

Pulsation of harmonic and $K\alpha$ emission from laser-produced plasmas

B. Luther-Davies and A. V. Rode

*Laser Physics Centre, Research School of Physical Sciences and Engineering, The Australian National University,
P.O. Box 4, Canberra, Australian Capital Territory 2601, Australia*

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We report simultaneous time-resolved measurements of second-harmonic and $K\alpha$ emission from layered, planar targets irradiated by 180 ps, Nd-laser pulses focused to an intensity of $(1-3)\times 10^{16}$ W/cm². We observe that pulsations of the second-harmonic emission appear to coincide with pulsations of the x-ray fluorescence with each burst lasting about 20 ps and separated by 30–50 ps. The x-ray pulsations indicate that fast electrons are produced in short bursts which can modulate the effective “temperature” in the quarter critical region thereby affecting emission from the two-plasmon decay instability. We suggest these pulsations appear because the laser intensity reaching the critical surface is modulated by a chaotic time-dependent Brillouin reflection from the underdense plasma. Some implications of such a process are discussed.

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I. INTRODUCTION

Pulsations of ω_0 , $2\omega_0$, and $\frac{3}{2}\omega_0$, and other harmonics emitted from laser-produced plasmas created by an intense ($I\lambda^2 \approx 10^{15} - 10^{17}$ W/cm²), short-pulse (20–200 ps), laser radiation incident on solid targets has been studied both experimentally [1–6] and theoretically [7–11] for some time. These emissions generally appear in randomly occurring bursts typically lasting 10–30 ps and separated by 20–50 ps. In our own work [1,2,7,12] we have found that the temporal bursts are always accompanied by the appearance of fine, random modulations in the corresponding time-integrated spectra. We pointed out that these particular spectral and temporal features alter only slightly with laser parameters (intensities between 10^{14} W/cm² and 10^{17} W/cm², pulse durations 30–1000 ps, and wavelengths from 0.35 to 10.6 μm) and target materials (from CH to ²³⁸U).

We argued in [7,12] that this insensitivity to the experimental parameters ruled out many of the mechanisms that had been previously suggested would produce pulsations. We further concluded that since the time-resolved and time-integrated data are related via Fourier transformation, the spectral and temporal features were only compatible if the emission swept in frequency during each burst. Such frequency sweeping was in fact measured for second-harmonic emission [2]. To explain the frequency sweeping we developed a model where the plasma contained two reflecting regions for the incoming light: one the critical surface and the other a Brillouin zone in the underdense plasma. The reflectivity of this latter region varied cyclically in time due to the influence of plasma expansion on the three-wave interaction detuned by a nonlinear phase mismatch caused by ion trapping on the intense sound waves generated by the Brillouin instability itself.

The complete description of the process is given in [1,7]. A direct consequence of this model is that the intensity of a laser light reaching the critical density sur-

face would be modulated in time since when the Brillouin reflectivity is high, little laser radiation penetrates to the critical surface, whilst when it is low, the intensity at critical approximates the vacuum intensity. Since many important plasma physics processes (resonance absorption, the parametric decay instability, etc.) are localized at the critical surface, pulsation of the laser intensity at the surface would be expected to become imprinted on those processes. Thus, for example, second-harmonic generation (SHG) at the critical surface should exhibit similar pulsations to those observed in the reflected laser light, as is indeed observed.

The consequences of this model are far reaching and could well affect the interpretation of much previous experimental data on plasma created using short-pulse intense laser light. For example, it would lead to the plasma absorption varying in time, especially since the strongest absorption generally occurs at densities close to critical. Our own measurements of absorption of short (20–150 ps) Nd laser pulses at intensities above 10^{14} W/cm² showed that the time-averaged absorbed fraction was typically 40% [13] and similar values have been reported from several other studies [14–16]. But this number was measured using calorimeters to determine the fraction of laser light not absorbed by the plasma. If, as is the case, the overall plasma reflectivity pulsates in time, at some times (those we would suspect as corresponding to the times the light penetrated to the critical density) the absorption must be much greater and at other times much less than this time-averaged value. (Note that the experiments have shown that the reflection pulsates synchronously over the whole plasma.)

This scenario leads one to examine other conclusions regarding absorption. For example, several experiments have shown that the absorption coefficient for short Nd laser pulses at high intensity is rather insensitive to laser intensity and has a value around 40% [13–16]. This result now has two possible interpretations: (1) that the absorption mechanism yields an intensity-dependent ab-

sorption of $\approx 40\%$, or (2) that the laser light only penetrates to a region of $\approx 100\%$ absorption for 40% of the time. Since the plasma reflectivity pulsates, it is this latter which is the more likely, but seldom discussed, scenario. Obviously, if the absorption instantaneously reached a value near 100%, it would be necessary to examine which absorption processes could lead to such high absorption. Resonance absorption, for example, which is often invoked as dominating in plasmas produced by picosecond duration pulses, provides a maximum of 50% absorption.

Other phenomena, such as fast-electron generation by resonance absorption or the parametric decay instability, would be also affected by modulation of the laser intensity at the critical surface. Thus fast electrons would be generated in bursts synchronized with other critical surface phenomena such as second-harmonic generation. Observation of fast-electron pulsations, therefore, would certainly lend support to the "two-reflection-point" model, especially if they correlated with pulsations of second-harmonic emission.

An additional motivation, however, underpinned the experiments reported here. We had also observed that $\frac{3}{2}\omega_0$ emission, associated with the two-plasmon decay instability, displayed the same complex temporal and spectral modulations found in the reflected laser light and second-harmonic emission. Hence we concluded that the $\frac{3}{2}\omega_0$ light had to be phase modulated just like the ω_0 and $2\omega_0$ emission. These observations have partly been reported elsewhere [4,17]. We include here examples of the $\frac{3}{2}\omega_0$ behavior. Figure 1(a) shows a typical time-resolved spectrum recorded at an intensity of 5×10^{15} W/cm². Notice that the separation of the usual red and blue peaks varies between bursts. Since it is normal to interpret this separation as reflecting the local plasma temperature (at the quarter critical density), the data suggests that this temperature varies from burst to burst within the range

1–6 keV [Fig. 1(b)], although it should be noted that the spectra are often quite broad introducing a large uncertainty in the temperature so determined. Nevertheless these values far exceeded the thermal temperature of the plasma as determined from x-ray measurements (≈ 0.5 – 0.8 keV) although this discrepancy should not be surprising since thermal x rays are generally produced from densities much higher than the quarter critical density leading to the possibility of large errors if thermal values are used to deduce the quarter critical temperature. One can, therefore, postulate that the effective temperature of the quarter critical region is increased by a flux of superthermal electrons, probably generated at the critical surface and ejected down the density gradient. The "effective" temperature T_{eff} is hence related to the average temperature of the thermal and superthermal groups weighted by their densities. Thus a small density of very energetic electrons can increase this effective temperature to several times the thermal value provided the superthermal electron energy is high enough. We reported earlier measurements of the total number and energy of the fast electrons generated in our plasmas and found that the superthermal electrons predominantly had energies in the 50–150-keV region and compromised some 3–5% of the total absorbed laser energy. These earlier measurements, therefore, provided firm evidence of the presence of superthermal electrons in the corona of the plasmas.

Figure 2 shows a typical time-integrated spectrum corresponding to time-resolved data similar to that shown in Fig. 1. Notable are the strong spectral modulations which when Fourier analyzed contain no regular structures but can be characterized as "pink" noise (see the inset).

As argued in detail in our previous work, fine spectral modulations over a wide spectral bandwidth are only consistent with the appearance of emission in short tem-

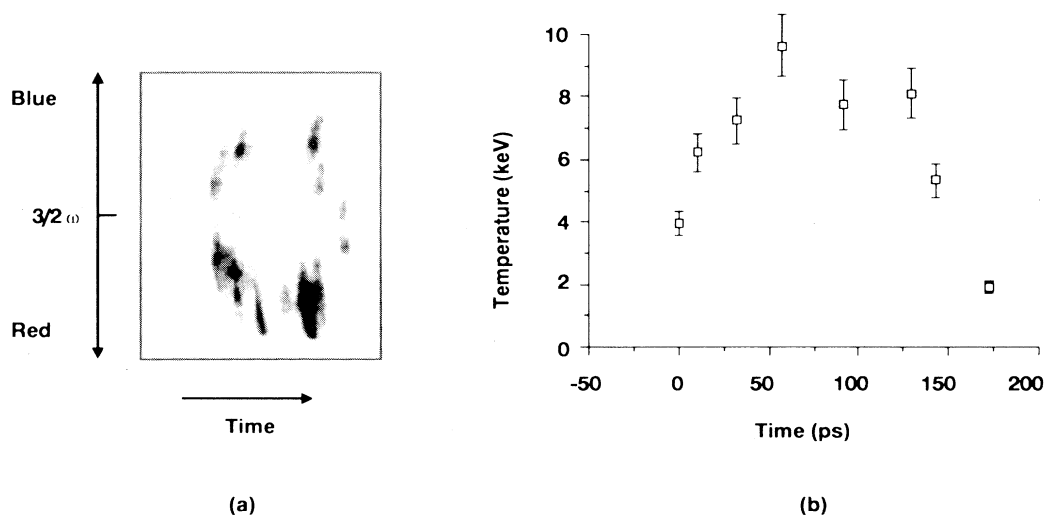


FIG. 1. Time-resolved spectrum of $\frac{3}{2}\omega_0$ emission (a) and the effective plasma temperature in the quarter critical region deduced from the separation of the red and blue spectral peaks in each burst (b).

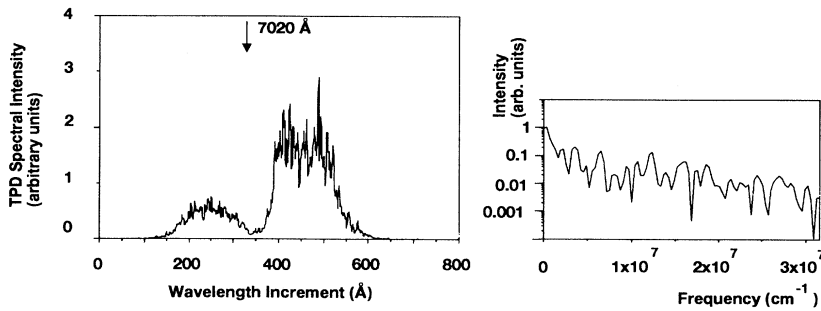


FIG. 2. Time-integrated $\frac{3}{2}\omega_0$ spectrum obtained at an intensity of 7×10^{16} W/cm² showing random spectral modulations (a); Fourier spectrum of these modulations (b).

poral bursts when the emission itself is phase modulated, i.e., the instantaneous emission frequency must sweep rapidly during each burst. Careful analysis of data such as that in Fig. 1 supports this view although not unequivocally because the bursts last only about twice the spectrograph response time.

The association of the separation of the red and blue peaks with the local plasma temperature indicates that this temperature must change rapidly. The two-plasmon decay instability has a sufficiently high growth rate for it to be able to follow temperature changes on the picosecond time scale. The most likely cause of rapid temperature modulation would be the appearance of bursts of fast electrons flooding into the quarter critical region.

The experiments reported here were designed to detect fast-electron pulsations both to support our previous model where pulsations ω_0 and $2\omega_0$ emission were a consequence of the two-reflection-point model, but also to provide a plausible explanation as to why the $\frac{3}{2}\omega_0$ emission frequency could sweep on the picosecond time scale.

II. EXPERIMENTS

Previous experiments on time-integrated $K\alpha$ emission [18] indicated that large numbers of fast electrons with energies in the range 50–200 keV are produced by irradiating planar solid targets with high intensity ($>10^{15}$ W/cm²), short (<100 ps) pulses of Nd-laser radiation. These electrons have sufficient energy to penetrate quite thick (>10 μm) low- Z transport layers and excite fluorescence in a high- Z fluorophore on the rear of the target. We, therefore, planned to search for fast-electron pulsations by streaking the intensity of the $K\alpha$ emission produced from fluorophores on the back of a layered, laser-irradiated, planar target (Fig. 3). Simultaneously, $2\omega_0$ emission was recorded by a second streak camera viewing from the front. Comparison of these images allowed us to search for a correlation which could indicate that both were being similarly affected by modulation of the pump (laser) intensity at the critical surface.

The experimental details were as follows. The targets were irradiated close to normal incidence by 7–9-J pulses of 180-ps duration using a single beam from a Nd:glass laser. The beam was focused using an $F=1$ aspheric lens to a spot 10–15- μm diameter on the target surface. In fact the target was positioned behind the position of the best focus of the $F=1$ lens since previous experiments

had shown this was close to the optimum for production of fast electrons and second-harmonic emission via resonance absorption [12,13]. The on-target laser flux density was $(1-3) \times 10^{16}$ W/cm².

The layered targets were made of 9- μm -thick aluminum and ≈ 5 - μm -thick carbon (in the form of an epoxy adhesive) as electron-transport layers and an iron layer as a fluorophore. We have shown that the energy spectrum of superthermal electrons which are responsible for $K\alpha$ production in layered targets irradiated by a short high-intensity laser pulse is well approximated by an electron beam with energy within the range 50–200 keV [18]. The choice of the atomic number and thickness of the transport layer was made taking this into account and using a simple analytic description [19] of energy transport and deposition in multilayer targets. The penetration depth Δ_{tr} for electrons with an energy E_e can be evaluated as $\Delta_{tr} \approx 2 \times 10^{-7} E_e$ for carbon, and $\Delta_{tr} \approx 1.5 \times 10^{-7} E_e$ for aluminum; here Δ_{tr} is in cm, E_e in keV. Therefore, the 9- μm -thick Al layer together with a 5- μm C layer filtered out those electrons with $E_e < 50$ keV.

Iron was chosen as the fluorophore since, firstly, K -shell ionization by Bremsstrahlung radiation from the plasma would not occur because its K edge is at an ener-

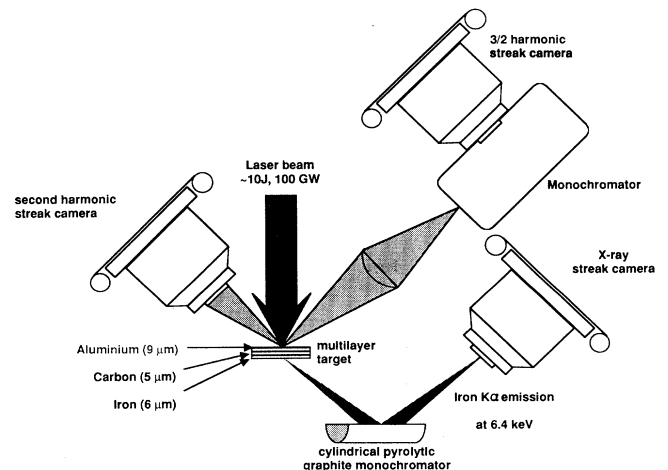


FIG. 3. Schematic of the apparatus used to measure hot-electron pulsations.

gy far above the thermal plasma temperature [13,20], and secondly, the $K\alpha$ line radiation was at an energy that could be conveniently collected by an available x-ray analyzer crystal and directed to the input slit of a Hadland Photonics X-Chron streak camera. The plasma thermal-electron temperature was estimated by recording the Al x-ray line spectrum and measuring the relative intensity of the resonance line Al XII and its j, k satellites [21]. The thermal temperature so determined was approximately 0.8 keV which is well below the Fe $K\alpha$ line radiation at 6.4 keV. The Fe-layer thickness, on one hand, had to be large enough to absorb the superthermal electrons with energies $E_e \leq 200$ keV, whilst being reasonably transparent to its own $K\alpha$ fluorescence. The optimum thickness was found using the electron-transport model [19] from which the penetration depth for iron is $\Delta_{tr} \approx 4 \times 10^{-8} E_e$ and Fe x-ray-absorption data [21]. An optimum thickness of 6 μm was deduced, taking into account the angle at which the monochromator crystal viewed the target ($\approx 70^\circ$ from normal).

A cylindrically curved pyrolytic graphite crystal with 35-mm radius was used in the Von Hamos geometry with x-ray focusing in the direction perpendicular to the dispersion direction (Fig. 3) to collect and focus the $K\alpha$ emission onto the streak camera. Graphite has, probably, the best [22] integrated reflectivity (2×10^{-3} rad for 2 \AA) in the spectral range below 3 \AA and good spectral resolution, $\lambda/\Delta\lambda \approx 10^3$, when used in the Von Hamos geometry. The width of the plasma $K\alpha$ image recorded on x-ray film was ≈ 1 mm which approximates the source size determined in previous two-dimensional measurements using a penumbral imaging camera [23]. The temporal behavior of $K\alpha$ emission was measured using an Hadland Photonics X-Chron x-ray streak camera fitted with a cesium-iodide photocathode. The temporal resolution of the system at 6.5 keV was not better than 15 ps. The expected period of the $K\alpha$ pulsations (based on the measurements of second-harmonic pulsations) was

about 10–30 ps; consequently, the relatively poor temporal resolution could have prevented very rapid pulsations, such as those often observed in second-harmonic emission at high intensities, being resolved [2]. However, sufficient numbers of $2\omega_0$ streak images had also been recorded where the bursts were separated by an easily detected 30–50 psec to make the experiment meaningful.

The $2\omega_0$ emission was collected by a $F=5$ lens viewing the target at 45° to the normal, and focused on a scattering screen located ~ 5 cm in front of the entrance slit of an S-20 Imacon 675 streak camera. The time resolution of this system was about 5 ps.

III. RESULTS

Data from several hundred laser shots were recorded. The major difficulty proved to be that high laser intensities were required to get an adequate signal level at the x-ray streak camera but in such conditions the second-harmonic pulsations were too short and occurred too rapidly for synchronized $K\alpha$ emission pulses to be resolved by the x-ray streak camera. By working with the minimum possible laser intensity (consistent with a good signal-to-noise ratio at the x-ray streak camera) it proved possible to obtain a situation where the second-harmonic split into bursts separated by more than 20 ps and in these conditions similar pulsations were clearly observed in the x-ray streak records (Fig. 4). When this occurred it also became apparent that the time history of both emissions was very similar. Unfortunately it was not possible to provide an absolute timing cross reference between the cameras with the necessary picosecond accuracy to prove definitively that the x-ray and second-harmonic pulsations were synchronized. However, the similarity between the records for those shots where structure could be clearly resolved makes us confident that this was, in fact, the case. Examination of a typical pair of records shown in Fig. 4 illustrates this. Both second harmonic

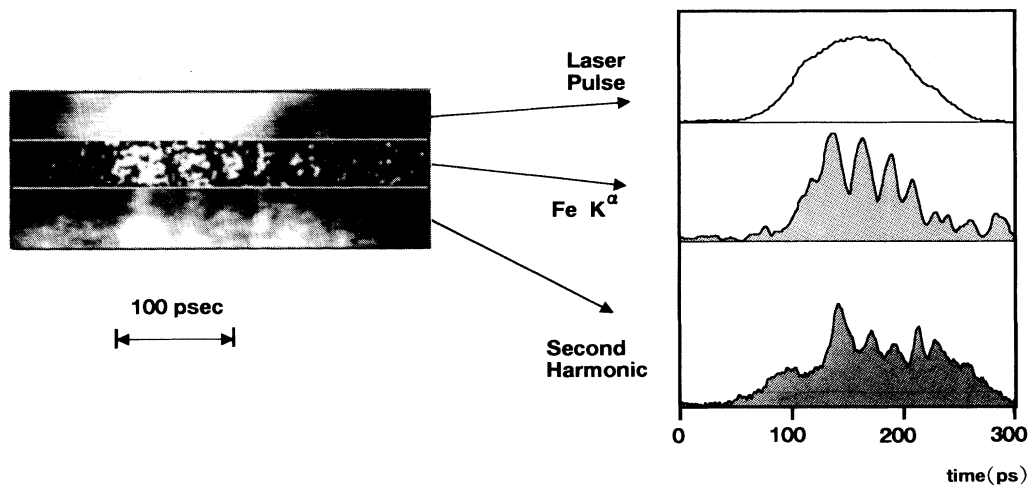


FIG. 4. Temporal behavior of the laser pulse, the x-ray fluorescence from the target, and the second-harmonic emission for the same laser pulse.

and x-ray signals last about the duration of the laser pulse (180 ps); both $K\alpha$ and $2\omega_0$ emissions occur in bursts about 20-ps long and separated by 25–30 ps; there appears a direct correspondence between the individual $K\alpha$ spikes and those in the second-harmonic emission.

IV. DISCUSSION

We have obtained evidence that x-ray fluorescence from layered targets pulsates on the 20–30 ps time scale. This implies that the fast electrons that create the fluorescence are also generated in short bursts. The bursts appear to correlate with bursts of second-harmonic emission as would be expected if both were responding to temporal modulation of the laser intensity at the critical surface. It is thus likely that bursts of fast electrons modulate the temperature at the quarter critical surface affecting the spectrum of the $\frac{3}{2}\omega_0$ emission. The data support the two-reflection-point model which predicts that the intensity of the laser light at the critical surface is modulated by pulsation of the reflectivity of the Brillouin zone. Before concluding we would like to make some further observations on the nature of the pulsations and elaborate the mechanism which we believe produces them.

Using the two-reflection-point model as the starting point for this discussion, it is worth examining the details of that model and how they affect the character of the pulsations. The basic linearized set of differential equations describing a three-wave parametric interaction has stable solutions for arbitrarily large pump powers. Hence calculation of the Brillouin reflectivity using those equations will yield a stable, but intensity-dependent solution. In [7] we pointed out, however, that as the amplitude of the ion sound wave grows, there are at least two processes (ion trapping and second-harmonic generation) that would give rise to the growth of a nonlinear phase

mismatch Δk_{nl} , which detunes the three-wave interaction from perfect phase matching (Δk_{nl} is proportional to the ion sound-wave amplitude). If one assumes a steady situation where the sound-wave amplitude is fixed, but allows the interaction length L (the size of the Brillouin zone) to increase in time, simulating plasma expansion, the Brillouin reflectivity varies cyclically as a function of $\Delta k_{nl}L$. The physics here is quite transparent and simply reflects the sensitivity of any parametric process to imperfect phase matching. Regular pulsations would be expected as L increases accompanied by movement of the reflection point for the laser light moves backwards and forwards between the Brillouin zone and the critical surface. This can be regarded as the zero-level approximation to the physics of the pulsations in these plasmas.

An important question remains, however; namely, is this model sufficient to explain all the details of the observations? Its major deficiency lies in the fact that it predicts regular pulsations rather than the randomness observed experimentally. To illustrate the randomness we have analyzed data on the second-harmonic pulsations from plasmas created by 400-ps duration pulses at an intensity $\approx 10^{16}$ W/cm² incident upon a planar glass target. A typical time streak record is shown in Fig. 5(a), whilst in 5(b) we present the corresponding Fourier power spectrum which shows that no dominant periodicity is present in the modulations. It is perhaps easy to gloss over this feature by imagining that the plasma expansion may not be linear with time, etc., but in that case the more realistic explanation may be missed. Furthermore, this simple model assumes that as the plasma expands the amplitude of the sound wave is constant, whereas in reality it will also vary affecting Δk_{nl} and introducing an additional nonlinear feedback to the system.

It is well established that such nonlinear feedback alters the fundamental nature of the solutions of the coupled wave equations and specifically permits a transition

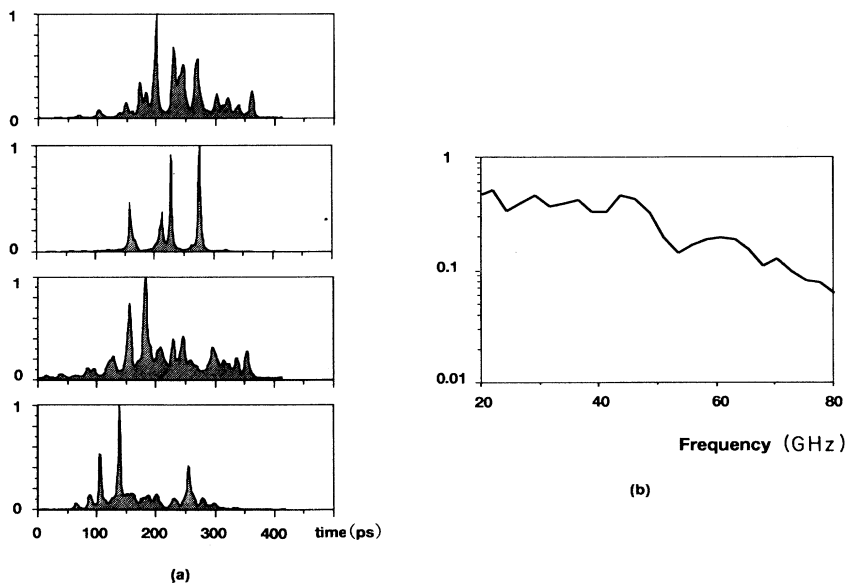


FIG. 5. Typical examples of second-harmonic pulsations (a) and the Fourier power spectrum of these pulsations (b).

to the stochastic (chaotic) regime. Kurin [10] discussed this possibility for stimulated Brillouin scattering (SBS) when second-harmonic generation on the ion sound waves provided the nonlinear feedback. Ion trapping would be expected to behave similarly. Most relevant to the situation described here, however, is the work of Randall and Albritton [11] who discussed the transition to chaos of the SBS instability in the presence of a reflection of the pump from the critical surface of the plasma. In this case the necessary nonlinear feedback was provided by pump-wave reflection rather than the "saturation" mechanisms mentioned above. More recently work by Gaeta, Sceldon, and Boyd [24] and Narum *et al.* [25] discussed the influence of counterpropagating pump beams on the transition of the SBS instability to chaos for a "simple" Brillouin-active medium. They found that even a low-intensity counterpropagating pump could induce a transition into the stochastic regime and in many circumstances this occurred at intensities considerably below those required for the normal SBS instability.

In other words, in a laser-produced plasma even when the laser intensity is below the normal SBS threshold, it is possible for nonlinear feedback of the pump to induce chaotic Brillouin scattering. In the absence of pump feedback, ion trapping, or second-harmonic generation on the ion sound waves can do the same thing. The more complex processes nevertheless produce very similar net results in terms of laser penetration into the plasma as does the simple two-reflection-point model. However, we are now dealing with a better, perhaps first-order, approximation to "reality." Obviously successively higher-order approximations can be envisaged, involving several nonlinear feedback processes acting simultaneously, including damping of the interacting waves and absorption for the light waves followed by a host of other linear and nonlinear plasma phenomena. It is doubtful that these higher-order approximations would add much to the understanding of the physics beyond the scenario described by Randall and Albritton [11] where under the influence of pump feedback caused by a reflection of some of the laser light from the critical density surface, Brillouin scattering in the underdense plasma undergoes a transition into the chaotic regime, resulting in random Brillouin reflectivity pulsations from these plasmas. In macroscopic terms it is useful to retain the two-reflection-point concept since then the origin of the phase modulations which lead to the fine structure in the time-integrated spectra becomes apparent. Furthermore, the two-reflection-point model predicts explicitly that the laser intensity at the critical surface will also be varying rapidly in time.

As a final point we illustrate some of the consequences of this model in interpreting data on other phenomena. The absorption mechanism for the laser light is of major importance in any study of laser plasma interaction physics. In the Introduction we outlined the way in which pulsation of the plasma reflectivity must alter thinking regarding the instantaneous absorption rate of the plasma. Clearly an average absorption of 40% implies the maximum absorption was close to 100% at times when the radiation penetrated to the critical density surface. Al-

though the maximum and minimum Brillouin reflectivity could not be deduced from the experiments, the general character of the pulsations, namely that the *total* plasma reflection is high for about 50% of the time and the maximum to minimum reflectivity is large (> 2), is only consistent with a time-averaged absorption of about 40% if the local absorption near the critical surface lies between 70% and 100% and at the same time the maximum SBS reflectivity is greater than 70%. Thus, resonance absorption can no longer be the dominant absorption mechanism, as stated in some of the earlier research [15,16], since it, at best, can provide only 50% absorption. Of the few other mechanisms available, enhanced collisional absorption localized near the critical density surface becomes the only viable mechanism. In previous work [18] we concluded independently that a wide range of data on absorption could only be explained when enhanced collisional absorption played the dominant role, but we left rather vague the origin of the long-wavelength ion turbulence essential for this mechanism to operate efficiently. In the context of the scenario presented here, it is possible to be more definite on this point. Not only would the pulsation of reflection-seeded SBS provide a source of long-wavelength ion waves in the underdense plasma that could convect up the density gradient to affect the critical surface, but the pulsating laser intensity at that surface could excite turbulence by cyclically "pushing" the critical surface via the ponderomotive pressure. Waves of dimensions c_s/τ_b (c_s being the ion sound speed and τ_b the approximate period of the bursts of radiation reaching the critical surface) would be preferentially excited, and these correspond to waves of dimension $\approx \lambda_0$ —the laser wavelength. It has been shown previously [13] that such waves result in strong, local absorption near the critical surface.

In conclusion, we have observed pulsations of $K\alpha$ emission from laser-irradiated planar targets which imply fast electrons are generated in bursts lasting around 20 psec within these plasmas. The bursts appear to correlate with bursts of second-harmonic emission and are consistent with the view that critical surface processes are responding to modulation of the pump intensity due to the nonlinear dynamics of SBS in the underdense region of the plasma. Furthermore, the appearance of bursts of fast electrons provides an explanation of the frequency sweeping of the emission from the two-plasmon decay instability which responds to the time-varying temperature of the quarter critical region caused by those electrons.

We suggest that the randomness in the bursts of both emissions reflects a transition of the SBS instability into the stochastic regime due to the nonlinear feedback provided by reflection of some of the pump laser light from the critical surface and/or by various nonlinear processes (ion trapping, second-harmonic generation) which influence the dynamics of the instability. Although some of our discussions is admittedly speculative, we are convinced that the weight of evidence supports our comments and that pulsation phenomena provide the essential clue for understanding the detailed interaction physics that really dominates the short-pulse, high-intensity regime.

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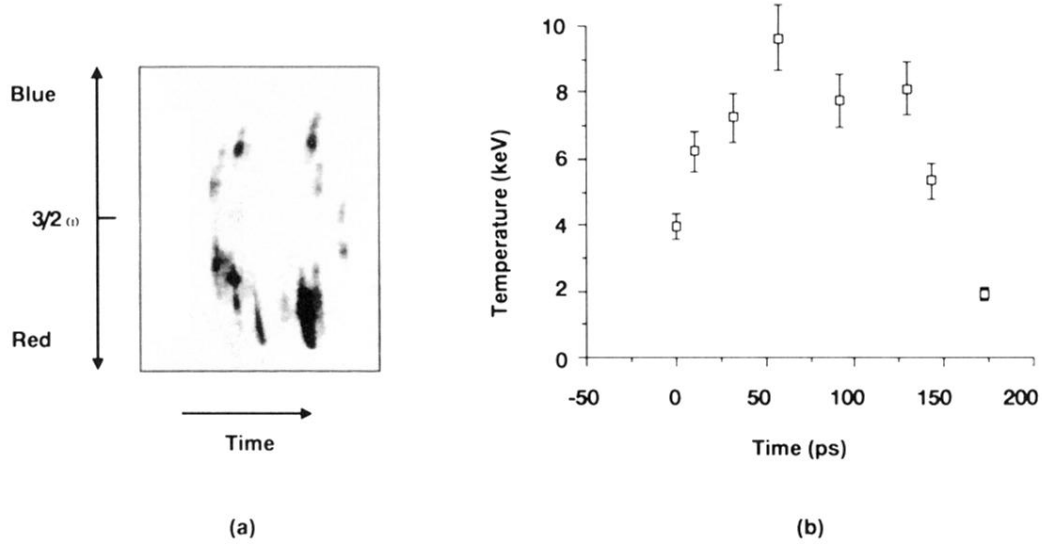


FIG. 1. Time-resolved spectrum of $\frac{3}{2}\omega_0$ emission (a) and the effective plasma temperature in the quarter critical region deduced from the separation of the red and blue spectral peaks in each burst (b).

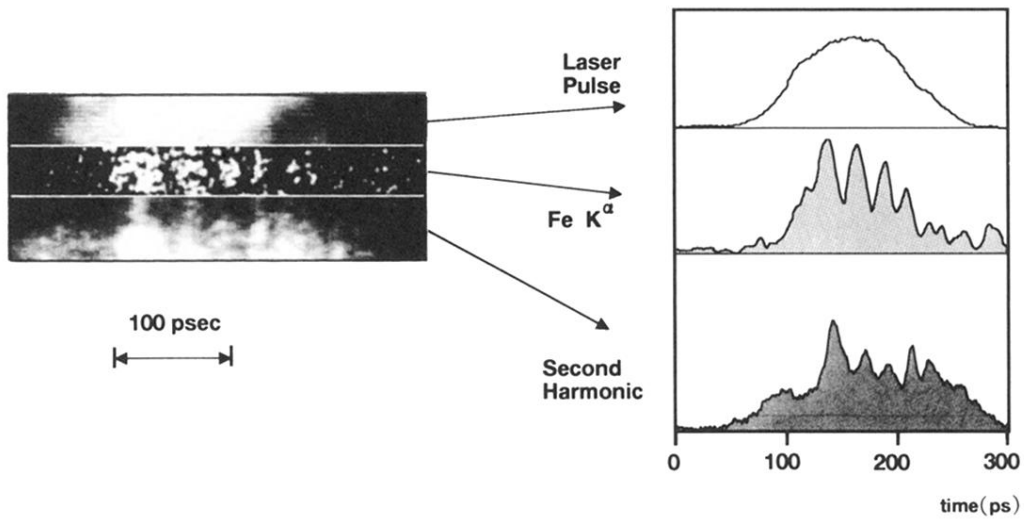


FIG. 4. Temporal behavior of the laser pulse, the x-ray fluorescence from the target, and the second-harmonic emission for the same laser pulse.