Self-defocusing of converging beams: circular "intensity waves" at the focus

Yu. K. Danileĭko, V. A. Milyaev, Yu. P. Minaev, A. M. Prokhorov,
A. V. Sidorin, and A. V. Shirkov

Institute of General Physics, Academy of Sciences of the USSR
(Submitted 3 December 1985)
Zh. Eksp. Teor. Fiz. **91**, 166–171 (July 1986)

The time behavior of the spatial distribution of the radiation intensity at a nonlinear focus is studied experimentally for self-defocusing of a converging neodymium laser beam ($\lambda = 1.06$ μm) in silicon. The initially bell-shaped distribution spreads out into a circular "wave" that travels outward at velocities of 10^5–10^6 cm/s. The wave moves so rapidly that the radiation pulse measured at an arbitrary point in the focal plane is much shorter than the laser pulse. This shortening must be taken into account when estimating how the defocusing alters the interaction of light with matter (photoionization, laser breakdown, etc.). Some possible defocusing mechanisms are discussed.

1. INTRODUCTION

The self-interaction of propagating light beams is one of the most actively studied areas in nonlinear optics. However, most of the interest has been in self-focusing and much less is known about self-defocusing, particularly in the time-dependent case. This is in spite of the fact that an understanding of self-defocusing is extremely important because it is commonly present when intense electromagnetic radiation interacts with matter, for example during photoionization of gases and solids (avalanche or multiphoton ionization), photochemical reactions, photoexcitation of excess carriers in semiconductors, etc.

One of the most striking manifestations of self-defocusing is the generation of a thin beam emerging from the focal region of a focused beam.[1] However, no experimental work has yet been reported concerning the structure of the nonlinear focus during self-defocusing.

The theoretical description of the interaction process in systems with self-defocusing requires that one analyze the Maxwell equations with suitable nonlinear material equations, which is very difficult. The published theoretical work is very scanty and treats only a few model nonlinearities.[2] It is therefore important to have experimental information on how the structure of the nonlinear focus evolves in space and time during self-defocusing of converging light waves.

2. EXPERIMENTAL TECHNIQUE

It is experimentally quite difficult to analyze the structure of the nonlinear distortions of the electromagnetic field near the focus in a system with self-interaction. This is because if the focal region (caustic) lies inside the material at a considerable distance from the entrance and exit surfaces, the intensity distribution outside the nonlinear medium is nearly symmetric relative to the focus even when self-defocusing occurs.[2] In this case it is almost impossible to detect any redistribution of the intensity at the focus when recording "into the beam." The distribution can be made asymmetric by positioning the focus near the exit surface of the nonlinear medium,[1–3] and in this case two methods can be used to find the intensity distribution in the boundary plane of the nonlinear medium passing through the focal region. The first method involves analyzing the far-field distribution and would appear to be quite simple experimentally. However, in this case one must solve an "inverse optics" problem which is computationally unstable, and the required accuracy in measuring the diffracted field is so great as to make this method impractical. The second technique is based on analyzing the near-field and requires a good spatial resolution, of the order of a few wavelengths; we chose this method in our experimental investigations.

The following requirements guided the choice of the nonlinear material. First, the self-defocusing must be time-dependent, because this is the situation usually encountered when radiation interacts with matter. Second, the nonlinearity of the material must be appreciable at relatively weak light intensities, so that breakdown of the exit face can be avoided. These requirements can be satisfied by irradiating semiconductors with photons of energy comparable to the width E_g of the forbidden band. We used a highly resistive p-type silicon single crystal without dislocations ($E_g = 1.09$ eV at $T = 300$ K) and a single-mode, single-frequency YAG:Nd^{3+} laser with photon energy $\hbar\omega = 1.17$ eV. The pulse length 10^{-8} s was shorter than the excess carrier recombination time, which ensured that the interaction was time-dependent.

The polarized laser beam was focused by a $\times 10$ microscope objective, and the focusing angle was set up by a diaphragm located immediately ahead of the focusing objective. The laser beam was first expanded to ensure that the intensity distribution was uniform over the aperture of the diaphragm. This method for changing the focusing angle made it much easier to align the system than would have been the case if lenses with various focal lengths were used. The beam was focused onto the exit surface of a silicon wafer of thickness 1 mm, and an infrared microscope ($\times 350$ magnification) monitored the error in positioning the focal plane coincident with the exit surface of the wafer. One end of a fiber optic probe was positioned in the plane of the image of the exit face of the wafer and directed the radiation onto an ava-

 0038-5646/86/070096-03$04.00

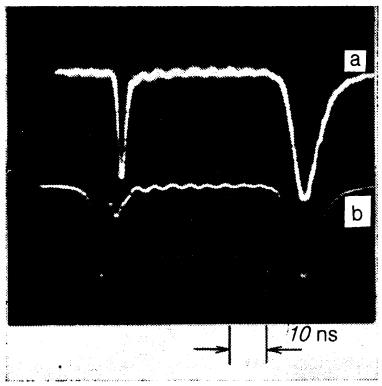

FIG. 1. Time behavior of the radiation intensity at a point in the focal plane at a distance r from the center of the caustic. The second pulse (incident radiation pulse) is used for time-clamping: a) $Q = 10^2\, Q_{\text{thr}}$, $2\theta = 8\cdot 10^{-2}$ rad, $r = 9\,\mu$m; b) $Q = 10^3\, Q_{\text{thr}}$, $2\theta = 0.14$ rad, $r = 0\,\mu$m.

lanche photodiode. A Glan prism was used to vary the incident power reaching the wafer. Analysis of the space-time dynamics required establishing a time reference, which was done by diverting part of the beam into an attenuator and thence into a light guide, where it was optically delayed before reaching an identical photodiode. The sum of the signals from the photodiodes was recorded on an oscilloscope; the time resolution was better than 10^{-9} s. The magnitude of the electric signal from the photodiode was proportional to the radiation intensity at the corresponding point in the focal plane of the focusing objective, with proportionality coefficient equal to $6\cdot 10^4$ W·cm^{-2}·mV^{-1}. Figure 1 shows some typical signal traces for pulse powers 100 and 1000 times above threshold. The pronounced shortening of the locally observed radiation pulse caused by the self-defocusing is striking.

The experimental arrangement enabled us to record a series of traces at fixed focusing angles and input powers, thereby obtaining information on the time behavior of the radiation intensity at a specified set of points in the focal plane at various distances from the center of the pattern. Since the time origin was known, the traces permitted us to reconstruct the intensity distribution on the exit face of the silicon wafer at arbitrary times.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The time behavior of the intensity in the focal plane deduced from the reconstructed distribution was as follows. Initially, the intensity increased at all points and had a bell-shaped distribution whose width increased slightly. The intensity then dropped at the center, and the resulting dip transformed the bell-shaped distribution into an expanding "ring" which continued to expand even on the trailing edge of the incident pulse. Figure 2 shows the reconstructed dynamics for $Q = 10^2 Q_{\text{thr}}$ and a diaphragm of diameter 4 mm, starting at the instant the intensity at the center of the caustic stopped increasing. The fact that the interaction region expanded indicates that self-defocusing rather than self-focusing occurred (the latter can also give rise to a ring structure). The time-dependence of the observed self-defocusing cannot be attributed to heating of the wafer by the laser radiation, because the refractive index of silicon increases with temperature and self-focusing would therefore be expected. On the other hand, the observed drop in the permittivity ε could be caused by excess carrier production, and the time-dependence could be due to the "integrating" character of the carrier generation (the long recombination time).

We compared the radial intensity distributions on the exit face of the wafer for two incident pulse powers differing by a factor of ten and found that the ring diameters recorded at various times were equal provided the transmitted energies were the same. This indicates that for fixed focusing conditions, the spatial distribution of the refractive index at any time is determined by the energy transmitted through the wafer up to that time. This agrees with the theoretical predictions for time-dependent self-defocusing in which the change in ε is proportional to the energy density of the radiation.[2] In this case one also expects that the energy threshold for self-defocusing will be the same for all focusing angles. Here the threshold energy is defined to be the radiation energy transmitted just prior to ring formation, i.e., prior to the instant the dip starts to form in the center of the distribution. Diaphragms of diameter 2.5 or 4 mm (corresponding to focusing angles $2\theta = 5\cdot 10^{-2}$ and $8\cdot 10^{-2}$ rad) were used in the experiments, and the threshold energy $Q_{\text{thr}} = 2\cdot 10^{-8}$ J was the same in both cases. For these focusing angles, the experimental findings also agreed with the theoretical conclusions in Ref. 2. In particular, at the geometric focus the radiation energy density transmitted prior to ring formation was proportional to the square of the focusing angle, and the peak intensity was roughly 50% of the value calculated by the formulas of linear optics for a given time t.

However, appreciable departures from proportionality $Q_{\text{thr}} \propto \theta^2$ were noted when the focusing angle increased to $2\theta = 0.14$ rad. The experimental values were 50 and 100% greater than predicted for $Q/Q_{\text{thr}} = 10$ and 10^2, respectively (the value Q_{thr} was determined for small focusing angles). Faster recombination of the excess carriers at higher powers (less "integration" associated with excess carrier generation) could account for this discrepancy. For silicon, Auger recombination can decrease the recombination time. We

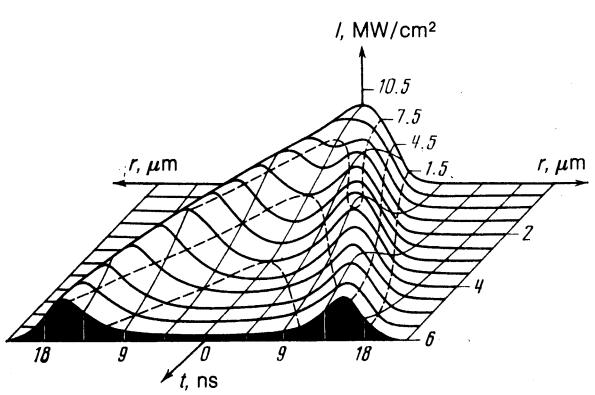

FIG. 2. Intensity distribution in the focal plane as a function of time t relevant to the instant the intensity at the center of the caustic stopped increasing; $Q = 10^2\, Q_{\text{thr}}$, $2\theta = 8\cdot 10^{-2}$ rad.

raised the incident pulse energy by another factor of ten ($Q = 10^3\, Q_{thr}$) to obtain additional information on the self-defocusing at $2\theta = 0.14$ rad. We observed a marked change in the shape of the trailing edges of the pulses recorded at points in the center of the focal spot (Fig. 1b). The entire time dependence of the intensity distribution in the focal plane was reconstructed and the following features were found. As in the experiments at lower Q, the initial bell-shaped distribution was transformed into an expanding ring; however, a new "bell" was then generated at the center of the ring, and another expanding ring was formed. This behavior demonstrates unambiguously that the carriers recombined more rapidly (there was no integrating effect caused by the accumulation of excess carriers).

To estimate the excess carrier concentration at which the Auger recombination time becomes comparable to the generation time ($\alpha I/\hbar\omega \approx qn^3$), we take $\alpha \approx 10$ cm^{-1}, $q \approx 10^{-31}$–10^{-30} cm^6/s, and $I \approx (2$–$4)\cdot 10^7$ W/cm^2. This gives $n \approx 10^{19}$ cm^{-3}, in agreement with the value

$$\frac{\alpha}{\hbar\omega}\int_0^{\tau} I\,dt$$

calculated from the trace (Fig. 1b); here I is the intensity at the center of the focal spot, and the integration is over all times t prior to "cutoff" τ.

Our investigations show that the nonlinearity of the refractive index is due to excess carrier generation. The results in Ref. 2 can be used to estimate the drop $\Delta\varepsilon$ in the permittivity of silicon required for the self-defocusing to be observable ($\Delta\varepsilon/\varepsilon \approx 5\theta^2$).

We mention three factors which are associated with free carrier generation and can decrease ε: 1) the electron plasma[4]; 2) charge transfer involving impurity centers, which can alter their polarizability[5]; 3) the time-dependent Burstein-Moss effect.[6]

One must consider the detailed band structure in order to quantitatively explain the mechanism for the nonlinear behavior of ε in single-crystal silicon irradiated by radiation at micron wavelengths. We are unaware of any theoretical work in which such an analysis has been undertaken. Numerical estimates based on the Drude theory suggest that at $\lambda = 1.06\,\mu$m, the electron plasma can decrease ε by only 10–20% of the amount required to explain the observed self-defocusing.

Because of the low polarizability of the impurity centers at frequencies near the edge of the fundamental absorption band, charge transfer involving impurities is unlikely to decrease ε significantly. Moreover, additional experiments showed that the self-defocusing was virtually independent of the level of doping.

As for the Burstein-Moss effect, we are not aware of any published work in which this effect has been analyzed for indirect-gap semiconductors.

4. CONCLUSIONS

Our experiments show that time-dependent self-defocusing greatly alters the spatial distribution of the field near the focus. The intensity distribution is smooth and bell-shaped at low energies but is transformed into one or more rapidly expanding rings at higher pulse energies. High energies are needed to generate electron concentrations of 10^{17}–10^{19} cm^{-3}. One should also bear in mind that the effective time for light-matter interaction at a fixed point in the focal region may be much shorter than the length of the laser pulse. The actual amount of energy deposited in the material may thus differ greatly from the value calculated using the linear theory of light propagation. For this reason, defocusing must not be neglected in studies of laser-induced ionization of transparent condensed media. For example, self-defocusing in silicon decreases the absorbed energy so greatly that laser pulses of wavelength $\lambda = 1.06\,\mu$m and length $\tau = 10^{-8}$ s are incapable of destroying the sample at room temperature. A similar conclusion was reached by Kelly *et al.*[7] for several other materials.

Although the main experiments were carried out for beams with a plateau-like intensity distribution, the time dependence of the field distribution is similar for Gaussian beams, and apparently for nearly all beams with a smooth intensity distribution. We note also that a similar time behavior was observed during self-defocusing of a neodymium laser pulse in ethanol, where the integrating nonlinearity is presumably of thermal origin.

[1]G. A. Askar'yan and M. A. Mukhamadzhanov, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 48 (1981) [JETP Lett. **33**, 44 (1981)].
[2]Yu. K. Danileĭko, L. M. Degtyarev, A. L. Kopa-Avdienko, and T. P. Lebedeva, Zh. Eksp. Teor. Fiz. **87**, 730 (1984) [Sov. Phys. JETP **57**, 417 (1984)].
[3]V. N. Lugovoĭ, Zh. Eksp. Teor. Fiz. **65**, 886 (1973) [Sov. Phys. JETP **38**, 439 (1973)].
[4]J. M. Ziman, *Principles of the Theory of Solids*, Cambridge Univ. Press (1969).
[5]R. Baltrameyunas, D. Veletskas, and I. Kalturauskas, Pis'ma Zh. Tekh. Fiz. **10**, 522 (1984) [Sov. Tech. Phys. Lett. **10**, 220 (1984)].
[6]D. A. B. Miller, S. D. Smith, and B. S. Wherrett, Opt. Comm. **35**, 221 (1980).
[7]P. J. Kelly, D. S. Ritchie, P. F. Braünlich, A. Schmid, and G. W. Bryant, IEEE J. Quant. Electr. **QE-17**, 2027 (1981).

Translated by A. Mason