SURFACE PHENOMENA IN TRANSPARENT DIELECTRICS UNDER THE ACTION OF LASER RADIATION

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This article presents some results obtained in an experimental study of the characteristics and causes of the luminescence at the surface of transparent dielectrics under the action of laser radiation.

The fluxes at which luminescence develops amount to $(2-5) \cdot 10^8$ W/cm^2 when the target is in an air atmosphere. The transparent dielectrics used in our experiments were quartz, type Ka8 glass, Plexiglas, rock salt, and Pyrex. We initially employed a ruby laser operating under a single-generation regime (the total energy was of the order of 1 J and the generation time was 10-12 nsec at the signal half-width).

With these fluxes, the luminescence creates a pulse at the transparent target amounting to 1-2 dyn·sec/cm^2, which corresponds to a pressure of several hundred atmospheres at the surface.* Such phenomena can cause surface destruction of glasses and other transparent dielectrics. This might explain the fact that dielectrics have a lower strength when their surfaces are exposed to laser radiation (in comparison with their volume strength).

The phenomenon itself is apparently associated with little-studied processes occurring in the thin boundary layers of the dielectric irradiated by the laser light. A number of studies have been made on the mechanism of this phenomenon [1-3].

1. Photographic and Spectral Measurements. The luminescence was photographed with an SFR-R scanning camera with a recording rate of $5 \cdot 10^7$ frames/sec. Development of luminescence at the surfaces of various dielectrics has a uniform spherical character; the front speed reaches 20 km/sec at the target surface with fluxes of 10^9 W/cm^2 and then rapidly decreases as the distance from the surface increases. When the flux density is reduced to $4 \cdot 10^8$ W/cm^2, the initial front speed decreases to 10 km/sec. Luminescence is accompanied by development of a shock wave, whose movement and detachment from the luminescent zone are well illustrated by a Tepler high-speed photograph (Fig. 1).

We also photographed the integral luminescence spectrum. The spectrum was found to consist of lines which corresponded to singly-ionized air components (nitrogen and oxygen). No lines corresponding to the target material were detected. The intense continuum observed in the spectrum, as well as the presence of lines for singly charged ions, indicate the temperature in the luminescence region to be high. Our investigations give us grounds for surmising that the luminescence is caused by ionized air.

The spectra for different targets were similar and had an "air" character. The luminescence is apparently not due to melting and vaporization of the target material, since it is not accompanied by noticeable (under 50-power magnification) surface damage, although in some cases there is slight damage consisting of slight streaks in the irradiated area.

A coaxial ÉLU-F photocell was used to record the integral luminescence pulse as the pressure of the air surrounding the target was varied (Fig. 2). The light flux remained almost unchanged when the pressure

*A. A. Kalmykov, Experimental Studies of the Processes Occuring under the Action of Laser Radiation on a Semitransparent Material [in Russian], Dissertation (1967).

Moscow. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, Vol. 13, No. 2, pp. 41-47, March-April, 1971. Original article submitted June 26, 1970.

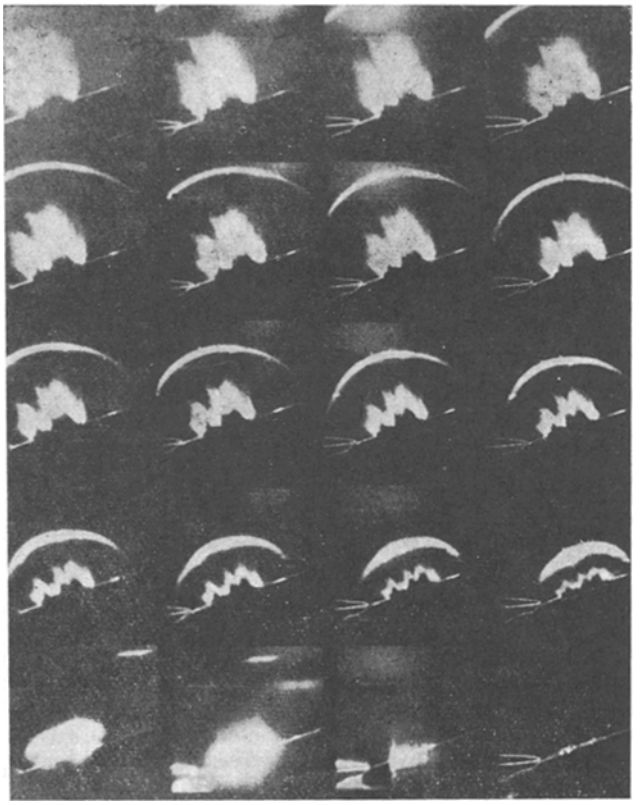

Fig. 1

was reduced from 760 to 100 torr, while the pulse duration substantially exceeded that of the laser pulse. At pressures below 100 torr, however, the pulse duration was reduced by a factor of 2 or more, and its amplitude also decreased.

The pattern became stabilized over the pressure range 1-0.1 torr, but the pulse duration was only 5-6 times that of the laser pulse. The integral light flux over the wavelength range 400-600 nm (the region of ÉLU-F sensitivity) decreased by an order of magnitude in comparison with that at atmospheric pressure. As a result of the spectral and photoelectric measurements, we obtained estimates for the luminescence threshold as function of the air pressure. The criterion for development of luminescence was appearance of signal from the photocell and the presence of a spectrum.

Some results in measuring the threshold density $\gamma \cdot 10^{-8}$ ($W \cdot cm^{-2}$) for a number of materials at different pressures p are given below (K8 represents glass, Q quartz, OG Plexiglas, and PS polystyrene):

	NaCl	K8	Q	OG	PS	p
$\gamma \cdot 10^{-8} =$	0.9	3.0	3.6	3.0	3.0	760
$\gamma \cdot 10^{-8} =$	—	5.6	5.6	5.6	—	100
$\gamma \cdot 10^{-8} =$	—	5.9	6.9	6.1	—	10
$\gamma \cdot 10^{-8} =$	—	5.2	4.7	12.3	—	1

At first glance, these data indicate that luminescence is caused by breakdown of the air, a process having features in common with the breakdown under the action of a focused laser beam in free space. The threshold levels for different dielectrics are similar. A change in air pressure from 1 atm to 1 torr leads to only a slight change in the luminescence threshold. Actually, the flux density required for breakdown in the case of sharp focusing with no obstacles is 10^{10}-10^{11} W/cm^2, i.e., exceeds the thresholds given above by more than two orders of magnitude.

It should be noted that, in breakdown with sharp focusing, the threshold changes far more abruptly when the pressure is varied.

If we assume that ionization and luminescence develop by an avalanche mechanism in the case under consideration, an electron avalanche obviously cannot develop at fluxes of $\sim 3 \cdot 10^8$ W/cm^2, since only one generation of electrons is produced during the action time of the laser (10-12 nsec). Moreover, the free path is comparable to the dimensions of the luminescence region in a vacuum of the order of 1 torr, and impact ionization can scarcely occur. It is obvious that there must be another mechanism that supplies the large number of free electrons needed in the luminescence zone.

The slight dependence of the luminescence threshold on the ambient of the air pressure indicates that the luminescence probably originates directly in the surface layers of the dielectric, possibly at gases adsorbed by the surface, then extending to the adjacent air.

2. Measurement of Currents for Dielectric Target Irradiated with Laser Pulse. In order to investigate the mechanism by which electrons are "supplied," we measured the currents at the surface of transparent dielectrics irradiated with ruby-laser light. The dielectric to be investigated 1 was located between the plates 2 of a flat capacitor, to which a constant electric field was applied. There were round apertures in the plates for entrance and exit of the laser beam 3. The current in the anode circuit was withdrawn from a 75-Ω resistor and recorded with oscillograph 4. The measurements were made at a pressure of 10^{-6} torr, freezing the vapor in a nitrogen trap. The form and energy of the laser pulse were monitored with FÉK-09 photocells.

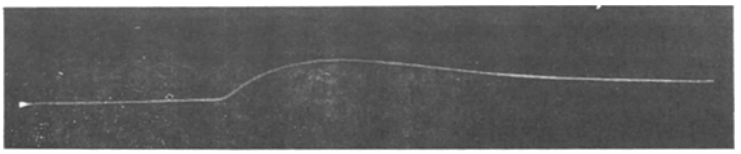

Fig. 2

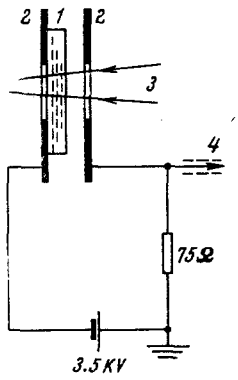

Fig. 3

Figure 4 shows a typical oscillogram for the current signal from the target. The signal from the target had two characteristic segments ("flashes"). The first flash developed during the operation of the laser and its maximum usually coincided with the laser-pulse maximum. The second pulse, which was generally of far greater amplitude, occurred after laser operation had ceased. The amplitude of the first pulse increased when the flux density was changed, although substantial scattering of its values was observed. The scattering probably depended on the state of the target surface, the type of surface treatment, and the surface finish.

However, the behavior of the targets differed, as can be seen from the following data:

	NaCl	K8	Q	P	PS	
$\gamma_{min}\cdot 10^{-8} =$	0.3	1	2	5	5	$w\cdot cm^{-2}$
$\gamma_{max}\cdot 10^{-8} =$	3	6	—	10	—	$w\cdot cm^{-2}$
j =	0.3—80	0.1—90	—	01.—40	—	$A\cdot cm^{-2}$

Here γ_{min} is the minimum flux at which current pulses appeared, the value γ_{max} being given to characterize the range of flux variation and the value of j characterizing the range of flux-density variation; P indicates Pyrex.

It must be noted that the high currents lead to a change in the color of the anode-plate surface in the immediate vicinity of the aperture after a series of experiments (the discoloration covered several field gradients).

The experiments were conducted in the region where there was no visual surface damage, i.e., below the destruction threshold. Isolated small scars were observed after a series of irradiations, but their area was small in comparison with the total area irradiated.

3. Time Sequence of Laser Signal, Current from Target, and Luminescence Signal. In order to compare the laser signal, the current from the target, and the luminescence pulse with respect to time, a "red mark" was supplied to the modulating electrode of the oscillograph tube. The accuracy in the time determinations was 2 nsec. Figure 5 shows the relative arrangement of the pulses: curve 1 is the laser signal, curve 2 is the current pulse, and curve 3 is the light pulse ("flash").

The maxima of the laser and current signals coincided, while the latter was received later than the former. This phenomenon was due to the nonlinear dependence of the current on the irradiation.

It should be noted that development of the current, like appearance of the flash, had a threshold character. The current densities (γ_{min}) at which current signals were detected are given above. No current was recorded when the flux density was slightly reduced. Similar experiments were conducted with longer laser pulses (of the order of 100 nsec at the half-width). The threshold current densities corresponding to the current signal remained unchanged. The duration of the current pulse was increased by a factor of approximately 5, while the front amplitude increased by a factor of 1.5-2.

4. Discussion of Results. We will now consider the probable causes of the appearance of a current at the target.

1) Multiquantum photoeffect. The possibility of photocurrent cannot be precluded but, in view of the stepwise dependence of the current on the electric-field strength of the light wave, the form of the current pulse should have been greatly altered (in comparison with experiments with short signals) when the laser pulse was prolonged; the current-pulse front should at least have been greatly prolonged. However, any photoeffect might be masked by another, more important phenomenon.

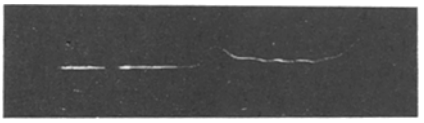

Fig. 4

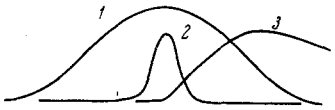

Fig. 5

2) Thermionic emission. The action of laser radiation on gases adsorbed on a dielectric surface can cause them to overheat, form a plasma, and emit electrons. If such heating occurs, the problem of how the energy necessary for this process is adsorbed remains unresolved.

We calculated the temperature necessary for electron emission from a plasma with current densities of 10-100 A/cm^2, using the Richardson-Dushman formula

$$I = en2\pi R^2 (k / 2\pi m)^{1/2} \exp(-e\varphi / kT)\sqrt{T}$$

where R is the radius of the radiating hemisphere and eφ is the electron work function for emission from the plasma.

The work function was calculated as eφ = e^2/d, where d is the Debye screening radius:

$$d = (kT / 4\pi ne^2)^{1/2}$$

When the air is at atmospheric pressure (the adsorbed air is assumed to be at this pressure, although its intensity at the surface can actually be above or below this figure) and its temperature is raised from 3500 to 10,000°K [4], the electron concentration changes from 10^{13} to 10^{20} cm^{-3}. In this case, $\exp(-e\varphi/kT) \sim 1$.

Calculation shows that a current density j = 100 A/cm^2 corresponds to a plasma temperature T = 5000°K, while j = 10 A/cm^2 corresponds to T = 4000°K.

3. Mechanism of plasma decay under the action of an applied electric field. Plasma decay begins with cold electron emission; the ionic components then begin to move. The plasma-decay processes depend to a large extent on the plasma temperature and on the charge-carrier concentration, the latter in turn being related to the amount of adsorbed gas subjected to heating. The presence of a second peak in the current pulse may be due to these phenomena.

Although we cannot hold with certainty to one view or the other regarding development of the current pulse, it can be assumed that current pulse 2 precedes development of the flash. This follows from Fig. 5.

The flash experiments conducted in air and the experiments involving measurement of the currents (with an electric field and a high vacuum, in which case there was no flash), were naturally different experiments. However, the appearance of free electrons in the dielectric during laser irradiation, a phenomenon that can lead to subsequent absorption of radiation and development of flashes, occurs without an external electric field.

In this connection, the mechanism by which the dielectric absorbs the laser radiation becomes extremely important. The dielectric specimens used (5 mm thick) exhibited almost no absorption in normal visible noncoherent light. When light fluxes of 10^8 W/cm^2 were supplied from a ruby laser, absorption occurred (the experiments were conducted below the breakdown threshold and the specimen was held in a vacuum of 10^6 torr to prevent flashing), constituting about 10% of the total incident energy.

It has been hypothesized that the absorption results from the surface state of the dielectric, which specifically depends on the surface finish and type of treatment (grinding, polishing with abrasive powder, etc.).

The development of a current pulse is independent of the surface treatment; measurements made with sections showed the pattern to remain unchanged. We are obviously dealing with nonlinear processes in the surface layers of the specimens, which do not behave like ideal dielectrics.

It is always necessary to take into account the influence of local levels in real dielectrics. Thus, for example, the forbidden-band width in Al_2O_3 is 10 eV. Transition of a substantial number of electrons from the filled band to the conductivity band begins at a temperature T = 2000°K, and it seems that Al_2O_3 should be an ideal insulator at room temperature. The resistivity of aluminum oxide is known to be 10^9-10^{12} $\Omega \cdot$cm. This comparatively low resistance is due to impurities in aluminum oxide, which form local donor levels.

The energy necessary for the transition from a local level to the conductivity band is substantially less than the forbidden-band width, amounting to 2 eV in Al_2O_3.

A similar pattern can occur in any dielectric, particularly in glasses, on whose band structure little research has been done.

In addition to the local volume levels, it is also necessary to take into account the superficial energy levels, as was first pointed out by I. E. Tamm.

All this indicates that, when a real dielectric is irradiated with ruby-laser light having a quantum energy of about 1.8 eV, the single-photon process in the conductivity band can by itself produce a sufficient number of free electrons, which are responsible for subsequent energy absorption and development of the phenomena under investigation. The existence of free electrons is indicated by the experiments of T. P. Belikova et al. [5], who determined the volume conductivity of ruby under the action of laser light. These authors took special measures to eliminate the surface conductivity, which is far greater than the volume conductivity.

It can thus be assumed that a sufficient number of free electrons appears in a dielectric under the action of laser irradiation, as is indicated by the current signal from the target; the presence of free electrons leads to further absorption of laser radiation and heating of the gases adsorbed on the target surface and the air surrounding the target. The latter leads to development of flashes.

This mechanism is supported by a comparison of the flux densities $\gamma^{\vdash}$ and $\gamma*$ [$W \cdot cm^{-2}$] corresponding to appearance of current and development of flashes for inorganic dielectrics (table salt, type K8 glass, and quartz):

	NaCl	K8	Quartz
$\gamma^{+} \cdot 10^{-8} =$	0.3	1	2
$\gamma_{*} \cdot 10^{-8} =$	0.9	3	3.6

It follows from these data that the currents at the target appear at lower flux densities than the flashes, i.e., development of a flash requires formation of a sufficient number of free charges in the dielectric.

This pattern was not observed for organic dielectrics, but it was necessary to work above the breakdown threshold in this case, since neither currents nor luminescence were recorded below the breakdown threshold.

The development of volume and surface damage in Plexiglas and styrene complicates the phenomenon under investigation. At the present stage of our research, it can thus be stated that it is necessary to have dielectrics with a crystal structure as close as possible to ideal in order to eliminate flashes at the surface or at least to increase the flash-development threshold; the forbidden band in the dielectrics must be sufficiently wide.

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