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LOCAL THERMODYNAMIC EQUILIBRIUM CONDITIONS IN SUPERHIGH-PRESSURE HELIUM PLASMAS PRODUCED BY LASER ACTION

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This paper presents an analysis of gas breakdown by laser action, assuming that the final state of the laser-beam–gas interaction is described by a plasma in local thermodynamic equilibrium. A simple formula is obtained which correlates the threshold electric field with the initial gas pressure and the temperature, in agreement with experiments.

Breakdown experiments in superhigh-pressure gases[1] irradiated by a focused Q-switched laser show a dependence of threshold electric field versus initial gas pressure in the rather large interval of 20 to 2000 atm. These curves present electric-field minima, which approximately agree with the values predicted by the well-known theory of electron-impact ionization. In other kinds of experiments performed on laser-produced plasmas,[2] it has been shown that local thermodynamic equilibrium (LTE) conditions may be approximately attained in time intervals as short as the laser-pulse duration ($\simeq$65-nsec half-width), working at energy density levels of 1000 J/cm^2 (power density $\simeq 1.5\times10^{10}$ W/cm^2). Measurements made at that laser energy density show that the plasma electron temperature, the gas temperature, and the blackbody radiation temperature converge to the same limit.

This Letter presents a theoretical analysis, attempting to define a unique local temperature T for electrons, ions, neutrals, and radiation, and to deduce a relation between threshold breakdown power, temperature, and pressure that may account for the experimental curves.[1]

Different mechanisms have been proposed to explain the gas heating: the radiation-supported shock-wave process[3,4]; the traveling ionization-breakdown–wave process[5]; or a possible luminous mechanism,[4] due to the short-wavelength radiation emerging from the initial heated focal region ionizing the adjacent layers and so allowing the absorption of laser radiation. Recently, Evans and Grey Morgan[6] presented the theory that the breakdown wave may be due to aberration effects in short-focal-length lenses. It is rather difficult to make a decision about the different interpretations: It might be that one or more of the proposed mechanisms predominate with different experimental parameters. Then we assume that, independent of the possible heating and plasma-formation process, the final state of the laser-beam–gas interaction, after the peak intensity passage of the laser pulse, is described by a model consisting of a plasma in LTE conditions. We adopt a spherically symmetric distribution of the plasma properties, surrounding the initially heated focal region, which is considered as a radiation source. The observed asymmetries in the plasma formation[3-5] are evidently due to the asymmetrical laser radiation propagation relative to the focus. In a thought experiment, with a symmetric distribution of the coherent radiation, it would be possible to obtain a plasma with spherical symmetry.

Fundamental equations.—In such a plasma, it is valid to apply the well-known equations of transfer and of radiative equilibrium, integrated over frequencies[7]:

$$F_r = -\frac{c}{\kappa\rho}\frac{d}{dr}(\tfrac{1}{3}aT^4), \quad (1)$$

$$\epsilon\rho = \frac{1}{r^2}\frac{d}{dr}(r^2F_r), \quad (2)$$

where F_r is the integrated radial flux of radiation; ϵ, the total amount of energy absorbed from the laser beam in the focal region by unit mass of gas in unit time; ρ, the gas density; κ, the "Rosseland mean-absorption coefficient;" and a, the Stefan constant. As far as Eq. (1) is valid, it indicates the existence of a distribution of temperature which is studied outside the focal region, where $\epsilon = 0$ and $r > r_0$ (focal radius). Hence, integration of Eqs. (1) and (2) is readily obtained on the following basis: (1) The "hydrostatic equilibrium" condition[7] is introduced, assuming an indefinite cold external atmosphere at initial pressure p_a and temperature T_a, surrounding the laser-induced discharge. (2) The state equation for an ionized gas is used, with the Debye-Hückel pressure correction p_c for Coulomb interaction.[8] The relation p_c/p_g has been computed in the intervals from 1 to 2000 atm and 2000 to 132 000°K; a maximum value of the order 5×10^{-2} has been found (p_g is the gas pressure after heating). (3) Integration is made in a region where $T < T_l = (3p_a/a)^{1/4}$, T_l being an upper limit of temperature.[9] Finally the temperature distribution, produced by the mean laser power $\overline{P}_L$, after breakdown is expressed by

$$T = \left[\frac{15\overline{P}_L\mu M\kappa}{16\pi cak}\frac{p_a}{r}\right]^{1/5}, \quad (3)$$

valid for $r > r_0$ and for $\overline{P}_L \geqslant \overline{P}_T$, the mean threshold laser power. M is the proton mass and μ, the mean atomic weight.

Threshold breakdown conditions. —We learn from experimental work[10] that, just at the threshold, emission lines superimposed over a continuous background are observed, belonging mainly to singly charged ions. Hence, it is not unreasonable to think that the distribution (3) will contain temperatures at which there is present a high percentage of singly charged ions relative to neutrals and other kinds of charged ions, at least in the outer layers of the spark. Obviously one may expect to find a higher density of multiply charged ions in the inner and hotter layers near the focal region, but, from the point of view of this model, the plasma matter-density increases in the radial sense toward the outer and colder layers. Hence singly charged ions will predominate on the whole, showing the corresponding emission spectrum. Then we may accept that (1) at a fixed distnace $r_i > r_0$, inside the laser-induced spark, we should find a temperature T_i that shows a high relative density of singly charged ions; (2) T_i must be in the neighborhood of T_m; and (3) we define T_m as the temperature of a plasma in thermodynamic equilibrium at which the mole fraction of the singly charged ion component is maximum. We now make the following assumption: The temperature distribution which corresponds to threshold breakdown conditions and which correlates the pair (T_i, r_i) is produced by the threshold electric field $\overline{E}_T$. Hence formula (3) enables us to obtain immediately the value $\overline{E}_T$, replacing T by T_i, r by r_i, and remembering that the electric-field amplitude is given by $E_L = (11\sqrt{2})P_L^{1/2}/r_0$, expressing P_L in watts, E_L in V/cm, and r_0 in cm.

Comparison with experiments. —This approach is checked against measurements in superhigh-pressure laser-produced helium plasmas.[1] Thermodynamic properties of helium were computed as functions of temperature in the pressure interval 1-2000 atm[11]; equilibrium composition of the species He I, He II, He III, and e^- was studied; and finally the values of T_m were obtained (Table I). The κ values were calculated using the Rosseland expression,[7] introducing the specific Planck intensity and κ_ν, the absorption cross section per atom. Straightforward calculations performed for these plasmas with the assumed LTE conditions and with the indicated temperature values (Table I) have shown that absorption due to free-free transitions is only a very small fraction of that due to bound-free transitions. On the other hand, integration of the measured intensities over solid angles[12] has shown that the laser radiation scattered by the spark was negligible compared with the energy removed from the laser beam by absorption. Therefore, we concentrated our attention on calculations of the continuous absorption due to bound-free transitions from ground and excited levels of He I and He II. Then it is readily found that the weighting function, based on Kramers's approximation $(1/\nu^3)$, $R(u) = u^7e^{-u}/(1-e^{-u})^2$, $u = h\nu/kT$, has a maximum when $u \simeq 7$. This means that for temperatures near the T_m values the high frequencies have the greatest weight in the Rosseland mean. Based on this previous

Table I. Summary of characteristic values of laser-produced helium plasmas, obtained with threshold electric field. Predicted electron densities n_e correspond to temperatures T_i. The κ values were obtained as an average in temperature intervals whose lower limits are the T_i values. The corresponding variation of the experimental threshold laser power $\bar{P}_T$ is included between 1.7×10^4 and 2.2×10^5 W (last two columns).

Pressure (atm.)	$10^{-4}T_m$ (°K)	$10^{-4}T_i$ (°K)	$10^{-6}\kappa$ (cm^2/g)	$10^{-18}n_e$ (cm^{-3})	$10^{-5}\bar{E}_T$(exp) (Volt/cm)	$10^{-5}\bar{E}_T$ (Theoret.) (Volt/cm)
27.2	4.22	4.88	21.4	2.4	15.0	8.9
102	4.64	5.07	14.4	7.0	8.3	5.8
167	4.85	5.07	11.6	12.0	5.5	5.0
408	5.29	5.29	7.24	27.0	4.4	4.3
612	5.47	5.47	5.62	39.0	4.4	4.3
828	5.64	5.64	4.52	52.0	4.2	4.5
1,120	5.78	5.78	3.71	69.0	4.4	4.8
1,360	5.90	5.90	3.30	81.0	5.0	5.1
1,675	6.02	6.22	2.75	96.0	4.9	5.5
2,000	6.13	6.33	2.46	112.0	6.2	6.1

analysis, κ values were computed in large frequency and temperature intervals around T_m; as expected, a strong mean absorption was found in the interval 60-227 Å that just corresponds to the He II first absorption edge (Table I).[13] The mean atomic weight μ was also computed in the same temperature intervals and it is easily found that a good approximation is given by $\mu = \frac{1}{2}A$ (we consider a plasma with a high relative concentration of singly charged ions), A being the mass number. Theoretical values of $\bar{E}_T$ were calculated at the measured pressures from formula (3) and compared with experimental values: The best fitting was obtained with the temperature indicated in column T_i from Table I. Good agreement is observed especially at the superhigh pressures (Fig. 1); at lower pressures, it might be that the predicted electron densities are lower than those necessary to assure LTE conditions.[14,15] The theoretical minimum, 4.2×10^5 V/cm for 550 atm, shows agreement with the broad experimental one, centered at about 610 atm with a value of the threshold E field of 4×10^5 V/cm.[1]

These theoretical results may be interpreted as follows: During the first nanoseconds of the laser pulse, the free electrons in the plasma absorb the laser radiation through predominantly free-free electron-photon processes (inverse bremsstrahlung) and are thus accelerated, so beginning the ionization-cascade process. Due to the high collision rate at superhigh pressures, the electrons are cooled performing collisions with ions and neutrals, until the plasma species reaches the same kinetic temperature. This statement is confirmed by calculations made on thermalization times[14] with the values of Table I. We obtain for ions $\tau \sim 5\times10^{-10}$ sec, and for neutrals $\tau \sim 10^{-11}$ sec, for the lowest pressure.

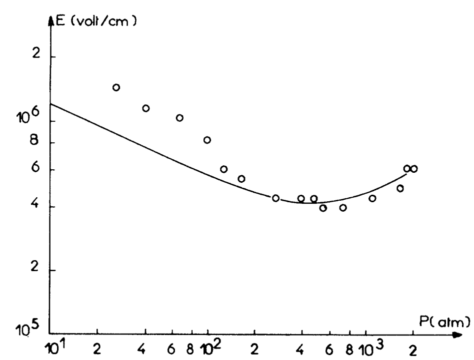

FIG. 1. Threshold electric field versus initial gas pressure. Experimental values (Ref. 1) are shown by the open circles; theoretical values (present work), by the solid line. Measured focal radius: $r_0 = 5\times10^{-3}$ cm. For r_i: Taking the value for the breakdown wave velocity $v \simeq 10^7$ cm/sec (Refs. 3, 4), after the 50-nsec half-width pulse duration (Ref. 1), the radius of the spark will be of the order of 0.5 cm; it is found from measured values and from Table I that the best fitting is obtained with a value of $r_i = 0.05$ cm. As expected, r_i value denotes a position inside the spark and it is true that $r_i > r_0$.

Hence a hot plasma in LTE conditions maintained by the laser power $\bar{P}_T$ is built up, due principally to collisional processes whose properties describe the final state of the laser-beam-gas interaction. The initial gas pressure plays an essential role, fixing the breakdown parameters. A more rigorous verification of this approach may be obtained in threshold breakdown experiments at different pressures with the aid of a high-energy laser operating in the spike mode,[16] thereby increasing the pulse duration. Use may be made of time-resolved spectrometry and Mach-Zehnder interferometry in order to measure gas, electron, and blackbody temperatures, and electron densities.

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