An Interferometric Study of CO_2-Laser-Produced Sparks

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Abstract—The expansion of sparks produced in various gas mixtures by a helical TEA CO_2 laser of modest power ($\sim$ 0.5 MW) has been examined using time-resolved photography and two-wavelength interferometry. The latter was accomplished with the aid of a synchronized ruby laser, the fundamental and second-harmonic frequencies of which illuminated a Mach–Zehnder interferometer. The interferograms have been analyzed using Abel inversion techniques and two-dimensional profiles of the electron density within the sparks obtained.

I. Introduction

In the past few years considerable effort has been made in the study of optical breakdown of gases induced by focused high-power CO_2-laser radiation. The availability of high-power laser radiation at 10.6 μ has permitted the evaluation of the frequency dependence of gas breakdown at a wavelength an order of magnitude longer than that of other lasers of sufficient power. Initial investigations of the absolute value of the threshold field for the occurrence of breakdown in the focal region and of its dependence on laser light frequency and gas pressure indicated the production of a highly ionized plasma could be explained on the basis of an electron-cascade model [1]–[3]. In this mechanism the electrons gain energy from the field by the inverse bremmstrahlung effect, and consequently produce further free electrons by ionization through collisions with gas atoms or molecules [4]. However, since the probability of direct multiphoton-ionization of the gas is much less than for the case of gases irradiated with visible wavelength radiation, and therefore does not readily explain the production of the first free primary electrons, the existence within the focal volume of particulate matter [5], residual ionization [1], [6], or absorbing foreign gases [7] was found necessary to initiate the plasma buildup. In addition, further studies of the dependence of the threshold on focal spot size and gas pressure indicate the existence of a loss mechanism during the cascade other than that of electron diffusion [5], [8]–[10]. The effects of an axial magnetic field on the radial electron diffusion during the electron cascade and the consequential reduction in the breakdown threshold have also been investigated [11], [12]. In general, these studies of the breakdown threshold with 10.6-μ radiation have particular relevance to the development of high-power high-pressure gas-laser systems where optical breakdown acts as a final limitation on the maximum energy density which can be sustained by the medium [13], [14], [8].

The development and maintenance of CO_2-laser-produced plasmas in a number of different gases have been studied for a variety of conditions. The expansion of the plasma produced by the focused, microsecond-, or submicrosecond-duration laser outputs both inside [15]–[17] and outside [18]–[22] the laser resonator has been investigated, and the production of sparks by the 3-ns pulses from the output of a mode-locked CO_2 laser reported [23]. In addition, the use of a high-power CW CO_2 laser to maintain a continuous optical discharge, initially created by a pulsed CO_2 laser in noble gases at a few atmospheres pressure, has also been demonstrated [24]–[26].

This paper describes in detail a time-resolved interferometric analysis of optical breakdown produced by a pulsed TEA CO_2 laser in air and other gases which has heretofore only been published in summary form [27]. In particular, an investigation of the conditions under which a previously observed plasma filament [18] propagates in the forward direction at later stages in the development of the plasma is described. The interferometric technique, which has already been employed in the analysis of plasmas produced by visible wavelength lasers [28], [29], utilized the fundamental and second-harmonic frequencies of a Q-switched ruby laser as sources for a Mach–Zehnder interferometer. Thus simultaneous interferograms of the plasma could be obtained at two wavelengths, and consequently, with the aid of Abel inversion techniques, it was possible to make two-dimensional maps of the electron distribution throughout the plasma at various times during its development.

II. Temporal Development of the Spark

The laser used to produce sparks in various gases was a TEA atmospheric-pressure device of the type developed by Beaulieu [30] consisting of a helical array of pin-to-pin resistively balanced 25-mm discharges. The laser resonator was approximately hemispherical and consisted of a fully reflecting Au-coated brass reflector having a radius of curvature of 5 m, and an uncoated Ge etalon 4 mm thick, having a nominal reflectivity of $\sim$77 percent. When operated with a CO_2, N_2, and He mixture of 5:5:90, respectively, the 5-mm-diameter almost-diffraction-limited beam, consisted of an initial pulse of $\sim$500-kW peak power and 200-ns full width at half power (FWHP), followed by a low-intensity tail of $\sim$50 kW which persisted

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for several microseconds. The beam was focused by an antireflection-coated Ge lens having a focal length of 39 mm in a pressure cell having NaCl input and exit ports and four quartz observation ports situated orthogonally to the beam axis. The focal-spot diameter was estimated to be $\sim 100\ \mu$. The temporal characteristics of the laser pulse were recorded with a photon-drag detector [31], and energy measurements made using a TRG cone calorimeter. The laser could also be operated without N_2 in the discharge, in which case the output consisted principally of the 200-ns-duration short pulse.

Studies of the breakdown produced in noble gases such as He, Ar, Xe, and mixtures of He and Xe, and of He and Ar, and of the molecular gases N_2 and air were made. A separate study of the development of sparks produced by a similar laser in a number of polyatomic molecular gases, and of the origin of a fluorescence halo surrounding such sparks has already been published [21]. Preliminary examination of the expansion of the sparks produced in air and other gases by a TEA laser having a peak power of 1 MW has revealed the appearance of a filament of plasma initiated sometime after breakdown propagating in the forward direction [18]. The general features of the expansion of the plasma can be seen in the streak photograph of the spark produced in N_2 at 400 torr, shown in Fig. 1. This photograph was obtained with the aid of a TRW image-converter camera observing the spark in a direction normal to the laser-beam axis. Upon the occurrence of breakdown at the focus, the plasma expands towards the lens, under the influence of the incident laser beam, with a velocity of $\sim 5 \times 10^6$ cm/s for a time of $\sim$200–300 ns, the duration of the main laser pulse. It can be seen from the degree of luminosity at the leading edge during this phase that most of the laser energy is absorbed in a thin layer of plasma propagating back toward the lens. On examination of the expansion of this luminous front as a function of time, Fig. 2, it is found that during this initial expansion phase the displacement of the front is dependent on the 0.6th power of time. This indicates that the observed expansion occurs as a result of a radiation-driven detonation wave of the type originally proposed by Ramsden and Savic [32]. It is perhaps interesting to note that this result is in close agreement with the recent results of Offenberger and Burnett, who found that sparks produced by laser pulses of power close to the breakdown threshold in low-pressure H_2 exhibited a similar behavior, whereas, for sparks produced by laser pulses of an amplitude significantly greater than the breakdown threshold, the spark initially propagated as a breakdown wave [33]. For the case of the spark shown in Fig. 1, breakdown occurred almost at the peak of the laser pulse, and consequently the latter phenomenon was not observed.

Following this initial propagation as a detonation wave for $\sim$ 200 ns, the expansion is less rapid, the displacement being related to the 0.25th power of time. This suggests that during this time the laser radiation is no longer absorbed in the thin expanding plasma boundary, but instead penetrates beyond into the center of the spark. An indication that this is the case is given in Fig. 1, where it can be seen that in the period between 400 ns and 1 μs the radiation is largely absorbed within the body of the plasma. Thus it is expected that during this time the plasma density in this region would be decreasing, and consequently the absorption length of 10.6-μ radiation increasing until a significant fraction of the laser energy is transmitted through the plasma, as has been observed in the case of sparks produced in He [18]. However, if the laser power density is sufficient it may induce the propagation of a fresh forward-going shock wave into the neutral gas, as can be seen in Fig. 1. The somewhat unexpected feature of this forward-propagating luminous boundary is its almost constant velocity. Although the laser has almost constant power ($\sim$50 kW) during this time the power density incident on the plasma filament would be expected to change by at least a factor of 4, as the plasma–gas interface is driven away from the focus. In addition it was found that, although the characteristics of this forward-going plasma filament were studied in a large number of different gases under different pressure and laser power conditions, the degree of linearity of its velocity remained the same. This is illustrated in Fig. 3, which shows the velocity of the forward-going plasma filament and its delay as measured from the time of initial breakdown, as a function of molecular mass for a constant pressure. These were determined from sparks produced in various mix-

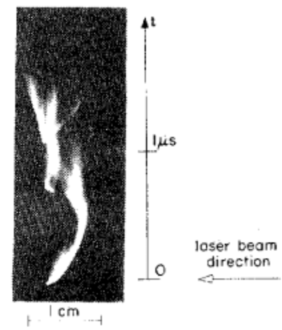

Fig. 1. Temporal development of spark produced by TEA CO_2 laser in nitrogen at 400 torr.

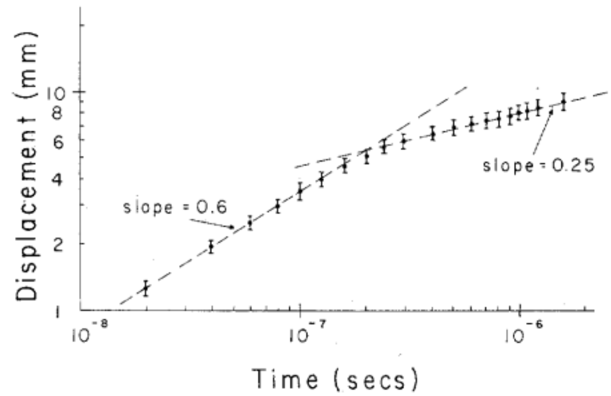

Fig. 2. Movement of the luminous front of the spark, shown in Fig. 1, as a function of time.

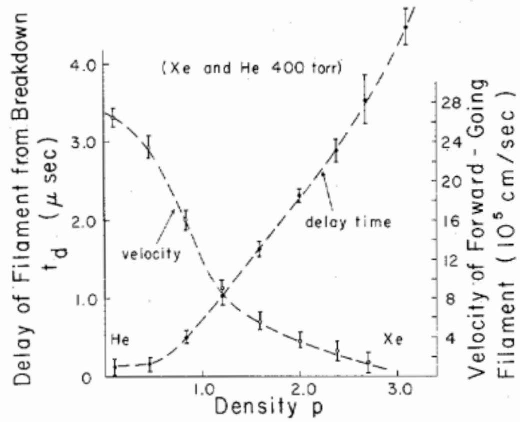

Fig. 3. Variation of the velocity of the forward-going plasma and of the time delay between initial breakdown and its onset as function of density for a fixed gas pressure of an He and Xe mixture.

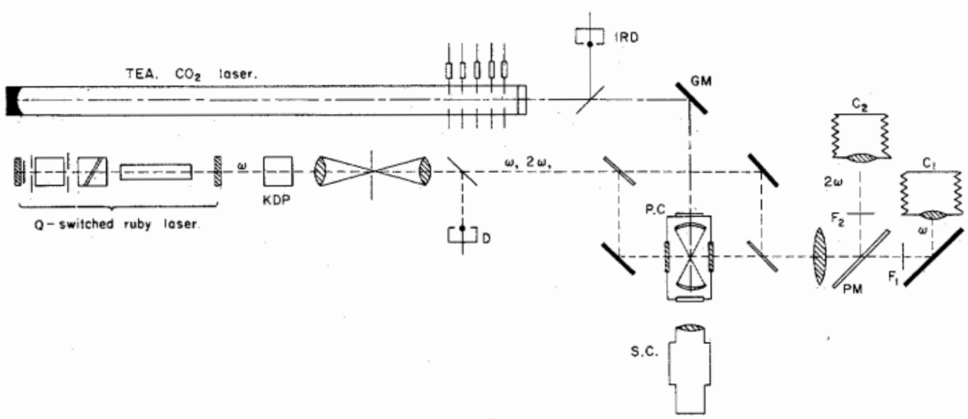

Fig. 4. Schematic of experimental setup used to obtain two-wavelength interferograms of the sparks produced by the TEA CO_2 laser. Legend: *PC*—pressure cell; C_1, C_2— identical cameras; *GM*—gold, fully reflecting mirror; *PM*—50-percent silver mirror; F_1, F_2—narrow-band filters for 6943 Å and 3472 Å, respectively; *SC*—streak camera; *D*—biplanar photodiode; *IRD*—infrared detector.

tures of He and Xe at a total pressure of 400 torr. Although the broad features of Fig. 3 are readily understood, the actual dependence of the velocity and the delay, or the "penetration time" of the laser beam in the plasma, cannot easily be explained. The velocity does not have the $\rho^{-0.33}$ dependence expected of a detonation-wave model, nor the $\rho^{-0.2}$ dependence of a blast wave. However, it is possible that the passage of the laser beam through the nearly transparent plasma may cause some modification in its characteristics. This might give rise to some progressive defocusing of the beam, resulting in a near-constant power density driving the forward-going plasma front.

III. Two-Wavelength Interferometry of CO_2-Laser-Produced Sparks

In order to determine more precisely the conditions under which the previously referred to plasma filament propagates, and to obtain two-dimensional density profiles of the plasmas produced in various gases at different times during their development, a simultaneous two-wavelength interferometric analysis was undertaken. The general experimental arrangement is shown schematically in Fig. 4. The sparks were produced, as before, in a pressure cell which was situated in one arm of a Mach–Zehnder interferometer. The latter was simultaneously illuminated by the fundamental (6943-Å) and second-harmonic (3472-Å) output pulses of a Pockels-cell *Q*-switched ruby laser. This laser has a multimode output of ~ 100 MW, with a pulse duration of ~ 20 ns at the fundamental frequency, and a second-harmonic output of ~ 10 MW produced in a phase-matched KDP crystal. However, the use of a spatial filter consisting of two lenses and a small aperture, as shown, severely limited the radiation incident on, and ensured uniform illumination of, the interferometer. Images of the spark in 6943-Å and in 3472-Å radiation were obtained with two similar cameras, a 50-percent reflecting mirror, and selective spectral filtering, as indicated in Fig. 4. The interferograms were obtained on Polaroid type-47 film, with a magnification of ~ 7, and the system had an effective spatial resolution of ~ 50 μ. In addition, streak photographs of the spark were obtained in a direction transverse to the 10.6-μ beam in order to monitor the time of occurrence of the forward-going plasma front, and also, by recording the ruby-laser light scattering off the plasma, to determine the time relative to breakdown when the interferograms were obtained.

A typical result obtained for a spark produced in atmospheric air is shown in Fig. 5. These interferograms were produced some 1700 ns after the initial breakdown, and ~ 500 ns after the initiation of the forward-going front. The latter can clearly be distinguished in the interferograms. The pointed nature of the leading front of the plasma filament is of interest, especially since it is ~ 3 mm from the focus. At this point, the beam should be ~ 0.5 mm in diameter, and yet the pointed nature of the interferogram would appear to indicate that the presence of the plasma significantly reduces the beam diameter in this region at this time. However, this feature of the forward-going plasma filament is not reproduced in all gases. As an example of this, Fig. 6 shows the development of the spark produced in argon at a pressure of 900 torr. It was found that in atomic gases, in general, the appearance of the forward-propagating plasma front was not as distinct as in air. However, the front can be distinguished from the main body of the plasma. In addition, it can be seen that at later times the freely expanding shock wave detaches from the plasma and progresses as a Taylor blast wave [34]. This behavior was noted to occur in all the gases studied, and has previously been referred to by George *et al.* [20], who observed such a "two component" plasma structure in sparks produced in helium by a TEA CO_2 laser.

The time dependence of the plasma density was obtained by producing simultaneously interferograms at two wavelengths of sparks created in several gases at different pressures. From comparison of the corresponding fringe shifts it was possible to unambiguously determine the electron density within the plasma, taking into account the contribution made by neutral particles to the local refractive index within the plasma. If S is the fringe shift produced by passage of radiation of wavelength λ through a disturbed region of thickness l then

$$S\lambda = \overline{(\mu_n - 1)l} + \overline{(\mu_e - 1)l}$$

where $\overline{(\mu_n - 1)}$ is the average change in refractive index caused by the neutral particles, and $(\mu_e - 1) \simeq -(\omega_p{}^2\lambda^2)/(8\pi^2c^2)$, where ω_p is the electron-plasma frequency. Hence

$$S\lambda = (\mu_n - 1)l - 4.46 \times 10^{-14}\lambda^2 n_e l.$$

Since the phase refractive index for the gases here considered may be represented by a single-form Cauchy equation of the type

$$(\mu_n - 1) = A(1 + B\lambda^{-2})n_n$$

where $B \ll \lambda^2$, the contribution of the neutral particles to the fringe shift may be considered to a first approximation

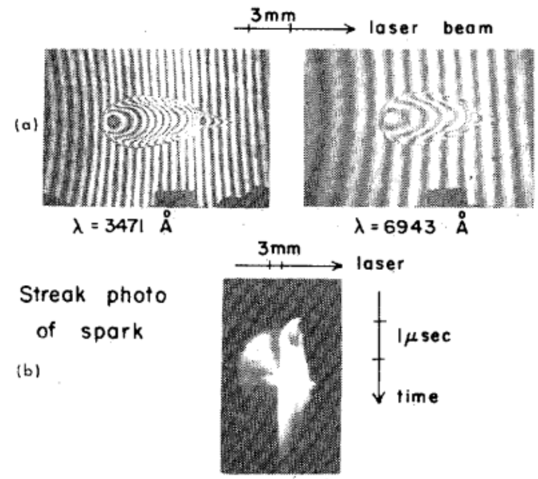

Fig. 5. (a) Interferograms at two wavelengths of CO_2-laser-produced spark in atmospheric air. (b) Corresponding streak photograph of spark. Scattered ruby-laser light indicates that interferograms were produced ~1.7 μs after breakdown.

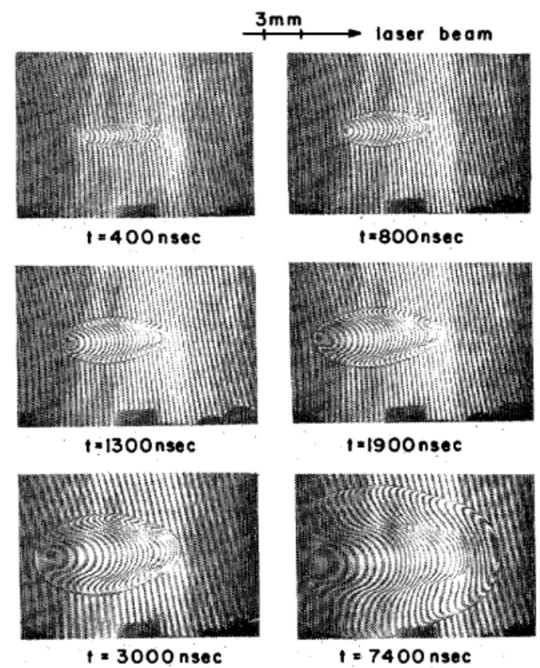

Fig. 6. Interferograms obtained at different times in the development of the sparks produced in argon at a pressure of 900 torr.

to be independent of wavelength. Hence, for the two fringe shifts S_1 and S_2 produced at different wavelengths λ_1 and λ_2,

$$S_1\lambda_1 - S_2\lambda_2 = -4.46 \times 10^{-14}(\lambda_1{}^2 - \lambda_2{}^2)\,\bar{n}_e l$$

from which the average value of the electron density ($\bar{n}_e$) can be determined. This was done for a number of sets of interferograms obtained at different times during the development of the spark. The path length l was estimated from the size of the phase disturbance, and the average electron density plotted as a function of time during the development of the spark. The case for sparks produced in argon at 150 torr is shown in Fig. 7. Maximum average

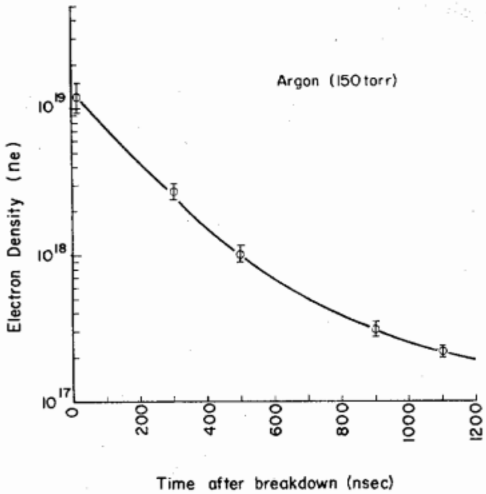

Fig. 7. Variation of the average electron density within the spark produced in argon at a pressure of 150 torr as a function of time.

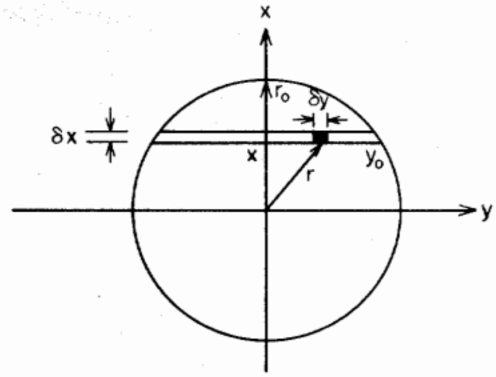

Fig. 8. Cross section of a circularly symmetric plasma, and its effect on the transverse optical path.

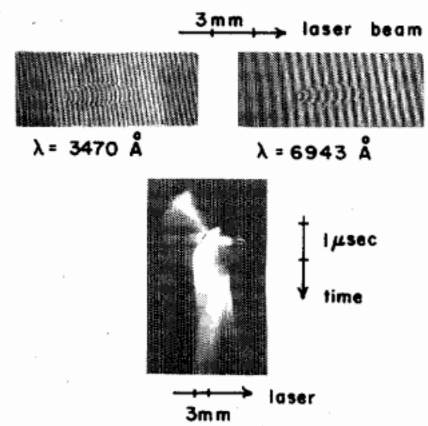

Fig. 9. Simultaneous interferograms at two wavelengths for the case of a spark produced in argon at 150-torr pressure. The corresponding streak photograph indicates that the interferograms were obtained immediately prior to the initiation of the forward-going plasma front.

electron densities above 10^{19} cm^{-3} were recorded and the low-density limit was $\sim 10^{17}$ cm^{-3}.

Moreover, considerable information on the spatial distribution of the density within the spark may be obtained from these interferograms by employing cylindrical transformation techniques. This has been done for a number of pairs of interferograms in different gases. If it is assumed that the plasma is centrally symmetric about the CO_2-laser-beam axis, then it is possible to obtain the radial electron-density distribution as a function of distance along the laser-beam axis. Assigning coordinates x and y to these axes perpendicular to the beam axis, viz., Fig. 8, then the fringe shift resulting from passage through an increment of thickness δx at distance x_0 in the y direction is

$$S(x)\lambda = 2k\lambda^2 \int_0^{y_0} n_e(r)\, dy + C$$

where $k = 4.46 \times 10^{-14}$ and C is the neutral atom contribution. Hence, for two wavelengths λ_1 and λ_2, where $\lambda_1 = 2\lambda_2 = 2\lambda$ (corresponding to the present experiment where $\lambda = 3472$ Å, the wavelength of the ruby-laser second harmonic), then

$$2S_1(x) - S_2(x) = 6k\lambda \int_0^{y_0} n_e(r)\, dy.$$

If now $N(x)$ is expressed as $[(2S_1(x) - S_2(x)]/3k\lambda$, then

$$N(x) = 2 \int_0^{y_0} n_e(r)\, dy.$$

By using Abel's transformation, this can be transformed into

$$n_e(r) = -\frac{1}{\pi} \int_0^{r_0} \frac{N(x)\, dx}{(x^2 - r^2)^{1/2}}.$$

A numerical solution to this transform has been calculated by Bockasten [35]. Several sets of interferograms were analyzed using this method, and, as an example, the two-dimensional electron-density distribution in the spark produced in argon at 150-torr pressure (Fig. 9) is shown in Fig. 10. These interferograms are of particular interest since they were observed ~ 300 ns after the occurrence of breakdown and immediately prior to the initiation of the forward-going plasma front. Although the technique does not permit electron densities much below 10^{17} cm^{-3} to be resolved, the broad features of the distribution are clearly visible. The density as expected is a maximum immediately behind the backward-moving plasma front and, from Fig. 11, which shows the peak electron density as a function of axial distance, decays almost exponentially with distance along the laser-beam axis. In addition, there is clear indication from Fig. 10 that, in the radial direction, there is a pronounced reduction in the electron density in the central region of the plasma immediately behind the backward-propagating plasma front. It is possible that the small peak, which occurs on axis in this region, may be an artifact of the transformation process, since in this region the transformation is most sensitive to local perturbations in $N(x)$ [35]. The calculated errors of these points are shown in Fig. 11, where it can be seen that they are greater

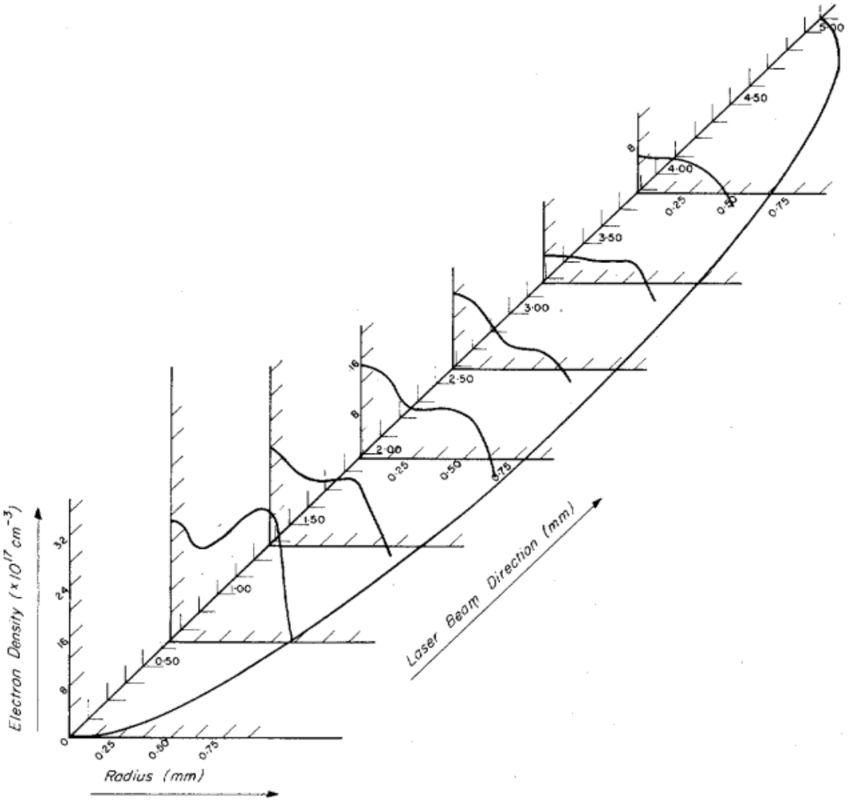

Fig. 10. Two-dimensional electron-density profile obtained from the interferograms of Fig. 9. The boundary shown is that of plasma–neutral gas interface as deduced from the interferograms.

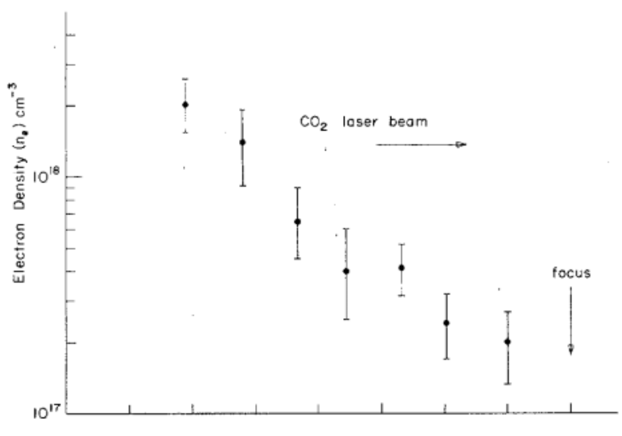

Fig. 11. Variation of the peak axial electron density as a function of axial distance through the spark produced in argon at a pressure of 150 torr some ~ 300 ns after breakdown.

than the amplitude perturbations produced on axis on the profile of Fig. 10. In addition, these density peaks were not observed on all the interferograms analyzed. However, the general feature of a broad minimum in the radial-density profile was common to all the interferograms, and therefore the existence of such can be relied upon to a greater extent. In addition, in this region, at larger values of r, the transformation is less sensitive to local variations in $N(x)$ resulting from the fringe measurement [35]. Unfortunately, the measurement technique will not permit the determination of the electron-density profile within the forward-moving plasma front. It was deduced that the electron density in most of this plasma region must be $<10^{17}$ cm^{-3}.

IV. Conclusions

The transform technique described is particularly suitable for the analysis of laser-produced plasmas which usually can be assumed to be axially symmetric. However, the small dimensions of the plasma studied in this experiment inhibited examination of the plasma distribution below electron densities of $\sim 10^{17}$ cm^{-3}. This has unfortunately meant that the density distribution within the forward-moving plasma front has not been accurately determined. In addition, although the analysis has established the existence of a minima in the radial-density distribution of the plasma, it has not led to a clear explanation of the linearity of the forward-moving plasma front. Modification of the character of the laser beam by this nonuniform plasma would still appear to be the most likely reason for the observed velocity dependence, but a greater knowledge of the density distribution and considerable numerical analysis would probably be required to verify this hypothesis.

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