Self-Phase Modulation of Light in a Laser-Breakdown Plasma

Eli Yablonovitch

Bell Telephone Laboratories, Holmdel, New Jersey 07733

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Self-phase modulation in a CO$_2$-laser breakdown plasma is observed to down-shift the wavelength from 10.6 to about 10.0 $\mu$m. This results from the sudden drop in refractive index, from 1 to 0, which accompanies the ionization of the gas. The pressure dependence of this effect in nitrogen is opposite to that in helium.

Under certain conditions, the growth rate of plasma density in a laser-breakdown experiment may be very rapid. It has previously been shown$^1$ that with clean gas and a small focal volume the laser intensity may exceed the normal breakdown threshold$^2$ by orders of magnitude before plasma nucleation occurs. An intensity of $10^{12}$ W/cm$^2$ and an avalanche$^5$ growth rate $\simeq 10^{12}$ sec$^{-1}$ are readily achieved.

In this high-intensity regime, the focal volume resembles an explosive medium. Once the plasma is triggered, the neutral gas becomes rapidly ionized. The index of refraction $n$ drops from a nominal value of 1 in the neutral gas to a value of 0 as the electron density exceeds the critical density. Thus the space and time dependence of the index $n(x,t) = (1 - \omega^2 / \omega_n^2)^{1/2}$ produces a sudden phase and amplitude modulation of the transmitted beam during the time required for the plasma to form. Here $\omega$ and $\omega_n$ are the laser and plasma frequencies, respectively. Since the change in index of refraction is negative, the phase modulation$^2$ may be expected to shift the incident light to higher frequencies.

The first observation of this effect$^1$ was of a symmetric spectral broadening, less than 1 cm$^{-1}$, about the incident frequency. We have now extended these observations to frequency shifts as high as 50 cm$^{-1}$, or about a 5% shift. As earlier expected, these experimental spectra are quite asymmetric, i.e., much stronger in the anti-Stokes direction.

It is interesting to compare the spectral broadening in transmission through a breakdown plasma with that seen using ultrahigh-power lasers in reflection from a solid target$^3$ in vacuum. In both cases the broadening is probably due to the rapidly varying index $n(x,t)$. In the latter case it comes from material motion in the target interaction region which causes the plasma density to be a function of space and time.$^6$ In the former case $n(x,t)$ varies as the neutral gas becomes ionized and actual material transport is not required. A radiative mechanism is sufficient to spread the plasma rapidly from its point of nucleation. Therefore the self-broadening effect can be very strong even with a laser of only moderate power.

The observation of self-phase modulation with large anti-Stokes shifts in a breakdown plasma produced by external focusing has important implications for the phenomenon of self-focusing. The suggestion$^2$ has been made that superbroadening$^5$ of light in self-focused filaments may re-
sult from the creation of a low-density plasma in the medium.²

Figure 1 shows the experimental configuration. The source is a Lumonics model 103 transverse-excitation–atmospheric (TEA) CO₂ laser equipped with an unstable resonator and capable of producing 5 to 10 MW of peak power. The light is brought to a focus by a well-corrected, f/1 germanium doublet,³ and then recollimated by a similar lens. A focal-spot diameter of 17 μm is produced. The beam is then transmitted through a Spex double monochromator and detected by a 400-MHz, liquid-helium cooled, Ge:Hg photoconductor. The electrical signal is transmitted via a 50-Ω coaxial cable from inside the detector Dewar to a Tektronix 519 oscilloscope. (For some of the measurements the focusing lens was mounted in a high-pressure cell.)

The incident pulse length is about 60 nsec. At some point during the rise time of the laser, a plasma nucleates, and abruptly cuts off any further transmission of light within 1 nsec, the system response time.

With the spectrometer tuned away from the incident wavelength, the trace in Fig. 2 is seen on the oscilloscope. This 1-nsec pulse appears exactly at the instant of plasma nucleation. Although its shape remains constant, its amplitude fluctuates greatly from shot to shot. The shape of the trace in Fig. 2 is actually the response of the detection system to a δ function in time. The 370-psec rise time is faster than the 1-nsec recombination time of the photoconductor, indicating that it is acting as an integrator. The rise time is then limited by the oscilloscope (300 psec) and the capacitance of the detector. The fall time is given by the recombination time, and the wiggles on the tail are due to ringing in the transmission line caused by the high-speed transient. From the observed rise time, we can place an upper limit of 300 psec on the optical-pulse duration.

In Fig. 3(a) is shown the result for the breakdown spectrum in nitrogen at 1 atm. The anti-Stokes side has about 10 times the energy of the Stokes side. From the large frequency shifts, up to 50 cm⁻¹, it is clear that the gas is becoming ionized on a picosecond time scale. With the pressure increased to 40 atm of nitrogen the spectrum becomes weaker and narrower, Fig. 3(b).

On the other hand, the pressure dependence in helium is exactly the opposite. An increase in pressure increases the energy in the sidebands and causes the spectrum to broaden, Figs. 3(c) and 3(d).

For comparison, Fig. 3(e) plots the spectrum $P/2\pi(\Delta \omega)^2$ which would result from an instantaneous cutoff of the beam. ($P$ is the laser power and $\Delta \omega$ is the frequency shift.) Such a step-function modulation is observed within the limited time resolution of the oscilloscope. Not surprisingly, the agreement is satisfactory only for small frequencies, corresponding to long times. Of course, this model contains no phase modulation and says nothing about the pressure dependence.

The spread of the plasma from its point of nucleation is thought to be responsible for the spectra in Figs. 3(a)–3(d). Raizer¹¹ has treated the problem of the radiative propagation of a zone of ionization under the influence of a laser beam. He did not consider, however, the regime in which the laser intensity greatly exceeds the breakdown threshold. In this case, the plasma density grows temporally at the avalanche growth rate $1/T$, and spreads spatially by means of ionizing radiation into the surrounding neutral gas. Secondary avalanches are therefore initiated a distance $L$ into the neutral gas, where $L$ is the mean range of ionizing radiation coming from the plasma. The speed of propagation of the ion-
FIG. 3. (a)–(d) Spectra produced by laser-induced breakdown in gases at various pressures. Note the pressure dependence in nitrogen is opposite to that in helium. The energy in the sidebands is measured relative to the incident laser power. Therefore the units on the ordinate axes are picoseconds per wave number. (e) The spectrum $1/2\pi(\Delta \omega)^2$ which would be produced by a step-function modulation of the beam.

The peculiar pressure dependence observed in nitrogen and helium may be associated with changes in $L$.

A more detailed understanding of this process awaits further experimentation. The most important unknown is the source of the initial electron, or the nucleation mechanism. Direct electron tunneling would require an intensity of $\sim 10^{14}$ W/cm$^2$ and is ruled out. The intensity at the instant of nucleation fluctuated greatly from shot to shot. When gas filters in the 100-Å range were employed, a factor 2 increase in the mean intensity was observed. This seems to indicate that small dust particles or large molecules were responsible. Further efforts in cleaning up the gas are likely to be rewarded by increased reproducibility as well as increased nucleation intensity. The regime near $10^{14}$ W/cm$^2$ is especially interesting since the ionization potential would be strongly Stark shifted in such an electric field.

The measurement of the spectrum induced by breakdown was complicated by the fluctuations. Each experimental point in Fig. 3 is the peak amplitude of the signal observed from twenty successive laser shots. The line drawn through the points is therefore the "envelope" of the peak values occurring in twenty successive spectra. It may be desirable to repeat this experiment with a visible laser and record the entire spectrum on photographic film in a single shot.

Techniques of short pulse generation will be treated in a subsequent paper. Suffice it to say that the approach described here is applicable to other laser wavelengths. The limit on pulse duration is the time required for the plasma to spread through the focal region, which at $10^{14}$ W/cm$^2$ is likely to be only a few optical cycles.

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Observation of the Stability Window in the Fast-Toroidal-Pinch Experiment

Kei-ichi Hirano, Shiro Kitagawa, and Mikio Mimura

Institute of Plasma Physics, Nagoya University, Nagoya, Japan
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A fast toroidal pinch with very small aspect ratio (2.6) is operated to test tokamak configuration. A narrow stability window appears between \( q = 1.3 \) and 1.5. In the stable region, the energy loss from the plasma is mainly governed by the decay of the plasma current, which, in the present case, is controlled by the external circuit. In the unstable region, the energy-loss rate observed under clean vacuum conditions is found to agree approximately with the calculated growth rate of the kink mode.

The application of \( \theta \)-pinch techniques to tokamak discharges is one of the most powerful and efficient ways to improve both the \( \beta \) and density values obtained in usual tokamaks which are limited by the relatively small input power of Joule heating. A symmetric toroidal pinch (STP) system has been constructed to check the theoretical and technical \( \beta \) limit of tokamaks. Since the \( \theta \)-pinch techniques are well established and simple, similar experimental efforts have been reported previously.

In order to improve the \( \beta \) value of a tokamak with circular cross section, the aspect ratio \( A \) and the safety factor \( q \) should be as small as possible. The reduction of \( A \) is limited for technical reasons, and too small a value of \( q \) leads to violent magnetohydrodynamic instability. In this Letter, the study of the relation between the growth rate of the instability and the \( q \) value is reported. When various factors are optimized, it is found that a lower technical limit of \( A \) is about 2.6 in the case of the STP.

A schematic drawing of the STP system is shown in Fig. 1. The major dimensions and parameters are indicated in Table I. In order to cancel the field error generated by the tab of the induction coil, which is the primary winding for inducing the plasma current, a double-copper shell structure is adopted. The operating time sequence of the STP system is shown in Fig. 2.

Figure 3 shows typical wave forms of the toroidal field and the plasma current, and also the negative one-turn voltage induced by the decay of the primary current. The toroidal field \( B_z \) and the primary current \( I_z \) are crowbarred simulta-

![FIG. 1. Schematic drawing of the STP system.](image-url)