Taking topological insulators for a spin

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Chapter 4

Investigation of helicity-dependent photocurrent generation in Bi$_2$Se$_3$

Optical manipulation of surface-state charge carriers in topological insulators can lead to new directions for electronic and spintronic applications. Therefore, the material’s response to polarized light requires a deeper understanding and can be monitored by the generated photocurrent, which indirectly yields information on the charge-carrier dynamics. In this chapter, generated photocurrents in Bi$_2$Se$_3$ are investigated by varying the helicity of incident infrared light which is expected to couple specifically to the surface states. Although such selectivity is currently not yet fully understood from literature, a helicity dependence is observed in our experiments. However, definitive conclusions on its origin are lacking due to technical difficulties.
4.1 Introduction

Optically accessing topological insulators has led to many interesting insights in this class of materials. The very first experiments, where (spin-resolved) ARPES has been used, have shown the existence of the theoretically proposed topological surface state located in the bulk band gap of Bi$_{1-x}$Sb$_x$ [1, 2]. Soon after, similar states were found for Bi$_2$Se$_3$ [3, 4], Bi$_2$Te$_3$ [5–7], and Sb$_2$Te$_3$ [7], triggering interesting new concepts in physics. Subsequently, ARPES on topological insulators has been extended in various ways. Two-photon photoemission spectroscopy (2PPES) enables probing of unoccupied bands where a second topological surface state has been observed that can enhance spin dynamics in topological insulators [8–11]. In addition, time-resolved 2PPES [or time-resolved ARPES (tr-ARPES)] experiments have shed light on the charge-carrier relaxation (scattering) dynamics relevant for photocurrent studies, using ultrafast optical pump and probe pulses [10, 12–29]. Besides tr-ARPES, by using circularly-polarized pump pulses, circular dichroism$^1$ in the angular distribution of ARPES has been claimed [30–32], but is under debate at the same time [33–36]. Another interesting feature is that the spin texture of surface states can be tuned by changing the polarization of the incoming light [33, 37–40], which can be useful for controlling the spin polarization of photoelectrons from topological insulators [37, 41] and is important for the analysis of ARPES spectra. Therefore, one could think of device schemes as those reported by Wunderlich et al. [42]. However, depending on the size of the device and required speed of the operation, strong spin ($\sim$100–500 fs) and momentum relaxation ($\sim$2 ps) might be the bottleneck for such applications [12, 24, 43]$^2$.

Besides 2PPES measurements, THz spectroscopy is a useful technique to unravel charge-carrier relaxation dynamics at time scales where scattering events as electron–phonon scattering occur [11, 44–53]. The origin of THz radiation is due to the transient charge current excited by a fs pulse that gives rise to an oscillating electric field emitting THz radiation. Therefore, THz spectroscopy serves as an indirect ultrafast photocurrent ammeter [51]. The low energy of THz radiation further allows to probe states close to the Fermi energy, which can be interesting when the Fermi level is tuned inside the bulk band gap and thus maximizes the sensitivity to surface state dynamics$^3$. Understanding these dynamics can in turn be interesting for high-performance THz photodetection applications [54, 55]. In order to increase the surface sensitivity, one can make use of the broken inversion symmetry at the surface ($C^{d}_{3v}$) relative to the bulk symmetry ($D^{d}_{5d}$) [56, 57]. Importantly, this symmetry consideration is different from experiments where symmetry arguments were used to describe surface-specific, second harmonic generation [12, 58, 59], which can be used to increase the surface state sensitivity in reflectivity measurements.

These temporal investigations give insights in the out-of-equilibrium dynamics of the charge carriers in topological insulators, which are useful for understanding and

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$^1$Circular dichroism is an effect that describes the difference in absorption for different circular polarizations of light. Such selectivity could lead to generation of a helicity-dependent photocurrent from the topological surface states.

$^2$The dynamics below 100 fs has been questioned by Boschini et al. [11].

$^3$However, bulk states will play a role since the pump pulse often has an energy above the band gap, exciting surface-state carriers to the bulk states. Reabsorption of the THz emission is an indirect measure for the surface state dynamics.
tuning photocurrents, i.e. currents that are driven by optical excitation. For spin-textured topological surface states, it has been proposed that photocurrents can be generated by using helical (i.e. circularly polarized) light, accessing different spin species leading to an asymmetric depopulation in \( k \) space \([60–62]\). The concept of the circular photogalvanic effect has earlier been reported for Rashba-textured, two-dimensional electron gases \([63,64]\) where it is emphasized that the observation of such effects depend on the symmetry of the system \([65]\). The control of photocurrent in Bi\(_2\)Se\(_3\) at room temperature has been shown for the first time by McIver et al. \([66]\), using excitation energies of \(\sim 1.7 \text{eV} \) that are way above the bulk band gap such that bulk effects cannot be ruled out. As became clear in later reports, the method presented in that work has to be treated with care since such photocurrents can have many different origins. It has been reported that asymmetric scattering \([25,52,56,57]\), photogalvanic effects from Rashba surface states due to band bending \([50,67]\), photon drag \([46,57,66]\), thermoelectric effects \([48,53,66,68,69]\), hexagonal warping effects \([69]\), and electrostatic potential fluctuations \([70]\) could also lead to photocurrents at the same time. Studies on the chemical potential dependence of these photocurrents have provided a better insight on the interplay between surface-state and bulk-state-related effects \([71,72]\).

Taking these considerations into account, we have studied photocurrent generation in Bi\(_2\)Se\(_3\) employing \(\sim 0.16 \text{eV} \) pump excitation with polarization modulation and by measuring the photocurrent through probing of the voltage created between Ohmic contacts. By studying the dependencies of the beam position, helicity, temperature, and fluence on the generated photovoltage, we have tried to exclude effects that mimic the expected behavior of the photovoltage on the circular polarization of the incoming light. Hampered by technical difficulties, careful conclusions on the results will be made.

### 4.2 Theory

In this section, basic concepts on the generation of photocurrents relevant for that in Bi\(_2\)Se\(_3\) are discussed. In the second part, a more detailed overview on the results from time-resolved experiments, as discussed briefly in the introduction, will be given. This section is aimed at giving a better understanding of effects that might affect or generate (additional) photocurrents.

#### 4.2.1 Generation of current through optical excitation

Although the exact mechanism for generation of photocurrents in topological insulators is still unresolved, an overview of the current work is given. Here, we consider topological insulators that have spin-momentum-locked surface states and spin-degenerate bulk bands that are illuminated by a light beam with a certain polarization and angle of incidence. It is important to realize that the photocurrent effects discussed here do not require an external bias to direct the photoexcited charge carriers.

\(^4\)However, the authors of the reported works focus on the surface states only and put the requirement of an out-of-plane spin orientation to generate a measurable effect.
but can be assisted by potential or thermal gradients nevertheless [68].

Generally, possible optical transitions are determined by electric–dipole selection rules based on the quantum numbers of the involved bands; a more detailed analysis can be found in for example [73]. The angular momentum \( j \) for electrons is built up from the orbital angular momentum \( l \) and spin angular momentum \( s \) with their \( z \) components specified by \( m_l \) and \( m_s \), respectively\(^5\). The orbital angular momentum quantum number \( l \) can be determined from the character of the orbital (s, p, d, f, g) and for electrons \( s = 1/2 \), giving \( m_s = \pm 1/2 \) (spin-up and spin-down). From this, the \( z \) component for the total angular momentum \( m_j \) can be found which takes integer steps from \(-j\) to \(j\) that in turn can have values from \(|l - s|\) to \((l + s)\). For any optical transition, one of the requirements is that \( \Delta l = \pm 1 \), related to a change in parity, whereas \( \Delta s = 0 \) and \( \Delta m_s = 0 \). One implication of this selection rule seems that the surface states of topological insulators, having a p-orbital character [74], have to couple with the s bands in the bulk. This means that optical transitions between the surface states and the first levels in the conduction band are not allowed since these both have a p-like character. However, higher-order phenomena might be present, which can lead to relaxation of the selection rules (i.e., spin–orbit coupling, mixing of states). Moreover, it has been proposed that due to the presence of a nonzero Berry curvature, spin flips (\( \Delta s \neq 0 \)) are allowed within the surface states [60, 62]. Most relevant for the optical excitations discussed here is \( \Delta m_j \) which depends on the polarization of the light. Circularly-polarized light carries angular momentum \( \pm \hbar \) (for \( \sigma^\pm \) polarization, depending on the relative orientations). In contrast, linear polarized light, which is a combination of \( \sigma^\pm \), carries zero angular momentum. During the light–dipole interaction, this additional angular momentum is transferred leading to \( \Delta m_j = \pm 1 \) for circular polarization, and \( \Delta m_j = 0, \pm 1 \) for linear polarization depending on the incident orientation.

Realizing spin-polarized photocurrents using different polarizations of light has been proven for quantum well structures where the resulting current depends on the exact symmetries of the system and the effective mass of the bands involved [65]. The photocurrent density \( J_\lambda \) due to such a circular photogalvanic effect can be phenomenologically described as:

\[
J_\lambda = \sum_{\mu} \gamma_{\lambda \mu} i (E \times E^*)_\mu, \quad (4.1a)
\]

\[
i (E \times E^*)_\mu = \hat{e}_\mu E_0^2 H_{\text{circ}}, \quad (4.1b)
\]

where \( \lambda, \mu = x, y, z \), \( E_0 \) the electric field amplitude and \( H \) denotes the helicity varying from \(-1\) to \(+1\). The elements of the second rank tensor \( \gamma \) are determined by the symmetry of the system. In the bulk of a topological insulator the tensor elements are all zero, but at the surface with broken inversion symmetry nonzero elements are obtained [72]. The angle of incidence can be related to the unit vector elements \( \hat{e}_\mu \) and allows one to distinguish the circular photogalvanic effect from the circular photon drag effect [72]. Furthermore, it has to be noted that, in the case for in-plane spin orientation in topological insulators, such transitions can generally only occur when

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\(^5\)Lower case letters indicate that electrons are treated [73].
the light is obliquely incident to the sample. However, this constraint is relaxed when deviations from this spin texture are present [25, 60, 61, 69].

The mechanisms behind optically accessing topological insulators with circularly polarized light with an excitation energy larger than the bulk band gap are still to be understood. The spin degeneracy of the bulk bands does not suggest any helicity-dependent effect similar to those observed in quantum wells a priori, which would mean that the spin-textured surface states have a key contribution in generation of the helicity-dependent photocurrent. In the picture sketched by McIver et al. [66], one of the branches of the Dirac cone would be selectively excited through circular dichroism as reported by the same group before [31], but that result is regarded controversial [33–36]. Instead, as recently reported by Pan et al. [72], it has been proposed that circularly polarized light is selective towards either transitions between bulk valence band and topological surface state or between topological surface state and bulk conduction band, provided that the excitation energy is high enough to overcome the band gap. This leads to an asymmetry in momentum in the surface states as well as the bulk states. In addition, tuning of the Fermi level further modulates the scattering between surface state and bulk conduction band leading to a change in the photocurrent, as suggested by Okada et al. [71]. A way to exclude such bulk-related effects is by using an excitation energy lower than the energy difference between the Fermi level and the conduction band minimum, provided that the Fermi level is below the charge neutrality point. However, such Berry-curvature-mediated photocurrents are theoretically predicted to be small unless the spin texture has a finite out-of-plane orientation [60, 61, 75]. Nevertheless, such photocurrents have been observed by Besbas et al. where the light was normally incident to the surface [69]. A similar result, but based on a different process, has been obtained by using mid-infrared pulses that are exciting carriers within the band gap in order to create a population asymmetry between +\(k\) and −\(k\) points [25, 26].

Other effects giving rise to photocurrents in topological insulators have been discussed in literature of which a brief overview will be given here. One candidate is the photon drag effect in which the (linear) momentum of the photon is transferred to the charge carrier. This effect is only present upon oblique incidence of the light on the sample [46, 57, 66], is connected to the symmetry at the surface [46], and can be dependent on the helicity [72]. Due to the momentum transfer, a momentum imbalance is created that can give rise to creation of a photocurrent (∼100 pA cm²/W [57]).

Showing similar features as the photon drag effect, the photogalvanic effect in topological insulators due to asymmetric scattering at the surface has been reported [56], which is observed especially in the THz regime where electron momenta get aligned along the optically created electric field. This electric field does not yield a photocurrent, but rather the asymmetric scattering of the electrons gives rise to such currents (∼40–1000 pA cm²/W [56]). The asymmetry is coming from the inversion symmetry breaking at the surface and the preferentially oriented wedges at the surface and can be indirectly proven by a vanishing photocurrent at twisted domains [52]. This effect and the photon drag effect have the same dependence on the orientation of the linear polarized light but can be distinguished by performing front and back illumination of the sample [52, 57].

Furthermore, circular photogalvanic effects could be coming from the selective
excitation of Rashba-split states located in the band-bent electronic structure at the surface (see chapter 3). The Rashba states can be visualized by split parabolic bands giving rise to partial cancellation of the spin at a certain energy. Nevertheless, it has been reported that Rashba states can give rise to (additional) circular photogalvanic effects [67], which could be distinguished from that due to the topological surface states by gating dependence [72]. Another commonly observed effect is that of laser-induced thermoelectric current (∼0.5–20 pA cm²/W [48, 66, 68]), which is due to the high Seebeck effect in Bi-based topological insulators with respect to that of the metallic contacts and is independent of the helicity [48,53,66,68,69]. This effect can be eliminated by illuminating the complete sample or by performing a line scan between the contacts to find the position where such effects are minimized. The possibility of having many effects thus requires a careful analysis on the origin of the generated photocurrent.

4.2.2 Transient charge-carrier dynamics in topological insulators

From tr-ARPES investigations and THz spectroscopy, one can get to know more about the charge-carrier dynamics in the system and more importantly the interaction between the bulk and surface states. Although the measurements reported in this chapter are performed using continuous illumination, and therefore the transient properties do not seem of importance, it can give additional information on the relaxation mechanisms that can be used to improve photocurrent generation in topological insulators. Furthermore, if one wants to use photocurrents for ultrafast applications then the dynamics become important. For this section, it is assumed that the (Sbdoped) binary compounds Bi₂Se₃, Sb₂Te₃, and Bi₂Te₃ have a similar behavior in their dynamics.

From reports on tr-ARPES measurements on topological insulators, it is often observed that after optical excitation with energies much larger than the bulk band gap the carriers thermalize through electron–phonon coupling within several ps towards the bottom of the conduction band [10,13–19,21,22,24,27–29], whereas cooling at the surface is limited [17]. From the bottom of the conduction band, acting as an electron source for the surface states, electrons are constantly supplied to the surface states. This occurs because phonon-mediated recombination (the highest phonon energy is ten times smaller than the bulk band gap) and direct recombination (appearing at time scales > 1 ns) are unlikely to occur, partially due to the presence of space charge layers [12,19,22,27]. The slow recombination leads to a metastable state present for several tens of ps. This metastable state is furthermore enhanced by a bottleneck effect at the Dirac point of the surface states where the density of states diminishes and therefore limits further relaxation [21]. Other proposed relaxation mechanisms are the diffusion of charge carriers away from the surface into the bulk [14,28] and Auger recombination [76]. Besides momentum relaxation, it is reported that the created (out-of-plane) spin polarization is removed within 500 fs, which indicates that due to strong spin–orbit coupling electron–electron scattering is dominant in the bulk [12,24,29]. Additionally, surface resonances seem to play a role [23,29].

The decay times on the order of picoseconds as reported above are confirmed by
Experimental Methods

THz measurements too [45,47,48,51], where the main part of the photocurrent decays in $\sim 3$ ps and completely vanishes around 10 ps [48]. In the same work by Kastl et al., it has been shown that the photocurrent survives up to 4 ps due to the helicity protection of the surface states that is directly linked to the spin-relaxation time and thus much longer than relaxation times found from tr-ARPES measurements. Beyond 4 ps thermoelectric effects come into play, in agreement with work by Seifert et al. [53]. It has been furthermore found in that work that the photocurrent moves with the group velocity at the Fermi level for Bi$_2$Se$_3$, which is important towards device applications. In the work by Sim et al. [45] and in agreement with the work by Valdés Aguilar et al. [47], it has been reported that charge-impurity scattering plays an important role at lower temperatures. Here, the injection of electrons from the bulk reservoir into the surface states is decreased, due to a suppressed electron–phonon/bulk-to-surface coupling, leading to a lower screening of such impurities. This seems to be in agreement with theoretical works that, despite the large dielectric constant that limits electron–electron and charge-impurity scattering, charge-impurity scattering can still play a role at low temperatures [77,78].

4.3 Experimental Methods

In this experiment, a thin film of the $n$-type topological insulator Bi$_2$Se$_3$ is illuminated by a light beam under oblique incidence and the photocurrent is measured via the voltage between two contacts. In this section, the experiment will be explained in more detail and sources of artifacts will be discussed.

To optically access the surface states of Bi$_2$Se$_3$ only, a low excitation energy with respect to the bulk band gap of Bi$_2$Se$_3$ should be used. However, due to the $n$-type character of Bi$_2$Se$_3$ where the Fermi level usually is positioned inside the bulk conduction band, bulk effects will always be present. In order to understand the polarization control of photocurrent in topological insulators and its connection to the bulk, we have performed such experiments using the free-electron laser at the electron linear accelerator with high brilliance and low emittance (FELBE), located at the Forschungszentrum Dresden-Rossendorf, Germany. The setup used for the experiment is shown in figure 4.1a. For the purpose of the experiment, wavelengths of around 8 $\mu$m have been used, corresponding to an infrared (IR) excitation energy of 0.16 eV (about half of the band gap of Bi$_2$Se$_3$). At the source, the power of the generated light beam is between 11 and 12 W and reduces to $\sim$8 W with which the beam enters the optical table. The beam is then oriented through a Fresnel rhomb that controls the light’s polarization, and does not suffer from birefringence effects that can be present in quarter-wave plates. In the experiment, the Fresnel rhomb is mounted on a rotation stage with which the polarization can be switched from left-circular to linear to right-circular polarization. After the Fresnel rhomb, the light is focused on the sample using a parabolic mirror that yields a final power of around 1 W at the sample position (the cryostat’s KBr optical windows are transparent in the IR range). The full width at half maximum (FWHM) of the beam is around 240 $\mu$m, as deduced from a screenshot of the beam profile, taken by the CCD camera (see figure 4.1b). The sample is oriented under an angle of 45° with respect to the
incoming beam, which leads to an effective beam width of \(240 \, \mu m / (\sqrt{2}/2) \approx 340 \, \mu m\). This value is a factor of 1.5 larger than observed from the sample’s reflection as registered by the IR camera, shown in figure 4.1c. Here, a spot size of around 240 \(\mu m\) is observed\(^6\), which is obtained from the known dimensions of the contacts on the Bi\(_2\)Se\(_3\) sample; the spot size is similar to the contact separation of 230 \(\mu m\). Differences between the two values can be related to differences in sensitivity or resolution of the cameras, but have to be taken into account when studying fluence dependencies.

The contacts, as schematically shown in figure 4.1d, are electrically connected to the chip carrier via wire bonding which has a finite chance of piercing through the Ti(5)/Au(70) layer and making direct contact with the Bi\(_2\)Se\(_3\) (see chapter 3). The legs of the chip carrier that is mounted with screws on a copper cold finger are then connected with one of the outputs of the cryostat in which the sample is mounted. Subsequently, the output of the cryostat can be connected to a switch box, where the right contact pads can be connected for voltage measurements, and is led to a lock-in amplifier via a voltage amplifier. The lock-in amplifier measures the voltage between two contact pads that is locked to the frequency of a rotating chopper (running at 150–200 Hz) that is positioned in front of the Fresnel rhomb. The chopper is a modified commercial version where 90% of the chopper is covered by reflecting Al foil that leads to a reduction in fluence of about 90%. By this lock-in technique, the photocurrent difference arriving at both contact pads generated by the chopped light beam can be analyzed from the measured photovoltage. It has to be emphasized that the current generation is different from that in a normal electrical circuit, such that the resistance (\(\sim 600 \, \Omega\) at 15 K) is not a good conversion between voltage and current. One has to rather look at the relaxation times of the photoexcited charge carriers. In contrast to many reports, there was accidentally no ground applied to one of the two contacts, which might lead to partial diffusion in two dimensions (i.e. also away from the contacts) and fluctuations in the local potential due to local charging. During the second measurement shift, the issue of 2D diffusion was being accounted for by making an alternative geometry as shown in figure 4.1e, where ferromagnetic AlO\(_x\)(0.9)/Co(35)/Au(100) contacts were included to investigate the spin character of the generated photocurrent\(^7\). Furthermore, it is important to realize that the measured voltage gives a difference in current arriving at two points and therefore can be considered as a different geometry compared to those reported where an ammeter is connected at one side of the device (I will use the term ‘net current’ hereafter) [66, 71, 72]. In these works, one does not know what current is collected at the other side of the device\(^8\).

\(^6\)This reflection further indicates that not all the light is absorbed. Determination of the reflected intensity was not possible with the tools at hand, but the reflectance is reported to be around 0.5 \([79]\).

\(^7\)The fabricated tunnel barrier, however, was not proper in the sense that the contact showed Ohmic behavior.

\(^8\)Another way to find this out, of course, is by varying the angle of incidence of the beam.
Figure 4.1: (a) Picture of the optics part of the setup used in the experiments. (b) Beam profile as recorded by the CCD camera. The pixel size is about 80 µm. (c) IR beam spot on the sample observed by IR camera indicated by the red arrow. The sample is mounted on the bottom of a copper plate and contains two rows of contact pads with dimensions defined in the following. (d) Schematics of Ti(5)/Au(70) contact pattern used for the first measurement shift for the sample as depicted in (c). A second row of such contacts is separated by 1.6 mm. (e) Schematics of contact pattern used for the second measurement shift. The brown contacts indicate $\text{AlO}_x(0.9)/\text{Co}(35)/\text{Au}(100)$ ferromagnetic (FM) contacts.
4.4 Results and Discussion

This section is divided in two parts. In the first part, the temperature dependence of the helicity-dependent photocurrent is investigated. Furthermore, thermal effects will be discussed that can play a role in the photocurrent generation as observed in our measurements. In the second part, the dependence on the position of the laser spot on the sample of the photocurrent is discussed.

4.4.1 Temperature dependence

As a quick check of the proposed helicity dependence, the voltage $V_{12}$ between two contacts has been measured over time where the helicity of the light beam has been changed every $\sim 50$ s using the contact geometry as depicted in figure 4.1d and more detailed in figure 4.2a. The beam has been positioned as central as possible in between the contacts by use of the cameras. As can be seen in figure 4.2b, switches in the photovoltage can be observed up to 55 K, which indicates that a helicity-dependent photocurrent difference between the two contacts is indeed generated. In addition, a decrease in the helicity-independent background voltage upon increasing temperature is visible. By plotting the difference $\Delta V_{\text{photo}} = \Delta V_{12,\text{right}} - \Delta V_{12,\text{left}}$ and the background photovoltage $V_{12,\text{bg}} = (\Delta V_{12,\text{right}} + \Delta V_{12,\text{left}})/2$ as function of temperature, a linear dependence is obtained where $V_{12,\text{bg}}$ decreases twice as fast compared to $\Delta V_{\text{photo}}$, as shown in figure 4.2c. Vanishing of the signal at such a narrow temperature range has also been reported by McIver et al. [66], where they have observed components in the modulation related to the bulk vanishing after $\sim 70$ K. They have ascribed the effect to the presence of photon drag and could be observed due to changes in the absorption for s-wave and p-wave light. However, the absorption has shown to be the same for circularly polarized light in contrast to what is observed here.

Other temperature-dependent effects exist that can yield vanishing of the helicity-dependent photovoltage beyond 55 K. From electrical characterization, as well as from time-resolved studies as discussed above, it is known that the mobility decreases with increasing temperature due to an enhanced electron-phonon coupling at elevated temperatures. From similar samples as will be discussed in chapters 5 and 6, the mobility shows a decrease of about 10 to 15% in the studied temperature range. The decrease in mobility can be attributed to an increase in electron-phonon scattering, which will lead to an increase in surface-to-bulk scattering. Therefore, it can be expected that $\Delta V_{\text{photo}}$ gets relaxed upon increasing temperature over such large contact separations. In addition, such a picture can explain the decrease in $V_{12,\text{bg}}$ upon increasing temperature since less photocurrent arrives at the contact and therefore the voltage difference will be smaller. The exact details of a relaxation mechanism that is related to the helicity-dependent photovoltage, if present, are unclear and cannot be concluded from the present study. For that, a comparison between the relaxation

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9For this experiment the polarity of $V_{12}$ was inverted with respect to that shown ($V_{21}$) in the figure. However, the phase detected by the lock-in amplifier was manually removed such that no statements can be made on the net flow direction of the optically induced current.

10Therefore it is difficult to discuss photocurrents instead of photovoltages for this data.
Results and Discussion

Figure 4.2: (a) Measurement geometry for first measurement shift. Despite the presence of multiple contacts, only three of them have been used that are located in the top left corner of the sample (figure 4.1c). Voltages $V_{23}$ and $V_{21}$ have been measured upon excitation under an angle of 45° with the polarities as shown in the figure. (b) Temperature dependence of photovoltage $V_{12}$ while varying between left and right-circularly polarized light ($\lambda = 8\,\mu m$ and $P_{circular} = 70\,mW$) in time, as indicated by the arrows on the top of the graph. Note: The polarities for the voltage probe as shown here are different for the line scan as shown in figure 4.3. For the results presented in this figure, $V_{12}$ with opposite polarity of the probes was measured instead of $V_{21}$. (c) Temperature dependence of extracted values for $\Delta V_{12}$ and $V_{12,bg}$. The solid lines are linear fits to the data. Slopes of $-1.4\pm0.1\,nV/K$ (black) and $-3.4\pm0.2\,nV/K$ (red) are found.

rates and the generation rate of the photocurrent is required.

Another origin of the observed helicity dependence could be related to changes in the beam position due to the performance of the Fresnel rhomb. We have observed slight shifts in the beam position upon changing the polarization as observed from slight intensity changes in pictures as in figure 4.1b. The shifts are estimated to be within 20% of the beam’s FWHM ($\sim50\,\mu m$), which is relatively large compared to the spacing of the contacts as quoted earlier. This could lead to an asymmetric temperature gradient induced by the beam and can yield a finite voltage drop between the contacts, partially due to the high Seebeck coefficient of Bi$_2$Se$_3$ [80]. It is possible to approximate the induced temperature difference $\Delta T_{\text{max}}$ (with respect to no laser-induced heating) at the center of the beam with power $P$ and waist $w$ (half of the spot size) incident on a material with thermal conductivity $\kappa$ as [81]:
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\[ \Delta T_{\text{max}} = \frac{P}{2\pi \frac{1}{2}\kappa w} \approx 3 \text{ K}, \] (4.2)

using \( P = 100 \text{ mW}, w = 125 \mu\text{m} \) and \( \kappa = 40 \text{ W/(m K)} \) at 25 K [80], where the thermal conductivity \( \kappa \) is strongly temperature dependent. Such a strong temperature-dependent thermal conductivity of Bi$_2$Se$_3$ could explain the change in the helicity-independent \( V_{12,\text{bg}} \) with temperature, since the induced \( \Delta T_{\text{max}} \) can vary strongly.

The simple model used here shows agreement with another work where the induced temperature was calculated [53], giving confidence on the applicability of this model that assumes that all the heat is generated in the surface layer. In a slightly modified version, the spatial dependence of the induced temperature difference \( \Delta T(R) \) can be calculated:

\[ \Delta T = \frac{P}{2\pi \kappa w} \frac{1}{R}, \] (4.3)

where \( R = r/w \) with \( r \) being the spatial coordinate. Since \( r_{\text{contact}} \approx w \), i.e. the total spot size is comparable to the separation between the contacts, small temperature differences between the center of the beam and the contact’s position are therefore expected.

An additional effect that could affect the laser beam position has its origin in the thermal expansion of the copper cold finger where the sample is mounted on. Copper at cryogenic temperatures has a thermal expansion coefficient around \( 10^{-6} /\text{K} \) [82] and thus a relative expansion of \( \Delta L/L_0 = 5 \times 10^{-5} \). By assuming a copper block of about 2 cm that is fixed on one side and is only heated very locally, \( \Delta L \) is about 1 \( \mu\text{m} \) with which the beam could be shifted relative to the contacts and therefore is not very likely to cause such a strong temperature dependence. Furthermore, the Seebeck voltage due to the temperature gradients discussed here is mainly canceled out because this effect is isotropic around the incoming beam and the voltage difference is measured. Nevertheless, assuming a Seebeck coefficient of \(-50 \mu\text{V/K} \) [80], any differences in the \( \mu\text{K} \) range can already give rise to photovoltages on the order of 1 nV, which correspond to beam displacements on the order of 1 nm with the settings used above. Interestingly, the beam shift due to polarization change is expected to be orders of magnitude larger, which would give rise to change of the photovoltage in the \( \mu\text{V} \) range.

However, treating laser-induced heating effects only in the lateral plane is not enough, because of the penetration depth of the laser. The penetration depth is inversely proportional to the complex part of the refractive index for the used wavelength [58] and is expected to be large [83]$^{11}$. Therefore, the laser will penetrate into the sapphire substrate which can act as a heat sink. Treating this problem only in the lateral plane is therefore not enough and one should consider heat transport in three dimensions. However, strong thermoelectric effects have not been reported in a very similar material system and geometry [69], where modulations in the photovoltage are on the order of 1 \( \mu\text{V} \) without hardly any background signal. The situation as sketched

$^{11}$Data on the complex part of the index of refraction in this wavelength range for Bi$_2$Se$_3$ is found to be limited.
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above is therefore too simplified to make quantitative statements, but heat-induced thermoelectric effects are important to be considered.

4.4.2 Position dependence

As can be concluded from the discussion in the previous section, the temperature dependence only does not give a conclusive picture on the origin of the observed effects, since artifacts can mimic the signals easily. In order to account for possible beam shifts, lasers scans are made along the $z$ direction while monitoring both $V_{23}$ and $V_{21}$ at $T = 15$ K as depicted in figure 4.2a. The scan is started on the right-hand side of contact 1 and continues beyond contact 3. In addition to possible beam shifts, it has been observed that the Fresnel rhomb transmits a different light power ($\sim 10\%$) for different helicities, indicating that the initially used azimuthal orientations of the rhomb (ideally $\pm 45^\circ$ for circularly polarized light and $0^\circ$ for linear polarization) were not correct\textsuperscript{12}. In order to account for changes in the power at a particular helicity of the light, the power after the Fresnel rhomb has been measured as function of the azimuthal orientation of the rhomb (the angles were off by 7–10$^\circ$). In this way, we reach a maximum power for linear polarization and equal power for both left and right-circularly polarized light. Furthermore, to avoid any problems with the beam position in the $z$ direction, the data has been corrected by finding the minimum voltages around the contact for every polarization that give the center position of the contact. The dependence of the $z$ position (read off from the micrometer screw on the translation stage) on $V_{21}$ and $V_{23}$ is shown in figure 4.3a and b, respectively.

The results will be discussed along with the schematics as depicted in figure 4.3c where, based on the combined data of $V_{21} = V_2 - V_1$ and $V_{23} = V_2 - V_3$, the relative voltage $V_i$ [related to the electronic potential via $-eV_i$] at every contact is depicted by the black dots and studied for different laser positions (red dots) and polarizations. First of all, it is observed that there is a background voltage present that is $\sim 10^3$ larger than observed for the temperature-dependent studies (figure 4.2b), showing the lowest voltage at contact 1 and highest voltage at contact 3. The background voltage can be related to a net electron current flow in the $+z$ direction, that is, more electrons are arriving at contact 1 compared to contact 3. This background current is strongly suppressed when the laser spot is located at the contact pads that strongly absorb the laser light\textsuperscript{13}, which indicates that the background current is mainly related to the interaction with Bi$_2$Se$_3$. Notably, it is important to realize that the net current always flows in the same direction, irrespective of the position of the laser spot, but the signal disappears if the beam is blanked. An obvious thermoelectric effect, as earlier discussed for the temperature-dependent studies, could therefore not explain the origin of the background current here. One possibility on the origin of such a current can be photon drag that would be related to the exact angle of incidence of the beam. If the beam is not fully within the $xy$ plane, the finite $z$ component can lead to such photon drag effects and can give rise to the directional current observed

\textsuperscript{12}This can be solved by using a tunable attenuator [68].

\textsuperscript{13}The laser spot size is comparable to the contact pad size so full absorption is unlikely, consistent with a residual signal of $\sim 0.1$ mV. The skin depth for Au at the used wavelength is calculated to be around 13 nm and thus yields full absorption at the pad.
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Figure 4.3: Laser position dependence of (a) $V_{21}$ and (b) $V_{23}$ according to the geometry as depicted in figure 4.2a for right (black) and left (red) circularly polarized light ($\lambda = 8$ $\mu$m, $P_{\text{lin}} = 170$ mW, and $P_{\text{circular}} = 110$ mW) at $T = 15$ K. The average of these two polarizations (blue) has been given as a guide to the eye for the relative changes. For convenience, the positions of the pads are indicated by the light gray rectangles. The dashed lines and the quoted voltages indicate the peak modulation when the laser is located at contact 2 compared to the satellite contacts. (c) Schematics depicting the relative changes (indicated by the arrows) in the voltage at the contacts (black dots), for different laser positions (red dots) and different polarization of the light as indicated on the right-hand side of each schematic, based on the results from (a) and (b). The light shade of gray of some of the arrows indicates that the voltage at these points most likely does not change. (d) Comparison of the position dependence of the averaged signal (left and right-circularly polarized light) and that for linear polarized light showing clear differences in magnitude.

here. However, a quantification of the effect is difficult. In order to check for such an effect, the angle of incidence should be varied such as to minimize/maximize photon drag effects.

Another observation is that when the laser is located at the contact, right-circularly polarized light always shows a larger voltage compared to left-circularly polarized light. This in agreement with the observation of an apparent brighter reflection from the sample for right-circularly polarized light, indicating differences in the power due to most probably limited calibration of the Fresnel rhomb. Furthermore, as can be
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observed from the results is that when the laser is incident on contact 2, $V_{21}$ and $V_{23}$ show remarkably different behavior. For $V_{21}$, a change in the polarity of the voltage is observed, whereas for $V_{23}$ a suppressed peak is shown compared to the satellite peaks when the laser spot is positioned at the other contact pad locations. Interestingly, when comparing the peak amplitudes (as indicated by the dashed line in figures 4.3a and 4.3b), the enhancement or suppression of the peak is very similar, i.e. the voltage at (most likely) contact 2 is lowered by $\sim 0.2$ mV when the laser spot falls on this contact. It has to be realized that the voltages measured are the differences between two contacts and since the exact mechanism of the background current is unknown it is difficult to tell where such a drop in voltage comes from. The effect could be related to an interaction between the bond wire and the laser spot which causes similar changes in $V_{21}$ when the spot is on contact pad 1. Since the polarity of the contacts is opposite, such interactions might enhance the voltage when the laser spot is one side and suppress the voltage when the laser spot is on the other side.

Looking at the response of Bi$_2$Se$_3$ when the laser spot is in between the contacts, something interesting is observed that is clarified with the schematics in figure 4.3c. As observed from the voltages $V_{21}$ and $V_{23}$, an enhancement in the voltage is found for left-circularly polarized light when the laser spot is located in between the contacts, which could come from helicity-dependent, surface-state-related photogalvanic effects as discussed in section 4.2.1. These changes in the voltage are translated in changes in the voltage at (either of) the contacts (figure 4.3c), as indicated by the arrows. The voltage at the third contact that is furthest away from the laser spot principally can change too but it is expected that the contact in between the laser spot and the third contact effectively screens such small differences (therefore these changes are indicated by a light gray arrow). From the relative shifts in $V_{i}$, it is suggested that left-circularly polarized light induces an additional net electron current in the $+z$ direction, whereas right-circularly polarized light induces an opposite net electron current in the $-z$ direction.

However, from these data it cannot be concluded whether this effect is a circular photogalvanic effect since there might be finite linear components in the polarization that can contribute to such generated currents related to the performance of the Fresnel rhomb. This can be validated by looking at the results for the laser’s position dependence on the voltage using linear polarized light, as shown in figure 4.3d. As noted before, the illumination power of linear polarized light is larger by 40% with respect to that of circularly polarized light, which can be concluded from the difference between the data of linear polarization and that of the average of left and right-circularly polarized light. This would require the data for linear polarization to be rescaled by the power, but due to the feature-rich position dependence an exact overlap is difficult to realize. A comparison with linear polarized light is thus difficult and would require a rotation dependence of the Fresnel rhomb on the generated voltage, which is subject to changes in the laser beam position and laser power on itself.

Effects such as the circular photogalvanic effect scale quadratically with the electric field and therefore should depend linearly on the laser intensity, as shown in equation (4.1). This relationship can be checked by measuring the generated photovoltage $V_{23}$ for left and right-circularly polarized light where the intensity is changed by
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inserting (a combination of) attenuators with a different optical density. The fluence dependence of $V_{23}$ is shown in figure 4.4 in which the fluence $F$ has been normalized for the maximum fluence ($\sim 200$ W/cm$^2$ without attenuators). The data has been acquired on a different moment such that the values are not to be compared with previous graphs in the same figure. The voltage $V_{23}$ seems to indeed follow a linear dependence for both helicities suggesting that the voltage is indeed generated by an effect that scales quadratically with the electric field. However, no ultimate statement can be made on the actual effect. Nonlinearities as those reported by McIver et al. have not been observed [66]. Interestingly, the fluence dependence does not show the same linear slope for both helicities, which can be expected if multiple (helicity-dependent) effects play a role in generating the photovoltage that have different linear dependence.

In the second measurement shift, we have used the contact scheme as indicated in figure 4.1e where the voltage differences in both in-plane directions can be measured as well as the spin character by the ferromagnetic contacts. This could give some insights into the preferential (perpendicular) direction of the photocurrent if it would be related to the surface states [66, 67]. First of all, it has been observed that the measured voltage differences are of similar magnitude as measured before. Besides that, it has been found that the switching of the signal was present up to at least 75 K; due to time constraints we have not been able to study the signal at higher temperatures. The main result of this measurement shift is shown in figure 4.5, where a two-dimensional laser position dependence was performed at 15 K for the horizontal and vertical voltage differences $V_{57}$ and $V_{64}$, based on the scheme depicted in figure 4.5a.

From the results in figures 4.5b and 4.5d, it is observed that the measurements for left-circularly polarized light are both influenced when the light is close to pads 5 and 7, which in contrast to the measurements for right-circularly polarized light (figures 4.5c and 4.5e). This observation might indicate that the orientation of incidence of the beam for left and right-circularly polarized light is different. This leads to the result that the pads or bond wires are being hit differently, yielding different reflections or thermal gradients. Neglecting these ‘hot spots’, it can be seen that $V_{64}$ shows
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similarities for both helicities (figures 4.5b and 4.5c): a positive voltage (\(\sim 0.15 \text{ mV}\)) when the laser spot is in the center of the pattern and negative voltage (\(\sim -0.15 \text{ mV}\)) when the laser spot moves away from the center in the \(y\) direction towards both inner edges of contacts 4 and 6. Considering the quoted spot sizes, this observation can be related to partial absorption of the beam, yielding a lower helicity-independent photovoltage. The negative sign of the photocurrent might be related to an ever present negative background voltage on top of which the modulations occur. A change in the

![Figure 4.5](image-url)

Figure 4.5: (a) Measurement geometry for second measurement shift. Yellow contacts 5 and 6 indicate Ti(5)/Au(70) layers (NM) and the brown contacts 4 and 7 indicate AlO\(_x\)(0.9)/Co(35)/Au(100) ferromagnetic (FM) contacts where contact 7 has been poled in the \(-y\) direction before loading into the cryostat. Voltages \(V_{57}\) and \(V_{64}\) have been measured upon excitation under an angle of 45° with the polarities as shown in the figure. Laser position dependence of \(V_{57}\) for (b) left-circularly polarized light and (c) right-circularly polarized light (\(\lambda = 8 \mu\text{m}\) and \(P_{\text{circular}} = 60 \text{ mW}\)). The rectangular regions indicate the contact pad position that is estimated from maxima in the reflection of the laser spot. Laser position dependence of \(V_{64}\) for (d) left-circularly polarized light and (e) right-circularly polarized light. Note that the \(z\) axis is inverted such as to realize an easy comparison with the scheme depicted in (a). Furthermore, note that the position of 0 V is not always in the middle and therefore the colors in the different plots cannot be directly compared.
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voltage when the laser spot crosses contact pad 4 in the $y$ direction is not expected since photovoltages generated from the topological surface states would develop in the $z$ direction, as is in agreement with the map for $V_{64}$ around contact pad 4. For $V_{64}$ in the case of right-circular polarization (figure 4.5c), one could state that crossing the contacts in the $z$ direction shows a clear difference. However, if this was due to the topological surface states the behavior of the photovoltage at contact 4 (yellow to blue) should be inverted at contact 6 (blue to yellow), which is not the case. Similar difficulties on assessing the (origin of the) photovoltage arise when the helicity dependence of $V_{57}$ is investigated (figures 4.5d and 4.5e). For the right-circularly polarized case (figure 4.5e), a strong effect is observed when the laser spot hits pads 4 and 6, similar to that observed for $V_{64}$. However, such an effect is absent for left-circularly polarized light where illumination at pads 5 and 7 dominate the photovoltage map. As observed from the scans in figure 4.3e, it is expected that the photovoltage will change in this case when crossing a contact pad in the $z$ direction (blue to yellow). However, this is only the case very locally and thus very dependent on the position.

The presence of artifacts (that yield stronger signals than when the spot is inside the pattern) and the complexity of the 2D scan (we observe changes in the photovoltage even when the beam is outside the contact pattern) makes it difficult to identify any helicity-dependent effects. The measured photovoltage is highly sensitive to the position and orientation of the incoming helical light, making it very difficult to reach to a definitive conclusion; also in regard of the earlier shown results.

4.5 Conclusions

In this chapter, an overview has been given of the most important work on photocurrent generation as well as the transient charge-carrier dynamics in topological insulators. The exact mechanism behind the optical control of (spin) currents in topological insulators using circularly polarized light is still under debate and requires more theoretical as well as experimental investigation to unravel such processes where bulk and surface states will be both contributing. In order to exclude bulk effects in the generation of surface-state-related photocurrents, we have examined the photovoltage by IR excitations from a free-electron laser. Although the results show interesting features that could be related to effects mediated by the surface states, it is difficult to draw final conclusions due to experimental difficulties on beam alignment, the type of readout, and the strong position dependence. Nevertheless, it is believed that our work gives reason for further investigation when the laser’s spot size is smaller or comparable to the contact separation. In performing the experiments in this way, the position dependence of the laser and factors that can influence beam alignment are important to be taken into consideration.

4.6 References


References


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References


