Opto-spintronics in InP using ferromagnetic tunnel spin filters

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Abstract
We demonstrate opto-spintronics using Fe-doped Indium Phosphide (InP). The method is based on optical orientation of InP conduction electron spins which are electrically detected in planar InP/oxide/Ni tunnel spin filters. We separate the optical excitation from electrical detection, avoiding thus additional interactions of photons with the ferromagnet. Interface engineering provides a surface iron accumulation and semiconducting Fe:In$_2$O$_3$ in the oxide tunnel barrier. The spin filtering effect switches to positive or negative asymmetry, depending on the Fe concentration in Fe$_x$:InP. With respect to the Fe-like electronic structure of these oxides, we can explain the opposite spin selection mechanisms as interface effects. In the temperature region where the InP mobility peaks, we find a maximum of spin-dependent asymmetry of $\sim 9\%$ in semi-insulating Fe:InP (001), and show the electrical spin detection in hyperpolarized InP also at room temperature. Such robust electronic spin detection in an InP nanodevice is planned to complement dynamic nuclear polarization experiments.

1. Introduction
Controlling spin polarization in modern semiconductors (SC) is a major quest in spin-based electronics and research [1]. In SC, the creation of electronic spin polarization can be conveniently achieved by optical excitation [2], without the obstacle of spin injection across metal/SC interfaces [3]. Indium phosphide (InP) is a perfect SC for selective spin excitation by circular-polarized photons due to its direct band gap (1.34 eV [4, 5]). If an efficient electrical detection of spins in InP could be realized, given its superior electron mobility [6, 7] InP can ultimately combine the concept of energy-saving opto-spinelectronics [8] with high-speed switching in excess of 250 GHz [9]. In addition, recent dynamic nuclear polarization nuclear magnetic resonance (DNP-NMR) experiments use optically hyperpolarized InP [10–17], which evokes the need of an easy electronic monitoring of spin polarization and its seamless on-chip integration.

In order to detect electrically the optically excited spins in a SC, tunneling the spins into a ferromagnetic (FM) spin filter contact has been proven advantageous [18, 19]. In the case of GaAs as the SC, tunnel spin injection of spin-pumped electrons into various FM metals was demonstrated with so-called spin diode structures [20–22], and recently theoretically described by Li [23]. The working principle of the ‘spin diodes’ was based on the irradiation of photons through a homogenous closed GaAs/Schottky/FM layer structure [8, 22, 24–26]. Here, the electrical detection of spins takes place at the point of optical excitation, overlayed by interactions of photons with the FM [26]. An advancement was recently shown by Crooker et al [27] for GaAs in the separation of light and spin detection, using localized spin filtering in planar FM contacts [28, 29]. For InP, however, no results of electrical control of optically excited spins are reported to date.

In this study, we realize the electrical spin detection of optically hyperpolarized InP. In contrast to earlier studies [8, 20–22, 24–28], we focus on the usage of tunnel spin filters by inserting an oxide, and on the material Fe$_x$:InP (001)—the semiconductor with superior electronic mobility [7]. In order to achieve an easy electrical detection of optically excited spins in a robust InP wafer structure, we decided to use a laterally arranged four-terminal geometry of localized spin filter contacts. In this way, we combine the three advantages of (i) using the...
highly efficient optical orientation of spins rather than electrical spin injection through a metal-semiconductor interface, (ii) avoiding a non-local spin detection geometry which would sensitively depend on the contact distances in the nanometer range, and (iii) separating the photoexcitation from the electrical spin detection area and thus avoiding magneto-optical interactions of polarized photons with the FM metal layer. The lateral geometry is predicted not to hamper spin detection, since the spin relaxation time $\tau$ of InP amounts up to 2 ns [30]. This leads to a spin diffusion path of up to 200 $\mu$m in our experiment [31, 32]—so that the spin-polarized electrons will readily bridge the gap from their excitation to the electrical detection.

We deposit the FM spin filter contacts of Ni on top of Fe$_x$:InP (001), since Ni on SC has proven successful for spin detection [33, 34]. At the heart of the spin-sensitive heterostructures is an ultrathin oxide, yielding the tunnel structure InP (001)/oxide/Ni. This tunnel barrier features a potential height that matches the kinetic energy of the optically pumped electrons, and thus primarily transmits the spin-polarized electrons from the selected $\sigma^\pm$ photon excitation.

In order to demonstrate the electrical spin detection in Fe$_x$:InP ($x = 0$–0.2%), we select two complementary specimens: $n$-type InP ($x = 0$) and Fe:InP ($x = 0.2\%$). The first system is intrinsically semiconducting and established for fast electronics [9, 35]. The latter compound is carrier-compensated by iron ions which bind most free electrons from excess In ions. Thus, Fe:InP is a semi-insulating model system; it is suitable for optoelectronics but has a large sensitivity to inhomogeneous Fe doping [36].

To provide evidence for the electrical spin detection in hyperpolarized InP tunnel structures, we analyze the asymmetry of the differential resistance with respect to the helicity of the polarizing photons. For the two complementary Fe$_x$:InP tunnel structures, we identify different ranges of asymmetry with an overall maximum of 9%. We identify a sign switching of the detected spin asymmetry and discuss this feature in terms of Fe-like and Ni DoS in the spin filter tunnel barriers. Finally, we outline the integration of such InP spin sensors with NMR—hereby providing a simple probe of electronic spin polarization for optically pumped dynamic nuclear polarization (OP-DNP) experiments.

2. The InP opto-spintronics experiment

Single-crystalline InP (001) wafer pieces with different Fe doping concentrations were used. A transparent oxide layer was formed in dry air, which serves as spin conserving tunnel barrier. We chose Ar-sputtered polycrystalline Ni ($d = 200$ nm) as FM for the spin filter contacts, in order to magnetize the Ni contacts perpendicular to the substrate due to weak magnetic anisotropy. For a successful spin injection into the FM Ni contacts [1, 19, 37] through the InP/oxide interface, we verified the atomic smoothness of the InP surface and of the surface oxide. The oxide interface is 18 Å thick and mainly In$_2$O$_3$. The FM spin filter contacts are Au-covered to isolate them from photons and laterally arranged with a central gap of 500 $\mu$m (figure 1(a)) in order to conduct four-terminal measurements. Spin-polarized conduction electrons are spin-filtered in exactly one of the inner $U_{\text{high}}/U_{\text{low}}$ spin detection contacts, with respect to forward or reverse bias. The differential resistances
deduced from voltage vs. current (IV) sweeps (figure 2) reveal the spin asymmetry of the optically pumped InP. Such IV curves represent a robust and easy transport experiment and provide statistically reliable access to the electronic conductances induced by either $\sigma^+$ or $\sigma^-$ photons (figure 1(b)) [38]. The photons ($\lambda = 833.5$ nm, $\nu = 1.488$ eV) originate from a laser diode and are focused to a spot size $\lesssim 0.5$ mm which matches the free InP area between the spin detection contacts. The dimensioning of the gap and the laser spot permits the investigation of the optically pumped spins from the InP gap, while any electrical spin injection from the 500 $\mu$m-distant counterelectrode is negligible. The InP tunnel structures are cooled by liquid He in a flow cryostat inside a VARIAN Fieldial Mark II magnet.

3. Results and discussion

First, to provide evidence for the true nature of the spin filtering in the FM contacts, we study the asymmetry $R = (R_{\sigma^-} - R_{\sigma^+})/R_{\sigma^+}$ under variation of the external field $\mu_0 H_0$ in figure 3. The spin filter effect in the InP/oxide/Ni tunnel structures is directly proportional to the perpendicular induction of the FM Ni contact. Indeed, when reducing the external perpendicular induction field, the curves of the spin asymmetry resemble the out-of-plane magnetization behavior of polycrystalline Ni films as reported in literature [39, 40]. At $H_0 = 0$, the magnetization of the FM Ni relaxes to in-plane domains due to its shape anisotropy, and the observable asymmetry $R$ ($H$) vanishes. This null of the observable spin asymmetry is equivalent to a control experiment which switches the magnetic field by $90^\circ$ while the photon irradiation remains perpendicular. In this way, we can conclude the absence of magneto-optical effects and the true nature of the observed electronic spin detection for both Fe$_x$InP ($x = 0$–$0.2$%) spin detection heterostructures.

Using the complete perpendicular magnetization ($\mu_0 H_0 = 1.0$ T) in the following, we investigate the detectable spin asymmetry for the two complementary Fe$_x$InP (001) semiconductors with different Fe doping ($x = 0$ or $x = 0.2$%).

3.1. n-type semi-conducting InP (001)

First, we discuss the standard InP (001) n-type semiconductor without any charge carrier compensation ($x = 0$%). From established electron diffusion characteristics of n-type InP [30–32, 41], we can expect a dramatic change of the electrically detectable spin asymmetry as a function of temperature. The spin relaxation time exceeds the time of travel from the excitation to the detection of spins only for low temperature (table 1) by
one order of magnitude. Consequently, a spin detection is principally impossible ($\tau_{\text{spin}}/\tau_{\text{travel}} = 0.03$) at room temperature for the semi-conducting InP heterostructures.

We compiled the experimental results of the electrically detected spin asymmetries $\mathcal{R}$ in figure 4(a). The $n$-type InP/oxide/Ni tunnel structures exhibit asymmetries $\mathcal{R}(T)$ in the range $-0.2\%$ up to $+4.5\%$. Primarily, we observe the maximal spin asymmetry at $T = 52$ K in an eye-catching peak-like structure (blue graph). This peak can be traced back to an intrinsic property of the InP: the electronic carrier mobility (as investigated in figure 5) explains this main amplitude feature of the temperature dependent asymmetry behavior. Secondly, the asymmetry $\mathcal{R}(T)$ in figure 4(a) increases steadily in the region $T \to 0$, while $\mathcal{R} \leq 0$ at $RT$. This trend of an increased detectable spin asymmetry towards lower temperatures can be explained by a model beyond the electronic mobility peak of InP.

For this, we relate the energy of exciting photons with the band gap, $\delta E(T) = \hbar \nu - E_{\text{gap}}(T)$. Previous studies reveal the optical spin polarization (i) to vanish at $\delta E > 0.2$ eV, and (ii) to become even reversed at $\delta E > 0.3$ eV due to dominant excitation from the InP split-off bands [4]. Indeed, the band gap in semiconducting $n$-type InP is significantly reduced, leading to almost metallic conduction at $RT$. Hence, in the range $T \geq 90$ K, condition (i) explains the vanishing of the spin asymmetry; and at $RT$, condition (ii) is fulfilled which in turn coincides with the small negative asymmetry $\mathcal{R}$ (300 K).

Finally, the spin-dependent resistance asymmetry $\mathcal{R}$ elucidates the spin filter mechanism in the Fe$_x$InP/oxide/Ni tunnel structures: its sign is positive for the current specimen with no iron doping, $x = 0$ (figure 4(a)).
Following the optical orientation rules for III–V SC [4, 12, 13, 43–45], electronic spins in InP (001) are excited antiparallel to the impinging photon spin (figure 1(b)). If we consider $\sigma^+$ photoexcitation [38], then spin-up electrons pass the oxide barrier via spin-conserving tunneling and fall into minority spin states of the Ni conduction bands (figure 4(a) inset). This mechanism reveals the measured positive asymmetry in a straightforward manner. We confirmed the true nature of spin filtering by the magnetized tunnel contacts by a control experiment without perpendicular field: The in-plane relaxed contacts do not show any spin asymmetry in that case, similar to figure 3. Hence, in the $n$-type InP/oxide/Ni heterostructures, the established principle of spin filtering for a ‘SC/oxide/FM metal’ junction [1, 22, 33] is successfully realized.

### 3.2. Compensated semi-insulating Fe:InP (001)

In a further step, we investigate semi-insulating Fe:InP, in which free $n$-type carriers are compensated by iron ions ($0.15\%$, as determined by the mass spectroscopy ICP-OES) — in that yielding an effectively unpopulated conduction band. Hence, the signal of spin-pumped conduction electrons is expected to be significant over intrinsic electrons at the point of excitation. For the spin transfer to the spin detection contacts, literature on semi-insulating InP characteristics (table 1) reveals that the spin relaxation time exceeds the time of travel from the excitation to the detection of spins by at least 2 orders of magnitude for lower temperatures. At room temperature, $\tau_{\text{spin}}/\tau_{\text{travel}} \approx 1$ lets us expect a reduced yet electrically detectable signal of electronic spin asymmetry for the semi-insulating Fe:InP heterostructures.

![Figure 5](image_url) Photoconductance for $n$-type semi-conducting InP (a) and semi-insulating Fe:InP (b) tunnel structures as a function of the temperature. InP carrier mobilities from earlier studies [31, 32, 42] provide a comparison (c) of carrier densities (Blue curves are reprinted with permission from J Appl Phys 31, 2659 (1980) and Appl Phys Lett 52, 117 (1988). Copyright 1980 and 1988, AIP Publishing LLC [31, 32]). The conductivity peaks in our study corresponds to a high concentration of $n$-type carriers, consistent with the model of optical excitation of conduction electrons during laser pumping in InP.

### Table 1. Electron transfer times and spin relaxation times for the $n$-type InP and the Fe:InP planar spin sensors.

| $n$-type InP, semi-conducting | $T = 10$ K | $T = 70$ K | $T = 300$ K | Reference$^a$ |
|------------------------------|------------|------------|-------------|--------------|
| electron mobility $\mu$ (cm$^2$ V$^{-1}$ s$^{-1}$) | n.a.       | $2 \times 10^4$ | $3 \times 10^3$ | $[31, 32], n_t \approx 10^{12}$ cm$^{-3}$ |
| electron velocity $v$ (cm s$^{-1}$) | $8 \times 10^5$ | $6 \times 10^4$ | $1.6 \times 10^9$ | $\nu = \mu E = -\mu \frac{\partial v}{\partial x}$ |
| transfer time $\tau_{\text{travel}}$ (s) | $1.3 \times 10^{-11}$ | $1.6 \times 10^{-9}$ | $\tau_{\text{travel}} = \frac{\Delta s}{v}$ |
| spin relaxation time $\tau_{\text{spin}}$ (s) | $2 \times 10^{-9}$ | $52 \times 10^{-12}$ | estimated after [30] |

| Fe:InP, semi-insulating | $T = 10$ K | $T = 70$ K | $T = 300$ K | Reference |
|-------------------------|------------|------------|-------------|-----------|
| electron mobility $\mu$ (cm$^2$ V$^{-1}$ s$^{-1}$) | $100 \times 10^3$ | $300 \times 10^3$ | $5 \times 10^3$ | $[31, 32], n_t \approx 10^{13}$ cm$^{-3}$ |
| electron velocity $v$ (cm s$^{-1}$) | $2.0 \times 10^6$ | $6.0 \times 10^6$ | $0.1 \times 10^9$ | $\nu = \mu E = -\mu \frac{\partial v}{\partial x}$ |
| transfer time $\tau_{\text{travel}}$ (s) | $5 \times 10^{-11}$ | $1.6 \times 10^{-11}$ | $10 \times 10^{-10}$ | $\tau_{\text{travel}} = \frac{\Delta s}{v}$ |
| spin relaxation time $\tau_{\text{spin}}$ (s) | $\gg 2 \times 10^{-9}$ | $>2 \times 10^{-8}$ | $>0.6 \times 10^{-10}$ | estimated after [30] |

$^a$ $E$ is calculated using the $\Delta x = 500 \mu m$ gap between the voltage contacts. $\tau_{\text{travel}}$ is calculated using a travel path $\Delta s$ of one micron.
In the experiment, we are able to observe a spin-dependent asymmetry of $R (300 \text{ K}) = 4.5\%$ (figure 4b) even at room temperature. We further investigate the Fe:InP/oxide/Ni tunnel structures at two representative lower temperatures: (i) at the electronic mobility peak of Fe:InP ($T \approx 77 \text{ K}$ as determined in [46]) we observe a doubling of the spin asymmetry to $R = 9\%$, and (ii) at lowest temperature ($T = 10 \text{ K}$) $R = 6\%$ (figure 4(b)), a value 1.34 times larger than at $RT$. The prominent doubling (i) of the spin asymmetry at low temperatures is attributed to the electronic carrier mobility characteristic of InP [31,32,42] as evidenced in our photoconductivity measurements in figure 5(b). The trend (ii) of decreased asymmetry at higher temperatures can be explained by the energy difference between photons and the band gap $\Delta E (T) = h\nu - E_{\text{gap}} (T)$. For semi-insulating Fe:InP, $\Delta E (T)$ vanishes at $T \rightarrow 0 \text{ K}$ and amounts to $\sim 0.15 \text{ eV}$ at $RT$, in consistence with observations reported earlier [4,43]. In this scheme, the spin-polarized photoexcitation from heavy and light hole valence bands of InP will also include excitation from the split-off band having opposite spin [47], for a narrowed band gap and constant photon energy. As a consequence, the spin asymmetry after photoexcitation is reduced at larger $\Delta E (T)$ (e.g. at $RT$). Remarkably, since the energy mismatch $\Delta E (T)$ in Fe:InP is only half that of $n$-type InP, the asymmetry $R$ throughout exceeds 4.5\%.

For an understanding beyond the coarse amplitudes of the measured spin asymmetry, we now elucidate the characteristics of the tunnel spin filter contacts.

### 3.3. Spin filtering in the Fe$_2$InP/oxide/Ni contacts

At low temperatures, tunnel processes are expected at the heart of the Fe:InP (001) spin filter contacts. Hence, a chemical and morphological analysis of the oxide barrier helps to unravel the transfer of charge and spin. The surface oxides of InP (001) wafers are composed of mainly InPO$_4$ and In$_2$O$_3$ [48], which were shown to form smooth and closed films [49]. In particular In$_2$O$_3$ has been used as an electrical tunnel barrier [50]. We confirmed a vanishing interface roughness of the 18 Å-thick surface oxide on top of the Fe:InP (001) basis by x-ray reflection (figure 6(a)). A further characterization by photoemission spectroscopy (XPS) revealed a dominant fraction of In$_2$O$_3$ with an iron accumulation of 1.3\% [46] in the surface layers (figures 6(b), (c))—a ferromagnetic SC which may be spin-functional.

The actual behavior of a single tunnel junction (Fe:InP/In$_2$O$_3$/Ni) was characterized by using an ohmic contact to the InP wafer (figure 7 inset). Without any excitation, the pristine spin filter shows a tunnel current within 100 nA via field emission with the curvature of a Schottky contact (figure 7(a)). Indeed, the tunnel transmission in a pristine state is highly insulating due to the concatenation of the oxide barrier and a Schottky interface from the oxide to the FM metal. By a short laser pulse ($d \leq 1 \text{ s}$, $P = 0.3 \text{ W}$) and subsequent relaxation, we can diminish the insulating Schottky behavior by adding a fraction of thermionic (field) emission [51] and, in that, are able to record a full IV-characteristic as shown in figure 7(b). In order to characterize the tunnel junction by means of potential barrier height and thickness, we used the Brinkman model [52] which includes a non-uniform barrier height $\phi$ in the WKB approximation.

$$
\sigma_{\text{Br}} = \sigma_0 \left(1 - \frac{A_0 \Delta \phi}{16 \sqrt{3} \phi^3} \phi V + \frac{9A_0^2}{128 \phi} \phi V^2\right),
$$

with the conductivity $\sigma_0 = 3.16 \times 10^{-10} \text{ d}^{-1} \sqrt{\phi} \exp(-1.25d/\sqrt{\phi})$, $A_0 = \frac{4\sqrt{2m_e d}}{3\phi}$, and the barrier thickness $d$ and the asymmetry $\Delta \phi$. Fowler–Nordheim tunneling [53] $\sigma_{\text{FN}} \propto V \exp(-4d/\sqrt{2m_e \phi^3/4eV})$ was applied for larger bias voltages. Least-squares fits reveal a consistent result for the barrier height of $\phi = 0.19 \text{ eV}$ and...
thickness \( d \approx 18 \ \text{Å} \). The tunnel barrier permits preferably electrons to pass with \( \delta E_c \geq h \nu - E_{\text{gap}} - \phi \); these are the InP conduction electrons excited by photons of \( h \nu = 1.488 \ \text{eV} \). The transport regime is then mainly based on thermionic (field) emission [51] over the oxide barrier, and figure 7(c) depicts the successful passing of the spin-pumped kinetic conduction electrons. These spin-polarized electrons are subjected to spin filter processes in the FM contacts.

The sign of the measured spin asymmetry \( R \) is the key to unravelling the spin filter mechanism, which is switchable by the presence of Fe-doping (see figure 4(a) versus (b)). Considering the case of \( \sigma^+ \) excitation (figure 1(b)), spin-up conduction electrons in the surface layer of InP are expected to pass preferably through the tunnel barrier into available minority spin states of the FM nickel contact, similar to the \( n \)-type InP structures. However, the opposite effect emerges in the Fe:InP/oxide/Ni spin filters: spin-down electrons (excited by \( \sigma^- \) ) pass through a spin filtering process (figure 4(b)). We revisit, hence, the magnetic environment of the spin-polarized electrons: inside the semi-insulating Fe:InP wafer, the iron ions have been shown to provide mainly unoccupied minority spin states around \( E_F \) [54], comparable to bulk Fe. Next, the chemical composition of the tunnel barrier (Fe(1.3%):In2O3 after XPS analysis) coincides with iron-doped indium oxide—a semiconducting compound that exhibits, too, iron-like ferromagnetism [55–57]. In proximity to the Fe Ni contacts, these iron-like compounds will be ferromagnetically aligned \(^1\) colinear with the magnetized Ni. Therefore, the spin filtering takes place in the iron-rich surface of Fe:InP and subsequently in the Fe:In2O3 tunnel barrier (figure 4(b) inset). In summary, the iron-like majority conduction bands in the surface and oxide barrier induce the spin filtering and explain the negative sign of the asymmetry \( R(T) \) for Fe:InP.

3.4. Increasing the electrically detectable spin asymmetry

A possible development might be achieved by alternatives designs, e.g. as a vertical stack in a spin diode structure [58, 59]. However, such vertically stacked devices have the disadvantages of dichroism effects and larger reflection of the incoming photons. Both lateral and vertical structures would take benefit from a magnetic electrode with perpendicular magnetic anisotropy (PMA) because the devices could work without any external magnetic field.

Another parameter would be the tuning of the photon intensity for the lateral spin detection structures in this study. Conduction electrons in InP have to be excited by photons of energy \( h \nu \geq E_{\text{gap}}(T) \). This is accomplished by a commercial laser diode which provides an output power of up to 1.4 W and a wavelength \( \lambda = 833 \ \text{nm} \), this being near infrared radiation. Consequently, the photon excitation using the infrared laser is a localized heating process. Due to that heating, Seebeck voltages at the contact points of the InP wafer with the FM contacts and also with the bond wires readily occur in proximity to the laser spot. These effects can falsify spin-dependent resistivity measurements, unless a thermal equilibrium can be maintained and the heat creation is minimized. In figure 8, we recorded the photoconductivity effect of the InP tunnel structures in the study at hand under variation of the laser power. The conductivity increases linearly beginning from a laser power of \( 0.1 \ \text{W} \). In our helicity dependent experiments, we limit the laser output power to 0.3 W, this being clearly in the significant photoconductivity regime yet only one fifth of the maximal laser output. In this regime we can achieve reliable spin asymmetry measurements in quasi-equilibrium with vanishing Seebeck effects.

We remark that by using larger laser pumping intensities, we would expect to electrically detect linearly increased photogenerated currents carrying the spin asymmetry information. Whether these enhanced signals provide larger spin asymmetries \( R \) is subject to current research. In addition, various Seebeck effects and heat dynamics would give rise to a variety of dynamic phenomena referred to as (spin-)caloritronics [60].

\(^1\) FM alignment due to direct FM exchange, since any possible NiO formation with exchange bias can be excluded after thermodynamic considerations.

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**Figure 7.** Characteristic \( IV \) curves reveal the electronic conduction of a single Fe:InP/oxide/Ni tunnel junction at \( T = 290 \ \text{K} \). The junction in dark pristine state (a), after a pump pulse fitted with a Brinkman model (b), and during persistent laser spin pumping (c).
3.5. InP opto-spintronics for dynamic nuclear polarization NMR

The optical excitation in conjunction with the electronic detection of spins in InP could lead to helicity-sensitive opto-spintronics devices integrated with established fast InP electronics. However, the main application idea is the electronic monitoring of spin hyperpolarization in optically excited InP (sketched in figure 9(a)), which we illustrate here. One way of boosting the inherently small NMR signal intensities by several orders of magnitude is the optically pumped electronic spin polarization, as explored in this study. The key feature of InP is the immediate transfer of electronic spin asymmetry to the nuclei via indirect hyperfine interactions, referred to as dynamic nuclear polarization (DNP) (figure 9(b)). Using nuclear hyperpolarization in InP obtained through optical pumping experiences increasing interest with promising applications to spintronics [61, 62], quantum computing [15–17], and NMR [10–14]. The selection of InP rather than GaAs has the advantage for DNP applications that the presence of the $I = \frac{1}{2}$ isotope $^{31}$P with 100% natural abundance makes InP a preferable candidate for biological DNP applications [10]. For future DNP applications, high nuclear polarization at the surface of the InP crystal is the key to hyperpolarize a material of interest in direct contact with the InP surface. While the polarization transfer to the P nuclei has its origin in various mechanisms based on the hyperfine contact interactions [63], they all depend on the initial spin polarization of the electronic system in InP, which needs to be monitored by a suitable technique.

As an easy-to-implement method, the spin filter detection of electronic spin polarization in InP in this study can perfectly complement upcoming optically pumped DNP experiments which use InP as hyperpolarized agent.
4. Conclusion

We succeeded in the electrical detection of optically pumped spins in InP (001) structures via tunnel spin filter contacts. Planar InP/oxide/Ni tunnel structures were designed which separate the optical spin excitation from the electrical detection and hence avoid magneto-optical effects. We unraveled different spin-selective mechanisms in semi-conducting n-type InP (001) and semi-insulating Fe:InP (001) tunnel structures: in n-type InP, the FM contact filters spins due to electron acceptance in the minority conduction bands of Ni; while on top of Fe:InP structures, a semiconductor with iron accumulation (Fe:In$_2$O$_3$) is formed which spin filters via the majority bands of Fe. These effects switch the electrical spin detection by variation of the Fe doping.

Maximal asymmetries of the spin-dependent resistance of $R = 9\%$ could be observed at $T \approx 80$ K. Even at room temperature, $R \approx 5\%$ demonstrates the applicability of the electrical tunnel spin detection in spin-pumped InP.

These spin-sensitive InP tunnel structures propel the field of opto-spintronics and may initiate the future integration of spin-functionality with high-speed InP-based electronics. For this, further research on the bias, temperature and field behaviors is motivated.

Moreover, such easy and robust electrical detection of electronic spin polarization in InP is a valuable complement to NMR [64], which are based on the $^{31}$P dynamic nuclear polarization of optically pumped InP.

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See the supplementary information.