# Experimental Evidence of Photon Acceleration of Ultrashort Laser Pulses in Relativistic Ionization Fronts

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The frequency up-shifts of ultrashort laser pulses (65 fs) propagating in opposite directions (20° and 160°) with respect to a relativistic ionization front (interface gas plasma) are measured for the first time. Up-shifts of the order of 25 nm are observed. A very good agreement is found with a two-dimensional ray-tracing theory. [S0031-9007(97)03381-4]

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In recent years, the concept of photon acceleration (or frequency up-shift by wave propagation in space and time-varying plasmas) has received considerable attention in plasma physics [1–5]. This is due to its potential use as a diagnostic tool for plasma based accelerators [6] and also, in a longer term, for the design of new types of tunable and ultrashort radiation sources [7]. One of the processes leading to a space and time-varying plasma is the production of a relativistic ionization front. Such a front can be created by optical-field induced ionization of a background neutral gas with an ultraintense laser pulse. This ionization front will have an ultrashort rise time (typically, a half pulse duration), and the plasma characteristics behind the front will only change in the recombination time scale ($\approx$ ms). The front will propagate with a velocity nearly equal to the group velocity of the laser pulse and, due to its relativistic velocity, will strongly interact with the probe photons (provided by a secondary, low energy laser probe pulse) and will up-shift their frequency significantly. It is now well understood that a significant frequency shift will occur even for plasma densities much lower than the cutoff plasma frequency [4,5]. This means that photon acceleration effects can be observed even if the probe photons are transmitted across the front. The same theory also predicts that the frequency up-shift will be much larger for photons copropagating with the front than for photons impinging on the front (or counterpropagating through it). Until now, experiments only dealt with a one-dimensional (1D) copropagation scheme [8].

In this Letter, we report the first experiments of photon acceleration in the optical domain, using a counterpropagating configuration. Comparison of the frequency shift in copropagation and counterpropagation enabled us to clearly identify the observed frequency up-shift as due to the photon acceleration mechanism and to perform the first detailed comparison between the theory and the experiments. In order to decouple the spectrum of the probe photon beam from the ionizing beam we chose an angle of incidence on the ionization front of 20°, for copropagation, and of 160° for counterpropagation. This means that the configuration of our experiments is intrinsically two dimensional (2D).

From a theoretical point of view, little attention has been paid to the collision of an incident (low intensity) laser beam with an ionization front in a 2D configuration. The generalization of the 1D results for 2D and for finite size ionization fronts can only be achieved in a straightforward way if the concepts of ray tracing or Hamiltonian dynamics are considered [5]. As before in the 1D models, it is possible to derive cutoff frequencies and the corresponding frequency up-shifts, for oblique photon collision with a semi-infinite ionization front. From the ray-tracing equations, it is quite easy to show that two constants of motion exist [5]: $I_1 \equiv \omega - \vec{k} \cdot \vec{v}_0$ and $I_2 \equiv \vec{k} \times \vec{v}_0/|\vec{v}_0|$, where $\omega$ is the local photon frequency, $\vec{k}$ the local wave vector, and $\vec{v}_0$ the velocity of the ionization front. Equating the frequency of the photons before and after the collision with the ionization front, and using the invariants $I_1$ and $I_2$, it is straightforward to derive the frequency up-shift for the several configurations of interest. Given a collision angle $\theta_0$, determined by $\vec{k}$ and $\vec{v}_0$, the maximum frequency up-shift $\omega_{\rm up}$ attained by a probe laser pulse, with initial frequency $\omega_0$, after collision with the front is [9]

$$\frac{\omega_{\rm up}}{\omega_0} = \frac{1 - \beta_f \cos(\theta_0)}{1 - \beta_f^2} + \frac{\beta_f |\beta_f - \cos(\theta_0)|}{1 - \beta_f^2} \tag{1}$$

for initial conditions, in vacuum, obeying the cutoff relation,

$$\omega_0 \leq \omega_\eta = \frac{\omega_{p0}\sqrt{1 - \beta_f^2}}{\sqrt{[1 - \beta_f \cos(\theta_0)]^2 - (1 - \beta_f^2)\sin^2(\theta_0)}}, \tag{2}$$

where $\beta_f = |v_0|/c$, and $\omega_{p0}$ is the maximum plasma frequency of the ionization front. Equations (1) and (2) reduce to the usual results for double Doppler shift in counterpropagation, if we take $\theta_0 = \pi$. Moreover, Eq. (2) shows that experimental conditions such that the probe laser pulse, propagating in the neutral gas region, is overtaken by the front and reflected back to the gas region can be easily achieved for $0 \leq \theta_0 \leq \pi/2$. This occurs in copropagation and leads to a double Doppler shift, which is, however, smaller than the well-known reflection in counterpropagation. The interest of this new reflection configuration is that it corresponds to the most favorable frequency shift and can occur for our experimental parameters. We shall call it the *copropagating relativistic mirror,* and describe it as a reflection in the phase plane $(k_z, \eta = z - v_0 t)$ [5]. When the cutoff condition (2) is not satisfied, and $\omega_0$ is larger than the cutoff frequency $\omega_\eta$, the frequency up-shift is given by a different expression, which can also be derived in a direct way using the constants of motion $I_1$ and $I_2$,

$$\frac{\omega_{\mathrm{up}}}{\omega_0} = \frac{1 - \beta_f \cos(\theta_0)}{1 - \beta_f^2} - \frac{\beta_f \sqrt{[\beta_f - \cos(\theta_0)]^2 - (\omega_{p0}^2/\omega_0^2)(1 - \beta_f^2)}}{1 - \beta_f^2}. \tag{3}$$

The analysis of Eqs. (1)–(3) shows that the frequency up-shift strongly depends on the incidence angle $\theta_0$. This situation would not occur if flash ionization [10], i.e., a spatial homogeneous time growing plasma, was the dominant frequency shift process. In fact, for flash ionization the maximum frequency shift is simply given by $\omega = \sqrt{\omega_0^2 + \omega_{p0}^2}$. Therefore, in order to clearly demonstrate the existence of photon acceleration by *moving* coherent plasma perturbations (in this case the ionization front), it is necessary to compare the frequency up-shift observed for two different values of the collision angle $\theta_0$.

The experimental setup is shown in Fig. 1. The photons to be accelerated (probe laser beam) correspond to a small fraction (0.05%) of the LOA dye laser-amplifier system. The main laser beam consists of nearly transform-limited pulses with an energy of 5 mJ and a duration of 65 fs. The central wavelength (in free space) is $\lambda = 620$ nm and the repetition rate is 10 Hz. The ionization front is produced by focusing 50% of the main laser beam, with an $f/8$ achromat (25 cm of focal length) in the border of a vacuum surrounded pulsed, supersonic, and high density ($10^{19}$ cm$^{-3}$) argon jet. A third laser beam (diagnostic beam) with a duration of 35 fs, central wavelength of $\lambda = 560$ nm at a repetition rate of 10 Hz is also produced by the LOA laser system. With this third beam we can measure the electron plasma density associated with the ionization front by using a Moiré interferometer represented in Fig. 1 by the two grids $G_1$ and $G_2$ [11]. In performing our experiments we have kept the probe laser beam in a fixed position. Copropagation and counterpropagation of this beam with respect to the ionization front was then obtained by changing the direction of propagation of the main laser beam (see Fig. 1). An angle of 20° between the two beams, the main and the probe beams, allowed us to decouple the observation of the blueshift of the probe photons from the expected blueshift of the main beam associated with the processes of plasma creation. This angle is then suitable to isolate, from an experimental point of view, the relativistic mirror effect in copropagation. The signal to noise ratio in the blueshifted signal is also increased by focusing the probe beam in the region of ionization front formation (which is the focal region of the main beam) by using an $f/6$ achromat (20 cm focal length). After the collision of the probe pulse with the front, the probe photons are sent to the spectrometer. The interaction region between the two beams is kept fixed but, by longitudinally moving the focusing achromat of the main beam optical circuit, we were able to introduce a variation of the electron plasma density. This is associated with a change of the diameter of the main pulse in the same region.

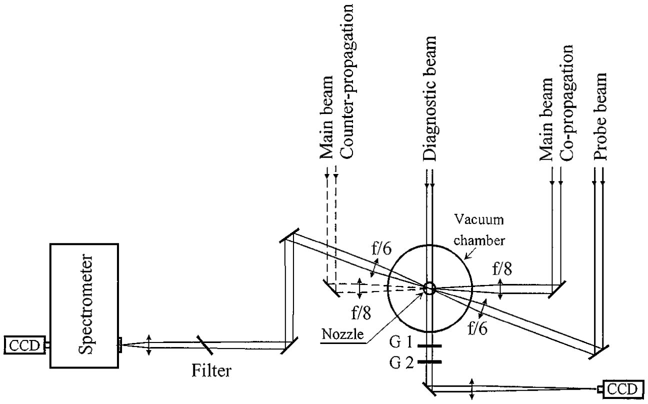

FIG. 1. Experimental setup.

As explained above, the experiments were performed in two different configurations: counterpropagation and copropagation, but in very similar experimental conditions. Typical spectra of the blueshifted probe beam are shown in Fig. 2 for counterpropagation and in Fig. 3 for copropagation. In Fig. 2(a), the focal point of the main beam is displaced in the forward direction 400 $\mu$m away from the interaction region (where the collision of the probe pulse with the ionization front takes place). The diameter of the main beam in this zone is then larger, by a factor of 2, than the minimum waist at the focal zone. In this configuration, the ionization front is well defined but the electron plasma density is not at its maximum value. A very clear displacement of the probe beam spectrum is observed, with a blueshift of 1.9 nm. If the interaction region approaches the focal zone of the main beam, when the electron plasma density associated with the ionization front gets closer to its maximum, this blueshift increases and approaches its maximum value, around 2.3 nm, as shown in Fig. 2(b). This effect is clearly associated with photon acceleration, as predicted by Eq. (3). But now, a second spectral shift of nearly double value, 4.8 nm in the same figure, is also observed and becomes dominant near

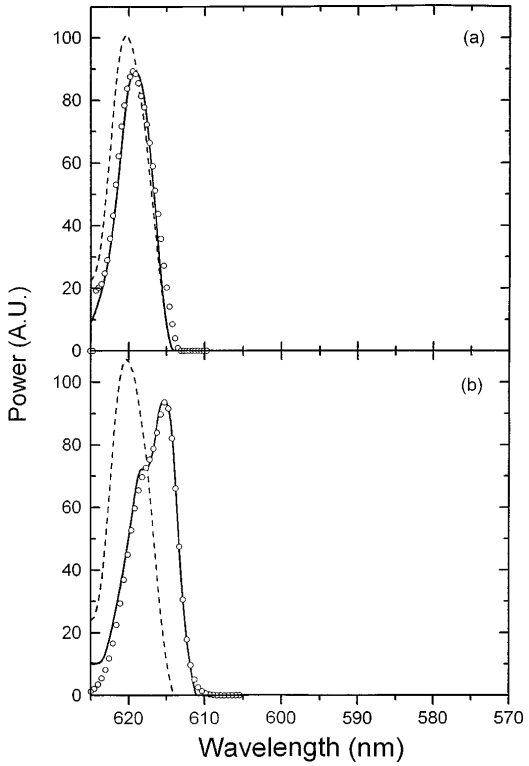

FIG. 2. Counterpropagating spectra ($\theta_0 = 160°$). (a) 400 $\mu$m away from the focal region: (dashed line) original pulse spectrum of the probe beam; (solid line) up-shifted spectrum; (circles) numerical simulation of the up-shifted spectrum for $n_e = 1.65 \times 10^{19}$ cm$^{-3}$ and $\beta_f = 0.941$. (b) At the focal region: (dashed line) original pulse spectrum; (solid line) up-shifted spectrum; (circles) numerical simulation of the up-shifted spectrum for $n_e = 4.32 \times 10^{19}$ cm$^{-3}$ and the same $\beta_f$, assuming that 56% of the photons experience flash ionization.

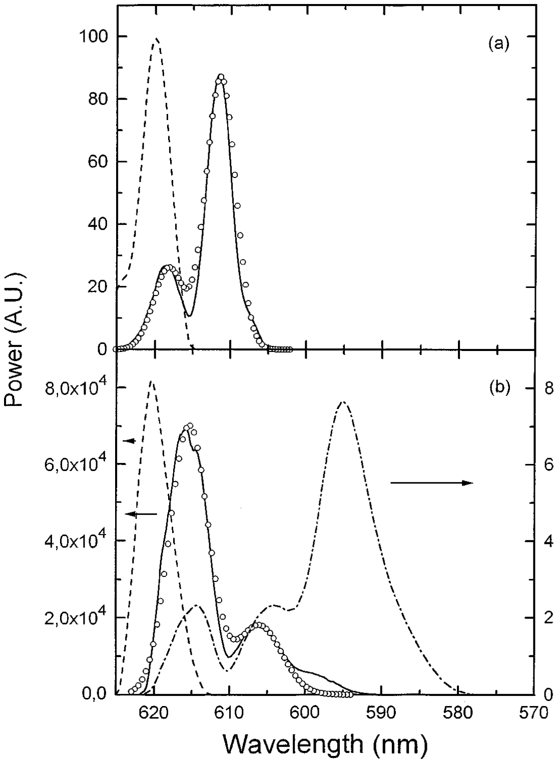

FIG. 3. Copropagating spectra ($\theta_0 = 20°$). (a) Same conditions as Fig. 2(a): (dashed line) original pulse spectrum of the probe beam; (solid line) up-shifted spectrum; (circles) numerical simulation of the up-shifted spectrum with the same parameters of Fig. 2(a), assuming that 23.1% of the photons experience flash ionization. (b) Same condition as Fig. 2(b): (dashed line) original pulse spectrum; (solid line) up-shifted spectrum; (dash-dotted line) filtered up-shifted spectrum; (circles) numerical simulation of the up-shifted spectrum with the same parameters of Fig. 2(b), assuming that 79.5% of the photons experience flash ionization.

the maximum shift. The most plausible explanation for this new spectral shift is flash ionization, because it corresponds to half of the electron plasma density, as measured by Moiré interferometry (see discussion of Fig. 4 below). When we get close to the focus, the front transverse dimension becomes comparable to the probe pulse waist. In this case, the ray-tracing theory is no longer valid and the phase effects become dominant. The incident probe photons no more experience the influence of a well-defined front, but they merely integrate over the entire plasma region which is suddenly being created and experience the well-known flash ionization blueshift [10]. As expected, the factor of about 2 between the two peaks in Fig. 2(b) corresponds to the factor $\beta/(1 + \beta)$ between the counterpropagating blueshift and flash ionization. An alternative explanation for the observed two peaks would be a two step ionization front, but that would not explain the copropagation spectra.

The same kind of behavior is also observed for copropagation. In Fig. 3(a) we can observe a blueshift of 8.6 nm, corresponding to the expected effect in copropagation, as described by Eq. (1). In this case the copropagating relativistic mirror is taking place. The difference between the blueshifts in copropagation and counterpropagation is a clear signature of the occurrence of photon acceleration by a relativistic front. From this we can estimate the value of the front velocity, as discussed below.

We see that nearly 75% of the photons are accelerated, but we can still observe a small peak which could also be explained as due to flash ionization. Of course, in an ideal configuration of a very large and homogeneous front all the probe photons would be equally accelerated. The observed blueshift of the peaks which we associate with flash ionization is equal in copropagation and counterpropagation, for the same plasma conditions, as in Figs. 2(b) and 3(b) ($\Delta\lambda_{\mathrm{flash}} = 4.8$ nm). We interpret this as a clear signature of the flash ionization effect. When we approach the main beam focus, the copropagating mirror effect becomes larger and we can observe blueshifts as high as 25 nm, as shown in Fig. 3(b). This spectrum was observed by filtering the radiation above 600 nm. If we correct the spectrum with the filter calibration curve we notice that the maximum of the blueshifted light is at 14 nm and that in this case of denser and thinner

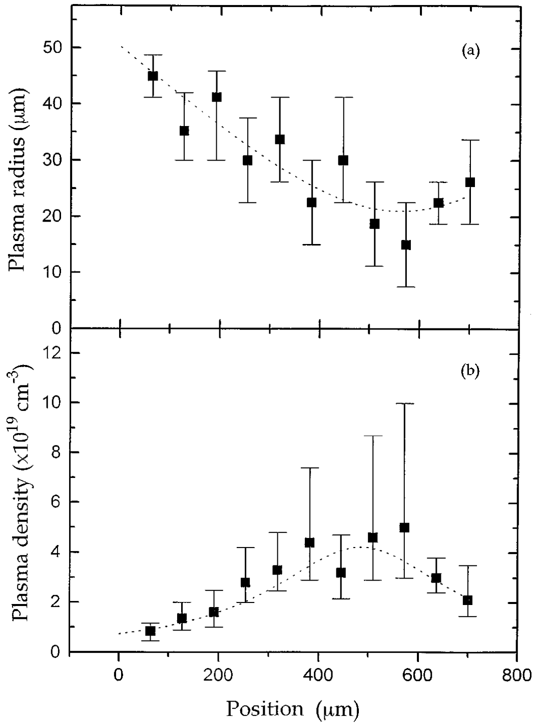

FIG. 4. (a) Plasma radius and (b) electron plasma density, as a function of the position along the laser propagation axis, measured by Moiré interferometry.

fronts the flash ionization effect is dominant, as already observed in counterpropagation.

In Fig. 4, we present the electron plasma density and the transverse width of the ionization front measured by Moiré interferometry. From this experimental data we can estimate the frequency up-shift which we associate with flash ionization for the two cases of Figs. 2 and 3: 4.75 nm near the focus (position $= 500\ \mu$m) and 1.33 nm away from the focus (position $= 100\ \mu$m). These results give some credibility to our conclusions about flash ionization. Furthermore, if we go back to Figs. 2 and 3, we can also derive the electron density and the velocity of the ionization front by comparing the maxima of the copropagating and counterpropagating "accelerated" spectra, and employing Eqs. (1)–(3). Near the focus the calculated electron density is $n_e = 4.32 \times 10^{19}$ cm$^{-3}$ and the velocity of the front is $\beta_f = 0.941$. On the other hand, 400 $\mu$m away from the focus, the electron density is $n_e = 1.65 \times 10^{19}$ cm$^{-3}$ and the front velocity is the same as before. We see that the electron density calculated using this method is in excellent agreement with the Moiré interferometry measurements (Fig. 4). The predicted front velocity lies within the experimental range, but is smaller by nearly 5% than the group velocity of the main laser beam if it propagated in a preformed plasma with the same density. Using the previous results for an infinite ionization front, we have also generated, by ray tracing [5], the full up-shifted spectrum for situations of Figs. 2 and 3, starting from the initial spectrum and assuming that a fraction of the incident photons experiences flash ionization. A very good agreement between the numerically generated spectra and the experimental spectra can then be observed in Figs. 2 and 3. We should state that such an agreement is only possible because the fraction of flash ionized photons was fitted to the experimental data. But this fraction increases from larger to smaller fronts, in agreement with the above qualitative arguments. The spectrum tail in Fig. 3(b), which is not predicted by our numerical simulation, can be due to finite front effects, in particular, to different incidence angles associated with the front curvature.

In conclusion, we have measured for the first time, the frequency up-shift of an ultrashort laser pulse colliding with an ionization front in two opposite directions. This allowed us to clearly demonstrate the occurrence of photon acceleration with relativistic ionization fronts. Efficiencies of frequency shift as high as 75% have been obtained.

With our 2D experimental setup, we have also observed, for the first time, the copropagating relativistic mirror, as predicted by the ray-tracing theory. The identification of the three frequency up-shift regimes (copropagation and counterpropagation, and flash ionization) in the up-shifted spectra allowed us to determine the velocity of the ionization front and its maximum electron density, which are in good agreement with other measurements and with numerical simulations. This clearly points to the feasibility of a photon acceleration diagnostic for relativistic coherent structures in laser produced plasmas.

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