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SECOND HARMONIC IN UNDERDENSE PLASMAS FROM GASES AND THIN FILMS AND ITS ANGULAR DISTRIBUTION

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Introduction

Second harmonic generation (SHG) in laser produced plasmas has been extensively used as plasma diagnostics. Usually we have SHG near the critical density layer of plasmas produced from solid targets. However SHG has been also observed in the corona, far from the critical region, and it's also been used as diagnostics [1], [2]. Here we report on SHG, and on its angular distribution, in experiments with underdense plasmas from solid targets (plastic thin films) and gases, generated by a neodimium laser with λ=1.06 μm and $I \leq 3\cdot10^{13}$ W/cm^2.

In an underdense plasma, the 2ω currents due to free electrons, which are the sources of SH, can be written as

$$\mathbf{j}_{2\omega} = j_0 [(\mathbf{E}\cdot\nabla n) + n \nabla E^2/4] \qquad (1)$$

where

$$j_0 = (\omega_p/\omega)^2 (v_E/L) E_0/4\pi i \qquad (2)$$

We consider a plasma with a density distribution $n_e = n_0 n(r)$, where n_0 is a dimensional constant (unperturbed electron density), interacting with an e.m. wave

$$\mathbf{E}_\omega = \mathbf{e}\, E_0\, E(\mathbf{r}) \exp(\mathbf{k}_\omega \mathbf{r} - i\omega t) = E_0\, \mathbf{E}(\mathbf{r},t) \qquad (3)$$

with a polarization vector $\mathbf{e}$ and a maximum amplitude E_0. Hence, ω_p and v_E are the electron plasma frequency at n_0 and the quiver velocity at E_0. L is a length to which all spatial coordinates will be referred to in the following. It can be taken as the length of the interaction region.

We note from (1) that no 2ω radiation can be generated if n_e is uniform, since in this case $\nabla n = 0$, while $n\nabla E^2$ is irrotational. SHG then implies the plasma to be inhomogeneous in the interaction region. In particular, 2ω will be produced by strong gradients as those generated by whole beam self-focusing and filamentation. Hence SHG is a preferential diagnostics for these processes.

Some theoretical works (e.g [3]) have shown that in underdense plasmas generated from gases, 2ω radiation should be emitted mainly in the forward direction, with no radiation on axis, with ring structures in the emission pattern and without 90^0-side emission.

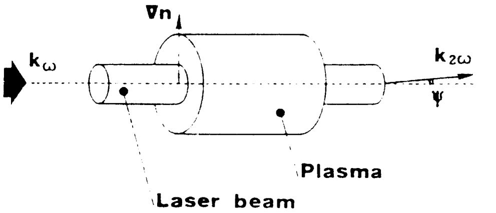

Fig. 1: Geometry of the experiment

We have explicitly calculated the angular distribution of 2ω radiation with a simple model of the physical situation of a laser produced spark as in fig 1

Angular distribution in experiments on gases

To get the angular distribution we write (1) as

$$\mathbf{j}_{2\omega} = \mathbf{j}^{(a)}_{2\omega} + \mathbf{j}^{(b)}_{2\omega} \qquad (4)$$

where

$$\mathbf{j}^{(b)}_{2\omega} = j_0 n \nabla E^2/4 = j_0 \nabla(nE^2)/4 - j_0 E^2 \nabla n/4 \qquad (5)$$

and we consider a linearly polarized electromagnetic field at frequency ω, wavevector $\mathbf{k}_\omega = k_\omega \mathbf{e}_z$ and linear polarization vector $\mathbf{e}_x$. A dielectric constant =1 (valid for the underdense plasma in our experiment) is assumed. Let

$$\mathbf{k}_{2\omega} = k_{2\omega}\mathbf{n} = k_{2\omega}(\sin\Psi\cos\Phi, \sin\Psi\sin\Phi, \cos\Phi) \qquad (6)$$

denote the wavevector of the 2ω radiation, let

$$\mathbf{r} = r\mathbf{e}_r + z\mathbf{e}_z = (r\cos\phi, r\sin\phi, z) \qquad (7)$$

be the position of a point emitting 2ω radiation in the cylinder and let

$$\mathbf{k} = 2\mathbf{k}_\omega - \mathbf{k}_{2\omega}. \qquad (8)$$

Using the far-field approximation, we compute the solution of the wave equation for $\mathbf{B}_{2\omega}$, with sources given by (4), at a point $R_0\mathbf{n}$

$$\mathbf{B}_{2\omega} = \mathbf{B}^{(a)}_{2\omega} + \mathbf{B}^{(b)}_{2\omega} \qquad (9)$$

where

$$\mathbf{B}^{(a)}_{2\omega} = B_0(\mathbf{n}\wedge\mathbf{e}_x) \int E^2(r)(\mathbf{e}\cdot\mathbf{e}_r)(\partial n/\partial r) \exp(i\mathbf{k}\cdot\mathbf{r})dV \qquad (10)$$

$$\mathbf{B}^{(b)}_{2\omega} = -(B_0/4) \int(\mathbf{n}\wedge\mathbf{e}_r)\, E^2(r)\, (\partial n/\partial r) \exp(i\mathbf{k}\cdot\mathbf{r})dV \qquad (11)$$

and

$$B_0 = k_{2\omega} [\exp(ik_{2\omega} R_0)]\, (\omega_p/\omega)^2\, (v_E/c)\, E_0/(4\pi R_0) \qquad (12)$$

It is now convenient to refer to a new orthogonal coordinate system having one axis along the direction of observation $\mathbf{n}$, one along the unit vector

$$\mathbf{e}_1 = (\mathbf{n}\wedge\mathbf{e}_x) = (0, \cos\Psi, -\sin\Psi\sin\Phi) \qquad (13)$$

and the third along the unit vector

$$\mathbf{e}_2 = \mathbf{n}\wedge\mathbf{e}_1 = (-\sin^2\Psi\sin^2\Phi - \cos^2\Psi,\ \sin^2\Psi\cos\Phi\sin\Phi,\ \sin\Psi\cos\Psi\cos\Phi) \qquad (14)$$

so that (10) and (11) become

$$\mathbf{B}^{(a)}_{2\omega} = B_0 \int\cos(\phi)Q(r) \exp(i\mathbf{k}\cdot\mathbf{r})\, dV\, \mathbf{e}_1 \qquad (15)$$

$$\mathbf{B}^{(b)}_{2\omega} = -\{[(\sin^2\Psi\sin^2\Phi + \cos^2\Psi)\int\cos(\phi)Q(r)\exp(i\mathbf{k}\cdot\mathbf{r})\, dV - \sin^2\Psi\cos\Phi\sin\Phi\int\sin(\phi)Q(r)\exp(i\mathbf{k}\cdot\mathbf{r})dV]\mathbf{e}_1 + [\cos\Psi\int\sin(\phi)Q(r)\exp(i\mathbf{k}\cdot\mathbf{r})dV]\mathbf{e}_2\}\, (B_0/4) \qquad (16)$$

where

$$Q(r) = E^2(r)\, (\partial n/\partial r) \qquad (17)$$

Since the dielectric constant of the plasma is close to 1, $|k_{2\omega}| \approx 2|k_\omega|$, and a straightforward, though lengthy calculation shows that

$$\int[\sin(\phi),\cos(\phi)]Q(r)\exp(i\mathbf{k}.\mathbf{r})dV = (2i\pi)\exp(i\chi)\cdot$$

$$\mathrm{sinc}(\chi)[\sin(\Phi),\cos(\Phi)]\int r dr Q(r) J_1(k_{2\omega} r \sin\Psi) \quad (18)$$

where

$$\chi = (k_{2\omega})\sin^2(\Psi/2) \quad (19)$$

$\mathrm{sinc}(x) = \sin(x)/x$ and $J_1(x)$ is the Bessel function. Let's now define the dimensional constant

$$\mathcal{E}_0 = ik_{2\omega}[\exp[i(k_{2\omega}R_0+\chi)]](\omega_p/\omega)^2(v_E/c)E_0/(2R_0) \quad (20)$$

and the form factor

$$\mathcal{F}(\Psi) = \mathrm{sinc}(\chi)\int r dr Q(r) J_1(k_{2\omega} r \sin\Psi) \quad (21)$$

equation (8) finally becomes

$$B_{2\omega}(\Phi,\Psi) = (\mathcal{E}_0/4)[(4-\cos^2\Psi)\cos\Phi\,\mathbf{e}_1 - \sin\Phi\cos\Psi\,\mathbf{e}_2]\,\mathcal{F}(\Psi) \quad (22)$$

From this, the Poynting flux averaged over Φ comes out to be

$$\langle S_{2\omega}\rangle = \mathbf{n}(|\mathcal{E}_0|^2/32)((4-\cos^2\Psi)^2 - \cos^2\Psi)\,\mathcal{F}^2(\Psi) \quad (23)$$

Assuming a gaussian density distribution and denoting its half-width by a, we can finally reproduce the dependence of the intensity on the elongation Ψ according to (23). This is reported in fig 2: note that at 90° there's no 2ω emission.

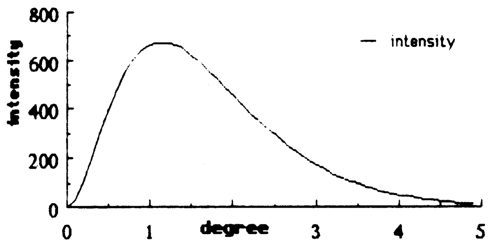

Fig.2: theoretical angular distribution for a=2.7 µm.

In practice, a is the filament radius and in our experimental conditions its value is 2.7 µm, as deduced from analysis of diffracted ω light [5]. The calculated distribution is in agreement with the experimental one [1] as observed in a helium spark at electron density $n_0 \approx n_c/100$ (n_c is the critical density for a Nd laser). This is another hint to the existence of filaments in the plasma. Furthermore, these have also been directly observed in 2ω time-resolved images of the interaction region [1].

Angular distribution in experiments on thin films

In underdense plasmas from solid targets the situation is very much different. As pointed out in [2], we can now have an additional fundamental wave

$$E^B_\omega = \mathbf{e}\,E^B{}_0\,E(\mathbf{r})\exp(-\mathbf{k}_\omega\mathbf{r} - i\omega t) = E^B{}_0\,\mathbf{E}^B(\mathbf{r},t) \quad (24)$$

propagating back toward the laser. This can arise from a reflection at an inner critical surface or from stimulated Brillouin backscattering (SBS). In this case, we have two additional terms in $j_{2\omega}$. The first one is analogous to (1) with E^B in place of E. This gives a 2ω wave in the backward direction ($\Psi = \pi$). The second term is

$$j_{2\omega} = j_0[E^B E + E E^B]\cdot\nabla n \quad (25)$$

where $E^B E$ and $E E^B$ are second order tensors. This term produces a contribution normal to the laser beam ($\Psi = \pi/2$). Indeed in this case the z-integration in the far field integral, which before gave the factor $\exp(i\chi)\mathrm{sinc}(\chi)$, now yields a factor proportional to

$$\int dz \exp(-2\,i\,k_{2\omega}\cos\Psi) \quad (26)$$

which has a maximum at $\Psi = \pi/2$, with an effective angular width $\Delta\Psi = \lambda/L$. This is a clear difference between experiments in gases and on solid targets. However the formation of density gradients perpendicular to the beam axis is still necessary for 90° emission and these could be again due to self focusing and filamentation.

In a recent experiment with plasmas generated from very thin plastic films [6] we collected 90°-side time resolved images in 2ω light of the expanding plasma. Fig. 3, shows one picture of 2ω source evolution for a Formvar film of 1.25 µm thickness. The extension of the 2ω sources in a region of the order of 1 mm perpendicular to the film confirms that SH originates from a plasma definitely underdense and hence (25) is the only possible origin of SHG, where E^B must be due to SBS.

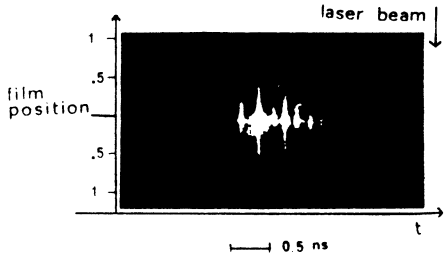

Fig.3: time resolved 2ω image for d=1.25 µm.

We note that the process (25) could be better defined as "sum of frequencies". Indeed the backscattered wave E^B has a frequency ω_B slightly different from ω

$$\omega_B = \omega + \omega_i \quad (27)$$

where ω_i is the frequency of the ion sound wave which is generated by SBS together with the backscattered e.m. wave E^B. This difference produces a red shift in 2ω radiation of the order

$$\Delta\omega = -(v_S\,\omega/c^2)(1-n_e/n_c)/(1+4k_\omega\lambda_D^2) \quad (28)$$

where v_S is the sound velocity in plasma and λ_D the plasma Debye length. Time resolved spectroscopy of the 2ω line is now in progress to see this effect.

In conclusion these notes can explain our experimental observations, showing forward annular (and no 90°) 2ω emission in plasmas from gases and 90° 2ω emission in underdense plasmas from thin solid targets.

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