Second Harmonic Generation of Stimulated Raman Scattered Light in Underdense Plasmas

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Experiments examining nonlinear scattering mechanisms of high intensity (2×10^{18} W/cm^2) laser light in underdense plasmas were performed. Redshifted second harmonic emission at 45° from the directly backscatterd direction was observed from field ionized plasmas having electron densities ranging up to 10^{19} cm^{-3}. This result is consistent with the doubling of stimulated Raman scattered light from large plasma electron density gradients produced by ponderomotive cavitation.

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The development of short pulse high intensity laser systems [1] in the past several years has enabled researchers to explore new and sometimes unexpected physics, which can occur in laser matter interactions as intensities approach 10^{19} W/cm^2. In this regime, conventional nonlinear optical theory is no longer valid [2] and, in fact, atoms in the target region will be ionized almost instantaneously by the leading edge of the laser field with subsequent interactions occurring in a plasma. Consequently, the interplay between atomic and plasma nonlinearities must be considered when undertaking experiments using such intense pulses [3]. Proposed applications for these lasers include electron acceleration, coherent x-ray generation, and laser "hole boring"—which has recently been postulated as a mechanism for enhancing the fusion rate in inertial confinement fusion (ICF) experiments [4]. An important tool in the study of such interactions is the phenomena of harmonic emission, and in this paper we report a new observation of second harmonic emission from a plasma that may be indicative of electron cavitation by ponderomotive forces [5] in the focal region of an intense laser pulse.

At the Naval Research Laboratory high intensity laser plasma interaction experiments were performed with pulsed gas jet targets to observe the angular distribution of scattered radiation in the visible and near infrared regions. In these experiments we used a table top terawatt laser system, which utilizes the technique of chirped pulse amplification [6], and which operates at a wavelength of 1.054 μm. The laser has a pulse length of 800 fsec, a maximum pulse energy of 1.2 J, and can be focused to intensities greater than 2×10^{18} W/cm^2 using an $f/3$ off-axis parabolic mirror. At such intensities, fully ionized underdense plasmas of hydrogen or helium can be created in the focal region by optical field ionization of the gas by the front edge of the high intensity laser pulse. Resulting plasma electrons in the laser field will have a significant mass increase from their relativistic "quiver" velocities [i.e., $\gamma = (1 + a_0^2/2)^{1/2} \approx 1.3$ for linearly polarized light where $a_0 = e|\mathbf{E}|/m_e \omega_0 c$ such that $\mathbf{E}$ is the laser electric field, m_e is the electron mass, and ω_0 is the laser frequency].

The scattered radiation emitted from these plasmas was examined by imaging the light onto the entrance slit of a spectrometer operating in the visible and near infrared region. Output from the spectrometer was directed into a streak camera having 10 psec temporal resolution with a sweep duration of 0.5 nsec. The radiation from plasma ion recombination (chiefly line emission) in these experiments occurs on a much longer time scale than the streak camera sweep and was not of interest, so only scattered light emitted at times close to the interaction of the laser with the target gas jet was measured. Scattered light was collected by a lens having an acceptance angle of 22°, and the spectrum was examined at various angles with respect to the incident laser direction as well as for various electron densities and laser intensities. The plasma electron density in the ionized gas jet ($n_e \approx 10^{18}$–10^{19} cm^{-3}) was inferred to within approximately 25% from observations of the backscattered stimulated Raman spectrum at relatively low laser intensities (10^{16} W/cm^2), where this backscattered light appears as a narrow peak shifted by approximately the electron plasma frequency, i.e., $\Delta\omega \approx \omega_p$ ($\Delta\lambda \approx 120$ nm at $n_e = 10^{19}$ W/cm^2). The atomic density in the gas jet was controlled by varying the backing pressure.

As the laser intensity increases, the spectrum of backscattered light in the region near the laser frequency becomes extremely broad and exhibits a narrow peak close to the incident laser frequency with a broad oscillatory structure surrounding the fundamental [7]. Such emission is likely due to the behavior of the stimulated Raman backscatter (SRS) [8] and stimulated Brillouin backscatter (SBS) [8] instabilities at high laser intensity. Emission was also observed at sidescattered angles; however, the growth rate of the SRS instability in sidescattered directions is less than that in the directly backscattered direction, and consequently the sidescattered signal is several orders of magnitude weaker and the spectral structure is not as broad.

When the emission spectrum at 45° from the directly backscattered direction is examined near the second harmonic frequency, a broad feature, redshifted relative to the exact second harmonic frequency, is observed. In Fig. 1,

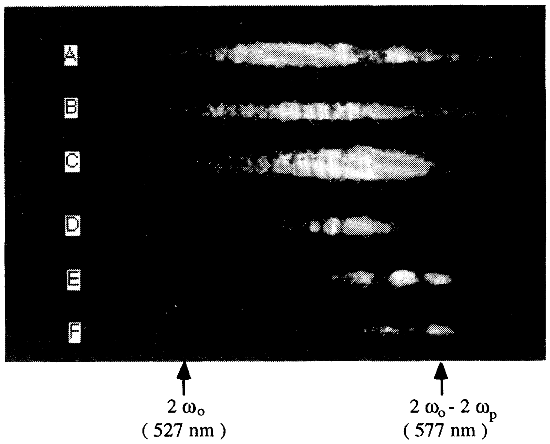

FIG. 1. Variation of second harmonic at 45° backscattered direction with respect to incident laser intensity: (a) 2.0×10^{18}, (b) 1.5×10^{18}, (c) 1.0×10^{18}, (d) 6×10^{17}, (e) 5×10^{17}, and (f) 3×10^{17} W/cm^2.

45° second harmonic spectra are shown for various peak laser intensities at an electron density of about 10^{19} cm^{-3} in hydrogen, indicating that the broadening varied with laser intensity. The signal was approximately 40 nm wide at maximum laser intensity (2×10^{18} W/cm^2) and exhibited an oscillatory structure similar to that shown in the sidescattered radiation near the fundamental. The spectra showed significant blueshifting as the intensity of the laser light increased. This is similar to the behavior of the directly backscattered SRS light, which also blueshifts and broadens as the laser intensity increases, and which has been observed previously [9]. In the experiments described here, however, the amount of blueshifting seems to "saturate" at intensities above 10^{18} W/cm^2. Note that for lower intensities the blueshift is small and the harmonic signal appears to be redshifted by $2\omega_p$.

Two scenarios are plausible for the generation of a redshifted second harmonic signal: (i) laser light may be frequency doubled as it propagates through the interaction region and subsequently redshifted due to stimulated Raman sidescattering in the plasma—leading to a shift of ω_p—or (ii) laser radiation may be first Raman sidescattered (redshifted by ω_p) and only frequency doubled as it passes out of the plasma—resulting in an apparent frequency shift of $2\omega_p$. In Fig. 2(a) sidescattered second harmonic wavelength shift is plotted against plasma density for $I = 10^{18}$ W/cm^2. It is clear that the observed redshift in the signal measured near the second harmonic is approximately $2\omega_p$ and that the second process is occurring, i.e., $(\omega - \omega_p) + (\omega - \omega_p) \rightarrow 2\omega - 2\omega_p \rightarrow \omega_{\text{meas}}$.

There have been predictions of the production of sidescattered second harmonic radiation—in particular, nonlinear Thomson scattering [10] due to relativistic effects. Theory and computer simulations of this mechanism for laser intensities of 2×10^{18} W/cm^2 also predict comparable levels of harmonic emission at other angles

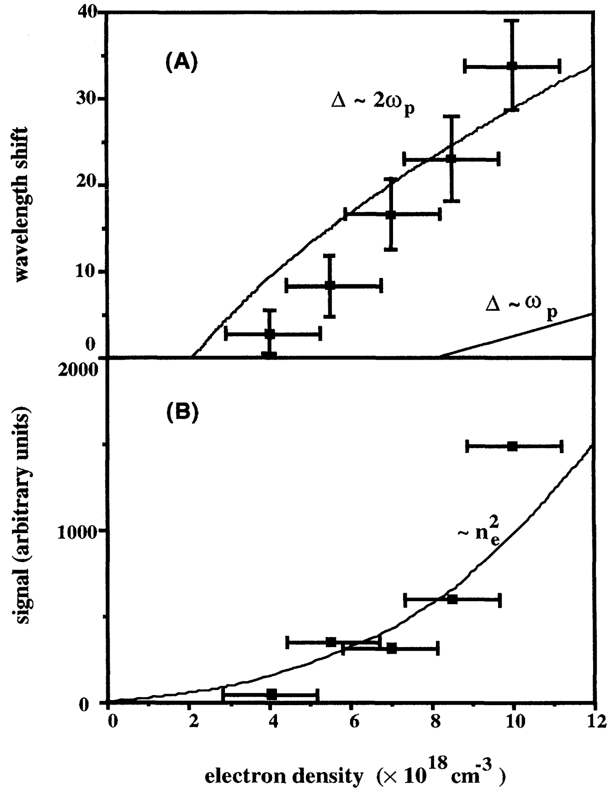

FIG. 2. (a) Wavelength shift of sidescattered second harmonic emission vs electron density (solid lines correspond to shifts of ω_p and $2\omega_p$ using 25 nm intensity dependent blueshift). Vertical error bars indicate FWHM of second harmonic signal. (b) Plot of second harmonic sidescatter emission intensity vs electron density [incident laser intensity was $\sim 1.0 \times 10^{18}$ W/cm^2 in both (a) and (b)].

and at higher orders—however, none were observed. In the experiments described here, emission in the second harmonic region was also measured at relatively low laser intensities of 2×10^{17} W/cm^2 where relativistic effects should be negligible. As shown in Fig. 2(b), in these experiments, the scaling of signal strength versus density is clearly nonlinear in contrast to relativistic Thomson sidescattering, which should show a linear scaling with electron density.

Changing the incident laser polarization from linear to circular does not affect the spectral shape or the signal level of emission in this region (to within the uncertainty in the measurement). There was also no emission in the third harmonic region. No significant emission in the visible and near infrared spectrum was observed at a 90° angle using the same detection system. These observations indicate that the source of the harmonics was neither relativistic effects [10] nor atomic processes.

Even though there are significant amounts of stimulated Raman scattered light in the directly backscattered direction, a second harmonic feature of this type was not clearly observable there. In fact, at intensities of 2×10^{18} W/cm^2 and 10^{19} cm^{-3} density the SRS spectrum in the directly backscattered direction is broadened to extend beyond the second harmonic (to about 450 nm) [7]. It is possible that at the highest intensities some of the scattered light near the second harmonic frequency is due to the doubling of directly backscattered light, although the efficiency is clearly less here than in the sidescattered direction.

It is likely that emission of the second harmonic in the sidescattered direction is due to propagation of the stimulated Raman backscattered or sidescattered light across the steep density gradients surrounding a region of cavitation in the gas jet—this is plausible due to the qualitative similarity of this feature to the "fundamental" sidescattered SRS. For very high intensity light a region of "cavitation" [5] can be caused by the expulsion of electrons due to the tremendous ponderomotive force of the focused laser light, while ions are approximately stationary since they are much more massive. The ponderomotive potential felt by the plasma electrons is given by $\Phi = e^2E^2/4m_e\omega^2$, and was 170 keV in this experiment for laser pulses at maximum intensity. Thus, significant charge separation can occur and a cylindrical shell of excess electrons, which have been expelled from the interaction region, is formed. The amount of cavitation can be simply estimated by a force balance between the ponderomotive force of the laser pulse and the space charge field. In this case, if r_0 is the laser spot size, the density perturbation is given approximately by $n_1 = \Phi/4\pi e r_0^2$, which is about 2×10^{17} cm^{-3} for the laser at maximum intensity. This is about (2–10)% of the electron densities used in this experiment. It should be noted that if self-focusing occurs and increases the ponderomotive potential, a larger density perturbation results.

The second harmonic generation process can be described by the solution of the wave equation with a source term due to the nonlinear current produced as the scattered light passes through the density gradient [11], i.e.,

$$(\nabla^2 - k^2)a_2 = k_p^2(n_1/n_0)a_1\,,$$

where n_0 is the plasma electron density, n_1 is the first order density perturbation, k_p $(=c\omega_p)$ is the plasma wave number, and a_1 and a_2 are the normalized laser intensities of the stimulated Raman sidescattered light and the doubled sidescattered light, respectively. From this equation it can be shown that the efficiency of the second order harmonic generation process is given by

$$P_2/P_1 = a_1^2[(L/4L_p)k_p^2/k_1^2]^2,$$

where L_p is the density gradient scale length, L is the interaction distance, and k_1 is the wave number of the scattered laser light. Using parameters from the present experiment, the predicted efficiency of this process is approximately $10^{-8\pm1}$. Using reasonable assumptions, the experimental conversion efficiency (i.e., the ratio of the second harmonic signal to the scattered fundamental passing through the collection optics) was measured to be approximately 2 orders of magnitude greater than this. This difference may be due to errors in the estimations used and the fact that cavitation-induced self-focusing in the plasma may produce density gradient scale lengths that are significantly less than the focal spot size.

There are other sharp plasma density gradients in the interaction region, especially that at the ionization edge at the side of the gas jet—although the intensity of sidescattered light should be too low at this boundary to produce significant doubling. As well, this gradient may be reduced since the plasma at the extreme edge of the interaction region is very cold and, at such low temperatures, high density plasmas can recombine on picosecond time scales [12] via three-body recombination. Harmonic generation is also critically dependent upon phase matching conditions in the medium [13]. In a plasma, free electrons cause a positive dispersion between the fundamental and the harmonic, and one can approximate the detuning length for the harmonic generation process. The plasma refractive index is given by $\eta = [1 - (\omega_p/\omega_0)^2]^{1/2}$, and the corresponding detuning length is

$$L_d = \pi/\Delta k = \pi c/q\omega_0\Delta\eta = 2\pi q c\omega_0/\omega_p^2(q^2 - 1)\,,$$

which becomes significantly shorter with increasing harmonic order (q). For the second harmonic this detuning length is approximately 70 μm, which is much longer than the density gradient scale length (approximately the laser spot size $\approx$10 μm) produced by the process of cavitation. The lack of a distinct second harmonic signal in the directly backscattered direction may be also due to this effect. The plasma gradient should be smaller in the backward direction—reducing the conversion efficiency—and the interaction distance should be either the laser pulse width or the Rayleigh length—whichever is less. Here the Rayleigh length is 250 μm and is greater than the detuning distance in this case. At high laser intensities, large amplitude plasma wakefields [14] may also be produced, which may increase the fluctuation levels in the plasma and prevent good phase matching, hence reducing the efficiency of this process. However, it is also possible that some of the second harmonic signal observed at sidescattered angles originates from directly backscattered light and exhibits a conical emission pattern as in Ref. [15].

Second harmonic generation has been previously observed at a 90° angle to the direction of the laser propagation during interactions of much lower intensity light (10^{14}–10^{15} W/cm^2) with preformed near-critical density plasmas [16]. This emission was associated with the filamentation instability and an interaction between the forward traveling fundamental and a backscattered wave due to SBS. Forward scattered second harmonic of CO_2 laser pulses (10^{14} W/cm^2) has also been observed by Meyer and Zhu [15] and was attributed to the filamentation instability. These results are in contrast to our experiments where SRS light is doubled—probably by passage through plasma density gradients caused by electron cavitation in the interaction region.

The production of second harmonic radiation can also occur from atomic processes—however, this requires the existence of a spatial asymmetry since, for isotropic media such as gases or liquids, generation of even harmonics cannot occur due to atomic parity considerations [13]. In this experiment, such spatial asymmetry can be caused by an instantaneous radial static electric field produced in the cavitation process. However, for hydrogen, in the region where significant static electric fields exist, the focused intensity is very high and there are no unionized atoms to be polarized. In the region where neutral atoms exist (i.e., where the laser intensity never exceeds about 10^{14} W/cm^2), the static electric field is very small. Previous work has observed second harmonic generation in the forward direction using metal vapors where the mechanism of charge separation was postulated [17]—and work with gas jet targets has also observed second harmonic generation in the forward direction (at laser intensities up to 10^{16} W/cm^2) due to this effect [18]. The efficiency observed ($\sim 10^{-14}$) in those experiments was found to be 2 orders of magnitude less than that for generation of the third harmonic.

In experiments described here, spectra were also recorded for forward scattered light at laser intensities of 2×10^{18} W/cm^2, and the emission of the forward second harmonic (with a bandwidth of 7 nm, which is consistent with the broadened forward scattered fundamental [7]) was found to be strong for helium gas targets (with an efficiency of $\approx 10^{-6\pm1}$) and may have been due to such atomic effects. However, no significant signal from the third harmonic was measured in our experiments. The forward second harmonic signal was also observed in our experiments to be blueshifted by 12 nm (much larger than the average blueshift of the forward scattered fundamental) indicating that such harmonics are produced only at the leading edge of the laser pulse where the ionization rate of the plasma is the highest. It is possible then that second harmonic emission is produced by the singly ionized helium atoms that remain in the laser electric field but that are also subject to an instantaneous static electric field produced by cavitation, although this emission could also be created by the mechanism of frequency doubling in a plasma density gradient as in Ref. [15]. In contrast, forward second harmonic generation was not observed for hydrogen gas. This can be explained since in hydrogen—after initial ionization and charge separation—there are no neutral atoms remaining to be polarized that could generate harmonics in the forward direction. Plasma density gradients would also be smaller in the forward direction in a hydrogen plasma.

In conclusion, this experiment provides the first measurements of frequency doubling of stimulated Raman scattered laser light from a plasma. Theoretical predictions of cavitation effects that result in large density gradients in the laser plasma interaction region may be used to explain the generation of harmonics of radiation scattered out of the interaction region. At high laser intensities, there may be a complex interrelation among the mechanisms of second harmonic generation, cavitation, and stimulated Raman scattering. Although, for the intensities and pulse duration used in our experiments, there is insufficient time for the ponderomotive force to push aside ions and create a "hole" in the plasma, such effects may occur at higher intensity and the phenomena observed in these experiments may become more pronounced. In fact, the measurement of the angular distribution of second harmonic backscatter may provide a useful diagnostic of "hole boring" in ICF experiments which utilize the "fast igniter" concept.

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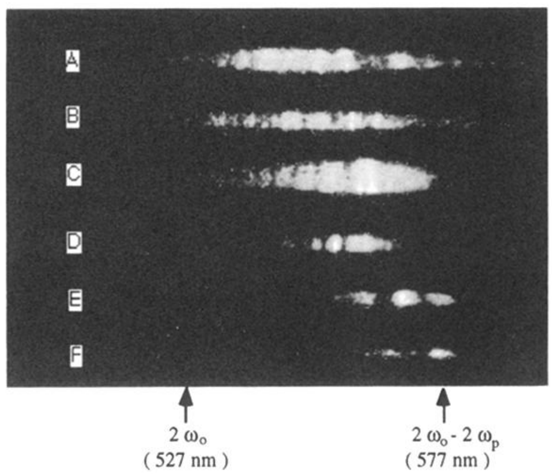

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