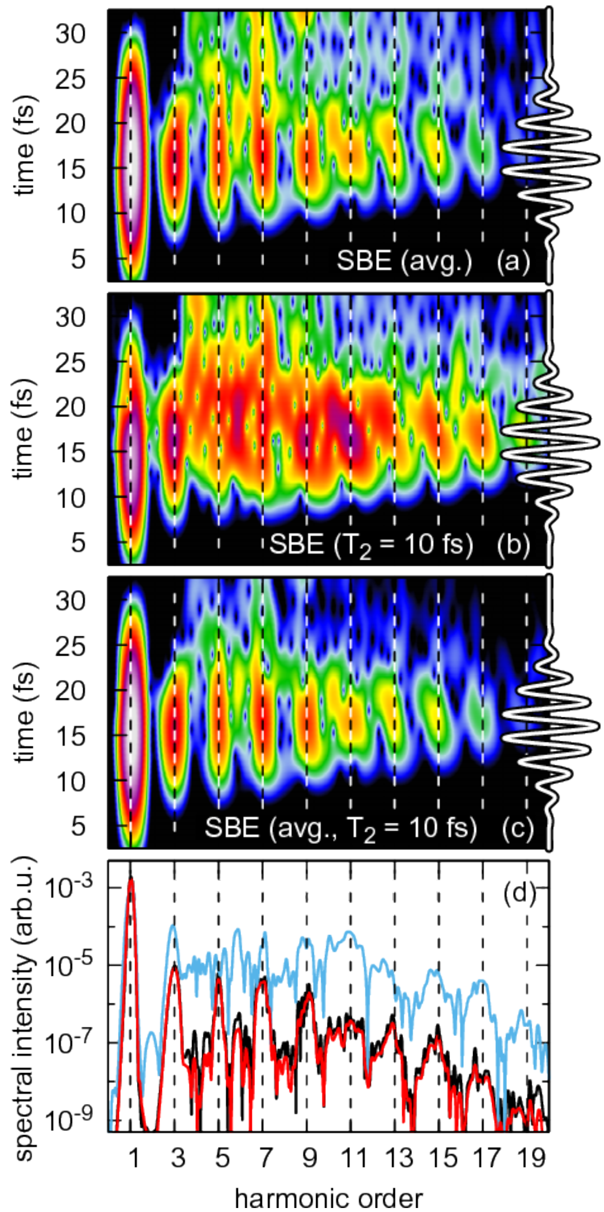


FIG. S2. (Color online) Time-frequency analysis (Gabor transform with width $\sigma = 2$ fs on a logarithmic color scale) of the radiation emitted from diamond; single-cell calculations for $\lambda = 800$ nm, $\tau_\mathrm{p} = 6.8$ fs (FWHM) and $I_0 = 2 \times 10^{13}$ W/cm$^2$ (intensity in the material) of the driving laser pulse shown on the right-hand side. (a) SBE calculation as in Fig. 2 (main text) without dephasing ($T_2 \to \infty$) but with averaging over the transverse field distribution [Eq. (6) in main text], (b) SBE calculation as Fig. 2 (main text) without field averaging but with dephasing ($T_2 = 10$ fs), (c) SBE calculation with both $T_2 = 10$ fs and transverse field averaging, (d) resulting harmonic spectra corresponding to (a), (b), and (c) shown in black, blue, and red, respectively. The spectra are scaled to match the intensity of the first harmonic. The dashed vertical lines indicate the odd multiples of the carrier frequency of the exciting laser pulse.

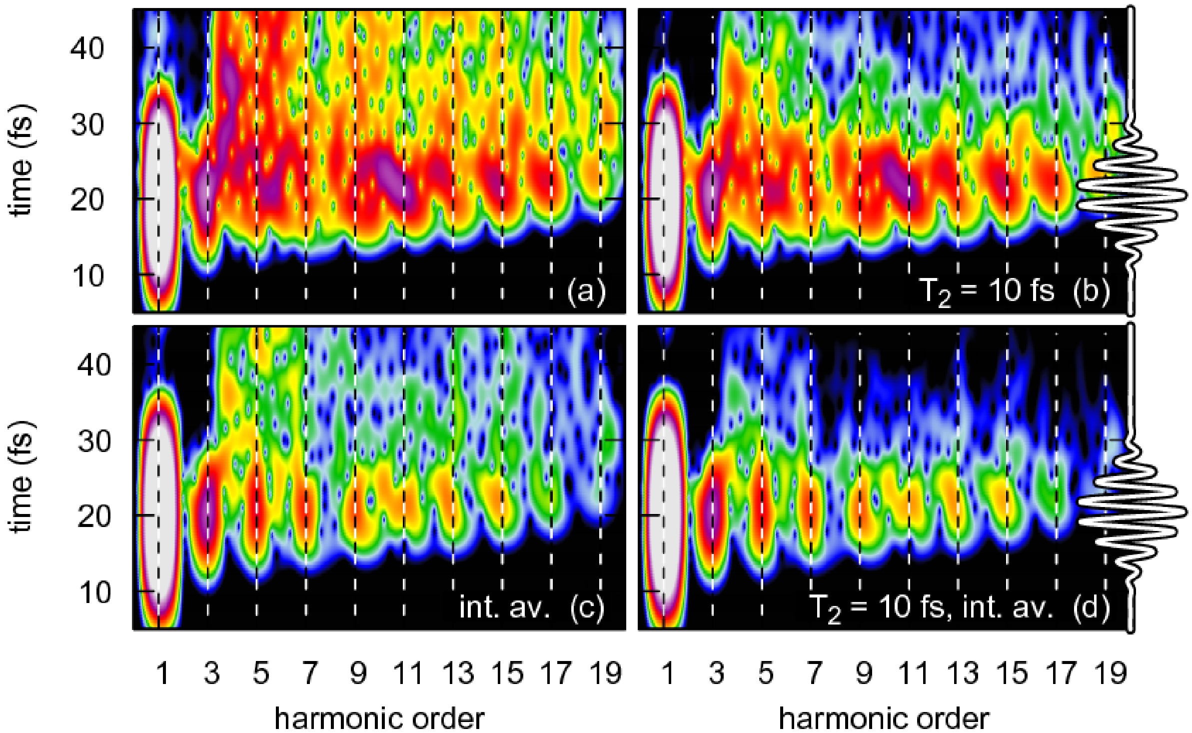


FIG. S3. (Color online) Time-frequency analysis (Gabor transform with width $\sigma = 2$ fs on a logarithmic color scale) of the emitted radiation induced by a laser pulse with $\lambda = 800$ nm, $\tau_\mathrm{p} = 6.8$ fs (FWHM) and incident (vacuum) intensity $I_\mathrm{vac} = 2 \times 10^{13}$ W/cm$^2$ (same parameters as single cell calculation in Fig. 2a and b) from self-consistently solving the Maxwell-Bloch equations for diamond with a thickness of $\lesssim 1$ nm (single layer) without ($T_2 \to \infty$) (a,c) and with dephasing ($T_2 = 10$ fs) (b,d). Without field averaging (a,b) and with field averaging (c,d). The dashed vertical lines indicate the odd multiples of the carrier frequency $\omega_\mathrm{IR}$ of the incident laser pulse. Note that for $\lesssim 1$ nm thick diamond both the reduction of intensity inside the material due to reflection as well as the blue shift of the driving frequency (see main text) are negligible.

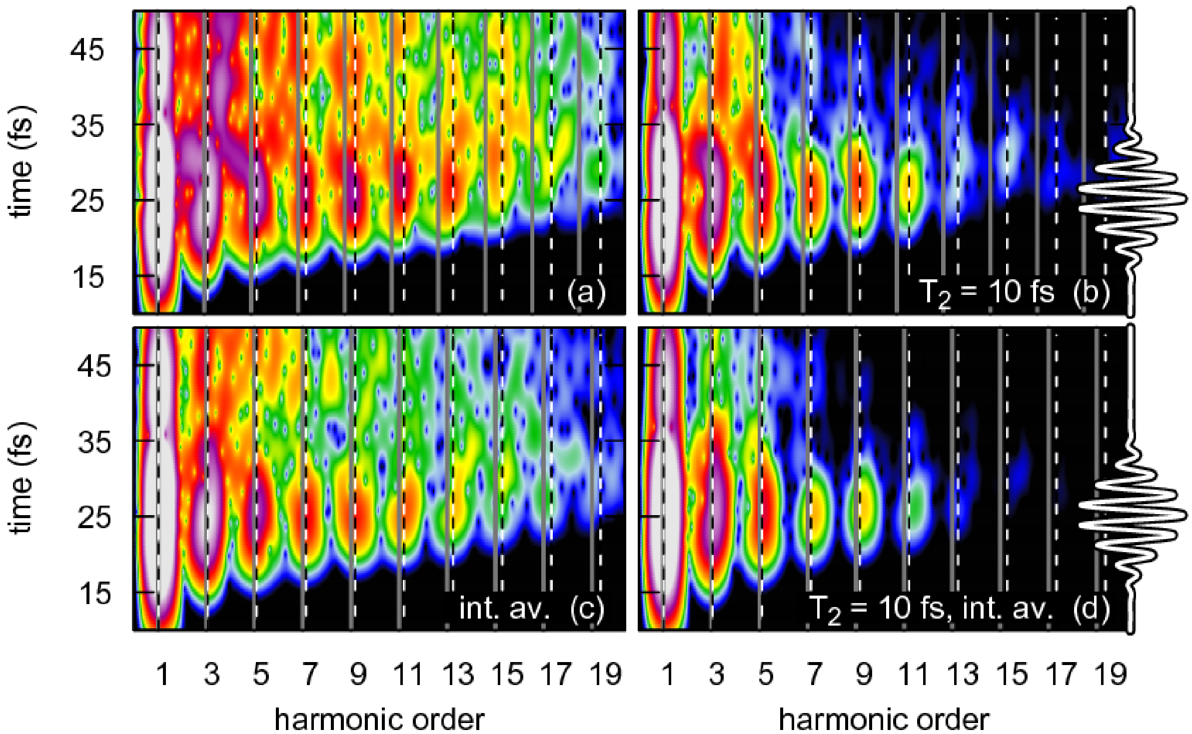


FIG. S4. (Color online) Same as Fig. S3 (logarithmic color scale extended by two orders of magnitude) but for a layer thickness of 1 $\mu$m. (a,d) correspond to Fig. 4a, 4b in the main text and are shown for comparison, (b) with dephasing ($T_2 = 10$ fs) and (c) with intensity averaging. The dashed vertical lines indicate the odd multiples of the frequency $\omega_{\mathrm{t}}$ of the blue shifted transmitted pulse (see main text). For comparison the odd multiples of the frequency $\omega_{\mathrm{IR}}$ of the incident pulse are shown as solid gray lines. It is worthwhile noting that the frequency shift $\delta\omega = \omega_{\mathrm{t}} - \omega_{\mathrm{IR}}$ strongly depends on the incident intensity and, thus, is reduced by intensity averaging.