Nonlinear far-infrared photoacoustic magnetospectroscopy of *n*-GaAs at low temperatures

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The far-infrared magnetoabsorption of shallow donors in *n*-GaAs has been investigated as a function of irradiation intensity applying a low-temperature photoacoustic cell. Optical cross sections and saturation intensities of various optical transitions were quantitatively determined yielding recombination times of excited electrons.

Nonlinear dynamical properties of carrier transport in semiconductors like self-sustained oscillations and chaotic fluctuations are of strong current interest.[1] In order to understand dynamical phenomena in high-purity semiconductors on a microscopic basis, detailed knowledge of the recombination kinetics of charge carriers is needed. In particular the magnetic field dependence of kinetic parameters are of importance as the magnetic field plays a crucial role for the occurrence of chaos.[2,3]

A powerful method to study the recombination kinetics of carriers bound to shallow impurities in high-purity semiconductors represents saturation spectroscopy applying high-power far-infrared lasers.[4–6] In general saturation yields the product $\sigma\tau$ where σ is the cross section of the involved optical transition and τ the recombination time. To determine τ the cross section σ must be known. As many high-purity semiconductor materials are only available in form of thin epitaxial layers, σ can be measured with sufficient accuracy by transmission measurements only for strong transitions like $1s\rightarrow 2p_{\pm 1}$ or cyclotron resonance in *n*-GaAs. Weaker structures and the photoionization of shallow impurities are usually not accessible by simple transmission measurements.

In the present letter we report on measurements using a photoacoustic cell which works immersed in liquid helium in the center of a splitcoil superconducting magnet yielding substantially higher sensitivity than plain transmission measurements. The device was used to measure quantitatively the absorption cross section of shallow donors in *n*-GaAs for various magnetic field strengths and to evaluate the saturation of optical transitions to metastable states[7,8] in comparison to the $1s\rightarrow 2p_{\pm 1}$ transition. Extremely low saturation intensities and correspondingly long recombination times of the order of 100 ns were observed.

The measurements of the absorption coefficient were performed at $\lambda = 164$ μm wavelength in order to rely on previous results obtained for the same wavelength by the use of the UCSB free-electron-laser.[5] In order to have sufficient intensity to observe saturation and getting steady-state conditions during irradiation a CH_3OH molecular laser optically pumped by an electrically pulsed CO_2-laser was used yielding 200 μs long pulses. Taking into account all losses of the optical arrangement including reflection at the sample surface, the maximum intensity in the sample was 70 mW/cm^2. The radiation was linearly polarized thus the right and left circular polarized component inducing optical dipole transitions with selection rule $\Delta m = \pm 1$ were of the same strength.

The photoacoustic cell behaves like an extended Helmholtz resonator[9] with lowest resonance frequency at 450 Hz for 4.2 K. As the duration of a laser pulse is small compared to the period of that frequency the response of the cell is a damped oscillation with the frequency of the first resonance. At 4.2 K the resonance is very sharp and weakly damped. This restricts the maximum repetition rate of the laser pulses to about 10 Hz. For measurements we detected the first maximum of the oscillation with a conventional boxcar, averaging over 30 to 100 pulses. The spectra obtained show up on a background which depends on the FIR wavelength but not on the magnetic field. The background signal is due to the absorption of the windows and must be measured separately to calibrate the photoacoustic cell.

The sample used is an *n*-GaAs epitaxial layer of 59 μm thickness grown on semi-insulating GaAs. The effective donor concentration and electron mobility at 77 K were $P_A = N_D - N_A = 5.1\times 10^{13}$ and $\mu = 114\,399$ V cm^2, respectively. The compensation ratio is $N_A/N_D = 0.83$.

In Fig. 1 the photoacoustic signal is shown in comparison to the photothermal conductivity as a function of the magnetic field obtained at 4.2 K with 70 mW cm^{-2} intensity in the sample. The photoconductive signal was measured in the usual way by biasing the sample with a voltage well below impact ionization nonlinearities whereas the photoacoustic spectrum was measured without bias. In both spectra the dominant structures are the $1s\rightarrow 2p_{+1}$ shallow donor transition at $B = 2.05$ T. Below these resonances a quasicontinuous spectrum is observed with weak structures which are the same in both recordings. The most pronounced difference is that the background of the photoconductive signal strongly increases approaching zero magnetic field in contrast to the absorption coefficient. The structures in the quasicontinuum are readily identified as optical transitions from the $1s$ donor ground state to metastable states or Coulomb resonances.[7,8] Such states are

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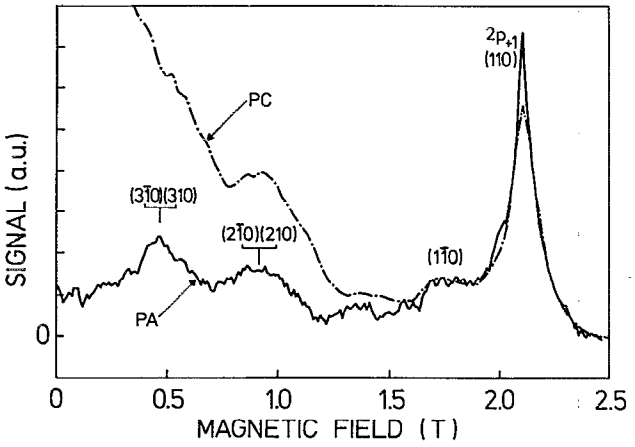

FIG. 1. The photoacoustic (PA) and, for comparison, the photoconductive (PC) magnetospectrum of n-GaAs at $\lambda=164\ \mu$m. Optical excitations out of the 1s donor ground state are labeled by the corresponding quantum numbers of the final states.

formed in atomlike systems subjected to a magnetic field below each (N,m)-Landau sublevel where N is the Landau level quantum number and m represents the quantum number of the angular momentum parallel to the magnetic field. Identified metastable states are indicated by the set (N,m,κ) where κ counts the knots of the wave function along the magnetic field.

The measurements demonstrate, that photoionization of shallow donors in a magnetic field at low temperatures is dominated by a two-step process like the photothermal conductivity caused by optical transitions to bound states. In the first step metastable states lying in the continuum spectrum of donors are optically populated and subsequently the exited donor is ionized. There are no indications for transitions into free states which should show a spectral structure resembling the density of states of Landau levels.[10]

Thus the quasicontinuum absorption and photoconductivity must be treated by at least a three level model in the same way as resonant lines due to excitations of bound states.[5,11] For intensities I much smaller that the saturation intensity I_s of transitions into free continuum states and assuming homogeneous broadening, the photoacoustic signal and the linewidth may be approximated by $signal \propto I/(1+I_s)$ and $\Delta B \propto (1+I/I_s)^{1/2}$. $I_s=\hbar\omega/(2\sigma\tau_{\rm eff})$ is the saturation intensity, and $\tau_{\rm eff}$ is the average time an electron needs to return to the donor ground state.[11] The measurements showed that the saturation intensity varies strongly with the magnetic field. Therefore, for the pronounced structures in Fig. 1, the photoacoustic signal was measured as a function of intensity and the nonsaturated cross section was determined by extrapolation to zero intensity. The response of the cell was calibrated by the absorption cross section of the $1s\rightarrow 2p_{+1}$ transition which was determined by transmission. To evaluate the measurements one must keep in mind that for $B\neq 0$ resonances only one half of the intensity is effective due to the $\Delta m=\pm 1$ selection rule whereas for the direct photoionization at $B=0$ the total intensity must be taken into account. The results are summarized in Table I. Because the transitions to (210) and (2$\bar{1}$0) metastable states are not well resolved the cross sections are given for the center of the corresponding spectral structures. The $1s\rightarrow 2p_{+1}$ absorption cross section agrees reasonably well to previous investigations.[12] The photoionization cross section at $B=0$ can easily be calculated by standard quantum theoretical methods[13] yielding 3×10^{-14} cm^2 in good agreement to the present experimental result. Two examples of saturation are shown in Figs. 2 and 3. The inverse of the observed effective optical cross section, $\sigma_{\rm eff}$, is plotted as a function of the intensity. In both cases a straight line, $\sigma_{\rm eff}^{-1}=\sigma^{-1}(1+I/I_s)$, is observed whose slope gives the saturation intensity. The plot of the $1s\rightarrow 2p_{+1}$ resonance also contains the square of the line width $(\Delta B)^2$ as a function of intensity. The ordinate scales were chosen in such a way that the averages of both graphs coincide. This demonstrates that both the absorption coefficient at the resonance center and the line width consistently have a common saturation intensity as expected for a homogeneously broadened line. This is usually not observed by photoconductivity due to excited state impact ionization of shallow donors caused by the unavoidable bias voltage.[11] For low intensities $(\Delta B)^2$ deviates from the straight line assuming somewhat larger values than given by the relation $\Delta B \propto (1+I/I_s)^{1/2}$. In fact it is well known from low power measurements that shallow donor transitions in GaAs are inhomogeneously broadened.[14,15] Our

TABLE I. Resonance field $B_{\rm res}$, unsaturated absorption cross section σ, saturation intensities I_s and effective recombination times $\tau_{\rm eff}$. (*) I_s of photoionization at $B=0$ T was determined by the UCSB-free-electron laser (**) $I_s>70$ mW cm^{-2}, larger than available intensity.

Transition	$B_{\rm res}$[T]	σ[cm^2]	I_s[mW/cm^2]	$\tau_{\rm eff}$[ns]
$1s\rightarrow 2p_{\pm 1}$	2.05	$1.4\pm 0.4\times 10^{-12}$	9.8 ± 2	46 ± 18
$1s\rightarrow$(1$\bar{1}$0)	1.79	$8.3\pm 3\times 10^{-13}$	3.4 ± 0.5	215 ± 92
$1s\rightarrow$(210) $1s\rightarrow$(2$\bar{1}$0)	0.90	$1.8\pm 0.3\times 10^{-13}$	26 ± 11	127 ± 70
$1s\rightarrow$(310) $1s\rightarrow$(3$\bar{1}$0)	0.47	$1.1\pm 0.3\times 10^{-13}$	(**)	(**)
$1s\rightarrow$continuum	0	$3.5\pm 1\times 10^{-14}$	6900 ± 500 (*)	5 ± 2

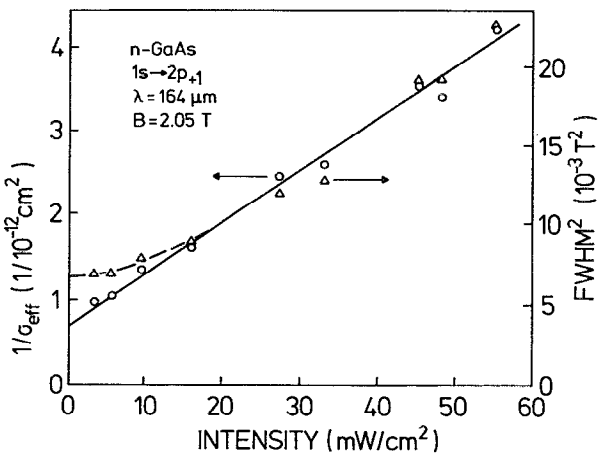

FIG. 2. Inverse of the cross section $\sigma_{\rm eff}$ and the line width of $1s\rightarrow 2p_{+1}$ as function of intensity.

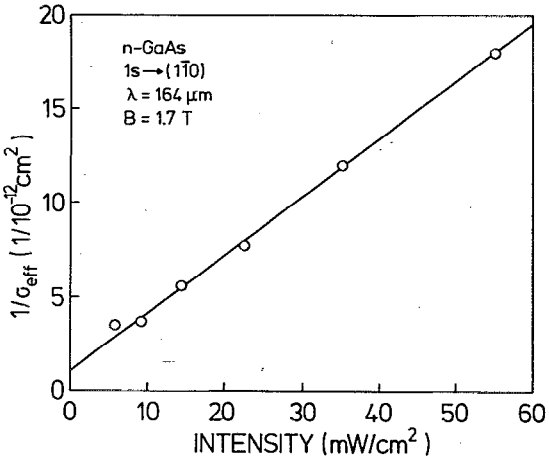

FIG. 3. Inverse of the cross section σ_{eff} for $1s\rightarrow(1\bar{1}0)$ as function of intensity.

measurements show that the homogeneous and the inhomogeneous line width are of the same order of magnitude and power broadening surpasses the inhomogeneous width at about 20 mW cm^{-2}. Extrapolating the high power values of $(\Delta B)^2$ to zero intensity we obtain the nonsaturated homogeneous line width $\Delta B=60$ mT corresponding to 0.8 cm^{-1}.

The resulting saturation intensities and recombination times are given in Table I for all investigated transitions. The signals at $B=0$ and for $1s\rightarrow(310)/(3\bar{1}0)$ were linear in the range of available laser power therefore no saturation intensity could be determined. The recombination times from the metastable states are surprisingly large, however, they are consistent with previous investigations of the nonlinear photoconductivity caused by $1s\rightarrow 2p_{+1}$ transitions at higher power levels. Electrons in an excited donor state may recombine either by remaining stuck on the donor (time constant τ_2) or by being transferred to the conduction band and subsequently captured (τ_1) . This leads to $\tau_{\text{eff}}=(1/2)p^*\tau_1+(1-p^*)\tau_2$ where p^* is the ionization probability of the excited state. In the case of the $1s\rightarrow 2p_{+1}$ at $\lambda=164$ μm $\tau_1=144$ ns with $p^*=0.18$ has been found.[5] In the case of metastable states p^* may be close to one, thus τ_{eff} approaches τ_1.

The saturation of photoconductivity was previously investigated at higher power levels in the same magnetic field range and wavelength using the UCSB free electron laser.[16] Saturation intensities in the range of 1 W cm^{-2} were observed to be much larger than those of the present investigation. This may be understood by competing optical transitions to true free states at the same energy like that of metastable resonances. Calculations have shown that the cross sections $1s\rightarrow$*free states* are about two orders of magnitude smaller than those experimentally obtained here for metastable states.[10] Thus at high intensities the transitions to metastable states are saturated and the photoconductive signal is governed by transitions to free states. Due to the small optical cross section large saturation intensities occur. At $B=0$ where no metastable states exist, only transitions to continuum states are possible. The saturation intensity obtained with the free-electron laser and the corresponding recombination time is also given in Table I.

In summary, far-infrared photoacoustic spectroscopy at low temperatures proved to be a reliable method to measure weak absorption structures in thin semiconductor layers. High purity *n*-GaAs has been investigated in a magnetic field by a molecular laser yielding absorption cross sections of transitions to metastable states for the first time. From the intensity dependence of the absorption saturation intensities and corresponding electron recombination times were determined. Financial support by the Deutsche Forschungsgemeinschaft is gratefully acknowledged. Work done at UCSB was supported in part by DOE grant No. ER45089, ONR-URI grant No. N000014-86-0110, and SDIO-ONR grant No. N000014-87-G-0026.

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