Time and spatially resolved electron spin detection in semiconductor heterostructures by magneto-optical Kerr microscopy

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We review on time and spatially resolved two-color pump–probe magneto-optical Kerr effect (MOKE) microscopy studies of electron spins in bulk n-GaAs and GaAs (110) quantum wells (QWs) at low lattice temperatures. The influence of photocarrier heating by above-bandgap optical spin excitation on the spatially resolved magneto-optical spin detection is considered in detail. We demonstrate that a continuous-wave (cw) measurement of the local Kerr rotation at a fixed arbitrary probe wavelength in general does not correctly reveal the local spin polarization when hot electrons are present. For bulk GaAs we determine the true lateral electron spin polarization profile from cw measurements of the spatial dependence of the full excitonic Kerr rotation spectrum. For the (110) QWs, we directly obtain the electron spin diffusion coefficient from picosecond real space imaging of the time evolution of an optically excited electron spin packet, which we observe with a spectrally broad probe pulse.

1 Introduction Spintronics demands for precise control of the storage, manipulation, and transfer of electron spin coherence in solid state systems [1, 2]. In semiconductors, the time and length scales available for these operations are characterized by the spin relaxation time $\tau_s$, the spin diffusivity $D_s$, and the related spin mobility $\mu_s$ [3, 4]. Pump–probe MOKE spectroscopy is a particularly valuable tool for the all-optical investigation of spin phenomena in semiconductors and the determination of these parameters. Because of its high temporal and spatial resolution it has been used extensively to study spin relaxation and spin transport in bulk semiconductors and low-dimensional systems [5–11].

Pump–probe MOKE spectroscopy and the related spin noise spectroscopy both infer information on the electron spin system of semiconductors by measuring small changes in the polarization of a linearly polarized probe laser. In spin noise spectroscopy, the influence of the specific probe wavelength on the experimental results has received significant attention in the past [12–15]. A similar critical assessment of the influence of the probe energy on results obtained from spatially resolved two-color MOKE spectroscopy, however, has been missing in the respective pioneering works.

We intend to close this gap by summarizing our recent results [16–20] on two-color pump–probe MOKE microscopy of electron spins in bulk n-GaAs and GaAs-based (110) QWs. Both materials are prototypical model systems for the magneto-optical spectroscopy of electron spin dynamics in semiconductors [9, 21–24].

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We focus on the fundamental aspect of spatially resolved MOKE spectroscopy – the reliable determination of the local electron spin polarization from pump–probe Kerr microscopy. For both systems, we demonstrate the
detrimental influence of pump-induced photocarrier heating and photocarrier scattering caused by above-bandgap optical spin excitation on the spatially resolved magneto-optical electron spin detection at low lattice temperatures $T_L$. We show that, in contrast to widespread assumption, a cw measurement of the local Kerr rotation at a fixed, arbitrary, probe wavelength in general does not correctly map the local spin polarization when hot photocarriers are present.

Instead, for bulk GaAs we determine the local excitonic spin splitting energy by analyzing the spatial dependence of the full excitonic Kerr rotation spectrum. From the local excitonic spin splitting, we obtain the true lateral electron spin polarization profile. For the examined sample, we further find that the true electron spin profile is observed by measuring the local Kerr rotation when the probe laser energy is tuned far below the excitonic resonance.

For the (110) QWs, the significantly more complex structure of the excitonic optical transitions impedes a quantitative analysis of the Kerr rotation spectrum. Instead, we directly measure the electron spin diffusion coefficient by picosecond real space imaging of the time evolution of an optically excited electron spin packet, which we observe with a spectrally broad probe pulse.

2 Materials and methods We present MOKE microscopy studies performed on a bulk n-GaAs epilayer and an intrinsic (110) GaAs QW sample.

The bulk n-GaAs sample has been provided by A. D. Wieck (University of Bochum). It was grown on a (001)-oriented GaAs substrate by molecular beam epitaxy (MBE). The sample structure consists of a 50 nm undoped GaAs buffer layer followed by a ten-period GaAs/AlAs superlattice with a period of 5 nm GaAs + 5 nm AlAs. The superlattice is followed by a 50 nm undoped GaAs layer and the top, 1μm thick, Si-doped n-GaAs layer. The room temperature electron density and mobility of the epilayer were determined by standard van der Pauw characterization as $n = 1.4 \times 10^{16} \text{ cm}^{-3}$ and $\mu_e = 5500 \text{ cm}^2/(\text{V s})^{-1}$.

The QW sample has been provided by P. V. Santos (PDI Berlin). MBE growth was initiated on a (110)-oriented GaAs substrate by a 200 nm GaAs buffer layer. The buffer is followed by a 500 nm Al$_{0.15}$Ga$_{0.85}$As layer and a series of five intrinsic GaAs QWs with a width of 20 nm. The QWs are each embedded in 64 nm wide Al$_{0.15}$Ga$_{0.85}$As barriers. The sample is capped by 120 nm Al$_{0.15}$Ga$_{0.85}$As followed by a top 2 nm GaAs layer.

A comprehensive description and detailed schematics of the two-color pump–probe Kerr microscopy setup employed in this work are given in Refs. [18–20]. In brief, for cw measurements the pump and probe beams are provided by two independent, continuously tunable Ti:sapphire lasers. For time-resolved spectroscopy, we use the output of a fiber-based pulsed ultrafast white-light supercontinuum source [25, 26] ($\lambda \approx 400–1400$ nm, 36.4 MHz repetition rate).

The pulsed pump and probe beams are derived from the supercontinuum by dividing the laser output in two components by a beamsplitter and performing individual spectral filtering. The pump laser is passed through a dielectric bandpass which results in an intensity spectrum centered at 7800 Å with a full width at half maximum (FWHM) of 100 Å. Tunable spectral filtering of the pulsed probe laser is realized by dispersing the supercontinuum with a prism and selecting the desired wavelength component by a pinhole placed in the common focal plane of a lens pair. The probe laser has a spectral FWHM of 25 Å ($\approx 5$ meV) for photon energies close to the GaAs band gap. A variable delay $\Delta t$ between the pump and probe pulses is introduced by a mechanical delay line. The overall time resolution of the setup is $\lesssim 3$ ps as directly determined from the observation of very fast initial rises of Kerr rotation transients [20].

The pump and probe laser beams are focused at normal incidence with the same apochromatic microscope objective (NA = 0.42, $f = 4$ mm) on the surface of the sample, which resides in an optical cryostat. Manipulation of the relative position of a lens pair arranged in confocal geometry by a piezo positioner enables lateral scanning of the probe with respect to the pump laser position. The net optical resolution of the setup as determined from the $(1/e)$-halfwidth of the convolution of the Gaussian pump and probe laser intensity profiles is 1.4 and 1.9 μm for cw and pulsed excitation, respectively.

For time resolved and cw measurements the pump laser is periodically modulated at 50.1 kHz between $\sigma^+$ and $\sigma^-$ circular polarization by a photoelastic modulator. Electrons with a net spin polarization $S_{\parallel}$ along the sample normal $z$ are locally excited by the focused pump laser. The local Kerr rotation $\theta_k$ imposed on the linearly polarized probe laser after reflection from the sample surface is measured by a balanced photoreceiver in combination with a standard lock-in detection scheme [27].

3 Continuous-wave MOKE microscopy of electron spins in bulk n-GaAs In two-color MOKE microscopy optical spin injection is achieved by a circularly polarized pump laser whose photon energy is tuned above the fundamental band gap \footnote{The practice of non-resonant excitation in two-color MOKE microscopy stems from the experimental requirement to detune the pump with respect to the probe laser to enable spectral filtering of the collinear pump and probe beams. This is necessary to prevent the saturation of the balanced photoreceiver by modulated pump light.} [9, 23, 28, 29]. It has been known for a long time that in semiconductors the electron temperature $T_e$ can significantly exceed the lattice temperature $T_L$ as a result of the deposition of excess energy in the electron system by such above-bandgap optical excitation [30–34]. This effect is most pronounced at very low lattice temperatures, for which the energetic coupling between the electron and the lattice system is weak, and vanishes with increasing $T_L$ [35].

An overheating of the electron system is observed at rather low power densities of 10 W cm$^{-2}$ for which no heating of the lattice system occurs [16]. Such power densities easily fall below excitation intensities typically required in pump–probe MOKE microscopy as a result of the necessity...
to strongly focus the pump laser to achieve high spatial resolution while exciting a detectable electron spin polarization. At low lattice temperatures pump-induced carrier heating therefore is inherent to the non-resonant optical spin excitation in two-color MOKE microscopy and virtually cannot be avoided.

We have previously used spatially resolved photoluminescence (SRPL) spectroscopy to demonstrate that for local excitation by a focused laser the thermal non-equilibrium between the electron and the lattice system can persist over length scales exceeding tens of microns in bulk GaAs [16]. Here we show that consideration of the presence of such gradients in the electron temperature is of importance for the correct magneto-optical electron spin detection at low $T_L$.

In two-color MOKE microscopy the probe laser is commonly tuned to the $E_0$ excitonic resonance and $\lambda_{probe}$ is chosen such that the observed Kerr rotation is maximized to facilitate the detection of the small polarization signals. Assuming a linear relation $\theta_k = \alpha S$, between the small Kerr rotation and the electron spin polarization, $S(r)$ then is determined by varying the relative distance $r$ between the focused pump and probe lasers and measuring $\theta_k(r)$.

If the magneto-optical response $\alpha$ does not depend on the distance $r$, MOKE microscopy correctly reveals the local electron spin polarization $S(r)$. The excitonic optical resonances in semiconductors, however, are strongly influenced by changes in the local charge carrier concentration and (electron) temperature [36–41], which both may vary significantly with the distance to the pump spot. The influence of this variation of spin-unrelated parameters on the local electron spin polarization. As a consequence, MOKE microscopy performed using an arbitrary probe wavelength – in general – does not correctly reveal the electron spin diffusion profile.

To demonstrate the spatial dependence of $\alpha(r)$ in a prototype MOKE microscopy experiment we measure the excitonic Kerr rotation spectrum $\theta_k(\lambda_{probe})$ for the bulk n-GaAs sample as a function of the radial distance $r$ between the pump and probe spot center. All cw measurements presented here are performed with a pump wavelength $\lambda_{pump} = 7800 \text{ Å}$. The pump and probe laser power are 10 and 50 $\mu$W, respectively.

In Fig. 1a, we compare normalized Kerr rotation spectra measured at the pump spot center and at $r = 25 \mu$m. The antisymmetric lineshape with a zero-crossing at $8189 \text{ Å}$ observed in both spectra is characteristic of an energetic splitting of the $\sigma^+$ excitonic resonances [27, 42]. Most important, we observe that for increasing distance to the pump spot the spectral width of the Kerr rotation resonance is strongly reduced, i.e., the resonance linewidth becomes significantly more narrow.

For $T_e \lesssim 25 \text{ K}$ the cooling of the electrons in GaAs is limited to inefficient emission of acoustic phonons [31]. For electron temperatures exceeding $\approx 25 \text{ K}$, however, longitudinal-optical (LO) phonon emission becomes energetically accessible and the thermal coupling between the electron system and the lattice strongly increases. For elevated lattice temperatures and moderate cw photoexcitation both systems are therefore in thermal equilibrium, i.e., $T_e = T_L$ [16, 35]. In this case, the electron temperature is spatially homogeneous and all effects on the Kerr rotation spectrum previously caused by local photocarrier heating vanish. In Fig. 1a, we show that for $T_e = 30 \text{ K}$ the lineshape of both normalized Kerr rotation spectra indeed becomes identical within experimental resolution, i.e., $\theta_k(\lambda_{probe})$ does not depend on the lateral position for a spatially homogeneous electron temperature.

The implications of this position dependence of the magneto-optical response $\alpha(r)$ for the spatially resolved electron spin detection by pump–probe MOKE microscopy are demonstrated in Fig. 1b. We show lateral Kerr rotation profiles $\theta_k(r)$ (each normalized to the peak value at the pump spot center) measured at $T_e = 8 \text{ K}$ for probe wavelengths between 8170 Å and 8230 Å in steps of 5 Å at low and high lattice temperature. Each profile is normalized to the amplitude at the center of excitation. (d) FWHM of the lateral Kerr rotation profiles as a function of probe wavelength. Adapted from [18].

![Figure 1](image-url)
The observed increase of the profile FWHM for probe wavelengths close to the excitonic resonance energy originates from the interplay of spin diffusion and hot photocarrier relaxation: Moving away from the pump spot center, \( \theta_K(r) \) is reduced by the lateral decrease of the electron spin profile \( S_e(r) \). However, this amplitude decrease is partially compensated by a concurrent transfer of oscillator strength within the Kerr rotation spectrum towards the central resonance energy. This spectral transfer stems from the influence of the spatially dependent photocarrier temperature on the excitonic linewidth and therefore is not spin-related. This leads to a widening of the Kerr rotation profile \( \theta_K(r) \) which masks the true spatial dependence of the local electron spin polarization \( S_e(r) \). Therefore cw measurements of the local Kerr rotation at fixed arbitrary probe wavelengths do not necessarily correctly reveal the local electron spin polarization when hot photoelectrons are present.

For \( T_L = 30 \, \text{K} \), the electron temperature is spatially uniform, the influence of hot carriers on the excitonic resonances vanishes and we observe no spatial variation of the magneto-optical response. In this case, \( \theta_K(r) \) is expected to reflect the actual spin polarization profile \( S_e(r) \) irrespective of the specific probe wavelength. This is observed in Fig. 1c where we show a series of \( \theta_K(r) \) profiles measured for the same probe wavelengths previously examined at low \( T_L \). In contrast to the low lattice temperature case, we find that the width and shape of the profiles only very weakly depend on the probe wavelength. This observation is supported by Fig. 1d where we show that the FWHM of the \( \theta_K(r) \) profiles now only varies between 12 and 15 \( \mu \text{m} \).

We next confirm that the spatial dependence of \( \theta_K(E_{\text{probe}}) \) is quantitatively explained by considering the interplay of the local spin polarization \( S_e(r) \) and the thermal broadening of the excitonic resonance which both depend on the distances to the pump spot. We therefore employ a model that relates the spectral dependence of the Kerr rotation \( \theta_K(h\omega) \) to spin-induced modifications of the dielectric functions \( \tilde{\epsilon}(h\omega) \) for \( \sigma^\pm \) light with photon energy \( E_{\text{probe}} = h\omega \).

From the phase jumps of the complex amplitude reflection coefficients of the electric field under normal incidence [43],

\[
\Delta^\pm = \arctan\left(-\frac{2k^\pm}{n^2 + k^2 - 1}\right),
\]

we can express the Kerr rotation [43]

\[
\theta_K = -\frac{1}{2} \left( \Delta^+ - \Delta^- \right)
\]

in terms of the dielectric functions via the well-known relation \( \tilde{n}^\pm = (\tilde{\epsilon}^\pm)^{1/2} \) for the complex index of refraction \( \tilde{n}^\pm = n^\pm - ik^\pm \).

Following the concept of Ref. [42], we model the dielectric function in the vicinity of the GaAs band gap in terms of a Lorentzian resonance representing the excitonic contribution and a static background \( \epsilon_0 = 11.95 \) [44] which results from higher-energy transitions:

\[
\tilde{\epsilon}^\pm(h\omega) = \epsilon_0 + \frac{A^\pm}{E_0^\pm - h\omega + i\Gamma}.
\]

Here, \( E_0^+ (E_0^-) \) and \( A^+ (A^-) \) are the resonance energy and amplitude for the optical transition involving \( \sigma^+ (\sigma^-) \) photons. The parameter \( \Gamma \) describes the linewidth of the excitonic resonance.

The microscopic origin of the Kerr rotation can be inferred from the distinct influence of a difference of the resonance amplitudes \( A^\pm \) and energies \( E_0^\pm \) on the spectral shape of the measured Kerr rotation spectrum [27]. A difference in the resonance amplitudes \( A^\pm \) is observed as a contribution to the Kerr rotation spectrum which is symmetric with respect to the resonance energy and does not change its sign. In contrast, an energetic splitting of the excitonic resonances \( \Delta E = (E_0^+ - E_0^-) \) results in an antisymmetric Kerr rotation spectrum with a zero-crossing at the mean resonance energy as observed in our experiment.

We now summarize the main results of our analysis of the spatial dependence of the excitonic Kerr rotation spectrum. For a detailed discussion of the Lorentzian resonance model and the fitting procedure we refer to Ref. [18]. In Fig. 2a, we show a series of local Kerr rotation spectra \( \theta_K(E_{\text{probe}}) \) measured at \( T_L = 8 \, \text{K} \) for distances between 0 \( \mu \text{m} \) and 25 \( \mu \text{m} \) to the pump spot center together with the best fits2 from the

\[\text{Figure 2} \] (a) Local Kerr rotation \( \theta_K(E_{\text{probe}}) \) for increasing distance between the pump and probe spot at \( T_L = 8 \, \text{K} \). Curves are shifted vertically. The distance to the pump spot is indicated above the spectra. Red solid lines are fits to a Lorentzian excitonic resonance model (see text). (b) Lateral profile of the excitonic spin splitting \( \Delta E(r) \sim S_e(r) \). (c) Local excitonic linewidth \( \Gamma(r) \) obtained from the Lorentzian resonance model fits. Adapted from [18].

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\[\text{To fit the spectra we assume a value of 2.35 meV for the unknown resonance amplitude } A^\pm, \text{ which is found in Ref. [45] for bulk GaAs with an impurity concentration comparable to our sample. This assumption introduces an uncertainty in the absolute values obtained for the spin splitting energy. However, the general form of the spatial dependence of } \Delta E(r) \text{ is}\]

\[\epsilon_0 = 11.95 \text{ meV}\]

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model Eqs. (1)–(3). The spatial dependence of the energetic splitting \( \Delta E(r) \) and the excitonic resonances linewidth \( \Gamma(r) \) obtained from these fits is shown in Fig. 2b and c.

In the following, we describe how we extract information on the local electron spin polarization and the photocarrier energy relaxation from both parameters. We first consider the spatial dependence of the excitonic resonance linewidth \( \Gamma(r) \). Moving away from the pump spot center, \( \Gamma(r) \) systematically decreases from a maximum value of 2.2 meV to 1.4 meV for distances far away from the pump spot. The strong enhancement of \( \Gamma \) close to the excitation center stems from thermal broadening of the excitonic resonance by the pump-induced local photocarrier heating. In Ref. [18], we have determined the intrinsic temperature dependence of the resonance linewidth \( \Gamma(T_c) \) from ultra-low excitation density measurements for which no heating of the electron system occurs. Using this calibration measurement we extract the local electron temperature profile \( T_e(r) \) from the spatial dependence of the excitonic resonance linewidth \( \Gamma(T_e(r)) \).

In Fig. 3a, we plot the lateral electron temperature profile \( T_e(r) \) which we obtain from this analysis. At the pump spot center we observe a high peak electron temperature of \( T_{e,\text{max}} = (27.3 \pm 2.8) \text{ K} \) which demonstrates the strong pump-induced heating of the carrier system in MOKE microscopy experiments. With increasing distance to the pump spot we observe the cooling of the electrons. \( T_e(r) \) reaches the lattice temperature of 8 K not until \( r \gtrsim 25 \mu \text{m} \). This macroscopic cooling distance significantly exceeds the pump spot size—a direct manifestation of heat conduction in the electron system [16].

We additionally confirm the electron temperature profile obtained from the local excitonic resonance linewidth analysis by comparison with SRPL spectroscopy. In Ref. [18], we have independently determined the local electron temperature from a lineshape analysis [46] of the electron to neutral carbon acceptor (e, \( \Lambda^0_e \)) transition. In Fig. 3a, we show that the spatial dependence of \( T_e(r) \) obtained from the measurement the local excitonic linewidth is well reproduced by the photoluminescence result which substantiates the validity of our approach.

We conclude the summary of our results for bulk GaAs by demonstrating how we determine the electron spin polarization profile \( S_e(r) \) from the local spin splitting \( \Delta E(r) \) of the excitonic resonance. This spin splitting of the excitonic optical resonances results from the presence of spin-polarized electrons. It has been directly observed previously in PL measurements on GaAs quantum wells [47–49] and recently for bulk GaAs and AlGaAs samples [50].

The energetic splitting of the excitonic resonances is proportional to the difference between the density of spin-up and spin-down electrons, i.e., \( \Delta E \sim (n_\uparrow - n_\downarrow) \) [48, 51, 52]. For the relatively high intrinsic carrier concentration of our n-type sample and our low excitation powers, the pump-induced change in the total electron density \( (n_\uparrow + n_\downarrow) \) is small. We therefore directly determine the local electron spin polarization \( S_e(r) = [n_\uparrow(r) - n_\downarrow(r)]/[n_\uparrow(r) + n_\downarrow(r)] \sim \Delta E(r) \) from the local exciton spin splitting.

In Fig. 3b, we show the spin splitting \( \Delta E(r) \) together with representative \( S_e(r) \) profiles observed at different probe wavelengths. Comparing both data sets, we find a strong difference between the lateral Kerr rotation profile \( \theta_{kerr}(r) \) measured at \( \lambda_{\text{probe}} = 8195 \text{ Å} \) and the spin splitting profile \( \Delta E(r) \sim S_e(r) \). Therefore probing the spin polarization with \( \lambda_{\text{probe}} \) tuned to the maximum amplitude of the Kerr spectrum is detrimental to a correct determination of the lateral spin polarization profile at low lattice temperatures. However, for the examined sample the Kerr rotation profile measured at \( \lambda_{\text{probe}} = 8210 \text{ Å} \) perfectly coincides with the position dependence of the excitonic spin splitting \( \Delta E(r) \). Comparing this result and Fig. 1d, we find that the Kerr rotation profiles \( \theta_{kerr}(r) \) measured with long probe wavelengths \( \lambda_{\text{probe}} \gtrsim 8210 \text{ Å}, \) i.e., detuned from the Kerr resonance energy, indeed correctly reflect the true local electron spin polarization. Therefore single-wavelength cw MOKE microscopy here indeed can be used to correctly measure \( S_e(r) \), however at the cost of strongly reduced signal amplitudes.

In this work, we have demonstrated the influence of hot carrier effects on the magneto-optical electron spin detection. However, at low \( T_e \) the temperature gradients induced by the strong pump-induced carrier heating also significantly affect the electron spin diffusion. We refer to Refs. [17, 19] for a detailed analysis of the influence of photocarrier heating on both stationary and time-dependent spin diffusion in bulk GaAs.

4 Continuous-wave MOKE microscopy of electron spins in (110) QWs

Spin phenomena in GaAs-based (110) QWs have attracted significant interest in the past because of the observation of strongly anisotropic spin relaxation rates and nanosecond spin coherence times at
room temperature. However, previous magneto-optical studies [8, 15, 53–55] on (110) QWs have mainly focused on aspects of the anisotropic electron spin dephasing results from by the suppression of Dyakonov-Perel relaxation for out-of-plane oriented electron spins. The small number of experimental results on electron spin diffusion in (110) QWs have mostly been obtained at elevated temperatures from the excitonic magneto-optical response of the (110) QW sample. In Fig. 4b, we show photoluminescence (PL) spectra measured at TL = 8 K and following text). This strongly suggests the presence of a low-density electron gas (2DEG) in the nominally undoped QWs. For low excitation densities peak B therefore might be attributed to the recombination of free heavy-hole excitons. Peaks B and C at 1.523 and 1.521 eV might result from the recombination of defect-bound excitons which saturate with increasing excitation density.

For peak B, however, an alternative transition is more likely: The (110) QW sample exhibits very long out-of-plane electron spin lifetimes T2 ≈ 30 ns (compare Fig. 5a and following text). This strongly suggests the presence of a low-density electron gas (2DEG) in the nominally undoped QWs. For low excitation densities peak B therefore might result from the recombination of negatively charged trions (X−), which can form from optically excited neutral excitons in the presence of excess electrons. Refs. [59, 60] report trion binding energies of 1.3 meV for GaAs QWs with identical widths as our sample. This binding energy coincides with the separation between the exciton peak A and peak B.
observed in our PL measurement, which further substantiates this interpretation.

In Fig. 4a, we compare two local cw Kerr rotation spectra $\theta_k(E_{probe})$ measured at $T = 8\,\text{K}$ for different distances (0 and 25 $\mu\text{m}$) to the pump spot. In both spectra, we see strong contributions from the (1e-1hh) and (1e-1lh) transitions which are also present in the PL spectra. At $\approx 1.534\,$eV we observe an additional weak resonance in the Kerr rotation spectrum which we attribute to the (1e-3hh) transition. This transition is absent in the PL since at low $T$, the lower valence subbands are not populated by holes. However, it is resolved in the Kerr rotation spectrum because of spin-dependent Pauli blockade and an energetic splitting of the $\sigma$ transitions caused by the spin-polarized electrons in the lowest (1e) conduction subband [61–63]. Furthermore, the individual contributions of the transitions A to C of the (1e-1hh) resonance fine structure are resolved in the Kerr rotation spectra. This underlines the significantly more complex structure of the Kerr rotation spectrum of the (110) quantum well as compared to the bulk GaAs sample.

The position dependence of the Kerr rotation spectrum is qualitatively similar to our observations in bulk GaAs: At the pump spot center the resonances are spectrally very broad, but become significantly more narrow when moving away from the pump spot. For the (110) quantum wells, however, the mechanism leading this spatial variation of the magneto-optical response is likely more complex than for the case of bulk GaAs: In addition to the carrier temperature $T$, the lower valence subbands additionally varying magneto-optical response on the electron spin polarization is [66–68]:

$$\frac{\partial S_z}{\partial t} = D_s \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial S_z}{\partial r} \right) - \frac{S_z}{T_z}.$$  \hfill (4)

Here $D_s$ is the electron spin diffusion coefficient and $T_z$ denotes the out-of-plane spin lifetime. Following pulsed excitation by a Gaussian pump spot, the time evolution of the spin polarization is [66–68]:

$$S_z(\Delta t, r) = \frac{\sigma_0^2 S_z(0)}{\sigma_0^2 + 4 D_s \Delta t} \exp \left( -\frac{r^2}{w_s^2} \right) \exp \left( -\frac{\Delta t}{T_z} \right).$$  \hfill (5)

with an initial (1/e) halfwidth $\sigma_0$ and amplitude $S_z(0)$. The time-dependent square (1/e) halfwidth of the spin packet

$$w_s^2 = \sigma_0^2 + 4 D_s \Delta t$$  \hfill (6)

increases linearly with the pump–probe delay $\Delta t$. It is evident from Eq. (5) that for pulsed excitation spin relaxation and diffusion are completely decoupled. Most importantly, the spatial shape of the spin packet is exclusively governed by the process of diffusion while spin relaxation only affects the overall packet amplitude.

In contrast to steady-state spectroscopy, time resolved Kerr microscopy therefore eludes auxiliary measurements of the spin relaxation time for the determination of spin diffusion coefficients [19, 67, 68]. A measurement of the squared spin packet width $w_s^2$ as a function of the pump–probe delay $\Delta t$ directly yields the spin diffusivity

$$D_s = \frac{1}{4} \frac{\partial}{\partial \Delta t} \left( w_s^2(\Delta t) \right).$$  \hfill (7)
To experimentally determine \( w_0^2(\Delta t) \) we observe the expansion of the electron spin packet by repeatedly raster-scanning the probe with respect to the pump spot position and measuring the local Kerr rotation \( \theta_k(r) \). From a systematic variation of the temporal delay \( \Delta t \) between the pump and probe pulse we then directly obtain the spin polarization profile \( S_z(\Delta t, r) \).

For time resolved Kerr microscopy on the (110) QW sample the central probe laser wavelength is tuned to the (1e–1hh) resonance at 8140 Å (compare Fig. 4). The time-averaged probe laser power is 12 µW and the pump power is varied between 1.5 and 5 µW as indicated in the text.

In addition to spin transport phenomena, picosecond Kerr microscopy also allows for the investigation of spin relaxation processes. To demonstrate this capability we first briefly examine the electron spin relaxation in the (110) QW. When probing \( S_z \) for short negative delays \( \Delta t \), i.e., before the arrival of the corresponding pump pulse, we measure a remanent spin polarization profile which results from excitation by previous pump pulses. This observation implies an electron spin lifetime \( T_\text{z}^r \) of the same order as the \( t_\text{rep} = 27.5 \text{ ns} \) pulse-to-pulse interval of the supercontinuum source. This rather long spin lifetime allows for an investigation of the relaxation dynamics by the resonant spin amplification (RSA) technique [6].

In Fig. 5a, we exemplarily show a representative RSA trace which we measure at \( T_c = 8 \text{ K} \) for a negative delay \( \Delta t = -120 \text{ ns} \). We minimize the influence of diffusion on the measurement by strongly defocusing the pump and probe spots \( (d \geq 50 \mu\text{m}) \). This results in a time-averaged excitation density of \( \approx 0.25 \text{ W cm}^{-2} \).

We determine the in-plane and out-of-plane spin lifetimes \( T_\text{z}^s \) and \( T_\text{z}^r \) from a RSA model which considers the anisotropic spin relaxation in the (110) QW [55, 70]. From the fit shown in Fig. 5a, we obtain \( T_\text{z}^s = (29 \pm 2) \text{ ns} \) and \( T_\text{z}^r = (9.4 \pm 0.4) \text{ ns} \), i.e., we indeed observe the strongly anisotropic spin dephasing expected for (110) QWs [8, 53]. From the RSA fit we additionally obtain the effective electron g-factor as \( |g^z| = (0.395 \pm 0.004) \).

We now turn to the measurement of electron spin diffusion coefficients by time resolved MOKE microscopy. Because of the long spin lifetimes \( T_\text{z}^s \approx t_\text{rep} \) of the (110) QWs, the spin polarization profile \( S_z(\Delta t, r) \) results from a superposition of the contributions of several previous pump pulses. To facilitate the following quantitative analysis, we suppress these previous pulse contributions by applying a small external in-plane magnetic field \( B_\text{ix} \). The external field is chosen such that \( S_z(\Delta t < 0 \text{ ns}) \) vanishes because of destructive spin interference (compare Fig. 5a). This previous-pulse suppression is exemplarily shown in Fig. 5b where we compare spin polarization profiles measured at \( \Delta t = -20 \text{ ps} \) for \( B_\text{ix} = 2 \text{ mT} \) and zero external magnetic field. We additionally show a single-pulse profile measured for a short delay \( \Delta t = 50 \text{ ps} \) and a Gaussian fit from which we determine the width \( w_0 \) of the spin packet.

To demonstrate an application of picosecond Kerr microscopy we determine the spin diffusion coefficient \( D_s \) of the (110) QW as a function of lattice temperature and excitation power. We therefore measure time traces of single-pulse spin polarization profiles \( S_z(\Delta t, r) \) for increasing pump-probe delays \( \Delta t \) between -20 ps and 814 ps.

In Fig. 6a, we exemplarily show a series of line cuts of the electron spin polarization profile along the [001] crystal direction together with Gaussian fits which we measure for \( P_{\text{pump}} = 3 \mu\text{W} \) at \( T_c = 8 \text{ K} \). The diffusive spreading of the spin packet is clearly observed from the increase in width of the \( S_z(r) \) profiles for longer delays. Comparison with the fits confirms that the shape of the spin packet remains Gaussian for all examined times.

In Fig. 6b, we plot \( w_0^2(\Delta t) \) extracted from the Gaussian fits. For the shortest delay of 13 ps the squared width coincides with our optical resolution, i.e., \( \sigma_0^2 = 3.8 \mu\text{m}^2 \) as indicated by the dashed line. The linear increase of \( w_0^2 \) with
$\Delta t$ expected from Eq. (6) is clearly observed. From the slope of a fit to a straight line we determine the spin diffusion coefficient $D_s = (100 \pm 3) \text{cm}^2\text{s}^{-1}$.

Previous studies of spin propagation driven by surface acoustic waves (SAW) have indicated anisotropic spin transport along the [001] and [110] crystal directions in (110) quantum wells in the presence of small external magnetic fields [71]. We have verified that a similar dependence of the spin diffusion on crystal direction was not present in our measurements, i.e., we observe isotropic diffusion in the QW plane. We therefore limit our following presentation to results obtained for measurements along the [001] direction.

The dependence of the spin diffusion coefficient of the (110) QW sample on pump power and lattice temperature is summarized in Fig. 6c. For $T_L$ between 8 and 40 K we measure the spin diffusivity for very low excitation powers $P_{\text{pump}} = 1.5, 3,$ and $5 \mu$W. For all examined excitation densities we observe that $D_s$ only varies weakly with the lattice temperature. We find that a reduction of the pump power from 5 to 3 $\mu$W leads to a systematic decrease of the spin diffusivity. A qualitatively similar decrease of $D_s$ with excitation density has been observed previously for a (110) QW at room temperature [58]. At present, the physical origin of this pump power dependence is not known. However, a significant decrease of the diffusion coefficient when further lowering the pump power to 1.5 $\mu$W is not observed. This suggests that we have reduced pump-induced influences on $D_s$ sufficiently to observe the intrinsic temperature dependence of the spin diffusivity.

We focus on $D_s(T_L)$ obtained for the lowest pump power of 1.5 $\mu$W for which we are closest to measuring the intrinsic spin diffusion coefficient. In contrast to the case of bulk GaAs where the spin diffusivity strongly depends on $T_L$ [17, 19], we find a constant value $D_s \approx 100 \text{cm}^2\text{s}^{-1}$ for all examined lattice temperatures. A similar temperature-independent spin diffusion has been observed previously for a GaAs (110) MQW sample up to room temperature [56].

We use the Einstein relation $\mu_s = (e/k_B T_L) D_s$ to calculate the temperature dependence of the spin mobility. Following Ref. [56] we infer the dominant scattering mechanism which limits the spin diffusivity in the (110) QW sample from $\mu_s(T_L)$. For the observed constant $D_s$, which is insensitive to changes of the lattice temperature, we find $\mu_s \sim T_L^{-1}$ as demonstrated in the inset of Fig. 6c. This temperature dependence is characteristic for a non-degenerate two-dimensional electron system where the mobility is limited by scattering with acoustic phonons with a rate which increases proportional to the lattice temperature [56].

6 Conclusions and outlook We have summarized our recent results on the spatially resolved electron spin detection in semiconductor heterostructures by magneto-optical Kerr microscopy. We have focussed on the fundamental aspect of spatially resolved MOKE spectroscopy in semiconductors – the reliable determination of the local electron spin polarization from pump-probe Kerr microscopy. We have addressed two particularly important model systems for the magneto-optical spectroscopy of electron spins in semiconductors, bulk n-doped GaAs and GaAs-based (110) QWs.

For both systems, we have found that the spatially resolved electron spin detection at low lattice temperature is strongly disturbed by pump-induced modifications of the magneto-optical response which are not related to the spin polarization. Contrary to a common assumption, as a result of these disturbances a measurement of the local Kerr rotation at a fixed, arbitrary probe wavelength in general does not reveal the electron spin polarization correctly.

In bulk GaAs, we have identified that the local disturbance of the magneto-optical response originates from the thermal broadening of the excitonic Kerr resonance by pump-induced photocarrier heating. For the (110) QW sample the microscopic origin of the local linewidth broadening of the Kerr resonance has not been determined yet. However, the strong influence of free-carrier-collision broadening on the excitonic linewidth in QWs [38] suggests that in QWs the spatially varying photocarrier density significantly contributes to the local disturbance of the magneto-optical response.

We have presented two alternative approaches to the spatially resolved electron spin detection by MOKE microscopy. For the bulk GaAs sample we have demonstrated how the lateral spin polarization profile can be determined correctly from a quantitative analysis of a spatially resolved measurement of the full excitonic Kerr rotation spectrum. For the (110) QW sample we have introduced time resolved real space imaging of the diffusion of optically excited spin packets by picosecond Kerr microscopy with a broadband probe laser for the direct all-optical determination of electron spin diffusion coefficients.

Having established reliable ways of measuring electron spin transport in semiconductors with high temporal and spatial resolution we are now in the position to quantitatively address the influence of hot photocarriers on the lateral electron spin diffusion. For bulk GaAs we have recently demonstrated the strong influence of the electron temperature on the optically induced spin diffusion both in cw spectroscopy [17] and in the time domain [19].

Surprisingly, similar signatures of hot carrier effects on the electron spin diffusion – namely a transient decrease of the spin diffusivity for long delays – have been absent in our measurements on the (110) QWs. However, we here have observed that an increase in the pump power results in a modest enhancement of the spin diffusion coefficient. For the future we therefore intend to perform a detailed time resolved investigation of the influence of the electron excess energy and excitation density on the spin propagation to unambiguously determine whether we indeed characterize the intrinsic temperature dependence of the electron spin diffusion coefficient of the (110) QW in the limit of low power excitation.

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