High-frequency breakdown of excitons and kinetics of free carriers and excitons in germanium in the presence of electron-hole drops

A. A. Manenkov, V. A. Milyaev, G. N. Mikhaĭlova, V. A. Sanina, and A. S. Seferov

P. N. Lebedev Physics Institute, USSR Academy of Sciences, Moscow
(Submitted August 15, 1975)
Zh. Eksp. Teor. Fiz. **70**, 695–701 (February 1976)

An investigation was made of the breakdown of excitons in Ge by a high-frequency electric field. A high-sensitivity method for detection of free carriers formed as a result of exciton breakdown made it possible to investigate the exciton kinetics at temperatures down to 1.3°K. This kinetics—the presence of a constant exciton concentration for a relatively long time—was explained by a model allowing for the existence of the liquid exciton phase in germanium in the form of electron-hole drops. This model was confirmed by the observation that the position of the breakdown threshold minimum on the time axis was independent of the frequency of the breakdown field. It was shown that the fall of the free-carrier density was due to a nonequilibrium mechanism, which was the release of electrons from the drops by the Auger process and their trappping by defects and impurities.

PACS numbers: 72.30.+q, 71.80.+j

1. INTRODUCTION

The microwave ($\sim 10^{10}$ Hz) breakdown of excitons was detected and investigated in detail in[1,2]. The experimental dependences of the breakdown field were explained by a model allowing for the fact that laser excitation of germanium at helium temperatures produces a three phase system composed of free carriers, free excitons, and electron–hole drops. In this model the drops act as free-carrier traps and have a considerable influence on the characteristics of the microwave breakdown. A detailed theoretical analysis of the model[2] made it possible to calculate the parameters of the drops and exciton gas from the breakdown characteristics and these parameters were found to be in good agreement with the results obtained by other methods.

One of the most important conclusions that follows from the theory of microwave breakdown of excitons based on the above model is the characteristic dependence (the existence of a minimum) of the threshold microwave breakdown power on the delay time of the microwave pulse relative to the exciting laser pulse. In this model the breakdown threshold minimum is due to the fact that the impact "ionization" of excitons by free carriers in a microwave field is initially impeded by the capture of carriers by electron–hole drops and other centers (impurities and structure defects). The threshold minimum appears after a delay sufficient for such a reduction in the total surface of drops by evaporation and carrier recombination that they cease to be effective traps.

In this model of the microwave breakdown of excitons it is assumed that, at a fixed temperature, the exciton concentration remains practically constant throughout the lifetime of electron–hole drops. This means that an equilibrium is maintained in the exciton–drop system in the course of carrier recombination and drop evaporation.

This theory predicts that the position of the breakdown threshold minimum should be independent of the breakdown field frequency. It should be noted that according to the model of exciton breakdown considered in[1] without allowance for the existence of electron–hole drops (pure exciton gas model) this minimum is attributed to the coincidence of the field frequency with the effective frequency of collisions of free carriers. In this case the position of the breakdown threshold minimum should depend on the field frequency.[3]

In view of these features of the microwave exciton breakdown, it would be interesting to investigate the frequency dependence of the breakdown threshold and to check experimentally the above hypothesis of the constancy of the equilibrium concentration of the excitons in the process of recombination of excited carriers in Ge.

We investigated the breakdown of excitons in Ge by an electric hf field (300–900 MHz). The presence of free carriers was detected with a microwave spectrometer operating at 10^{10} Hz. Measurements of the hf and microwave thresholds under the same experimental conditions (in particular, in the presence of the same optical excitation) made it possible to investigate accurately the frequency dependences of the exciton breakdown characteristics, particularly of the position of the breakdown threshold minimum. In addition to the hf breakdown and exciton kinetics, we studied the kinetics of free carriers in the presence of electron–hole drops. This was of interest in the identification of the various drop decay processes (in particular, evaporation and Auger recombination of carriers).

The microwave method of detection of the presence of free carriers had a number of advantages (high sensitivity, absence of contacts), which—in combination with the pulse method of exciton breakdown—enabled us to determine easily the low exciton concentrations and to study the kinetics in the range of concentrations

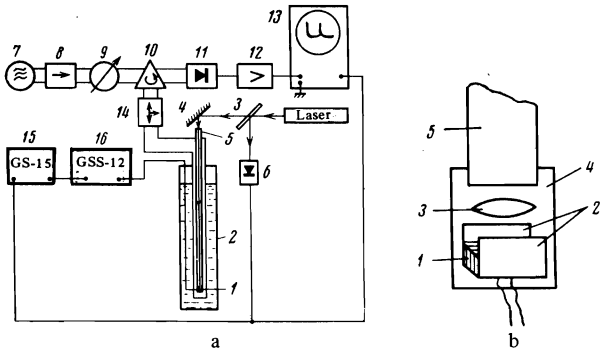

FIG. 1. a) Block diagram of the apparatus: 1) sample in a casette; 2) cryostat; 3) plane-parallel plate; 4) mirror; 5) optical fibers; 6) photodiode; 7) clystron; 8) ferrite isolator; 9) attenuator; 10)ferrite gyrator; 11) microwave detector; 12) wide-band amplifier; 13) oscillograph; 14) impedance transformer; 15) square-pulse generator; 16) hf oscillator. b) Sample holder: 1) germanium sample; 2) capacitor plates; 3) lens; 4) Teflon casette; 5) optical fiber.

($\sim 10^{12}$ cm^{-3}) in which the sensitivity of the optical methods was insufficient. The kinetics of free carriers in the presence of electron–hole drops could be studied by no other method except the microwave conductivity.

2. APPARATUS

We investigated the pulsed hf breakdown of excitons in Ge at high optical excitation levels using the apparatus shown schematically in Fig. 1a.

An investigated Ge sample was in a parallel-plate capacitor, which was subjected to an hf (300–900 MHz) electric field pulse which caused exciton breakdown. The capacitor was placed either in a section of a short-circuited waveguide or in a reflecting rectangular resonator of a microwave spectrometer (operating at 10^{10} Hz). In both cases we recorded the change in the reflected microwave power due to a change in the conductivity of the sample. The waveguide variant of the microwave spectrometer was used in the observations of exciton breakdown in an hf field. In this case the amplitude of the spectrometer signal was made a linear function of the recorded reflection signal associated with the change in the conductivity of the sample, which was proportional to the carrier density. Selection of a suitable phase of the reflected wave (by tuning a shorting plunger) made it possible to record either the

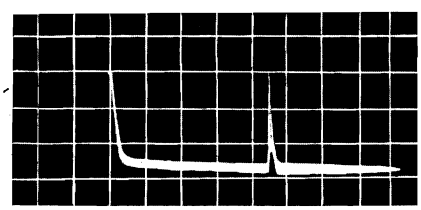

FIG. 2. Oscillogram of a signal representing the microwave conductivity of a germanium sample. The initial moment corresponds to a laser pulse; after some delay, an hf pulse is applied and this causes exciton breakdown. The scale along the abscissa is 20 μsec/div. and that along the ordinate is relative.

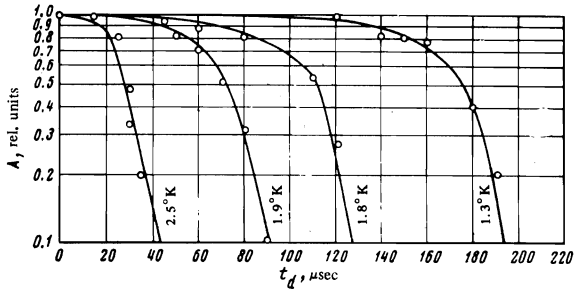

FIG. 3. Experimental dependences of the amplitude of the breakdown peak I on the delay time of the breakdown pulse t_d relative to the laser pulse, obtained at various temperatures. The maximum amplitude is normalized to unity.

total complex conductivity of the sample or its real part representing the microwave power absorption.[1)]

The resonator variant of the microwave spectrometer was used in simultaneous studies of the microwave and hf breakdown processes.

Germanium was excited optically with pulses produced by a YAG: Nd^{3+} ($\lambda = 1.06$ μ) laser. We employed a sample of $2\times2\times4$ mm dimensions in which the net impurity concentration was $N_A - N_D \sim 10^{10}$ cm^{-3}.

3. EXPERIMENTAL RESULTS AND DISCUSSION

We observed the microwave conductivity signals representing the kinetics of free carriers after a laser excitation pulse, which were in the form of two exponential functions with characteristic times of 2 μsec and 50 μsec. The fast exponential decay represented the process of binding of carriers into excitons and their condensation into electron–hole drops, whereas the slow decay represented the drop kinetics.[2]

The application of an additional hf field pulse of frequency in the 300–900 MHz range and of 1–10 μsec duration caused exciton breakdown, as manifested by the appearance of a sharp peak (spike) of the conductivity of Ge (Fig. 2). The breakdown was a threshold process in respect of the hf field intensity. The breakdown threshold minimum was observed after a delay t_d of the hf pulse relative to the laser pulse and, in the microwave breakdown case,[2] this delay time was a function of the excitation intensity. A typical value of t_d at an excitation rate corresponding to a carrier density of $\sim 10^{15}$ cm^{-3} was 100 μsec. It was found that, under given conditions, the position of the breakdown threshold minimum on the time axis was independent of the breakdown field frequency. (The hf oscillator frequency was varied within the range 300–900 MHz and microwave breakdown was investigated at 10^{10} Hz.)

We studied the dependence of the amplitude of the breakdown peak on the delay time of the hf pulse under advanced breakdown conditions, i.e., when the hf field intensity was much higher than the threshold value. The amplitude of this peak was found to be constant for a relatively long time interval (Fig. 3). This interval increased when the sample was cooled. After a long delay, the amplitude of the breakdown peak depended

exponentially on t_d and the characteristic time was 8 μsec.

The experimental results can be explained quite readily by the existence, after a laser excitation pulse, of three phases in the system: free carriers, exciton gas, and electron–hole drops. In fact, the amplitude of the breakdown peak represents the density of free carriers formed as a result of exciton dissociation. Consequently, under advanced breakdown conditions, when all the excitons are dissociated, the amplitude of the breakdown peak gives information on the exciton gas concentration in a sample.[2)] As in any other liquid–vapor system, at a given temperature the density of excitons which are in phase equilibrium with electron–hole drops should remain constant as long as the liquid phase exists. After the disappearance of the liquid phase (electron–hole drops) as a result of carrier recombination and evaporation, the exciton concentration decays at a rate characterized by a constant equal to the lifetime of free excitons.

We shall now consider in greater detail the kinetics of electron–hole drops, excitons, and free carriers. It is described by the equations[5–7]

$$\frac{dR}{dt}=-\frac{R}{3\tau_0}+\frac{v_T}{n_0}(n-n_T), \tag{1}$$

$$\frac{dn}{dt}=-\frac{n}{\tau_{ex}}-4\pi R^2N_dv_T(n-n_T)+\beta n_e^2, \tag{2}$$

$$\frac{dn_e}{dt}=-\frac{n_e}{\tau_e}-4\pi R^2N_dv_{eT}n_e+4\pi R^2\Delta N_dw_An_0-\beta n_e^2. \tag{3}$$

Here, R and τ_0 are the radius and lifetime of electron–hole drops; n, v_T, and τ_{ex}, are, respectively, concentration, thermal velocity, and lifetime of free excitons; n_e, v_{eT}, and τ_e are, respectively, the corresponding parameters of free carriers (τ_e is related to the capture by impurites, defects, etc.); N_d is the concentration of electron–hole drops; $n_0=2\times10^{17}$ cm^{-3} is the density of carriers in a drop; β is the cross section for binding carriers into excitons; w_A is the probability of the Auger recombination of carriers in electron–hole drops; Δ is the thickness of a surface layer in an electron–hole drop from which the Auger electrons are emitted; n_T is the thermal-equilibrium value of the concentration of free excitons evaporating from a drop:

$$n_T=g\left(\frac{MkT}{2\pi\hbar^2}\right)^{3/2}e^{-\psi/kT}, \tag{4}$$

where g is the multiplicity factor of the ground state of an exciton, M is the density-of-states effective mass of an exciton, and ψ is the work function of excitons emitted from a drop.

Equations (1)–(3) ignore the process of thermal dissociation of excitons as unlikely at low temperatures. Moreover, the thermal evaporation of carriers from a drop is neglected compared with the Auger process. This is justified since an estimate of the thermal-equilibrium concentration of free carriers, obtained using a formula analogous to Eq. (4), gives $n_{eT}\lesssim10^4$ cm^{-3} when the work function of the drops is $\psi_e=6$ meV and temperature is in the range $T<3.5$ °K, whereas the density of the Auger electrons is $n_{eA}\sim10^{10}$–10^{11} cm^{-3}. It should be noted that the sensitivity of our microwave spectrometer makes it possible to detect free carriers in densities $n_e\gtrsim10^{10}$ cm^{-3} and consequently, the observed slow component of the microwave conductivity signal (Fig. 2) is due to the Auger electrons.[8]

An analysis of Eq. (2) shows that a quasisteady solution for excitons ($dn/dt\approx0$) corresponds to n^{st}:

$$n^{st}=\frac{\beta n_e^2+4\pi R^2N_dv_Tn_T}{4\pi R^2N_dv_T+1/\tau_{ex}}. \tag{5}$$

We can see that under the conditions

$$\beta n_e^2\ll4\pi R^2N_dv_Tn_T,\quad 1/\tau_{ex}\ll4\pi R^2N_dv_T \tag{6}$$

we have

$$n^{st}\approx n_T.$$

If we assume that $R_0=10^{-3}$ cm, $N_d=5\times10^4$ cm^{-3}, $v_T=3\times10^6$ cm/sec,[2] $\tau_{ex}=8\times10^{-6}$ sec, $n_T=10^{12}$ cm^{-3} ($T\approx1.5$ °K)[7]), $w_A=0.8\times10^4$ sec^{-1},[9] $\beta\approx6\times10^{-4}$ cm^3/sec,[10] we find that the conditions of Eq. (6) are readily satisfied. The exciton concentration remains constant in time until $R(t)$ falls to the value

$$R\sim(\beta n_e^2/4\pi N_dv_Tn_T)^{1/2}\approx1\ \mu, \tag{7}$$

which occurs ~100–200 μsec after the end of the exciting pulse. When the drop radius becomes sufficiently small, we find, in accordance with Eq. (2), that the exciton concentration decays exponentially with a time constant τ_{ex}.

We shall now analyze the kinetics of free carriers [Eq. (3)]. It follows from the experimental results (from the fall of the microwave conductivity signal) that the density of free carriers changes significantly in a time ~10^{-4} sec, whereas the characteristic times for the capture of carriers by drops and impurities, of binding of carriers into excitons, and emission of carriers from drops because of the Auger process are all of the order of 10^{-6}–10^{-7} sec. Therefore, we can obtain a solution for n_e which varies slowly with time if we substitute $dn_e/dt\approx0$ in Eq. (3). When the trapping of carriers by impurities and defects predominates over their binding into excitons [i.e., when the term βn_e^2 in Eq. (3) can be ignored], this solution is of the form

$$n_e\approx\frac{4\pi R^2\Delta N_dw_An_0}{4\pi R^2N_dv_{eT}+1/\tau_e}. \tag{8}$$

Hence, it follows that if

$$1/\tau_e>4\pi R^2N_dv_{eT}, \tag{9}$$

which is usually well satisfied,[2] the density of free carriers is $n_e(t)\propto R^2(t)$, i.e., this density should decay with a characteristic time ~$(3/2)\tau_0$. If the process of binding of carriers into excitons predominates, the fall of the density n_e should have a characteristic time $3\tau_0$.

The observed microwave conductivity signals show that the former is true in our samples, i.e., it is more likely that carriers are captured by defects and impurities.

4. CONCLUSIONS

The breakdown of excitons in an hf electric field was observed and investigated. A high-sensitivity method used to detect free carriers resulting from the breakdown to excitons (a microwave spectrometer was used) made it possible to study the kinetics of excitons at temperatures down to 1.3 °K. The experimental results, demonstrating constancy of the exciton concentration for a relatively long time interval, were explained by a theory allowing for the existence of the liquid exciton phase in germanium (electron–hole drops). This model was confirmed by the observation that the position of the breakdown threshold minimum on the time axis was independent of the frequency of the breakdown field. The exponential decay of the free carrier density in the absence of exciton breakdown was attributed to the Auger recombination of carriers in electron–hole drops.

The authors are grateful to A. M. Prokhorov and L. V. Keldysh for their valuable advice.

[1)]A detailed analysis of a circuit for measuring the microwave conductivity of semiconductors is given in[4].

[2)]It should be noted that complete breakdown of an exciton gas results in a fairly interesting physical situation when electron–hole drops exist in an "exciton vacuum."

[1]A. A. Manenkov, V. A. Milyaev, G. N. Mikhaĭlova, and S. P. Smolin, Pis'ma Zh. Eksp. Teor. Fiz. **16**, 454 (1972) [JETP Lett. **16**, 322 (1972)].
[2]L. V. Keldysh, A. A. Manenkov, V. A. Milyaev, and G. N. Mikhaĭlova, Zh. Eksp. Teor. Fiz. **66**, 2178 (1974) [Sov. Phys.-JETP **39**, 1072 (1974)].
[3]A. D. MacDonald, Microwave Breakdown in Gases, Wiley, New York, 1966 (Russ Transl., Mir, M., 1969).
[4]B. V. Zubov, A. A. Manenkov, V. A. Milyaev, G. N. Mikhaĭlova, T. M. Murina, and A. S. Seferou, Fiz. Tverd. Tela (Leningrad) **18**, 706 (1976) [Sov. Phys.-Solid State **18**, 406 (1976)].
[5]Ya. E. Pokrovskiĭ and K. I. Svistunova, Fiz. Tekh. Poluprovodn. **4**, 491 (1970) [Sov. Phys.-Semicond. **4**, 409 (1970)].
[6]L. V. Keldysh, in: Éksitony v poluprovodnikakh pod red. B. M. Vula (Excitons in Semiconductors, ed. by B. M. Vul), Nauka M., 1971, p. 5.
[7]R. M. Westervelt, T. K. Lo, J. L. Shachli, and C. D. Jeffries, Phys. Rev. Lett. **32**, 1051 (1974).
[8]J. C. Hensel, T. G. Phillips, and T. M. Rice, Phys. Rev. Lett. **30**, 227 (1973).
[9]K. Betzler, B. G. Zhurkin, A. L. Karuzskii, and B. M. Balter, Preprint No. 71, Lebedev Physics Institute, Academy of Sciences of the USSR, M., 1975.
[10]P. S. Gladkov, Kandidatskaya dissertatsiya (Thesis for Candidate's Degree), Lebedev Phsyics Institute, Academy of Sciences of the USSR, M., 1972.

Translated by A. Tybulewicz

Dragging of excitons and electron-hole drops by phonon wind

V. S. Bagaev, L. V. Keldysh, N. N. Sibel'din, and V. A. Tsvetkov

P. N. Lebedev Physics Institute, USSR Academy of Sciences
(Submitted August 19, 1975)
Zh. Eksp. Teor. Fiz. **70**, 702–716 (February 1976)

Dragging of excitons by nonequilibrium short-wave phonons is studied theoretically. It is shown that in multivalley semiconductors with degenerate valence bands (such as Ge or Si) the cross section for scattering of short-wave phonons by indirect excitons may exceed by several orders of magnitude the scattering cross section calculated for the case of isotropic and nondegenerate bands. The kinetics of electron-hole drop (EHD) growth is also considered with allowance for the diffusion of excitons to the drop surface and dragging of the excitons by the phonon wind. By invoking the dragging effect one can explain a number of experiments in which the EHD "diffusion" coefficient is measured: moreover, the dragging effect may restrict the growth of the EHD. Results are presented of experimental observation of EHD motion induced by the phonon wind.

PACS numbers: 71.80.+j

Despite the appreciable progress in the understanding of the properties of electron-hole drops (EHD),[1–3] many facts connected with the kinetics of drop formation and growth and with the spatial distribution and motion remain unclear. These include experiments aimed at determining the coefficient of EHD "diffusion"[2,4–7] and the limitation of the EHD radius at temperatures close to threshold.[8,9] The EHD diffusion coefficient was measured by different workers,[2,4–7,9–12] but there are colossal discrepancies (six orders of magnitude) between the results of different measurements. In most studies the diffusion coefficient was determined by measuring the dimensions of the region occupied by the EHD. In[2,4,7], where a relatively low excitation intensity was used, a value $D < 1$ cm^2/sec was obtained for the diffusion coefficient. A value $D \approx 150$ is cited