Stretching magnetism with an electric field in a nitride semiconductor

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The significant inversion symmetry breaking specific to wurtzite semiconductors, and the associated spontaneous electrical polarization, lead to outstanding features such as high density of carriers at the GaN/(Al,Ga)N interface—exploited in high-power/high-frequency electronics—and piezoelectric capabilities serving for nanodrives, sensors and energy harvesting devices. Here we show that the multifunctionality of nitride semiconductors encompasses also a magnetoelectric effect allowing to control the magnetization by an electric field. We first demonstrate that doping of GaN by Mn results in a semi-insulating material apt to sustain electric fields as high as 5 MV cm\(^{-1}\). Having such a material we find experimentally that the inverse piezoelectric effect controls the magnitude of the single-ion magnetic anisotropy specific to Mn\(^{3+}\) ions in GaN. The corresponding changes in the magnetization can be quantitatively described by a theory developed here.
It has been known for a long time that piezoelectricity specific to crystals with no inversion symmetry offers a spectrum of outstanding functional properties. For instance, various nаноэлектромеханического and energy harvesting devices employing wurtzite II–VI and III–V semiconductor compounds have recently been demonstrated. At the same time, nano-composite or hybrid piezoelectric/magnetic systems allow for the electric control of magnetization.

In Ga$_{1-x}$Mn$_x$N system a single-ion magnetic anisotropy dominates for Mn$^{3+}$ ions that assume in GaN a high-spin configuration with spin and orbital momentum $S=2$ and $L=2$, respectively. For the experimental values of the lattice parameters $a$ and $c$ the easy axis is perpendicular to the polar [0001] direction (c axis) of the wurtzite structure. At the same time a mid-gap Mn$^{4+}$/Mn$^{3+}$ level traps electrons introduced by Si or residual donors, so that the material becomes semi-insulating for $x \approx 0.05\%$ (refs 8,9). Because of Anderson–Mott and Hubbard–Mott localization of carriers residing in the Mn impurity band, the semi-insulating character of Ga$_{1-x}$Mn$_x$N permits us to the highest available Mn concentrations. In this dilute magnetic insulator, the exchange coupling between Mn$^{3+}$ ions is dominated by short-range ferromagnetic superexchange interactions leading to a ferromagnetic ordering at $T_c \approx 1$ K for $x \approx 3\%$ (ref. 10).

Here we find experimentally the existence of a strong coupling between piezoelectricity and magnetism in the dilute magnetic insulator Ga$_{1-x}$Mn$_x$N and develop a theory that describes the effect quantitatively. The measurements are performed at $T \geq 2$ K and $x \leq 3\%$, that is in the paramagnetic regime. Under these conditions, the inverse piezoelectric effect by stretching the elementary cell along the c axis affects the magnitude of the uniaxial magnetic anisotropy and, thus, of the magnetization. We reveal this novel magnetoelastic effect by direct magnetization measurements employing a new detection method. Our work bridges two fields of research so far independent: piezoelectricity of wurtzite semiconductors and electric control of magnetization in magnetic insulators. More specifically, the findings presented in this Communication lead to the conclusions: (i) the magnetoelastic effect generated by piezoelectricity can exist in a homogenous crystalline compound, not only in hybrid or nanocomposite systems, (ii) the multifunctional capabilities of Ga$_{1-x}$Mn$_x$N extend into the core of spintronic functionalities, like the manipulation of magnetization by the electric field.

**Results**

**Samples.** The observation of the piezoelectromagnetic effect (PEME) is built on our recent progress in the metal-organic vapour phase epitaxy (MOVPE) and nanocharacterization of wurtzite Ga$_{1-x}$Mn$_x$N and Ga$_{1-x}$Mn$_x$N:Si layers, which led us to fabricate a single phase homogenous alloy with Mn atoms substituting Ga sites up to $x = 3.5\%$ (ref. 8). The structures studied here are grown by MOVPE onto c-plane 2" sapphire substrates and have the c axis parallel to the growth direction and Ga-face polarity. They consist of 1 μm GaN buffer, a 500 nm-thick Si doped n$^-$-GaN bottom contact layer ($n \approx 10^{18}$ cm$^{-3}$), and a 30 nm-thick Ga$_{1-x}$Mn$_x$N or Ga$_{1-x}$Mn$_x$N:Si film to which a vertical electric field can be applied. Structures with three different films have been deposited, namely: Ga$_{1-x}$Mn$_x$N, Ga$_{1-x}$Mn$_x$N:Si having $x = 2.4$ and 2.5% as established from magnetization data—denoted as A and B, respectively—and a reference structure containing Ga$_{1-x}$Mn$_x$N:Si with $x = 0.1\%$ (denoted as R). Samples of dimensions $(5 \times 5)$ mm$^2$ are cut out from the wafers, and systematically characterized by atomic force microscopy (AFM), high-resolution x-ray diffraction (HRXRD), secondary-ion mass spectrometry, high-resolution transmission electron microscopy (HRTEM), and superconducting quantum interference device (SQUID) magnetization measurements.

**Magnetization as a function of the magnetic field.** Because of a relatively small film thickness and low Mn content, an accurate determination of magnetization $M(T, H)$ for our Ga$_{1-x}$Mn$_x$N layers by SQUID magnetometry has required an extension of the previous experimental methodology$^{71}$. The modifications are outlined in the section Methods.

The circles in Fig. 1 represent the experimental data $M(T, H)$ per unit sample area $A$ for the A and B Ga$_{1-x}$Mn$_x$N films at two orientations of the magnetic field $H$ with respect to the c axis. No hysteretic behaviour is found down to the experimental noise level $\sigma_{\text{ME}}(H \geq 0) \leq 1 \times 10^{-6}$ emu cm$^{-2}$, where $A = 0.25$ cm$^2$ at any investigated temperature (2, 5, 10 K—presented in Fig. 1—and at 50 and 300 K, not shown). This points to the absence of a ferromagnetic phase that might originate from Mn-rich aggregates. This conclusion is further supported by results of HRXRD and HRTEM studies on these samples presented in Methods, and which—within the ultimate limits specific to these techniques—rule out the presence of any Mn-rich phases either in the form of nanometric precipitates or of regions of spinodal decomposition$^{12}$.

Actually, both layers exhibit magnetism characterized by $M(T, H = 0) = 0$, $M(T, H \rightarrow \infty)$ tends to saturate, and $\chi(T) \propto M(T, H = 0)/dH$ increases with lowering temperature according to $\chi(T) \propto 1/(T - T_0)$. This behaviour demonstrates that the studied samples at $T \geq 2$ K are in the paramagnetic phase, though ferromagnetic coupling between neighbouring Mn spins starts to be visible and leads to $T_0 \approx 0$. At the same time, a sizable uniaxial magnetic anisotropy is observed, $M(T, H \parallel c) > M(T, H \perp c)$, which is specific for Mn$^{3+}$ ions in wurtzite GaN (refs 6,7). The starting point of the theory aiming in description of $M(T, H)$ in the paramagnetic phase is a single-ion Hamiltonian in the form implied by the group theory for Mn$^{3+}$ ions in wurtzite GaN (refs 6,7). This Hamiltonian and the corresponding crystal-field, spin-orbit and Jahn-Teller parameters employed to generate theoretical curves in Fig. 1 are presented in Methods. The theoretical procedure, developed earlier for purely paramagnetic Ga$_{1-x}$Mn$_x$N ($x < 0.6\%$) containing Mn$^{3+}$ ions, is modified here to describe samples with higher Mn concentrations. In particular, we introduce an effective temperature $T = T - T_0(T)$ as a fitting parameter, where
interpretation of magnetic anisotropy. The single-ion magnetic Mn\textsubscript{3}T(T) and for Mn\textsubscript{3}T(T) the lattice parameters \(a\) uniaxial anisotropy originates primarily from the trigonal

\begin{align*}
\text{B}_1 & = \left(1 + \frac{1}{3} \right) G_{\text{Ga}} \\
\text{B}_2 & = \left(1 - \frac{1}{3} \right) G_{\text{Ga}}
\end{align*}

While another fitting parameter \(x\) controls the magnitude of the slope \(dM(T, H=0)/dH\).

Especially relevant for our work is the quantitative interpretation of magnetic anisotropy. The single-ion magnetic anisotropy is virtually negligible for Mn\textsuperscript{2+} ions, as the orbital momentum vanishes in this case \((S = \frac{5}{2}, L = 0)\). In contrast, for Mn\textsuperscript{3+} ions in GaN \((S = 2, L = 2)\) a sizable single-ion uniaxial magnetic anisotropy has been reported\(^6\). In GaN:Mn, the uniaxial anisotropy originates primarily from the trigonal deformation of the tetrahedron formed by anion atoms next to Mn, as illustrated in the panels b and c of Fig. 2. In terms of the lattice parameters \(a\) and \(c\), the deformation is described by the parameter \(\xi = \sqrt{a} - \sqrt{8}/3\). In particular, for \(\xi < 0\), the case of GaN:Mn\textsuperscript{3+}, the easy axis is perpendicular to the wurtzite \(c\) axis, whereas in the opposite case \(\xi > 0\), the easy axis assumes the direction of the \(c\) axis. According to the theoretical model of Mn\textsuperscript{3+} ions (see Methods), the effect of trigonal deformation is described by two crystal field parameters, \(B_1\) and \(B_2\), for which, the relevant small values \(\xi\), can be assumed to be linear in \(\xi, B_1 = \gamma_1 \xi\). The values of \(\gamma_1\) are known from previous investigations of thick

\(T_\text{g}(T) > 0\) takes phenomenologically into account the presence of spin-spin coupling in dilute ferromagnets above \(T_\text{C}\). While another fitting parameter \(x\) controls the magnitude of saturation magnetization, the Curie-Weiss-like temperature \(T_\theta(T)\) determines, at given \(x\), the magnitude of the slope \(dM(T, H=0)/dH\).

The lattice parameter \(a\) of our thin films is pinned by the GaN buffer. According to X-ray rocking curves and reciprocal space mapping (RSM) \(a = (3.1842 \pm 0.0003)\) Å in our samples, and this value is employed further on for the data analysis. In contrast, the minute thickness of our layers precludes a sufficiently precise experimental determination of the \(c\) parameter by HRXRD, so that we evaluate it from the magnitude of low-temperature magnetic anisotropy in sample A. This procedure leads to \(\xi = -0.0058\).

The corresponding changes of the Jahn-Teller describing the distortion of tetragonal symmetry are unknown but a better fit is obtained by increasing their absolute values by 15% with respect to their magnitudes for \(x = 0.4\%\) (ref. 7), for which the fitting of magnetic anisotropy led to \(B_1 = 4.2\) meV and \(B_2 = -5.56\) meV, whereas HRXRD measurements to \(a = 3.191, c = 5.1863\) Å and, thus, \(\xi = 0.077\).

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According to data in Fig. 1a,b magnetic anisotropy is weaker in Sample B. As established previously\(^3\), the doping with shallow Si donors reduces the oxidation state of a fraction \(z\) of the Mn ions from the \(3+\) to the \(2+\) charge state, as the Mn\textsuperscript{2+/3+} state resides in the mid gap region. As recalled in the theoretical part of Methods, the magnetization for Mn\textsuperscript{2+} ions in GaN is described
by the isotropic Brillouin function for $S = 5/2$. The magnitudes of $x$, $z$, and $T'$ are determined by fitting the combined theoretical magnetization for Mn$^{3+}$ ions and Mn$^{2+}$ ions with weights $1 - z$ and $z$, respectively, to the whole-set of experimental data.

The most appropriate account of the experimental finding is obtained by taking $x = 2.4\%$ and $z = 0$ for sample A, whereas $x = 2.5\%$ and $z = 0.1$ for sample B. The magnitudes of the effective temperatures $T'$ obtained from the fitting are displayed in Fig. 1. As indicated in Fig. 1 by solid lines, the employed approach with the quoted values of the input parameters provides a correct reproduction of the experimental data.

**Structure processing and experimental set-up.** Having parameterized $M(T, H)$, we turn to the measurements of the magnetic moment as a function of the electric field. In order to minimize the risk of structure damage by an electrical short via, for example, threading dislocations, two precautions are undertaken, as sketched in Fig. 2a. First, after removing by reactive ion etching (1 $\mu$m$^2$ square down to the GaN:Si bottom contact layer, atomic layer deposition is employed to fabricate a 100 nm gate oxide (Al$_2$O$_3$/HfO$_2$), which after appropriate masking is covered by Ti/Au to serve as a top insulating material. For these evaluations we employ the value of the oxide dielectric constant $E_{\text{Ga}N}$ expected because, according to the comparison with $E_{\text{Ga}N}$, the samples' capacitance as well as the dielectric strength and losses are measured for each of these capacitors, and only those showing no leaks are connected in parallel and studied at low temperatures. The capacitance is below the detection limit in the case of sample R (Fig. 3a), which contains more than twenty times less Mn ions than samples A and B.

In order to detect directly the effects of an electric field on magnetism we place the structured sample chamber. By redirecting the output of the SQUID electronics to the input of a digital lock-in nanovoltmeter, we look for the magnetic signal $M_E$ in phase with the a.c. electric field $E$ of low frequency $f$, as shown in Fig. 2a. Alternatively, the film is biased by a d.c. electric field up to $E = 2.8$ MV cm$^{-1}$ and the phase sensitive detection of the slope of the $M_E(E)$ is measured with an a.c. component $E_{\text{mod}} = 0.26$ MV cm$^{-1}$. A noise-limited resolution of $3 \times 10^{-11}$ emu at $f = 1$ Hz and for $2 \mu$s of lock-in output averaging has been achieved in the absence of magnetic field. Both the signal and noise increase with the magnetic field, but acquisition times up to $2 \mu$s ensure an adequate signal to noise ratio under our experimental conditions. No dependence of $M_E$ on $f$ is found between 0.1 and 200 Hz, however $f$ is kept below 3 Hz. The whole set-up is calibrated against a copper coil of known dimensions fed with an a.c. current. Experiments with the coil demonstrate also the linearity of the output with the magnetic moment over the relevant range of magnetic moments independently of an external magnetic field $H$ up to at least 20 kOe.

**Magnetization as a function of the electric field.** Having calibrated the system, we turn to the experimental determination of the effects of an applied electric field on magnetization. The results obtained at 2 K are summarized in Fig. 3. As indicated, no magnetic signal generated by the electric field is detected in the absence of magnetic field. This is expected as the time reversal symmetry is maintained in the paramagnetic phase and $z = 0$ in the lowest order magnetoelectric term $M_E = 2E$ (ref. 14). However, for $H \neq 0$ a sizable electric-field-induced magnetic signal $M_E$ is seen for samples A (all panels in Fig. 3) and sample B (panel a). Importantly, this signal is substantially smaller for the easy axis configuration $H \perp c$, as exemplified in Fig. 3b for sample A. Finally, we note that the magnitude of $M_E$ is below the detection limit in the case of sample R (Fig. 3a), which contains more than twenty times less Mn ions than samples A and B.

The magnitude of $M_E$ decays rapidly with increasing temperature, as exemplified in Fig. 4 for sample B. This is an expected behaviour because, according to the comparison presented in Fig. 3c, the PEME signal is directly related to the easy and hard axis magnetization difference of the material, $M_L - M_H$, which, on the other hand, has already been shown to vanish at $T \approx 20$ K (Fig. 10 in ref. 7). Additionally, the samples' magnetization response to the electric field gets sizably enhanced at very low temperatures (that is on approaching $T_C \lesssim 1$ K in our

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**Figure 3** | Magnetization changes generated by an electric field as a function of magnetic field. Symbols: experimental data, solid lines: theory.

(a) Hard axis $H \parallel c$, areal density of the magnetic moment amplitude $M_E/A$ induced by electric field of the amplitude $E = 0.66$ MV cm$^{-1}$ for all studied samples. (b) Easy axis $H \perp c$, response to $E = 0.26$ and 0.4 MV cm$^{-1}$ (sample A). (c) Response to $E = 0.66$ MV cm$^{-1}$ for the hard axis $H \parallel c$ and difference in magnetization for the hard and easy axis $(M_L - M_H)/A$ in a wide magnetic field range (sample A).
dependence of ME for both experimental configurations. The results obtained at 2 K
momentum\textsuperscript{18–20}. Another mechanism, particularly relevant for
the presence of a d.c. bias of magnitude \( E \) determined by integrating the a.c. magnetic response obtained in
\( E \) via ferroelectricity, that is, in multiferroic systems, the electric field
induced by the electric field depends on the orientation of the
data presented in Fig. 3, the magnitude of magnetization changes
and modifies the trigonal distortion—as shown schematically in
\( c \)
explain magnetoelectric effects in various magnetic systems \textsuperscript{15,16}.

A number of physical mechanisms has been invoked in order to
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of the exchange coupling between magnetic ions \textsuperscript{17} or the
origin of this discrepancy is unclear.

**Discussion**

A number of physical mechanisms has been invoked in order to
explain magnetoelectric effects in various magnetic systems\textsuperscript{15,16}.
The proposed models include a variation of the carrier density at
the interface, due to gating. Such a change alters either the strength
of the exchange coupling between magnetic ions\textsuperscript{17} or the
occupancy of carrier subbands with a given orientation of orbital
momentum\textsuperscript{18–20}. Another mechanism, particularly relevant for
strong electric fields adjacent to a ferromagnetic metal/oxide
interface is the electromigration of atoms, which affects the surface
magnetic anisotropy\textsuperscript{21}. In magnetic compounds showing
ferroelectricity, that is, in multiferroic systems, the electric field
leads to an atom rearrangement that modifies the magnetization via
variations in magnetic anisotropy or spin-spin coupling\textsuperscript{2,13,16,22}.

We propose here, and support our conjecture with direct
quantitative computations that the changes in magnetization
generated by the electric field in \( Ga_{1-x}Mn_xN \) films are caused by
the inverse piezoelectric effect that alters the lattice parameter \( c \) and
modifies the trigonal distortion—as shown schematically in
Fig. 2b, c—and, thus, modifies the uniaxial magnetic anisotropy.

According to this model, and in agreement with experimental
data presented in Fig. 3, the magnitude of magnetization changes
induced by the electric field depends on the orientation of the
magnetic field in respect to the easy axis. A weak influence of the
electric field is expected for the magnetic field along the easy axis,
as in such a configuration \( M(H) \) follows approximately the
Brillouin function for \( S = 2 \) (see Fig. 1), meaning that it depends
only weakly on the trigonal distotion \( \xi \) and, thus, on \( c(E) \). By
contrast, the magnetic response for the hard configuration \( H \parallel c \)
varies strongly with \( \xi \), as it is much reduced for \( \xi \neq 0 \), and tends
to the Brillouin function if \( \xi \rightarrow 0 \), that is, when the uniaxial
magnetic anisotropy vanishes.

To quantify all our \( E \)–dependent results, we adopt the value of
the piezoelectric coefficient \( d_{33} = 2.8 \text{ pm V}^{-1} \), as obtained for a
clamped epitaxial GaN film\textsuperscript{23} since there is no lateral deformation
in thin films deposited on thick buffer layers. Having beforehand
parameterized \( M(T, H) \), as reported in Fig. 1, and knowing \( c(E) \) and,
thus, \( \xi(E) \) we are in the position to determine \( M_\parallel(H, T) \) with
no additional adjustable parameters. Theoretical results obtained
in this way are represented by solid lines in Figs 3–5.

As seen, the model proposed here gives a good account of all
experimental results as a function of the three experimentally
controllable parameters: \( H \), \( T \) and \( E \). The level of achieved
agreement indicates that the PEME dominates in the studied

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**Figure 4 | Temperature and magnetic-field dependence of magnetization generated by the electric field.** Areal density of the magnetic moment amplitude \( M_\parallel/A \) induced by electric field of the amplitude \( E = 0.66 \text{ MV cm}^{-1} \) (sample B). Inset: temperature dependence in 5 kOe (sample B). Points and solid lines: experimental and theoretical results, respectively.

**Figure 5 | Magnetization as a function of the electric field.** (a) Areal density of the magnetic moment amplitude induced by electric field \( M_\parallel/A \) in the magnetic field along the hard axis is determined in two ways: (i) by applying a low-frequency sinusoidal electric field (open symbols) or (ii) by modulating the d.c. electric field by an a.c. electric field of amplitude \( E_{\text{mod}} = 0.26 \text{ MV cm}^{-1} \) and displaying the integrated signal (closed symbols). This configuration is investigated at 2 and 5 kOe (sample A). Solid lines: theory. (b) Results for the easy axis configuration investigated at 5 kOe by the second method. The upper scales show the gate voltage \( V_G \) corresponding to the electric field \( E \).
Ga$_{1-x}$Mn$_x$N films. At the same time, it is clear that in some cases there is only qualitative agreement between experimental results and theoretical expectations (see, for example, high-field data in Fig. 3c, at 5 K in Fig. 4, and in high electric fields in Fig. 5). On the experimental side, as already noted, surface roughness detected by AFM may lead to lateral non-uniformities of the applied electric field, which can affect the behaviour of the magnetoelectric effect. It is also possible that the observed discrepancies indicate that the piezoelectric stretching not only changes the magnitude of trigonal distortion but also alters, for example, the Jahn-Teller effect or spin-spin coupling. Finally, the discrepancies may herald an onset of a contribution from a not yet identified phenomenon beyond PEME.

To put our findings in a more general perspective, it is worth recalling that dilute ferromagnetic semiconductors, such as Ga$_{1-x}$Mn$_x$As, have been playing a major role in seeding new concepts of spintronic devices and in developing our quantitative understanding of the interplay between magnetic and semiconducting properties. Among these concepts particularly far reaching is the demonstration of magnetization control by an electric field. In the present paper, this methodology has been extended to the wurtzite dilute magnetic insulator Ga$_{1-x}$Mn$_x$N, in which the Fermi energy is pinned by Mn ions in the mid-gap region and the Mn$^{3+}$ ions show strong single-ion anisotropy. By applying a new detection scheme, we have demonstrated magnetization control by the electric field via the inverse piezoelectric effect and developed a theory describing the PEHE quantitatively. In this way, our work bridges two fields of research developed so far independently: piezoelectricity of wurtzite III–V and II–VI semiconductors and electrical control of magnetization in hybrid and composite magnetic structures containing piezoelectric components.

According to the results presented here, the PEHE shows a strong increase for $T \rightarrow T_C$. Since $T_C$ grows rapidly with $x$ in Ga$_{1-x}$Mn$_x$N ($T_C \propto x^{2.2}$, refs 10,26), a further progress in the incorporation of Mn into GaN will shift the magnetization control by an electric field towards higher temperatures. It is worth mentioning in this context that in addition to the influence of the inverse piezoelectric effect upon single-ion magnetic anisotropy dominating in the case considered here, an impact of the electric field upon spin–spin interactions may dominate at higher Mn concentrations. While we have detected magnetization changes by direct magnetization measurements, spin-dependent tunnelling$^{27}$, spin–Hall magnetoresistance$^{28}$ and magnetooptical phenomena$^{29}$ may also serve to probe magnetization in magnetic insulators.

Another worthwhile outcome of the present work is the notion that the magnitude of the PEHE, or more pictorially the depth of the magnetic anisotropy modulation, which depends on $E$ and on the $d_{33}$ coefficient of the given material, will be the stronger the larger is the $\Delta \zeta(E) = \zeta(E) - \zeta(0)$ change with respect to $\zeta(0)$. An inversion of the magnetic anisotropy of the material (from in-plane to perpendicular, or vice versa) is expected when $\zeta$ changes sign, $\zeta(E)\zeta(0)<0$. As for many materials small magnitudes of dielectric strength or $d_{33}$ may preclude obtaining a sufficiently large $\Delta \zeta(E)$, advancements in material science may allow to engineer such a strain in layers with $\zeta(0) \geq 0$ and so the condition $\zeta(E)\zeta(0)<0$ could be met and the easy magnetization axis switching could be realized with accessible electric fields. For the Ga$_{1-x}$Mn$_x$N films studied here a use of compressive strain generated by an Al$_x$Ga$_{1-x}$N buffer would be a viable option.

Finally, we note that according to our results Mn doping is the method to turn GaN into a high-quality semi-insulating material that sustains a strong electric field. Importantly, Mn doping does not deteriorate structural properties, as shown in Methods, up to Mn concentrations $x \approx 3\%$ achievable by MOVPE. This is in contrast to other transition metals, such as Fe (ref. 30) and Cr (ref. 31) that tend to aggregate into transition-metal-rich nanocrystals for $x \lesssim 0.5\%$ (refs 32,33), the process destroying their carrier trapping capabilities. Similarly, heavy doping with C acceptors results in the dislocation formation and the appearance of self-compensating defects.

Methods

Atomic force microscopy. The GaN and Ga$_{1-x}$Mn$_x$N layers co-doped with Si are grown in an AIXTRON 200RF horizontal tube MOVPE reactor. Information on the morphology of the sample surface is obtained from tapping-mode AFM performed with a VEECO Dimension 3100 AFM system. The $40 \times 40 \mu m^2$ area scans for samples A, B and R are reported in Fig. 6. The root mean square surface roughness for samples A and B doped with a nominal Mn concentration of 3%, is 7.9 and 6.8 nm, respectively. For sample R with Mn concentration 0.1%, the surface roughness is estimated to be 5.5 nm.

High-resolution x-ray diffraction. HRXRD measurements have been carried out in a PANalytical X'Pert PRO materials research diffractometer equipped with a hybrid monochromator with a 1/4° divergence slit. The diffracted beam is collected by a solid state PixCell detector with a 9.1 mm anticounter slit. Radial $2 \theta$–$\omega$ scans between 30° to 80° are acquired along the growth direction of GaN [0001], covering the symmetric (0002) and (0004) Bragg diffraction peaks and are shown in Fig. 7a–c. The 20–$\omega$ scans confirm the high crystallinity of the epitaxial layers with no precipitation of secondary phases such as Mn$_2$N or SiN. The lattice parameters are calculated from rocking curve measurements of the symmetric (0002) and asymmetric (–105) Bragg peaks. Furthermore, the full-width half maxima of the symmetric (0002) and asymmetric (–1015) reflections are estimated to be 150–160 arcsec.

Secondary-ions mass spectrometry. The SIMS measurements have been performed on a Cameca IMS 6 F using either $^{133}$Cs$^+$ or $^{16}$O$_2^+$ ions, and are presented in Fig. 8 for samples A, B and R. The profiles confirm the nominal thickness of the Mn-containing layers (30 nm) and are consistent with the Mn concentration established from the saturation moment at 2 K (2.4 and 2.5% for samples A and B, respectively, as given in the main text).

Figure 6 | Atomic force microscopy surface imaging. Tapping mode images for all three studied layers.
High-resolution transmission electron microscopy. HRTEM measurements have been carried out employing a FEI Titan Cubed 80–300 scanning transmission electron microscope (STEM). High-angle annular dark-field images (top row of Fig. 9) show a contrast between GaN and the top Ga$_1$Co$_x$Mn$_x$N, whose thickness is evaluated to be 32 nm for both samples, in a good agreement with the nominal value of 30 nm. No hint of Mn aggregation is detected. HRTEM images (bottom row in Fig. 9), similarly to HRXRD, demonstrate the absence of crystalline phase separation in the studied layers.

SQUID magnetization measurements. Among the many factors limiting the credibility of magnetometry in nanomagnetism, exhaustively discussed previously, the presence of bulky substrates with a magnetic moment typically larger than those of the measured films is the most detrimental one. At first glance, the most appropriate approach calls for a direct subtraction of the magnetic response of the clear substrate at the matching fields and temperatures and then scaled according to its mass. However, such procedure yields a proper result only when the investigated sample and a reference have commensurate shapes as, actually, the real moment of the measured specimen is $m(T, H)/\gamma$, where $\gamma \approx 1$ is the coupling factor of a uniformly magnetized body with finite pick-up coils of a SQUID magnetometer. For a point object $\gamma = 1$. Table 1 in ref. 11 lists $\gamma$ values for some of the most typical sample shapes, indicating that actually it is the substrate’s highly nonsymmetrical shape that introduces the most significant error in the magnetic anisotropy data. Indeed, $(\gamma - 1)/\gamma \approx 5\%$ for a typical tile shaped samples, introducing an error easily exceeding the magnitude of the investigated anisotropy, particularly in strong magnetic fields.

In order to mitigate this challenge, both the original (5 × 5 × 0.3) mm$^3$ samples and a piece of matching sapphire substrate have been diced into four ~1.2 mm wide strips and glued to form a cuboid of a square cross-section, (1.2 × 1.2 × 5) mm$^3$, as illustrated in Fig. 10. In this way, both in-plane and perpendicular measurements can be carried out upon a simple rotation of the specimen by 90° along its length, thus preserving $\gamma$, and using the same sample holder, the procedure assuring the required reproducibility of the experimental conditions.
strongly compensated bulk crystals of very low Mn content but high O and Mg concentrations\(^6,35\) or to thick (unstrained) epitaxial layers of significantly smaller Mn contents\(^7\) than in our case. Therefore, as already pointed out in the main text, the parameter values are adjusted to reproduce the initial magnetization data (Fig. 1) for epitaxially strained epilayers with a relatively high Mn concentrations studied in our work. As explained in the main text, a particularly sizable modification is expected for the parameters \(H_V^i\) and \(H_G^i\), as they describe the trigonal deformation, so that their linear in \(\xi = c/a - \sqrt{3}/3\) dependence, \(y_G = y^G_0 + y^G_1 \xi\). Namely, we use: \(B_1 = 11.44 (11.44) \text{ meV} \quad \xi = -0.0058 \quad (0.0077)\), \(y_G = -550 \quad (550) \text{ meV} \quad y^G_0 = 73 \quad (73) \text{ meV}\), \(H_V^i = -5.9 \quad (5.1) \text{ meV}\), \(H_G^i = -1.2 \quad (1.02) \text{ meV}\), \(x^i_{\text{GaN}} = 5.5 \quad (5.0) \text{ meV}\), \(\lambda^i_{\text{GaN}} = 11.5 \quad (10.0) \text{ meV}\), and \(\lambda^i_{\text{Mn}} = 0 \quad (0)\), where the numbers in parentheses correspond to the very dilute Ga\(_{1-x}\)Mn\(_x\)N unstrained epilayers\(^8\).

In calculations of the PEEM, the value \(\xi(0)\) is obtained from \(\xi(0) = -0.0058 \) and the clamped value of \(a = 3.1842\) Å, known from the HRXRD measurements.

Data availability.

The authors declare that the supporting files of the studies are available from the corresponding author upon request.

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Author contributions
The work was planned and proceeded by discussions among A.B., M.S., and T.D. Samples growth by MOVPE and their structural characterization by AFM and HRXRD were carried out by R.A. and A.B. Atomic layer deposition and RIE were performed by K.K., R.K., and A.P., whereas R.J. and T.L. carried out SIMS and TEM studies, respectively. K.G. and M.S. performed SQUID magnetization measurements, whereas D.S., M.F., G.P.M. and M.S. processed the samples and made SQUID studies in the electric field with an input of M.Z. Analysis and interpretation of the data were accomplished by D.S. and T.D. The manuscript was written by D.S., M.F., G.P.M., A.B., M.S., and T.D. with inputs from all the authors.

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