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Spin-dependent terahertz nonlinearities at inter-valence-band absorption in p-Ge

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Abstract

Far-infrared absorption and photon drag effect have been investigated in p-type germanium as a function of intensity of a high-power pulsed far-infrared molecular laser. Optical transmission shows saturation independent of the state of polarization of the radiation whereas in the photon drag effect a pronounced linear–circular dichroism occurs. Linear–circular dichroism is caused by spin-selective excitations as a result of selection rules and, equally important, multi-photon transitions with a rate comparable to one-photon absorption. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Far-infrared; Multi-photon absorption; Saturation; Spin

1. Introduction

Spin polarization and spin relaxation are the subject of intensive studies with respect to the development of spin devices [1]. So far, spin polarization has been achieved by circular polarized optical inter-band excitations leading to spin-polarized electrons and holes [2,3]. Inter-subband transitions have been proposed as a means of monopolar spin orientation [4] but not yet experimentally realized. Here we report on spin-dependent multi-photon absorption and saturation of one-photon absorption in p-Ge. Optical transmission and, as a very sensitive method to determine absorption, longitudinal photon drag effect have been investigated applying high-power terahertz radiation. We demonstrate that inter-valence-subband absorption of intense circular polarized radiation causes spin-dependent nonlinearities and might become a method to detect monopolar spin orientation.

2. Experimental methods and results

The measurements have been carried out in the temperature range between 40 and 300 K on p-Ge bulk samples with carrier densities from 10^{13} to 10^{16} cm^{-3}. The radiation source used was a TEA-CO_2-laser pumped NH_3-laser operating in the wavelength range from 28 to 280 μm with pulse duration $\tau \approx 40$ ns and intensity up to 2 MW/cm^2.

In Fig. 1 the photon drag current for $\lambda = 90$ μm is shown as a function of the intensity for circular and linear polarized light. For both polarizations, the photon drag current changes sign with increasing intensity. At room temperature, the intensity at which inversion occurs is five times smaller for

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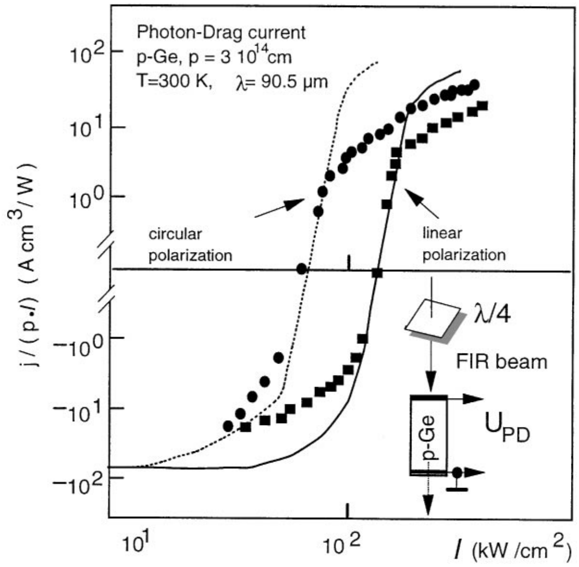

Fig. 1. Intensity dependence of the photon drag current j normalized by the hole concentration p and the intensity I for linear and circular polarized radiation. Solid lines show calculations taking into account one- and two-photon transitions. Inset shows the experimental setup.

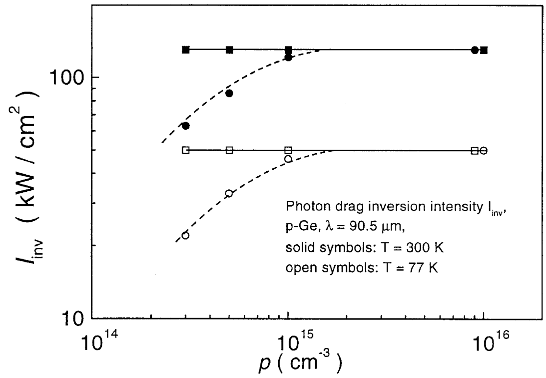

Fig. 2. Photon drag current inversion intensity for circular (circles) and linear (squares) polarized radiation.

circular than for linear polarized light and is independent of carrier density (Figs. 1 and 2).The same behavior has been observed for $\lambda = 76\,\mu m$. However, at longer wavelengths between 148 and 280 µm no sign inversion could be detected. At low temperature the inversion of sign is extended to longer wavelength and does occur also at $\lambda = 148\,\mu m$ but not at 280 µm. In contrast to room temperature the inversion intensity depends on the hole density at low temperatures. For 77 K and $\lambda = 90.5\,\mu m$ this is shown in Fig. 2.

In contrast to the photon drag current, the transmission shows no polarization dependence. At room temperature no intensity dependence of transmission has been observed. At low temperatures and for wavelengths shorter than about 100 µm saturation of transmission was observed. The saturation intensity depends on free carrier concentration, temperature and wavelength.

3. Discussion

The linear circular dichroism detected in the photon drag effect arises at high intensities and only when it is caused by direct inter-subband transitions which is the case for shorter wavelengths. Direct inter-subband transitions by circular polarized light de-populate and populate selectively spin states at certain energies in the valence band due to selection rules and energy and momentum conservation (see Fig. 3). In contrast, optical transitions induced by linear polarized light are not spin selective. High intensities lead to nonlinear absorption due to saturation [5,6] of one-photon and/or multi-photon absorption [7]. Both may show linear circular dichroism.

Saturation of one-photon absorption of circular polarized light may cause mono-polar spin orientation in the ground state spin doublet. This situation occurs if the saturation of absorption is either caused by Rabi-oscillations [8] or by slow relaxation of the photo-excited carriers when the spin relaxation time τ_s in the ground state is larger than the energy relaxation time τ_0 of photoexcited carriers (see Fig. 3). Saturation of the one-photon absorption caused by slow relaxation is given by $K^{(1)}(I) = K^{(1)}(0)/(1 + I/I_s)$ where $K^{(1)}(I)$ and I_s are the absorption coefficient as a function of the radiation intensity I in the crystal and the saturation intensity, respectively. The saturation intensity $I_s = 1/(\sigma\tau)$ is determined by the absorptions cross section σ per hole and a time constant τ which corresponds to the average time a photo-excited carrier needs to return to the initial state. For circular polarized radiation the relaxation time can be

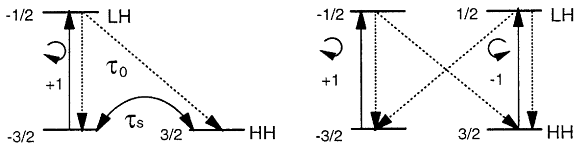

Fig. 3. Schematic representation of one-photon inter-valence-subband transitions and relaxation (dashed lines) at excitation by circular (left) and linear (right) polarized radiation.

equal to either the spin relaxation time τ_s or to the energy relaxation time τ_0 depending on the relative magnitude of the two time constants. For linear polarized radiation the return time τ is equal to the energy relaxation time τ_0 because spin up and spin down states are always equally populated. Therefore, saturation can show linear circular dichroism if $\tau_s \gg \tau_0$ and thus allows to conclude on spin relaxation time.

The fact that transmission which is controlled by one-photon excitation is independent of the polarization rules out saturation as the reason of linear circular dichroism in the photon drag and shows that the spin relaxation time is substantially smaller than the energy relaxation time of photo-excited carriers. Note that the absence of saturation of transmission at room temperature is due to the fact that in this case absorption is dominated by indirect *intra*-valence-band transitions (Drude absorption), whereas photon drag effect is due to *inter*-valence-band transitions.

Another spin-selective excitation process which must be taken into account is multi-photon absorption which causes the linear–circular dichroism of the photon drag observed here. Multi-photon absorption is of particular importance in the far-infrared due to the dependence of the ratio of n-photon to $(n-1)$-photon absorption, $\eta_n = K^{(n)}/K^{(n-1)} \propto I\omega^{-3}$ on radiation frequency ω and intensity I. In Ref. [7] it was shown that simultaneous n-photon transitions can occur in p-Ge with comparable contributions to the total absorption at intensities of about 1 $\mathrm{MW/cm^2}$ and $\lambda = 100\,\mu\mathrm{m}$. The photon drag current is proportional to the number of absorbed photons, KI, the photon momentum, the effective mass of carriers, and the momentum relaxation time. As a result of the interplay of these parameters, two- and three-photon drag currents have opposite direction to the one-photon contribution at wavelengths in the range 76–148 μm. The comparable input of two- and three-photon processes and their superlinear intensity dependence lead to a change of sign of the total photon drag current at high intensities.

At longer wavelengths, where no sign inversion has been observed, the photon drag current is caused by indirect intra-band transitions overriding the effect of direct one- and multi-photon absorption. The solid lines in Fig. 1 show the result of calculations after [8] taking into account one- and two-photon transitions. The calculations have been carried out for all wavelengths used here yielding a good agreement to measurements. The shift of the inversion intensity to smaller values at low temperatures and low carrier densities (Fig. 2) is caused by polarization-independent saturation of one-photon absorption.

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