Observation of the dynamics of electron plasma oscillations in femtosecond laser-produced plasmas

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A microplasma is produced on the surface of a glass sample by a 120 fs laser excitation pulse. The optical second harmonic from the plasma is measured using a weak delayed probe pulse. It is shown that the rise and decay of electron plasma oscillations can be mapped out by measuring properly selected second harmonic components as a function of delay time. © 1995 American Institute of Physics.

It is well known that one of the mechanisms of second harmonic (SH) generation in plasmas is coherent scattering of the fundamental electromagnetic wave from the electron density fluctuations associated with electron plasma waves. By making use of the characteristic polarization selection rules of SH generation and selecting the proper SH components it should be possible to directly observe electron plasma waves.

Here we wish to report an experiment in which measurements of the SH were used to directly observe the ultrafast dynamics of electron density oscillations in a microplasma produced on the surface of a solid target during the interaction with an intense femtosecond laser pulse.

We used an excite-and-probe scheme where a strong pump pulse produced a microplasma on the surface of a glass substrate, and a weak probe pulse reflected from the excited surface interrogated the system. The laser pulses were obtained from an amplified CPM dye laser system (λ = 620 nm). The duration, the focal diameter, and the peak intensity of the pump beam on the target surface were, respectively, 120 fs, 7 μm, and 6 × 10^13 W/cm^2. The peak intensity of the probe pulse was down by more than a factor of 10, but otherwise pump and probe were the same.

The experimental geometry is shown in Fig. 1. The p-polarized pump beam and the s-polarized probe beam were incident on the target surface at an angle of q_pump = 37° and q_probe = 50°, respectively. A diaphragm positioned approximately in the direction of the bisector of the reflected beams blocked the latter and passed the reflected SH due to sum frequency mixing of pump and probe. A polarization analyzer placed in the SH beam after the diaphragm suppressed scattered light from the strong pump beam and ensured that only s-polarized SH reached the detector.

Second harmonic generation in plasmas can be described by a hydrodynamic model. However, in contrast to most conventional plasmas, in a microplasma produced by a femtosecond laser pulse the energy of the ions is much lower than the electronic energy during the first few hundred femtoseconds of the plasma lifetime. The hydrodynamic picture of SH generation treats the ions as an immobile positive background charge. Very similar models have been used to treat surface SH generation in free electron metals.

Using a perturbation description the second order induced electronic current density responsible for SH generation can be written:

\[ J^{(2)}_{\text{elec}} = \frac{i e}{4 \pi m \omega} \left( \frac{\omega_p^2}{4 \omega^2} \nabla E_{\text{el}}^2 + E_{\text{el}} \nabla \cdot E_{\text{el}} \right) \]  \hspace{1cm} (1)

Here, \( E_{\text{el}} \) is the electric field vector of the fundamental wave at the frequency \( \omega \), and \( \omega_p \) is the local plasma frequency. The other symbols have their usual meaning.

The second term in (1) is directly proportional to the first order electron density perturbation \( n^{(1)}_{\text{el}} \) associated with the electron plasma wave driven by the fundamental field, \( n^{(1)}_{\text{el}} \propto \nabla \cdot E_{\text{el}} \). For a p-polarized pump field \( E_{\text{pump}} \) and an s-polarized probe field \( E_{\text{probe}} \) the component of the SH current density corresponding to s polarization reduces to

\[ J^{(2)}_{\text{elec}} = \alpha E_{\text{probe}} \nabla \cdot E_{\text{pump}}, \]  \hspace{1cm} (2)

where the constant \( \alpha \) follows from Eq. (1). It can be shown from the conservation of the wave vector components parallel to the target surface that the angle \( \theta_{2\omega} \) of the SH generated by this current is given by

\[ \sin \theta_{2\omega} = \frac{1}{2} (\sin \theta_{\text{pump}} + \sin \theta_{\text{probe}}), \]  \hspace{1cm} (3)

which is to a good approximation the direction of the bisector of the reflected beams. It follows that for the chosen experimental configuration the selected SH component should be proportional to the squared amplitude of the electron plasma wave generated by the pump pulse.

In the experiment we have observed a clear s-polarized SH signal. The SH was critically dependent on the temporal and spatial overlap between pump and probe. The signal disappeared when either the temporal or spatial overlap was spoiled. By measuring the SH as a function of the position of

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the diaphragm it has been verified that the $s$-polarized SH was indeed centered between the reflected pump and probe beam near the bisector. We conclude that the observed $s$-polarized SH exhibits the behavior expected for a source term of the form given by Eq. (2) and thus provides direct evidence of the presence of density fluctuations associated with electron plasma waves.

A typical example of the observed time dependence is shown in Fig. 2, where the measured $s$-polarized SH is plotted as a function of the delay time. Figure 2 demonstrates that the $s$-SH is concentrated in a sharp peak of about 200 fs, which appears to be somewhat asymmetric and slightly up-shifted towards positive delay times.

For a more detailed picture of the time dependence, the measured $s$-polarized SH from three different experimental runs is compared in Fig. 3 with the intensity autocorrelation of the laser pulses, which has been measured using a thin KDP crystal. The autocorrelation data can be represented, to a good approximation, by a squared hyperbolic secant function (thin dotted line in Fig. 3). The comparison shows that there is a clear deviation between the SH from the plasma and the autocorrelation of the laser pulses. Although the dynamic range of the $s$-polarized SH was limited to approximately one order of magnitude due to leakage from the very strong background SH generated by the pump pulse, it appears that the rise and particularly the decay of the SH is significantly slower than that of the autocorrelation.

The observation of a distinct difference between the time dependence of the SH from the plasma and the autocorrelation of the laser pulses provides clear evidence that a two-step process is responsible for SH generation and that the SH signal reveals the dynamics of the electron density oscillations. The two-step process can be described as follows. Firstly, during the interaction with the pump pulse a thin surface layer of the material is ionized and a hot electron plasma is formed which starts to expand rapidly. When the polarization of its electric field vector lies in the plane of incidence ($p$ polarization), the pump pulse couples with the longitudinal modes of the plasma and drives coherent electron density oscillations at the frequency of the light. Secondly, the probe pulse is scattered from these charge density fluctuations. This process gives rise to coherently scattered anti-Stokes light at the second harmonic frequency. Thus, in a pump–probe experiment measuring this SH signal the rise and decay of the electron density oscillations is mapped out.

It is interesting to discuss dynamics of the electron density oscillations observed in our experiment. In a weakly excited, weakly inhomogeneous plasma the damping of long wavelength electron plasma waves is determined by the electron–ion collision frequency. For an electron temperature of several hundred eV the inverse collision frequency at the critical density is much less than the duration of the laser pulses in our experiment. For a linear coupling scheme the electron plasma oscillations would then be expected to follow the driving optical field in a quasi-steady-state fashion. In this case the SH should be proportional to the autocorrelation of the laser pulse. However, the situation is probably quite different for a strongly inhomogeneous, rapidly expanding plasma produced by an intense femtosecond laser pulse. Only two points will be mentioned: (i) For short times, when the pump pulse is still present, a linear coupling scheme is probably inadequate to describe the interaction, e.g., wave breaking phenomena are likely to play a role. (ii) The plasma expansion should possibly be taken into account because the probe pulse sees changing coupling conditions when the delay time is varied. To our knowledge, a theoretical model describing the dynamics of the electron plasma waves under these circumstances is still lacking.

In conclusion, it has been demonstrated that measurements of the SH can be used to detect electron density oscillations in femtosecond laser-produced plasmas. Pump–probe experiments using this method have revealed the femtosecond dynamics of electron plasma excitations.

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