Resonant circular photogalvanic effect in GaN/AlGaN heterojunctions

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The resonant circular photogalvanic effect is observed in wurtzite (0001)-oriented GaN low-dimensional structures excited by infrared radiation. The current is induced by angular momentum transfer of photons to the photoexcited electrons at resonant inter-subband optical transitions in a GaN/AlGaN heterojunction. The signal reverses upon the reversal of the radiation helicity or, at fixed helicity, when the propagation direction of the photons is reversed. Making use of the tunability of the free-electron laser FELIX we demonstrate that the current direction changes by sweeping the photon energy through the intersubband resonance condition, in agreement with theoretical considerations.

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I. INTRODUCTION

Wide bandgap GaN has been extensively investigated for applications as blue and ultraviolet light sources as well as for high temperature and high power electronic devices. The commercial fabrication of blue and green LEDs has led to well established technological procedures of epitaxial GaN preparation and sparked a great research activity on the properties of heterostructures based on GaN and its alloys with AlN and InN. Most recently two-dimensional GaN attracted growing attention as a potentially interesting material system for semiconductor spintronics since, doped with manganese, it is expected to become ferromagnetic with a Curie temperature above room temperature. Gadolinium doped GaN is mostly caused by the spin-dependent mechanism. The thickness of the AlGaN layers was varied between 30 nm and 100 nm. An undoped 33 nm thick GaN buffer layer grown under a pressure of 40 Pa at temperature 550°C is followed by an undoped GaN layer (∼2.5 μm) grown under 40 Pa at 1025°C; the undoped Al0.3Ga0.7N barrier was grown under 6.7 Pa at 1035°C. The mobility and density in the two-dimensional electron gas measured at room temperature are μ = 1200 cm²/Vs and Nc ≈ 10¹³ cm⁻², respectively. To measure the photocurrent two pairs of contacts are centered at opposite sample edges with the connecting lines along the axes x || [1120] and y || [1100], see inset in the top of Fig. 1. In order to excite resonantly transitions between the size quantized subbands ε1 and ε2 and to obtain a measurable photocurrent it was necessary to have a tunable high power radiation source for which we used the free electron laser “FELIX” at FOM-Rijnhuizen in the Netherlands oper-
FIG. 1: Spectral dependence of the transversal photocurrent $J_y$ measured at room temperature at oblique incidence ($\theta_0 = 15^\circ$) for right-handed circularly polarized radiation. The data are obtained by the free electron laser FELIX (dots) and TEA CO$_2$ laser (square). The radiation power used for these measurements was about $35 \, \text{kW}$ for the CO$_2$ laser and about $100 \, \text{kW}$ for FELIX. The magnitude of the current measured at FELIX is fit at this plot to that obtained with the CO$_2$ laser, assuming that the current depends linearly on the radiation power at used power level. The solid line represents a fit by the sum of asymmetrical and symmetrical contributions to Eq. (2). The inset in the bottom corner shows the temporal structure of the current in response to the radiation of FELIX. The inset in the upper corner demonstrates the experimental geometry.

The output pulses of light from FELIX were chosen to be about 3 ps long, separated by 40 ns, in a train (or “macropulse”) of duration of 7 $\mu$s. The macropulses had a repetition rate of 5 Hz. The linearly polarized light from FELIX was converted into left handed ($\sigma^-$) and right handed ($\sigma^+$) circularly polarized radiation by means of a Fresnel rhomb. A rotation of the optical axis of the Fresnel rhomb’s plate by an angle $\varphi$ with respect to the laser radiation polarization plane results in a variation of the radiation helicity as $P_{\text{circ}} = \sin 2\varphi$. Here the angle $\varphi = 0$ corresponds to the setting where the position of the polarizer optical axis coincides with the incoming laser polarization. Radiation is applied at oblique incidence described by an angle of incidence $\theta_0$ varying from $-15^\circ$ to $+15^\circ$, see the top inset to Fig. 1.

III. EXPERIMENTAL RESULTS AND PHENOMENOLOGICAL DESCRIPTION

On illumination of the low-dimensional structures by circularly polarized radiation at room temperature at oblique incidence in $(xz)$-plane we observe a current signal in the $y$ direction perpendicular to the plane of incidence, Fig. 1. The current reverses its direction by switching the sign of the radiation helicity. Signals in response to the left handed and right handed polarized radiation are shown in the lower inset of Fig. 1. Using short 3 ps pulses of FELIX we observed that the response time was determined by the time resolution of our set-up which therefore sets an upper limit to the response time. This fast response is typical for photogalvanics, where the signal decay time is expected to be of the order of the momentum relaxation time being in our samples at room temperature typically about 0.1 ps. Figure 2 demonstrates the dependences of the photocurrent on the angle $\varphi$ for two angles of incidence $\theta_0 = \pm 15^\circ$. The photocurrent signals generated in the unbiased devices were measured via an amplifier with a response time of the order of 1 $\mu$s, i.e. averaged over the macropulse. The current closely follows the radiation helicity $P_{\text{circ}} = \sin 2\varphi$. The signal proportional to the helicity is only observed under oblique incidence. The current vanishes for normal incidence and changes its polarity when the incidence angle changes its sign, see Fig. 2. The photocurrent in the layer flows always perpendicularly to the direction of the incident light propagation and its magnitude does not change by rotating the sample around the growth axis.

Investigating the spectral dependence of the photocurrent we observe that for both, left and right handed circular polarizations, the current changes sign at a photon energy $\hbar \omega \approx 93 \, \text{meV}$, see Fig. 1. Figure 4 also shows that the spectral behaviour of the current can be well described by the sum of the derivative of the Lorentzian-like absorption spectrum and an additional contribution proportional to the absorption spectrum itself.

A phenomenological analysis for the $C_{3v}$ point group symmetry relevant to the structures under study shows that the $\varphi$-dependence of the transverse photogalvanic current density $j$ under excitation at oblique incidence...
in \((xz)\)-plane is given by \(12,13\):

\[
j_y(\varphi) = E_0^2 t_p t_s \sin \theta \left( \gamma \sin 2\varphi - \frac{x}{2} \sin 4\varphi \right). \quad (1)
\]

Here \(E_0\) is the amplitude of the electromagnetic wave, \(\theta\) is the refraction angle related to the incidence angle \(\theta_0\) by \(\sin \theta = \sin \theta_0/n_\omega\), where \(n_\omega\) is the refractive index, and \(t_s\) and \(t_p\) are the Fresnel amplitude transmission coefficients from vacuum to the structure for the \(s\) and \(p\)-polarized light, respectively. The coefficient \(\gamma\) is a component of the second-rank pseudotensor \(\chi\) describing the helicity dependent current, comprising the CPGE and the optically induced spin-galvanic effect, \(12,13,23\) and \(\chi\) is a component of the third-rank tensor \(\chi\) describing the linear photogalvanic effect. \(12,13,23\) In systems of \(C_{3v}\) symmetry the tensor \(\chi\) has one linearly-independent component, namely \(\chi_{xy} = -\chi_{yx} \equiv \gamma\). Thus, the CPGE current flows always perpendicularly to the plane of incidence. The components of the tensor \(\chi\) are given by \(\gamma \equiv \chi_{zxx} = \chi_{yyx},\) where \(z\) \(||\) \([0001]\) is the \(C_3\) axis. We take into account that the second linearly-independent component of \(\chi\) in \(C_{3v}\) systems is much smaller than \(\chi\), so that the corresponding photocurrent is negligible at oblique incidence. \(12,13\)

The fits of the experiment to Eq. (1) at \(\gamma/\chi = 1.4\) are shown in Fig. 2 for \(\theta_0 = \pm 15^\circ\) by solid lines demonstrating a good agreement with the experimental data. Figure 2 shows that the dominant contribution to the photocurrent is due to the helicity dependent current given by the first term on the right-hand side of the Eq. (1). The fact that the magnitude of the photocurrent does not change by rotating the sample around the growth axis is in agreement with the fact that in structures of \(C_{3v}\) symmetry the tensor \(\gamma\) has one linearly-independent component.

IV. MICROSCOPIC MODELS AND DISCUSSION

Two microscopic mechanisms are known to give rise to helicity-dependent currents exhibiting sign reversal for photon energies matching the energy separation between size-quantized subbands. The first is caused by an asymmetry of the momentum distribution of carriers excited by direct optical transitions in the system with the Rashba splitting of energy subbands. \(12,13\) Spectral inversion at resonance is a characteristic feature of this CPGE mechanism and has also been observed in GaAs QWs. \(20,26\) The second mechanism is of orbital nature and spin-independent. It is caused by the quantum mechanical interference of various pathways in Drude absorption. \(28\) This effect has most recently been demonstrated in Si-MOSFET inversion layers, where spin dependent mechanisms are absent due to vanishingly small spin-orbit interaction in silicon. \(29\) The spectral inversion in orbital mechanism is caused by a difference of the virtual states in the excited subband needed for the pathway with direct virtual optical transitions. Below we consider both mechanisms and compare their contributions to the total photocurrent.

The spin-dependent mechanism of the CPGE caused by direct inter-subband transitions at oblique incidence is illustrated in Fig. 3 for \(\sigma_+\) radiation. In \(C_{3v}\) symmetry the \(\alpha\sigma_xk_y\) term in the Rashba Hamiltonian splits the subbands in the \(k_y\) direction into two spin branches with the spin projection \(\pm 1/2\) oriented along \(x\). Due to selection rules, like in \((001)\)-grown GaAs QWs of \(C_{2v}\) symmetry, the absorption of circularly polarized radiation is spin-conserving. \(26\) It turns out, however, that under oblique excitation with circularly polarized light the rates of inter-subband transitions are different for electrons with the spin oriented parallel and antiparallel to the in-plane direction of light propagation. \(26\) This is depicted in Fig. 3 by vertical arrows of different thickness. In systems with \(k\)-linear spin splitting such processes lead to an asymmetrical distribution of carriers in \(k\)-space, i.e. to an electrical current. The inversion of photon helicity driven current is a direct consequence of \(k\)-linear terms in the band structure of subbands together with energy and momentum conservation as well as optical selection rules for direct optical transitions between size quantized subbands. At photon energy \(\hbar \omega_1 < \varepsilon_{21}\) of right circularly polarized radiation the most intense optical transition occurs at negative \(k_y\) resulting in a current \(j_y\) shown by an arrow in Fig. 3 (a). Here \(\varepsilon_{21}\) is the intersubband energy separation. Increase of the photon energy shifts the transition toward positive \(k_y\) and reverses the direction of the current, see Fig. 3 (b). In the frame of this model the sign reversal of the current takes place at the
photoexcited electrons in the band minima. The CPGE current at direct inter-subband transitions is given by

\[ j_{\text{spin}} = \Lambda (\alpha_1 - \alpha_2) \frac{e IP_{\text{circ}}}{\hbar} \times \left( (\tau_p^{(1)} - \tau_p^{(2)}) \frac{d\eta_21}{d\omega} + \tau_p^{(2)} \eta_21 (\omega) \right). \]

Here \( \alpha_{1,2} \) are the Rashba constants for electrons in the first and second subbands, the factor \( \Lambda \) describes monopolar spin orientation of carriers under resonant transitions, \( \tau_p^{(1)} \) and \( \tau_p^{(2)} \) are the momentum relaxation times in the initial and final state of optical transition, \( E \) is the average kinetic energy of carriers, \( \eta_21(\omega) \) is the intersubband absorbance which in the model of an infinitely-deep rectangular quantum well is given by

\[ \eta_21(\omega) = \frac{512 N e^2 \hbar}{27 \pi \epsilon_{21} - (\omega) \epsilon_{21}^2 + \Gamma^2}, \]

where \( m \) is the electron effective mass and \( \Gamma \) is the peak width. The 2DEG in GaN structures is almost degenerate even at room temperature due to the large Fermi energy: \( E \approx 100 \text{ meV} > k_B T \). Therefore, in contrast to GaAs based structures, the optical phonon emission by photoexcited electrons in the \( \epsilon_2 \) subband is suppressed (also due to the high frequency of the optical phonon in GaN). As a result, both initial and final states have momentum relaxation times of the same order, so that both symmetrical \( [\chi \eta_21(\omega)] \) and asymmetrical \( [\chi d\eta_21/d(\omega)] \) terms give comparable contributions to the CPGE. This is due to the fact that GaN based low dimensional structures have a large energy of optical phonons and typically a rather large Fermi energy compared to the GaAs QWs.

The orbital mechanism of the CPGE is caused by Drude-like absorption which is usually rather small at infrared frequencies used in the experiment. However, its contribution to the total photocurrent may be comparable with the spin-dependent mechanism and, therefore, should be analyzed. Following Refs. \[25,26\] the orbital mechanism of the CPGE is described by

\[ j_{\text{orb}} = \xi \frac{4 \pi e^3 N e}{\omega m \epsilon_{21}} \frac{\epsilon_{21}}{(\epsilon_{21} - \omega)^2} IP_{\text{circ}}, \]

where \( \xi \) is the factor of scattering asymmetry, and \( \kappa \gtrsim 1 \).\[25\] This equation shows that the CPGE due to the orbital mechanism also changes the photocurrent sign for photon energies matching the energy separation.

To compare spin-dependent and orbital contributions to the CPGE we take magnitudes of the photocurrent at the wings of the absorption contour (\( |\hbar \omega - \epsilon_{21}| = \Gamma \)). Equations \(2\) and \(3\) give an estimate for the ratio of the contributions as

\[ j_{\text{spin}} / j_{\text{orb}} \sim \frac{\Lambda (\alpha_1 - \alpha_2) \omega \tau_p^{(1)} E}{\xi \Gamma \epsilon_{21}}. \]

The factor \( \Lambda \) describing monopolar spin orientation of carriers under resonant transitions with account for both crystal (\( \Delta_{cr} \)) and spin-orbit (\( \Delta \)) splittings of the valence band provided \( \Delta \ll \Delta_{cr} \ll E_g \) (\( E_g \) is the fundamental energy gap) is given by

\[ \Lambda = \frac{\epsilon_{21} \Delta}{3E_g^2}. \]

It can be estimated for investigated GaN based structures as \( \Lambda \sim 0.03 \). This substantially smaller value of \( \Lambda \) in GaN structures compared to GaAs QWs is caused by a small spin-orbit interaction in the nitrogen atom, yielding \( \Delta/E_g \approx 10^{-3} \) for GaN. Taking \( \epsilon_{21} = 100 \text{ meV} \) and \( \Gamma = 6 \text{ meV} \) from the photocurrent spectrum, \( \epsilon_{21} \approx 10 \text{ Å} \), \( |\alpha_1 - \alpha_2| \sim \alpha_1 \approx 10^{-10} \text{ eV cm} \) and \( \xi \approx 10^{-2} \), as in Ref. \[26\], we obtain that the spin-dependent contribution to CPGE caused by resonant direct transitions is about five times larger than the CPGE due to the orbital mechanism caused by quantum interference in Drude absorption.

This conclusion is also supported by the shape of the CPGE spectrum. Indeed, Fig. \[1\] shows that the shape of the spectrum is substantially asymmetric. The orbital mechanism described by Eq. \(3\) yields only a slight asymmetry while for the spin-dependent mechanism in GaN structures we obtain that the photocurrent is a superposition of comparable symmetric \( [\chi \eta_21(\omega)] \) and asymmetrical \( [\chi d\eta_21/d(\omega)] \) parts, Eq. \(2\). Figure \[1\] shows that Eq. \(2\) describes well the whole spectral behaviour. We note that, when considering the spin-dependent contribution to the helicity dependent photocurrent, one should also take into account a possible admixture of the spin-galvanic effect which is proportional to the radiation absorbance and \( P_{\text{circ}} \). The interplay between CPGE and the spin-galvanic effect caused by resonant inter-subband transitions has been reported for GaAs QWs.\[27\]

To summarize, we demonstrate that in GaN low dimensional structures resonant intersubband transitions result in a circular photogalvanic effect with a dominant contribution by the spin dependent mechanism. The specific feature of the resonant CPGE in GaN heterojunctions is that the symmetric and asymmetric components of the photocurrent have comparable strengths.

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