Laser-induced breakdown of argon at 0.35 μm

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Laser-induced breakdown of argon and other gases at a wavelength of 0.35 μm has been measured in the pressure range 200 to 2000 Torr. The results of short-pulse ($\tau_p = 0.4$ ns) and long-pulse ($\tau_p = 500$ ns) measurements in argon indicate that at $p = 1$ atm the breakdown threshold I_{th} scales as $\tau_p^{-0.7}$. Practically complete absorption of the laser beam by the broken-down plasma has been observed at the highest pressures studied. A model for breakdown in Ar is described which gives satisfactory agreement with the data.

The use of ground-based high-power lasers to propel rockets into space is an attractive concept since a laser beam that is focused into the nozzle can heat the propellant to very high temperatures. The laser can provide a specific impulse (exhaust velocity divided by the acceleration of gravity) several times larger than that attainable by the best chemical propellants. The objective of this work is to determine whether the concept of a pulsed laser-heated thruster, which was first formulated for the CO_2 laser[1] and has been experimentally demonstrated at that wavelength,[2] can be extended to much shorter wavelengths. High-power excimer lasers which radiate in the uv appear particularly attractive for laser propulsion since diffraction of the beam, which presents a limitation to the effective range for propulsion, scales as the wavelength squared. Breakdown thresholds and plasma absorption lengths are, however, both expected to be larger in the near uv than at 10.6 μm. Since these quantities will influence the design of the nozzle and the pressure of the propellant gas required for optimum performance, it is important that they be determined.

Breakdown thresholds as a function of gas pressure have been measured previously for argon (and other gases) at $\lambda = 347$ nm (doubled ruby wavelength) for laser pulse lengths τ_p of 20 ps,[3] 8 ns,[4] and 20 ns.[5] We present in this paper breakdown and absorption measurements obtained using an *e*-beam pumped excimer device (Maxwell Laboratories, Inc., Maximer™ 10-1) operated as an XeF laser ($\tau_p = 500$ ns, $\lambda = 353$ nm, rise time =100 ns, energy =5 J) and using a frequency-tripled Nd glass laser ($\lambda = 351.3$ nm, $\tau_p = 0.4$ ns) made available to us by the National Laser User Facility, Laboratory for Laser Energetics, University of Rochester, Rochester, New York.

The experimental arrangement used for the XeF breakdown and absorption measurements is shown in Fig. 1. Chamber fill pressures were monitored with standard Bourdon dial gauges. High-purity gases were introduced into the test volume through a gas manifold constructed of copper tubing. The gases could be further purified by passing them through a column of molecular sieve 13X to remove any residual water vapor or hydrocarbons. The laser beam entered the test chamber through a fused silica window. In order to obtain a diffraction-limited beam using the XeF laser, we had to mask the 10×10 cm^2 annular output beam of the laser so that only a 2.5×2.5 cm^2 segment was allowed to pass. Using a 1.6-m-focal-length mirror, we found the dimension of the central lobe in the focal region (containing 80% of the power) to be 5×10^{-3} cm. The focused spot was measured by imaging it through an optical magnification system and then recording the image photographically. (To prevent overexposure of the film, the input beam was attenuated in several stages to achieve an overall energy reduction of more than six orders of magnitude.) The maximum intensity at focus that could be reached with our experimental setup was 2×10^{11} W/cm^2. Several diagnostic measurements were employed to monitor the gas breakdown and subsequent laser absorption. Firstly, breakdown was detected by visually observing or pho-

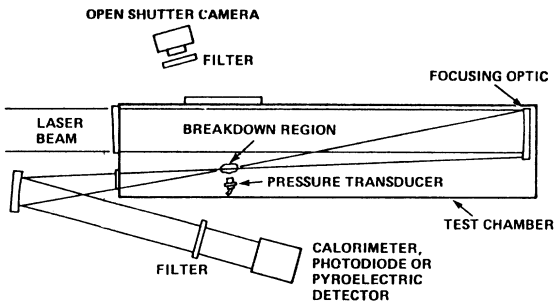

FIG. 1. Schematic diagram of experimental setup.

tographically recording the formation of a bright spark in the laser focal region. Secondly, measurements were made of the laser radiation transmitted beyond focus. Both time-integrated and time-resolved measurements were performed using, respectively, a large-area calorimeter and a high-speed uv-enhanced silicon photodiode (EGG UV-040B). A similar scheme was used to identify the onset of breakdown in the short-pulse data.

Experimental results for Ar, CH_4, and N_2 are shown in Fig. 2. We also attempted breakdown measurements in H_2 and NH_3, but were unable to achieve breakdown in these gases for pressures below 3 atm. The data on breakdown intensity versus laser-pulse duration for argon at 1 atm are shown in Fig. 3. Breakdown measurements using the $\tau_p = 0.4$ ns, tripled Nd glass laser yielded a threshold of $(6 \pm 4) \times 10^{12}$ W/cm^2. The large error limits are the result of uncertainties in the beam spatial distribution at focus (obtained from two-dimensional densitometer measurements in the focal region of an equivalent diagnostic beam) as well as difficulties that were encountered in precisely defining the onset of "breakdown." For the result quoted here, the breakdown threshold is defined as the lowest power density at which (1) measurable attenuation ($\sim$10%) of the transmitted beam was observed, and (2) a "bright" visible glow was seen in the laser focal region.

A detailed model of the physical processes leading to breakdown in argon, which is the simplest gas to analyze of those studied, has been developed and will be presented elsewhere.[6] We present the essential features below. The excited states of Ar that play a role in the model are shown in Fig. 4. The electron concentration builds up in three stages.

In stage 1, initial electrons are formed by multiphoton ionization of impurities (organic impurities having an ionization potential $E_I \simeq 8$ eV) and argon.

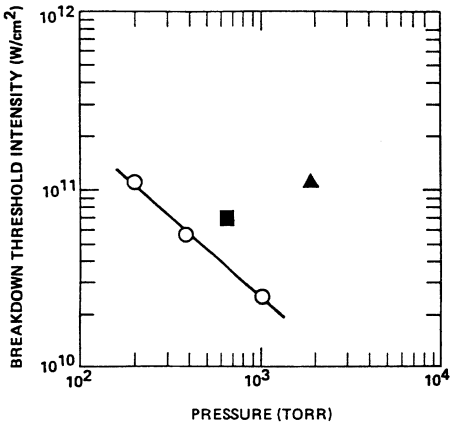

FIG. 2. Experimental laser-induced breakdown thresholds for several gases at $\lambda = 0.35$ μm, $\tau_p = 0.5$ μs. ○ Ar, ■ CH_4, ▲ N_2.

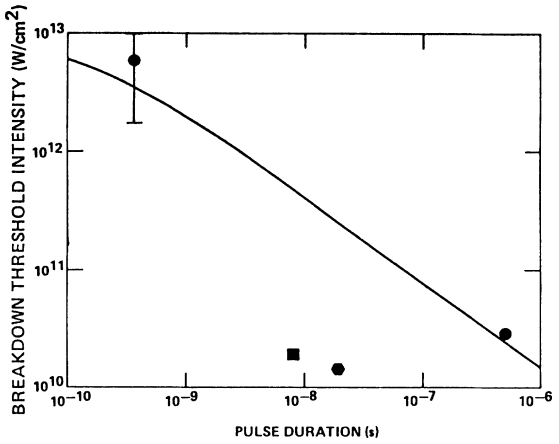

FIG. 3. Laser-induced breakdown threshold for argon, $\lambda = 0.35$ μm, $P = 1$ atm. ● Our measurements, ■ Alcock *et al.* (Ref. 5, $\lambda = 0.345$ μm), ● Buscher *et al.* (Ref. 4, $\lambda = 0.345$ μm), —— model calculations.

Multiphoton ionization of argon results in a linear growth of electrons with time, $n_e = Q\phi^5 nt$ (ϕ is the laser flux in photons s^{-1} cm^{-2}, n is the argon density), where, from the data of Kracyuk and Pashinin,[3] $Q = 1$ to 2×10^{-145} cm^{10}s^4. One expects from this buildup early in the laser pulse a sufficient concentration of electrons ($n_e \geq 10^{11}$ cm^{-3}) in the focal region so that losses by diffusion are ambipolar and can be neglected during stages 2 and 3 of the breakdown process.

In stage 2, electron cascade growth becomes the dominant mechanism. The electrons absorb laser energy by inverse bremsstrahlung collisions with neutrals. The electrons thus heated collisionally excite the 4*s* and 4*p* levels of argon (see Fig. 4). The 4*p* state is immediately photoionized (lifetime $= 10^{-11}$ s at a laser intensity of $I = 5 \times 10^{10}$ W/cm^2), whereas the 4*s* state requires the absorption of two photons and becomes ionized at the rate w_{sf} (s^{-1}) = 1.1

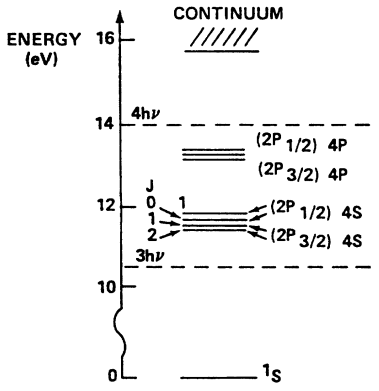

FIG. 4. Energy levels of argon.

$\times 10^{-13} I^2$. The lifetime of the 4s states, W_{sf}^{-1}, is not much shorter than the laser pulse time ($\tau_p = 500$ ns) at the intensities at which breakdown was found to occur. The excitation rates were obtained by using a Boltzmann code written by Morgan at the Joint Institute for Laboratory Astrophysics, with properly scaled excitation cross sections derived[7] from the data of Shaper and Scheibner.[8] The excitation rates for the s and p states in the intensity range $10^{10} < I < 5 \times 10^{11}$ W/cm^2, could best be fit by the relation $\nu_{s,p} = k_{s,p} n$, where k_s (cm^3/s) $= 5 \times 10^{-23} I$ and

$$k_p(\text{cm}^3/\text{s}) = \frac{2.5 \times 10^{-23} (I/10^{10})^{1.28}}{1 + (I/10^{10})^{0.28}} ,$$

I being in W/cm^2.

Stage 3 begins when electron-electron collisions are sufficiently rapid that the tail of the electron distribution function becomes populated. Population of the tail causes an increase in the inverse bremsstrahlung absorption rate. At $p = 1$ atm, this occurs for $n_e \geq 10^{13}$ cm^{-3}. Inclusion of electron-electron collisions in the Morgan code for an electron density $n_e \simeq 10^{14}$ cm^{-3} results in an absorption rate that is two to three times larger than that predicted in the absence of these collisions. At a density $n_e > 10^{14}$ cm^{-3} electron-impact ionization of the 4s excited states becomes more probable than two-photon photoionization of these states. We have modeled stage 3 by solving the kinetic equations for a two-temperature gas (T_e and T) and solving for the population of Ar, Ar*(4s), and electrons as a function of time.

The time to breakdown, which we have defined as the time to reach an electron concentration of 10^{17} cm^{-3} is obtained by adding the duration times of stages 1, 2, and 3, where, in phase 3, the electron density is allowed to grow from 10^{13} to 10^{17} cm^{-3}. The result of our calculation is shown in Fig. 3 as a solid line. Agreement with our data is seen to be quite good, although the breakdown fluxes measured at a slightly different wavelength by Buscher *et al.*[4] and Alcock *et al.*[5] are an order-of-magnitude lower. We cannot explain this discrepancy within the framework of our theoretical model. One may argue that nonlinear effects, such as self-focusing, may result in localized regions of high field amplitude where breakdown would occur on a faster time scale. Alcock *et al.*[9] have observed self-focusing in their beam during the breakdown process. Self-focusing may occur when the population of excited states is high enough to affect the third-order polarizability. It is not expected, however, to play a role during phases 1 and 2, which dominate the induction time to breakdown.

Measurements were made to determine the fraction of XeF laser-pulse energy that could be absorbed in the breakdown plasma. The data shown in Fig. 5 were obtained from time-integrated optical-pulse-transmission measurements using a large-area

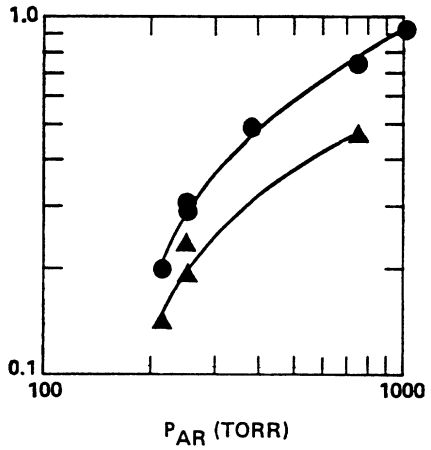

FIG. 5. Fraction of XeF laser-pulse energy deposited in argon breakdown plasma vs ambient gas pressure, $\tau_p = 0.5$ μs. ● from optical transmission measurements, ▲ from strength of blast wave.

calorimeter. The results of the time-resolved measurements showed that subsequent to breakdown the laser-produced plasmas were, in general, only partially absorbing to 0.35-μm radiation (except at the highest argon pressures studied). In order to confirm that the laser-beam attenuation observed in these optical-transmission experiments was dominated by plasma absorption rather than scattering, a separate determination of the absorbed laser energy was made by measuring the strength of the resulting blast wave. A pressure transducer (Kistler Model 211 B4) mounted 1 cm from the focus and approximately perpendicular to the optical axis was employed to measure the arrival time and amplitude of the laser-driven pressure wave. Applying the theory for a spherical blast wave in a constant density background,[10] we then used the measured transit time and shock pressure to infer the energy in the blast wave. An effective absorptance (shown as solid triangles in Fig. 5) was thus determined. The absorptance is found to be somewhat lower (by 33% to 50%) than that obtained from optical-transmission measurements. Considering energy losses by radiation and energy stored in excited (4s) states that is not immediately released, we may conclude, however, that both measurements are in substantial agreement.

The large absorption observed in the laser-produced plasmas cannot be explained on the basis of electron-ion inverse bremsstrahlung absorption. The probable dominant absorption mechanism late in the breakdown process is photoionization of highly excited states formed by the three-body recombination reaction

$$e^- + e^- + \text{Ar}^+ \rightarrow e^- + \text{Ar}^* \quad . \qquad (1)$$

The rate for reaction (1) has been calculated by Pitaevskii[11] and is sufficiently fast so as to maintain a large population of excited states which can effectively absorb the laser radiation. The absorption coefficient for a given electron temperature scales as the gas density[12] and is of the order of several inverse centimeters at atmospheric density.

In conclusion, we have found that both the model and data indicate a breakdown intensity threshold in argon at 0.35 μm that scales with pulse duration as $I_T \propto \tau^{-(0.72 \pm 0.05)}$. The model, however, is not able to explain breakdown thresholds measured in other experiments[4,5] with doubled ruby laser pulses. The present experiments have also demonstrated that substantial absorption of 0.35-μm radiation can be achieved in a laser-produced plasma.

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