Laser heating and vaporization of electron-hole drops in germanium

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We report the observation of the vaporization of electron-hole drops in germanium by CO_2 laser radiation, observed by the extinction of the luminescence of the electron-hole drop. The results are explained by heating of the electron-hole system and the crystal lattice by 10.6-μm radiation and spreading of the nonequilibrium carriers under the action of the phonon wind, generated with the absorption of radiation and relaxation of hot carriers.

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Investigation of the interaction of strong electromagnetic fields with electron-hole liquid in semiconductors is of great interest for clarifying the nature and properties of the liquid. In this work, we studied the action of powerful CO_2 laser radiation ($\lambda = 10,6\,\mu$m, $E \simeq 5 \times 10^{-3}$ J, and $\tau \simeq 100$ ns) on a system of nonequilibrium carriers in

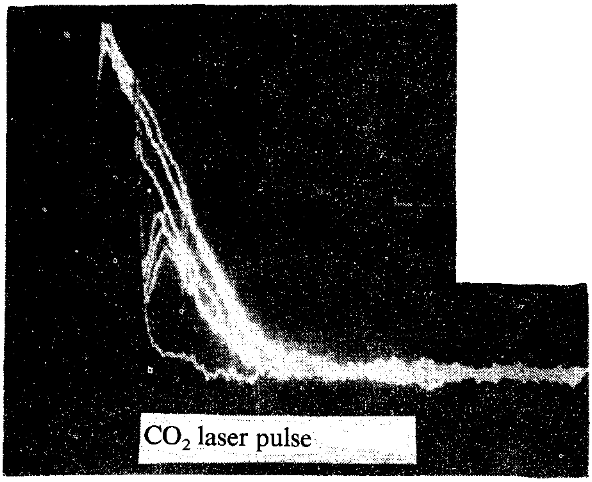

FIG. 1. Oscilloscope traces of signals showing the time dependence of luminescence of an electron-hole drop in Ge at different levels of CO_2 laser intensity (in W/cm^2). Bottom to top: 1) 5×10^5; 2) 1.2×10^5; 3) 6×10^4; 4) 3×10^4; 5) 1.4×10^4; 6) 2.4×10^3; 7) 0. The scale is 10 μs/division. The delay of the CO_2 laser pulse is $t_d = 10\ \mu s$. $T = 1.6$ K. The spectral resolution is 1.5 meV.

pure dislocation-free Ge under conditions of formation of electron-hole drops at $T = 4.2$ and 1.6 K. In order to generate carriers, we used in the experiments a pulsed YAG Nd^{3+} laser ($\lambda = 1.06\ \mu m$, $E \simeq 10^{-5}$ J, and $\tau \simeq 10$ ns), which yielded an average initial concentration in the specimen 10^{15}–10^{16} cm^{-3}. A controlled delay was introduced between the exciting YAG laser pulse and the CO_2 pulse in the range $t_d = 0$–$50\ \mu s$.

We studied the time dependence of the intensity and spectra of the recombination radiation of the specimen before and after the CO_2 laser pulse. Figure 1 shows oscillograms of the signals corresponding to the time-dependent luminescence of the electron-hole drop with different CO_2 laser intensities (W). It is evident that under the action of a CO_2 laser pulse with $W \sim 10^5$ W/cm^2, a sharp drop is observed in the intensity of the recombination radiation: irreversible for $W \geqslant 5\times10^5$ W/cm^2 and with partial regeneration for $W \gtrless 10^5$ W/cm^2.

It was found that the suppression of luminescence is not a threshold phenomenon with respect to the CO_2 laser intensity and does not depend on the delay time of the CO_2 pulse relative to the YAG laser pulse. The luminescence spectrum of the specimen after the action of the CO_2 laser radiation is strongly modified: The intensity of the electron-hole drop line (709 meV) decreases and a free exciton line (714 meV) appears (see Fig. 2). The spectra obtained are characteristic of an electron-hole system near the critical point of the liquid-gas phase transition ($T \sim 6$ K).[1]

A detailed investigation of the kinetics of these lines (Fig. 3) gives the following results: the intensity of the electron-hole drop line first increases (up to $t \sim 5\ \mu s$) after the CO_2 pulse and then decreases much more rapidly than in the case of natural recombination. The time dependence of the free-exciton line also first increases and then decreases nonexponentially, [We note that due to the adjustment occurring at the

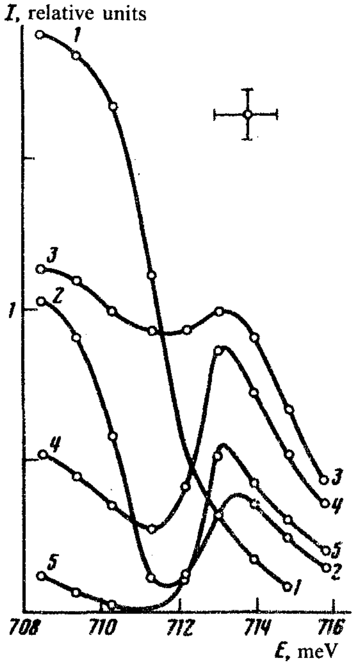

FIG. 2. Short wavelength part of the luminescence spectra of Ge at 1.6 K at different times relative to the YAG laser (t^*). Curve 1 is without CO_2 laser action at $t^* = 7.5\ \mu s$. Curves 2–5 are after the CO_2 laser pulse ($W = 1.5 \times 10^5$ W/cm^2, $t_d = 5\ \mu s$): 1) $t^* = 7.5\ \mu s$; 3) $t^* = 10\ \mu s$; 4) $t^* = 12.5\ \mu s$; 5) $t^* = 20\ \mu s$.

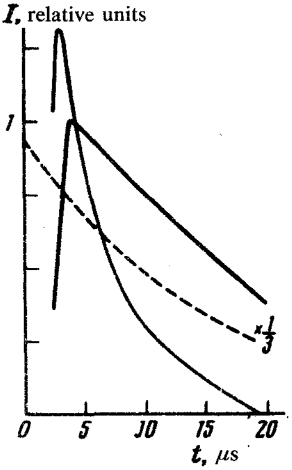

FIG. 3. Time dependence of the drop (a) and exciton (b) lines of recombination radiation of Ge after the CO_2 pulse with $W = 1.5 \times 10^5$ W/cm^2. The beginning of the measurement corresponds to the time that the CO_2 pulse terminates. $T = 1.6$ K. The dashed line shows the luminescence as a function of time for an electron-hole drop in the absence of CO_2 laser radiation.

time the CO_2 laser is switched on, the detection system does not detect the signal at the time of the CO_2 pulse (~ 100 ns), but 1–2 μs later.]

Thus, these results indicate that the electron-hole drop disintegrates under the action of CO_2 radiation and repeated condensation at elevated crystal temperatures.

In order to explain the experimental results, we must examine three processes: heating of the electron-hole system by 10.6-μm radiation, effect of the phonon wind, and the overall heating and subsequent cooling of the specimen.

During the action of the CO_2 laser, the electron-hole system is heated as a result of absorption of CO_2 laser quanta with energies $\hbar\omega = (2\pi\hbar c/\lambda) \simeq 117$ meV by the carriers. The average energy $\mathscr{E}$ of the carrier can be estimated by equating the energy absorbed from the field [absorption cross section $\sigma = 2 \times 10^{-16} \mathrm{cm}^2$ (Ref. 2)] to the losses due to phonon emission. For $W = 10^4 - 10^5$ W/cm^2, the rate of absorption of CO_2 radiation quanta by each electron-hole pair is of the order of $\nu = (\sigma W/\hbar\omega) \sim (10^9 - 10^{10})\ \mathrm{s}^{-1}$. During the time between successive acts of absorption, a carrier emits, within time $\tau_{\mathrm{opt}} \sim 10^{-12}$ s (Ref. 3), optical phonons with energy $\hbar\omega_{\mathrm{opt}} \sim 34$ meV[4] and, then, acoustic phonons with long characteristic times.[1)] Since $\nu \ll \tau_{\mathrm{opt}}^{-1}$, the balance condition can be written in the form

$$\frac{\mathscr{E}}{\tau_{ac}(\mathscr{E})} = \hbar\nu(\omega - 3\omega_{opt}), \tag{1}$$

where $\tau_{ac}(\mathscr{E})$ is the relaxation time of a carrier with energy $\mathscr{E}$ into acoustic phonons, which is approximately given by

$$\tau_{ac}^{-1}(\mathscr{E}) \simeq \eta\,\tau_p^{-1}(T)\sqrt{\frac{\mathscr{E}}{kT}}\ , \tag{2}$$

where $\tau_p(T) \sim 10^{-9}$ s is the average relaxation time at $T \sim 3$ K and $\eta \sim 1$ is a numerical factor.[5] Thus we have

$$\mathscr{E} = \left[\sigma W\left(\frac{\omega - 3\omega_{opt}}{\omega}\right)\sqrt{kT}\,\tau_p(T)\right]^{2/3} \simeq 8.6\,\mathrm{meV}, \tag{3}$$

which corresponds to a temperature ~ 100 K.

Thus, for CO_2 laser radiation intensities $W \sim 10^5$ W/cm^2, electron-hole drops are vaporized and an electron-hole plasma is formed with temperature ~ 100 K.

In the process of thermalization of high-energy carriers and subsequent decay of the acoustic quanta formed in this case, powerful phonon fluxes appear. These fluxes push part of the carriers to the surface of the crystal, where they rapidly recombine. It follows from the experiments that for $W \lesssim 10^5$ W/cm^2 a significant number of carriers is again concentrated in the electron-hole drop. Estimates show that the fraction of such phonons, which efficiently interact with carriers and push them to the boundaries of the crystal, in the total energy of emitted phonons is very small $\beta < 10^{-3}$ [the corresponding number for phonons, formed as a result of recombination, is much larger $\beta \sim 0.1$ (Ref. 6)]. This indicates that the phonons, which were produced as a result of thermalization and subsequent decay of acoustic quanta, interact weakly with the electron-hole plasma. These are presumably transverse acoustic phonons.[7]

The CO_2 radiation energy, absorbed by the electron-hole system, finally goes over into the lattice and leads, together with lattice absorption, to heating of the specimen. We shall estimate this additional heating. With an average carrier concentration $\bar{n} \sim 10^{15}$ cm^{-3} and crystal thickness $x = 2$ mm, we have $E_{eh} = \sigma \bar{n} x E_0 \simeq 5 \times 10^{-2} E_0$. The absorption in the lattice is $E_L = \alpha x E_0 \simeq 2 \times 10^{-3} E_0$ with $\alpha \simeq 0.01$ cm^{-1}.[8] The total absorption is $E_a = E_{eh} + E_L \simeq 5.2 \times 10^{-2} E_0$. For $E_0 \simeq 5 \times 10^{-3}$ J, the temperature of the crystal can be as high as ~ 10 K.[9]

The experimental results agree well with the analysis of the interaction of 10.6-μs radiation with the electron-hole liquid. Indeed, at relatively low intensities W, the specimen has time to cool rapidly.[2)] After the CO_2 laser pulse, the excitons recondense and the emission line of the electron-hole drop reappears (Fig. 1). At high intensities W, the additional heating of the crystal is appreciable and drops do not form again. In addition, the phonon wind under these conditions is sufficiently intense to push all carriers to the boundaries of the specimen.

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[1)] Strictly speaking, direct absorption of 117-meV quanta is forbidden by the law of conservation of quasimomentum and indirect transitions of electrons and holes with the emission of phonons must play a determining role in absorption. For energy balance, however, this is not important.

[2)] The time dependence of cooling of Ge specimens in liquid helium is studied in Ref. 9.

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