NONLINEAR ABSORPTION OF LASER PULSES BY A PARTIALLY IONIZED GAS

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The experimental arrangement and method used for investigating the absorption of laser pulses by a plasma are described below, together with the results obtained; these investigations led to the discovery of new effects in the dependence of absorptive power on the light intensity [1, 2].

A ruby laser was employed, operating in the modulated Q mode. The investigations were performed in a shock tube, behind the front of the reflected shock wave. The experiments showed that the plasma has pronounced transmission properties at light intensities $I = 10^7 - 10^9 \text{ W/cm}^2$, while its transparency falls at $I = 10^9 - 10^{11} \text{ W/cm}^2$. The phenomenon will be described in qualitative terms.

<u>1. Experimental System and Method.</u> Before light absorption by a plasma can be measured, a homogeneous plasma with known, accurately reproducible properties must be obtained. One of the best ways of doing this is to use a shock tube. The set-up shown schematically in Fig. 1, and consisting of a shock tube, laser, and measurement system, was used for the experiments.

The shock tube consists of a high pressure chamber 1 of length 1.7 cm and a low-pressure chamber 2 of length 7.0 m. The latter is a ground channel 80 mm in diameter, consisting of two sections each 3 m long, and two measurement sections each 0.5 m long. The measurement sections have windows, for carrying out optical measurements, together with ionization and piezoelectric pick-ups 3, mounted in their walls. Between the high- and low-pressure chambers was located the diaphragm unit, connected with the systems for pumping out the chambers and filling them with the impacting and the studied gas. Hydrogen 11 was used as the impacting gas; the studied gas was xenon 12. The low-pressure chamber was pumped out to a pressure of $6 \cdot 10^3$ mm Hg; the inleakage during the measurements did not exceed 10^{-4} mm Hg.

The shock wave velocity was measured to an accuracy of $\pm 1.0\%$ by means of the ionization and piezo-electric pick-ups.

The windows were located opposite one another, each 1 cm from the end of the tube; the light whose absorption by the plasma was being measured passed through them.

The high-intensity light source was the ruby laser 4 with modulated Q. Its active element was a $120 \times 12 \text{ mm}^2$ ruby rod. The resonator consisted of two plane mirrors parallel to one another, with reflection coefficients of 99 and 20% respectively. The resonator Q was modulated by means of a passive shutter (solution of phthalocyanine). The pump tube was the type IFPP-7000. The giant pulse duration was 50 nsec, and the laser power in the modulated Q condition was roughtly 20 MW.

Measurement of the light absorption over a very wide range of intensities was required. The high intensities were obtained by focusing the laser beam into the plasma by a lens with a focal length of 3 cm. The diameter of the focusing circle, found by measuring the intensity distribution over the focus cross section, was $1.35 \cdot 10^{-2}$ cm, while the light intensity at the focus reached $1.4 \cdot 10^5$ MW/cm². The light was attenuated by means of neutral light filters. To obtain the low intensities, a lens with a focal length of 23 cm was used; the light pencil within the tube was then almost parallel. The range of investigated intensities covered seven orders (from 10^{-2} to 10^5 MW/cm²). The radiation passing through the plasma was collected by a lens at the center of the wide open slit of the DFS-12 monochromator 5, which enabled the inherent

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Fig. 1

plasma illumination to be eliminated. Behind the monochromator output slit was mounted the FEU-52 photomultiplier 6, the signal from which was connected to the nanosecond oscillograph I2-7 7.

A plasma is well known to cool behind the front of shock waves due to radiation losses, and its parameters correspondingly change in the course of time. To ensure that the laser pulse passed through a plasma with the same properties during the numerous measurements, the laser had to be triggered at a precisely determined instant after the passage of the reflected wave past the inspection windows. This time interval was 300 μ sec in one series of measurements, and 100 μ sec in another. The synchronization system 8 was used for this purpose; it operated as follows: at the instant when the incident shock wave went past the pressure pick-up,

spaced 3 m from the measurement section, a triggering pulse was generated, and applied to the G5-15 generator. The square pulse from this generator, delayed by an adjustable time relative to the reference signal, triggered the firing circuit of the pump tube IFPP-7000. The giant pulse developed roughly 800 μ sec after firing the tube. At the same instant that the IFPP-7000 was fired, the OK-17 M oscillograph 9 was triggered; one scan of the latter was used for recording the signal from the ionization pick-up, mounted in the cross section where the absorption was being observed, while the other scan was used for recording the instant when the giant pulse formed. This last was done as follows: the plate 10 with plane parallel sides was mounted at a certain angle to the laser radiation beam, with the function of allowing the FEU-52 photomultiplier to pick up part of the light pulse. The nanosecond voltage pulse from the FEU-52 was applied to the input of the G5-3B generator, converted into a 10 μ sec square pulse, and re-corded by the second scan of the OK-17 M oscillograph. By using this method, it was possible to determine the plasma region behind the front of the reflected wave, through which the laser pulse passed.

Any point behind the shock wave front could be studied, due to the fact that the measurements were accurately reproducible, as regards both the shock wave parameters and the instant of formation of the giant pulse.

It is well known that, when intense laser radiation is focused in a cold gas, an intensity will eventual ly be reached, the size of which depends on the type of gas and its density, at which the gas breaks down, i.e., violent ionization develops in it and a plasma is formed. To compare the measurements of high-intensity light absorption in a plasma with the transmission of such light through a cold gas when breakdown occurs, the breakdown in cold xenon at the same density as the plasma was investigated in the same apparatus.

Special attention was paid to the pulse shapes during the investigations.

2. Results. The studies of the interaction between laser radiation and a plasma were carried out behind the front of the reflected shock wave in xenon. The equilibrium parameters of xenon were found by the usual calculation, using the equations of conservation of mass, momentum, and energy, together with the equations of state and Saha's equation. Allowance was made for the drop in the ionization potential of xenon in a plasma. The measurements were performed withan initial xenon pressure of 10 mm Hg and the same incident shock wave velocity of $1.82 \text{ mm}/\mu \text{sec}$. This corresponded to the following equilibrium parameters of xenon behind the front of the reflected shock wave: temperature $11,250^{\circ}$ K, electron density $0.97 \cdot 10^{18}$ cm⁻³, and neutral atom density $5.6 \cdot 10^{18}$ cm⁻³. The true values of the plasma parameters in the investigated region were somewhat different from the equilibrium figures just quoted, due to radiation losses.

When laser radiation passed through ionized xenon, an attenuation of the light was observed, the size of which depended on the incident radiation intensity. As an example, Fig. 2a shows oscillograms of the giant pulse in the absence of a plasma, and after it has passed through the plasma; the radiation intensity was $I = 5.6 \cdot 10^2 \text{ MW/cm}^2$.



Fig. 2



The results of these measurements are presented as curves in Fig. 3, showing light transmission, or more precisely, the ratio P_1/P_0 of the power transmitted by the plasma to the incident power, against the power of the beam incident on the plasma. For convenience, the latter is expressed in relative units P_0/P_t , where the scaling factor P_t is the beam power causing breakdown in cold xenon of the same density as the plasma when the beam is focused by the lens of 3 cm focal length. In the measurements $P_t = 20$ MW. Curve 1 (Fig. 3) refers to the plasma parameters 300 μ sec after passage of the reflected wave. The plasma cools by roughly 2000° K in this time. The ranges of the experimental points corresponding to low intensities are indicated in Fig. 3, while the circles on the curve represent the means of 7-8 measurements.

It can be seen from Fig. 3 that nonlinear, i.e., power-dependent, absorption of the laser radiation occurs. As the incident power increases, roughly from 10^{-4} Pt, the plasma "transparency" increases slightly at first, then the absorption gradually increases, roughly from 20 to 90%.

For purposes of comparison with the data obtained on nonlinear absorption of laser radiation in a plasma, auxiliary measurements were made with the same equipment, on the breakdown in xenon at room temperature, but at the same density as behind the wave front. This density corresponds to a cold gas pressure of approximately 200 mm Hg. The results from these latter measurements are also shown in Fig. 3 (curve 2). Notice first that the figure $I_t = 1.4 \cdot 10^5$ MW/cm² obtained for the intensity corresponding to breakdown threshold agrees well with the data of [3]. In addition, Fig. 3 shows clearly the very sharp threshold, which is quite different from the case of a plasma. For instance, while at most 10% of the incident beam energy passes through the breakdown region with $P_0 \approx P_t$, virtually no light attenuation is observed when $P_0 \approx 0.57P_t$.

Another interesting point is that, in the case of breakdown in cold xenon, the transmitted pulse has the steep trailing edge typical of breakdown in gases (Fig. 2b), whereas in the "transparent" region and in the initial "darkening" region of the plasma, the transmitted pulse retains to a first approximation the shape of the initial pulse, the only difference being in its amplitude (Fig. 2a). At higher light intensities, at which the plasma darkens considerably, the transmitted pulse undergoes some change of shape; its duration is somewhat reduced, and its maximum moves slightly towards the start of the pulse.

Notice, however, that the giant pulse amplitude was not very stable, i.e., at low absorptions, in the region where the "illumination" effect is observed, the method is not sufficiently accurate for reliable quantitative data to be obtained. A series of experiments aimed at improving the method was made in this connection; in this series, the shapes of the initial and transmitted pulses could be observed simultaneously, thus avoiding the inaccuracy just mentioned. In addition, to facilitate processing the results, these experiments were performed with a long-focus lens (f = 230 mm), so that the light beam was virtually parallel within the tube, while its mean diameter was approximately 1.5 mm. And finally, to strengthen the effect, the plasma was probed by the laser beam at a point closer to the reflected wave front, where the absorption was greater. This point corresponded to a period of 100 μ sec after passage of the reflected wave past the windows. The plasma was estimated to have cooled by only 1000° K in this time. The results of this series of experiments are shown in Fig. 4. As distinct from the first series, the results are here expressed as ratios of the intensity corresponding to the mean beam diameter, since illumination here occurs almost throughout the length of the beam, and not at the point of focusing. This led to some displacement of the experimental curve towards lower intensities as compared with the former processing used in [2], where the results were referred to the light intensity at the focus. Account should also be taken of the reduction in intensity due to light absorption on passing through the plasma, but the resulting corection is small.

It can be seen from the curves that, at low light intensities (powers), the ratio of transmitted to incident power is independent of the intensity, indicating that the plasma absorption coefficient is constant. In other words, the absorption is "linear" at low intensities. As the intensity rises to a certain (not par-ticularly high) level the absorption falls somewhat, i.e., the plasma is more transparent and "illumina-tion" takes place. At even higher intensities the transparency falls sharply and the absorption becomes much higher than linear. At powers higher than $10^{-2}P_t$ it becomes difficult to perform measurements with a long-focus lens, due to destruction of the windows under the action of the laser pulse.

To verify that the light was in fact absorbed in the plasma during the measurements, the intensity of the light reflected backwards was measured. The results shows that only a very small fraction of the energy, of the order 10^{-3} , is reflected or scattered in the plasma. The supplementary plasma radiation, linked with the laser beam action, was also investigated. An example of an oscillogram is given in Fig. 2c. This records the laser pulse at the start of the scan and the plasma radiation in the region of 5000 Å. The time pip repetition period is 50 nsec. These experiments showed that the plasma recovery time from laser pulse excitation is roughly 1.5 μ sec; an interesting point is that the radiation relaxation time in the plasma experiments were roughly half the time for a breakdown plasma.

3. Physical Causes of the Effect. The above experimental data reveal two contradictory effects in different ranges of light intensities: "illumination", i.e., a reduction in the absorptive power of the plasma, and "darkening," i.e., an increase in absorption. Both effects are linked with the action of the absorbed light on the plasma, which results in the plasma going over to a different, nonequilibrium, state.

There are two main mechanisms of light absorption in a partially ionized gas: photo-ionization, and a process the reverse of bremsstrahlung, which for brevity may be termed stopping absorption.

The coefficient of photo-ionization absorption $\varkappa p_h$ is the sum of partial coefficients corresponding to the contributions from atoms in different excited states but with binding energies less than $h\nu = 1.78$ eV:

$$\kappa_{\rm ph} \sim \sum N_n \sigma_n$$

where N_n is the number of atoms per cm³ staying at the n-th energy level, and σ_n is the corresponding effective cross section of the photo-effect.

The coefficient if stopping absorption is proportional to the number of collisions between electrons and ions and inversely proportional to the square root of the electron temperature:

$$\kappa_T \sim N_s^2 T^{-1/s}$$

Here N_e is the electron density and T the electron temperature. The overall coefficient of true absorption is $\varkappa = \varkappa_{Ph} + \varkappa_{T}$. In addition to the acts of absorption, processes occur involving forced emission of light quanta, of the same magnitude and direction as in the laser pulse, and manifesting itself in a certain reduction of the resultant absorption.

Consider first how the plasma illumination occurs. As a result of absorption of the laser light, the electron gas heats up and its temperature rises. Estimates show that substantial heating will occur at precisely those intensities at which illumination starts.

The temperature rise assists the ionization of excited atoms during collisions with electrons; this ionization takes place very rapidly, in the order of 10^{-11} sec at temperatures of $10,000^{\circ}$ K, which is much shorter than the laser pulse duration. The temperature rise also leads to simultaneous acceleration of the acts of atomic excitation from the fundamental state due to electron collisions. However, the process whereby the atoms are excited from their fundamental state (and ionized) takes place much more slowly than the ionization of the excited atoms, since a much greater energy expenditure is required. In ordinary (equilibrium) conditions this second process is nevertheless quite rapid, so that the population of higher atomic states is increased. In experiments with short laser pulses the situation is different. The excitations of the atoms from their fundamental states are unable to take place during the short pulse, and the primary effect of rapid ionization of excited atoms results in the population of higher levels falling. But the fall is not indefinite, since simultaneously with the ionization, inverse processes take place just as rapidly, involving capture of electrons by ions at higher levels during triple collisions in which electrons participate as third particles. A new quasi-equilibrium state is established, in which there is an approx-

imate equilibrium between the free electrons and excited atoms, but no equilibrium between the excited and unexcited atoms. In this new state the concentration of excited atoms, which are in fact the principal light absorbers, becomes less than the initial concentration, thus leading to reduced light absorption.

The disappearance of the excited atoms is also assisted by the process of photoelectric absorption of light quanta. This plays a secondary role at the very beginning of the "illumination" range. But its importance increases as the light intensity increases further. Notice that, along with the photo-ionization under the light action, a process of forced photo-recombination rapidly evolves, in which electrons are captured by ions in the laser light field and quanta of the same frequencies and directions are emitted.

Another effect, namely, darkening of the plasma, comes into play at higher intensities. In this range there is a very marked rise in the electron gas temperature, which tends to speed up the excitation (and ionization) of atoms from the fundamental state. This causes an increase in the degree of ionization and the density of the free electrons, and as a result, an increase in the absorptive power of the plasma.

An interesting point is the gradual rise in the absorptive power of the plasma, in contradistinction to the case of breakdown in a cold gas, where there is no absorption at all initially, then a very rapid rise in absorption as the intensity increases (see curve 2 of Fig. 3). This difference may be explained on the basis of the avalanche-like mechanism of cold gas breakdown.

The nonlinear effects in light absorption by a plasma can substantially affect the screening of laser radiation by ionized vapor when a solid target is bombarded. It should also be taken into account in methods where a laser is used for plasma diagnosis, and intense radiation is involved, since at such intensities the light has a significant effect on the plasma and changes its state.

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