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Phase transition of a photogenerated electron gas in semiconductors

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We study a nonequilibrium phase-transition-like behavior of a photogenerated electron gas in semiconductors. The kinetics of the electron gas is given by a set of nonlinear rate equations. For low temperatures we show that they have three steady-state solutions when the photoexcitation energy is in a certain interval that depends on the electron-electron interaction. Two of them are stable and the other is unstable. By defining a generalized free potential we obtain the Maxwell construction that determines the order parameter, namely, the difference of the electron population in the bottom of the conduction band of the two steady-stable states. Hence, this phase transition is a nonequilibrium first-order phase transition.

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Nonequilibrium phase transitions in semiconductors have been studied in the past decades mainly in connection with the nonlinear generation-recombination mechanism, including impact ionization.[1–3] Many experimental studies have been done on hot photoexcited electron systems.[4,5] However, to the best of our knowledge no systematic study has been done as to as certain how the energy of the photoexciting pump influences the steady state of these systems.[5,6] In Ref. 7 a qualitative theoretical account of the effects of the excitation energy was given. The authors showed that the electron population in the bottom of the conduction band strongly depends on the excitation energy. The purpose of this report is to show that, for low temperatures, there exists a first-order nonequilibrium phase transition between two homogeneous stable steady states of the electron gas that appears when the energy of the pump is varied. The order parameter is the difference between the electron populations in the bottom of the conduction band of the stable states. It depends upon the effectiveness of the electron-electron (e-e) interaction. We obtain the order parameter by defining a "potential" that allows us to make a construction similar to the Maxwell construction for the equilibrium phase transition of a van der Waals gas.[2,8]

Electrons in the bottom of the conduction band of a semiconductor play an important role in the dynamics of the whole-conduction electron gas. In general, electrons with an energy in excess less than the longitudinal optical (LO) phonon energy cannot make transitions by emitting LO phonons. In the case in which the emission of LO phonons is one of the dominant mechanisms, the nonequilibrium kinetics of the electron gas in the conduction band of a semiconductor is given as follows.[9] We define a set of energy levels, each one of them representing an energy interval of width $\Delta\epsilon$ of the conduction band. Although not strictly necessary, for simplicity $\Delta\epsilon$ is set equal to $\hbar\omega_{\mathrm{LO}}$, the LO phonon energy.[9] We set the electron population in these energy levels and, based on the main interaction mechanisms, the nonlinear rate equations that give the temporal behavior of these populations are obtained.

Then, we have the following set of rate equations that describes the kinetics of a photoexcited electron gas in semiconductors.[9]

$$\begin{aligned}\frac{d\chi_i}{dt} &= \nu_o^+(\chi_{i+1}-\chi_i)+\nu_o^-(\chi_{i-1}-\chi_i)\\ &\quad +ZN_{\max}\chi_{\mathrm{tot}}(\chi_{i+1}-2\chi_i+\chi_{i-1})\\ &\quad +ZN_{\max}\chi_0(\chi_i-\chi_{i-1})+g_p\delta_{i,i_p}-w\chi_i, \end{aligned} \tag{1}$$

for $i\neq 0$. For $i=0$, since the emission of LO phonons by an electron is not possible, we have

$$\begin{aligned}\frac{d\chi_0}{dt} &= \nu_o^+\chi_1-\nu_o^-\chi_0+ZN_{\max}\chi_{\mathrm{tot}}(\chi_1-\chi_0)\\ &\quad +ZN_{\max}\chi_0\chi_0+g_p\delta_{0,i_p}-w\chi_0. \end{aligned} \tag{2}$$

The terms with ν_o^+, in Eqs. (1) and (2), describe the contribution of the electron-LO phonon emision interaction to the rate of change of the population χ_i at level i. The terms with the factor ν_o^- correspond to absorption of a LO phonon. The e-e interaction contributes to the terms that contain the factor $ZN_{\max}$. Their form come from considering the contribution to the rate of change of the population χ_i of the interaction between electron populations of all the energy levels and from the use of the energy conservation.[9] The electron populations χ_i have been normalized to the maximum reachable electron concentration $N_{\max}$ and $\chi_{\mathrm{tot}}=\Sigma\chi_i$. In the steady state $N_{\max}$ is constant and $\chi_{\mathrm{tot}}=1$. The last two terms in the right-hand member of Eqs. (1) and (2) are the generation and recombination contributions, respectively. The main interaction mechanisms, the generation and recombination terms that are given by the collision frequencies $\nu_o^{\pm}$ and $ZN_{\max}$, and by g_p, and w, respectively, depend on the lattice temperature, carrier concentration, and material parameters. We give the explicit expresions for $\nu_o^{\pm}$ and $ZN_{\max}$, which are needed for the following discussion and refer the reader to Ref. 9 for the expressions of the other frequencies. Then, for $\nu_o^{\pm}$ we have[9,10]

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$$\nu_{\mathrm{o}}^{\pm}(\epsilon)=\frac{\sqrt{m}e^{2}\hbar\omega_{\mathrm{LO}}}{\sqrt{2}\hbar^{2}}\left(\frac{1}{\mathcal{E}_{\infty}}-\frac{1}{\mathcal{E}_{\mathrm{s}}}\right)\left(N_{q}+\frac{1}{2}\pm\frac{1}{2}\right)\times\frac{1}{\sqrt{\epsilon}}S_{\mathrm{LO}}\ln\left[\frac{1+\sqrt{1\mp\frac{\hbar\omega_{\mathrm{LO}}}{\epsilon}}}{\pm1\mp\sqrt{1\mp\frac{\hbar\omega_{\mathrm{LO}}}{\epsilon}}}\right],\qquad(3)$$

where $\mathcal{E}_{\infty}$ and $\mathcal{E}_{\mathrm{s}}$ are the static and optical dielectric constants, respectively. The electron effective mass and charge are m and e, respectively, and $\hbar$ is the Planck constant. The upper (lower) sign is for emission (absorption). Phonon population effects may be taken into account in N_q, which is the phonon population at wave vector of magnitud q. The screening effects in the electron-LO phonon interaction are included in the factor S_{LO}, which in the random-phase approximation (RPA) is given by[11]

$$S_{\mathrm{LO}}^{\gamma}=\left[1+\left(\frac{\chi_{\mathrm{tot}}N_{\mathrm{max}}}{N^{\mathrm{c}}}\right)^{2}\right]^{-1},$$

where $N^{\mathrm{c}}=\mathcal{E}_{\infty}m(\hbar\omega_{\mathrm{LO}})^{3}/3^{3/2}8\pi e^{2}\hbar^{2}k_{\mathrm{B}}T_{\mathrm{e}}$ is the threshold value for the concentration in the conduction band at which the screening becomes important.[11] T_{e} is the effective electronic temperature and k_{B} is the Boltzmann's constant. The collision frequency (3) is a smooth function of the energy and the solution to Eqs. (1) and (2) can be considerably simplified if we substitute it by its average over the conduction band.

For ZN_{max}, following ideas of Takenaka *et al.*[12] and Collet and co-workers,[13] we use static RPA and obtain[9]

$$ZN_{\mathrm{max}}=\frac{e^{2}\sqrt{\pi mk_{\mathrm{B}}T_{\mathrm{e}}}}{2^{2}\hbar^{2}\mathcal{E}_{\infty}}\left[\frac{1}{1+\frac{\chi_{\mathrm{tot}}N_{\mathrm{max}}}{N_{\mathrm{ee}}}}\right],\qquad(4)$$

The square brackets factor takes into account the screening effects. These become important when the carrier concentration N_{max} reaches a critical value $N_{\mathrm{ee}}=4m\mathcal{E}_{\infty}(k_{\mathrm{B}}T_{\mathrm{e}})^{2}/\pi^{2}\hbar^{2}e^{2}$.

The expression for ZN_{max}, Eq. (4), is a simple approximation that allows us to determine, in an easy way, the ranges of the carrier concentration and electronic temperature, in which the energy exchange through e-e scattering is the dominant mechanism in the kinetics of the system. The use of static RPA is justified for experiments that take place on longer time scales.[14]

We have to point out that the structure of Eqs. (1) and (2) does not depend on this approximation and we may use in a more complete calculation a more general expresion for the e-e frequency. However, we expect that expressions (3) and (4) give good results for steady-state situations and not very high carrier concentrations as is the present case here and where many body effects and occupation effects are not important.[6] Degeneracy effects change the structure of Eqs. (1) and (2), but it is well known that for concentrations below $1\times10^{18}\ \mathrm{cm}^{-3}$ their effects are negligible.[6] Due to the e-e interaction, this set of rate equations is nonlinear. Finally, we should say that Eqs. (1) and (2), which are the main equations of our model, came from a more general theory published in Ref. 9. These general equations formulated in the energy space take into account degeneracy effects and have the mathematical structure that other formulations have (See, for example, Refs. 13 and 14). They can be generalized to take into account quantum effects, e.g., exchange, by using more appropiate scattering frequencies. However, quantum effects are expected to be important in very short times and very small distances.[6,14]

Let us consider the finite differences as derivatives,

$$\chi_{i+1}-\chi_{i}\rightarrow\frac{d\chi}{d\epsilon}\Delta\epsilon$$

$$\chi_{i+1}-2\chi_{i}+\chi_{i-1}\rightarrow\frac{d^{2}\chi}{d\epsilon^{2}}(\Delta\epsilon)^{2}$$

$$\chi_{i}-\chi_{i-1}\rightarrow\frac{d\chi}{d\epsilon}\Delta\epsilon-\frac{d^{2}\chi}{d\epsilon^{2}}(\Delta\epsilon)^{2}.$$

Then, we obtain the partial-differential equation

$$\frac{\partial\chi}{\partial\tau}=(\chi_{\mathrm{tot}}-\chi_{0}+\mu)\ \frac{\partial^{2}\chi}{\partial\varepsilon^{2}}+(\nu-\mu+\chi_{0})\ \frac{\partial\chi}{\partial\varepsilon}+\chi_{p}\delta(\varepsilon-\varepsilon_{p})-\omega\chi,\qquad(5)$$

where we have defined $\tau\equiv tZN_{\mathrm{max}}$, $\nu\equiv\nu_{\mathrm{o}}^{+}/ZN_{\mathrm{max}}$, $\mu\equiv\nu_{\mathrm{o}}^{-}/ZN_{\mathrm{max}}$, $\omega\equiv w/ZN_{\mathrm{max}}$, $\chi_{p}\equiv g_{p}/ZN_{\mathrm{max}}$, and $\varepsilon\equiv\epsilon/\Delta\epsilon$. By this normalization, we have eliminated one parameter. For low temperatures,[9] $\mu\ll\nu$. Also, in steady state, $\chi_{p}=\omega$. Hence, in this case, we have only two relevant parameters, ν and ε_{p}.

The electron distribution function $\chi(\varepsilon,\tau)$ is continuous and positive definite in the whole interval $0\leqslant\varepsilon<\infty$ and has a discontinuity in its first derivative at $\varepsilon=\varepsilon_{p}$. In addition, it must satisfy the conditions

$$\chi_{\mathrm{tot}}=\int_{0}^{\infty}d\varepsilon\,\chi(\varepsilon,\tau),\qquad\chi_{0}=\int_{0}^{1}d\varepsilon\ \chi(\varepsilon,\tau).\qquad(6)$$

The steady-state solution to Eq. (5) is[7]

$$\chi^{s}(\varepsilon)=\begin{cases}Ae^{\alpha\varepsilon}+Be^{-\beta\varepsilon}, & 0\leqslant\varepsilon\leqslant\varepsilon_{p}\\ Ce^{-\beta\varepsilon}, & \varepsilon_{p}\leqslant\varepsilon,\end{cases}\qquad(7)$$

where

$$\left.\begin{matrix}\alpha\\ \beta\end{matrix}\right\}=\frac{[(\nu-\mu+\chi_{0}^{s})^{2}+4\omega(1-\chi_{0}^{s}+\mu)]^{1/2}\mp(\nu-\mu+\chi_{0}^{s})}{2(1-\chi_{0}^{s}+\mu)}.\qquad(8)$$

In the steady state $\chi_{\mathrm{tot}}^{s}=1$. The electron population at the lowest level χ_{0}^{s} is determined self-consistently by the equation

$$\chi_{0}^{s}=\frac{\omega}{\alpha}e^{-\alpha\varepsilon_{p}}\frac{e^{\alpha}-e^{-\beta}}{\alpha+\beta}\,\frac{1}{1-\chi_{0}^{s}}.\qquad(9)$$

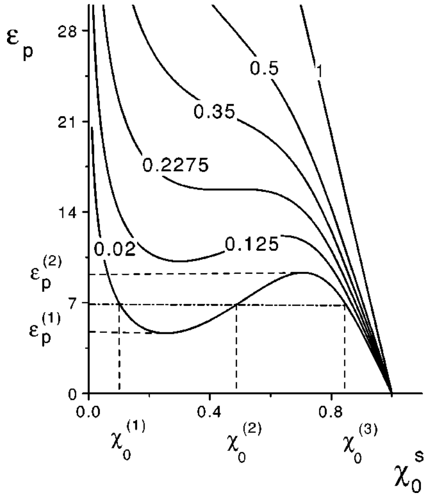

FIG. 1. Photoexcitation pump energy ε_p as a function of χ_0^s. Numbers on each curve are the values of ν.

The coefficients *A*, *B*, and *C* are obtained using the continuity in $\chi(\varepsilon)$, the discontinuity in its first derivative at $\varepsilon=\varepsilon_p$, and Eq. (6).

In Fig. 1 we show the low temperature dependence of ε_p on χ_0^s for several values of ν. We take ω=0.02 for the recombination. We must notice that we first set ε_p and then find self-consistently χ_0^s. However, we chose to plot ε_p as a function of χ_0^s. The reason will be apparent below. Also notice that, for example, the curve with ν=0.02, for $\varepsilon_p^{(1)}<\varepsilon_p<\varepsilon_p^{(2)}$ has three possible values of χ_0^s, that are labeled $\chi_0^{(1)}$, $\chi_0^{(2)}$, and $\chi_0^{(3)}$ (the interceptions with the horizontal dashed-dotted line). We will show that two of them are stable and the other one is unstable. In addition, for $\nu_c\approx0.2275$ we have the critical curve that separates curves that have regions of ε_p with three possible solutions for χ_0^s from curves with just one solution. We must point out that $\nu=\nu_o^+/ZN_{\max}$, then Fig. 1 can be seen as a family of curves in which the *e*-*e* interaction is changed, for example, by changing the electron concentration. For GaAs at 4 K we obtain $\nu\approx0.02$ using $T_e=300$ K and $N_{\max}=4.1\times10^{17}$ cm^{-3} in expressions (3) and (4). For a carrier concentration of 6.11×10^{16} cm^{-3} we obtain approximately the critical curve.

Next, we consider the perturbation to the steady state, $\chi(\varepsilon,\tau)=\chi^s(\varepsilon)+\delta\chi(\varepsilon,\tau)$ produced by a time-dependent excitation, $\chi_p(t)=\chi_p^s+\delta\chi_p(t)$, where χ_p^s is the constant excitation that produces the steady state. Notice that χ_{tot} and χ_0 are time dependent and they are given by the relations $\chi_{\text{tot}}=1+\delta\chi_{\text{tot}}$ and $\chi_0=\chi_0^s+\delta\chi_0$, respectively.

From Eq. (2) for χ_0 and assuming that $\varepsilon_p>1$, we have for its steady state

$$\nu\chi_1^s-\mu\chi_0^s+\chi_1^s-\chi_0^s+\chi_0^{s2}-\omega\chi_0^s=0, \tag{10}$$

and the time evolution of its perturbation $\delta\chi_0$ is given to first order by

$$\frac{d}{d\tau}\delta\chi_0=\left[\nu\frac{\delta\chi_1}{\delta\chi_0}-\mu+\frac{\delta\chi_1}{\delta\chi_0}-1+2\chi_0^s-\omega\right]\delta\chi_0. \tag{11}$$

We now define the function $\psi(\chi_0)$ that allows us to perform the stability analysis. From Eq. (10)

$$\psi(\chi_0)\equiv\nu\chi_1-\mu\chi_0+\chi_1-\chi_0+\chi_0^2-\omega\chi_0 \tag{12}$$

and the steady-state condition [Eq. (10)] becomes $\psi(\chi_0^s)=0$. Performing the derivative of $\psi(\chi_0)$ with respect of χ_0 we find that the factor in square brackets in Eq. (11) is equal to this derivative evaluated at $\chi_0=\chi_0^s$. Therefore,

$$\frac{d}{d\tau}\delta\chi_0=\left.\frac{d\psi}{d\chi_0}\right|_{\chi_0=\chi_0^s}\delta\chi_0 \tag{13}$$

and the solution for $\delta\chi_0$ is

$$\delta\chi_0(\tau)=\delta\chi_0(0)\exp\left[\left.\frac{d\psi}{d\chi_0}\right|_{\chi_0=\chi_0^s}\tau\right]. \tag{14}$$

A steady state, given by a χ_0^s, is stable if

$$\left.\frac{d\psi}{d\chi_0}\right|_{\chi_0=\chi_0^s}<0. \tag{15}$$

Equation (9) can be written as

$$-\frac{1}{\alpha}\ln\left[(1-\chi_0^s)\chi_0^s\frac{\alpha}{\omega}\frac{\alpha+\beta}{e^{\alpha}-e^{-\beta}}\right]-\varepsilon_p=0. \tag{16}$$

In the steady state, Eqs. (12) and (16) are equivalent, therefore

$$\psi(\chi_0)=-\frac{1}{\alpha}\ln\left[(1-\chi_0)\chi_0\frac{\alpha}{\omega}\frac{\alpha+\beta}{e^{\alpha}-e^{-\beta}}\right]-\varepsilon_p. \tag{17}$$

In Fig. 1 we plotted ε_p as a function of χ_0^s, which means that we plotted the function $\psi(\chi_0)$ and this establishes the stability of the states $\chi_0^{(1)}$, $\chi_0^{(2)}$, and $\chi_0^{(3)}$. The middle root is an unstable steady state, while $\chi_0^{(1)}$ and $\chi_0^{(3)}$ are stable steady states.

A thermodynamic system remains homogeneous and stable if the criteria of instrinsic stability is satisfied,[15] $(\partial P/\partial V)<0$, where *P* is the pressure of the system and *V* is its volume. When this condition is violated a phase transition ocurrs.[15] Then, from Eq. (15), we have the following correspondence: $\chi_0\rightarrow V$, $\varepsilon_p\rightarrow P$, and $\nu\rightarrow T$, where *T* is the temperature of the thermodynamic system.[2] We see that Eq. (16) turns out to be the equation of state. The homogeneous states $\chi_0^{(1)}$ and $\chi_0^{(3)}$ are nonequilibrium stable steady states of the system. Therefore, we call the transition between states $\chi_0^{(1)}$ and $\chi_0^{(3)}$, a first order out of equilibrium phase transition. The order parameter is $\chi_0^{(3)}-\chi_0^{(1)}$ and still remains unknown.

So far, we have assumed that the populations χ_0, χ_1, and so on, are constants and homogeneous in space. Now, we assume that there exist spatial inhomogeneities, $\chi_0=\chi_0(\mathbf{r})$, which produce spatial gradients, and diffusion of this population. We also assume, from the structure of Eq. (5), that the spatial and temporal behavior of the populations χ_i, $i>0$, is given through χ_0. Then, we have the following equation

$$\frac{\partial \chi_0}{\partial \tau} = \psi(\chi_0) + \kappa \frac{\partial^2 \chi_0}{\partial z^2}. \tag{18}$$

We suppose, for simplicity, that χ_0 depends only on the spatial coordinate z. Here, κ is the χ_0 diffusion constant.

Let us introduce the "potential" $\Phi(\chi_0)$ with the definition[3,8]

$$\psi(\chi_0) = \frac{\partial}{\partial \chi_0} \Phi(\chi_0). \tag{19}$$

Then, the steady state satisfies the equation

$$\kappa \frac{\partial^2 \chi_0}{\partial z^2} = -\frac{\partial}{\partial \chi_0} \Phi(\chi_0). \tag{20}$$

We have seen from Fig. 1 that, when $\nu = 0.02$ and ε_p is in the energy interval, $\varepsilon_p^{(1)} < \varepsilon_p < \varepsilon_p^{(2)}$, the system has two homogeneous stable steady states. Let us find a solution $\chi_0(z)$ such that $\chi_0(+\infty) = \chi_0^{(1)}$ and $\chi_0(-\infty) = \chi_0^{(3)}$. In such a case two steady states coexist. Obviously, the "potential" $\Phi(\chi_0)$ has two maxima in $\chi_0^{(1)}$ and $\chi_0^{(3)}$. Coexistence ocurrs for a value of ε_p such that the two maxima are indistinguishable for the system, $\Phi(\chi_0^{(1)}) = \Phi(\chi_0^{(3)})$. Then

$$0 = \Phi(\chi_0^{(3)}) - \Phi(\chi_0^{(1)}) = \int_{\chi_0^{(1)}}^{\chi_0^{(3)}} d\chi_0 \psi(\chi_0). \tag{21}$$

The last equation is the Maxwell construction for the vapor pressure in the van der Waals gas from which the order parameter $\chi_0^{(3)} - \chi_0^{(1)}$ can be calculated. We also notice that $-\Phi$ corresponds to the Hemholtz free potential. Moreover, the condition that Φ is at a maximum in a stable steady state corresponds to that the generalized free potential $H = -\Phi$ is at a minimum. This is consistent with the condition that an equilibrium thermodynamic system is in a state of minimum Hemholtz free energy.[15]

In summary, we have found a low-temperature nonequilibrium phase transition between two homogeneous stable steady states of an electron gas in semiconductors. We introduced the function ψ and the stability condition for these states was given by Eq. (15). A generalized free potential was defined by Eq. (19) from which we obtained Eq. (21) that corresponds to the vapor pressure Maxwell construction of a van der Waals gas. The order parameter, the difference of the electron population in the bottom of the conduction band, is then calculated and goes to zero when ν approaches the critical value $\nu_c \approx 0.2275$. This is the reason we call this phase transition a first-order nonequilibrium phase transition. For GaAs at 4 K, this critical value corresponds to an electron effective temperature of 300 K and an electron concentration of 6.11×10^{16} cm^{-3}. The bistability occurrs in the range $\nu < 0.2275$, which corresponds to carrier densities greater than 6.11×10^{16} cm^{-3}. We expect that the equations of the model are valid for polar semiconductors, such as GaAs, under low-temperature cw photoexcitation that creates a not-so-high carrier concentration (less than 1×10^{18} cm^{-3}), where many body and occupation effects are not important and the electron-LO phonon and e-e interactions dominate the dynamics of the carriers.[6] Moreover, the static RPA used to calculate these interactions is applicable under the present conditions.[11–13] We have to point out that the mathematical form of Eqs. (1) and (2) and the e-e interaction, which gives the nonlinear character of the rate equations, are the necessary main ingredients for the existence of the phase transition. The e-e interaction may be improved by using a more general approximation for the scattering frequencies that take into account, for example, exchange effects.[6,13] For higher concentrations degeneracy effects are easily taken into account as is described in Ref. 7. Also, the assumption that the distribution function changes very slowly over an energy interval equal to the LO phonon energy may be improved by sampling the distribution function over several tens of points. Finally, we may use a nonstatic RPA in the evaluation of the scattering mechanisms but at the cost of computing several integrals.[6,13]

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