rate of spin-polarized hydrogen points the way to obtaining higher densities. By compressing a sample of spin-polarized hydrogen in the pure state it may be possible to observe the Bose-Einstein condensation. For example, at 300 mK the transition should occur at a density of $8\times10^{19}$ $cm^{-3}$. Our results suggest that the lifetime of spin-polarized hydrogen under these conditions is about three seconds, long enough for useful experiments.

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# Far-Infrared Two-Photon Transitions in *n*-GaAs

W. Böhm, E. Ettlinger, and W. Prettl
*Institut für Angewandte Physik, Universität Regensburg, D-8400 Regensburg, West Germany*
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Two-photon transitions in the far-infrared spectral range have been observed for the first time. The magnetophotoconductivity of $n$-GaAs measured by a pulsed high-power $CH_3F$ molecular laser at $\lambda = 496$ $\mu$m exhibited two-photon transitions between 1$s$ and 2$s$ shallow donor states and two-photon cyclotron resonance. The experimental results and symmetry considerations indicate that the observed two-photon cyclotron resonance is caused by impurities.

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Low-energy electronic transitions in semiconductors can be detected by far-infrared photoconductivity with high sensitivity. Previous experiments have been performed by low-power far-infrared sources involving one-photon cyclotron resonance and one-photon transitions between impurity levels. In this paper we report on the first observation of two-photon transitions in the far-infrared spectral range. The experiments were carried out on $n$-GaAs at liquid-helium temperature employing a pulsed high-power $CH_3F$ molecular laser at the wave number $\tilde{\nu} = 20.2$ $cm^{-1}$ ($\lambda = 496$ $\mu$m). Shallow donor states in $n$-GaAs to a high accuracy obey the simple hydrogenic effective-mass theory with an effective Rydberg constant $R^*$ of 46.1 $cm^{-1}$.[1-3] The wave number 20.2 $cm^{-1}$ corresponds to a photon energy of 2.5 meV $= 0.44R^*$ which is too small to cause one-photon transitions between the 1$s$ donor ground state and excited states. The energy levels of shallow donors and the cyclotron frequency in $n$-GaAs can be tuned by moderate magnetic fields in such a way that the energy of two photons $2\hbar\omega$ fits the $1s$-$2s$ energy-level separation or the energy difference between adjacent Landau levels.[3] We observed photoconductivity resonances at magnetic field strengths just satisfying these resonance conditions, which provides direct evidence for two-photon absorption between $1s$ and $2s$ shallow donor levels and for two-photon cyclotron resonance.

The employed samples were high-purity epitaxial layers with (100) crystallographic orientation of the coated faces. The samples were mounted in a metallic light pipe at the center of a superconducting solenoid and immersed in liquid helium. The photoconductivity was measured in Faraday configuration with the [100] crystallographic direction of the samples parallel to the magnetic field. The $CH_3F$ laser was optically pumped by a pulsed transversely excited atmosphere $CO_2$ laser producing far-infrared pulses of the order of 1 kW peak power. The laser power was varied by attenuators down to $\frac{1}{10}$ of the peak power. Lower intensities could not be detected because of electrical interference in the detection electronics from the electric discharge of the transversely excited atmosphere laser. In addition to this the photoconductivity was also measured using a continuously working $CH_3F$ laser emitting approximately 1 mW power and being pumped by a conventional cw $CO_2$ laser.

In Fig. 1 the magnetic field dependence of the photoconductivity measured by laser pulses of the order of 100 W peak power is compared with a recording obtained by using low-power radiation of about 1 mW. Resonant structures arise on a continuous photoconductivity background. In both spectra one-photon cyclotron resonance occurs at the magnetic field strength $B_{CR} = 1.43$ T, as expected from the effective electron mass $m^*/m = 0.067$ and the applied laser frequency $\omega$. In the high-power spectrum two additional resonances are observed. The line at $B = 1.15$ T below the cyclotron resonance agrees well with the magnetic field strength anticipated for $1s$-$2s$ two-photon transitions of shallow donors.[3] The other peak appears just at $B = 2B_{CR}$ indicating two-photon cyclotron resonance. The structures of the photoconductivity resonances show a remarkable difference concerning their line shapes. The line due to $1s$-$2s$ transitions and the one-photon cyclotron resonance line are symmetric as is expected for resonant transitions between equidistant energy levels, whereas the photoconductivity peak at $2B_{CR}$ is evidently asymmetric. In the latter case the photosignal grows smoothly with increasing magnetic field and drops steeply as soon as the cyclotron frequency $\omega_{CR}$ exceeds twice the laser frequency above $2B_{CR}$.

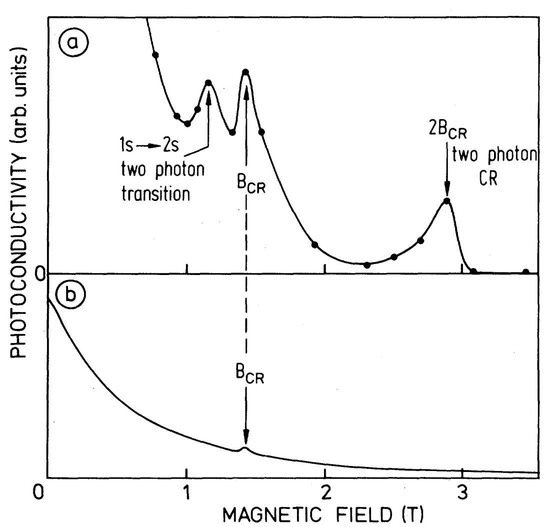


FIG. 1. Comparison of the photoconductive response as a function of the magnetic field obtained (a) by a pulsed laser of about 140-W peak power and (b) by a 1-mW cw laser. $B_{CR}$ is the resonant magnetic field of cyclotron resonance.

The evidence for the two-photon transition interpretation of the observed photoconductivity resonances comes from the good agreement between theoretical and experimental two-photon transition energies and the absence of the structures at low laser power. Usually, two-photon transition intensities are proportional to the corresponding transition rates and thus vary like the square of the optical power. However, extrinsic photoconductivity is expected to be proportional to the transition rate at low power levels only, whereas for increasing power, saturation effects become important. It seems to be reasonable to assume that the $1s$-$2s$ two-photon transition is observed in photoconductivity because of the photothermal process.[4,5] In $n$-GaAs cyclotron resonance absorption yields a photoconductive signal due to an increase of the electron density by thermal repopulation of optically depleted Landau levels from bound donor states.[6] Therefore all three resonant structures observed in our experiments result from changes in free-electron concentration $\Delta n$ and thus saturate as $\Delta n \propto N_D - N_A$, if at high power levels the number of neutral donors is exhausted. In Fig. 2 the photoresponse is plotted for various laser powers up to approximately 1 kW peak power. These measurements

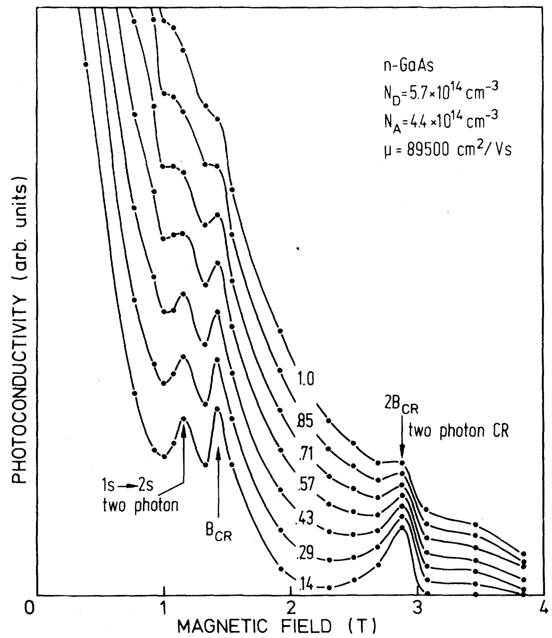


FIG. 2. Photoconductivity at various laser powers. Numbers identifying the curves denote the peak power of the laser pulses, where 1 corresponds to about 1 kW.

clearly show that in the power range accessible in our experiments saturation occurs and thus the two-photon transition intensities must not be proportional to the square of the optical power.

The selection rules of two-photon transitions require intermediate states which must be connected by electric dipole matrix elements with the initial and final states of the transition. For the $1s$-$2s$ shallow donor transition all $np_\pm$ donor states may act as intermediate states. Contrary to this clear situation, the underlying physical mechanism of two-photon cyclotron resonance is not evident, as this optical process is forbidden for free electrons in the conduction band of GaAs if our experimental conditions—Faraday configuration with $B$ parallel to [100]—are taken into account. This selection rule is valid for the continuous symmetry of electrons in a constant potential as well as for the correct crystallographic symmetry of GaAs.[7] The only reasonable physical mechanism causing the relaxation of this selection rule is due to impurities. This can be shown by group-theoretical considerations which are applicable independent of the particular model describing the electron states. The space group of a free conduction electron in a homogeneous magnetic field is given by $C_{\infty h}\otimes T$, where $T$ denotes the invariant subgroup of translations. The presence of impurities destroys the translational invariance and lowers the symmetry to the point group $C_{\infty h}$. This means that the conservation of the wave vector $k_\parallel$ parallel to the magnetic field $B$ and the independence of the energy with respect to translations of the cyclotron orbit perpendicular to $B$ are relaxed. As a result the full space-group irreducible representations $D_m{}^{*k}$ of $C_{\infty h}\otimes T$ are reduced according to $D_m{}^{*k}\to\Gamma_m{}^+\oplus\Gamma_m{}^-$, where $\Gamma_m{}^\pi$ are irreducible representations of the point group $C_{\infty h}$. Here $*k$ denotes the star of the wave vector, being a circle around the direction of the magnetic field with $k_\parallel$ = const; $m$ is the angular momentum quantum number, and $\pi=\pm$ represents the parity. Thus each Landau subband condenses into a continuum of energy levels which contains states of both parity $\pi=\pm 1$ and angular momentum $m\leq n$, where $n$ is Landau energy quantum number. The density of continuum states equals the density of states of the Landau levels for free electrons in a translational invariant potential. We must keep in mind, however, that the selection rules in the impurity case are quite different. Electric dipole transitions are governed by the less restrictive parity and angular momentum selection rules only. Essentially, electric dipole transitions are now possible between energy levels of any spacing. Zawadzki came to similar conclusions by calculating one-photon magneto-optical resonances in the presence of impurities by a perturbation treatment. He explained by this approach cyclotron resonance harmonics observed in InSb.[8] If donors as impurities are taken into account, discrete bound states appear below and within the energy continuum. This situation is schematically displayed in Fig. 3, which corresponds to the present experimental conditions in $n$-GaAs. It is now obvious that there are many ways for subsequent virtual electric dipole transitions yielding impurity-induced two-photon cyclotron resonance. Intermediate states may be bound impurity states of both parities and continuum states as indicated in Fig. 3. When we consider the zinc-blende structure of GaAs, a completely analogous result is found. The space group $S_4\otimes T$ (Ref. 7) is reduced by impurities to $C_2$ and the energy continuum deduced from each Landau subband contains states of both representations $\Gamma_1$ and $\Gamma_2$ of $C_2$. Dipole matrix elements exist[9] between $\Gamma_1$ and $\Gamma_2$ and thus all that was stated above about continuous symmetry is also valid for this case.

The usual cyclotron resonance of electrons re-

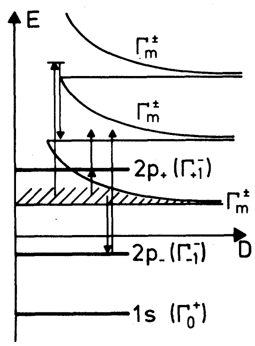


FIG. 3. Schematic energy-level diagram of bound and free donor states of point symmetry $C_{\infty h}$. Possible initial, intermediate, and final states for impurity-induced two-photon cyclotron resonance are indicated by arrows.

sults from transitions between energy levels equidistantly spaced by $\Delta E = \hbar\omega_{CR}$ and therefore yields a line of Lorentzian shape whose width is determined by the electron momentum relaxation. In impurity-induced two-photon cyclotron resonance the limitation of equal spacing of the initial and final states is lost. Thus at low temperatures and quantizing magnetic fields the line shape is determined by the density of states of the energy continuum deduced from the $n=1$ Landau subband and the magnetic field dependence of the relevant matrix elements. At fixed laser frequency two-photon transitions are already possible at magnetic fields $B<2B_{CR}$ and the transition probability must drop down sharply above $2B_{CR}$ so long as only a narrow energy range at the bottom of the lowest Landau continuum is occupied by electrons. Our measurements just show a lineshape being asymmetric in this sense for the resonance at $2B_{CR}$ as pointed out previously. This result strongly supports our interpretation of this resonance as due to impurity-induced two-photon cyclotron transitions.

In conclusion we have carried out the first two-photon absorption measurements in the far-infrared spectral range in a semiconductor. Our measurements demonstrate that the method of two-photon spectroscopy can be extended to the long-wavelength infrared with presently available lasers. Two-photon spectroscopy may be particularly useful to study excited impurity states for which electric dipole transitions from the ground state are parity forbidden.

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