Even if the pre-exponential factors of the present theory are wrong the "dynamical scaling" re $sult^{12}$ (4) and the critical slowing down can be checked.

The methods used in our theory near T_c will be applied' also to the discussion of (homogeneous) nucleation far below T_c .

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New Observations of Dielectric Breakdown in Air Induced by a Focused Nd^{3+} -Glass Laser with Various Pulse Widths

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Dielectric breakdown in air induced by a train of subnanosecond laser pulses was found to occur as a sequence of strong point explosions, with each pulse producing breakdown at a different spot. The angular dependence of the scattered laser light was found to be due to the reflection and diffraction from the breakdown spots which were approximately spherical with diameters of about 9μ m. For atmospheric breakdown, analysis of results indicates that a plasma density as high as 7×10^{20} electrons/cm³ was created in the breakdown region,

This Letter reports some new observations of dielectric breakdown in air induced by a focused Nd'+-glass laser with various pulse widths. While it has been eight years since the first observation of optical dielectric breakdown in air,¹ many important aspects of the phenomenon remain to be understood. Most notable among them are the angular dependence of the scattered light at the laser frequency' and the dependence of the threshold power on the pulse width.³ Using laser pulses with a width of I nsec or shorter, we have found that the scattering pattern can be explained quantitatively in terms of the reflection and diffraction from plasma "bubbles" created in the breakdown region. At atmospheric pressure, these bubbles are about $9 \mu m$ in diameter and have a

plasma density as high as 7×10^{20} electrons/cm³. Of particular interest is the observation that with a train of such pulses each individual pulse induced breakdown at a different spot in air. The evolution of these breakdown spots was found to be characteristically similar to that of the shock wave generated by a strong point explosion.

The experiments were performed with a Nd^{3+} glass laser with intracavity acoustic modulation (Fig. 1). ^Q switching was achieved with a saturable-dye cell (or a rotating prism), resulting in a train of pulses about 20 psec (or 1 nsec) in duration and separated by 12.5 nsec, the round-trip transit time of the cavity. With the saturabledye cell (rotating prism), the half-width of the train is about 200 nsec (100 nsec). In both cases,

FIG. 1. Schematic of the experimental setup.

the output beam from the laser was about 2 mm in diameter, and was focused with a 3.2-cm-focal-length lens to induce dielectric breakdown in air. The energy per pulse at the threshold for breakdown was about 3 mJ in both cases. Study of the transmitted and scattered light indicates that less than half of this energy was transmitted through the breakdown region, and about 25% scattered in all directions.

Figure 2 shows photographs of the breakdown region taken with radiation at the laser frequency and at right angles to the laser beam. Figures 2(a) and 2(b) were obtained with trains of 20-psec and 1-nsec pulses, respectively. In both cases, the breakdown occurred in discrete spots extending over a distance about 0.3 cm in length. By blocking a selected portion of the magnified image of the breakdown region and observing the time-resolved intensity on a Tektronix model No. 519 oscilloscope, it was established that each spot was generated by a different pulse in the train and that the later the pulse was in the train, the closer the corresponding spot was to the laser. The spacing between the spots was found to decrease from 0.02 cm in the focal region to 0.015 cm toward the laser. The spot size is estimated to be about 10 μ m in diameter, the measurement being limited by the resolution of the photographic process. Figure 2(c) was obtained with the 100-nsec Q-switched pulse without acoustic modulation, and is similar to that reported previously.³

The angular dependence of the scattered light at the laser frequency is shown in Fig. 3. The scattered light was detected with a photodiode placed 17 cm away from the focal point and subtending a solid angle of approximately 0.7×10^{-4} sr. With the incident laser radiation polarized either in the plane of scattering or perpendicular to it, the scattered light was found to be completely polarized as reported previously,⁴ and was strongest near the forward direction. For laser light polarized in the plane of scattering, a minimum exists near 105' from the forward direction. The data in Fig. 3 were taken with

FIG. 2. Photographs of the breakdown region (a) with a train of 20-psec pulses, (b) with a train of 1-nsec pulses, and (c) with a 100-nsec Q -switched pulse.

pulses of 1 nsec duration. The angular dependence with longer (100 nsec) and shorter (20 psec) pulses was qualitatively the same both in terms of the existence of a minimum near 105 for the laser light polarized in the plane of scattering.

One possible explanation for the results in Fig. 2 is that breakdown occurs as a sequence of strong explosions induced by the individual laser pulses: With subnanosecond excitation, the expansion of the breakdown region is negligible

FIG. 3. Plots of the scattered light as a function of the scattering angle for both polarizations. The solid curves are the plots of Eq. (2) with $d=9.3 \mu m$ and n =0.7-0.45i . The error bars indicate the experimental uncertainties.

during the span of the excitation, $^{\rm 5}$ so that these explosions can be considered as point explosions. The first explosion occurs where the energy density exceeds the threshold for the development of an electron cascade,⁵ and sends out a spherical shock wave. The ionization products associated with the shock front then serve as the nucleus for breakdown for the next pulse. This process repeats progressively for all the subsequent pulses until the energy density of the pulse is too weak to produce a breakdown spot even in the presence of ionization.

For a point explosion the radius R of the shock front varies with the time t as

$$
R \approx (E/\rho_0)^{1/5} t^{2/5},\tag{1}
$$

where E is the energy absorbed per pulse and ρ_0 $= 1.3 \times 10^{-3}$ g/cm³ is the density of air at standard conditions. Equation (1) states that R is relatively insensitive to the amount of absorbed energy and to the initial density. With $t = 12.5$ nsec and $E = 1.5$ mJ, the value of $R = 0.018$ cm predicted from Eq. (1) compares favorably with the observed spacing of 0.02 cm in the focal region. When account is taken of the decrease in energy density and the resulting decrease in the energy absorbed per pulse, Eq. (1) is also consistent with the observed small decrease in spacing away from the focal region.

It is possible to account quantitatively for the angular dependence in Fig. ² in terms of the scattering from the breakdown spots if the latter are approximated as spheres with an average refractive index n . It is well known in geometrical optics that scattering from spheres large compared to the wavelength can be regarded as the sum of that arising from diffraction and that from reflection. The former is important only near the forward direction and is independent of the state of polarization of the light beam. Within this approximation of geometrical optics, the intensity of the scattered light, $I_s(\theta)$, is given by⁶

$$
I_s(\theta) \propto x^2 I_0 [J_1^2(x\theta)/\theta^2 + r_i^2(\theta)], \qquad (2)
$$

where I_0 is the intensity of the incident laser beam with wavelength λ ; $x = \pi d/\lambda$, d being the diameter of the scattering sphere; θ is the scattering angle; $J_1(x \theta)$ is the Bessel function of order 1; and $r_i(\theta)$ is the Fresnel reflection coefficient appropriate for light waves polarized either in or perpendicular to the plane of reflection and reflected into the direction θ from a large sphere with refractive index n . According to this equa-

tion, the width of the forward lobe is determined by the size of the scattering sphere, whereas the general shape of $I_s(\theta)$ away from the forward direction is determined by the refractive index of the sphere through the Fresnel reflection coefficients. To a first approximation, the value of the real part of n may be deduced from the location of the minimum in $I_s(\theta)$.

In Fig. 3, the solid curves are the plots of Eq. (2) with $d = 9.3 \mu m$ and $n = 0.7 - 0.45i$. The experimental data for both polarizations are consistent with our geometrical optical approximation. ' The value of $d = 9.3 \mu m$ also compares favorably with that deduced from direct photographic evidence. The value of the real part of $n = 0.7-0.45i$ corresponds to an electron density⁸ of 7×10^{20} cm^{-3} , and is in agreement with the previously⁹ deduced average density of 3×10^{20} cm⁻³ when proper account is taken of the over-density near the shock front and the absorption within the spherical region. The value of $-0.45i$ for the imaginary part of n corresponds, in the case of predominantly electron-ion collisions in a fully ionized plasma, ' to an electron temperature of about 4 eV. This temperature is much lower than the maximum temperature of 60 eV deduced from the recombination radiation spectrum in the soft x-ray region,⁹ but is consistent with that deduced from the observed broadening of the 'deduced from the observed broadening of the
scattered laser radiation.^{9,10} This apparent discrepancy could be explained, as has already been noted, ' if the scattered laser radiation is from electrons in a colder region. For a strong explosion, it is well known that the temperature of the ionized product increases very rapidly from behind the shock front to the center. Since the scattered laser radiation is primarily due to electrons near the shock front, the change in electron temperature from 4 eV near the shock front to 60 eV near the center is consistent with this characteristic of a strong explosion.

We have also studied the threshold energy density for the development of an electron cascade as a function of the duration of the laser beam. The establishment that short pulses in a train break down at discrete spots makes it meaningful to compare the corresponding threshold energy per pulse with that of a Q -switched laser pulse.³ Figure 4 shows the threshold energy density per pulse obtained with pulses of various widths. The pulsewidth was varied by changing the cavity length and the acoustic modulation. Within the experimental uncertainty of about 30%, the threshold energy density W was found to be de-

FIG. 4. Plot of threshold energy density for breakdown in air as a function of pulse width. The solid curve is a plot of Eq. (3) with $\omega_c = 1.5 \times 10^9 \text{ sec}^{-1}$.

scribable by the phenomenological relation

$$
W = A\omega_c \Delta t / [1 - \exp(-\omega_c \Delta t)], \qquad (3)
$$

where A is a constant, Δt is the pulse width, and $\omega_c = 1.5 \times 10^3$ sec⁻¹. Equation (3) states that the threshold energy density ^W is constant for short pulses with $\omega_{n}\Delta t \ll 1$, and that the threshold power density $W/\Delta t$ is constant for long pulses with $\omega_c \Delta t \gg 1$. Similar dependence on pulse width was also observed in dielectric breakdown in the mialso observed in dielectric breakdown in the m
crowave regime.¹¹ Note that Eq. (3) can be inferred from the rate equation for the electron energy¹² under the assumptions that the threshold for breakdown corresponds to the attainment of certain electron-energy values within a given duration of the excitation, and that the rate of energy loss is proportional to the electron energy. For breakdown in air, the electron-energy losses may be due to diffusion of the electrons away from the focal region, elastic collisions with the air molecules, and the excitation and dissociation of these molecules. Under the experimental conditions of Fig. 4, the rate of energy loss is estimated to be about 1.3×10^8 sec⁻¹ for diffusion away from the focal region, and about 2×10^8 sec⁻¹ for elastic collision. Both of these loss mechanisms are too slow to account for the observed rate of 1.5×10^9 sec⁻¹. The dynamics of energy loss due to excitation and dissociation remains to be understood, but it appears from the

above discussion that they play a major role in the breakdown process.

The observations reported in this Letter should find use in the study of shock waves and in the simulation of strong explosions involving release of high energy densitites. However, the fact that high electron density may easily be created in the breakdown region implies that a large fraction of the incident energy will be reflected from the breakdown region, thus imposing an ultimate limit on the amount of energy absorbable from the laser beam.

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FIG. 2. Photographs of the breakdown region (a) with a train of 20-psec pulses, (b) with a train of 1-nsec pulses, and (c) with a 100–nsec $Q\!-\!$ switched pulse.