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Avalanche Ionization and the Limiting Diameter of Filaments Induced by Light Pulses in Transparent Media*

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A limiting intensity is shown to exist for light propagation in transparent liquids and solids. In pure bulk materials it is determined by avalanche ionization over a wide range of pulse durations, wavelengths, and band gaps. The ionization rate per unit time is deduced from the thickness dependence of the dc breakdown. The negative real part of the index of refraction of the carriers stabilizes the size of self-focused filaments.

The investigations of laser-induced breakdown have been very numerous, but in most cases the breakdown was preceded by self-focusing, or was initiated by absorbing inclusions.[1,2] Recently, experiments have been performed on damage in alkali-halide crystals with laser pulses for which the maximum power was 1 or 2 orders of magnitude less than the critical power for self-focusing.[3,4] A reproducible field strength, characteristic of the *bulk* material, could be established. The effect of impurities and inclusions was eliminated by the small scale of volume elements and time intervals in which the breakdown occurred. Data taken with pulses from a TEA CO_2 laser established the following relationship: $E_{\mathrm{rms}}{}^{\mathrm{br}}(10.6\ \mu\mathrm{m}) = (1.25 \pm 10\%)\ E_{\mathrm{dc}}{}^{\mathrm{br}}$ for eleven different alkali halides. [$E_{\mathrm{rms}}{}^{\mathrm{br}}(10.6\ \mu\mathrm{m})$ is the rms breakdown field at the 10.6-μm wavelength and $E_{\mathrm{dc}}{}^{\mathrm{br}}$ is the dc breakdown field.] The precise value of the factor 1.25 is not important since the dc breakdown field is known[5] to be somewhat sensitive to the experimental technique used to measure it. A similar relationship exists for data[4] taken with a Nd:YAlG laser (1.06 μm) and a ruby laser (0.68 μm). It should be noted that the breakdown field changes almost a factor of 5 in going from NaF to RbI. Furthermore, the breakdown field strength is about the same at all wavelengths from dc up to the visible red. These data strongly suggest that the breakdown mechanism in the laser pulses is the same as for dc breakdown. This mechanism is avalanche ionization.

The nature of dc electric breakdown has been studied in a quantitative fashion for hot-electron phenomena in semiconductors.[6] In reverse-biased p-n junctions, avalanche ionization has been observed.[7] The same mechanism is responsible for electric breakdown in alkali halides.[5] In particular, the dependence of $E_{\mathrm{dc}}{}^{\mathrm{br}}$ on sample thickness corroborates this conclusion.[8-10]

In an avalanche, a free electron becomes heated in the high electric field until it has sufficient energy to ionize a second one. These two repeat the process, and the electron number N grows exponentially as the electrons drift from cathode to anode, $N = N_0 \exp[\alpha(E)t]$, where $\alpha(E)$ is the rate of ionization per unit time per electron and N_0 is the number of starting electrons. If in the available time the avalanche grows to critical proportions, then breakdown takes place.[11] The available drift time $T = x/v$, where v is the drift velocity and x is the thickness between electrodes. Then the critical multiplication number M_c can be written

$$M_c = \exp[\eta(E)x], \tag{1}$$

where $\eta = \alpha/v$. For extremely thin samples the breakdown fields become so high that another mechanism, Zener tunneling, takes over. This transition to tunneling breakdown has been observed in Zener diodes with extremely thin p-n junctions.[7]

The thickness dependence, observed [8-10] for the dc breakdown field in NaCl, can, in fact, be used to estimate the ionization coefficient. Inverting Eq. (1) gives $\eta = x^{-1} \ln M_c$. For a given breakdown field x, the sample thickness is known; therefore, the only unknown on the right-hand side is M_c. Because it appears under the logarithm, only a crude estimate of M_c is sufficient to given reasonable values for η. Thus, the experimental thickness dependence of the breakdown field was used[12] to obtain the ionization coefficient η versus E, curve a in Fig. 1.

These concepts are also applicable to optical electric fields. In a high-power laser beam there are always a small number N_0 of free electrons, available from thermal ionization of shallow traps, multiphoton ionization, thermionic emission from specks of dirt, etc. This initial electron number will grow exponentially[13] by the usual avalanche process: $N = N_0 \exp[\int \alpha(E)\, dt]$. In this case, the critical avalanche size for breakdown is given by the condition that the laser should be significantly attenuated by it (requiring more than 10^{16} or 10^{17} electrons/cm^3). This number must be reached during the pulse duration of the laser. Similarly, the time available for the dc avalanche to form is limited because the electrons may interact with the field only a short time before being swept out of the crystal. Thus, it is possible to use the thickness dependence of the dc breakdown field to predict the pulse-length dependence of the laser-induced breakdown.

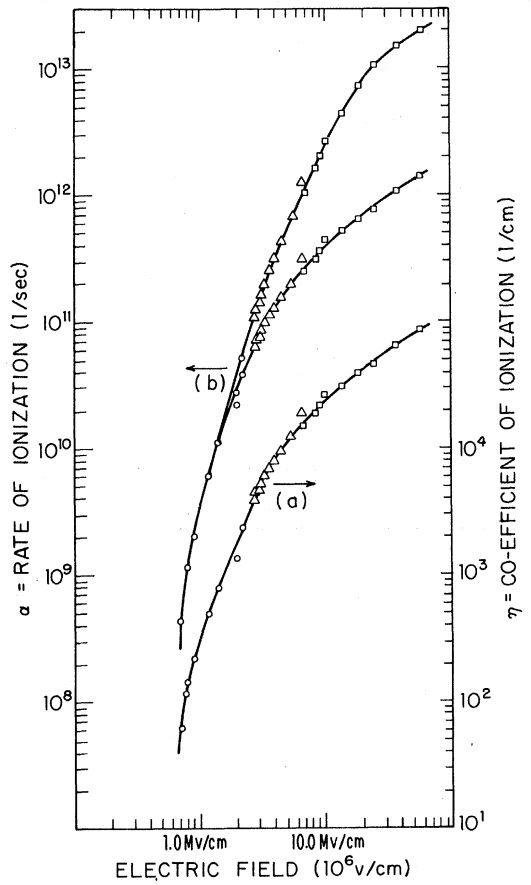

FIG. 1. The lower curve a is the coefficient of ionization η with the scale on the right. The points are determined from the empirical thickness dependence of the breakdown strength in NaCl. The upper curve b is the rate of ionization α with the scale on the left. At high fields the curve has two branches due to the uncertainty in the drift velocity v. The actual value of α lies somewhere between the two branches. Triangles, from Ref. 8; circles, from Ref. 9; squares, from Ref. 10.

A knowledge of the drift velocity $v(E)$ is necessary to determine $\alpha(E)$, the ionization per unit time, from $\eta(E)$, the ionization per unit length, plotted in Fig. 1, curve a. Although the low-field mobility μ is known[14] for electrons in NaCl, the scattering is certain to increase when the field is so great that

$$\tfrac{1}{2} m v^2 \gtrsim \hbar \omega_L, \tag{2}$$

the energy of an LO phonon. Therefore, in the high-field region, an upper limit on the drift velocity is $v = \mu E$, and a lower limit is Eq. (2). This determines $\alpha(E)$, plotted as curve b in Fig. 1. The curve has two branches at high electric fields, corresponding to the two limits on the drift velocity whose exact value is unknown. The curve predicts an ionization rate of 10^9 per sec per electron at an electric field of 10^6 V/cm in reasonable agreement[3,4] with the breakdown fields observed for nanosecond laser pulses. To produce breakdown in the picosecond regime, requiring an ionization rate of 10^{12} sec per electron, Fig. 1, curve b shows that fields of about 10^7 V/cm are sufficient.

These interesting predictions depend on the idea that the electron heating is essentially the same as in the dc limit even for optical frequencies. The energy absorption of an electron gas is $\vec{J} \cdot \vec{E}$, where the current J may be written in terms of the well-known ac conductivity. Then the energy absorption rate per electron is

$$\frac{dW}{dt} = \frac{e^2 \tau}{m(1+\omega^2\tau^2)} E^2, \tag{3}$$

where τ is the electron collision time. This formula describes the heating of electrons, and is correct even in the quantum limit,[15] where the en-

ergy absorption mechanism is often called "inverse bremsstrahlung." For NaCl, $\omega\tau \lesssim 1$, where ω is the CO_2 laser frequency and for τ determined from the room-temperature mobility.[14] For hot electrons, with kinetic energy exceeding the optical-phonon energy, the collisions are even more frequent.[16] A plausible estimate[17] is what $\omega\tau \lesssim 1$ remains true for hot electrons in NaCl even up to the ruby laser frequency.

Numerical estimates show that for a large class of wide band-gap insulators, comprising ionic crystals, glasses, and transparent liquids, up to red-light frequencies, and times down to 1 psec, the avalanche intensity limiting mechanism dominates both Zener tunneling[18] and multiphoton ionization.[18,19]

It is well established that smooth long pulses produce a moving focal spot, while picosecond pulses produce a quasistationary self-focused filament at the tail end of the pulse.[20] In either case, each material volume element experiences a high-intensity light field during a picosecond time interval. The question of the limiting diameter of self-focused filaments is still under active discussion.[21,22] Saturation of molecular reorientation and multiphoton absorption processes are inadequate to explain the observed filament diameters, which lie between 3–6 μm in a large variety of dielectrics.

Since an avalanche can build up the electron density to critical levels in a picosecond, it forms an effective limiting mechanism with an exponential contribution to the index of refraction,

$$(n+i\kappa)_{exp}=\frac{2\pi e^2\tau(i-\omega\tau)}{n_0 m\omega(1+\omega^2\tau^2)}N_0\exp\left[\int_0^t\alpha(E)\,dt\right]. \quad (4)$$

Here n_0 is the linear index of refraction, and m is the effective mass. The exponential field dependence enters through the ionization rate α. Equation (4) only takes account of the intraband contribution, omitting the effect of interband transitions. The subscript exp indicates that (4) is suddenly going to dominate all other nonlinear contributions, such as $n_2|E|^2$, etc. It should be noted that the real part n_{exp} is negative, and the imaginary part κ_{exp} represents the absorption in the ionized volume elements.

It is important to distinguish between laser breakdown and white-hot spark formation. By breakdown, we mean that the avalanche generates a sufficiently dense plasma (10^{16}–10^{17} cm^{-3}) to affect the laser-beam propagation. A white-hot spark with optical damage requires, in addition, that enough energy is deposited to cause catastrophic heating of the volume element. With $N \simeq 10^{17}$ cm^{-3}, for example, the temperature rise is less than 10°C for a picosecond pulse and the optical absorption depth is about 1 cm^{-1}, according to Eq. (4). Nevertheless, the nonlinear nature of the self-focusing problem makes it difficult to predict under what conditions a spark will be observed.

For self-focusing in glasses the diameter of the light appears to be trapped in regions of about 5 μm diameter, while the fossil damage tracks may have a diameter of only 1 μm.[23] The light intensity is only slightly higher on axis, but the electron density and heating is much larger there.

Accurate determinations of filamentary diameters in various liquids with different values of n_2 and different viscosities[24] support the proposal that avalanche ionization builds up in a filament. In spite of fairly large variations in the critical power and filament diameter, the maximum field strength remains constant,[24] near 10^7 V/cm. This number is in accord with Fig. 1, curve *b*. The following, admittedly oversimplified, argument leads very quickly to the filament size: The power in the filament is on the order of P_{cr}, the field strength on the order of E^{br} (for $t_p \approx 10^{-12}$ sec). The diameter is consequently given by $d \approx 1.22\lambda/4n_2^{1/2}|E^{br}|$. Numerical values lead immediately to sizes for d between 3 and 10 μm.

The filament can readily be stabilized by the negative real part of the index of refraction for electron densities less than 10^{17}/cm^3. This is sufficiently low that heating and sparks are avoided. For a small perturbation of the filament radius, Δr, the change in intensity-dependent index of refraction is balanced by an opposite change in plasma index [Eq. (4)], plus a small relative change in diffraction, which may be ignored, to estimate the required value of n_{exp}. Hence, $2n_2|E|^2 + n_{exp}(\partial \ln N/\partial \ln E) = 0$. A relative change of 10% in E may change N by a factor of 2. Since $n_2|E|^2 \approx 10^{-3}$ in the filament, one obtains at the equilibrium radius $n_{exp} \approx 10^{-4}$ corresponding to $N \lesssim 10^{17}$/cm^3. The length of the filaments may well be related to electronic absorption.

Finally, the stronger-than-anticipated frequency dependence of the Raman gain and other nonlinear processes inside filaments[25] may be determined in part by the fact that the maximum field strength will be higher at higher frequencies according to Eq. (3).

It is concluded that avalanche ionization is a generally occurring phenomenon which sets an

upper limit to the pulsed light intensity transmitted in a transparent dielectric and which limits the diameter and length of self-focused filaments.

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Anomalously Narrow Mössbauer Linewidth in ^{197}Au†

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Mössbauer measurements of the 77.34-keV transition in ^{197}Au are reported for thin absorbers of AuCN in *p*-azoxyanisole, AuCN dispersed in an inert powder, and $KAu(CN)_2$ similarly prepared. The average linewidth is 1.54 ± 0.05 mm/sec. This result is only 80% of that obtained from lifetime measurements, but the latter are in good agreement with results in absorbers of gold metal obtained elsewhere and corroborated here.

In the course of a study of the velocity spectra of Mössbauer absorbers aligned by the collective diamagnetism of liquid-crystal hosts, it was observed that AuCN in *p*-azoxyanisole (PAA) showed a linewidth appreciably less than the commonly accepted natural width.[1] The latter is $2\Gamma_0 = 1.85 \pm 0.01$ mm/sec, a value obtained in a recent careful measurement on Au metal,[2] and corresponds to an excited-state half-life of 1.92 ± 0.01 nsec. It is in excellent agreement with the electronic measurements of $T_{1/2}$ namely, 1.9 ± 0.2 nsec[3] and 1.95 ± 0.06 nsec.[4] It is in agreement with earlier Mössbauer measurements of gradually increasing accuracy[5] and is in fact in good agreement