Electron Acceleration by Relativistic Surface Plasmons in Laser-Grating Interaction

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The generation of energetic electron bunches by the interaction of a short, ultraintense ($I > 10^{19}$ W/cm$^2$) laser pulse with “grating” targets has been investigated in a regime of ultrahigh pulse-to-prepulse contrast ($10^{12}$). For incidence angles close to the resonant condition for surface plasmon excitation, a strong electron emission was observed within a narrow cone along the target surface, with energy spectra peaking at 5–8 MeV and total charge of $\sim$100 pC. Both the energy and the number of emitted electrons were strongly enhanced with respect to simple flat targets. The experimental data are closely reproduced by three-dimensional particle-in-cell simulations, which provide evidence for the generation of relativistic surface plasmons and for their role in driving the acceleration process. Besides the possible applications of the scheme as a compact, ultrashort source of MeV electrons, these results are a step forward in the development of high-field plasmonics.

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Surface plasmons [1,2], also named surface waves, are electromagnetic (EM) modes localized at the interface of different media which allow local field confinement and enhancement. Surface plasmons are the core of the vibrant research field of plasmonics [3], with applications ranging from light concentration beyond the diffraction limit [4], to biosensors [5] and plasmonic chips [6]. The extension of plasmonics into the regime of high fields, where nonlinear and relativistic effects arise, is largely unexplored. An example is provided by the multiterawatt laser-driven excitation of unipolar surface plasmons by transient charge separation [7,8], with potential application to the generation of intense THz pulses [8,9].

In the optical or near-infrared frequency range, surface plasmons can be excited by laser light incident on a sharp material interface having a periodic modulation, e.g., a grating, to allow phase matching. However, most experiments so far have been restricted to intensities below $10^{16}$ W/cm$^2$ [10] because of the prepulses inherent in high-power laser systems which can lead to an early disruption of the target structuring. The development of devices for ultrahigh contrast pulses [11,12] now allows us to explore the interaction with targets structured on a submicrometric scale at laser intensities high enough for the electron dynamics to become relativistic [13,14]. In particular, a strong increase of the cutoff energy of protons accelerated from the rear surface of grating targets was observed and related to surface plasmon-enhanced absorption [15]. While a detailed theory is still lacking for nonlinear and relativistic surface plasmons, numerical simulations also showed surface plasmon-related effects in this regime [16,17], including electron acceleration at weakly relativistic intensities [18] and, more recently, surface plasmon-enhanced high harmonics [19] and synchrotron radiation [20] in gratings.

In this Letter, we demonstrate that relativistic surface plasmons accelerate high-energy electrons along a grating surface. The acceleration process is related to two basic surface plasmon properties, i.e., the subluminal phase velocity and the longitudinal field component. The energy and number of electrons in gratings irradiated at an incidence angle close to the resonant value for surface plasmon excitation are strongly enhanced with respect to flat targets. At intensities $I = 5 \times 10^{19}$ W/cm$^2$, corresponding to a relativistic parameter $\alpha_0 = (I \lambda^2/10^{18}$ W cm$^{-2}$ µm$^2)^{1/2}$ and $\lambda$ is the laser wavelength, the electron emission was concentrated in a narrow cone with energy spectra peaking at 5–8 MeV and reaching up to $\sim$20 MeV.

The basics of surface plasmon generation and electron acceleration may be described as follows. At high laser intensities ($I > 10^{18}$ W/cm$^2$) a solid target is ionized within one laser cycle, thus the interaction occurs with a dense plasma. Assuming a dielectric function $\varepsilon(\omega) = 1 - \omega_p^2/\omega^2 \approx 1 - \alpha$ (where $\omega_p$ is the plasma frequency) the phase velocity of a surface plasmon is $v_p = \omega_0/\kappa = c(\alpha - 2)^{1/2}/(\alpha - 1)^{1/2}$ where $k$ is the surface plasmon wave vector.
and $\alpha > 2$ holds. The condition for resonant excitation of a surface plasmon on a periodically modulated target (grating) by an incident EM wave $[1,2]$ of the same frequency is $\lambda/\lambda_g = (1 - \alpha)^{1/2}/(2 - \alpha)^{1/2} - \sin(\phi_g)$ where $\lambda_g$ is the grating period and $\phi_g$ is the angle of incidence. Notice that these equations neglect the effects of finite temperature and nonsteplike density profiles $[21]$ as well as of collisions and possible relativistic nonlinearities $[22]$; thus, in principle the resonance could be expected at somewhat different angles.

Electron acceleration up to relativistic energy by the longitudinal surface plasma field requires the phase velocity $v_p$ to approach $c$ in order to minimize dephasing; thus, $\alpha \gg 1$ is required as expected for a solid-density plasma and optical frequencies. The basic process may be described similarly to the well-known acceleration in wake plasma waves $[23]$, but with the difference that the transverse field component of the surface plasmon (i.e., the field component perpendicular to the wave vector) pushes an electron on the vacuum side, so that the process is two dimensional and eventually the electrons are emitted at an angle with respect to the surface plasmon propagation direction. In a frame $L'$ moving with velocity $v_p = v_p\hat{y}$ with respect to the laboratory frame $L$, the surface plasmon field is electrostatic in the vacuum region ($x > 0$) and can be derived from the potential

$$\Phi' = -\frac{E_{SP}}{k'} e^{-k'x} \sin k'y',$$

where $k' = k/\gamma_p$, $\gamma_p = (1 - v_p^2/c^2)^{-1/2} = (\alpha - 1)^{1/2}$, and $E_{SP}$ is the amplitude of the longitudinal surface plasma field ($E_x$) in $L$. In the $L'$ frame the process is simply described as the electron going down the potential energy hill $-e\Phi'$. Because the evanescent field component $E'_x = -\partial_x \Phi'$, electrons are predominantly accelerated towards the vacuum side with velocity almost normal to the $x = 0$ surface. The condition of "optimal" injection corresponds to an electron placed initially in $L'$ at the top of the potential hill ($x' = 0$, $y' = \pi/2k'$) with $v'_z = 0$, i.e., with an initial velocity $v_p$ in $L$. Such an electron will acquire in $L'$ the energy $W' = eE_{SP}\gamma_p/k$. If $a_{SP} \equiv eE_{SP}/m_e c^2 \sim 1$ then $W' \gg m_e c^2$. In this limit, the energy momentum in $L'$ is $p'_p = (W', W'/c, 0, 0)$ and thus $p_p = (\gamma_p W', W'/c, \gamma_p W'/c, 0)$ in $L$. The final energy $\mathcal{E}_f$ and emission angle $\phi_e$ are given by

$$\mathcal{E}_f \equiv eE_{SP}p'_p/k = m_e c^2 a_{SP}, \quad \tan \phi_e \equiv p_x/p_y \equiv \gamma_p^{-1}.$$

Thus, strongly relativistic electrons ($\mathcal{E}_f \gg m_e c^2$) are emitted at small $\phi_e$, i.e., close to the target surface. The acceleration length $\ell_a \equiv \mathcal{E}_f/eE_{SP} \approx \lambda a/2\pi$, showing that electrons may reach the highest energy over a few microns. For linear conversion of the laser pulse into the surface plasmon, $a_{SP} \propto a_0$ is expected. Notice, however, that for high amplitudes $a_{SP}$ may be limited by wave breaking effects.

The experimental setup was carried out at the CEA Saclay Laser Interaction Center (SLIC) facility with the UHI100 laser system (see Ref. $[24]$ for a preliminary presentation of the experimental results). The laser pulse had 0.8 $\mu$m wavelength, 25 fs duration, and 2.3 J energy before compression. A double plasma mirror $[25]$ yielded a pulse contrast $\sim 10^{12}$ managing $\sim 50\%$ energy loss. The pulse was focused with $P$ polarization using an off-axis $f/3.75$ parabola in a focal spot of $\approx 4 \mu$m FWHM containing $\sim 60\%$ of the total energy in the $1/e^2$ spot diameter, which lead to an average intensity of $\sim 5 \times 10^{19}$ W/cm$^2$. Focal spot optimization was performed with an adaptive optical system. The schematic view of the experimental setup is shown in Fig. 1. A compact CMOS-based spectrometer, specifically designed for this experiment, was mounted on a motorized tray able to change the angle $\phi_{spec}$ within the range $0^\circ$–$60^\circ$ from the tangent, while remaining aligned to the interaction center. The entrance lead pinhole had a diameter of 500 $\mu$m and was placed at 8 cm from the interaction point. A pair of permanent magnets dispersed the electrons on the large (49.2 $\times$ 76.8 mm$^2$) triggered 12bit CMOS with 48 $\mu$m pixel size. The energy detection range was $\sim 2$–30 MeV. A scintillating Lanex screen ($16 \times 5$ cm$^2$) was used to collect the electron spatial distribution in the angular range $\phi = 0^\circ$–$90^\circ$. The screen was placed with an angle of 45$^\circ$ with respect to the target and its center was at 8 cm from the interaction point. The green light emitted by the Lanex was selected using a 546 nm band-pass filter and recorded by a 12bit CCD. When in use, the Lanex screen excluded the electron spectrometer. In addition to the electron diagnostics, a Thomson parabola was used to detect protons emitted along the rear target normal, as in a previous experiment $[15]$. The proton energy cutoff was used as a reference to optimize the target position.

The grating targets were produced at Czech Technical University, Prague by heat embossing of Mylar™ foils using a metallic master. Mylar was chosen considering its high damage threshold for prepulses. In the following we show results obtained with targets with a resonant angle...
absolute calibration of the Lanex screen, resulting on
bunch along the target surface was estimated from the
tions. The amount of charge accelerated in the collimated
design and alignment is foreseen to eliminate the fluctua-
in fluctuations of the direction of maximum emission.
of the target or nonexact perpendicularity of the grating
also found in previous measurements [15]). Local bending
firms the survival of the grating during the interaction, was
grating diffraction of the high-intensity pulse, which con-
image) are observed in the directions of specular reflection
at the same angle of incidence. Two minima (holes
larger than the maximum intensity observed for flat targets

FIG. 2. Images on the Lanex screen for simple flat target (top)
and for grating target (bottom), both irradiated at \( \phi_i = 30^\circ \)
incidence. A 3 mm Al foil was placed in front of
the screen to filter out electrons with energy \( E < 1.2 \text{ MeV} \).
The parabolic dashed lines give the local \( \theta \) angle corresponding to
the position on the screen. In order to highlight the features of
the signals, the upper panel has been amplified 2\times\ compared
to the bottom image. The right end (\( \Phi > 72^\circ \)) of the bottom image
has been amplified too (4\times\) to bring out the first-order diffraction
minimum.

of \( \approx 30^\circ \), i.e., \( \lambda_g = 2\lambda \) having assumed \( \omega_p \gg \omega \). The
average thickness was 10 \( \mu \text{m} \) and the peak-to-valley depth
of the grooves 0.25 \( \mu \text{m} \). Flat foils with the same average
thickness were used for comparison. In a limited number of
shots, gratings with a resonance angle of 15\( ^\circ \) (\( \lambda_g = 1.35\lambda \))
and 45\( ^\circ \) (\( \lambda_g = 3.41\lambda \)) were also used, obtaining similar
results.

Figure 2 shows the spatial distribution of the electrons
for both gratings irradiated at angles near resonance and the
flat foils. The emission from the flat foil is rather diffused,
with a local minimum in correspondence of the specular
reflection direction, as if electrons were swept away by the
reflected pulse. The signal is maximum in an annular region
around the hole. In contrast, for a grating at resonance the
emission is strongly localized on the plane of incidence
(\( \theta \approx 0^\circ \)). The maximum intensity of the electron signal (in
units of 1/sr) is close to the target tangent and is \approx 10\ times
larger than the maximum intensity observed for flat targets
at the same angle of incidence. Two minima (“holes” in
the image) are observed in the directions of specular reflection
and first-order diffraction of the laser pulse (evidence of
grating diffraction of the high-intensity pulse, which con-
firms the survival of the grating during the interaction, was
also found in previous measurements [15]). Local bending
of the target or nonexact perpendicularity of the grating
grooves to the plane of incidence may result in shot-to-shot
fluctuations of the direction of maximum emission.

Depending on the individual foil, the average angular shift
in \( \theta \) was in the 1\( ^\circ \)-5\( ^\circ \) range. An optimization of target
design and alignment is foreseen to eliminate the fluctua-
tions. The amount of charge accelerated in the collimated
bunch along the target surface was estimated from the
absolute calibration of the Lanex screen, resulting on
average in 100 \pm 15 \text{ pC}, spread in a 8.5\( ^\circ \) full angle cone.
As a comparison, the overall emission from the flat foil
around the specular reflection (\( m = 0 \)) was estimated in a
square region (35\( ^\circ \) full angle in \( \phi \) and \( \theta \)), resulting in
about 60 \text{ pC}.

The energy spectra were obtained placing the spectrom-
eter at \( \phi_{\text{spec}} = 2^\circ \). The angle of incidence \( \phi_i \) was varied
from 20\( ^\circ \) to 52\( ^\circ \). Figure 3 shows spectra obtained for
\( \phi_i \geq 30^\circ \), as for smaller angles no signal above the noise
level was collected. The aforementioned fluctuations of the
direction of the electron beam lead to a shot-to-shot
variability of the intensity of the signal. Nevertheless,
the most intense signals are detected only close to the
resonance angle (\( \approx 30^\circ \)). Moreover, spectra collected at 30\( ^\circ \)
and 35\( ^\circ \) are characterized by higher maximum energies
and a peculiar distribution with a dip at lower energies
(3–4 \text{ MeV}) and a broad peak at 5–8 \text{ MeV}, and a high-
energy tail extending up to \approx 20 \text{ MeV}. Electron spectra
above the noise level from flat targets at \( \phi_{\text{spec}} \approx 2^\circ \) were not
observed for any incidence angle. Spectra obtained for
\( \phi_i = 30^\circ \) and changing \( \phi_{\text{spec}} \) in the 1\( ^\circ \)–35\( ^\circ \) range showed a
similar shape in all positions of the spectrometer when
\( \phi_{\text{spec}} \lesssim 20^\circ \). The signal monotonically decreased in
intensity with respect to \( \phi_{\text{spec}} \) and was visible up to \( \phi_{\text{spec}} \approx 30^\circ \),
in agreement with the signal collected on the Lanex screen.

3D simulations were performed for flat targets at \( \phi_i = 30^\circ \)
and for gratings at \( \phi_i = 30^\circ, 35^\circ, 40^\circ \). For computa-
tional feasibility, the target thickness was \( \ell_t = 1\lambda \) and the
electron density was \( n_e = 50n_i \) (where \( n_i = \pi m_e c^2/\epsilon^2 \lambda^2 \)
is the cutoff density), while the other parameters corre-
sponded to the experimental ones. The simulations were
performed on 16 384 cores of the FERMI supercomputer
using the open-source, particle-in-cell code “PICCANTE”
[26,27]. The numerical box size was \( 80\lambda \times 80\lambda \times 60\lambda \),
wide enough for the boundaries not to affect the results.
A resolution of \( 70 \times 51 \times 34 \) points per \( \lambda \) and 50 particles
per cell were used.
Figure 4 shows a snapshot of a 3D simulation of a grating irradiated at the resonance angle in the same geometry of Fig. 1. The electric field component $E_x$ is represented together with the isosurface corresponding to the electron density. A surface plasmon propagating along the $-\hat{y}$ direction is excited. Figure 5(a) shows the simulated electron energy spectra $dN/dE$ at $\phi_{\text{spec}} = 2^\circ$ for the flat target irradiated at $\phi_i = 30^\circ$ and for gratings at $\phi_i = 30^\circ, 35^\circ, 40^\circ$. With respect to the gratings, the signal for the flat target is much weaker and the energy cutoff is $\sim 10$ times lower. The spectrum for the grating at $\phi_i = 30^\circ$ shows the peculiar shape observed for $\phi_i = 30^\circ, 35^\circ$ in the experiment (Fig. 3), while for larger $\phi_i$ the low energy dip disappears, as for $\phi_i \geq 40^\circ$ in the experiment. Figure 5(b) compares the spectra obtained in 2D and 3D simulations, showing that details such as the broad peak with low energy dip are reproduced only in 3D. The angular distribution on the screen Fig. 5(c) also reproduces the experimental data (Fig. 2) including the hole in the specular reflection direction.

The 3D simulation also shows a correlation between electron energy and the emission angle. Electrons at energies lower than the peak value are emitted at some angle with respect to the propagation direction of the surface plasmon, so that the 3D spectrum integrated over the whole range of $\theta$ resembles the 2D case. This is consistent with interpreting the fluctuations in the energy spectra (Fig. 3) as related to those in the electron beam direction.

In the simulation, $a_{SP} \approx 1$ showing that the surface plasmon is relativistic. Inserting such a value and $\alpha = 50$ in Eq. (2) we obtain a maximum energy $E_k \approx 25$ MeV and $\phi_e \approx 8^\circ$, in fair agreement with the observations considering the simplicity of the model. 2D simulations show that $E_f \propto a_0$ for $a_0 \lesssim 10$ while the scaling is slower for higher $a_0$, presumably due to nonlinear effects. Details of the spectrum will be dependent on the distribution of injection velocities. The Supplemental Material [28] includes movies from 2D simulations showing the acceleration of electrons along the surface. In a very recent work [29], self-injection and phase locking of electrons in the surface plasmon wave is studied with a test-particle approach.

The proposed scheme, with further optimization, may provide an intense ultrashort electron source in the MeV range, with characteristics not easily attainable with other techniques and potential applications including photoneutron generation [30,31] or ultrafast electron diffraction [32–34]. The study of relativistic surface plasmons may be further pursued in various schemes inspired by ordinary plasmonics, such as tapered waveguides for energy concentration and field amplification [35,36], and exploiting various target structurings including optically controlled transient gratings [37]. Such development of high field plasmonics will open new possibilities for the control and manipulation of ultraintense laser pulses and their interaction with matter.

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[28] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.116.015001 for animations showing the motion of some selected electrons in the fields near the grating surface. The color of the particles changes with the energy, showing that the strongest acceleration occurs along the surface. Also an animation showing the temporal evolution of the electric field in 3D is provided.


