Self-focusing of ruby-laser radiation in single-crystal silicon carbide

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The dynamics of self-focusing of the radiation of a ruby laser in SiC(6H) and SiC(15R) is investigated, as is the dependence of the self-focusing on the density of the free carriers in the conduction band. It has been observed that the changes of the refractive index of silicon carbide with increasing laser-beam intensity take place within not more than 10^{-9} sec. It is shown that the nonlinearity of the refractive index of the silicon carbid is due mainly to the free electrons at 300 K and to the nonlinear polarizability of the bound electrons at 77 K.

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Self-focusing of laser radiation was first observed by us in[1], where the possibility of recording the dynamic phase holograms in silicon carbide single crystals was studied. Both effects are of the same nature, and are due to nonlinear changes in the refractive index in the field of a strong laser beam; these changes, moreover, as was shown in the estimates, were rather substantial—of the order of 10^{-4} to 10^{-5}. It is therefore of interest to identify the mechanism that causes the nonlinearity of the refractive index of silicon carbide, since it determines both the relaxation time of the refractive index and the value of the coefficient n_2; these in turn determine the operating speed and diffractive efficiency of the dynamic holograms.

Our purpose was to study the dynamics of the self-focusing and to determine the nature of the nonlinearity of the refractive index of silicon carbide single crystals.

1. EXPERIMENTAL METHOD

We used the setup shown in Fig. 1 to observe the self-focusing and to study its dynamics. A single-mode Q-switched ruby laser with pulse power ~1 mW and duration ~20 nsec was used as the source. The laser beam, attenuated by neutral filters to the required power, was focused by a lens ($F=14$ cm) on a sample placed at a distance of 5 mm from the focus. The samples were plane-parallel plates of silicon carbide single crystals 0.5 mm thick polished surfaces and of high optical quality (α-SiC(6H), α-SiC(15R), and others, where H is the hexagonal and R is the rhombohedral structure of the unit cell of the SiC crystals). The beam cross section at the exit from the crystal was magnified 30 times with a microscope and photographed on film. A fraction of the beam leaving the microscope was diverted and registered with a coaxial photocell and an oscilloscope with a resolution time 10^{-9} sec. A diaphragm of 0.5 mm diameter (spot diameter 0.4 mm) was placed in front of the photocell in the plane corresponding to the film plane. The diaphragm isolated various portions of the magnified cross section so that the dynamics of the change in intensity of these portions could be studied during the time of the laser pulse. A calibrated calorimeter-galvanometer system was used to measure the energy of the laser radiation. The absolute error in the measurement of the energy amounted to 10–15%.

2. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2a shows photographs of cross sections of the beam leaving the crystal at various intensities of the light incident on the crystal. A narrowing of the beam is observed as the intensity is increased. At high intensities, a ring structure appears, testifying to the appearance of rather large aberrations in the self-focusing beam. Figures 2b and 2c show oscillograms obtained by placing the diaphragm at the center of the beam cross section (b) and by shifting the diaphragm 1 mm away from the beam axis (c). Narrowing the pulse and a sharpening of its edges are observed when the diaphragm is centrally located, as a result of the abrupt increase in the intensity at the beam axis in the course of the self-focusing. This is particularly evident at high power densities. The last photograph in column b shows for demonstration purposes the oscillograms obtained by superposition of the incident pulse (1) on the pulse obtained from the axial portion of the cross section of the beam emerging from the crystal (2).

In the second case, when the diaphragm is shifted to the periphery of the beam cross section, a different situation is observed. At powers lower than the critical self-focusing power, the change in the beam intensity at the exit from the crystal is similar to the changes at the

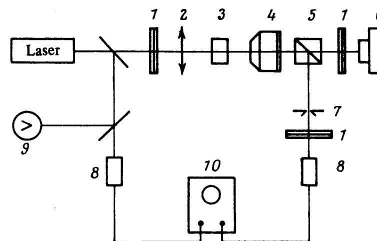

FIG. 1. Schematic diagram of the experimental setup: 1—neutral filters, 2—lens, 3—sample, 4—microscope, 5—beam splitting prism, 6—camera, 7—diaphragm (0.5 mm dia), 8—coaxial photocell FK-1, 9—calorimeter, 10—oscilloscope.

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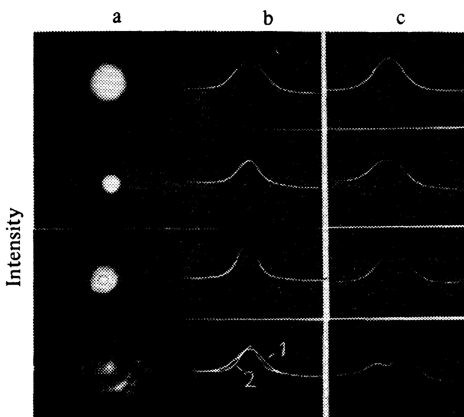

FIG. 2.

input. At incident beam intensities somewhat higher than critical, i.e., when the gradient of the refractive index over the beam cross section becomes significant, the pulse from the peripheral portion of the beam becomes flattened as a result of the shift of a portion of the light to the axis, and consequently, an increase in the intensity produces a dip in the pulse maximum, corresponding to a maximal change in the refractive index at the beam axis. When the intensity concentrated at the beam axis exceeds the threshold value, the crystal is damaged, the light is scattered, causing an overshoot on the trailing edge of the pulse, as can be clearly seen in the last oscillogram in column c.

In both cases, the dynamics of the intensity change at the crystal exit corresponds to the self-focusing process, so that we can deduce the dynamics of the change in the refractive index in the field of an intense light wave. Recognizing that the pulses on the oscillograms are symmetric, with edges steeper than the corresponding edges of the incident pulse, and also that the maximum change in the refractive index occurs at maximum intensity, and that it is not shifted relative to the maximum of the incident beam (the dip is at the center of the pulse), it can be concluded that in silicon carbide crystals the refractive index "follows" the intensity of the light wave with a time constant not larger than 10^{-9} sec, which is the resolution limit of the recording system. Thus, the mechanism of the nonlinearity of SiC, which leads to a positive change in n, turns out to be essentially instantaneous in comparison with the duration of the laser pulse.

We have estimated the nonlinear coefficient n_2 by using the measured diffraction efficiency η of the holographic phase grating recorded in the silicon carbide and the relation[2]

$$I_m = I_0 J_m^2(2\pi\Delta n T/\lambda),$$

where I_m is the intensity of the m-th order of diffraction, I_0 is the intensity of the recording pulse, J_m is the value of the m-th order Bessel function with an argument equal to the phase shift over the crystal thickness T, and Δn is the depth of modulation of the refractive index. The diffraction efficiency $\eta = I_1/I_0$ amounted to 3% for $I_0 = 50$ MW/cm². Here, $\Delta n = 7.9\times10^{-5}$ g and $n_2 = 7\times10^{-11}$ cgs esu.

The analysis of possible nonlinear mechanisms in silicon carbide reveals that the striction and thermal mechanisms can be disregarded if their inertia is taken into account. In fact, the relaxation time of the electrostriction nonlinearity is equal to $\tau_{str} = r_0/v_{ac}$, where r_0 is the radius of the light beam and v_{ac} is the velocity of sound in the crystal. In our experimental conditions, this time was equal to 10^{-7} sec. The relaxation time of the thermal nonlinearity is determined by the thermal diffusion time[3]

$$\tau_d = \rho C_p d^2/8k,$$

where ρ is the density, C_p is the specific heat, k is the thermal conductivity of the medium, and d is the diameter of the light beam in the crystal. In our case, the time is of the order of 2×10^{-6} sec. Consequently, neither density relaxation, nor all the more heat dissipation will manage to occur during the laser pulse duration $\tau_p = 20$ nsec. The pulse on the oscillograms should have had "delayed" trailing edges, which was not observed in the experiment.

The study of the self-focusing dynamics gives grounds for assuming that the nonlinearity of the refractive index may be caused by high-speed electron mechanisms. The positive change in n may be due to the nonlinear polarizability of the coupled electrons. In this case n_2 can be estimated at[4]

$$n_2 = \frac{4\pi}{\chi^{(1)} n_0}\left\{\left[\frac{2}{3}\chi^{(2)}\right]^2 - \frac{2[\chi^{(1)}]^3}{E_g N_v}\right\},$$

where N_v is the number of valence electrons per unit volume, $\chi^{(1)}$ and $\chi^{(2)}$ are the linear and nonlinear susceptibilities of second order, E_g is the width of the forbidden band, and n_0 is the refractive index. In SiC(6H) these take the following values: $\chi^{(1)} = 0.45$ cgs esu, $\chi^{(2)} = 4.9\times10^{-7}$ cgs esu, $E_g = 3$ eV, and $n_0 = 2.68$. Calculation yields $n_2 = 1.1\times10^{-12}$ cgs esu.

A positive change in n may also be caused by mechanisms that are governed by free electrons in the conduction band and result from the fact that the nonparabolic dispersion of free carriers causes the carrier relaxation time to depend on the energy and on the intervalley carrier transport. Even specially undoped single crystals of silicon carbide contain, for technological reasons, an uncontrolled nitrogen impurity that acts as a weak donor with an ionization energy ~ 0.08 eV.[5] The impurity concentration is such that at room temperature there are 10^{17}–10^{18} free carriers per cm³.

Investigations of the dependence of the self-focusing threshold on the concentration of the nitrogen impurity in SiC crystals have shown that the self-focusing threshold increases with decreasing nitrogen concentration. The results of these investigations are shown in Fig. 3. At the same time we can change the concentrations of the carriers by varying the temperature of some particular sample. We have measured the self-focusing thresholds at room temperature and at liquid-nitrogen

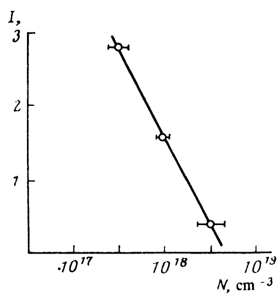

FIG. 3. Dependence of the threshold intensity (in relative units) of self-focusing on the concentration of nitrogen impurity in silicon carbide crystals.

temperature, at which the carrier density was shown by the estimates, should be one-quarter the value at 300 K. The self-focusing threshold at 77 K increased approximately one order of magnitude. Estimates of the nonlinear coefficient n_2 for the mechanisms listed above have yielded values comparable with those obtained for n_2 from the data of the diffraction efficiency of a dynamic grating.

The contribution caused by the energy dependence of the scattering can be estimated by the formula[8,9]

$$\varepsilon_2 = -\frac{ne^2}{4m^*\bar{E}_0\omega}\frac{4\pi Ne^2}{m^*\omega^2}$$

where e is the charge of the electron, ω is the frequency, $m^* = 2.28\times10^{-28}$ g is the effective mass of the electron, $N = 5\times10^{18}$ is the carrier density, $\bar{E}_0$ is the total average energy of charge carriers, equal to $(3/2)kT$ for a nondegenerate semiconductor, and n is an integer whose value and sign depend on the type of scattering. In the case of a SiC(6H) single crystal we obtain $n_2 = 5.1\times10^{-11}$ cgs esu.

The possible contribution to the refractive index, caused by the nonparabolic nature of the dispersion law, can be estimated by using the following equation[6,7]

$$\varepsilon_2 = \frac{3}{8}\left(\frac{e}{m^*c^*\omega}\right)^2\frac{4\pi Ne^2}{m^*\omega},$$

where $c^* = (E_g/2m^*)^{1/2}$ is the equivalent velocity of light. For the same values as in the preceding case, we obtain $n_2 = 2\times10^{-12}$ cgs esu.

The contribution of the intervalley mechanism can not be estimated at the present time, since the values of the electron mass effective for different valleys enter in the expression for ε_2.[10] These values have not yet been measured experimentally.

Thus, on the basis of the above results, we can conclude that the nonlinearity of the refractive index of silicon-carbide crystals at room temperature is mainly due to the free carriers in the conduction band, the predominant contribution being caused by the energy dependence of the scattering and by the smaller contribution of the nonlinear polarizability of coupled electrons, which does not depend on the temperature and becomes the principal one at 77 K.

We note in conclusion that if our ideas concerning the nature of the nonlinearity of the refractive index of silicon carbide are correct, then the relaxation time of Δn should have a value 10^{-12}–10^{-13} sec, which is characteristic of interband electron processes. This means that the operating speed of dynamic holograms recorded in silicon carbide will be much larger than that of the holograms in all recording media known at the present time.

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