Quasi-nondegenerate pump–probe magneto-optical experiment in GaAs/AlGaAs heterostructure based on spectral filtration

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Abstract
We report on a quasi-nondegenerate pump–probe technique that is based on spectral-filtration of femtosecond laser pulses by a pair of mutually-spectrally-disjunctive commercially available interference filters. The described technique enables to obtain pump and probe pulses with wavelengths that are spectrally close but distinct. These contradictory requirements, which are dictated, for example, by a suppression of stray pump photons from the probe beam in spin-sensitive magneto-optical experiments in non-magnetic semiconductors, can be fulfilled at very low cost and basically no requirement on space. Especially the second feature is important in pump–probe microscopy where collinear propagation of pump and probe pulses is dictated by utilization of a microscopic objective and where the setups are typically quite complex but suffer from a limited size of optical breadboards. Importantly, this spectral-filtration of 100 fs long laser pulses does not affect considerably the resulting time-resolution, which remains well below 500 fs. We demonstrate the practical applicability of this technique by performing spin-sensitive magneto-optical Kerr effect (MOKE) experiment in GaAs/AlGaAs heterostructure, where a high-mobility spin system is formed after optical injection of electrons at wavelengths close to the MOKE resonance. In particular, we studied the time- and spatial-evolutions of spin-related (MOKE) and charge-related (reflectivity) signals. We revealed that they evolve in a similar but not exactly the same way which we attributed to interplay of several electron many-body effects in GaAs.

1. Introduction
The pump–probe technique is well established stroboscopic optical method allowing to measure ultrafast dynamical response of various materials [1]. Here, a short intensive laser pulse (pump) excites the investigated system and its relaxation toward an equilibrium is measured by a weaker time-delayed laser pulse (probe) in a reflection or transmission geometry. If this technique is combined with an optical microscope, it is possible to achieve relatively high spatial- and time-resolutions simultaneously [2]. The spatial-resolution of such pump–probe microscope is determined mainly by a numerical aperture of the objective and by a wavelength of the laser light [3]. The time-resolution is primarily given by a duration of the laser pulses. In a traditional form of degenerate pump–probe experiment, pump and probe pulses are obtained by an intensity splitting of the output from a pulsed laser, i.e., their wavelengths are exactly equal. This variety of pump–probe experiment is probably the most frequently used one not only due to its simplicity (and the resulting low cost) but also due to the fact that the signal-to-noise ratio in the measured data is determined predominantly by a performance of the laser itself, which is usually rather good. The most frequently employed detection scheme involves a chopper-induced intensity modulation of a pump beam and monitoring the pump-induced changes of a probe beam by a lock-in technique [4]. However, to achieve a good signal-to-noise ratio in this experiment, it is necessary to separate the signal due to probe photons, where the required information is encoded, from the signal due to (scattered) pump photons,
which acts as a noise. In principle, this separation could be done electronically if both pump and probe beams are modulated at two distinct frequencies and a signal at the sum (or difference) frequency is measured by the lock-in amplifier. Nevertheless, much better signal-to-noise ratio is usually obtained if pump photons are prevented from reaching the photodetector at all. If pump and probe pulses have different angles of incidence (see figure 1(a)), this can be achieved quite straightforwardly by spatially filtering the probe beam after the sample. The situation is much more complicated if pump and probe beams are collinear, which is usually the case of experiments with a high spatial resolution where microscopic objective lenses have to be used (see figure 1(b)). In this case, one can separate the pump and probe photons by setting their polarization to mutually-orthogonal linear polarizations and placing a polarizer in front of the detector. However, this option is not compatible with magneto-optical (MO) experiments where the measured signal is encoded in the probe polarization rotation [3]. Similarly, it cannot be applied in a time-resolved research of molecular dynamics where a separation of pure population dynamics from molecular structural changes and rotational diffusion is achieved by setting the ‘magic angle’ between the polarization planes of pump and probe photons [5, 6]. Therefore, the most efficient solution is to use pump and probe pulses with a different wavelength and to remove scattered pump photons from the probe beam by placing a spectral filter, which is transparent only for probe photons, in front of the photodetector. Moreover, if excitation and/or detection channels are strongly wavelength dependent, which is the case for MO effects [3], the independent spectral tunability of pump and probe pulses is very favorable also for the experimental setup performance optimization.

There are several well-established methods how to obtain different wavelengths of pump and probe pulses for nondegenerate pump–probe experiment. In principle, this can be achieved by an electronic synchronization of the pulse trains emitted from two independent laser systems [7, 8]. However, due to a presence of the timing jitter in the lasers, the achievable time-resolution in this case is limited to several picoseconds only, unless a rather complicated detection scheme is used [9]. When advanced digital electronics is used, a high-speed asynchronous optical sampling with sub-50 fs time resolution could be achieved [10]. As an easier (and cheaper) alternative how to reach a femtosecond resolution, it is to start with a single femtosecond laser which output is split into two (or more) parts. Using this approach, the simplest way how to change the wavelength of one of the beams is to use a second harmonic generation (SHG) in a nonlinear crystal. This method serves its purpose very well but only when the very different wavelengths of pump and probe pulses (e.g., in a case of Ti:sapphire laser, around 400 and 800 nm, respectively) and/or the inability to tune independently the wavelengths of the beams are not an issue. Considerably more versatile solution, which enables to achieve a spectral tunability in a very broad range, is to use one of the beams to pump an optical parametric oscillator (OPO) or optical parametric amplifier (OPA) [2, 11]. The disadvantage of this solution is a high cost and a large footprint of OPO/OPA. To avoid this, very efficient light color conversion can be obtained also in a photonic crystal fiber [12]. However, as several distinct nonlinear effects take place simultaneously in photonic fibers, the achievable time-resolution in this case is limited to several picoseconds [12]. Moreover, because nonlinear optical effects are used in a photonic crystal fiber, OPO and OPA, in all these cases the resulting signal-to-noise ratio is usually considerably worse than the one obtained when the fundamental frequency output from the laser is used in the degenerate pump–probe experiment.

![Figure 1.](image-url)
Another convenient option to obtain synchronized pump and probe pulses at different wavelengths is to select two distinct spectral regions within a spectrum of the femtosecond laser pulse. In reality, this seemingly straightforward solution is considerably complicated by the fact that spectral- and temporal-profiles of light pulses are mutually interconnected through the Fourier transform [13]. Selecting a rectangular spectral-profile from a laser pulse (by a monochromator, for example) leads to a sinc² time-profile and, consequently, to a very poor time-resolution. To avoid this, more complicated approaches based on a grating-based pulse shaper [14, 15] or a prism-pair [2] were used. But even here the experimentally demonstrated time-resolutions were in the picosecond time scale only [14, 15]. In this paper we show that sub-picosecond time resolution can be achieved in a very simple quasi-nondegenerate pump–probe experiment where an output of a femtosecond oscillator is filtered by a pair of well-chosen mutually-spectrally-disjunctive interference filters. We also demonstrate that this cost- and space-efficient solution is very useful not only in the case of pump–probe microscopy, as we have already reported recently [16], but also in a standard time resolved spin-sensitive experiments in semiconductors.

The paper is organized as follows: first, we describe briefly experimental setups where this experimental technique was tested. Then we study spectral- and temporal behavior of the filtered femtosecond laser pulses. Finally, we demonstrate the utilization of this technique for a measurement of electron spin dynamics at the heterointerface GaAs/AlGaAs.

2. Experimental setup

As a light source we used a computer-controlled femtosecond Ti:sapphire oscillator (Mai Tai, Spectra Physics) with a repetition rate of 80 MHz that generates ≈100 fs laser pulses with a spectral width of about 8 nm (full width at half maximum, FWHM). Each laser pulse is split to two parts with a typical intensity ratio between pump and probe pulses equal to 10:1. The intensity of the pump beam is modulated by a chopper at frequency of ≈2 kHz. A time delay between pump and probe pulses, Δt, is set by a computer-controlled delay line. The sample is mounted in an optical cryostat, where temperature was set to 15 K, and placed between poles of an electromagnet providing in-plane magnetic field up to 500 mT. Light reflected from the sample is led to a detection part of the setup. Here the MO signal corresponding to the probe polarization rotation and the differential reflectivity are measured as difference and sum of signals from detectors in the optical bridge, respectively [17]. Two possible implementations of the pump–probe setup are schematically depicted in figure 1. In figure 1(a) we show the version with a non-collinear incidence of pump and probe beams—the angles of incidence to the sample surface are <1° and ≈7° for the pump and probe beams, respectively. Both beams are focused by a single converging 10D lens on the sample to overlapped spots with a size of ≈25 μm (FWHM). The major advantage of this configuration is that it enables, even without any spectral-filtering described in this paper, to filter out spatially the scattered pump photons from the probe beam, which leads to a very good signal-to-noise ratio in the measured data [18–21]. If higher spatial-resolution is needed, a microscopic objective lens has to be used instead of the focusing lens. However, the correct functionality of the objective lens requires pump and probe beams to be collinear. This collinearity of the beams requires the pump and probe beams to be spatially overlapped before the objective lens and the reflected probe beam to be separated from the incident beams by a non-polarizing beam splitter cubes (BS)—see figure 1(b). The insertion of a BS can, however, modify the polarization state of the laser beams due to a different transmission (reflection) amplitude and phase of the BS for light polarized in the plane of incidence (p-polarization) and perpendicular to this plane (s-polarization). If horizontal and vertical linear polarization of (pump and probe) beams are used solely, the straightforward solution of this problem is a precise (angular) alignment of the BS in such a way that the incident horizontal and vertical polarizations coincide with the p- and s-polarizations, respectively, which are not mixed in refraction and/or reflection at BS. If circular polarizations are needed, it is necessary to select such a BS that has very similar transmission (reflection) amplitude and phase for p- and s-polarizations in the utilized spectral range. In our particular case of magneto-optical Kerr effect (MOKE) experiment, we employed a circularly polarized pump beam and linearly polarized probe beam. Consequently, we had to secure the preservation of the circular polarization during the transmission through two BSs for the pump beam only (see figure 1(b)). We verified experimentally that the selected BS (BS014 from Thorlabs) did not degrade the quality of circular polarization in the spectral range relevant for the studied material system (see figure 4) considerably: Without BSs, the pump circular polarization contained 97% of photons with the required handedness. With the two BSs inserted, this fraction decreased to 92%, which is still sufficient for the experiments. In fact, it is also possible to use BSs that modify the polarization of the beams. In this case, to obtain a circular polarization on the sample, it is necessary to pre-compensate the BS-induced polarization changes by the waveplates located in the beams before the BSs.
Figure 2. Examples of spectrally-disjunctive laser pulses (solid lines) obtained by filtering femtosecond laser pulses from the oscillator (filled curves) with central wavelengths of (a) 774 nm, (b) 791 nm, (c) 798 nm, (d) 820 nm, (e) 824 nm, and (f) 900 nm. The dashed lines show transmission spectra of the corresponding interference filters.

Similarly, the eventual polarization distortion of the linearly polarized probe beam after the reflection from the sample can be compensated by the waveplates located in the detection part of the setup. However, as the polarization response of BSs can be strongly wavelength dependent, this polarization change compensation has to be verified at all used wavelengths individually.

Another problem, which is induced by the collinearity of the pump and probe beams, is that it is not possible to separate them by a spatial-filtration. This in turn leads to a very bad signal-to-noise ratio unless other means of separating pump and probe photons are implemented. As we show below, a very compact and cost-efficient solution of this problem, which does not affect significantly the achievable time-resolution, is to use pump and probe beams prepared from the output of a femtosecond oscillator by a spectral-filtering using a pair of mutually-spectrally-disjunctive interference filters. The resulting quasi-nondegenerate experiment can be used very efficiently in experiments with a high spatial-resolution; see reference [16] where a near-infrared objective with magnification $20 \times$ and numerical aperture 0.4 was used to create a spot size $<2 \mu m$. In addition, as we demonstrate below, this rather small variation of the experimental setup can be used to enhance considerably the measured time resolved MO signal in a case of materials with spectrally narrow MO spectrum, or even to measure this MO spectrum in a pump–probe experiment.

3. Spectral filtration of femtosecond laser pulses

In our experiment, we used a rather typical commercial femtosecond Ti:sapphire oscillator which produces $\approx 100$ fs laser pulses with a central wavelength tunable in the infrared spectral region. The major problem connected with a spectral filtration of these femtosecond pulses is their relatively narrow spectrum with a width of about 8 nm (FWHM). To prepare a pair of mutually-spectrally-disjunctive laser pulses, it is necessary to use pairs of spectral filters with a sufficiently separated transmission edges (see figure 2). Moreover, as already discussed in the introduction, the filter transmission edges could not be too steep, otherwise the mutual interconnection between the spectral- and temporal-profiles of light pulses would lead to a considerable prolongation of the filtered laser pulse in a time-domain. Simultaneously with this, the transmission edges have to be rather steep and located relatively close to each other (i.e., within the spectrum of the oscillator output) otherwise the intensity of transmitted laser pulses would not be sufficient.
Table 1. Summary of properties of original and filtered femtosecond laser pulses. For each oscillator output, which is labeled by a corresponding central wavelength ($\lambda_{\text{central}}$), the properties of laser pulses after passing through a given interference filter are shown: spectral position of maximal intensity ($\lambda_{\text{max}}$), transmitted fraction of the laser pulse ($I/I_0$), temporal length of correlation trace ($\Delta t_{\text{FWHM}}$) measured by SHG. The filters are labeled by the notation of Thorlabs: Bandpass filters are denoted as 'FB', notch filters as 'NF', long pass filters as 'FEL', and short pass filters as 'FES'; the letter 'H' depicts that the filter is a premium version of a filter with improved properties (higher transmission, steeper cut-on and cut-off slopes). The number behind the letters describes a spectral position of the filter and the second number for bandpass filters stands for their spectral width (in nanometers).

| $\lambda_{\text{central}}$ (nm) | Filter   | $\lambda_{\text{max}}$ (nm) | $I/I_0$ (%) | $\Delta t_{\text{FWHM}}$ (fs) |
|-----------------------------|----------|-----------------------------|-------------|-----------------|
| 774 nm                      | —        | —                           | —           | 126             |
|                            | FBH780-10| 777                         | 24.6        | 204             |
|                            | NF785    | 772                         | 25.3        | 220             |
| 791 nm                      | —        | —                           | —           | 124             |
|                            | FBH780-10| 786                         | 5.9         | 206             |
|                            | FBH800-10| 795                         | 19.8        | 240             |
| 798 nm                      | —        | —                           | —           | 114             |
|                            | NF808    | 794                         | 8.8         | 230             |
|                            | FBH810-10| 805                         | 9.7         | 215             |
| 820 nm                      | —        | —                           | —           | 145             |
|                            | FBH810-10| 815                         | 7.3         | 193             |
|                            | NF808    | 826                         | 31.2        | 390             |
| 824 nm                      | —        | —                           | —           | 121             |
|                            | FBH800-40| 818                         | 6.4         | 180             |
|                            | FBH850-40| 832                         | 20.9        | 172             |
| 900 nm                      | —        | —                           | —           | 189             |
|                            | FELH0900 | 903                         | 46.5        | 272             |
|                            | FESH0900 | 896                         | 19.5        | 241             |

for the pump–probe experiment. Several combinations of commercially available interference filters (bandpass, notch, long pass, and short pass) sold by Thorlabs, which are matching these mutually antagonistic criteria, are listed in table 1 and their spectral dependences of transmittances are shown in figure 2 as dashed lines. The spectra of transmitted laser pulses are dependent also on the spectral profile of pulses that are incident on the filters. The filled curves in figure 2 depict the spectral profiles of the unfiltered oscillator outputs at the sample position for several selected central wavelengths. Apparently, at certain wavelengths, the propagation in air (for about 5 m) and/or properties of optical components in the beam path modify partially the pulse spectrum from the Gaussian shape of transform-limited laser pulses, which are leaving the oscillator cavity. The solid lines in figure 2 represent the measured spectra after insertion of the corresponding filters to positions depicted in figure 1(a). Note that for a given filter pair, the relative intensity of the transmitted pulses in the pump and probe arms of the setup, respectively, can be fine-tuned by slightly changing the central wavelength of the oscillator output.

We stress that the ability to create a pair of spectrally-disjunctive laser pulses by spectrally filtering an incident femtosecond laser pulse is not a guarantee of their applicability in the pump–probe experiment. The interference filters are rather complicated layered structures that can, in principle, distort severely the time profile of the laser pulses. To address this issue, we measured the intensity auto- and cross-correlations [13] of the pulses as described below. We used the setup shown in figure 1(a) where we replaced the sample by a beta barium borate (BBO) nonlinear crystal. We detected (in transmission geometry) light produced by SHG when pump and probe pulses, which were set to have the same intensity, were present in the BBO crystal simultaneously. The filled curves in figure 3 correspond to the correlation traces of the unfiltered oscillator outputs for several selected central wavelengths. As expected, these curves are symmetrical with respect to a zero-time delay, which is a signature that properties of pump and probe pulses in our experimental setup are rather similar—i.e., these curves can be regarded as auto-correlation traces for the oscillator outputs. The three-peak structure observed for the central wavelength of 900 nm indicates that at this wavelength there is a weaker satellite with a time-spacing of about 0.5 ps relative to a position of the major laser pulse. The widths (FWHM) of the measured auto-correlation traces, which correspond to the actual time-resolution in our pump–probe experiment, are listed in table 1. The precise shape of the laser pulses envelope is not known in our case. If we assume $\text{sech}^2$-shaped pulses for simplicity, the pulse duration is $\approx 0.65$ times the width of the measured autocorrelation signal, which corresponds to 80–120 fs for the wavelengths studied. To study the properties of the filtered laser pulses in a time domain, we have measured their auto-correlation traces and also their cross-correlation traces with the original (unfiltered)
Figure 3. Correlation traces measured by non-collinear SHG in BBO crystal for unfiltered (filled curves) and filtered (solid lines) laser pulses.

laser pulses. In figure 3 we are showing the cross-correlation traces that were measured when the selected filter was inserted in one of the beams only. The advantage of the cross-correlation traces is that they (unlike the auto-correlation traces) could be asymmetrical in time and, therefore, enable to distinguish between the leading and trailing edge broadening. For certain filters, the obtained correlation trace remained nearly symmetrical with respect to a zero-time delay, which shows that the time-profile of the filtered laser pulse was not modified significantly by the filter (see figure 3(e)). However, for a majority of the filters the measured correlation trace is clearly asymmetrical in time, which can be interpreted as a broadening of the trailing edge of the laser pulse in the interference filter. Nevertheless, for all used filters the achievable time resolution in the corresponding pump probe experiment is well below 500 fs (see table 1).

Last but not least, we would like to mention one additional advantage of this filter-based method. Unlike the case of OPO and OPA, the spectral position of the obtained laser pulses does not fluctuate in time because the position of its spectrally-sharp edge is set by a position of the filter transmission edge (see figure 2). This is very important in all experiments when ‘seemingly small’ thermal spectral fluctuations of laser pulses (e.g., for about 1 nm) could lead to a large change of detected signals. As an example, we can mention MO experiments in the field of semiconductor spintronic [22–25]. Here, imaging of the lateral spin transport is based on MOKE the spectrum of which is rather narrow and has a dispersive character, i.e., the corresponding MO coefficient changes a sign around the semiconductor bandgap energy (see inset in figure 1 in reference [24] or figure 4(e)).

4. Quasi-nondegenerate pump–probe experiment in GaAs/AlGaAs heterostructure

4.1. Description of studied material system
As an example, in this chapter we demonstrate the practical utilization of our quasi-nondegenerate pump–probe scheme for an investigation of electron spin dynamics in a material system where a spectrally close excitation and probing is necessary. We performed a time-resolved spin-sensitive MO experiment in a non-magnetic semiconductor GaAs where the spectrum of MOKE is rather narrow (with an spectral width
Figure 4. Utilization of quasi-nondegenerate pump–probe experiment for an investigation of the dynamics of spin-polarized electrons in GaAs/AlGaAs heterostructure. (a) Sketch of the layer structure of the studied sample (the sample surface, on which the light is incident, is on the left side) together with the energetic diagram of the conduction and valence bands, showing the band bending before (solid curves) and after the illumination (dotted curves). The squares depict the surface and bulk localized charged states, the dashed line the Fermi energy $E_F$. Circles stand for photoinjected spin-polarized electrons and holes that are photoinjected by circularly-polarized pump pulses, the arrows indicate their drift motion. The studied long-lived and highly mobile spin-polarized electron spin system is self-consistently formed near the upper GaAs/AlGaAs interface where it was detected by probe pulses using MOKE signal. (b) External magnetic field $H$ applied in a direction perpendicular to the electron spin orientation induces the spin precession, which is detected as an oscillatory MOKE signal that was measured using a spatial-filtration by laser pulses with central wavelength of 820 nm (red points) and the same pulses that were spectrally-filtered (FB820-10 and FB830-10 in probe beam and FB810-10 in pump beam) (green points); $\mu_0H = 500$ mT, sample temperature 10 K, fluence of pump pulses $8 \mu$J cm$^{-2}$. The solid lines are fits by a damped harmonic function. (c) Comparison of the relative noise in data shown in part (b). (d) Demonstration of MO signal phase change measured in a quasi-nondegenerate pump–probe experiment due to probe pulses wavelength change. The vertical line depicts the time delay of 470 ps where the polarization rotations displayed in part (e) were recorded. (e) Spectral shape of MOKE in the studied electron sub-system that was derived from the measured time resolved MO signals in the quasi-nondegenerate experiments. The position and horizontal bar width for each data point correspond to the maximum and spectral width of the spectrally-filtered probe pulses, respectively. The filled curves depict the original and spectrally-filtered spectrum of laser pulses that were used to measure the data shown in part (b).

of several tens of meV) and located around the semiconductor bandgap energy $E_g$ (see figure 4(e)), which restricts the spectral position of probe pulses. Pump pulses are used to photoinject spin-polarized electrons by the optical orientation effect [26], which limits the photon energy $h\omega$ of pump pulses because this phenomenon occurs only when $E_g < h\omega < E_g + \Delta SO$, where $\Delta SO$ is the energy of the spin–orbit splitting that is $\approx 340$ meV in GaAs (see the one-photon excitation case in figure 2 in reference [25]). In general, even more stringent restrictions on pump and probe wavelengths can appear when the studied structure has a more complex excitation spectrum, for instance due to pronounced excitonic or impurity bands (such as in transition metal dichalcogenides) or due to a spatial confinement of charge carriers (e.g., in quantum wells or quantum dots). Overall, the requirement of a mutual spectral proximity of pump pulses, probe pulses and band gap energy is a common feature for all spin-sensitive pump–probe experiments in any non-magnetic semiconductor.

As a test sample, we choose a self-confined electron system that was recently reported to be formed in the vicinity of an undoped GaAs/AlGaAs heterointerface [16, 27], which is depicted in figure 4(a). A 100 nm-thick undoped $Al_{0.4}Ga_{0.6}As$ barrier was deposited by molecular beam epitaxy on top of an insulating GaAs buffer and a GaAs substrate. The barrier was then covered by another undoped 800
When the wavelength of probe pulses is changed, not only the precession signal amplitude but also its phase can change—see figure 4(d). This strong sensitivity of the amplitude and phase of the measured dynamical MO signal on the relative spectral positions of probe laser pulses and MOKE spectrum can be used for a crude reconstruction of the MOKE spectrum in the studied sample from the measured time resolved data, as demonstrated in figure 4(e). This procedure is extremely useful in the case of the studied material system where the observed long-lived electronic spin sub-system is present only in a very thin layer.
of GaAs close to the GaAs/AlGaAs interface (see figure 4(a)). The standard (time-unresolved) experimental techniques for measuring the MOKE spectrum [28] would provide information only about the depth-averaged MO properties of the sample, which would be dominated by the properties of undoped GaAs in our case. On the contrary, the MOKE spectrum shown in figure 4(e) corresponds solely to the studied photo-injected electrons with the very long spin lifetime. The absolute spectral position of the MOKE spectrum is strongly dependent on the stress-induced band edge shifts in GaAs [24] and, therefore, its comparison is rather difficult between different experiments. The most apparent distinction between the MOKE spectrum in our material system, when electrons are injected to GaAs optically, and the n-doped GaAs epilayer [24] is the spectral width—the MO coefficient is non-zero only within the material system (see figure 2(d) in reference [24]) while in our case the spectrum is considerably broader (see figure 4(e)).

4.3. Exemplary experimental data obtained using the collinear setup

For the final test experiment, we used the experimental setup with a collinear incidence of pump and probe beams. The simultaneous high spatial and time resolutions of our technique allows us to study exotic spin-transport phenomena in the high-mobility regime of an optically excited GaAs/AlGaAs interface. An example of a contra-intuitive behavior of spin diffusion is shown in figure 5 where the relative positioning of pump and probe beams on the sample surface was controlled by a movable mirror (see figure 1(b)). As pointed out in reference [29], the interpretation of MO experiments in terms of spin dynamics is truly meaningful only past the temporal overlap between pump and probe pulses and/or after the characteristic electronic dephasing time in the studied material. Therefore, the utilization of ‘as short as possible’ laser pulses is essential for investigation of the initial stages dynamics of photo-injected carriers and spins. The measured spatially-resolved traces of Kerr rotation and transient reflectivity for several selected time-delays $\Delta t$ are shown in the left and right panels of figure 5(a), respectively. We note that it is necessary to measure both these quantities for disentangling the dynamics of spin and charge [30]. We observed that a few picoseconds after the photo-excitation of the sample by a pump pulse with a Gaussian spatial profile both the MOKE and transient reflectivity signals show the expected Gaussian-like distribution of spins and carriers (see the traces for $\Delta t = 5$ ps in figure 5(a)). However, after another 10 ps, the profiles of both signals change dramatically and considerable modulation and even a sign change occur in the measured traces. We also note that the dynamics of spin (MOKE) and charge (transient reflectivity) is apparently rather different—see, e.g., the traces for $\Delta t = 95$ ps in figure 5(a).

The observed spatial evolution of the (spin-sensitive) MOKE signal for time delays <100 ps shares rather prominent similarities with the spatially-resolved time-integrated photoluminescence (PL) measured under comparable conditions in p-doped GaAs [31–33]. Similar to our data, there was a dip in the PL polarization profile observed at the center of the photo-excitation spot if degeneracy of the electron gas was achieved (i.e., for a high concentration of electrons at low temperature). The authors attribute it to the Pauli blockade and the spin-degeneracy pressure if the photoexcited carrier density surpasses the steady-state density. In this high-degeneracy regime, the scattering probability is reduced due to the higher occupation of end-states of the scattering process. When the photocarriers are spin-polarized, it leads to different mobility and diffusion constants and, thus, different diffusion profile for minority and majority spin populations which forms a characteristic dip at the center of excitation. The interpretation of the observed oscillatory behavior in the measured traces as a consequence of electron many-body effects in GaAs [31–34] is corroborated by the fact that these effects are apparent only at high excitation intensities (see figure 5(b)). As our interfacial system features much longer spin life-time (10’s of ns comparing to ≈200 ps in reference [31]), we can observe the evolution of the system up to nanosecond time scale where a complete inversion of the MOKE signal is found (see left panel in figure 5(a)). However, this interpretation does not explain the evolution of the transient reflectivity signal, which is shown in right panel in figure 5(a), where we observed similar trends but time-shifted (accelerated) with respect to the MOKE signal. The reflectivity signal is connected with a carrier-induced change of complex index of refraction, which is sensitive to the density of photo-injected charge-carriers and their energetical position within the semiconductor band structure. Within the Pauli blockade picture, the difference in the diffusion profiles for both spin populations should lead to a simple Gaussian profile in the charge-carrier distribution, which is clearly not the case in our sample. The precise identification of the exact underlying effect(s) in the reflectivity signal is well beyond the scope of this article. We only note that non-Gaussian ring-like features were previously reported for GaAs-based QWs [35–38] and transition metal dichalcogenides [39, 40] where they were interpreted as a consequence of indirect excitons, Coulomb interaction and/or Auger recombination. In the context of this paper, we just would like to mention that it is the very high quality of the achieved experimental data (i.e., their low noise) that opens the possibility to study all these many-body effects experimentally by pump–probe microscopy in future.
Figure 5. Utilization of quasi-nondegenerate pump–probe experiment for an investigation of the time- and spatially-resolved spin and charge diffusion in GaAs/AlGaAs heterostructure using the experimental setup with a collinear incidence of pump and probe beams. (a) Kerr rotation (left panel) and differential reflectivity signals (right panel) as functions of pump–probe spot distance for various pump–probe time delays (central labels) for circularly polarized pump pulses with fluence $305 \, \mu J \, cm^{-2}$. (b) Same signals measured at time delay 45 ps for various pump powers. Filters NF808 and FBH810-10 (see figure 2(d)) were used in pump and probe beams, respectively. Measured curves are vertically shifted for clarity.

5. Conclusions

We demonstrated that it is very simple to transform a standard degenerate pump–probe MO experiment with $\approx 100$ fs laser pulses into its quasi-nondegenerate form using spectral-filtration of the laser pulses by appropriately-selected interference filters. The major advantage of this approach is that it is cost- and space-efficient and, simultaneously, it does not affect considerably the resulting time-resolution, which remains well below 500 fs. The utilization of this approach leads to the improved signal to noise ratio in the pump–probe experiment and it enables to use an arbitrary polarization of pump and probe pulses, because pump and probe photons do not have to be separated by polarization means. The full potential of this technique lies in pump–probe microscopy where collinear propagation of pump and probe pulses is dictated by the utilization of a microscopic objective and where, consequently, the possibility to remove stray pump photons from the probe beam by a spectral-filtration is essential for achieving a good
signal-to-noise ratio. To demonstrate the capability of our experimental technique for pump–probe microscopy, we studied the spin and charge dynamics in the highly mobile self-confined electron subsystem formed at the undoped GaAs/AlGaAs heterointerface. We observed a rather contra-intuitive behavior of the spin and carrier diffusion which can be attributed to interplay of many-body effects in GaAs.

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