Frequency mixing of high intensity laser light with stimulated Raman backscattered radiation in underdense plasmas

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Sidescattered radiation from the interaction of an ultraintense laser pulse (*P*.2 TW) with underdense helium plasmas ($n_e \approx 10^{19} \text{ cm}^{-3}$) is measured and emission at 90° from the axis of laser propagation is observed to be broadened significantly near the second harmonic $(\Delta\lambda=30-40 \text{ nm})$. This result is consistent with sum frequency mixing of forward propagating laser light and the broadened backscattered stimulated Raman scattered radiation that occurs within transient filamentary structures produced by the ponderomotive force of the focused laser pulse. $[S1063-651X(98)03009-8]$

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There has been much interest recently in ultrahigh intensity laser plasma interactions, especially with regard to x-ray laser $[1]$, advanced accelerator $[2]$, and inertial confinement fusion applications $[3]$. Consequently, it is important to have a comprehensive understanding of the many complex and competing processes that may occur during such interactions in order to properly evaluate the feasibility of these potential applications.

In this paper we report results of recent experiments examining high intensity laser interactions with underdense plasmas and particularly observations of the properties of scattered light generated during the interaction. We observed a large broadening of the second harmonic radiation produced at 90° to the direction of laser propagation. This sidescattered spectrum is likely due to sum frequency mixing of forward going laser light (fundamental) with backward going stimulated Raman scattered (SRS) light, which occurs within the transient density channel caused by high intensity charge displacement (cavitation) effects in the plasma. Broadening of the sidescattered second harmonic light is similar to that observed in the SRS light. Measurements of this emission may be useful as a diagnostic of the stimulated Raman scattering instability and density gradients in such plasmas.

For these experiments, we used the Table Top Terawatt laser system at the Naval Research Laboratory (λ) =1.054 μ m, τ 400 fsec) operating at a power of 2.5 TW. The beam was focused to an intensity of about 6 $\times 10^{18}$ W/cm² (in vacuum) using an *f*/4 off-axis parabolic mirror. A target jet of helium gas was produced in the experimental chamber by operation of a pulsed gas valve and supersonic nozzle that had a diameter of about 3 mm. The plasma had a typical electron density of approximately 1.4 $\times 10^{19}$ cm⁻³ ($n_e \sim 0.01 n_{\text{critical}}$) when fully ionized. During our experiments, the high intensity laser beam was focused 1–2 mm above the edge of the nozzle and in the front portion of the gas jet to minimize the effects of ionization induced refraction.

Light directly backscattered by the SRS instability was measured using a 3% reflecting MgF_2 beam splitter and lens combination that imaged backscattered radiation from the interaction onto the slit of a 0.25-m Czerny-Turner spectrometer. At an angle of 90° to the axis of laser propagation (and 45° to the laser polarization vector) the scattered light was collected by a simple lens (angle of acceptance $\sim 5^{\circ}$) and imaged onto the slit of a similar spectrometer. A twodimensional charge coupled device (CCD) array that was sensitive to radiation in the visible and near infrared region was used as a detector. The experimental setup is shown in Fig. 1.

The spectrum of directly backscattered SRS light is shown in Fig. 2 for 2.5 TW. The spectrum has been corrected for spectrometer and detector efficiency. At lower intensities $(I \sim 10^{16} \text{ W/cm}^2)$, the backscattered SRS radiation is a relatively narrow spectral line, which is frequency downshifted by approximately the electron plasma frequency $(\omega_{pe}^2 = 4 \pi e n_e e^2/m_e)$. For a plasma density of 10^{19} cm⁻³, the spectral line is centered at 1170 nm. As the laser intensity is increased the SRS instability becomes ''strongly coupled'' and in the experiment the backscattered light is observed to become broadened, blueshifted, and highly modulated $(4,5)$. We have previously measured the amount of backscattered light to be about 10% in the directly backward direction $[5]$

FIG. 1. Experimental setup for scattering measurements.

FIG. 2. Spectrum of stimulated Raman backscattered light using 2.5-TW incident power $(I \sim 6 \times 10^{18} \text{ W/cm}^2)$. The plasma electron density is $\sim 10^{19}$ cm⁻³.

at maximum incident laser intensity. The blueshifting that we observe in the present experiments at incident powers of 2.5 TW (Fig. 2) is much larger than that previously measured. Note that the peak of the SRS backscattered spectrum has been shifted from 1170 nm to the short wavelength side of the fundamental near 1000 nm (i.e., the shift is greater than 100 nm). These features have been attributed to the nature of the SRS instability in the strongly coupled regime $[4]$. Other experiments have measured blueshifting and broadening of the backscattered spectrum related to the transition from SRS light to stimulated Compton scattering (SCS) [6]. However, it is unlikely that SCS is important in our experiments since this instability only occurs at relatively high temperatures. For our experimental parameters $(\lambda = 1.054 \mu m, n_e)$ \sim 10¹⁹ cm⁻³) the electron temperature should be greater than 500 eV for stimulated Compton scattering to occur (i.e., a temperature somewhat larger than that measured by other groups under similar conditions $[7]$.

Sidescattered emission at 90° near the fundamental frequency from the interaction region is shown in Fig. $3(a)$. Using a two-dimensional CCD array as the detector, it is possible to obtain low resolution images of the interaction region along with spectral information. The interaction of the high intensity laser pulse with the gas jet plasma was characterized by channeling over distances of several millimeters $(1.5-2$ mm in helium) as shown in Fig. 3(a), which is caused by the combined effects of relativistic and ponderomotive

FIG. 3. Spectrally resolved images of sidescattered emission at 90° (P_{laser} ~ 1.5 TW) near (a) the fundamental (1054 nm) and (b) the second harmonic (527 nm) . Note that the laser pulse is propagating from left to right.

self-guiding $\lvert 8 \rvert$. Emission near the fundamental $\lvert \text{Fig. 3(a)} \rvert$ is attributed to incoherent Thomson scattering of the laser light. The blueshift in the scattered light in Fig. $3(a)$ is induced by the rapid field ionization of the plasma by the laser. The blueshift increases as the laser propagates through the plasma.

Second harmonic radiation can be generated in high intensity laser-plasma interaction experiments by the interaction of intense laser radiation with large plasma density gradients as previously observed in other experiments $[9-11]$. Ions move much less than electrons during the interaction and density gradients are produced by the ''snowplow'' effect as plasma electrons are displaced out of the laser path by its ponderomotive force. This effect can also give rise to laser channeling and filamentation effects [12]. Previous work [9] has shown that $2\omega_0$ radiation emitted at 90° angles can be generated as a result of plasma density gradients caused by laser filamentation. However, phase matching arguments imply that the production of appreciable emission of second harmonic normal to the laser axis of propagation requires both forward and backward propagating electromagnetic waves. Those previous experiments were conducted at much lower laser intensity ($\sim 10^{15}$ W/cm²) and in near critical density plasmas where coupling of stimulated Brillouin backscattered (SBS) light to the forward going fundamental was observed $[9]$ and 90° second harmonic emission was used as a diagnostic of filamentation during the interaction. In our case, however, there is little SBS light $(Fig. 2)$ because of the shortness of the laser pulse $({\sim}400$ fsec) and the backward wave involved in the frequency mixing process is most likely the significant backscattered SRS radiation produced during these interactions.

The same 90° CCD-spectrometer setup as used to image scattered light in the spectral region near the fundamental was used to image radiation near the second harmonic [Fig. $3(b)$ on different laser shots. Significant shot-to-shot variability was observed in the fine scale structure of both spectral region images, although the overall emission patterns (two bright regions and increasing blueshift with increasing propagation distance near the fundamental and one bright region with blue and red frequency shifts of 30–40 nm near the second harmonic) were consistent from shot to shot. The overall emission patterns in each spectral region have obvious qualitative differences. Since the qualitative structure of the sidescattered second harmonic is different from that near the fundamental, the production of second harmonic in this direction is probably not caused by a direct frequency doubling of the sidescattered fundamental. The 90° second harmonic emission could be due to incoherent Thomson scattering of the forward propagating second harmonic light. However, the second harmonic generated in the forward direction during such laser plasma interactions $[13]$ is significantly weaker than the forward propagating fundamental (by more than three orders of magnitude) and would consequently result in much weaker 90° second harmonic than that observed in our experiments. In addition, the spectrum of the forward propagating second harmonic is much narrower and not blueshifted.

The combined effect of filamentation and backscattered SRS light may explain the large broadening of the 90° second harmonic observed in these experiments since the backscattered SRS light also exhibits significant broadening (Fig. 2). The spatial localization of the second harmonic emission region $\left[\text{Fig. 3(b)} \right]$ may occur because of the existence of regions where large density gradients are present due to charge displacement effects and the self-focusing behavior of the intense laser pulse.

The second harmonic radiation emitted from a long $(L \gg \lambda)$ filamentary region in the plasma is given by [9]

$$
\mathbf{E}_{2\omega} = (2ik_0/cR)(\mathbf{\tilde{1}} - \hat{\mathbf{R}}\hat{\mathbf{R}}) \cdot \int dz \exp(-2ik_0z \cos \gamma)
$$

$$
\times \int d^2 \mathbf{r}_\perp \exp(-2ik_0\mathbf{R} \cdot \mathbf{r}_\perp)\mathbf{j}_{2\omega}(\mathbf{r}_\perp,z), \qquad (1)
$$

where **R** is the radial vector to the observer, γ is the observation angle, and $\mathbf{j}_{2\omega}$ is the source current given by

$$
\mathbf{j}_{2\omega}(\mathbf{r}_{\perp},z)=(ie/2\omega)\mathbf{V}_{\omega}(\mathbf{V}_{\omega}\cdot\mathbf{\nabla}n),
$$

where V_{ω} is the quiver velocity of the plasma electrons in the electromagnetic field. If V_{ω} is only due to one forward propagating wave then second harmonic emission from Eq. (1) averages to zero at an observation angle of $\gamma=90^\circ$. However, if the quiver velocity is due to both forward and backward propagating electromagnetic waves such that

$$
\mathbf{V}_{\omega}(r_{\perp},z) = \frac{e}{im} \left[\frac{\mathbf{E}_1 \exp(ik_0 z)}{\omega} + \frac{\mathbf{E}_2 \exp(-ik_0 z)}{\omega + \Delta \omega} \right],
$$

where \mathbf{E}_1 is the amplitude of the forward going fundamental

and \mathbf{E}_2 is the amplitude of the backward Raman scattered light, then correct phase matching is possible in the 90° sidescattered direction [9]. Since the backscattered Raman light is strongly blueshifted, the central wavelength is close to the fundamental, i.e., $\Delta \omega \sim 0$ instead of $\Delta \omega \sim -\omega_{pe}$. The 90° second harmonic generated by this mechanism therefore also centers around the second harmonic of the fundamental as shown in Fig. $3(b)$.

In conclusion, we have observed broadening and blueshifting of directly backscattered SRS radiation larger than that previously measured. We have also made spectrally resolved measurements of radiation scattered at 90° to the axis of laser propagation. The observed emission near the second harmonic region is not due to Thomson scattering of the forward propagating second harmonic. It is the sum frequency mixing of the stimulated Raman backscattered light with the forward propagating fundamental in the presence of density gradients caused by cavitation effects along a transient filamentary structure produced by the propagation of the high intensity laser pulse and is consistent with previous theory. This effect may be useful as a technique for diagnostic measurements of stimulated Raman scattering and channel formation during such high intensity laser-plasma interactions.

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