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Relativistic high-harmonic generation and correlated electron acceleration from plasma mirrors at 1 kHz repetition rate

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We report evidence for the first generation of XUV spectra from relativistic surface high-harmonic generation (SHHG) on plasma mirrors at a kilohertz repetition rate, emitted simultaneously and correlated to the emission of energetic electrons. We present measurements of SHHG spectra and electron angular distributions as a function of the experimentally controlled plasma density gradient scale length L for three increasingly short and intense driving pulses: 24 fs (9 optical cycles) and $a_0 = 1.1$, 9 fs (3.5 optical cycles) and $a_0 = 1.8$, and finally 4 fs (1.7 optical cycles) and $a_0 \approx 2.0$. For all driver pulses, we observe relativistic SHHG in the range $L \in [\lambda/25, \lambda/10]$, with an optimum gradient scale length of $L \approx \lambda/15$.

Surface high-harmonic generation (SHHG) [1] from relativistic plasma mirrors is a promising method for greatly enhancing the available energy of attosecond XUV pulses. This is motivated by the absence of an inherent limitation for the driving intensity such that extremely large numbers of photons from ultra-high intensity lasers can be converted into attosecond XUV pulses. In strongly relativistic conditions with a normalized vector potential $a_0 = \sqrt{I[\text{W cm}^{-2}] \lambda_0^2[\mu\text{m}^2]/(1.37 \times 10^{18})} \gg 1$, where I is the laser intensity and λ_0 the central wavelength, this is expected to occur with extremely high, percent-level conversion efficiencies [2–4]. Reported experimentally observed laser-to-XUV conversion efficiencies for plasma mirrors with $a_0 \sim 1$ are $\sim 10^{-4}$ [5–8], but are expected to increase with higher-intensity drivers.

Reaching relativistic SHHG regime with $a_0 > 1$ requires an on-target intensity of $\gtrsim 10^{18}$ W/cm² for an 800-nm laser while retaining a very steep surface plasma density profile, i.e. a profile $n(x) = n_c \exp[x/L]$, with a scale length L of a small fraction of the driving laser wavelength. Here n_c is the nonrelativistic critical plasma density for the driving wavelength and x is the coordinate in the target normal direction. Technically this requires a highly focusable terawatt-class driver laser with a temporal contrast of $\gtrsim 10^{10}$. These conditions are typically met by Joule-class amplifier chains with dedicated contrast filters [9, 10] and operating at ~ 10 Hz repetition rate [4–9].

Many applications as well as parametric studies of this regime would benefit from a higher repetition rate. At LOA, we have developed a unique laser chain with power-scaled hollow-core-fiber postcompression system [11] operating at 1 kHz repetition rate. Using this kHz-laser, which achieves ultra-high intensities with few-mJ pulse energy and few-cycle pulse duration, we have demonstrated laser-plasma interaction in the relativistic regime through laser-wakefield acceleration of electrons both in underdense gas jets [11, 12] and in the underdense part of a smooth plasma density gradient on a plasma mirror [13]. Here we report on the first experimental demonstration of relativistic SHHG at kHz-repetition rate, the arguably most demanding application in terms of laser performance as it depends critically on the spatio-temporal pulse quality and the temporal contrast.

For relativistic driving intensities, $a_0 \gtrsim 1$, the SHHG emission mechanism is described by a push–pull process [14], also dubbed “relativistic electron spring” [15, 16], repeating once per driving laser period. The incident laser field first pushes electrons into the plasma, piling up a dense electron bunch and creating a restoring internal plasma field. As the laser field changes sign, the combined plasma and laser fields accelerate the electron bunch to a relativistic velocity towards the vacuum. SHHG

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emission then either results from a pure phase modulation of the incident laser field by the relativistically moving critical-density plasma surface [“relativistic oscillating mirror” (ROM)] [2, 17, 18], or as a coherent synchrotron emission (CSE) from the relativistically moving dense electron bunch as it gets accelerated orthogonally by the laser electric field [3, 19, 20].

For $a_0 \sim 1$, this relativistic SHHG emission needs to be discriminated from coherent wake emission (CWE) [1, 21], generated already at sub-relativistic intensities, $a_0 < 1$. Once per laser field cycle, Brunel electrons [22] are accelerated out and back into the surface plasma to form, via collective trajectory crossings, a spatially localized electron density peak. This peak traverses the overdense part ($n(x) > n_c$) of the plasma density gradient and in its wake excites plasma oscillations at the local plasma frequency, which finally lead to the emission of one attosecond light pulse per laser cycle. Clear criteria are known for the discrimination of the two types of SHHG emission.

(i) *Spectral extent*: CWE emission extends no further than the maximum plasma frequency given by the solid-target density, which corresponds to a cutoff-photon-energy of ≈ 30 eV for an SiO₂ target. In contrast, for sufficiently high driving intensities, relativistic SHHG can extend well beyond this CWE cutoff.

(ii) *Intensity dependence*: CWE is a linear process, i.e. the laser-to-SHHG conversion efficiency of CWE is constant over a large driving intensity range $> 10^{15}$ W/cm² [1], whereas relativistic SHHG is highly nonlinear, i.e. the conversion efficiency increases rapidly over a large a_0 -range from $a_0 \sim 1$ until a saturation in the ultra-relativistic limit $a_0 \sim 100$ [1, 20]. Note however that in the experimentally still well out-of-reach ultra-relativistic regime, the radiation reaction force may significantly modify SHHG [23].

(iii) *Phase properties*: The intensity-dependence of the Brunel trajectories leads to increasing delays between successive attosecond pulses during the driving pulse envelope, and thus to frequency-broadened negatively chirped individual harmonics [21, 24]. A positively chirped driving pulse can partially compensate the effect and consequently lead to spectrally narrower CWE-harmonics [21, 25]. In contrast, for moderately relativistic $a_0 \sim 1$, the relativistic SHHG emission is Fourier-limited [1] such that despite its much steeper intensity dependence and consequently shorter temporal envelope, its harmonic spectral width is narrower than that of CWE harmonics generated in the same conditions [1, 9, 26]. Only for higher $a_0 \gtrsim 10$, plasma surface denting due to radiation pressure will again induce an intrinsic harmonic chirp, with the opposite sign compared to that of CWE [1, 27].

(iv) *Dependence on the plasma density gradient*: In CWE, the electron density peak formation and conversion of plasma oscillations to light are both optimized for rather steep plasma density gradients. The optimal scale length is $L \sim \lambda_0/100$ for higher-order CWE harmonics, followed by a rapid drop for increasing L [1]. Furthermore, the intrinsic harmonic chirp and thus the spectral width of CWE-harmonic peaks increases with L [21, 25]. In contrast, the optimal L for relativistic SHHG is longer and has been consistently found in single-shot experiments near a scale length $L \approx \lambda_0/10$ [4, 5, 26, 28]. This can be shown to be consistent with particle-in-cell simulations, which often treat initially step-like plasma density profiles with variable peak density n . This adjusts the similarity parameter $S = n/(n_c a_0)$, which scales laser plasma-interaction in the relativistic regime [29]. The optimal conditions for the relativistic push-pull process, relying on the formation and dynamics of a sharp and high electron density spike, are predicted for $S \approx 1$ to 5, i.e. a few times overdense plasmas [15, 16, 20]. In the experimentally relevant case for solid targets of a smooth surface plasma density profile rising up to $\sim 100n_c$ bulk density, one may define an effective S_{eff} , as demonstrated in ref. [30] for a linear density ramp.

Considering instead the exponential density profile $n(x)$ defined in the introduction, the balance of the laser’s Lorentz force and the electrostatic restoring force exerted by the plasma ion background [14, 27] yields a position of the electron density spike, averaged over the “pushing cycle” of the laser field, of $x_b = L \ln \{ [a_0(1 + \sin \theta) \cos \theta] / [2\pi(L/\lambda_0)] \}$, where θ is the laser incidence angle on the plasma surface. Experimentally varying the plasma density gradient scale length L thus corresponds to adjusting the effective plasma (ion) density $n[x_b(L)]$. This yields an effective similarity parameter $S_{\text{eff}} = n(x_b)/(n_c a_0) = (1 + \sin \theta) \cos \theta / [2\pi(L/\lambda_0)]$. The range $S_{\text{eff}} \in [1, 5]$ is thus achieved for $L \in [\lambda_0/30, \lambda_0/6]$, independently of the

laser peak intensity. This is perfectly consistent with the existing experimental results as well as with those we report in this letter. Finally, for longer L , SHHG disappears due to the onset of chaotic electron dynamics in the long low-density [4].

(v) *Simultaneous electron emission*: The same optimal $L \approx \lambda_0/10$ has been found for the emission of high-energy electrons from a plasma mirror [31], which is therefore found to be correlated to relativistic SHHG [4] and be anti-correlated to CWE [32].

The experiment was carried out using the 1-kHz Salle Noire laser system at LOA, delivering pulses with durations adjustable between 24 and < 4 fs and temporal contrast ($> 10^{10}$) [11]. For SHHG, the p-polarized pulses are focused by an $f/1.5$, 30° off-axis parabola onto a rotating optically flat fused silica target at an incidence angle of $\theta = 55^\circ$ to an $\approx 1.8\text{-}\mu\text{m}$ FWHM spot. With a pulse energy of 2.6 mJ on target, this yields, supposing absence of spatio-temporal distortions, peak intensities of 2.5×10^{18} W/cm² ($a_0 = 1.1$ for $\lambda_0 = 800$ nm), 7×10^{18} W/cm² ($a_0 = 1.8$ for $\lambda_0 = 800$ nm), and 1.0×10^{19} W/cm² ($a_0 = 2.0$ for $\lambda_0 = 720$ nm) for 24-fs, 9-fs and 4-fs pulses, respectively. We thus operate in an intensity regime where both CWE and relativistic SHHG can be expected to give significant contributions to the generated harmonic signal. A spatially superposed pre-pulse, created by picking off and then recombining $\approx 4\%$ of the main pulse through holey mirrors, is focused to a much larger $13\ \mu\text{m}$ FWHM spot in order to generate a homogeneous plasma expanding into vacuum. A pre-pulse lead time τ of a few ps lets us density gradient scale length $L = L_0 + c_s\tau$. The expansion speed c_s is measured with spatial-domain interferometry (SDI) [33] and L_0 is the scale length increase owing to the limited temporal contrast of the main pulse. The fact that we do observe CWE emission (see below) justifies the assumptions that $L_0 \lesssim \lambda_0/50$.

The emitted SHHG radiation in the specular direction is recorded using an angularly resolving XUV spectrometer, including a gold-coated flat-field grating (600 lines/mm, 85.3° incidence) and micro-channel-plate (MCP) and phosphor screen detector. The MCP is time-gated for ≈ 250 ns synchronously with the laser pulses so as to suppress longer background plasma emission. The phosphor screen is finally imaged by a charged-coupled-device (CCD) camera to record angularly resolved ($[-30, 30]$ mrad) SHHG spectra. This only detects the central part of the the emitted SHHG beam, which in our conditions, owing in particular to the small source spot, has a FWHM-divergence of $\gtrsim 70$ mrad. The SHHG-spectra shown in the following are angularly integrated over the full range and represent the MCP signal without correction for the spectral response of the XUV spectrometer so as to enhance the visibility of the higher harmonic orders.

Synchronously, we detect the angular emission profile of electrons with energies $> 150\text{keV}$ with a lanex-screen [34], placed between the specular and the target-normal directions imaged by another CCD camera. The distributions along the azimuthal angle (in the incidence plane) shown in the following are integrated over the polar angle (perpendicular to the incidence plane).

All data shown are acquired by integrating over 100-ms long bursts of pulses at 1 kHz repetition rate, i.e. over 100 shots.

Figure 1(a) shows results obtained with a near-Fourier-limited 24-fs driver pulse. The pre-pulse lead time is scanned between 0 and 10 ps. Via an SDI measurement, this translates to a change of plasma density gradient scale length from L_0 to $L_0 + \lambda_0/10$, where $\lambda_0 = 800\text{nm}$. Two L -regimes are clearly distinguished. At the shortest gradients $L - L_0 > \lambda_0/50$, the harmonic peaks are broad and their intensity quickly drops with increasing L , as expected for CWE emission. At higher L , the harmonic peaks are remarkably narrower and their intensity goes through an optimum located at $L - L_0 \approx 0.06\lambda_0$, consistent with the theoretical expectations for relativistic SHHG emission as well as with earlier experimental observation with similar multi-cycle driver pulses [4, 5, 26, 28]. We also remark that no harmonics are visible beyond the CWE-cutoff at 30 eV, even for softer gradients. This is not very surprising as the laser intensity is only just around the relativistic threshold, $a_0 = 1.1$. Finally, we find a stronger electron emission at softer gradients, correlated to the (presumed) relativistic SHHG emission.

We can corroborate this interpretation by using a ‘‘Dazzler’’ acousto-optic programmable dispersive filter in our amplifier chain to add 500 fs^2 GDD to the driving pulses, thus chirping them positively, stretching them to 85 fs duration and consequently decreasing the intensity to 0.7×10^{18} W/cm² ($a_0 = 0.6$ for $\lambda_0 = 800$ nm). As

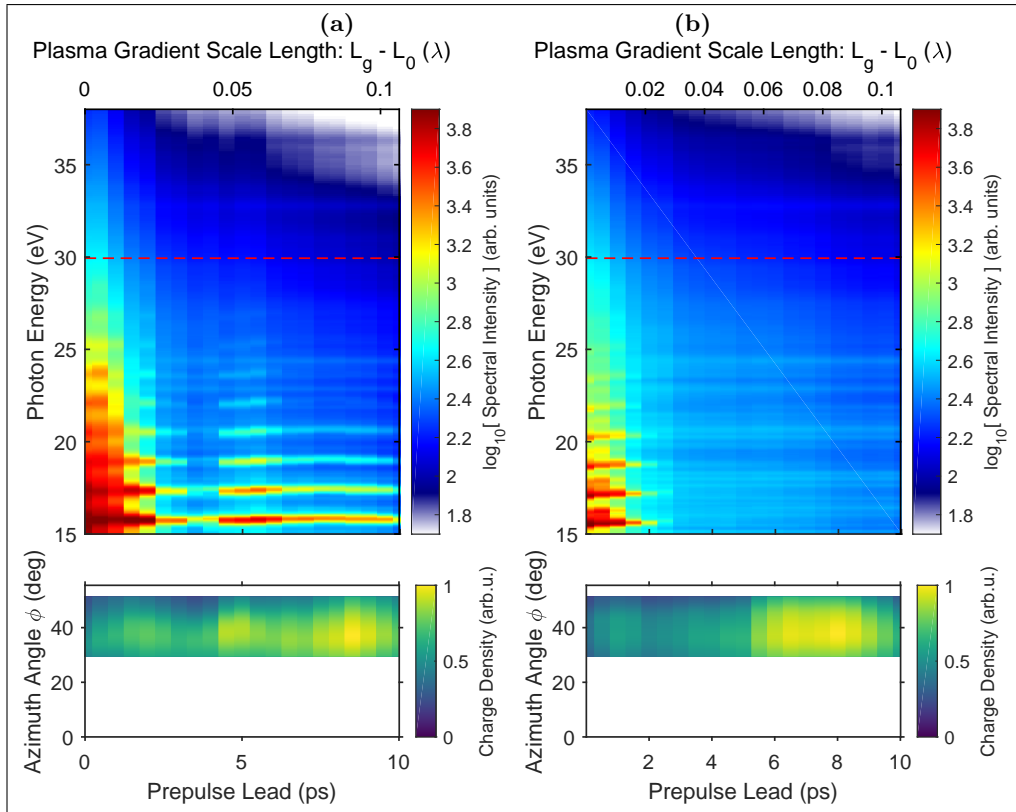


FIG. 1. Synchronously measured SHHG (upper, log-scale) and electron (lower) emission as a function of the plasma gradient scale length. The dashed red line marks the CWE cutoff energy. The normal and specular directions correspond to $\phi = 0$ and $\phi = 55^\circ$. (a) For a compressed, 24-fs, $a_0 = 1.1$ driver pulse, (b) for the same pulse with added 500 fs^2 GDD, 85 fs duration, $a_0 = 0.6$.

shown in figure 1(b), the harmonic spectral widths reduce significantly at the shortest L , but remain at similar intensity. This is consistent with the compensation of the aperiodicity of the CWE attosecond pulse train and the linearity of the CWE process. The SHHG emission at softer gradients however disappears completely, consistent with a highly nonlinear intensity-dependence of relativistic SHHG. Electrons are still emitted preferentially for the softer gradients and the resulting anti-correlation of harmonics and electrons is reminiscent of our earlier sub-relativistic-intensity results reported in ref. [32].

Reducing the driving pulse duration to 9 fs at constant pulse energy increases the peak normalized vector potential to $a_0 = 1.8$ and leads to the results shown in Figure 2. The same features as for the 24-fs driver can be observed, i.e. a transition from spectrally broad harmonics at the shortest L towards a “cleaner” well-modulated harmonic spectrum around an optimal $L - L_0 \approx 0.06\lambda_0$, and a correlation of the relativistic SHHG with electron emission for softer gradients. Additionally, we now observe a spectral extent of the SHHG signal slightly beyond the CWE-cutoff, which is definite proof for relativistic SHHG, reported here for the first time with a 1-kHz repetition rate. For longer L , the relativistic SHHG signal persists until at least $L = \lambda/5$ but their intensity and spectral extent gradually decreases.

An even shorter 4-fs driving pulse may be expected to strongly boost the SHHG signal beyond the CWE cutoff. Self-steepening in the helium-filled hollow-core fiber used for post-compression however reduces the central wavelength to $\lambda_0 = 720\text{nm}$ [11] so that despite a constant on-target pulse energy, the peak normalized vector potential only increase to $a_0 = 2.0$. Furthermore, such short pulses become extremely sensitive to spatio-temporal couplings which can drastically reduce the achievable peak intensity. On many days we thus observe reduced spectral extent of the SHHG spectra generated with $\lesssim 4$ -fs pulses as compared to those obtained with 9-fs pulses shown in Fig. 2, as was the case for the experiments reported in ref. [35].

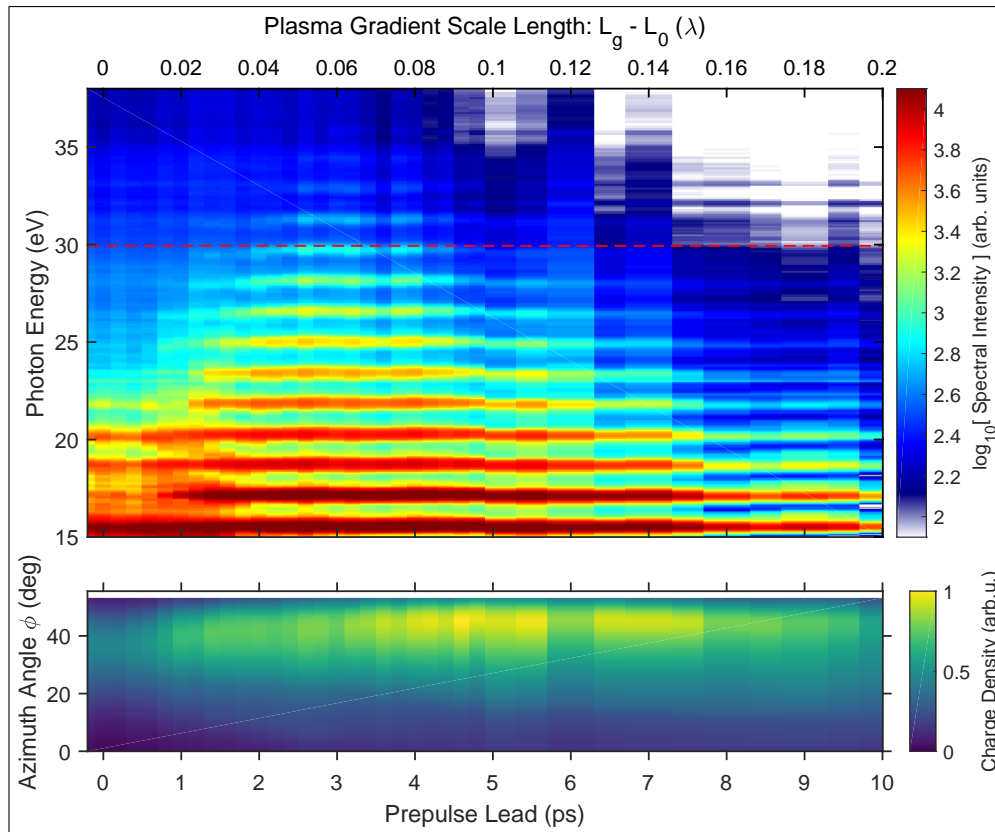


FIG. 2. Synchronously measured SHHG (upper, log-scale) and electron (lower) emission as a function of the plasma gradient scale length for a 9-fs, $a_0 = 1.8$ driver pulse. The dashed red line marks the CWE cutoff energy. The normal and specular directions correspond to $\phi = 0$ and $\phi = 55^\circ$.

We did nonetheless succeed in recording the dataset shown in Figure 3 with a 4-fs driving pulse and shot-to-shot randomly varying carrier-envelope phase. As for the 9-fs result, relativistic SHHG is optimized for plasma density gradient scale lengths between $L - L_0 \approx \lambda/25$ and $\lambda/10$, where $\lambda = 800$ nm, consistent with the theoretical prediction. The optimal relativistic SHHG spectra now extend well beyond the CWE cutoff. The spectra still present a discernible harmonic modulation, in particular for the softer gradients, and one can make out small gradual L -dependent shifts of the harmonic peaks. These may result from a varying temporal structure of the generated very short attosecond pulse-train. While all recorded spectra are continua, i.e. the harmonic peaks spectrally overlap, the absence of CEP-stability however precludes any further discussion of this temporal structure [35].

In conclusion, we have reported clear evidence for the first generation of XUV spectra from relativistic SHHG on plasma mirrors at a kilohertz repetition rate, emitted simultaneously and correlated to the emission of energetic electrons. We have presented and discussed measurements of SHHG spectra and electron angular distributions as a function of the experimentally controlled plasma density gradient scale length L for three increasingly short and intense driving pulses: 24 fs (9 optical cycles) and $a_0 = 1.1$, 9 fs (3.5 optical cycles) and $a_0 = 1.8$, and finally 4 fs (1.7 optical cycles) and $a_0 \approx 2.0$. For all driver pulses, we observe relativistic SHHG in the range $L \in [\lambda/25, \lambda/10]$, with an optimum gradient scale length of $L \approx \lambda/15$.

These results open the route to detailed examination and optimization of relativistic SHHG exploiting the high kHz-repetition rate and exceptional stability of our laser system. In particular for the shortest driver durations and with added CEP-locking, we should become able to harness the favorable phase properties of relativistic SHHG for the generation of powerful isolated attosecond pulses from plasma mirrors [35].

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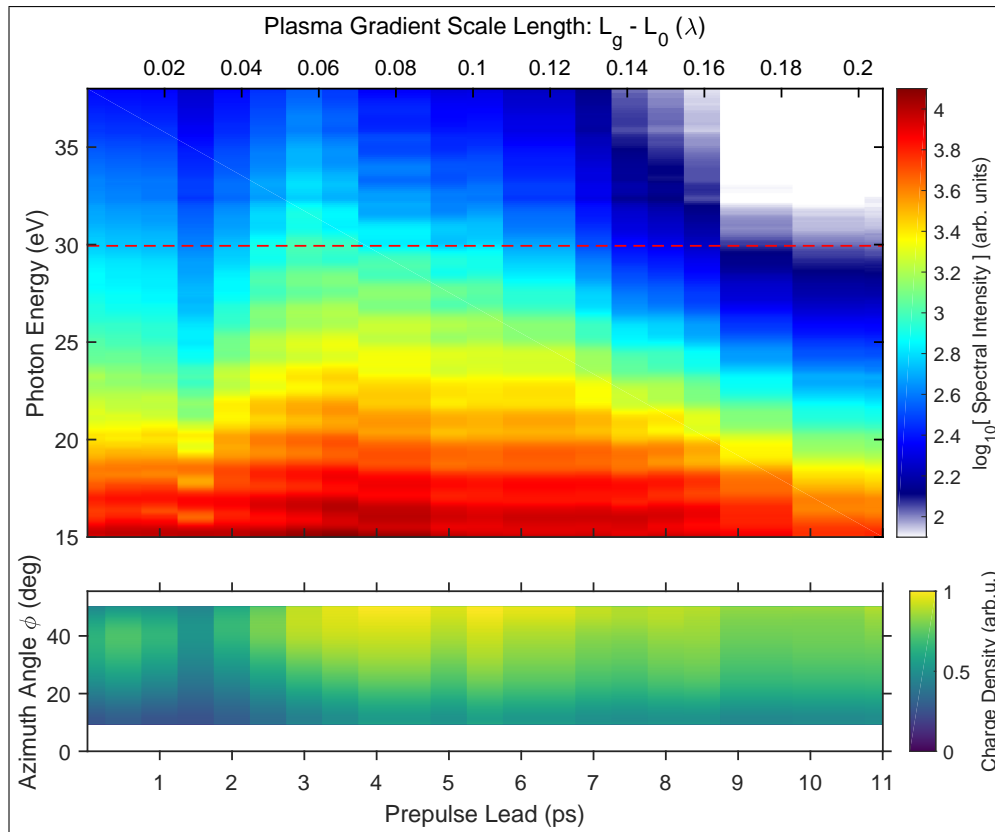


FIG. 3. Synchronously measured SHHG (upper, log-scale) and electron (lower) emission as a function of the plasma gradient scale length for a 4-fs, $a_0 = 2.0$ driver pulse. The dashed red line marks the CWE cutoff energy. The normal and specular directions correspond to $\phi = 0$ and $\phi = 55^\circ$.

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