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Impact ionization of excitons in Ge and Si†

D. L. Smith, D. S. Pan, and T. C. McGill*
California Institute of Technology, Pasadena, California 91125
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A charged free carrier in an applied electric field may pick up enough energy from the field to allow it to impact ionize an excition. For this to occur, the carrier must have an energy greater than the exciton binding energy. At low temperatures ($T \lesssim 10$°K in Ge and $T \lesssim 30$°K in Si) and modest electric fields ($E \sim 2$ V/cm in pure Ge and $E \sim 20$ V/cm in pure Si), the energy of a significant number of carriers exceed this threshold energy. Once this happens, the impact-ionization process can change the relative concentration of excitons and free carriers; the equilibrium law of mass action is no longer satisfied. Calculations of exciton concentration (for fixed carrier concentration) as a function of temperature and applied-field strength show that a sudden drop in exciton concentration occurs when electric fields exceed a temperature-dependent critical field.

I. INTRODUCTION

An exciton in a semiconductor can be ionized by a number of processes: phonon absorption, collision with an impurity, collision with a free carrier, etc. In pure materials with relatively low carrier concentrations in thermal equilibrium, phonon absorption is the dominant ionization process.[1] In thermal equilibrium each ionization process is balanced by a corresponding formation process; corresponding ionization and formation rates are related by the principle of detailed balance. If the exciton lifetime (recombination of electron and hole) is much longer than the total ionization time, the law of mass action is satisfied.

The application of an external electric field will change the balance between ionization and formation processes and can, therefore, modify the relative concentration of excitons and free carriers. Impact ionization by free carriers is especially sensitive to electric fields; even quite small fields can greatly increase the rate of this process. A critical field is reached when the impact-ionization process first dominates both exciton recombination and ionization by phonon absorption. Exciton concentration will plunge if fields larger than the critical field are applied.

Impact ionization of excitons has been observed in low-temperature photoconductivity experiments in Ge.[2-4] Gurnee, Glicksman, and Yu[2] have observed finite photoconductivity in Ge at low temperature (~2 °K) and low photoexcitation levels when the mass-action law predicts electrons and holes should combine to form excitons and electron-hole droplets with overwhelming probability. Impact ionization of excitons and electron-hole droplets by field-excited free carriers could account for this observation. Asnin, Rogachev, and Ryvkin[3] have measured photocurrent as a function of applied electric field at constant temperature (~2.5 °K) in Ge. They observed a large (about two orders of magnitude) increase in photocurrent when the field is increased from about 3 to about 7 V/cm and attributed this observation to impact ionization of excitons. More recently Yao, Inagaki, and Maekawa[4] have repeated this experiment at 4.2 °K; they also observed a large increase in photocurrent, but their measured critical fields are somewhat lower (~0.6 V/cm) and depend on the photoexcitation level. Impact ionization should also be important in double-injection devices at low temperatures,[5,6] and preliminary measurements[7] indicate that the relative concentration of excitons and free carriers is field dependent and not given by the law of mass action. The purpose of this paper is to investigate theoretically the effect of an external electric field on exciton concentration at different temperatures.

Section II contains a qualitative discussion of the effect of impact ionization on exciton concentration. Quantitative calculations are presented in Sec. III. Our results are summarized and compared with experiment in Sec. IV.

II. QUALITATIVE PHYSICS

Only those free carriers whose energy is greater than the exciton binding energy can impact ionize an exciton. At low temperatures ($kT \ll \epsilon_B$) few free carriers are this energetic and the impact-ionization rate is very small. An external electric field accelerates free carriers and increases the number of carriers which are sufficiently energetic to impact ionize. This carrier heating greatly increases the impact-ionization rate. Whereas the impact-ionization rate is greatly increased by an electric field, exciton ionization by phonon absorption (the dominant ionization process at thermal equilibrium) is

not signifcantly affected by an electric field. As long as the impact ionization rate is slower than the phonon absorption rate the applied field will have little effect on the excition concentration, but once the field becomes large enough that the impact-ionization rate exceeds both the phonon-absorption and exciton-annihilation rates, the exciton concentration will drop sharply with increasing field.

The rate of change of the exciton concentration is controlled by the rate of exciton recombination and the thermal (phonon absorption) and impact-ionization and formation rates. Assuming spatial uniformity (i.e., no exciton diffusion), the rate of change of exciton concentration is described by the rate equation

$$\frac{dn_{\mathrm{Ex}}}{dt}=A_T n_e n_h + A_e n_e^2 n_h + A_h n_e n_h^2 - \frac{n_{\mathrm{Ex}}}{\tau} - B_T n_{\mathrm{Ex}} - B_e n_{\mathrm{Ex}} n_e - B_h n_{\mathrm{Ex}} n_h \ , \qquad (1)$$

where n_{Ex}, n_e, and n_h are, respectively, the exciton concentration, the electron concentration and the hole concentration; A_T (B_T), A_e (B_e), and A_h (B_h) are, respectively, the coefficients for thermal, electron-impact, and hole-impact formation (ionization); and τ is the exciton lifetime. Assuming equal concentrations of electrons and holes ($n_e = n_h \equiv n$; this will be the case in intrinsic materials and double-injection devices), the steady-state exciton concentration is given by

$$n_{\mathrm{Ex}}=\frac{[A_T+(A_e+A_h)n]n^2}{B_T+1/\tau+(B_e+B_h)n} \ . \qquad (2)$$

In a state of thermal equilibrium (no external field), the ionization and formation coefficients are related by the principle of detailed balance,

$$A_i/B_i=K \ , \qquad (3a)$$

$$K=\left(\frac{2\pi m^* kT}{h}\right)^{-3/2} e^{\epsilon_B/kT} \ , \qquad (3b)$$

$$m^*=\frac{m_t^{2/3} m_l^{1/3} m_h}{(m_t+m_h)^{2/3}(m_l+m_h)^{1/3}} \ . \qquad (3c)$$

Here K is the equilibrium constant; m_t, m_l, and m_h are, respectively, the transverse-electron, longitudinal-electron, and hole (density-of-states) effective masses; and ϵ_B is the exciton binding energy. If the exciton lifetime (electron-hole recombination) is long compared to ionization times, the exciton concentration is given by the law of mass action

$$n_{\mathrm{Ex}}=Kn^2 \ . \qquad (4)$$

In any exciton ionization process, some particle (free carrier, phonon, exciton) must have an energy greater than the exciton binding energy. In thermal equilibrium, the number of such energetic particles decreases exponentially with temperatures (for $kT \ll \epsilon_B$); therefore, the exciton-ionization rate decreases approximately exponentially with temperature. At sufficiently low temperature ($T \lesssim 5$ °K in Ge, $T \lesssim 17$ °K in Si) the exciton-ionization rate will become longer than the exciton lifetime and the mass-action law [Eq. (4)] will not be satisfied. In Fig. 1, we plot exciton density versus temperature in Ge for infinite exciton lifetime (mass-action law) and for the experimental lifetime. There is a significant deviation at low temperature. At these low temperatures, the rate of exciton loss [in Eq. (1)] is governed by the temperature-independent exciton lifetime; the rate of exciton gain is a slowly varying function of temperature and therefore the exciton concentration varies slowly with temperature in this low-temperature region.

The cross sections for thermal ionization and formation and impact formation are smoothly varying functions of carrier energy; therefore, the coefficients A_T, B_T, A_e, and A_h are slowly varying functions of the external electric field.

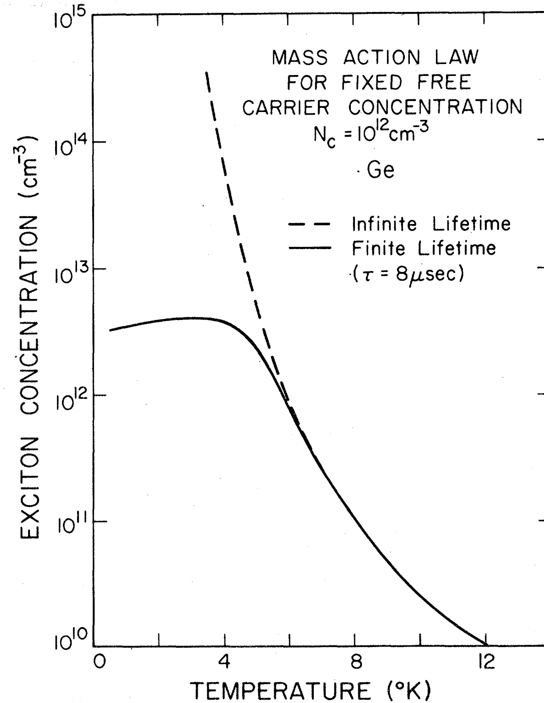

FIG. 1. Exciton density vs temperature for Ge in thermal equilibrium computed using Eq. (2); dashed line for infinite exciton lifetime, solid line for an exciton lifetime of 8 μsec . The concentration of electrons and of holes was taken to be 10^{12} cm^{-3}.

The impact-ionization cross section, however, contains a threshold (the exciton binding energy); therefore, as the field excites carriers to states with energy greater than the exciton binding energy the impact-ionization coefficients B_e and B_h will increase sharply. In Fig. 2, we show the electron distribution function at several values of applied field and the impact-ionization cross section as a function of electron energy. At zero field, a very small fraction of the electrons are sufficiently energetic to impact ionize an exciton; as an external field is applied, the fraction of electrons capable of impact ionization is greatly increased. The effect this carrier heating has on the impact-ionization rate is shown in Fig. 3. The sudden increase in the impact-ionization coefficient depends only on the existence of a threshold in the impact-ionization cross section. Because the impact-ionization coefficient increases very rapidly with field strength, small uncertainties in the thermal-ionization coefficient (which is not affected by the field) will have very little effect on the predicted behavior of the exciton concentration. The effect of the field will be especially dramatic at low temperatures, where few carriers in thermal equilibrium would be energetic enough to impact ionize, the thermal-ionization rate is low, and mobilities are high (so carriers are easily excited).

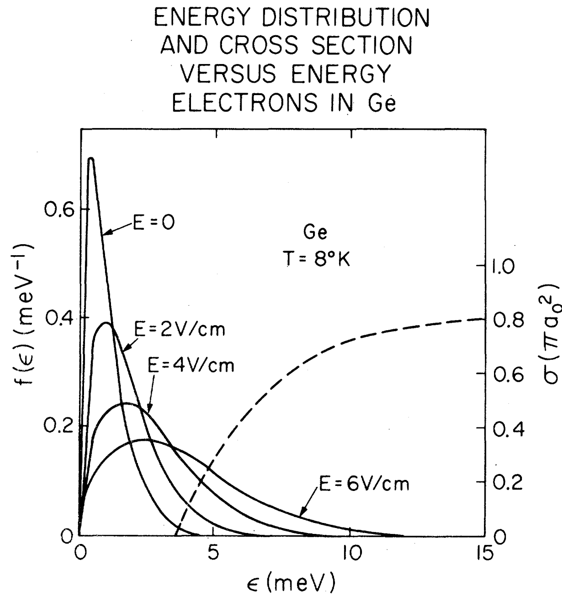

FIG. 2. Electron energy distribution at several applied electric fields (solid lines) and exciton impact-ionization cross section (dashed line) measured in units of the geometrical cross section vs electron energy in Ge at 8 °K. The threshold in the impact-ionization cross section is the exciton binding energy (3.6 meV). At zero applied field, a very small fraction of the electrons are sufficiently energetic to impact ionize an exciton; as an external field is applied, the number of electrons capable of impact ionization is greatly increased.

III. CALCULATION OF EXCITON CONCENTRATION

In order to determine the steady-state exciton concentration from Eq. (2), it is necessary to compute the kinetic coefficients A_T, A_e, A_h, B_T, B_e, and B_h (the exciton lifetime is known experimentally). We first consider the thermal formation coefficient,

$$A_T = \sum_{\vec{k}_e, \vec{k}_h} \sigma_T(\epsilon_e, \epsilon_h) \, | \vec{V}_e - \vec{V}_h | \, f_e(\vec{k}_e) f_h(\vec{k}_h) \, , \tag{5a}$$

$$\sum_{\vec{k}} f(\vec{k}) = 1 \, , \tag{5b}$$

where σ_T is the cross section for exciton forma-

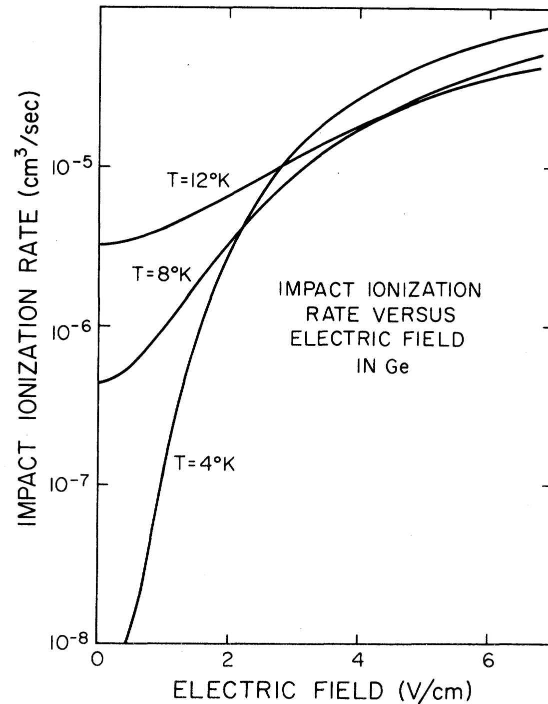

FIG. 3. Impact-ionization rate ($B_e + B_h$) vs applied electric field at several temperatures in pure Ge. As the field strength is increased, the fraction of carriers energetic enough to impact ionize rises rapidly and the impact-ionization rate is greatly increased. The effect is especially large at low temperature, where the fraction of thermal carriers (zero applied field) sufficiently energetic to impact ionize is very small and carrier mobilities are high so that they are easily excited by an electric field.

tion, V is the carrier velocity, and f is the distribution function. The cross section σ_T has been calculated by Lipnik[1]; he found that it was essentially independent of the carrier energies and of the order of 10^{-13} cm^2 for both Ge and Si. The external field increases the average carrier velocity and therefore the rate at which electrons and holes collide; since the probability for forming an exciton in an electron-hole collision is roughly independent of carrier energy, the thermal formation coefficient A_T is a slowly increasing function of field.

The thermal-ionization coefficient is not affected by a small external electric field and can therefore be determined from the thermal-formation coefficient at zero field using the principle of detailed balance,

$$B_T = A_T(E=0)/K . \qquad (6)$$

The impact-ionization coefficient is taken to be

$$B_E = \sum_{\vec{k}} \sigma_I(\epsilon) |\vec{V}| f_e(\vec{k}) , \qquad (7)$$

where σ_I is the impact-ionization cross section. To estimate σ_I, we use Percival's empirical formula for the electron impact-ionization cross section of the hydrogen atom[8] scaled by effective masses and the dielectric function

$$\sigma_I(\epsilon) = \pi a_0^2 \, (1.19 \ln x + 5.26)(x-1) \times (x^2 + 1.67x + 3.57)^{-1} \Theta(x-1) , \qquad (8a)$$

$$x = \epsilon/\epsilon_B , \qquad (8b)$$

where a_0 is the exciton radius. Because of the threshold in the impact-ionization cross section, $A_{e(h)}$ is a rapidly increasing function of electric field.

In this calculation of B_e, exciton motion has been ignored; this approximation introduces two sources of error, both of which are small and which affect the result in opposite directions. First, part of the energy necessary to ionize the exciton can be supplied by the exciton kinetic energy so that impact ionization can occur in a collision with a free carrier whose energy is less than the exciton binding energy. From this point of view, the impact-ionization coefficient should be larger than that computed using Eq. (7). With an applied external field, however, the free-carrier energies will be statistically larger than exciton kinetic energies (which are not changed by the field) and, therefore, we expect this effect to be small. Second, the exciton (ionized) will recoil following collision with the carrier so as to conserve total crystal momentum. Therefore, a carrier colliding with a stationary exciton must have energy somewhat greater than the exciton binding energy in order for ionization to occur. From this point of view, the impact-ionization coefficient should be somewhat lower than that computed using Eq. (7). This recoil effect is small for ionization owing to impact with the light carrier. In particular, for an electron moving in the light-mass (transverse) direction (the direction in which an electron is most easily accelerated) this recoil effect is quite small. While these considerations should be included in a detailed very accurate treatment of the ionization process, they have no effect on the qualitative results presented here.

The impact-formation coefficient is a slowly varying function of field; at zero field it can be determined from the impact-ionization coefficient and the principle of detailed balance. To determine this coefficient at finite fields, the energy dependence of the three-body recombination cross section must be known. Sclar and Burstein[9] have considered this problem for shallow impurities and we use their result here:

$$A_{e(h)} = \frac{B_{e(h)}}{K} \left(\sum_{\vec{k}} \frac{f(\vec{k})}{\epsilon(\vec{k})} \Big/ \sum_{\vec{k}} \frac{f_B(\vec{k})}{\epsilon(\vec{k})} \right)^2 , \qquad (9)$$

where f_B is the Boltzmann distribution for zero field. The impact-formation coefficient decreases with increasing field strength. For the carrier concentrations we will consider, the impact-formation rate will be slower than that of thermal formation.

In pure Ge and Si at low temperature, intravalley acoustic phonon scattering is the most important carrier scattering mechanism. In the temperature range we are considering, optical phonons and acoustic phonons with wave vectors large enough to cause intervalley scattering are too energetic to be thermally excited and carriers in a thermal distribution are not energetic enough to emit these phonons. An external electric field heats the carriers, but impact ionization of excitons become important before optical or intervalley phonon scattering because the exciton binding energy is smaller than both the optical phonon energy and the lowest-energy phonon with momentum large enough for intervalley scattering (TA mode). In pure Ge and Si (less than about 10^{13} impurities/cm^3), acoustic intravalley phonon scattering is more important than neutral impurity scattering for temperatures greater than about 1 °K.[10,11] Therefore, for temperatures and fields where impact ionization begins to become important, intravalley acoustic phonon scattering is the important scattering mechanism.

An analytic expression for the distribution function may be found assuming longitudinal

acoustic intravalley scattering, Boltzmann statistics, that the mean energy loss per collision is small, and that equipartition holds for the phonons,[12]

$$f(\epsilon)=N(\epsilon/kT+y)^y e^{-\epsilon/kT}, \qquad (10a)$$

$$y=(E/E_0)^2, \qquad (10b)$$

$$E_0=(6ms^2kT)^{1/2}/el, \qquad (10c)$$

$$N=\pi^{1/2}\Big/4\left(\frac{2\pi mkT}{h}\right)^{3/2}\int_0^\infty (x+y)^y x^{1/2}e^{-x}\,dx. \qquad (10d)$$

Here s is the velocity of sound, m is the carrier density-of-states effective mass, and l is the carrier mean free path. The mean free path due to acoustic phonon scattering is inversely proportional to the temperature; we set

$$E_0=CT^{3/2} \qquad (11)$$

and determine the parameter C by comparing computed with measured electron drift velocities.[13] In Fig. 4 we show the result of this comparison in Ge; a good fit is possible for fields less than about 25 V/cm. In the case of Si, it was possible to fit the measured drift velocities for fields less than about 100 V/cm. We set the hole distribution function equal to the electron distribution function. (We expect electron impact ionization to be somewhat more important than hole impact ionization owing to slightly higher electron mobilities; therefore, this approximation to the hole distribution function can introduce at most a factor-of-2 error in the kinetic coefficients. However, our estimates of the cross sections are no more accurate than a factor of 2.) Comparison of the simple distribution function with detailed Monte Carlo calculations[14] for electrons in Si show good agreement for fields less than or equal to about 25 V/cm at 20 °K. At a field of 50 V/cm, the simple distribution function overestimates the effect of the field owing to the neglect of intervalley phonon scattering, which begins to become important at these higher fields.

After the distribution function and cross sections have been determined, the kinetic coefficients can be computed by a numerical integration. Once the kinetic coefficients are known, the steady-state exciton concentration is found from Eq. (2); the results for Ge and Si are shown in Figs. 5 and 6, respectively. Parameters used

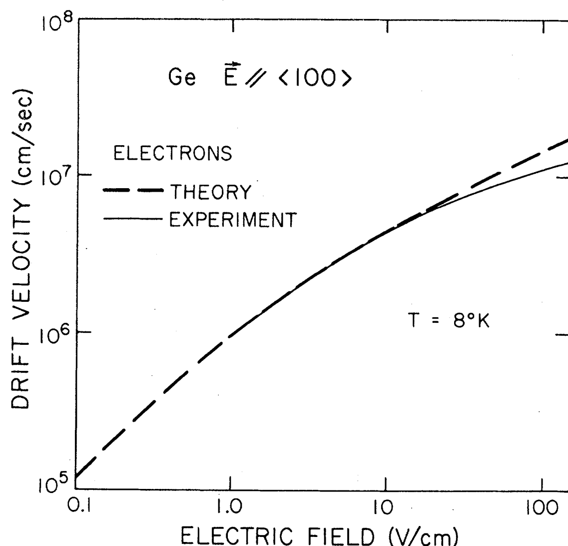

FIG. 4. Electron drift velocity vs applied electric field (applied parallel to ⟨100⟩ direction) in Ge at 8 °K. The dashed line was computed using the distribution function given in Eq. (10) and E_0 given in Eq. (11); the parameter C was fit to give the best agreement with the experimental drift velocity (solid line) measured by Nava *et al.* (Ref. 13). We see that a good fit is possible for fields less than about 25 V/cm. In the case of Si, it was possible to fit the measured drift velocities (electrons, 20 °K, field parallel to ⟨111⟩, data from Ref. 13; the quality of the fit is comparable to that for Ge) for fields less than about 100 V/cm.

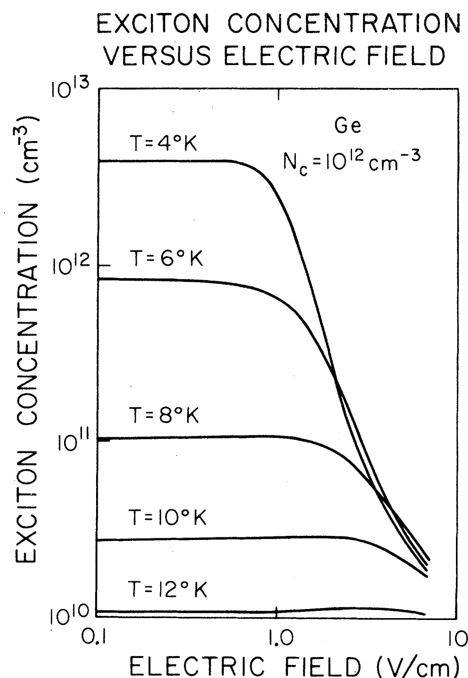

FIG. 5. Exciton concentration vs applied electric field in Ge at several temperatures; the concentration of electrons and holes was taken to be 10^{12} cm^{-3}. The sharp drop in exciton concentration when the applied field exceeds a temperature-dependent critical field is due to impact ionization by field excited carriers.

in the calculation are summarized in Table I. The carrier concentrations are typical of those appearing in low-temperature double-injection experiments.[5,6] For both Ge and Si a sharp drop in exciton concentration (due to impact ionization) occurs when fields greater than a temperature-dependent critical field are reached. The dependence of critical field on carrier concentration can be found from Fig. 3. The critical field is reached when the impact-ionization rate first exceeds both the thermal-ionization and exciton-annihilation rates. If the number of carriers is increased, the impact ionization rate per carrier (plotted in Fig. 3) at the critical field is decreased by the same factor. For example, if at $T=4$ °K and $E_c=1$ V/cm the carrier concentration is increased by an order of magnitude, the critical field decreases by about a factor of 2. The drop in exciton concentration with external field is more dramatic at low temperatures, where ionization rates in thermal equilibrium are slow; at higher temperatures ($T \gtrsim 12$ °K in Ge, $T \gtrsim 30$ °K in Si) the thermal-ionization rate is fast enough that this process is competitive with impact ionization even with relatively large applied fields. The

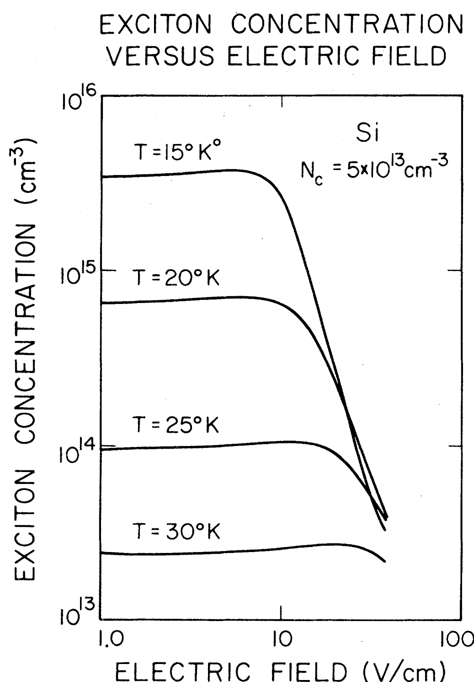

FIG. 6. Exciton concentration vs applied electric field in Si at several temperatures; the concentration of electrons and holes was taken to be 5×10^{13} cm^{-3}. The sharp drop in exciton concentration when the applied field exceeds a temperature-dependent critical field is due to impact ionization by field-excited carriers.

numerical differences between Ge and Si are almost due entirely to the larger exciton binding energy in Si.

IV. SUMMARY AND DISCUSSION

At low temperatures ($T \lesssim 10$ °K in Ge and $T \lesssim 30$ °K in Si), relatively modest electric fields ($E \sim 2$ V/cm in Ge and $E \sim 20$ V/cm in Si) can excite free carriers to the point that impact ionization of excitons becomes more important than thermal ionization; as a result the exciton concentration drops rapidly with increasing field. The sensitivity of the impact-ionization process to external fields is due to the existence of a threshold in the impact-ionization cross section. Because of the rapid increase in the impact ionization rate with applied field, small uncertainties (factor of 2) in the magnitude of the impact-ionization or thermal-ionization cross sections would not significantly affect the results of the calculation.

The results of our calculation appear to be in qualitative agreement with observations of the field dependence of low-temperature photoconductivity measurements in Ge; however, the interpretation of these measurements may be complicated by the formation of electron-hole droplets. (The boundary conditions used in the calculation,

TABLE I. Parameters used in the calculations: ϵ_B is the exciton binding energy; a_0 is the exciton radius; τ is the exciton lifetime; σ_T is the thermal recombination cross section; n is the carrier concentration; C is carrier scattering parameter; m_t is electron transverse effective mass; m_l is electron longitudinal effective mass; and m_h is hole density of states effective mass.

	Ge	Si
ϵ_B	3.6 meV [a]	14.7 meV [b]
a_0	180 Å [c]	50 Å [c]
τ	8 μsec [d]	2.6 μsec [e]
σ_T	10^{-13} cm^2 [f]	10^{-13} cm^2 [f]
n	10^{12} cm^{-3}	5×10^{13} cm^{-3}
C	5.9×10^{-2} V/cm	8.3×10^{-2} V/cm
m_t	$0.082m_0$ [g]	$0.19m_0$ [g]
m_l	$1.6m_0$ [g]	$0.97m_0$ [g]
m_h	$0.3m_0$ [g]	$0.5m_0$ [g]

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fixed carrier concentration and spatial uniformity, are really more appropriate to double-injection-device experiments, where exciton impact ionization is also expected to be important.) Our calculated critical fields (about 0.6 V/cm at $T=4$ °K) is somewhat smaller than that observed by Asnin *et al.*[3] (about 3 V/cm at $T=2.5$ °K) but in good agreement with the more recent results of Yao *et al.*[4] (about 0.6 V/cm at $T=4.2$ °K) and of Gurnee *et al.*[2] (about 1 V/cm at $T=2$ °K). The calculated decrease in exciton concentration between fields of 1 and 5 V/cm is about two orders of magnitude, in agreement with the increase in photoconductivity observed by Yao *et al.* between these two fields and also in agreement with Asnin *et al.* (for fields of E_c to E_c+4 V/cm). The dependence of critical field on carrier concentration (an order-of-magnitude increase in carrier concentration decreases the critical field by about a factor of 2) agrees with the observation of Yao *et al.* (Asnin *et al.* reported that the critical field does not vary with exciting light intensity). Finally, we note that in the experiments of Gurnee *et al.* and Asnin *et al.* the excitation levels were probably high enough to form electron-hole droplets which are also sensitive to an applied electric field[15] and may have contributed to the observed increase in photoconductivity in these experiments.

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