

## Simultaneous Measurements of Hard X Rays and Second-Harmonic Emission in fs Laser-Target Interactions

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The interaction of 150 fs laser pulses with very thin plastic targets at an intensity of  $5 \times 10^{17}$  W/cm<sup>2</sup> was investigated experimentally. Second-harmonic (SH) radiation was found to be emitted only in the specular direction. Both SH intensity and hard x-ray yield were found to be strongly dependent upon the laser polarization. The main features of SH emission are in agreement with a theoretical model which assumes resonance absorption as the source mechanism of electron plasma waves. Measurements suggest that, in conditions of maximum energy absorption, wave breaking of resonantly excited electron plasma waves takes place.

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The interaction of high power femtosecond laser light with matter is now recognized as a promising way of generating short intense x-ray pulses with photon energies extending beyond 1 MeV [1]. Intense subpicosecond terahertz radiation can also be generated in these conditions [2].

Detailed analytical studies and numerical simulations show that collisionless processes including resonance absorption [3], vacuum heating [4], and anomalous skin-layer heating [5] are the key factors in the laser-target coupling. A fundamental role is expected to be played by the polarization and the angle of incidence of the laser light [6]. The effect of polarization has indeed been observed in fs interactions by studying either x-ray or second-harmonic (SH) emission processes. The temporal evolution of SH emission was investigated in a pump-probe experiment [7]. In the case of a *p*-polarized probe, SH clearly showed the effect of resonant enhancement of the electric field at the critical density typical of resonance absorption.

On the other hand, recent experimental investigations [8,9] have established the link between polarization of laser light and x-ray emission. These experiments provide striking evidence of enhanced coupling of *P*-polarized light with the plasma, showing that collisionless absorption processes are of crucial importance in fs interactions. However, it has been pointed out [10] that, on the basis of the experimental results presently available, it is impossible to discriminate between the various collisionless processes named above.

In this Letter we report on the results of a study on both SH and x-ray emission from high intensity fs laser-matter interaction. To our knowledge, these are the first simultaneous measurements of SH and hard x-ray emission as a function of laser polarization. Our measurements clearly show the dramatic dependence of plasma emission properties upon polarization. We strongly believe that

the observed behavior of SH and x-ray emission is a convincing evidence of the key role played by the classical resonance absorption in the interaction processes under investigation.

A Ti:sapphire laser at Laboratoire d'Optique Appliquée operated at a wavelength of 0.8  $\mu$ m and delivered up to 30 mJ on target in a 150 fs FWHM pulse [11]. The linearly polarized beam was focused on an 800 Å thick plastic (Formvar) foil target by using an *f*/4 reflective optics in an off-axis configuration, at an angle of incidence of 20°. The intensity on target was typically  $5 \times 10^{17}$  W/cm<sup>2</sup>. The polarization of the laser light was varied gradually from *S* to *P* by rotating a half-wave plate on the incident beam, before the focusing optics.

The laser pulse was characterized by a nanosecond pedestal originating from amplified spontaneous emission (ASE) that resulted in an intensity on target of approximately  $5 \times 10^9$  W/cm<sup>2</sup>. The effect of this prepulse was investigated experimentally by firing the laser system without injecting the fs pulse in the amplifier chain and focusing the ASE pulse on the target. Our measurements clearly showed that no damage was induced on the target, even when a sequence of 20 ASE pulses was fired on the same position of the target. This result is consistent with previous measurements in the nanosecond regime performed using thin plastic targets ([12] and references therein) that showed a damage threshold intensity for nanosecond pulses of the order of  $10^{10}$  W/cm<sup>2</sup>. Therefore we have experimental evidence that the laser-target interaction was free from preformed plasma.

The second-harmonic radiation was collected by an *f*/5 optics, selected by using narrow band filters and detected by a photodiode. The x-ray radiation emitted by the plasma was studied by using an x-ray charge-coupled device (CCD) array set to work in a single photon detection mode. In fact, the x-ray flux incident on the device was low enough to ensure that the average number of x-ray

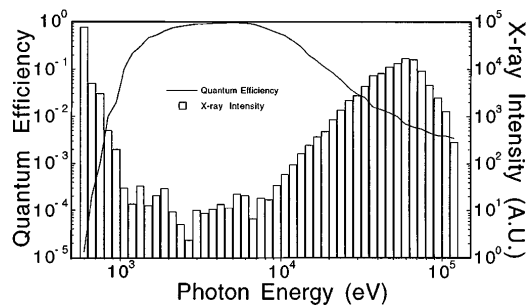


FIG. 1. A typical spectrum of the x-ray emission obtained by the x-ray CCD in single photon detection operation, in conditions of mixed laser polarization ( $S/P$ ). The spectrum was corrected for the quantum efficiency of the detector, also plotted in the same graph.

photons per pixel was much smaller than 1, so that the charge induced in each pixel could be related to the energy of the incident photon.

The spectral distribution of the x-ray radiation could be immediately obtained from the charge distribution function, provided that charge diffusion to neighboring pixels was taken into account. The CCD detector was filtered by a  $25\ \mu\text{m}$  Be foil in order to cut off photons at energies lower than 1 keV. A careful calibration of the detector in the 1–10 keV spectral range was carried out and a detailed description of this study can be found elsewhere [13]. The main properties of the detector including energy response linearity, spectral resolution, and quantum efficiency were found to be in good agreement with the predictions of numerical models based on the x-ray absorption properties of the detector elements. The behavior of the detector in the high energy range ( $h\nu > 10\ \text{keV}$ ) was therefore calculated taking into account the Si mass absorption coefficient and the depth of the active region in which x-ray photons are absorbed with consequent production of photoelectrons [13]. The overall detector quantum efficiency as a function of incident photon energy is presented in Fig. 1.

A typical spectrum of the x-ray emission, corrected for the quantum efficiency of the detector, is shown by the histogram of Fig. 1 where the number of photons

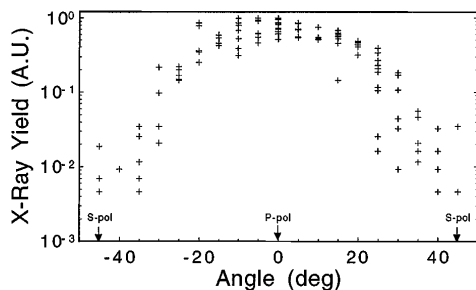


FIG. 2. Dependence of the x-ray energy detected by the CCD detector as a function of the polarization of the incident laser light. The polarization was varied continuously from  $S$  to  $P$  and back to  $S$  again by means of a half-wave plate placed on the incident beam. The zero angle of the wave plate corresponds to  $P$ -polarized incident laser light while  $\pm 45^\circ$  correspond to  $S$ -polarized laser light.

detected per energy interval is plotted as function of photon energy. This spectrum was recorded with the incident laser light set in a mixed  $S/P$  polarization, that is, with the half-wave plate rotated by  $20^\circ$  with respect to the  $P$ -polarization configuration.

The behavior of the total x-ray energy as a function of the angle of the half-wave plate is shown in Fig. 2, where the zero angle corresponds to the condition of  $P$  polarization of the incident laser light. Each data point was obtained by integrating the charge induced by x rays on the CCD array over the whole active area of the detector. Provided that the spectrum of the x-ray radiation does not change, this value is proportional to the total energy of the x-ray photons detected by the CCD array. We observe that our detector enabled us to measure simultaneously the spectrum and the intensity of the x-ray radiation. Therefore, spectral features were accounted for in the evaluation of the total x-ray energy when unfolding the data by using the quantum efficiency reported in Fig. 1.

According to these results, the hard x-ray emission observed in our experiment is characterized by a strong spectral component centered at photon energies of  $\approx 60\ \text{keV}$ , while the contribution of lower energy photons to the total x-ray spectrum in the observed spectral window increases rapidly as the energy decreases. In fact, most of the *thermal* x-ray emission arising from the plasma is expected to peak at several hundred eV. Moreover, the plot of Fig. 2 shows that the total x-ray yield is found to be maximum when the laser light is  $P$  polarized, its value being approximately 2 orders of magnitude higher than in the case of  $S$ -polarized light.

On the other hand, SH emission also exhibits a well defined behavior as shown in the plot of Fig. 3 where the intensity of the SH light is reported versus the angle of the half-wave plate, as in the case of Fig. 2. The main features of SH emission can be outlined as follows: (i) SH emission was found only in the specular direction; (ii) its intensity was found to be strongly dependent upon the polarization of the laser light; (iii) regardless of the laser polarization, SH emission was found to be always predominantly  $P$  polarized, though with different degrees of polarization; (iv) SH intensity shows an absolute

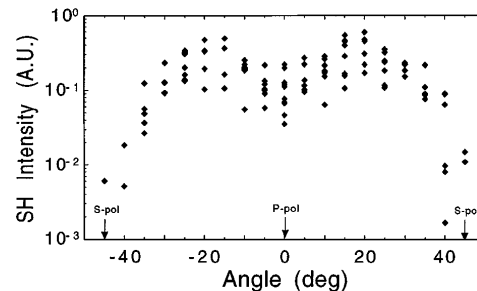


FIG. 3. Specularly reflected second-harmonic radiation as a function of polarization of the incident laser light. The polarization was varied by means of a half-wave plate placed on the incident beam as in the case of Fig. 2.

minimum in condition of  $S$  polarization and increases with increasing  $P$  component of the laser light; (v) at close-to- $P$ -polarization conditions, however, SH emission suffers a depletion and shows a relative minimum for exact  $P$  polarization.

The polarization of SH light was analyzed for the two cases of  $S$ - and  $P$ -polarized laser lights. A weak  $S$  component of SH light was always present and its intensity was found to be basically independent from the polarization of the laser light. In the case of  $P$ -polarized laser light, the ratio between the  $P$  and the  $S$  components of SH was approximately 100, while in the case of  $S$ -polarized laser light such a ratio was approximately 2.

The experimental results presented here clearly show that laser polarization is a key factor in the interaction regime under investigation. A strong enhancement in the production of SH radiation and hard x rays is observed in the case of  $P$ -polarized laser light, indicative of a more efficient laser-plasma coupling. In addition, we note that the total energy emitted in the hard x-ray region, from 1 to  $\approx 100$  keV, in the case of  $P$  polarization, is of the order of  $10^{-3}$  of the total laser energy. Indeed, according to Fig. 1, most of this x-ray energy is concentrated in a narrower spectral region around 60 keV. Therefore, we believe that the energy coupling mechanism responsible for such an intense hard x-ray emission process is of primary importance in the dynamics of the interaction process under investigation.

As already pointed out above, SH radiation was found to be emitted only in the specular direction. This is a distinctive feature of SH emission generated by electron plasma waves produced by a linear conversion mechanism. In fact, it is well known [7] that electron plasma waves arising from nonlinear processes, i.e., parametric decay instability, would lead to a distribution of directions of the SH emission which was not observed experimentally. In addition, the occurrence of a nonlinear mechanism in the generation of electron plasma waves would also imply the existence of a threshold value of the incident laser intensity for the production of SH radiation. Indeed, we found no experimental evidence for the existence of such a thresholdlike behavior in the SH emission process.

According to the model described below, energy absorption is mostly accounted for by resonance absorption. Electron plasma waves generated by the resonance process account for SH emission as well as for the production of very energetic electrons from which hard x rays originate.

It can be shown that by combining Maxwell equations, the continuity equation, and the plasma equation of motion and applying a perturbative method, the SH current density is related to the local electric field by the following expression:

$$\vec{j}^{(2)}(2\omega) = \frac{in^{(0)}e^3}{4m_e^2\omega^3} \vec{\nabla}(\vec{E} \cdot \vec{E}) + \frac{ie^3}{m_e^2\omega^3} \left( \frac{\vec{\nabla}n^{(0)} \cdot \vec{E}}{1 - \omega_p^2/\omega^2} \right) \vec{E}, \quad (1)$$

where  $\vec{E}$  is the local electric field oscillating at the laser frequency,  $n^{(0)}$  is the unperturbed electron density,  $\vec{\nabla}n^{(0)}$  is the longitudinal electron density gradient,  $\omega$  is the laser angular frequency, and  $\omega_p$  is the plasma angular frequency. According to this model, and following simple considerations [7] on the conservation of the component of the wave vector parallel to the target, SH radiation is emitted in the form of a coherent beam collinear with the specularly reflected fundamental beam.

According to Eq. (1) there are two main source terms which give rise to a current density oscillating at the SH frequency. The first term  $A$  originates from the gradient of the intensity of the electric field. The second term  $B$  gives a component of the SH current parallel to the local electric field and proportional to  $\vec{\nabla}n \cdot \vec{E}$ . The intensity ratio between the terms  $A$  and  $B$  of the SH current given by Eq. (1) can be simply written as

$$\frac{A}{B} \approx \frac{\varepsilon}{4 \cos \alpha}, \quad (2)$$

where  $\varepsilon = 1 - \omega_p^2/\omega^2$  is the plasma dielectric constant and  $\alpha$  is the angle between the density gradient and the electric field. Since in our experimental conditions SH is generated in the proximity of the critical density region where the dielectric function  $\varepsilon \rightarrow 0$ , provided that  $\alpha \neq \pi/2$ , that is, provided that laser radiation is not  $S$  polarized, the ratio given by Eq. (2) also vanishes, indicating that the term  $B$  is expected to give a dominant contribution to SH emission. Therefore, when the laser is  $P$  polarized, strong enhancement of the local electric field occurs due to resonance absorption and the term  $B$  of Eq. (1) accounts for intense  $P$ -polarized SH emission. The small fraction, approximately 1%, of  $S$ -polarized SH radiation observed experimentally can be easily explained in terms of contribution to SH emission given by the first term of Eq. (1) as a consequence of transverse gradients of the electric field. Such gradients are expected in the interaction region due either to the finite spot size or to intensity nonuniformities in the focal spot.

In the case of  $S$ -polarized laser light, the electric field is no longer enhanced by the resonance process and, in agreement with experimental observations,  $P$ -polarized SH emission is no longer largely dominant over the  $S$ -polarized one. As in the previous case,  $S$ -polarized SH emission is again accounted for by the first term of Eq. (1) due to transverse gradients of the local electric field while  $P$ -polarized SH emission occurs as a consequence of the residual  $P$ -polarized laser light due to the spreading in the angle of incidence introduced by the focusing optics.

The validity of the model presented here is further corroborated by the following observation concerning the behavior of SH emission for close-to- $P$  laser polarization discussed above. According to the model, SH intensity and x-ray emission is expected to peak in the condition of maximum coupling between laser radiation and electron

plasma wave. Indeed, according to Fig. 2, x-ray emission is found to be maximum when the laser light is  $P$  polarized. In contrast, Fig. 3 shows a strong depletion of SH emission for close-to- $P$  polarization conditions. This effect suggests that the process from which such an emission originates is partly inhibited in this configuration. However, we observe that SH generation has been modeled here by using perturbation theory which requires that the charge density fluctuation induced by the resonant process must be small compared to the critical density. The ratio of the induced charge density fluctuation to the critical density can be estimated by using the following expression [14]:

$$\frac{\delta n}{n_c} = \frac{x_{\text{osc}}}{L}, \quad (3)$$

where  $x_{\text{osc}}$  is the oscillation amplitude of electrons in the laser field and  $L$  is the electron density scale length. For the experimental conditions outlined above the resonantly induced density fluctuation is expected to be of the order of the critical density. Therefore, the conditions for the perturbation theory to be valid can be violated, especially early during the laser pulse, before a sizable plasma has developed.

Following these considerations, another mechanism must be considered in the analysis presented so far, which can strongly influence the evolution of plasma waves and, consequently, the generation of second-harmonic radiation. It is well known that, for sufficiently high laser intensity, electron plasma waves produced by resonant absorption are subject to wave breaking [14]. In fact, if the electric field  $E$  associated with the electron waves is very high, Landau damping is no longer applicable and the wave damping process becomes nonlinear. Background electrons, regardless of their initial velocity, are rapidly accelerated at velocities comparable to the phase velocity of the electron wave in a fraction of the period of the wave oscillation. The wave is therefore heavily damped, its energy being efficiently converted into very energetic electrons which escape from the interaction region.

One can easily verify that for the experimental conditions considered here, assuming an electron temperature in the close-to critical density plasma region of 200 eV and a density scale length  $\approx 250 \text{ \AA}$ , the wave breaking condition is satisfied in the interaction region and strong nonlinear damping of resonantly excited plasma waves can occur. From the point of view of the interpretation of the experimental results, this process can explain the observed behavior of SH and x-ray emission shown in Figs. 2 and 3. According to Eq. (1), as the polarization of the incident laser light varies from  $S$  to  $P$ , the component of the driving electric field along the density gradient increases. Consequently, the amplitude of the resonantly excited wave also increases, resulting in stronger SH emission, until the amplitude of the driving field becomes so high that the wave breaking condition is reached.

As a consequence of the collapse of the electron wave, the source current described by Eq. (1), from which SH radiation originates, is disrupted. A minimum of the SH emission intensity is therefore expected in conditions of maximum resonance, that is, for  $P$ -polarized laser light. On the other hand, since the energy lost by the wave is efficiently converted into electron kinetic energy, bremsstrahlung x-ray emission, originating from these hot electrons, is expected to reach its maximum in this condition. This result is also in agreement with the plot of Fig. 2 which shows that the maximum x-ray yield is indeed reached in conditions of resonance.

In summary, the interaction of intense femtosecond laser light with a highly confined plasma has been studied. SH radiation was emitted only in the specular direction. The intensity of SH and hard x-ray emissions was found to be strongly dependent upon the polarization of the incident light. On the basis of the overall agreement between our experimental observations and the model presented here, we conclude that, among the various mechanisms that can occur under the conditions of the present experiment, resonant absorption plays a key role in the laser-target coupling process. The behavior of SH emission at maximum resonance conditions strongly suggests that wave breaking of the resonantly produced electron plasma waves occurs in the interaction regime under investigation.

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