Orientation of electron spins in hybrid ferromagnet–semiconductor nanostructures

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The spin orientation of electrons is studied in ferromagnet (FM)–semiconductor (SC) hybrid structures composed of a (Ga,Mn)As ferromagnetic layer, which is placed in the direct vicinity of a non-magnetic SC (In,Ga)As quantum well (QW). It is shown that the polarization of carriers in the SC QW is achieved by spin-dependent tunnelling into the magnetized ferromagnetic layer. This leads to dynamical spin polarization of the electrons, which can be directly observed by means of time-resolved photoluminescence. We find that the electron spin polarization grows in time after excitation with an optical pulse and may reach values as large as 30%. The rate of spin-dependent capture grows exponentially steeply with decreasing thickness of the spacer between ferromagnetic layer and QW, and it persists up to the Curie temperature of the (Ga,Mn)As layer. From time-resolved pump–probe Kerr rotation data, we evaluate a value of only a few μeV for the energy splitting between the electron Zeeman sublevels due to interaction with the ferromagnetic (Ga,Mn)As layer, indicating that the equilibrium spin polarization is negligible.

1 Introduction The first application of spintronics became possible by the discovery of the giant magnetoresistance effect by the groups of Fert and Grünberg in 1988, where the resistance of a metallic structure consisting of a three-layer, ferromagnet (FM)/paramagnet/FM sequence depends on the mutual orientation of the magnetic is of the adjacent FMs [1,2]. Readout heads in modern hard drives are operated on the basis of this effect [3]. Nowadays large efforts are directed towards implementation of magnetism into the semiconductor (SC) architecture of modern computers. The most self-evident approach in this respect is to construct a universal object combining
ferromagnetic and semiconducting properties. Following the pioneering research from the 1960s on Eu chalcogenides and Cd–Cr spinels [4], recent active studies have focused on III–V ferromagnetic SCs [5, 6]. Nevertheless, FM SCs such as (Ga,Mn)As and (In,Mn)As exhibit poor mobility for charge-carrier transport and rather low Curie temperature. Therefore, search for room-temperature FM SCs using different compounds such as Cr-doped II–VI diluted magnetic SCs deserves great interest [7, 8].

There is an alternative strategy which combines two types of material, i.e. a FM and a SC. In such FM–SC hybrid structures, each component retains its basic properties without deteriorating the properties of the other component: high charge carrier mobility in the SC, and magnetic properties in the FM [9]. Selection of an optimal FM–SC combination out of a large variety of materials is the key challenge here. One of the most commonly used devices are direct spin injection structures [10, 11], where spin-oriented carriers are injected from the FM, having still an active role, into the SC [12]. Spin polarization could be obtained in that way with an efficiency of up to 50% [13, 14], but challenges remain such as the quite high electrical resistance mismatch, low Curie temperatures, and the control of the FM’s magnetization.

Besides readout of FM state, an essential point for integration of the FM–SC hybrid structures into the electronic devices is the control of FM magnetization by external stimuli. Electrical control of the FM by means of the SC is very appealing. This would allow reading and writing of the information in the FM without use of magnetic heads. Towards this goal, one might use spin transfer torque control by a high-density electric current [15–18], a combination of magnetic order and ferroelectricity [19, 20], or application of the magnetoelectric effect [21]. Korenev [22] proposed a structure comprising a FM near a SC quantum well (QW) with a two-dimensional hole gas, which is schematically shown in Fig. 1a. The p–d exchange interaction of the magnetic atoms in the FM layer with the QW holes induces an equilibrium spin polarization of the holes, which, in turn, influences the orientation of the magnetization \( \mathbf{M} \). The magnetization direction can be altered by applying a bias to the gate through changing the hole concentration or the overlap of the hole wave function and the magnetic layer, i.e. the value of the exchange constant.

The unified FM–SC spin system is highly flexible, as it is possible to control the magnetic anisotropy both optically and electrically. Optical tools for reading and writing of the FM state would allow ultra-fast operation on a sub-ps time scale. Although conventional optical techniques are limited in spatial resolution by the diffraction limit (in the order of hundreds of nm), recent developments in near-field optics by means of plasmonic antennas allow one to overcome this obstacle [23, 24]. In addition, optical control of magnetization by means of the SC has a huge potential. First, SCs possess excellent optical properties where the spin polarization of photoexcited carriers can be addressed optically by means of optical orientation [25]. This in its turn can influence the magnetization of the FM [26]. Second, there is a large variety of powerful optical phenomena for measuring the magnetization in the FM as well as spin orientation of carriers in the SC. It includes polarized photoluminescence (PL), magneto-optical Faraday/Kerr rotation, and their combination with time-resolved techniques, which can also be efficiently used for spin control [25, 27, 28]. Currently, there is a large amount of experimental and theoretical work that has been done in this direction. The main results regarding FM–SC hybrids include (i) read out of magnetization of the FM by means of spin-polarized carriers in the SC [29–33] and (ii) optical control of the coercive field of the FM and its magnetization direction [21, 34–37].

Recently, we have shown that in parallel to well-established s(p)–d exchange interaction between the
electrons (heavy holes) in the SC QW and the magnetic ions in the FM there is an alternative mechanism involving spin-dependent tunnelling of electrons from the SC into the FM [38]. This leads to fundamentally different dynamical spin polarization of the electrons in contrast to equilibrium polarization of the electrons (holes) in the case of s(p)-d exchange interaction. The schematic illustration of these two mechanisms is given in Fig. 1b for equilibrium spin polarization of carriers and in Fig. 1c for dynamical spin-dependent capture (SDC) of electrons. Optical excitation with linearly polarized light creates unpolarized photoexcited electrons and holes. If the FM layer is magnetized along the direction perpendicular to the sample plane (z-axis), the resulting PL is circularly polarized in both cases. However, in Fig. 1b, it is due to recombination of spin-polarized electrons and heavy holes, which at low temperatures populate the lower conduction band and the upper valence band states, respectively. The electron and hole subbands are split due to s–d and p–d exchange interaction with the FM layer. In Fig. 1c SDC of electrons in the FM layer leads to accumulation of the electron spins and subsequent recombination with unpolarized holes. This process is dynamical and most efficient if the SDC rate exceeds the radiative recombination of the electrons in holes in the QW. Note that the holes may tunnel into the FM layer as well. However, their spin relaxation is typically much faster and consequently the resulting spin polarization is weak.

In this article we present an overview of FM-induced orientation of electron spins in FM–SC (In,Ga,Mn)As-based structures. We discuss the main mechanisms responsible for spin orientation of the electrons in the QW. Time-resolved optical techniques based on polarization-resolved PL and magneto-optical Kerr rotation allow us to uncover the dynamical properties of the FM–SC hybrid structures and to evaluate the strength of several contributions. The experimental data clearly show that SDC is the dominating mechanism in the studied structures.

2 FM-induced spin polarization in quantum well

The experiments were carried out on GaAs-based structures, whose design is schematically shown in Fig. 2a. The Mn δ layer is separated from the 10 nm wide (In,Ga)As QW with about 10% of In content by a 5 or 10 nm thick spacer. In the following, we will label the samples according to this spacer thickness d_s, ‘5 nm’ and ‘10 nm’, respectively. Due to diffusion of Mn ions, the δ layer is transformed into an ≈1 nm thick ferromagnetic layer of Ga_{1-x}Mn_xAs with a Mn content x ≈ 5% [39]. The Mn ions supply holes with a density ≈ 10^{13} to 10^{14} cm^{-2}, mediating the ferromagnetism of the (Ga,Mn)As layer with a Curie temperature T_c ≈ 35 K. A relatively small fraction of the holes with concentration ≈ 10^{11} cm^{-2} may populate the QW. A detailed description of the samples is given in Ref. [40].

The magnetic properties of the (Ga,Mn)As layer were characterized by means of the single-beam magneto-optical Kerr effect (MOKE). The magnetization curve was measured in an external magnetic field B applied along the growth z-axis of the sample [001] and parallel to the incident beam (Faraday geometry). The rotation angle of the polarization plane of the reflected beam is proportional to the magnetization z-axis component M_z. The signal is homodyne detected using a polarization bridge in combination with a balanced photodiode and a lock-in amplifier. The typical magnetization curve at the temperature T = 2 K for the ‘10 nm’ sample is presented in Fig. 2b. It shows a hysteresis loop with coercive field B_c ≈ 10–20 mT. The form of the magnetization curve and the value of B_c are independent of the photon energy of the incident beam in the wide range from 1.4 to 1.8 eV. This indicates that the MOKE signal originates from the FM layer rather than the QW.

Figure 3a shows the PL spectrum from the QW centred at around 1.417 eV. This line corresponds to recombination of electrons with heavy holes from the lowest QW subbands. Here, it is essential that there is no optical orientation of photoexcited carriers under linearly polarized excitation. The spin polarization of the carriers in the QW is probed by the degree of circular polarization \rho_+ = (I_+ - I_-)/(I_+ + I_-), where I_+ and I_- are the \sigma^+– and \sigma^-–polarized intensities, respectively. In Fig. 3a, the points show the spectral dependence of \rho_+ in an external magnetic field of 100 mT in Faraday geometry. It follows that the PL line is polarized with \rho_+ ≈ 4%. Note that in the structures without Mn in such weak magnetic fields the PL polarization is absent. Figure 3b shows the polarization hysteresis loop \rho_+(B), measured at the PL maximum under linearly polarized excitation. The coercive force is determined to be B_c ≈ 10 mT, and the polarization loop closes approximately at the magnetic field of 60 mT. The shape of the hysteresis loop is similar to the MOKE data in Fig. 2b. Hence, the appearance of hysteresis demonstrates that the polarization degree of QW emission is controlled by the (Ga,Mn)As FM layer. The magnetization of this layer is,
Figure 3  (a) Spectrum of photoluminescence (PL) intensity (solid line) and degree of circular polarization \( \rho_c \) (open dots) for the ‘10 nm’ sample measured under linearly polarized above-barrier excitation with photon energy \( h \omega_{\text{exc}} = 1.92 \text{ eV} \) and excitation density \( \mathcal{P}_{\text{exc}} \approx 20 \text{ W cm}^{-2} \). (b) Polarization hysteresis loop in the ‘10 nm’ sample measured in Faraday geometry by PL circular polarization \( \rho_c(B) \) (open circles) and PL intensity modulation \( \eta \) (solid squares). The excitation photon energy \( h \omega_{\text{exc}} = 1.44 \text{ eV} \), which corresponds to quasi-resonant below-barrier excitation of the QW. This allows us to perform excitation and detection through the GaAs substrate in order to eliminate the magnetic circular dichroism effect in the FM layer (see inset) [38]. Temperature \( T = 2 \text{ K} \).

However, detected by carriers localized in the QW. Therefore, circular polarization of the PL from the QW reflects the FM state, i.e. the read out of FM magnetization by means of the carriers localized in the QW is accomplished.

3 Equilibrium spin polarization in effective exchange field versus spin-dependent capture  It is essential to answer the following two questions: which charge carriers in the QW detect the magnetization of the FM, and what is the origin of their spin orientation? One may assume that an equilibrium spin polarization of charge carriers in the SC is established by the effective magnetic field due to the s(p)–d exchange interaction of the carriers with Mn ions [22, 32, 33]. The degree \( \rho \) is given by the spin polarizations of electrons and/or holes, which, in turn, are proportional to the out-of-plane magnetization component \( M_z(B) \) of the FM. An example of equilibrium spin polarization of holes (electrons) due to the thermal population of spin levels split by the effective exchange magnetic field is schematically shown in Fig. 1b. Under linearly polarized excitation unpolarized photoexcited electrons (holes) recombine with equilibrium spin-polarized heavy holes (electrons). Consequently, PL is circularly polarized. The sign of the polarization degree is positive (\( \rho_c > 0 \)) when the magnetic field is applied in the forward direction, e.g. \( B = +60 \text{ mT} \). In the ferromagnetic (Ga,Mn)As layer, the equilibrium Mn spin with a Landé factor of \( g = +2 \) is directed against the external magnetic field. Thus, the equilibrium spin of electrons (holes) in the QW would be oriented against (along) the magnetic field, due to the ferromagnetic (antiferromagnetic) s–d (p–d) coupling with Mn spins. According to the selection rules for optical transitions, the contribution of both charge carriers to the PL polarization would have to be positive, which is in accord with experimental observations.

Another dynamical mechanism, which is based on the spin transfer between FM and SC layers, induces the non-equilibrium polarization [38]. Here, the FM works as a spin separator redistributing the spins spatially (see Fig. 1c). We assume that tunnelling processes are more efficient for the electrons due to their small effective mass. The resulting SDC leads to an accumulation of electrons with spin opposite to those leaving the QW faster. Symmetry arguments suggest that the SDC rates \( \gamma_+ \) and \( \gamma_- \) should be different for electrons with spins along and against the magnetization. In earlier experiments, the non-equilibrium polarization of free electrons in MnAs/bulk-GaAs structures was observed by means of time-resolved Kerr rotation [41]. It was explained by their spin-dependent reflection from the FM–SC heterointerface [42]. In our case, spin polarization appears under below-barrier excitation with photon energies \( h \omega_{\text{exc}} \) smaller than the GaAs band gap \( E_{\text{gGaAs}} = 1.52 \text{ eV} \), which means that photoexcited carriers are generated in the QW only.

An important consequence of SDC is reflected in the dependence of the electron population in the QW on the helicity of exciting light. For \( \sigma^- \)-polarized excitation, photoexcited electrons are generated with spin oriented along or against the magnetization \( \mathbf{M} \). We introduce a modulation parameter \( \eta = (I^+ - I^-)/(I^+ + I^-) \), which characterizes the difference of the photoexcited carrier population when the helicity of exciting light changes. Here, \( I^+ \) and \( I^- \) correspond to the total PL intensity measured under excitation with \( \sigma^+ \)– and \( \sigma^- \)-polarized light, respectively. Note that the total intensity is proportional to the total number of electrons in the QW. The modulation parameter is sensitive to excitation of light and should change sign in the case where the spin-dependent rates are reversed. This is because the total PL intensity depends on the tunnelling rate. So, it should behave in a similar way to the degree of circular polarization \( \rho_c \) under linearly polarized excitation. In contrast, for small equilibrium spin polarization of carriers in the QW the total intensity is independent of the helicity of the excitation, i.e. \( \sigma^- \)-polarized excitation would give the same total PL intensity as \( \sigma^+ \) excitation in the case where no SDC takes place. Below, we show that SDC is the main mechanism which is responsible for FM-induced circularly polarized QW emission.
Quantitatively, the optical orientation of electrons by SDC is described by the rate equations

\[
\begin{align*}
\frac{dn_+}{dt} &= \frac{G}{2} \left(1 + P_i\right) - \gamma_s n_+ - \frac{n_+ - n_-}{2\tau_s}, \\
\frac{dn_-}{dt} &= \frac{G}{2} \left(1 - P_i\right) - \gamma_s n_- - \frac{n_- - n_+}{2\tau_s},
\end{align*}
\]

where \(n_\pm\) are the numbers of electrons with spin projections \(S_z = \pm 1/2\), \(G\) is their generation rate, and \(P_i\) is their initial polarization determined by the selection rules for circularly polarized light and by their spin relaxation before thermalization. For resonant excitation of the heavy hole subband by \(\sigma^+\)-polarized light the polarization \(P_i = \mp 1\) [28]. \(\tau\) and \(\tau_s\) are the times of radiative recombination and spin relaxation of electrons; \(\gamma_s^\mp\) are the rates of SDC of electrons with spin projections \(S_z = \pm 1/2\) into the FM. Equation (1) neglects the polarization of holes in the QW (the time \(\tau\) is independent of spin) and the thermal orientation of charge carriers in the exchange field of the FM. Under steady-state conditions the concentration of electrons \(n = n_+ + n_-\) and their spin polarization \(P = (n_+ - n_-)/(n_+ + n_-)\) are given by

\[
\begin{align*}
n & = \frac{G}{\gamma + \gamma_s^+ + \gamma_s^- + \gamma \beta P}, \\
P & = \frac{(\gamma + 1/\tau)P_i - \gamma \beta}{\gamma + 1/\tau + 1/\tau_s - \gamma \beta P_i},
\end{align*}
\]

where \(\gamma = (\gamma_s^+ + \gamma_s^-)/2\) gives the mean capture rate of electrons by the FM, and the parameter \(\beta = (\gamma_s^+ - \gamma_s^-)/(\gamma_s^+ + \gamma_s^-)\) characterizes the asymmetry of the capture rates for spin-up and spin-down electrons. The parameter \(\beta\) is an odd function of the magnetization \(M_i\) of the FM. It is seen from Eq. (2) that the concentration of electrons depends on the mutual orientation of \(P\) and \(M_i\), which becomes most pronounced for \(\gamma \tau \gg 1\), when the capture into the FM dominates. Accordingly, the electron polarization \(P\) in Eq. (2), besides the usual term \(\propto P\) describing optical orientation, contains an additional contribution, \(-\beta\). The minus sign indicates that the electron spin in the QW is directed opposite to the electron spins that are caught faster by the FM. Following Eq. (2) and taking into account the selection rules for optical transitions between the electron and the heavy hole subbands, we obtain the PL polarization degree under linearly polarized excitation \((P_i = 0)\) and intensity modulation parameter

\[
\begin{align*}
\rho & = -P = -\frac{n_+ - n_-}{n_+ + n_-} = \frac{\gamma \beta}{\gamma + 1/\tau + 1/\tau_s}, \\
\eta & = \frac{n(-|P_i|) - n(+|P_i|)}{n(-|P_i|) + n(+|P_i|)} = \frac{\gamma \beta |P_i|}{\gamma + 1/\tau + 1/\tau_s}.
\end{align*}
\]

Therefore, it follows that in the case of the SDC mechanism there is a remarkable relation between the degree of circular polarization and the modulation parameter:

\[
\eta(B) = |P_i|\rho_s(B).
\]

For resonant excitation of the QW heavy hole subband, the value of \(P_i\) is close to unity so that the magnetic field dependences of \(\rho_s(B)\) and \(\eta(B)\) coincide. Indeed, it follows from Fig. 3b that magnetic hystereses of \(\rho_s(B)\) and \(\eta(B)\) are the same under below-barrier excitation with \(\hbar\omega_{exc} = 1.44\,\text{eV}\). As mentioned above, the modulation parameter is not influenced by the equilibrium spin polarization. Therefore, we conclude that the main contribution to the spin-induced polarization is given by spin separation of photoexcited carriers, e.g. spin-dependent tunneling.

\section*{4 Dynamics of FM-induced spin polarization}

Time-resolved PL allows us to monitor directly the accumulation of electron spins in the QW and to obtain dynamical parameters of this process: the PL decay rate, e.g. the total decay rate of electrons \(\Gamma = 1/\tau + \gamma\) through the capture into the FM and radiative recombination, the spin-relaxation time \(\tau_s\), and the spin-acumulation rate \((\gamma_s^+ - \gamma_s^-)/2\) due to SDC. The electron spin evolution after excitation with an optical pulse can be described by Eq. (1) without generation term \((G = 0)\). For solving Eq. (1), we should set the initial conditions for electron spin-up and spin-down populations \(n_\pm(t = 0) = n_{0 \pm}\). Under linearly polarized excitation, \(n_0^+ = n_0^-.\) Under circularly polarized excitation and ideal selection rules, \(n_\pm = 1\), and \(n_\pm = 0\) for \(\sigma^\pm\) excitation. The solutions \(n_\pm(t)\) are quite bulky. However, as follows from the experiment the intensity transients \(I(t)\) show a significantly faster decay than the dynamics of both polarization \(\rho_s(t)\) and modulation parameter \(\eta(t)\). This means that \(\Gamma \ll 1/\tau_s\), \((\gamma_s^+ - \gamma_s^-)/2\). In this case, the formulas become much simpler. The intensity decays exponentially with time, while over the whole range of PL emission the polarization \(\rho_s(t)\) and the parameter \(\eta(t)\) evolve linearly with time:

\[
\begin{align*}
I(t) &= (n_0^+ + n_0^-) \exp(-\Gamma t), \\
\rho_s^\pm(t) &= -\frac{n_0^\pm - n_0^-}{n_0^+ + n_0^-} \left(1 - \frac{t}{\tau_s}\right) + \frac{2n_0^\pm n_0^\mp \gamma_s^\pm - \gamma_s^-/2}{(n_0^+ + n_0^-)^2} t, \\
\eta(t) &= \frac{\gamma_s^- - \gamma_s^+}{2} t.
\end{align*}
\]

From Eq. (5), it is seen that the intensity decay is determined by the total escape rate \(\Gamma\), and the intensity modulation is given by \((\gamma_s^+ - \gamma_s^-)/2\). It is interesting to consider the polarization evolution under different excitation conditions. For example, for circularly polarized excitation, say \(\sigma^+\), the degree of polarization

\[
\rho_s^+(t) = 1 - \frac{t}{\tau_s}.
\]
is given at the initial stage by the spin-relaxation time $\tau_s$. This result can be understood in the following way. The electrons are excited fully polarized with spin down and the SDC mechanism (given by the $\gamma c n_s$ terms in Eq. 1) does not mix the populations of spin-up and spin-down states. Therefore, PL depolarization occurs initially only due to spin relaxation. On the contrary, under non-polarized excitation ($n_s^0 = n_d^0$) the degree of polarization

$$\rho_c(t) = \frac{\gamma_c - \gamma_d}{2}$$  \hspace{1cm} (7)

is given by the asymmetry of the capture rates into the FM. The populations of spin-up and spin-down states are equal at $t = 0$ and therefore the spin-relaxation term $\propto (n_d - n_s)/2\tau_s$ does not contribute, while the growth of electron spin polarization is attributed solely to SDC. Therefore, time-resolved measurements allow one to measure directly the main dynamic parameters $\Gamma, \gamma_c - \gamma_d$, and $\tau_s$.

For time-resolved PL measurements, we used a tunable mode-locked Ti:sapphire laser emitting pulses with a duration of 1 ps at 76 MHz rate and a single monochromator, equipped with a streak camera providing an overall time resolution below 20 ps. All PL experiments were performed in backscattering geometry. Optical excitation and detection of either $\sigma^+$ or $\sigma^-$ polarization were selected by rotating a λ/4 plate in conjunction with a Glan–Thompson prism.

The intensity transients $I(t)$ for ‘5 nm’ and ‘10 nm’ samples are shown in Fig. 4a. The PL intensity decays exponentially and, using Eq. (5), we find that the time constant $\Gamma^{-1}$ decreases significantly from 440 to 30 ps when the spacer $d_s$ is reduced from 10 to 5 nm. It follows that $\Gamma$ varies exponentially steeply with $d_s$, which hints at the strong contribution of the electron capture rate $\gamma$ by the FM. The time evolutions of the circular polarization $\rho_c(t)$ under linearly polarized excitation and of the modulation parameter $\eta(t)$ under below-barrier excitation ($\hbar\omega_{exc} < 1.52$ eV) are summarized in Fig. 4b. At the time of pulse arrival ($t = 0$), both quantities are zero, as there is no equilibrium spin polarization in the QW. Both $\rho_c$ and $\eta$ follow then the linear increase in time, and change their sign when the external magnetic field is reversed. From a linear fit of $\rho_c(t)$ using Eq. (7), we obtain for $(\gamma_c - \gamma_d)^{-1}$ the values of 5.5 ns and 240 ps in samples ‘10 nm’ and ‘5 nm’, respectively. In the ‘5 nm’ sample the SDC is about 25 times faster than in ‘10 nm’, which leads to electron dynamical spin polarization as large as 30%.

The evaluated SDC rates are much longer than the corresponding electron lifetimes and hence the linear approximation in Eq. (5) holds (the spin-relaxation time $\tau_s$ is also long and will be discussed in the next section). In this case of pulsed excitation, the remarkable relation of Eq. (4) between $\rho_c(t)$ and $\eta(t)$ also holds (see Eqs. 5 and 7). Indeed, in the ‘10 nm’ sample, the dependences of polarization degree and modulation parameter coincide, i.e. $P_i \approx 1$. In the ‘5 nm’ sample the appearance of $\rho_c$ is steeper than in the case of $\eta$, which can be attributed to the lower efficiency of optical spin polarization in the QW when the excitation energy is increased from 1.44 to 1.50 eV ($P_i < 1$).

Figure 4. Time-resolved data for the ‘5 nm’ and ‘10 nm’ samples. (a) PL intensity transients (dots) fitted with an exponential decay (lines). $\Gamma^{-1} \approx 30$ ps for ‘5 nm’ and $\Gamma^{-1} = 440$ ps for ‘10 nm’. (b) Appearance of circular polarization $\rho_c$ (solid symbols) under linearly polarized excitation and modulation parameter $\eta$ (open symbols) for $B = +125$ mT. The excitation photon energies are 1.50 and 1.44 eV for samples ‘5 nm’ and ‘10 nm’, respectively, which correspond to below-barrier excitation $\hbar\omega_{exc} < 1.52$ eV. (c) Appearance of $\rho_c$ in the ‘10 nm’ sample for different excitation energies $\hbar\omega_{exc}, T = 2$ K.

The dependence on the photon excitation energy indicates that quasi-resonant excitation is very important for clear identification of the SDC mechanism in the FM–SC hybrid structures. This is demonstrated in Fig. 4c, which shows the time evolution of $\rho_c$ under linearly polarized excitation for various excitation energies in the ‘10 nm’ sample. It follows that $\rho_c(t)$ undergoes significant changes when the photon energy becomes larger than 1.52 eV, i.e. the photoexcited carriers are generated in the GaAs barrier rather than in the QW. First, the degree of polarization does not equal zero at the time of pulse arrival ($\rho_c(t = 0) \approx 2\%$). Second, the
polarization rise is slower. At first glance, the appearance of a non-zero initial polarization indicates the existence of equilibrium spin polarization. However, it is most likely due to additional mechanisms of electron spin polarization in the barriers: the electrons get polarized before being captured into the QW. One of such mechanisms is the spin-dependent reflection from the FM [41]. In this case, linearly polarized optical excitation generates first spin-polarized electrons in bulk GaAs, which subsequently get captured into the QW. Thus, in accordance with Eq. (5), the dependence of the polarization degree remains linear in time but its slope should be smaller due to the electron spin relaxation process. This is in accordance with experimental data presented in Fig. 4c. In addition, the decrease of the slope might be related with delayed capture of electrons from the barriers into the QW, which is consistent with the appearance of a rise time in the intensity transients under above-barrier excitation.

5 Optical orientation in FM–SC structures

Time dynamics of circular polarization under circularly polarized excitation $\rho^c(t)$ and its dependence on the FM magnetization allow us to determine the main mechanisms which are responsible for spin relaxation of the carriers. These data are summarized in Fig. 5 for ‘5 nm’ and ‘10 nm’ samples under below-barrier excitation with photon energies 1.50 and 1.44 eV, respectively. In a magnetic field the average $\rho^c(t) = [\rho^c(t, +B) + \rho^c(t, -B)]/2$ measured for opposite field directions corresponds to the optical orientation signal. This allows us to exclude odd-$B$ effects related to SDC. In the ‘10 nm’ sample, within the first 20 ps, there is a fast decay, which is attributed to spin relaxation of the holes. It is not observed in the ‘5 nm’ sample, which means that the spin relaxation of holes is even faster here. Such fast dynamics is not seen in the spin-separation transients in Fig. 4. Hence, we can exclude the hole contribution to the spin-separation effect [38].

The long decay of $\rho^c$ is due to the electrons. It can be well fitted with a linear dependence in accordance with Eq. (6). In the ‘10 nm’ sample we evaluate $\tau_s = 9$ ns at $B = 0$. The electron spin relaxation time increases up to 30 ns in an external magnetic field of 125 mT. Such a field converts the multi-domain FM into a single magnetic domain. It leads to the suppression of the QW electron spin dephasing owing to the stray fields of FM domains [38]. The electron spin relaxation time decreases with increase of the temperature down to 1 ns at 20 K (see the inset in Fig. 5a). Therefore, the Dyakonov–Perel’ mechanism starts to dominate at higher temperatures [25]. The spin-relaxation time decreases significantly with reduction of the spacer thickness, being about 1 ns for the sample with $d_s = 5$ nm. Here, $\tau_s$ is much shorter and it does not change in a magnetic field of 125 mT. Hence, the spin-relaxation mechanism is different as compared with the ‘10 nm’ sample. It is most likely related to spin relaxation on magnetic ions due to exchange interaction [43, 44]. These centres can be decoupled from the FM layer due to diffusion of Mn into the spacer and therefore the spin relaxation is independent of the magnetization of the FM. Note that the initial value of $\rho^c(t = 0)$ is smaller in the ‘5 nm’ sample. As stated before, this can be attributed to lower efficiency of optical orientation in the QW when the excitation energy is increased from 1.44 to 1.50 eV ($P_i < 1$).

The Hanle effect [25] and time-resolved pump–probe Kerr rotation [45, 46], in a magnetic field applied along the QW plane (Voigt geometry), enable one to detect the ferromagnetism through the spin precession about the effective magnetic field $B_{ex}$ generated by the FM. These measurements therefore allow us to estimate the electron spin splitting $\Delta E_{ex}$ due to the interaction with the FM. The splitting $\Delta E_{ex}$ originates mainly from the exchange interaction with the FM and the hyperfine interaction with dynamically polarized nuclei [38].

For time-resolved pump–probe Kerr rotation we used the same pulsed laser split into pump and probe beams and tuned to the resonance with the QW exciton transition (photon energy 1.417 eV). Optical orientation of photoexcited carriers, by circularly polarized pump pulses, initiates a difference in the refraction indexes for $\sigma^+$- and $\sigma^-$-polarized light. As a result, linearly polarized probe pulses undergo a rotation of their polarization plane by an angle $\theta_R$ that is proportional
Figure 6  Kerr rotation transients in Voigt geometry. The time corresponds to delay between pump and probe pulses and the data are shown for various magnetic fields. The scheme of the experiment is illustrated in the left-hand inset. The data are measured at temperature $T = 8 \, \text{K}$ in the ‘10 nm’ sample. The curves are shifted vertically for clarity. The dashed line for zero field ($B = 0$) data is a fit using a double-exponential decay. Right inset shows the magnetic field dependence of the Larmor precession frequency. A linear fit gives $|g_s| = 0.53$, corresponding to the electron g-factor. The intersection of the linear fit with the y-axis gives an estimate of the electron splitting $\Delta E_{ex} = 2.4 \, \text{\mu eV}$ owing to interaction with the FM layer [38].

to the photoinduced average spin $S$. The pump-beam polarization was modulated between left and right circular with a photelastic modulator operated at 40 kHz. The polarization rotation was homodyne detected in reflection using a polarization bridge in conjunction with balanced photodiodes. Application of a magnetic field in Voigt geometry allows us to resolve electron and hole spin precession and relaxation.

Typical Kerr rotation signals for different transverse magnetic fields, applied to the ‘10 nm’ sample at $T = 8 \, \text{K}$, are shown in Fig. 6. The signal is observed only when the excitation photon energy is tuned to the QW exciton resonance. This indicates that the experimental data monitor the spin dynamics of photoexcited carriers in the QW, not FM. At $B = 0$ the signal is well approximated by a double-exponential decay (see the dashed line in Fig. 6). The initial fast response is attributed to the spin relaxation of holes with $\tau^{-1}_h = 20 \, \text{ps}$, which is in agreement with time-resolved PL data in Fig. 5. The slow response with long decay time $\tau_s = 0.3 \, \text{ns}$ corresponds to the lifetime of optically oriented electrons. The inverse electron spin lifetime $1/\tau_s = 1/\tau_s + \Gamma$. The value of the spin lifetime is in reasonable agreement with time-resolved PL data at $T = 8 \, \text{K}$ with $\Gamma = 3 \, \text{ns}$ and $\Gamma^{-1} = 0.4 \, \text{ns}$.

Application of an external magnetic field $B$ leads to oscillatory behaviour of the long-living component. The frequency of oscillations is given by the Larmor frequency $\Omega_L = |g_s| \mu_B (B + B_{ex})/\hbar$ and can be well described by the form $\cos(\Omega_L t) \exp(-t/\tau_s)$. Note that the electron spin precession frequency $\Omega_L$ is determined by the sum of the external magnetic field $B$ and the internal magnetic fields generated by the FM $B_{ex}$. It follows from Fig. 6 that the linear dependence of $\Omega_L(B)$ does not go through the zero point. The intersection point with the frequency axis gives an estimate of $\Delta E_{ex} = 2.4 \, \text{\mu eV}$. A similar value follows from continuous-wave Hanle-effect measurements [38]. The equilibrium spin orientation of electrons due to this splitting is $P_e = \Delta E_{ex}/(2k_B T) \approx 0.7\%$, at temperature $T = 2 \, \text{K}$, and cannot explain the much more strongly polarized PL signals with $4\%$ polarization (see Fig. 3). Hence, the equilibrium polarization of electrons is negligible in the studied sample. The equilibrium spin polarization of photoexcited holes is also negligible but for another reason. As discussed above, the short spin-relaxation time of holes is not seen in the spin-separation dynamics.

Finally, the SDC rates $\gamma_s - \gamma_s$ and the energy splitting $\Delta E_{ex}$ show clear correlation with ferromagnetic ordering of the (Ga,Mn)As layer. The temperature dependences of these
quantities are presented in Fig. 7. In addition, we show the single-beam MOKE signal in Faraday geometry at saturation field $B = +125$ mT, which is proportional to the out-of-plane magnetization component of the FM layer. The latter shows the step-like dependence with the kink around the Curie temperature $T_c \approx 35$ K. In agreement with our expectations, the SDC rates and energy splitting disappear above $T_c$. This demonstrates that the FM-induced spin polarization persists up to the Curie temperature [38].

6 Conclusions We have studied the spin-polarization dynamics of electrons and holes in (In, Ga)As/GaAs QW structures located a few nanometers apart from the FM (Ga,Mn)As layer. We have considered two main contributions to the FM-induced spin polarization of the SC charge carriers: equilibrium spin polarization due to thermal population of spin levels split by an effective exchange field and dynamical polarization arising from SDC of electrons through the FM–SC interface. Time-resolved PL and time-resolved pump–probe Kerr rotation allowed us to distinguish between these two contributions and to measure directly the main dynamical parameters such as spin-accumulation rates and energy splitting in the effective exchange field. The data clearly demonstrate that the main mechanism of spin polarization in the investigated structures is due to electron spin dependent capture. In the structure with 5 nm thick spacer, the electron spin polarization reaches values of up to 30%. The spin separation originates from the QW electrons and therefore it would take place even for the FM–QW hybrid embedded into an insulating (that is, non-injection) structure.

An important feature of the SDC mechanism is its robustness against the temperature as compared to thermal spin polarization in an effective magnetic field. Efficient spin separation persists up to elevated temperatures as far as the spin relaxation of electrons is small compared with tunneling rates. In contrast, equilibrium spin polarization requires the splitting of electron sublevels, which is significantly larger than the thermal energy $k_B T$. Therefore, it follows that the proper choice of high-temperature ferromagnetic material would allow us to build a spin-separation device, which is operational at elevated temperatures.

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