Spectral Broadening in the Light Transmitted through a Rapidly Growing Plasma

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It has been found that CO_2-laser intensities greatly exceeding the normal breakdown fields may be transmitted in many gases. When breakdown does occur at these high intensities, the initial plasma growth is so violent as to cause frequency shifts of ~ 1 cm^{-1} in the beam transmitted through the focus. The interpretation of the spectrum induced by this plasma nonlinearity indicates that CO_2-laser pulses of duration as short as 30 psec were produced.

Although TEA (transverse electric atmospheric) CO_2-laser–induced breakdown in gases has been studied for some time,[1] the factors which determine the threshold intensity are still rather obscure. In most theoretical treatments[2,3] one assumes a starting electron, and then simply calculates the intensity I_{aval} required for a sufficiently fast avalanche multiplication rate.

While this approach may be valid in certain experiments,[4] it is now thought that the plasma frequently nucleates on dust particles,[5] and then spreads through the focal region in the form of an "optical detonation wave."[6]

In the experiments reported here, the emphasis was placed on reducing the influence of the impurities. To this end, a high flow rate was sustained through the gas cell, making available a fresh supply of gas for each laser shot. Moreover, very tight focusing, of aperture ratio $f/3$, caused the focal region to be very small, thereby helping to avoid dust particles which are present in finite concentration only.

This approach permitted the transmission of a CO_2-laser intensity of close to 10^{11} W/cm^2 in 55 atm of helium gas. Such a high intensity greatly exceeds the normal breakdown threshold I_{aval} which has been measured[4] in the presence of preionization at 1 atm. In fact, it exceeds by more than 2 orders of magnitude the intensity I_{aval} appropriate to helium at 55 atm. The scaling law[3] for pressure implies that the avalanche e-folding rate $1/T$ is close to 10^{12} sec^{-1} in this experiment.

Since breakdown usually did not occur under these conditions, it is safe to say that frequently there was not even a single free electron available in the focus to initiate the avalanche. Similar behavior was observed in nitrogen and argon.

In the remainder of this paper, we will exploit the following concept: When breakdown finally does occur at these ultrahigh intensities, the plasma growth will be so violent as to produce substantial phase modulation on the transmitted light beam. This form of plasma nonlinear optics is particularly suited to the CO_2 laser. Its significance is shown by the fact that at this wavelength a free electron has 10^5 times greater refractivity than a bound electron.

Since the nonlinear techniques used in the visible region are generally unsuitable at the CO_2 wavelength, the plasma nonlinearities will play an important role in exploiting the full potential of this laser. As an example, we will show how these nonlinearities may be used for pulse shaping to produce a picosecond pulse.

The experimental configuration is shown in Fig. 1. The TEA laser output of about 2 MW is focused by an $f/3$ aspheric Irtran 2 lens of 1 in. focal length to an area about 2×10^{-5} cm^2 in a high-pressure gas cell. The light is recollimated, spatially filtered, reflected off an uncoated Ge etalon 1 mm thick, and focused into the slits of a Spex $\frac{3}{4}$-m grating monochromator. The Ge Fabry-Perot[7] is used as a prefilter for the monochromator, and is set for a minimum in reflection of the incident laser beam.

Figure 2(a) shows a typical oscilloscope trace of the light at the exit slit when the monochromator wavelength is centered on that of the incident laser beam. The modulation periodic at 6 nsec represents longitudinal mode beating. The instant of breakdown is defined within a nano-

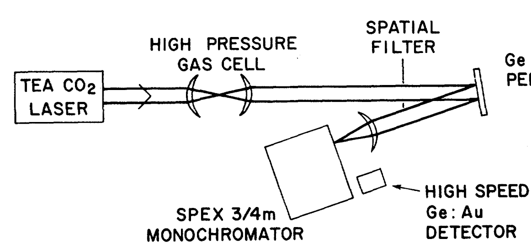

FIG. 1. Experimental configuration.

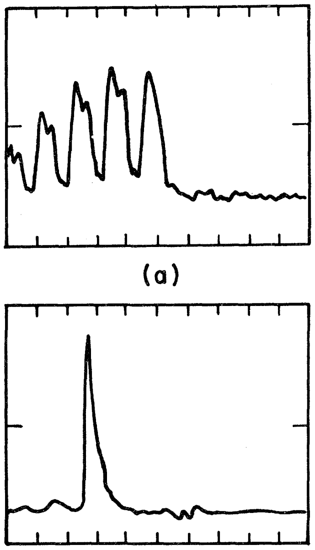

FIG. 2. (a) Transmitted light at the incident laser wavelength as observed with a pyroelectric detector and a 7904 oscilloscope. The sudden drop in intensity indicates formation of a laser breakdown plasma. The periodic modulation is longitudinal mode beating. (b) Transmitted light at a frequency shifted 0.5 cm^{-1} from the laser as monitored on a Ge:Au detector. The spike occurs at the instant of breakdown. The precursor structure is leakage of laser light through the monochromator. After the spike one observes reflections in the transmission line leading to the scope. Time scale: 5 nsec/div.

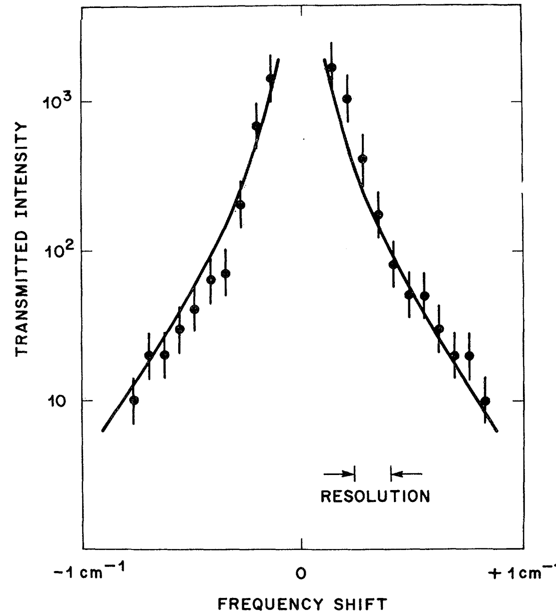

FIG. 3. Intensity of the spike observed in Fig. 2(b) plotted as a function of frequency shift from the incident beam. This particular observation was made in He at 55 atm and a laser intensity of $\sim 10^{11}$ W/cm^2. Solid line, theoretical curve based on a simple step-function model for the amplitude modulation of the transmitted light. We conclude that the fall time for the amplitude was only 20 psec.

second by the sudden drop in transmitted light. The actual fall time is not resolved.

Figure 2(b) shows the transmitted light when the monochromator is detuned from the incident beam about 0.5 cm^{-1}. At the instant of breakdown, a huge spike appears at the shifted wavelength. The structure before the spike is leakage of incident laser light through the monochromator. The structure after the spike represents electrical reflections in the transmission line leading to the scope. The true width of the spike is not resolved. Estimates shown below indicate the pulse width to be ~30 psec.

Figure 3 gives the frequency spectrum of the spike amplitude showing a symmetric broadening about the laser frequency. The inherent linewidth[8] of the TEA laser output, 0.02 cm^{-1}, is negligible. It should be emphasized that there were tremendous fluctuations in the width of the spectrum and the spike amplitude from shot to shot. This is characteristic of a nonlinear process. On some laser shots no broadening was observable at all. Figure 3 was obtained by averaging only those shots with strong broadening. The fluctuations will be explained below.

The frequency broadening represents either a phase or amplitude modulation of the beam caused by a rapidly changing plasma index $\Delta n(t)$. Under our conditions, it is safe to assume that $\omega_p \ll \omega$, in which case $\Delta n(t)$ is simply proportional to the plasma density $N(t)$. Here ω and ω_p are the laser and plasma frequencies, respectively. The contribution to the phase ψ of the electromagnetic wave is $-\Delta n(t)z\omega/c = \frac{1}{2}(\omega\tau - i)N(t)z\sigma$, where τ is the electron-atom collision time, z is the propagation path length, and σ is the free-electron absorption cross section. The imaginary part of the phase represents amplitude modulation caused by absoprtion of the wave in the plasma. Because of the avalanche growth, the free-electron density is $N(t) = N_0 \exp(t/\mathcal{T})$, where $\mathcal{T}$ is rather short as discussed earlier.

The power spectrum of a wave of such frequency and amplitude modulation is $(\pi T/2\omega')$ $\times \{\exp[(\pi - 2\theta)\omega' T] - \exp[-(\pi + 2\theta)\omega']\}^{-1}$, where ω' is the frequency shift and $\theta = \arctan(\omega\tau)$ in the first quadrant. For $\omega\tau \gg 1$, which is true at these pressures, the spectrum is asymmetric, i.e., much stronger in the anti-Stokes direction because of the negative real part of the plasma index dominating the absorptive part.

These predictions disagree with Fig. 3, where the spectrum is symmetric indicating a pure amplitude modulation. Furthermore, the broadening predicted above is greater than that actually observed.

The reason for these discrepancies is interesting and is related to the spatial inhomogeneity of the plasma which was completely ignored in the theory presented above. The inhomogeneity arises for two reasons: The plasma must begin at a nucleation center and then spread through the focal region. Depending on its rate of spreading it will be more dense closer to its point of nucleation. Additionally, the avalanche growth rate depends on intensity; therefore the plasma will become dense more quickly in the center of the focus.

The phase shift ψ for rays traversing regions of differing plasma density will be different and will attain maximum value of $\omega\tau$ before absorption sets in at $\frac{1}{2}Nz\sigma \approx 1$. Each part of the beam will suffer a different phase shift between 0 and $\omega\tau$, and because $\omega\tau \gg 1$ they will interfere destructively when all their contributions are superposed; i.e., the beam will be scattered out of the forward direction by the index inhomogeneity.

The net effect is a falling off in the amplitude of the transmitted beam during the time required for the plasma to spread through the focal region. The method of short-pulse generation is to set a monochromator to transmit a sideband but to block the carrier (laser) frequency. Before breakdown nothing is transmitted; during the sudden fall in amplitude the sidebands are produced, and afterward again nothing is transmitted. Thus a short pulse is created of duration approximately equal to the fall time.

Let us represent the amplitude by the steplike function $\frac{1}{2}(1-\tanh t/\mathcal{T})$ whose fall time is $\sim 2.2\mathcal{T}$. The power spectrum of this amplitude modulation is $\frac{1}{2}\pi\mathcal{T}\sinh^2(\frac{1}{2}\pi\omega\mathcal{T})$. It is fitted in Fig. 3 for a value of $\mathcal{T}=9$ psec. The fall time is about 20 psec. If a partial Fourier transform is taken, including only a sideband but excluding the laser frequency, the result in the time domain is a Lorentzian pulse with full width at half-maximum of $\pi\mathcal{T} \approx 30$ psec. The above results are expected to be insensitive to the precise details of the exact shape of the amplitude function, but depend mainly on its actual fall time. Therefore, we conclude that the duration of the unresolved spike in Fig. 2(b) is ~ 30 psec.

The speed of propagation of the plasma front may also be estimated. The transverse radial dimension of the focus divided by the fall time yields a speed about 10^8 cm/sec.

It remains to explain the large shot-to-shot fluctuations in the width of the spectrum. The cause is thought to be the difference in location of the nucleation center of the plasma on successive pulses. Since the laser intensity is so high, the plasma may nucleate on dust particles well outside the focal region. In those shots the fall time will not be as rapid and the broadening not as great. For better consistency, measures must be taken to ensure nucleation always at the center of the focus.

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[7]I would like to thank Michel Duguay for pointing out to me the usefulness of a Fabry-Perot in the reflection mode.

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