Time-resolved lateral spin-caloric transport of optically generated spin packets in n-GaAs

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Abstract
We report on lateral spin-caloric transport (LSCT) of electron spin packets which are optically generated by ps laser pulses in the non-magnetic semiconductor n-GaAs at $T \lesssim 35$ K. LSCT is driven by a local temperature gradient induced by an additional cw heating laser. The spatio-temporal evolution of the spin packets is probed using time-resolved Faraday rotation. We demonstrate that the local temperature-gradient induced spin diffusion is solely driven by a non-equilibrium hot spin distribution, i.e. without involvement of phonon drag effects. Additional electric field-driven spin drift experiments are used to verify directly the validity of the non-classical Einstein relation for moderately doped semiconductors at low temperatures for near band-gap excitation.

Keywords: spin caloritronics, spin transport, time-resolved Faraday rotation, GaAs

Supplementary material for this article is available online
(Some figures may appear in colour only in the online journal)

1. Introduction
Transport of spins is a crucial functional process of spintronic devices as spin injection, manipulation and detection usually take place in different parts of a device [1–10]. In semiconductor spintronics, the spin transport parameters (diffusivity, velocity and dephasing time) determine the length and time scale for manipulation and propagation of the non-equilibrium spin distribution after spin injection or optical excitation with excess energy above the band gap. Most important is that the spin diffusion length exceeds the device dimensions. With the discovery of the spin Seebeck effect (SSE) [11], a new scheme of triggering spin transport by temperature gradients was demonstrated, initiating the field of spin caloritronics [12, 13].

The spin Seebeck physics is intensively studied in magnetic metals, semiconductors and insulators [11, 14, 15]. The SSE can be probed in a transverse configuration where a thermal gradient is applied to a ferromagnetic material which excites the spins out of thermal equilibrium. This results in thermal spin diffusion into an adjacent normal metal perpendicular (transverse) to the direction of heat flow where it is detected [13]. In contrast, in the longitudinal Seebeck effect, the direction of thermal spin injection into the metal is parallel to the temperature gradient [16, 17]. The latter is only well-defined for insulating ferromagnets as parasitic contributions may lead to the anomalous Nernst effect [18]. Moreover, the spin-dependent Seebeck effect has been measured across ferromagnet-to-metal interfaces where a heat current across the interface results into a spin accumulation on the metal side [19]. The measurement scheme is similar to lateral non-local spin-valve [20] with the electrical injection being replaced by thermal spin injection. A thermal spin flow can also be generated across a tunneling barrier between magnetic and non-magnetic electrodes of different temperatures which results in Seebeck spin tunneling [21]. A giant SSE in the non-magnetic semiconductor InSb was explained by the same processes, except that here the phonon-mediated out-of-equilibrium magnetization is carried by a magnetic field-induced spin polarization of conduction band (CB) electrons [22]. More recently, magnetic insulators have been used to measure the
spin Peltier effect [23] and the spin Nernst effect [24] in Pt-yttrium-iron-garnet heterostructures. Although other spin-caloritronic effects are also predicted for non-magnetic semiconductors [25–27], experimental work on these materials is still lacking.

Here, we present diffusive transport of an electron spin polarization in a local temperature gradient in non-magnetic n-GaAs. In contrast to the SSE observed in magnetic materials using inverse spin Hall effect detection, in our experimental approach, we take advantage of the spatial and temporal resolution of optical pump-probe experiments. These enable to directly map and detect lateral transport of electron spin packets driven by laser-induced local temperature gradients in n-GaAs. The basic transport feature is a spin-polarized analog of the charge-based Seebeck effect, except for the detection scheme. A fascinating and unique feature of our lateral spin-caloric transport (LSCT) under local laser heating is that the hot-electron driven spin transport is free of phonon drag contributions. The method yields direct access to the spin transport parameters, such as the spin diffusion velocity \( v_s \), the spin diffusion coefficient \( D_s \), and the spin dephasing time \( T^* \). We show that the diffusive LSCT is corroborated by the dependence of the spin diffusion velocity \( v_s \) on the heating laser position, on the lattice temperature \( T_L \) and on the heating laser power. Further evidence stems from the independence of the LSCT-Seebeck coefficient from the lattice temperature \( T_L \) as well as of the spin diffusion constant \( D_s \) from the electric field and the overheating of the electron system \( \Delta T = T_e - T_L \) with \( T_e \) being the electron temperature. The dependence of \( D_s \) and of the spin dephasing time \( T^*_s \) on the heating laser spot position gives evidence for enhanced electron densities resulting in electron–electron scattering and screening effects. The spin transport parameters are contrasted with corresponding values from our electric field-driven spin drift experiments. The latter allow to verify the non-classical Einstein relation which was predicted for moderately doped semiconductors at low temperatures, but has not been experimentally verified with respect to the electronic states involved [28, 29].

The donor band (DB) states, modeled by a Gaussian density of states, play a crucial role in this validation.

### 2. Experimental setup

The GaAs sample is Si-doped close to the metal-to-insulator transition (MIT) with \( n = 2 \times 10^{16} \text{ cm}^{-3} \) providing spin dephasing times exceeding 100 ns at low temperatures [5, 30–34]. A specimen of 2 mm \( \times 14 \text{ mm} \times 350 \mu \text{m} \) with electrical contacts at its ends is mounted in a helium flow cryostat. As depicted in figure 1(a), electron spins are excited and probed by Faraday rotation \( \theta_F \) of time-delayed, linearly polarized pulses (0.5 mW) and probe pulses from the laser being focused to a 35 \( \mu \text{m} \) spot onto the sample with the pump being positioned at variable distances \( \xi_H \) from the probe spot (figure 1(a)). This yields local heating of the electron system by hot carrier excitation into the CB while leaving the lattice almost unaffected [36]. The local effective electron temperature \( T_e \) is determined from photoluminescence (PL) spectra after low intensity excitation by the heating laser with \( P_H = 0.1 \text{ mW} \), as shown in figure 1(b) for lattice temperatures \( T_L = 10, 20 \) and 30 K. The CB-to-acceptor transitions (\( e, A^0 \)) at the high-energy side of the main emission line around 1.495 eV can be fitted by spectral profiles \( I(E) \propto D(E) \cdot f(E, T_e) \) (red lines), given by the CB occupation function with \( D(E) \) being the density of states and \( f(E, T_e) \) the Fermi function [37]. The extracted \( T_e \) values are plotted as a function of \( T_L \) in figure 1(c). The strongest increase \( \Delta T = T_e - T_L = 23 \text{ K} \) is found at \( T_L = 6 \text{ K} \), decreasing for larger \( T_L \) and vanishing at \( T_L \approx 35 \text{ K} \) due to efficient thermalization of the hot electrons with the lattice by optical-phonon emission [36, 38]. For the following we assume a \( T_e \) profile with the Gaussian shape of the heating laser spot but slightly increased width \( \sigma_H \) due to electronic heat diffusion, \( T_e(\Delta \xi, \xi_H) = T_L + \Delta T \cdot \exp \left[ - (\Delta \xi - \xi_H)^2 / (2 \sigma_H^2) \right] \) [37]. Thus, electron temperature gradients of approximately 0.5 K \( \text{µm}^{-1} \) can be achieved by local laser heating.

### 2.2. Analysis method for LSCT

For the investigation of LSCT in n-GaAs, a dedicated measurement and analysis method is developed allowing us to observe the spatio-temporal evolution of the electron spin packets on time scales comparable to the spin dephasing time, i.e. of \( T^*_s \gg 100 \text{ ns} \) at low temperatures, with high spatial resolution of the spin diffusion. Due to the long spin dephasing time of \( T^*_s \gg 100 \text{ ns} \) as compared to the laser repetition interval of \( T_{\text{rep}} = 12.5 \text{ ns} \), Faraday rotation measures the superposition of many electron spin packets excited by consecutive pump pulses. The Larmor precession of the spin packets in an applied magnetic field \( B \) results in resonant spin amplification (RSA) [30, 31, 39]. In figure 2(a), we show a series of RSA measurements taken at various pump-probe separations \( \Delta t \) for a heating laser distance \( \xi_H = 25 \mu \text{m} \) (see also figure 1(a)) and at a fixed pump-probe delay of \( \Delta t = -50 \text{ ps} \). This yields the \( B \)-dependent superposition of the spin packets, \( \theta_B(\Delta \xi, B) = \sum_n \theta_B(\Delta \xi) \cdot \cos \left( g \mu_B B t_n / h \right) \), with the electron g-factor \( g \), Bohr’s magneton \( \mu_B \) and Planck’s constant \( h \). Each spin packet is characterized by its specific age \( t_n = n T_{\text{rep}} + \Delta t \) and precesses with its Larmor frequency \( \omega = g \mu_B B / h \) about the \( B \) field direction. Because \( |\Delta t| \lesssim T_{\text{rep}} \), the RSA traces can be treated as a Fourier series with coefficients.
The Gaussian width of both pump and probe laser spots is $\sigma \approx 10 \, \mu m$. Consequently, the fast Fourier transformation (FFT) of the RSA scans in figure 2(b) shows resonances which are the amplitudes $\theta_n(\Delta x)$ of the spin packets $n = 1, 2, 3$ etc at the pump-probe separation $\Delta x$. As each spin packet has its specific age $t_n$, the corresponding FFT spectra in figure 2(b) directly show the time evolution of the spin polarization at $\Delta x$. While the FFT amplitude decreases most rapidly at $\Delta x = 1 \, \mu m$ (upper curve in figure 2(b)) due to the presumably combined effects of spin dephasing and diffusion away from the center of the spin distribution at $\Delta x = 0$, the decrease becomes less at larger $|\Delta x|$ (lower curves in figure 2(b)) as the effect of spin dephasing is partially compensated by diffusion of the spins to the tail of the spin distribution. By plotting the integrated FFT amplitudes of each spin packet as a function of $\Delta x$, see figure 2(c), we can reconstruct the lateral profiles of the spin packets at times $t_n$ that show their spatio-temporal evolution. The centers of the spin packets (black dots) move away from the heating laser centered at $\xi_H$ (dashed black line), revealing heat-induced spin diffusion. The heating laser provides a temperature gradient which is controlled by the heating laser position $\xi_H$. Lines are linear fits $\xi_H(t) = v_s t$. The fit range is indicated by the grey shaded area. (c) Effective electron temperature $\Delta T = 23 \, K$.

Figure 1. (a) Optical pump-probe setup to measure lateral spin transport in laser-induced temperature gradients or applied $E$ fields in GaAs. (b) PL spectra and fits of the $(e, A')$ transitions (red lines) at different lattice temperatures $T_L$. The fit range is indicated by the grey shaded area. (c) Effective electron temperature $T_e$ as a function of lattice temperature $T_L$. Below $T_L \approx 35 \, K$ the electron system becomes locally hotter than the lattice, creating an electron temperature gradient of approximately $0.5 \, K \, \mu m^{-1}$ at $T_L = 6 \, K$. The grey line indicates the thermal equilibrium condition $T_e = T_L$.

Figure 2. (a) RSA traces at $T_L = 6 \, K$ for different pump probe separations $\Delta x$ with the heating laser at $\xi_H = 25 \, \mu m$. (b) Corresponding FFT spectra. The amplitude of each spin packet characterized by its specific age $t_n = n t_{spr}$, is given by the integrated FFT resonances (colored and grey points). Both RSA traces and FFT spectra are offset for clarity. (c) Lateral spin profiles are reconstructed from the integrated FFT resonances as indicated by the red, green and blue guide lines between panels (b) and (c). The centers of the spin packets (black dots), extracted from Gaussian fits to the lateral spin profiles (colored and grey lines), shift away from the heating laser spot (broken black line). (d) The center shifts $\xi_H(t)$ of the spin packets change sign with the direction of the $T$ gradient which is controlled by the heating laser position $\xi_H$. Lines are linear fits $\xi_H(t) = v_s t$.
can be extracted from the center shifts \( x_c(t) = v_s t \), the profile broadening \( 2 \nu(t)^2 = 4 \sigma^2 + 4 D_s t \) and the decrease of the profile amplitude \( \theta_c(t) = \theta_0 / [1 + D_s t / \sigma^2] \exp (-t / T_s^2) \), respectively. Figure 2(d) shows the temporal evolution of \( x_c \) of the spin packets for different heating laser positions \( \xi_H \). When the heating laser is moved from \( \xi_H = +25 \mu m \) to \( \xi_H = -36 \mu m \), thereby reversing the sign of the temperature gradient \( dT / dx \), this result in a reversal of the diffusion direction because the spin packets always diffuse away from the heating laser spot as expected for LSCT. At \( \xi_H = 0 \), no significant spin diffusion (center shift \( x_c \)) is observed as the laterally averaged temperature gradient in the probe area is zero. Within the error bars, we are able to resolve center shifts of \( \approx 0.5 \mu m \) even with laser spots of Gaussian widths of \( \sigma \approx 10 \mu m \).

Systematic errors of the spin transport parameters due to idealizations in the analysis model, such as the neglect of the finite pump-probe delay \( \Delta t = -50 \) ps and of the magnetic field dependence of the spin dephasing time \( T^* \), which can be seen by the decrease of the RSA resonances with the magnetic field in figure 2(a), are quantified by spin transport simulations presented in the supplementary material (stacks.iop.org/JPhysD/51/214003/mmedia), together with further details of the FFT analysis. These systematic errors are less than 7% of the input parameters of the simulations, confirming the good reliability of our analysis method.

### 3. Experimental results

#### 3.1. Hot-electron driven lateral spin-caloric transport (LSCT)

We now explore the dependence of the hot-electron driven LSCT on the temperature gradient by investigating the behavior of the spin transport parameters \( v_s, D_s, \) and \( T^*_s \) on the heating laser position \( \xi_H \) in figures 3(a)–(c). LSCT is measured for spin excitation both into the CB states with pump energy \( E_L = 1.497 \text{ eV} \) (filled squares) and into the donor band (DB) states with \( E_L = 1.474 \text{ eV} \) (open squares), corresponding to the \((e, A^b)\) and \((D^h, A^b)\) transitions in figure 1(b), respectively. For both spin excitation energies, \( |v_s| \) first increases with increasing distance \( \xi_H \) of the heating laser, reaching a maximum of \( |v_s| \approx 1000 \text{ cm s}^{-1} \) at \( |\xi_H| \approx 25 \mu m \), and decreases for larger \( |\xi_H| \). The sign of \( v_s \) changes with the inversion of \( \xi_H \). This behavior results from the profile of the local temperature gradient which is averaged over the probe spot at \( \xi_H = 0 \). The black line in figure 3(a) is a plot of

\[
\nu_s \propto (-dT/dx) \propto |\xi_H| \exp\left[-\frac{\xi_H^2}{2(\sigma_H^2 + \sigma^2)}\right] \quad (2)
\]

with \( \sigma_H = 23 \mu m \) (corresponding to \( 54 \mu m \) FWHM) and \( \sigma = 9 \mu m \) being the width of the pump beam. The data clearly follow this model which confirms the observation of LSCT. In contrast to \( v_s \), both \( D_s \) and \( T^*_s \) show a symmetric dependence on \( \xi_H \). While \( D_s \) is lowest around \( \xi_H = 0 \) and increases with larger distance of the heating laser, the opposite behavior is observed for \( T^*_s \). This is surprising, as \( T^*_s \) is known to decrease with higher \( T \) [30] which is also seen in our sample (figure 3(g)). Within the Dyakonov–Perel (DP) spin dephasing mechanism [40], such an enhancement of \( T^*_s \) at the heating area might result from enhanced electron–electron scattering which reduces the momentum scattering time between scattering events and thus increases the spin dephasing time. DP spin dephasing also predicts that the spin dephasing time should increase with increasing magnetic field. However, such dependency has never been observed in n-GaAs for doping concentrations close to the metal-to-insulator transition [30]. The overall larger \( D_s \) values and lower \( T^*_s \) times observed for \( E_L = 1.497 \text{ eV} \) (filled squares in figures 3(b) and (c)) indicate that the spins might occupy CB states, while for 1.474 eV (open squares) less mobile states in the donor band (DB) might be excited [32, 41, 42].

The strongest verification of hot-electron driven LSCT is given by the dependence of \( v_s \) on the lattice temperature \( T_L \) (black squares in figure 3(d)) which was measured with the heating laser at \( \xi_H = +25 \mu m \), i.e. at maximum \( v_s \) (see arrow in figure 3(a)). Most strikingly, \( |v_s| \) (black squares) shows the same dependence on \( T_L \) as the effective electron temperature increase \( \Delta T \) deduced from PL measurements (red data points), confirming equation (2). Although PL was measured under low intensity (\( P_H = 0.1 \text{ mW} \)) of the heating laser (compared to \( P_H = 100 \text{ mW} \) used for the LSCT measurements), the inset of figure 3(d) indicates that spin diffusion is already in saturation for \( P_H \ll 0.1 \text{ mW} \), justifying the comparison of
[\nu_s] and \Delta T. The saturation results from the fact that the mean electron excess energy per absorbed photon from the heating laser, \varepsilon, will be rapidly distributed among the whole electron system due to electron–electron scattering which leads to both an increase of the effective electron temperature and the effective chemical potential. When these electrons diffuse away from their excitation spot they are out of equilibrium with the lattice [36]. Therefore, the laser-induced increase of the effective electron temperature is given by

\[ \Delta T \propto \frac{n_{h\text{el}}}{n + n_{h\text{el}}} \propto \frac{P_H}{P_S + P_H} \]  

with \( n \) and \( n_{h\text{el}} \) denoting the electron densities from doping and from optical excitation, respectively, and \( P_S \) being the saturation power. The black line in the inset of figure 3(d) is a plot of equation (3) with \( P_S = 40 \mu W \). The dependencies of \( \nu_L \) on the heating laser position \( \xi_H \), on the lattice temperature \( T_L \), and on the heating laser power \( P_H \) prove that spin diffusion originates from a local temperature gradient generated by a hot electron distribution as described by equations (2) and (3).

The Seebeck coefficient of the hot-electron-driven LSCT, \( S_{\text{LSCT}} \), can be estimated by comparing thermally-driven spin diffusion velocities with those from electric field-driven spin drift measurements. The latter were additionally carried out with electric fields \( E_x \) applied along the x-direction without the heating laser (see figure 1(a)). Figure 4(a) shows a linear dependence of \( \nu_s = -\mu_s E_x \) with a spin mobility of \( \mu_s = 1160 \text{cm}^2 \text{V}^{-1} \text{s}^{-1} \). The maximum spin diffusion velocity \( |\nu_s| \approx 1000 \text{ cm s}^{-1} \) of the LSCT measurements in figure 3(a), which was measured at \( \xi_H = 25 \mu m \) with \( (dT/dx) \approx 0.5 \text{ K} \mu\text{m}^{-1} \), is obtained in our electric field-driven experiments at \( E_x = 0.86 \text{ V cm}^{-1} \) (see figure 4(a)), from which we estimate \( |S_{\text{LSCT}}| \approx 170 \mu\text{V K}^{-1} \) at \( T_L = 6 \text{ K} \). These values are well below typical Seebeck coefficients \( |S| \) in GaAs exceeding 500 \( \mu\text{V K}^{-1} \) at 10 K [43]. The small dependence of \( |S_{\text{LSCT}}| \) on the lattice temperature \( T_L \) in figure 3(e) reaffirms that our LSCT is purely driven by a hot electron distribution without involvement of phonon drag effects. This is a unique feature of our experimental approach using local laser heating, in contrast to typical charge-based Seebeck effect measurements where phonon-drag results in a distinct extremum in the temperature dependent Seebeck coefficient at low temperature which we do not observe in our LSCT measurements [43, 44].

The dependence of \( D_s \) on \( T_L \) (figure 3(f)) is the same for both LSCT (black squares) and electric field-driven experiments (open and filled green squares, see legend of figure 3(a)) indicating that spin diffusion is independent of \( \Delta T \) and \( E_x \). The measured \( T^*_2 \) times in the electric field-driven spin transport (figure 3(g), green squares) differ increasingly from the LSCT data (black squares) for both spin excitation energies starting from \( T_L = 25 \text{ K} \) towards \( T_L = 6 \text{ K} \). At \( T_L = 6 \text{ K} \) this deviation is in agreement with the thermal equilibrium values of \( T^*_2 \) for both excitation energies at large \( \xi_H \) in the LSCT measurements in figure 3(c). This indicates that different spin states are excited and probed for \( E_L = 1.497 \text{ eV} \) and 1.474 eV which become indistinguishable at elevated temperatures (\( T_L \geq 25 \text{ K} \)), independent of the excitation energy and the heating laser, most likely resulting from thermal energy redistribution.

3.2. Testing the Einstein relation using spin drift transport parameters.

A closer insight into which states are involved in the measured spin transport can be gained from the temperature dependence of electric field-induced spin transport. We first note that \( |\nu_s| \)
shows the same increase with $T$ (here $T_c = T_L$) in figure 4(b) as the electrical conductivity $\sigma_D$ (black line) measured by 2-point $I$–$V$-curves. This implies that the electron density does not depend on temperature as expected for itinerant CB or DB charge carriers. From the spin drift velocities in figure 4(b) together with the spin diffusion constant in figure 3(f), we can test the Einstein relation $R \equiv eD_\perp/(\mu kB_0 T)$ that was predicted to deviate from the classical value $R = 1$ for non-degenerate particles, i.e. doped semiconductors close to the MIT at low temperatures [28, 29]. The results in figure 4(c) verify the predicted increase of $R \gg 1$ at low temperatures for fermionic particles. The blue broken line shows the classical case for Boltzmann-distributed systems. The black solid and broken lines are simulations of the generalized Einstein relation according to [28] for CB transport of Fermi–Dirac-distributed electrons with the Fermi-energy $E_F = \hbar^2/(2m_{CB}^*)(3\pi^2n)^{2/3}$ for carrier densities $n$ between $1 \times 10^{16}$ cm$^{-3}$ (lower curve) and $3 \times 10^{16}$ cm$^{-3}$ (upper curve). Remarkably, all experimental data (green squares) clearly exhibit consistently larger $R$ values and thus do not follow simple CB transport. In contrast, our data can be described much better by DB transport over the whole temperature range when assuming a Gaussian density of states of width $\Delta E = 6 \text{ meV}$ with $E_F$ in the center of the band (see green solid line in figure 4(c)). For comparison, we included respective curves with $\Delta E = 5 \text{ meV}$ (lower green broken line) and $7 \text{ meV}$ (upper green broken line). Surprisingly, even spins which are directly excited into the CB at $E_L = 1.497 \text{ eV}$ (full green squares in figure 4(c)) are best described by this DB approach despite their slightly different spin dephasing times at low temperatures as discussed for figure 3(g). Alternative interpretations like ambipolar diffusion [28] or spin Coulomb drag [45] have been ruled out [46].

4. Conclusions

In conclusion, we have demonstrated lateral spin-caloric transport of optically generated spin packets in n-type non-magnetic GaAs at low temperatures. This diffusive spin transport is driven by local temperature gradients from a hot electron distribution due to laser heating. By mapping the spatio-temporal evolution of the spin packets, our experiments give access to the spin transport parameters and thus a microscopic insight into the spin analog of the charge-based transport in the Seebeck effect. As a unique feature of our experimental approach, the driving mechanism of the LSCT is solely governed by excitation of a local hot electron distribution in the CB and therefore free of significant phonon-drag contributions yielding a nearly temperature-independent Seebeck coefficient of the LSCT. Furthermore, we tested the non-classical Einstein relation for moderately doped semiconductors and found a temperature dependence which can be well described by donor band transport. Our results strongly suggest that also thermally-induced transverse spin currents due to spin Nernst effect might be observable by our all-optical spin transport measurement technique.

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