Cyclotron-Resonance-Induced Nonequilibrium Phase Transition in *n*-GaAs

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A thresholdlike behavior of the far-infrared photoconductivity due to cyclotron resonance and a drastic deviation of the cyclotron-resonance line shape from a Lorentzian has been observed in *n*-GaAs at low temperatures by applying a high-power cw far-infrared laser. Both effects may consistently be explained in terms of generation-recombination–induced nonequilibrium phase transitions showing that, besides impact ionization of impurities, cyclotron resonance can critically control the conductivity of the semiconductors.

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The most important autocatalytic process in extrinsic semiconductors is impact ionization of impurities, which is responsible for the breakdown observed in the current-voltage characteristics of many high-purity semiconductors.[1–4] At low temperatures almost all carriers are bound to shallow impurities, and this yields a low conductance of the sample. At a critical electric field strength free carriers gain sufficient energy so that the impact ionization rate of shallow impurities exceeds the recombination rate for low carrier concentration, which results in a rapid increase of the current. The transition from the low-conducting state to the high-conducting state has been recognized as a nonequilibrium phase transition governed by nonlinear generation-recombination rate equations.[5,6] The order parameter of the transition corresponds to the steady-state free-carrier concentration n. By increase of the electric field E, and thereby the impact ionization probability per electron, the system is driven from the phase of low carrier concentration into that of high concentration. Thus the electric field can be identified as a control parameter of the phase transition. Dependent upon the material parameters and the excitation conditions, the phase transition may be of first or second order, which corresponds to a discontinuity in $n(E)$ or dn/dE, respectively. The first case is connected with hysteresis of the current-voltage characteristic, and spatial phase coexistence,[6] while the second case corresponds to passing through a critical point at a threshold field E_c, analogous to the Curie point of a ferromagnetic phase transition. Other types of nonequilibrium phase transitions associated with impurity breakdown at low temperatures, in particular the onset of chaos,[7–9] have also been observed recently.

In a previous investigation of *n*-GaAs it was shown that photoconductivity due to low-power far-infrared (FIR) excitation of cyclotron resonance probes a generalized susceptibility of the second-order nonequilibrium phase transition following a classical Curie-Weiss law.[10] In the present work we investigate high-power FIR laser irradiation under cyclotron-resonance conditions, and establish that the optical excitation probability of cyclotron resonance σF is an additional control parameter of the nonequilibrium phase transition, where F is the photon flux density and σ is the cyclotron-resonance absorption cross section which depends upon the magnetic field and the FIR frequency. By analyzing the σF-E plane of control parameters we find that the FIR irradiation shifts the threshold of the impact ionization instability E_c (i.e., the critical point) to lower values, which thus generates a whole line of critical points in the σF-E plane similar to the λ line of equilibrium phase transitions in superfluid helium.[11] On crossing the critical line from the low-conducting state at constant E the photoconductivity shows a thresholdlike behavior due to the combined action of cyclotron-resonance excitation and impact ionization. This novel highly nonlinear photoconductive mechanism gives further insight into the kinetics of electrons bound to shallow donors and may be useful as a threshold detector and optical correlator, opening up a new field of nonlinear FIR optoelectronics. Furthermore, the impact-ionization coefficient X as a function of the electric field strength and the lifetime of electrons in the $N=1$ Landau level could experimentally be determined.

The measurements were carried out on a high-purity *n*-GaAs epitaxial layer with alloyed Au-Sn Ohmic strip contacts on opposite edges of the sample in order to get a homogeneous electric field. The sample was mounted in a metallic light pipe and immersed in liquid helium at the center of a superconducting solenoid. Cyclotron resonance was excited by the $\lambda = 570$ μm line of CH_3OH laser pumped by a pulsed CO_2 laser. The duration of the laser pulses was 300 μs, much larger than any expected relaxation time; thus steady-state conditions during optical excitation may be assumed. Crosschecks of the laser power with several calibrated pyroelectric detectors and a Golay cell allowed the determination of the power with an ac-

curacy of about 10%. Photoconductivity was measured in Faraday configuration with the electric field normal to the magnetic field by application of a standard load resistor circuit. The load resistance in all cases was chosen to be much smaller than the sample resistance. Therefore, and because the mobility of n-GaAs at low temperatures is not appreciably affected by cyclotron-resonance absorption,[12] it follows that $\Delta V/V \propto \Delta n$, where ΔV and Δn are the changes due to irradiation of the voltage across the sample and of the free-electron concentration, respectively.

The photoconductivity signal at the center of the resonance is shown in Fig. 1 for various fixed electric field strengths below the critical field $E_c^{(0)}$ as a function of the intensity $I=\hbar\omega F$. The photosignal obviously sets in at a threshold intensity $\hbar\omega F_c$ and saturates at high intensities. Both the intensity threshold and the saturation intensity decrease with rising electric field. Figure 2 shows the cyclotron-resonance line for three different electric field strengths and in each case for various laser intensities. For the lowest electric field [Fig. 2(a)] and for low intensities the cyclotron resonance shows up as a Lorentzian-shaped line of half-width $\Delta B=15$ mT, as it is usually observed in high-purity n-GaAs under application of low-power lasers. With increasing intensity the linewidth broadens and the line shape deviates from a Lorentzian. This effect is more drastically shown at higher electric field strengths [Fig. 2(b)]. In particular, close to $E_c^{(0)}$ [Fig. 2(c)] the line shape does not at all resemble a Lorentzian. This strange behavior of the line shape is an immediate consequence of the fact that cyclotron-resonance excitation critically controls the sample conductance and leads to second-order phase transitions along the critical line $(E_c, (\sigma F)_c)$. At constant intensity and field the optical transition probability σF varies with the magnetic field strength B like a Lorentzian centered at the resonance field B_{CR}. With approach to B_{CR} the signal vanishes or is very small at nonzero but low temperatures as long as $\sigma F < (\sigma F)_c$, where $(\sigma F)_c$ is independent of the magnetic field strength but depends upon E_c. When, upon further increase of B, σF crosses the threshold $(\sigma F)_c$, the sample is converted into the high conducting phase and the conductivity rapidly increases upon further increase toward B_{CR}. Thus, when the control parameter σF is modulated, by the magnetic field B according to a Lorentzian $\sigma(B)$, the photoconductivity (the order parameter) shows a distinctly non-Lorentzian structure with a base width ΔB which is given by the condition $\sigma(B_{CR}-\Delta B/2)F=(\sigma F)_c$. Hence $\Delta B \propto [\sigma(B_{CR})F/(\sigma F)_c - 1]^{1/2}$ increases with increasing F and with decreasing $(\sigma F)_c$, which corresponds to rising field strength. The situation is quite similar to the tuning of a laser through resonance, and the observed photoconductivity lines closely resemble the tuning curves for different pumping rates of a single-mode laser, which is a more familiar example of a nonequilibrium phase transition.[13]

We apply a three-level model including the donor ground state and the two lowest Landau levels. The concentrations of electrons bound to donors and in the $N=0$ and $N=1$ Landau levels, and the density of ionized donors are denoted by n_D, n_0, n_1, and p_D, respec-

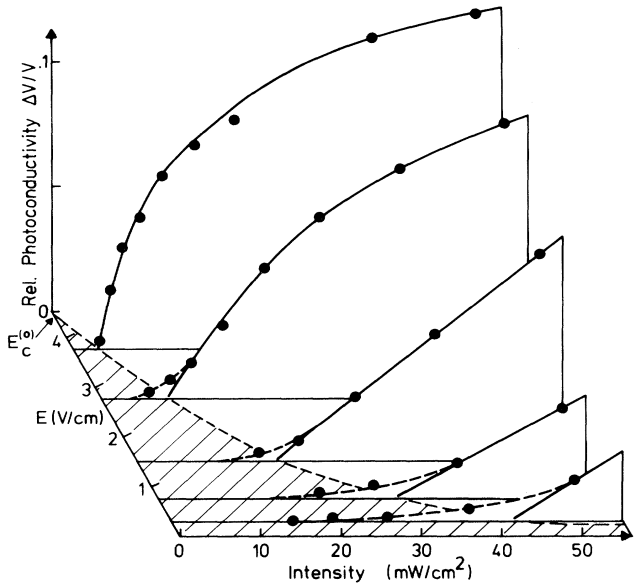

FIG. 1. Relative photoconductive signal $\Delta V/V \propto \Delta n$ at 4.2 K in the center of the resonance for various electric field strengths E as a function of the effective intensity of cyclotron-resonance-active polarization in the sample. Effective donor concentration of the sample, $N_D - N_A = 8.3 \times 10^{13}$ cm^{-3}; compensation, $N_A/N_D = 0.7$.

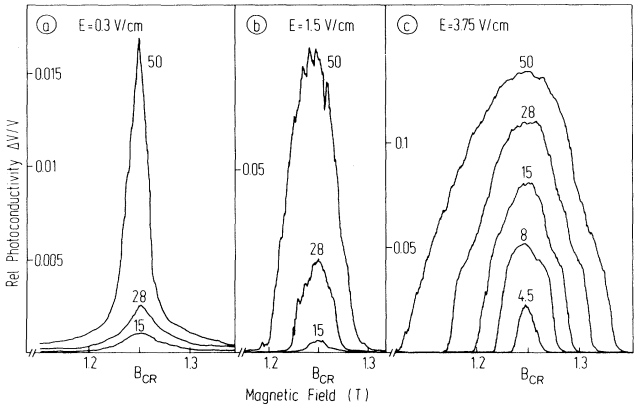

FIG. 2. Cyclotron-resonance-induced photoconductivity at 4.2 K for three different electric bias fields (a)–(c) and various irradiation intensities. The numbers identifying the curves give the effective optical intensity in the sample in units of milliwatts per squared centimeter.

tively. Then the rate equations are given by[10]

$$\dot{n}_1 = X_2^s n_0 - T_2^s n_1,$$
$$\dot{n}_0 = X_1^s n_D + X n n_D + T_2^s n_1 - T_1^s n_0 p_D - X_2^s n_0, \quad (1)$$

where X, X_1^s, T_1^s, X_2^s, and T_2^s are generation-recombination coefficients corresponding to impact ionization, thermal ionization, and recombination of donors, and excitation and relaxation of the $N=1$ Landau level, respectively. We assume that the $N=1$ Landau level is populated by optical transitions solely, ignoring thermal excitations, and we take into account stimulated emission to allow for saturation of the cyclotron-resonance absorption. Hence $X_2^s = \sigma F$ and $T_2^s = \tau_1^{-1} + \sigma F$, where τ_1 is the lifetime of electrons in the $N=1$ Landau level. In the steady state ($d/dt = 0$) and under the local neutrality condition Eq. (1) may easily be solved, which gives the free-electron concentration $n(F) = n_0 + n_1$.

At low temperature the probability of impact ionization nX exceeds that of thermal ionization X_1^s of the donors for electric field strengths even well below E_c; therefore X_1^s will be neglected. In this case for $F=0$ the stable solution is $n=0$ as long as $X < X_c^{(0)} := T_1^s N_A / P_A$, and thus photoconductivity is due to $\Delta n = n(F)$ probing the free-electron concentration. Under this condition Eq. (1) has two stable solutions,

$$n(F) = 0 \quad \text{for } \sigma F < (\sigma F)_c,$$
$$n(F) = \Delta n = \eta P_A \tau_{\text{eff}} [\sigma F - (\sigma F)_c](1 + F/F_s)^{-1} \quad (2)$$
$$\text{for } \sigma F > (\sigma F)_c,$$

which represent the order parameter in the two phases and exhibit the observed thresholdlike behavior of the photosignal. The critical optical excitation probability depends on X as

$$(\sigma F)_c = \tau_1^{-1} (X_c^{(0)} - X)/(2X - X_c^{(0)}) \quad (3)$$

and determines the critical line in the σF-E plane when X is given as a function of E. Cyclotron-resonance-induced transitions are possible in the range $X_c^{(0)}/2 < X < X_c^{(0)}$, where $(\sigma F)_c$ diverges at $X_c^{(0)}/2$ because of the saturation of cyclotron resonance and vanishes at $X_c^{(0)}$. In Eq. (2), $P_A = N_D - N_A$ is the effective donor concentration, $F_s = (\sigma \tau_{\text{eff}})^{-1}$ is the saturation photon flux density, $\tau_{\text{eff}} = \tau_1 [(2X + T_1^s)/(X + T_1^s)]$ is an effective lifetime of the electrons in the conduction band, and $\eta = (2X - X_c^{(0)})/(2X + T_1^s)$ is a dimensionless quantum efficiency.

The experimentally observed intensity dependence of the photosignal has been fitted by the relation given above (see solid lines in Fig. 1), which yields $(\sigma F)_c$ and F_s for various electric field strengths. At lower fields and low laser intensities the condition $X_1^s \ll nX$ obviously is not fulfilled. Here the photosignal first increases linearly (dashed lines in Fig. 1) with intensity as a result of electrons thermally excited into the $N=0$ Landau level via X_1^s. At higher intensities, however, the signal proceeds superlinearly into the transition region. Also, in this case $(\sigma F)_c$ can be estimated from our measurements by extrapolation of the high-intensity data. The softening of the sharp second-order phase transition by thermal generation is analogous to the smoothing out of the laser threshold by spontaneous emission.[14] The second-order phase transition at $(\sigma F)_c$ becomes sharper, i.e., $(dn/dF)_{(\sigma F)_c}$ becomes larger, as E increases towards $E_c^{(0)}$ and diverges as

$$\left(\frac{dn}{dF}\right)_{(\sigma F)_c} \propto |E - E_c^{(0)}|^{-1} \quad \text{for } E \to E_c^{(0)}. \quad (4)$$

This identifies a critical exponent $\gamma = 1$ of the generalized susceptibility $\chi \propto dn/dF$, in agreement with our previous findings in the low-FIR-power regime.[10]

In Fig. 3 the critical optical transition probability $(\sigma F)_c$ obtained by these procedures is plotted as a function of the electric field strength E, and it separates both phases in the σF-E plane. $(\sigma F)_c$ vanishes at $E = E_c^{(0)} = 4.25$ V/cm, corresponding to $X = X_c^{(0)}$. The inverse of $(\sigma F)_c$ has been proven to extrapolate to zero at $E_1 = 0.25$ V/cm, corresponding to $X = X_c^{(0)}/2$. By application of Eq. (3) the impact ionization coefficient X in units of $X_c^{(0)}$ as a function of E has been evaluated. The result, shown also in Fig. 3, may be fitted, with excellent agreement, by the simple relation $X = X_0 \exp(-E_0/E)$ derived from Shockley's "lucky electron" model,[15] with $E_0 = 0.18$ V/cm and $X_0/X_c^{(0)} = X_0 P_A / T_1^s N_A = 2.0$. The inverse of $T_1^s N_A$ is just the lifetime τ_0 of the electrons in the

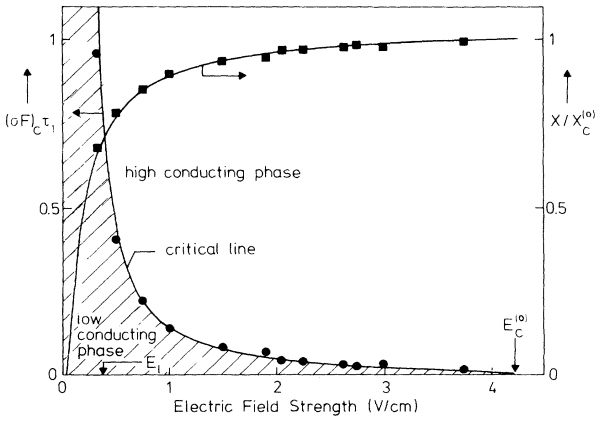

FIG. 3. The critical line of the optical transition probability $(\sigma F)_c$ in units of τ_1^{-1} and the resulting impact ionization probability per electron X in units of $X_c^{(0)}$ as functions of the electric field strength for 4.2 K. The circles and squares are derived from the measurements. The solid lines are calculated with the Shockley formula.

$N=0$ Landau level at zero electric field. This time constant has been estimated from the decay of the current through the sample after application of short electric pulses. For the present sample we found $\tau_0 \simeq 5$ ns, which yields $X_0 P_A \simeq 10^8\ s^{-1}$.

The lifetime of the electrons in the $N=1$ Landau level τ_1 can easily be derived from the saturation intensity for X close to $X_c^{(0)}$. In this case $\tau_{\text{eff}}=\tau_1(1+N_A/N_D)$, being free from the inherent uncertainties of X and T_1^s. We obtained in this case for the saturation intensity $I_s=\hbar\omega F_s=9$ mW/cm^2, which yields an electron lifetime $\tau_1=1.9$ ns, of the same magnitude as recently observed by the saturation of absorption at a higher magnetic field strength than in this work.[16]

The three-level model worked out here is a reasonable approximation as long as the population of the $N=2$ Landau level may be neglected compared to that of $N=1$. The splitting of radiative transitions between subsequent Landau levels due to nonparabolicity is too small to justify this assumption. However, the fact that we observe saturation and a good agreement between the calculated photoconductivity and the experimental results (Fig. 1) indicates that the above condition is satisfied, most probably because of an even shorter electron lifetime in the $N=2$ Landau level. In general, optical excitation of higher Landau levels increases the saturation intensity and lowers $X_c(F\rightarrow\infty)$ below $X_c^{(0)}/2$. In a more refined model, the effective impact ionization rate Xnn_D, which represents an average over different Landau bands, should be replaced by more detailed expressions, and more sophisticated expressions for the field dependence of the impact ionization coefficients[17] could be used. However, even such refined rate-equation models will not alter the basic features of the threshold behavior of the photoconductivity, since the behavior of equilibrium as well as nonequilibrium systems near critical points falls into only a few universality classes,[18] independent of the microscopic mechanisms. This is the reason why second-order phase transitions can be characterized by universal critical exponents, and a justification for the use of simple models. In fact, our rate equations (1) under steady-state conditions can be expanded near the critical line into the universal form[19]

$$n^2-\tau n+h=0, \tag{5}$$

where $h\propto X_1^s$ is analogous to an "ordering field," and $\tau\propto\sigma F-(\sigma F)_c$ is a measure for the distance from the critical line in the σF-E control-parameter plane, similar to the reduced temperature in equilibrium systems.

In conclusion, we have found a novel highly nonlinear threshold behavior of the cyclotron-resonance-induced FIR photoconductivity under impact-ionization conditions. We have interpreted this effect as a second-order nonequilibrium phase transition which corresponds to the crossing of a line of critical points in the control-parameter plane of the optical excitation probability σF and the electric field E. This novel example of critical phenomena far from equilibrium can be potentially important for applications in nonlinear quantum optics.

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