

representative samples are sufficient.

A further study of Reactions (1) and (2) as a function of primary ion energy has been made using the method described above and the 6.5-V dissociative attachment peak for production of H^- or D^- . The full width at half-maximum of the primary ion energy distribution was kept to less than 0.2 eV by the application of a modulated retarding potential difference to the electron beam and the use of phase-sensitive ion-current detection. Variation of the electron energy over the main dissociative attachment peak produced primary ion energies in the range 1.2 to 2.3 eV. The results will be described in more detail in a later publication but indicate that though the cross section increases rapidly with decreasing energy,¹¹ only about 30% of the isotope effect above may be accounted for by the velocity differences between the H^- and D^- primary ion energy distributions.

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¹J. A. Rutherford and B. R. Turner, *J. Geophys. Res.* **72**, 3795 (1967). T. L. Bailey, in *Proceedings of the Second International Conference on Physics of Electronic and Atomic Collisions, Boulder, Colorado, 1960* (W. A. Benjamin, Inc., New York, 1960), p. 54. K. Peuckert-Kraus, *Ann. Physik* **18**, 288 (1966).

²F. C. Fehsenfeld, E. E. Ferguson, and A. L. Schmeltekopf, *J. Chem. Phys.* **45**, 1844 (1966).

³J. F. Paulson, in *Ion Molecule Reactions in the Gas Phase: A Symposium*, Advances in Chemistry Series 58 (American Chemical Society, Washington, D. C., 1966), Chap. 3. J. G. Dillard and J. L. Franklin, *J.*

Chem. Phys. **48**, 2349, 2353 (1968), have measured rates of some negative-ion-molecule reactions by a method similar to that used by the present authors. Their results appeared after this work had been completed.

⁴R. A. Challinor and R. A. Duncan, *Australian J. Phys.* **20**, 633 (1967). See also Ref. 3. R. K. Curran, *Phys. Rev.* **125**, 910 (1962), has examined the formation of NO_2^- in a mass-spectrometer source region by charge exchange from several negative ions. A very low extraction field was used; however, the initial kinetic energies of the primary ions and their energy distribution during the reaction period were not well known. A drift-tube mass-spectrometer study of negative-ion-molecule reactions reported by J. L. Moruzzi and A. V. Phelps, *J. Chem. Phys.* **45**, 4617 (1966), also suffers from uncertainties in the knowledge of the primary ion energy distribution.

⁵R. N. Compton and L. G. Christophorou, *Phys. Rev.* **154**, 110 (1967).

⁶First reported by E. E. Muschlitz and T. L. Bailey, *J. Phys. Chem.* **60**, 681 (1956).

⁷L. G. Christophorou, R. N. Compton, G. S. Hurst, and P. W. Reinhardt, *J. Chem. Phys.* **43**, 4273 (1965).

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⁹P. J. Chantry and G. J. Schulz, *Phys. Rev. Letters* **12**, 449 (1964), and *Phys. Rev.* **156**, 134 (1967).

¹⁰This description is accurate if the dissociation products are not excited. It may be used in the present case since G. J. Schulz, *J. Chem. Phys.* **33**, 1661 (1960), has shown that the OH radical produced in $e + H_2O \rightarrow H^- + OH$ is not significantly excited.

¹¹Reactions (1) and (2) are exothermic by ~ 0.4 eV and may, therefore, take place at zero primary ion energy.

CREATION OF A SPARK BY A SINGLE SUBNANOSECOND LASER PULSE*

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The production of a spark by focusing a single subnanosecond laser pulse in air and other gases has been investigated. Streak photographs reveal that the plasma development is significantly different from that observed with conventional Q-switched laser pulses. Preliminary studies of the pressure dependence of the breakdown threshold in argon and nitrogen are reported.

Although the sparks produced by focusing high-intensity laser pulses with durations greater than several nanoseconds have been studied extensively, the production of a plasma by a single subnanosecond laser pulse has not yet been reported. There is considerable interest in extend-

ing the investigation of laser sparks to the region of gigawatt peak powers and subnanosecond pulsewidths, since cascade ionization, including both single- and multiple-photon interactions, and direct multiphoton ionization may play competing roles in the development of the plasma.

In this paper we wish to report the observation and preliminary investigation of a spark created by a single pulse having a duration of $\sim 10^{-11}$ sec. The pulse was obtained by cavity dumping a mode-locked laser in a manner similar to that which has already been reported.^{1,2}

A schematic diagram of the experimental arrangement is shown in Fig. 1. A Nd:glass laser consisting of an 8-in. long, $\frac{5}{8}$ -in. diam, Brewster-ended rod was simultaneously Q switched and mode locked by means of Kodak 9740 saturable dye. The optical resonator was formed by two wedged, dielectric mirrors separated by a distance of 120 cm and having reflectivities of 99 and 75% at the laser wavelength. A sample of the output from the 75% reflectivity mirror was detected by an ITT F4000 photodiode and used to trigger a high-voltage, cold-cathode thyatron switch. The resulting voltage step, which had a rise time of ~ 2 nsec, was applied to a Pockels cell situated within the cavity, and provided that the pulse reached the cell when the quarter-wave voltage was applied to it, most of the radiation was coupled out of the cavity by the Glan-Thompson polarizer placed between the laser rod and the Pockels cell. In the majority of cases, the laser output consisted of a single pulse.

The intensity of the prepulse background radiation, due to reflection losses from the polarizer, was of the order of 1/50th of the intensity of the main pulse peak and was further reduced by passing the beam through a second dye cell. The

pulse then entered an amplifier stage, a 9-in. $\times \frac{3}{4}$ -in.-diam Nd:glass rod, resulting in a peak power gain of ~ 8 . In this manner energies, as measured with a calibrated photodiode, of the order of 100 mJ were obtained.

Measurements of the pulse width for each laser shot were made by means of the two-photon fluorescence technique³ using an ethanol solution of Rhodamine 6G. The train of pulses transmitted through the 75% mirror, prior to the Pockels cell being switched, was split into two beams which were made to collide within the fluorescent dye. This method indicated that the length of the individual pulses was typically 10^{-11} sec. Since there is no apparent way by which the switching operation can increase the pulse duration, this value may be taken as an upper limit for the single pulse coupled out of the cavity.

Sparks were produced in air, nitrogen, and argon by focusing the beam within a pressure cell with a lens of 2-cm focal length. The development of the sparks was observed by means of a Space Technology Laboratories image-converter camera, and the use of a 1-mm slit parallel to the direction of the laser beam resulted in a time resolution of the order of 0.5 nsec. The streak photographs revealed that breakdown occurred at the time of the main pulse, no observable effect being produced by the small amount of background radiation which preceded it. The photographs obtained in all three gases investigated were quite similar and a typical result is shown

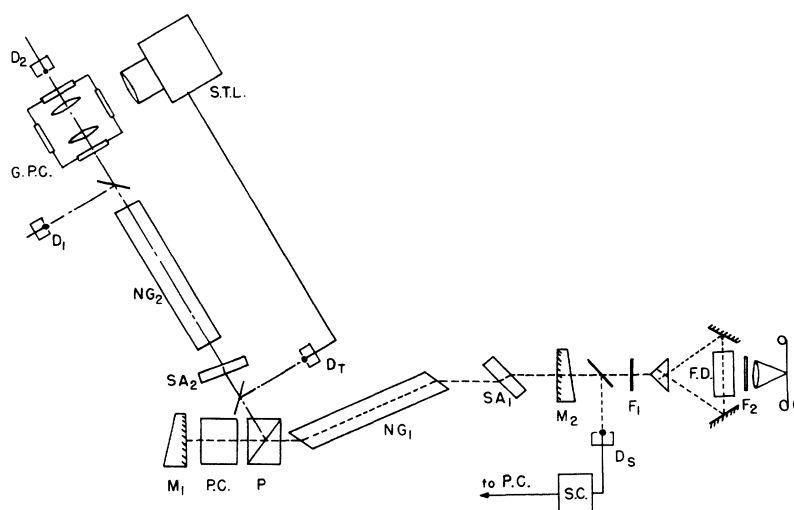


FIG. 1. Schematic diagram of the experimental arrangement used for the observation of sparks produced by a single subnanosecond laser pulse. NG, Nd:glass rods; SA, saturable absorbers; M_1, M_2 , dielectric mirrors; P.C., Pockels cell; P, Glan-Thompson polarizer; SC, fast high-voltage switch; D_5, D_4, D_1, D_2 , photodiodes; G. P. C., pressure cell; F_1, F_2 , filters; F. D., fluorescent dye; C, camera; STL, image-converter streak camera.

in Fig. 2(a), where it can be seen that the development of the spark is significantly different from that observed when a conventional Q-switched laser pulse is used.⁴ In all cases the photographs revealed the existence of the following distinct regions: There is a central luminous core whose initial development could not be resolved by the streak camera and which then does not appear to increase appreciably in diameter; its luminosity persists for several tens of nanoseconds and in many photographs the core appears to be hollow. From the photographs it can be seen that the initial radius of the central core is less than 0.1 mm. Situated outside this region is a luminous shell which expands initially with a velocity of $\sim 2 \times 10^8$ cm/sec, reaches a maximum diameter in ~ 1 nsec, and then collapses with a velocity of $\sim 2 \times 10^7$ cm/sec. The shell was surrounded by a weakly luminous region, which expands initially with a velocity exceeding 4×10^8 cm/sec corresponding to the temporal resolution of the streak camera and decays thereafter in a few nanoseconds. A possible explanation for this luminosity is the excitation or ionization of atoms by short-wavelength radiation emitted by the central core. Additional streak photographs taken with the slit normal to the laser beam revealed the development in the transverse direction to have the same features.

The effect of a second mode-locked pulse incident on the spark, approximately 8 nsec after its formation, is shown in Fig. 2(b). The central core expands asymmetrically in the direction of the incident beam and its diameter increases by ~ 0.2 mm in a time less than the resolution of the camera. At the same time a second luminous shell and surrounding region of weak luminosity is produced.

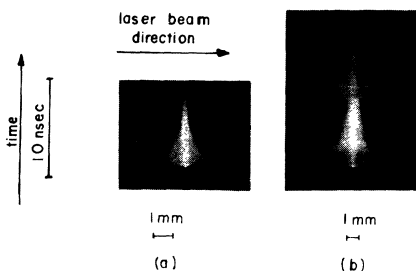


FIG. 2. (a) Streak photograph of spark in nitrogen, at a pressure of 3300 mm Hg, produced by a single subnanosecond laser pulse. (b) Streak photograph of spark in air, at atmospheric pressure, produced by two subnanosecond laser pulses.

Photodiode measurements in front of and behind the focus revealed that approximately 80% of the incident beam was transmitted through the spark. The assumption that much of the remaining 20% was absorbed by the plasma indicates that an energy of $\sim 10^{17}$ eV was released within the focal volume.

From the measured beam divergence of 2 mrad (full width at half-power) the calculated breakdown threshold in air at atmospheric pressure is $\sim 3 \times 10^{14}$ W/cm². This exceeds by more than three orders of magnitude the threshold observed with laser pulses having a duration of a few tens of nanoseconds.⁵ Although there is insufficient evidence to conclude that the production of the plasma is governed by a single-photon cascade ionization process, the present result is not inconsistent with the theory of Zel'dovich and Raizer⁶ which predicts a breakdown threshold inversely proportional to pulse length.

Measurements of the minimum power density required for breakdown in nitrogen and argon over the pressure range 500-6000 Torr are plotted in Fig. 3, where the principal error arises from the inaccuracy in measurement of the pulse duration. It can be seen that the pressure dependence of the breakdown threshold is similar to that obtained with nanosecond pulses. In view of this it seems unlikely that multiphoton ionization can be solely responsible for the production of the plasma. The conditions under which this can

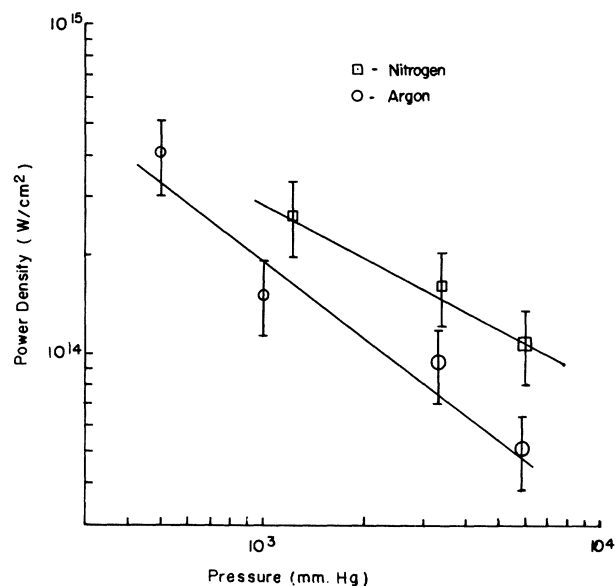


FIG. 3. Pressure dependence of breakdown threshold power density in nitrogen and argon.

occur have been examined recently by Bunkin and Prokhorov⁷ who consider only the relative importance of multiphoton ionization and a single-photon inverse bremsstrahlung cascade. However, their analysis cannot be applied directly to the gases studied in the present experiment since the measured power densities are well above the predicted threshold for multiple-photon bremsstrahlung processes.

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EFFECT OF COLLISION BROADENING UPON MAGNETIC RESONANCE IN A He-Ne LASER*

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The optical saturation of atomic laser transitions depends upon two linewidths Γ_1 and Γ_2 which have not been independent observables in previous experiments. We have obtained an independent measurement of the linewidth Γ_1 while observing magnetic resonance in a stimulated-emitting state.

Bennett, Chebotayev, and Knutson¹ determined the cross sections for He-Ne and Ne-Ne collisions from the spontaneous-emission linewidth of the 0.633- μ m transition in neon. The observed collisional contribution to the linewidth of the atomic transition was $(185 \pm 54)p_{\text{He}} + (870 \pm 370)p_{\text{Ne}}$ MHz, with the helium and neon partial pressures expressed in Torr. The collisional contribution to the linewidth has also been measured in stimulated emission by observing the optical saturation of atomic laser transitions.²⁻⁴ However, the optical saturation of an atomic laser transition depends upon two linewidths Γ_1 and Γ_2 , which are analogous to T_1 and T_2 relaxation times in solids. It has been necessary in previous experiments to make some assumption regarding the relative values of Γ_1 and Γ_2 in the data analysis, and the agreement between the collisional contribution to the linewidths deduced from these stimulated-emission experiments and the spontaneous-emission value reported by Bennett, Chebotayev, and Knutson depends strongly upon the assumptions made. In this Letter we show that in a magnetic resonance experiment involv-

ing a stimulated-emitting state, the linewidth Γ_1 may be measured independently of Γ_2 , and we report the measurement of Γ_1 . Our result confirms the conclusion by Bennett, Chebotayev, and Knutson¹ that the phenomenologically treated collision-dependent linewidth must contain relatively large collision contributions.

Phenomenological modifications of the Lamb theory of an optical maser⁵ to account for atomic collisions have been introduced by several authors.²⁻⁴ The laser intensity is given by the ratio of α , the "linear gain coefficient," to β , the saturation parameter. In the He-Ne laser, collision-dependent contributions to the atomic linewidths primarily modify β ; they have little effect upon the Doppler-broadened "linear gain coefficient" α . The saturation parameter may be written²

$$\beta = C \left(\frac{1}{\Gamma_1^2} + \frac{\Gamma_2/\Gamma_1}{\Gamma_2^2 + (\omega - \nu)^2} \right), \quad (1)$$

where C contains the square of the matrix element of the atomic transition, other physical and atomic constants, and an atomic lifetime depen-

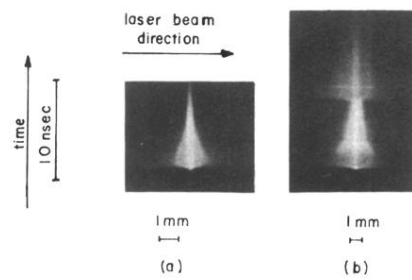


FIG. 2. (a) Streak photograph of spark in nitrogen, at a pressure of 3300 mm Hg, produced by a single subnanosecond laser pulse. (b) Streak photograph of spark in air, at atmospheric pressure, produced by two subnanosecond laser pulses.