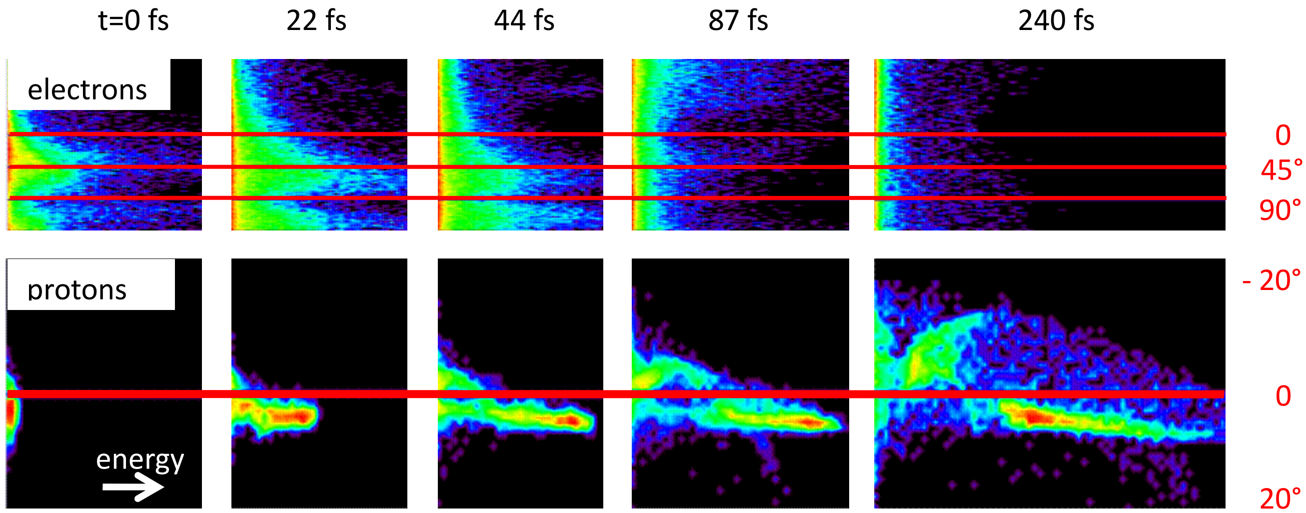
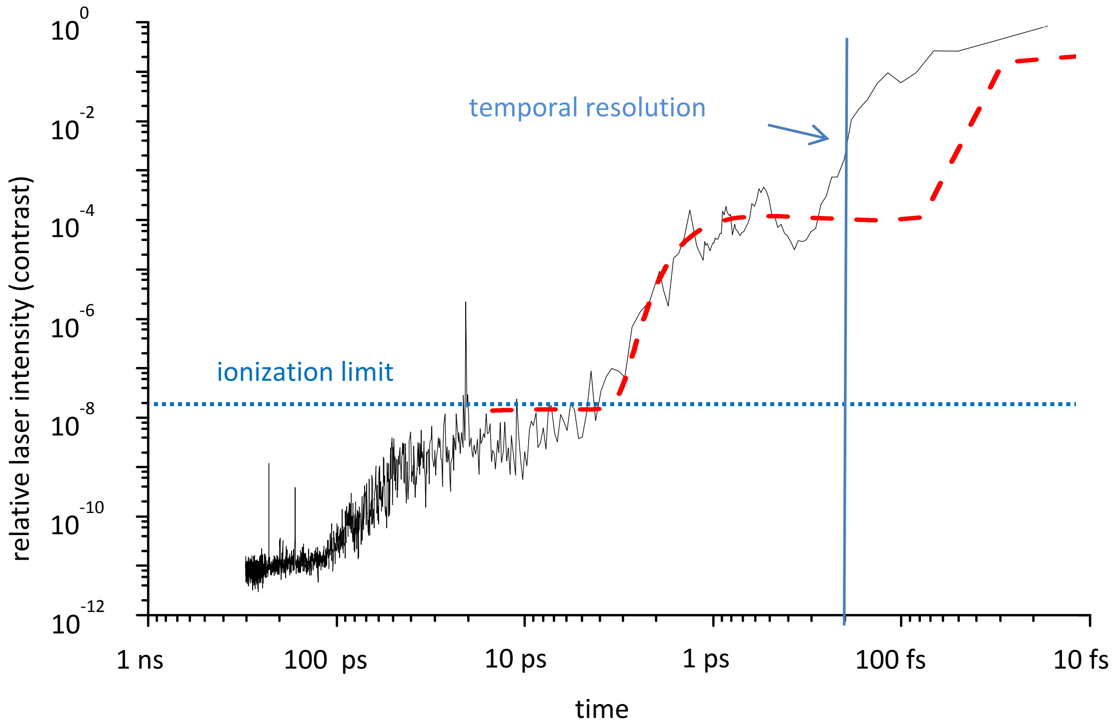
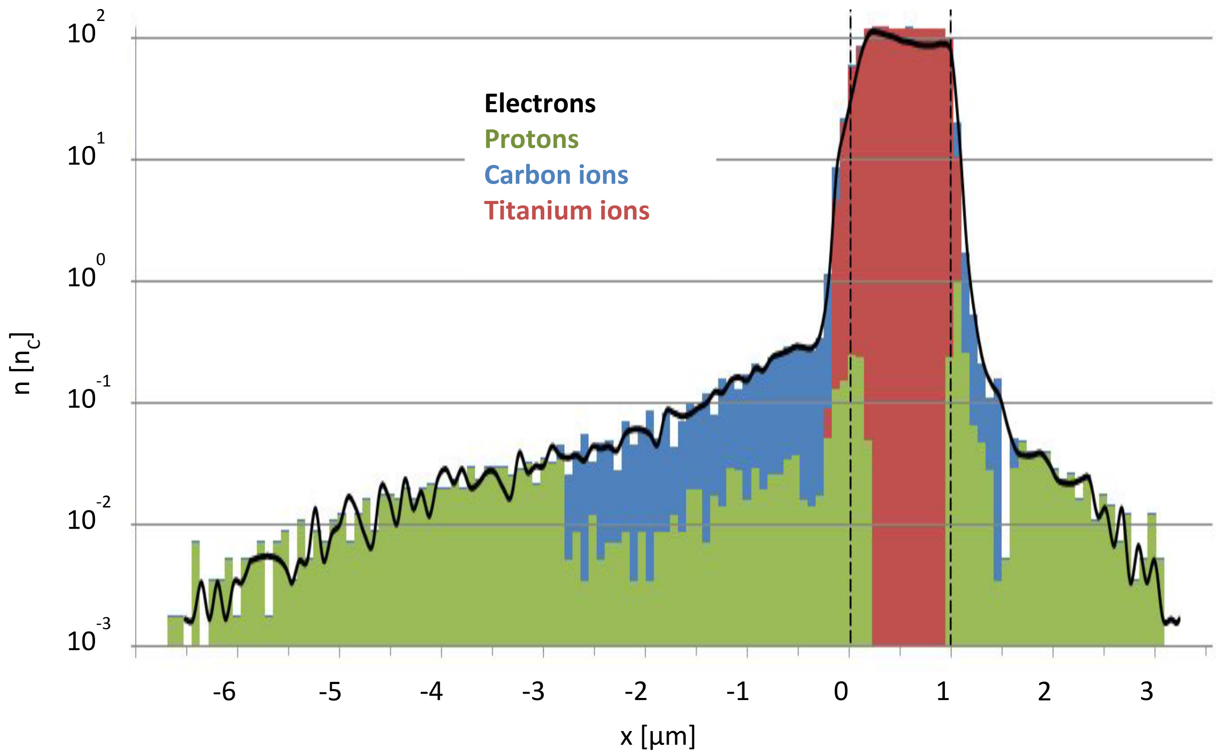


Supplementary Fig. S1 Observables of the intra-pulse acceleration dynamics. Colour coded (logarithmic rainbow scale) images of simulated spatial energy distributions for electrons (upper row) and protons (lower row) are plotted as a function of the emission angle for consecutive time steps. At t=0 fs the peak of the laser pulse reaches the target front surface. Electrons are ejected from the target rear along the direction of the incident laser beam (45°). Almost immediately (refer to t=22 fs) protons start to gain energy and follow the highest field gradients. Whereas the asymmetry in the electron distribution completely vanishes at later time steps t>44 fs, the proton distribution conserves the initial deflection angle during the later expansion phase of the sheath. This simulation illustrates how the intra-pulse dynamic of the promptly accelerated electrons translates into a signature that can be experimentally detected any time later, the deflection of the angular proton spectrum. The spectral modulations visible in the proton distribution can be attributed to multi-species effects.



Supplementary Fig. S2 Temporal laser pulse contrast. The contrast as measured by third order autocorrelation (black line) and the simplified curve taken as input for the particle-in-cell simulation (dashed red line) are shown in double logarithmic scaling. Note, that experimental data between 10 and 100 fs is resolution limited due to the measurement technique. In this range a SPIDER (Spectral phase interferometry for direct electric-field reconstruction) measurement yields a pulse length of 30 fs. Two shoulders in the pulse contrast are characteristic: The first is reached around 50 ps before the main pulse at a relative level of 10^{-8} and thus below the ionization threshold and the second extends from 3 ps until the arrival of the main laser pulse at a level of 10^{-4}. The simulation starts 3.5 ps in advance of the main laser pulse in order to self-consistently include all contributions above the ionization threshold.



Supplementary Fig. S3 Snapshot of the density distribution (in units of the critical density) 80 fs before the main pulse arrives. Originating from the surface of a 1 µm thick titanium foil protons on the front illuminated (left) side are leaking out into the vacuum for up to 6 µm. Thus, the main pulse interacts with underdense plasma for several micrometers before reaching the Ti-foil. The back side slope is steeper and protons can be found up to 2 µm behind the foil. The critical density surface has only moved by a few hundred nanometers. Significant deformations of the target rear side influencing the emission characteristics of energetic protons can therefore be safely neglected.