Magnetotransport and magnetization dynamics of GaMnAs thin films and magnetic tunnel junctions

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We describe the comprehensive characterization of GaMnAs epitaxial thin films and magnetic tunnel junctions (MTJ) by complementary techniques ranging from high-resolution imaging to magnetometry, magnetotransport, time-resolved magneto-optics, and coplanar broadband ferromagnetic resonance (FMR). Magnetometry and magnetotransport on macroscopic samples allow deriving the saturation magnetization and quasi-static reversal fields. Perpendicular transport experiments on patterned MTJ pillars further reveal the tunneling magnetoresistance ratio. Additionally the precessional dynamics are characterized by time-resolved magneto-optics and broadband network analyzer FMR yielding the effective Gilbert damping and the sample anisotropies. The combination of the different techniques allows a comprehensive characterization of the key magnetostatic and dynamic material parameters of GaMnAs-based thin films and devices.

1 Introduction Starting from the discovery of the giant magnetoresistance (GMR) [1, 2] in 1988, the field of spintronics [3] has been in rapid development. It has allowed using the spin degree of freedom for information technology, especially for high-density data storage and readout [4]. Various aspects of spintronics have been studied since then such as spin transfer [5], semiconductor spintronics [6], molecular spintronics [7], or single-electron spintronics [8]. Here, semiconductor spintronics could offer the opportunity to design hybrid devices that can combine the two key branches of information technology: semiconductor logic and magnetic storage. Such semiconductor spintronics would ideally be based on ferromagnetic semiconductors, which are compatible with technologically important materials like Si, Ge, or III–V compounds such as GaAs. In semiconductor materials, one way to induce ferromagnetic properties is the insertion of magnetic impurities into the host matrix. This concept was first realized in the ferromagnetic compound InMnAs [9, 10]. Later, the growth of ferromagnetic GaMnAs [11, 12] was realized by Ohno et al. [13, 14] yielding a Curie temperature $T_C = 60 \text{ K}$ and a saturation magnetization of $M_S = 40 \text{ mT}$ at 5 K. Progress in sample growth and annealing procedures have since then led to significantly improved values of $M_S$ and to higher Curie temperatures up to $T_C = 190 \text{ K}$ [15]. Regardless of this progress, the preparation of GaMnAs with ferromagnetic properties at room temperature has not been successful yet [16]. Despite of this GaMnAs is still considered as the most important model system for the studies of spintronic properties of ferromagnetic semiconductors [17].

The properties and quality of GaMnAs thin films, multilayers, and devices, can be evaluated based on various sets of material parameters. Depending on the envisaged applications different material parameters are relevant and could be independently optimized. In the first place, the preparation of high-quality GaMnAs epitaxial layers relies on the quality of the low-temperature (LT) molecular beam epitaxy (MBE) growth and on the crystalline quality of the thin films and interfaces. The key parameters concerning the magnetostatic properties of GaMnAs films are $M_S$ and $T_C$. Both parameters are directly correlated and can be optimized using post-growth annealing procedures. For reliable magnetization reversal in well-defined magnetic fields the magnetic anisotropy constants $K$ need to be
characterized and controlled. Additionally, magnetotransport parameters such as the tunneling magnetoresistance (TMR) ratio of magnetic tunnel junction (MTJ) devices are important for device operation [18–26]. Last but not least, the ultrafast precessional magnetization dynamics, namely the ferromagnetic resonance (FMR) frequencies $f_{\text{FMR}}$ and the Gilbert damping, become important when considering high-bandwidth operation of future GaMnAs-based spintronics devices. Hence for complete characterization of GaMnAs thin films and devices a broad variety of characterization tools and procedures is required. Although many studies of various aspects of GaMnAs material and device properties have been carried out over the last years most of these studies only address a small subset of the above parameters [17–31].

Here, we review how the comprehensive characterization of magnetostatic and dynamic properties of GaMnAs thin films and devices can be achieved by combining complementary characterization techniques. These techniques span from imaging of the crystalline structure via magnetometry and magnetotransport for the characterization of the magnetostatic parameters to the characterization of the precessional magnetization dynamics by optical and electrical methods. The different characterization techniques and procedures are carried out and compared on a set of as-grown and annealed epitaxially grown GaMnAs thin films.

The article is structured as follows: after this Introduction the low-temperature MBE growth and annealing procedures of GaMnAs thin films and MTJs and the typical crystalline characterization are described in Section 2. In Section 3, the critical material parameters saturation magnetization $M_S$ and Curie temperature $T_C$ are derived from superconducting quantum interference device (SQUID) magnetometry. Section 4 describes magnetotransport characterizations of macroscopic samples. From measurements of the planar Hall effect (PHE) [27, 28], the domain wall nucleation energy is derived. From transport experiments on microstructured MTJ pillars additionally the TMR ratio is determined (Section 5). Furthermore, the precessional magnetization dynamics of different GaMnAs thin films are investigated by time-resolved magneto-optical pump probe experiments (Section 6) yielding the effective Gilbert damping. Broadband coplanar FMR experiments (Section 7) further yield the thin film and crystalline anisotropy constants [29–32]. The combination of these measurements thus allows a complete characterization of all important magnetostatic and dynamic parameters material parameters of GaMnAs ranging from saturation magnetization to Gilbert damping.

2 GaMnAs growth and annealing The GaMnAs epitaxial layers were grown by MBE in a Mod Gen II MBE system. The lowest possible $A_S$-partial pressure of about $2 \times 10^{-8}$ mbar was used in order to maintain two-dimensional (2D) growth and suppress the formation of As-antisite defects. The 2D growth mode was indicated by streaky electron diffraction patterns obtained by in situ reflection high-energy electron diffraction (RHEED). The growth procedure was optimized for comparably thick films in the range between 25 and 100 nm, which are well suitable both for dc characterization as well as for broadband inductive characterization of precessional dynamics. The latter requires a sufficiently large total magnetic moment of the samples, which can be obtained by increasing the magnetic layer thickness. The wafer temperature during growth $T_g$ was measured by a commercial band edge thermometry system (kSA BandiT) [33]. After growth of a 100 nm high-temperature (HT) GaAs buffer layer at $T_g = 560$ °C, the substrate temperature was lowered during a 10 min growth interruption. The following 100 nm LT-GaAs buffer was grown to stabilize $T_g$ at the value for the subsequent GaMnAs growth. Samples with 25 and 100 nm GaMnAs layers were grown and $T_g$ was kept constant within $\pm 2$ and $\pm 5$ °C, respectively, during GaMnAs growth. The total Mn concentration $x_{\text{Mn}}$ in the Ga$_{1-x}$Mn$_x$As layers was estimated from Mn growth rate calibration using RHEED and X-ray diffraction (XRD) of GaMnAs calibration layers. During our growth studies $x_{\text{Mn}}$ was varied between 4% and 7%.

GaMnAs-based MTJs were fabricated by the following growth procedure: after a GaAs buffer layer as described above a 100 nm thick highly carbon-doped p-GaAs non-magnetic bottom contact was grown at $T_g = 560$–580 °C. During a 10 min growth interruption, $T_g$ was reduced to the lower GaMnAs growth temperature. The subsequent MTJ layer sequence consists of a 100 nm GaMnAs bottom layer, a tunnel barrier of 1 nm GaAs, 2 nm AlAs, and 1 nm GaAs, and finally a 50 nm GaMnAs top layer. For the MTJs, a Mn concentration of 5% was chosen for both GaMnAs layers.

The crystalline quality of the GaMnAs epitaxial growth was evaluated by transmission electron microscopy (TEM) in combination with energy dispersive X-ray analysis (EDX). Figure 1 shows typical cross-section images for a 100 nm thick GaMnAs epilayer with $x_{\text{Mn}} = 4\%$. Figure 1a shows a low-magnification TEM image covering the sample range from the GaAs wafer via the two 100 nm buffer layers to the 100 nm GaMnAs top layer. No contrast between GaMnAs and GaAs is found and the layer structure cannot be resolved. Here, the use of additional EDX analysis allows a clear distinction of the two materials as shown in Fig. 1c. The intensity of the Ga L-line is encoded in green and the intensity of the Mn K-line is encoded in orange allowing a clear distinction of the GaMnAs epilayers from the GaAs buffer. Note that the even distribution of the two colors in GaMnAs speaks for a high growth quality without Mn clustering. This high quality of the GaMnAs layers can also be seen in the high-resolution TEM image of Fig. 1b showing the well-defined atomic crystalline structure of GaMnAs.

To improve the magnetic parameters $M_S$ and $T_C$ of the GaMnAs thin films post-growth annealing was applied [34–36]. In our experiments, the samples were annealed for different annealing times (18–90 h) at 200 °C in ambient atmosphere.

The effect of post-growth annealing is generally explained by the out-diffusion of excessive interstitial...
manganese to the surface [30]. These interstitial Mn donors compensate the hole-induced ferromagnetism of Mn on the Ga sites and effectively reduce \( M_S \) and \( T_C \) [16]. Based on optimized annealing procedures, \( T_C \) values of up to 190 K have been reported for a 10 nm-thick film with 10.1% effective Mn concentration [14]. However as the diffusion of defects to the GaMnAs surface should be thickness limited, it was not clear whether post-growth annealing would also be suitable for improving the magnetic properties of rather thick GaMnAs films up to a layer thickness of \( t = 100 \) nm. Here, recent studies [37] have shown that also for films of up to 100 nm film thickness comparable values of \( T_C \) and \( M_S \) can be obtained as reported in the literature for lower thickness of 25 nm [38].

3 Magnetometry The Curie temperature \( T_C \) and the saturation magnetization \( M_S \) of as-grown and annealed samples can be reliably derived from SQUID magnetometry at variable temperatures. In our experiments samples of 5 mm \( \times \) 5 mm lateral dimensions were cut from the wafers and characterized by SQUID. Two typical easy axis (e.a.) SQUID hysteresis loops of an as-grown and an annealed (18 h at 200 °C) 100 nm GaMnAs sample are shown in the inset of Fig. 2. The hysteresis loop of the annealed sample shows a significant increase of the saturation magnetization value \( M_S \) to about 70 mT compared to 30 mT for the as-grown sample, exemplifying the effect of annealing. Note that the loop of the annealed sample reveals a small intermediate plateau around 0 mT. This is most likely due to a small misalignment of the applied field with respect to the easy axis resulting in the temporary rotation of the magnetization by 90° along the secondary crystalline easy axis.

The diffusion process also results in a change of the structural properties and weakening of the domain wall nucleation energy, which result in a reduction of the coercive field and hence in a narrower hysteresis loop [39]. In GaMnAs the increase of \( M_S \) is also directly linked to an increase of the Curie temperature. This can be seen in Fig. 2 where \( M_S \) (as derived from SQUID magnetometry at a saturation field of \( \mu_0 H = 100 \) mT) is plotted as a function of temperature. A strong increase of \( T_C \) from 50 K to about 120 K is found following the annealing procedure.

Figure 3 compiles temperature-dependent magnetization data derived from SQUID magnetometry for a set of samples resulting in the temporary rotation of the magnetization by 90° along the secondary crystalline easy axis.
with 100 nm GaMnAs layer and Mn concentration $x_{\text{Mn}} \approx 4\%$ grown at various $T_g$. It is clearly seen that with increasing $T_g$ from about 218–267°C the magnetization for the lowest accessible temperature of 4 K increases up to 40 mT but decreases again for higher growth temperature of 286°C. At $T_g = 267°C$, the shape of the magnetization curves follows in good approximation a calculated curve from the mean-field theory as indicated by the red dashed line in Fig. 3. The model assumes an averaged exchange field over nearest neighbors and includes the GaMnAs typical hole-mediated ferromagnetic coupling. The curve is calculated for $p = 10^{20} \text{cm}^{-3}$ and $x_{\text{Mn}} = 5\%$ [40]. The good agreement between the experimental and the model curves, which assumes a homogeneous ferromagnet indicates a homogeneous Mn distribution in our GaMnAs layers for this growth temperature. For lower growth temperatures ($T_g \leq 260°C$), the magnetization curves show a different behavior with an opposite curvature and an elongated magnetization tail before the magnetization vanishes at $T_C$. In particular, the sample with $T_g = 260°C$ shows the difference very clearly. Although a similar $T_C$ value is observed as in the sample grown at 267°C, the shape and especially the curvature differs strongly. This behavior is attributed to inhomogeneous Mn or hole distribution in the GaMnAs films, in particular in the surface layer as indicated by etching experiments of GaMnAs samples [41]. For these lower growth temperatures, a homogeneous Mn distribution is not obtained and the local $T_C$ and $M_S$ values can strongly vary across the sample.

In our growth experiments, $T_g = 286°C$ was the highest accessible growth temperature before transition from a streaky RHEED pattern to a spotty one indicating MnAs clustering. For $T_g = 286°C$, again a reduction of $M_S$ is observed, which we attribute to a lower solubility of Mn at elevated temperatures. However, the effective reduction of the Mn content leads to a lower $M_S$ than for $T_g = 267°C$.

The derived dependence of $T_C$ versus $T_g$ for this set of samples with $x_{\text{Mn}} \approx 4\%$ is plotted in the inset of Fig. 3 as full dots. $T_C$ strongly increases with increasing $T_g$ from $T_C = 20 K$ and saturates around $T_C = 70 K$ at the highest $T_g$. This is attributed to the fact that at low temperatures the portion of interstitially incorporated Mn is larger, which counteracts hole-mediated ferromagnetism. At higher growth temperatures, a portion of those interstitial Mn is annealed out during growth, thereby strengthening ferromagnetism.

Additionally, the inset of Fig. 3 displays the results for a few further 100 nm thick GaMnAs samples with higher Mn concentrations. At low growth temperature of $T_g = 220°C$, a maximum Mn concentration $x_{\text{Mn}} \approx 7\%$ (down triangle) can be achieved which results in a $T_C$ value of 40 K, thus twice as high as compared to $x_{\text{Mn}} \approx 4\%$. Higher Mn concentrations are not possible under these growth conditions since the observed spotty RHEED pattern reveals a 3D growth mode indicating MnAs clustering.

Also for higher growth temperatures, the Curie temperature can be increased by increasing $x_{\text{Mn}}$. Here however, at $T_g = 270°C$ only a maximum $x_{\text{Mn}} \approx 5\%$ can be achieved before clustering occurs. This behavior is qualitatively in good agreement with the phase diagram for GaMnAs growth since higher temperatures enhance Mn segregation and reduce Mn incorporation [42]. However, since the Curie temperature is already comparably high for higher $T_g$ the relative increase of $T_C$ from 65 to 78 K is less significant.

Since a fraction of Mn is incorporated as compensating interstitial Mn, which weakens the hole-mediated ferromagnetism of GaMnAs films the out-diffusion of such Mn is an effective way to improve the magnetic properties. This is displayed in Fig. 2 where the changes of $M_S$ and $T_C$ upon post-growth annealing are shown for one sample. A summary of $M_S$ and $T_C$ values before and after annealing of different samples is plotted in Fig. 4. The GaMnAs layers are grown at low $T_g = 220°C$ where the density of Mn interstitials is high, promising a strong annealing effect. As can be seen the annealing process of the 100 nm GaMnAs sample (squares in Fig. 4) leads to a very strong increase of $M_S$ from about 30 to 70 mT and to an increase of $T_C$ from nearly 50 to 120 K as indicated by the black arrows. These high values of $M_S$ and $T_C$ are remarkable since they are comparable to those of thinner reference samples with 25 nm GaMnAs layer thickness (triangles). It was argued before that Mn out-diffusion is thickness limited and thus less effective for thicker GaMnAs layers. Our result shows that for 100 nm thick GaMnAs layers, Mn out-diffusion is yet not significantly thickness limited. Note that for longer annealing times we do not observe any significant further improvement of $T_C$ and $M_S$ (red arrows). Only $T_C$ of the 25 nm sample slightly increases after 90 h annealing time. However, for $M_S$ even a reduction is observed both for 25 and 100 nm films.

The correlation of $T_C$ and $M_S$ for our samples in the as-grown and the annealed states is in good agreement with the
In Fig. 5, two typical hysteresis loops for both the as-grown (black dots) and the annealed sample (red dots) of Fig. 2 are shown. During the sweep $\vartheta = -25^\circ$ was constant corresponding to an in-plane angle $\varphi_H$ of the applied field $\mathbf{H}$ with respect to [100] of $\varphi_H = 30^\circ$. Let us first consider the hysteresis loop of the as-grown sample. For negative fields, the Hall resistance is negative and $\mathbf{M}$ is basically oriented along the negative e.a.1 orientation. At positive fields of about 10 mT (arrow 1), the Hall resistance switches to a positive value. Here, $\mathbf{M}$ switches to positive e.a.2 orientation. At about 20 mT field (arrow 2), the resistances switches back and $\mathbf{M}$ rotates to positive e.a.1 orientation. Such low-field 90° reversal usually takes place by domain wall nucleation and propagation characterized by the domain wall nucleation energy $\varepsilon$ [44]. For the reverse field sweep, the reverse switching process is observed at negative fields. Also the annealed sample (red) shows a similar behavior, however with different switching fields and different Hall resistance amplitude.

The PHE data can be comprehensively analyzed by compiling such measurements at various field angles $\varphi_H$ in a polar plot. For each angle $\varphi_H$, only the positive field range of the up sweep ($0 \rightarrow 30 \text{ mT}$) is color encoded (see color scale bar in Fig. 5, right-hand side). Figures 6 and 7 show such polar plots for the as-grown and the annealed sample, respectively.

The four rectangular checkerboard patterns in the center are characteristic for the cubic anisotropy. They represent the boundaries for the first 90° switching of Fig. 5. The diagonals of the rectangular patterns are along e.a.1 and e.a.2, which are close to [100] and [010]. The length of the diagonal of the rectangles yields the domain wall nucleation energy $\varepsilon$ in units of $M_S$ as marked. From the plot we derive $\varepsilon/M_S = 12 \text{ mT}$.

The tilt of the diagonals with respect to the crystalline axes as well as the opening of the outer corners of the...
The main crystalline orientations, the easy axes (e.a.1, e.a.2) and one hard axis (h.a.) are marked. The different switching properties and anisotropies of the MTJ system they do not allow deriving the TMR ratio of the MTJ. The standard way to derive the TMR, which is a very relevant parameter for potential applications, is to carry out perpendicular-to-plane magnetotransport measurements on patterned MTJ devices. Such data is shown in Fig. 9. It was carried out on a lithographically patterned MTJ pillar of 20 µm diameter having separate electrical contacts to bottom and top GaMnAs electrode. The hysteresis loop is taken along the [100] easy axis. In contrast to the PHE signal, the TMR signal is sensitive to the relative orientation of the two magnetic layers of the MTJ. Two distinct TMR peaks are found corresponding to the two possible antiparallel orientations during the up- and down-sweep as marked by the thicker bottom layer. From this, we derive that the thinner top layer has lower switching field and is thus magnetically softer. The domain wall nucleation energy of the top and bottom layer are $\varphi_e/M_S = 11$ mT and $\varphi_o/M_S = 14$ mT, respectively. Also, the high-field reversal shows distinct switching boundaries of the two layers speaking for slightly different cubic anisotropy fields $K_c/M_S$.

While PHE measurements allow analyzing the reversal properties and anisotropies of the MTJ system they do not allow deriving the TMR ratio of the MTJ. The standard way to derive the TMR, which is a very relevant parameter for potential applications, is to carry out perpendicular-to-plane magnetotransport measurements on patterned MTJ devices. Such data is shown in Fig. 9. It was carried out on a lithographically patterned MTJ pillar of 20 µm diameter having separate electrical contacts to bottom and top GaMnAs electrode. The hysteresis loop is taken along the [100] easy axis. In contrast to the PHE signal, the TMR signal is sensitive to the relative orientation of the two magnetic layers of the MTJ. Two distinct TMR peaks are found corresponding to the two possible antiparallel orientations during the up- and down-sweep as marked by

5 Magnetotransport of magnetic tunnel junctions

Such PHE characterization of large unpatterned samples can also be applied to more complex samples such as MTJ. In this case the current will pass through both layers in parallel and the PHE signal will be composed of the PHE contributions of both layers. The PHE contribution of each layer can be derived by considering parallel conduction through the two GaMnAs layers of given thickness and conductivity. Such PHE characterization of GaMnAs MTJs has been described by Ge et al. [47] to study interlayer exchange coupling in GaMnAs MTJs.

Figure 8 shows a PHE polar plot of a GaMnAs MTJ. As described in Section 2, the bottom and top GaMnAs layers of the MTJ are 100 and 50 nm thick, respectively, and are separated by a GaAs/AlGaAs/GaAs tunnel barrier. The PHE of the MTJ signal basically shows a similar switching behavior as the as-grown GaMnAs thin film. However, in contrast to the single layer the switching boundaries reveal a stepped transition from high to low resistance indicating independent switching of the two layers. The larger signal amplitude of the two PHE signal changes can be attributed to the thicker bottom layer. From this, we derive that the thinner top layer has lower switching field and is thus magnetically softer. The domain wall nucleation energy of the top and bottom layer are $\varphi_e/M_S = 11$ mT and $\varphi_o/M_S = 14$ mT, respectively. Also, the high-field reversal shows distinct switching boundaries of the two layers speaking for slightly different cubic anisotropy fields $K_c/M_S$.

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![Figure 7: Polar Hall plot of the annealed sample of Fig. 2 at 10 K. The data is compiled from field sweeps as shown in Fig. 5 for various angles. The main crystalline orientations, the easy axes (e.a.1, e.a.2) and one hard axis (h.a.) are marked.](image)

![Figure 8: Polar Hall plot of a magnetic tunnel junction at 10 K. The main crystalline orientations, the easy axes (e.a.1, e.a.2) and one hard axis (h.a.) are marked. The different switching fields and of the two GaMnAs layers are clearly resolved.](image)
MTJ wafers, on pillars with different sizes, ranging from in-plane measurements on simpler devices. It was shown [48, 49] that the TMR can also be reliably derived from in-plane measurements on simpler devices at 0.39 K [20]. Note however that for our measurements the maximum bandwidth or switching rate of magnetic thin films is limited by the precession frequency [52–55]. The maximum TMR value obtained in such experiments on our devices was 89% on a 2 μm² to 300 × 300 μm². All these devices showed a similar behavior. The maximum TMR value obtained in such experiments on our devices was 89% on a 2 μm² at 2 K. Using a simple Jullièrre model (TMR = 2P²/(1 − P²)) where P is the spin polarization of GaMnAs, we derive a value of P = 55.5%, which is compatible with the literature [50, 51]. The record value of TMR in GaMnAs-based structures was 290% at 0.39 K [20]. Note however that for our measurements in the current-in-plane configuration, the derived TMR generally does not exceed a few percent.

6 Magneto-optical pump probe measurements

Precessional magnetization dynamics of magnetic thin films are very relevant for a variety of magnetic device applications such as sensor or memory devices. For example, the maximum bandwidth or switching rate of magnetic devices is limited by the precession frequency [52–55]. Optical pump–probe experiments based on excitation using fs laser pulses and time-resolved measurement of the Kerr rotation have shown to be a very convenient way to study time-resolved precessional dynamics of GaMnAs thin films [56–62] and to investigate all-optical magnetization switching [63]. In such experiments, the excitation by an optical pulse induces a transient change in hole concentration and temperature of the GaMnAs. This, in turn, changes the magnetic anisotropy parameters and leads to an ultrafast photoinduced reorientation of the in-plane easy axis [56–59] thereby exciting a coherent magnetization precession.

Our experiments were carried out using a Ti:Sa oscillator producing 130 fs long pulses with a center wavelength of 800 nm and a repetition rate of 76 MHz. The laser beam was split into a pump beam and a probe beam and both beams were focused onto the same position on the sample through the same lens with a focal length of 50 mm. The angle of incidence of the probe beam was roughly 6° off from normal incidence, thus realizing a longitudinal Kerr effect configuration [64]. The magnetic-field-induced phase change of the reflected probe beam was analyzed in a conventional detection setup comprising a quarter-wave plate, a polarizing beam splitter, and a differential detector. Time resolution was achieved by moving a stepper motor, which in turn changes the propagation path length of the probe beam before having been focused onto the sample.

Different excitation schemes using a circularly and linearly polarized pump beam were tested. Figure 10 shows a typical measurement of the time-resolved optically induced precession observed in our films. The sample is the as-grown film of Fig. 2. The data in Fig. 10a were acquired at T = 5 K. The Kerr rotation detected by the probe pulses (30 pJ energy) is plotted in arbitrary units versus time. The excitation pulse hits the sample at t_ex = 65 ps. Two different excitation energies of 450 pJ (blue) and 900 pJ (black) were used. In both curves, the light-induced damped precession of the magnetization following the excitation pulse is well observed. The measured Kerr oscillation can be well described by an exponentially damped sinusoid (red lines). From the fit parameters of the damped sinusoid, the

Figure 9 [100] easy axis TMR loop of a patterned MTJ pillar measured in the current-perpendicular-to-plane geometry. T = 10 K. The orientation of the magnetization of top (gray) and bottom (black) layers is marked by the arrows.

Figure 10 (a) Pump-probe-based time-resolved magneto-optics of 100 nm Ga_{1−x}Mn_xAs, x = 0.05 at 5 K for two different pump energies of 450 pJ (blue) and 900 pJ (black). The damped precession around the crystalline anisotropy axis is well observed and can be fitted by a damped sinusoid (red line). (b) Temperature dependence of the time-resolved measurements, showing that the oscillations vanish near the Curie temperature.
precession frequency $f_{\text{FMR}} = 3.56 \pm 0.35\,\text{GHz}$ and the effective Gilbert damping $\alpha_{\text{eff}} = 0.165 \pm 0.033$ can be derived. In the measurement of Fig. 10a, no external magnetic field is applied. The magnetization thus precesses about the crystalline easy axis along (100) and the precession frequency is determined by the intrinsic anisotropies and by $M_S$. The field and angular dependence of $f_{\text{FMR}}$ will be described in more detail in Section 7.

The effective Gilbert damping parameter derived from our experiments agrees well with the literature values as, e.g., published by Qi et al. [56] (where values of the damping ranged from 0.12 to 0.21) revealing the good crystalline quality of our samples. Here, inhomogeneities of the crystalline structure would lead to an enhanced dephasing of the coherent precession and hence to an extrinsically enhanced effective damping $\alpha_{\text{eff}}$. The determination of the intrinsic damping $\alpha_{\text{int}}$ [56] would require the measurement of $\alpha_{\text{eff}}$ over a broad range of $f_{\text{FMR}}$ (and thus a broad field range) and extrapolation of $\alpha_{\text{eff}}$ to zero frequency. However, in our present setup, no external magnetic field could be applied and the intrinsic damping could therefore not be determined.

When increasing the temperature toward $T_C$, the magnetization $M_S$ is reduced and the measured Kerr signal becomes weaker. This is seen in Fig. 10b. Here, time-resolved Kerr data is plotted for $T = 5, 20, 40\,\text{K}$. As in Fig. 10a at $5\,\text{K}$, the damped precessional Kerr signal is well observed (black). At $20\,\text{K}$ (red), the measured Kerr amplitude is already significantly reduced. Furthermore, the precession frequency $f_{\text{FMR}} = 3.21\,\text{GHz}$ is lower than at $T = 5\,\text{K}$. Both effects can be explained by the reduced magnetization and the reduced crystalline anisotropy at higher temperatures. Finally, at $40\,\text{K}$, the sample is close to $T_C$ and no precessional signal is detectable in our setup.

As mentioned before the precessional dynamics in these pump probe experiments in zero applied field are determined by the intrinsic magnetic material parameters of saturation magnetization and crystalline anisotropy. The dependence of $f_{\text{FMR}}$ on an applied magnetic vector field will be discussed in the following section.

### 7 Broadband ferromagnetic resonance

In the magneto-optical experiments described above, no external magnetic field could be applied. As a consequence, only the precession about the equilibrium orientation along the main crystalline axis could be investigated. In the presence of an applied magnetic field $(H_x,H_y)$, in the plane of the sample the precession frequency $f_{\text{FMR}}$ strongly depends on the amplitude $H$ and the in-plane angle $\phi_H$ of the field with respect to the crystalline axis [100]. Cavity-based FMR is a way to access this field and angular dependence of $f_{\text{FMR}}$. A detailed review of cavity-based FMR in GaMnAs and the influence of the various anisotropy parameters has been given by Liu and Furdyna [65].

However, cavity-based FMR is limited to a fixed frequency and hence does not allow accessing the detailed field dependence of $f_{\text{FMR}}$. This is possible by vector network analyzer-based ferromagnetic resonance (VNA-FMR), where the FMR cavity is replaced by a coplanar waveguide (CPW) as the broadband inductive antenna. Such VNA-FMR has become a versatile tool for all electrical characterization of precessional dynamics of various metallic magnetic thin films and multilayers over the last years [66, 67]. Based on a LT VNA-FMR setup [68] recently also VNA-FMR investigations of GaMnAs have been demonstrated [69].

Figure 11 sketches the setup used in our experiments. For VNA-FMR measurements, $5 \times 5\,\text{mm}^2$ samples of $100\,\text{nm}$ thick GaMnAs are placed on the center of a CPW with suitable geometry [70]: The CPW is placed inside a variable temperature cryostat with base temperature $1.5\,\text{K}$. The experiments described here are carried out at a temperature of $10\,\text{K}$. The CPW is connected to the two ports of a VNA situated outside the cryostat by suitable high bandwidth cables. In-plane static vector fields $\mu_0(H_x,H_y)$ up to $0.5\,\text{T}$ field amplitude are applied inside the same cryostat used for the transport measurements. As sketched in Fig. 11, the GaMnAs sample is placed diagonally on the CPW, so that the high-frequency field $H_{\text{HF}}$ is aligned with the [010] easy axis. At a fixed static field, the frequency output of the VNA is swept between 1 and 18 GHz, at the maximum power of nominally $20\,\text{dBm}$ to maximize the inductive signal. Note that the actual power at the sample is only about $1\,\text{dBm}$ due to the damping of the long HF cables inside the cryostat. For this applied power, nonlinear effects can be ruled out. The forward scattering signal $S_{21}$ is then measured. Under resonance conditions the GaMnAs magnetization is excited into FMR precession and the signal transmission is reduced leading to a Lorentzian absorption line in the VNA sweep. After reference subtraction of a $S_{21}$ measurement at a non-resonant field, the FMR frequencies $f_{\text{FMR}}$ are subsequently derived for different in-plane magnetic field amplitudes $H$ and angles $\phi_H$.

Figure 12 shows typical angular dependent data of $f_{\text{FMR}}$ for the two samples of Fig. 2 for field amplitudes of $0.2\,\text{T}$ (green symbols) and $0.3\,\text{T}$ (red symbols). The data of the as-grown sample is shown in Fig. 12a and the data of the annealed sample in Fig. 12b. The angular dependence of $f_{\text{FMR}}$...
anisotropy derived by the different methods. Note that in Table 1 the parameter values derived from the model fit well captures the measured FMR behavior. The parameter values derived from the model fit are given in Table 1 together with the other material parameters. The corresponding anisotropy parameters can be derived from a numerical fit to an extended Kittel model including in-plane cubic anisotropy $K_c$ and uniaxial anisotropy $K_u$ [65, 71] and the so-called effective thin film magnetization $M_{eff}$ resulting from the interplay of $M_S$ and the thin film shape anisotropy. More details can be found in Ref. [69].

The lines in Fig. 12 are fits to the FMR data using such a model. The fit well captures the measured FMR behavior. The parameter values derived from the model fit are given in Table 1 together with the other material parameters derived by the different methods. Note that in Table 1 the anisotropy fields $H_{ani} = 2K_{ani}/M_S$ are given instead of the anisotropy constants $K_{ani}$. These derived values are in agreement with literature values of GaMnAs thin film anisotropies obtained by conventional cavity FMR experiments on annealed samples [46]. The data of Table 1 again shows the strong impact of annealing on all important material parameters, as already discussed in the previous sections. Please note that while $M_S$ increases upon annealing the effective magnetization $M_{eff}$ is reduced. The latter is due to the competing effects of $M_S$ and the thin film anisotropies on the value of $M_{eff}$.

In Table 1, only the effective damping $\alpha_{eff}$ derived from the magneto-optic experiments of Section 6 is displayed. As mentioned above, a full analysis of the intrinsic damping would require a measurement of the FMR line width over a broad frequency range and subsequent extrapolation of the line width to zero frequency [66]. However, the given VNA-FMR data only allowed a line width analysis over a rather narrow field range around 0.3 T. Therefore, no detailed analysis of the intrinsic damping was carried out. However, please note that for the given narrow field range a significant lower effective damping $\alpha_{eff} = 0.018$ was found. Here a further optimization of the VNA-FMR setup should allow a comprehensive VNA-FMR line width analysis over a broad frequency range allowing to distinguish intrinsic and extrinsic contribution to the FMR line width and to derive the true intrinsic damping $\alpha_{int}$ of GaMnAs thin films [65, 66].

### 8 Conclusions

We have described a comprehensive set of experimental methods for the complete characterization of the magnetostatic and dynamic properties of GaMnAs thin films. The characterization procedures have been exemplified with respect to an as-grown and an annealed 100 nm thick GaMnAs thin film. As shown by the compiled results of Table 1, only a combination of a broad range of complementary characterization techniques allows a complete characterization of all important magnetostatic and dynamic material parameters. These range from material composition to Curie temperature, and via saturation magnetization, domain wall nucleation energy, to crystalline anisotropies and Gilbert damping.

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**Table 1** Experimental values of the magnetic parameters for the as-grown and the annealed 100 nm thick GaMnAs thin films of Fig. 2. The table compiles the data derived by the different methods EDX, SQUID magnetometry, PHE transport, magneto-optics (MO), and VNA-FMR. The FMR data are calculated using a gyromagnetic ratio of $g = 1.71 \times 10^{11}$ s$^{-1}$ T$^{-1}$ [69].

| parameter     | as-grown | annealed | method   |
|---------------|----------|----------|----------|
| $x_{Ms}$      | 7%       | 7%       | EDX      |
| $M_S$         | 30 mT    | 74 mT    | SQUID    |
| $T_C$         | 50 K     | 120 K    | SQUID    |
| $\omega M_S$  | 12 mT    | 6 mT     | PHE      |
| $\alpha_{eff}$| 0.165 ± 0.033 | - | MO |
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