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Room temperature electrical spin injection into GaAs by an oxide spin injector

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Spin injection, manipulation and detection are the integral parts of spintronics devices and have attracted tremendous attention in the last decade. It is necessary to judiciously choose the right combination of materials to have compatibility with the existing semiconductor technology. Conventional metallic magnets were the first choice for injecting spins into semiconductors in the past. Here we demonstrate the electrical spin injection from an oxide magnetic material Fe_3O_4, into GaAs with the help of tunnel barrier MgO at room temperature using 3-terminal Hanle measurement technique. A spin relaxation time $\tau \sim 0.9$ ns for n-GaAs at 300 K is observed along with expected temperature dependence of τ. Spin injection using Fe_3O_4/MgO system is further established by injecting spins into p-GaAs and a τ of ~0.32 ns is obtained at 300 K. Enhancement of spin injection efficiency is seen with barrier thickness. In the field of spin injection and detection, our work using an oxide magnetic material establishes a good platform for the development of room temperature oxide based spintronics devices.

Ever since the proposal of spin field effect transistor by Datta and Das in the year 1990[1], the research on spin injection and detection has been one of the central themes in condensed matter physics and materials science[2–6]. Initially, optical methods of spin injection and detection in semiconductors[7,8] were studied extensively by several groups; but, later on as the research progressed in this field, electrical injection methods were attempted and became successful[9–11] though detection techniques still remained as optical. The non-local electrical measurements on metallic spin-valves by Jedema F. J. *et al*[12] in 2002 demonstrated that both spin injection and detection can be carried out completely by electrical methods, which proved to be more efficient because of the ease of integration and practical usage of these devices.

However, in case of spin injection into semiconductors, lack of sufficient spin injection efficiency due to the conductivity mismatch between the metallic magnets and the semiconductors were the initial hurdles encountered by the research community. Hence, overcoming the conductivity mismatch[13,14] between the spin injector and the semiconductor or using an injection material with high spin polarization would be the efficient ways of injecting spins into semiconductors. It is now established that one can inject spins into conventional semiconductors such as Si, Ge, and GaAs *etc.* using ferromagnetic electrodes such as CoFeB, Co, NiFe *etc.* through a tunnel barrier[4–6]. Recently, J. Bae *et al.*[15] also observed electrical spin injection into GaAs using perpendicularly magnetized metallic magnetic layers using MgO barrier.

Of late, oxide magnetic materials are emerging as potential candidates for substituting the established metallic spintronics devices because of their versatility and flexibility in tuning the properties which are essential for fabricating superior devices[16–18]. Hence, it is perceived that oxide spintronics may play a vital role in the next generation electronics. In this regard, spin injection into an oxide and from an oxide need to be realized as it is crucial for the development of oxide spintronics. Recently, Wei Han *et al.*[19] showed that it is possible to inject spins from metallic CoFe into oxide materials such as La doped or Nb doped $SrTiO_3$. Now what remains to be addressed is spin-injection from an oxide magnetic material. In this regard, room temperature magnetic oxides like Fe_3O_4 and LSMO ($La_{1-x}Sr_xMnO_3$) would be the first choice, since they have the advantage of theoretically predicted half metallic nature which makes them perfect candidates for efficient spin injection due to high spin polarization. Stable room temperature ferrimagnetism and high ferrimagnetic Curie temperature (~860 K) of Fe_3O_4 make it more attractive from application point of view in addition to the half metallicity. Moreover, spin-polarized photoelectron spectroscopy experiment on epitaxial Fe_3O_4 thin films supports the theoretical prediction of half-metallicity with measured spin polarization $\sim 80 \pm 5\%$[20]. In the present study we benchmark the spin injection from Fe_3O_4 into the well studied conventional semiconductor GaAs using MgO as

tunnel barrier. In GaAs, the spin relaxation studies have been carried out by various groups in the past from the optical as well as electrical spin injection point of view. Due to the non-centro symmetric crystal structure of GaAs, an additional field in the lattice similar to magnetic field of spin-orbit interaction influences the spin scattering (D'yakonov-Perel mechanism[21]) more apart from the usual momentum scattering and impurity scattering when compared with centro symmetric crystals like Si. From earlier reports it is seen that all electrical spin injection and detection using in-plane magnetized ferromagnetic systems in GaAs is observed only up to ~120 K[2–4,22,23]. In spite of these facts, we have demonstrated spin injection and detection in in-plane magnetized Fe_3O_4/MgO/n-type GaAs and p-type GaAs devices at room temperature. In the recent past, T. Saito *et. al.*[24] and J. Bae *et. al.*[15] were also successful in observing spin signal in GaAs at room temperature by electrically injection method with the help of highly spin polarized material like Heusler alloy and perpendicularly magnetized layer in combination with MgO barrier respectively. Since we have used an oxide magnet for the demonstration of spin injection, our efforts with spin injection and detection using an oxide material will be of importance in establishing these oxides for the future spintronics devices.

In the recent years, electrical 3-terminal Hanle technique (as depicted in Figure 1) has attracted a lot of attention since it is one of the simple yet powerful techniques to detect spins in semiconductors. Figure 1(a) represents the schematics of the 3-terminal measurement geometry with the device at the centre being common for one of the current as well as voltage lead. Unlike the four terminal measurements, this technique directly probes the spin injection right at the Fe_3O_4/MgO/GaAs interface[5]. A constant injection current will help in accumulating these injected spins at the interface between the semiconductor and the barrier. The magnitude of these accumulated spins is given as $\Delta\mu = \mu_{\uparrow} - \mu_{\downarrow}$, where $\mu_{\uparrow}$ and $\mu_{\downarrow}$ are the chemical potentials corresponding to the majority and minority carriers respectively. These accumulated spins precess within the semiconductor as a function of perpendicular magnetic field leading to the decay of the accumulated spins (shown in Figure 1b) with its functional form as a Lorentzian, $\Delta\mu(B_z) = {}^{\Delta\mu(0)}/_{1+(\omega_L\tau)^2}$, where ω_L is Larmor frequency of precession of spins, it is defined as $\omega_L = {}^{g\mu_B B_z}/_{\hbar}$, with Lande's g-factor value of GaAs to be −0.44 and τ being the effective spin relaxation time. The magnitude of the accumulated spin is measured as ΔV, which is related to $\Delta\mu$ as $\Delta V = {}^{TSP\times\Delta\mu}/_{2}$, where TSP is the tunnel spin polarization of the Fe_3O_4/MgO combination.

Results

Fe_3O_4 (a_{Fe3O4} = 8.396 Á) also has an interesting low temperature transition at 120 K known as the Verwey transition[25], which is a first order transition with structural, magnetic, electrical ordering within the material[26]. Growth of Fe_3O_4 and MgO on GaAs system follows a perfect epitaxial relation Fe_3O_4 (100) [011]//MgO (100) [001]//GaAs (100) [001][27,28] in addition to all the attractive features listed above. With a_{MgO} = 4.212 Á, the lattice mismatch between Fe_3O_4 and MgO is only about only 0.33% because of the 45° in-plane rotation growth of Fe_3O_4. In our study, a stack of Fe_3O_4 and MgO thin films were grown on Si- doped n-type GaAs with 1×10^{18}/cm^3 doping and Zn-doped p-type GaAs with 1×10^{19}/cm^3 (see the methods for sample preparation). SEM micrograph of the typical Hanle structure is as shown in Figure 1(c). The quality of Fe_3O_4/MgO/GaAs interface has been verified with TEM, and the typical image is shown in Figure 1(d). From the X-ray diffraction measurements, it is seen that the growth of Fe_3O_4/MgO on GaAs is oriented (Figure S1 of supplementary material) and epitaxial. Figure S2 of supplementary material further confirms the Fe_3O_4 film growth and the absence of satellite peak at 719 eV which confirms the absence of Fe_2O_3. Also, the mag-

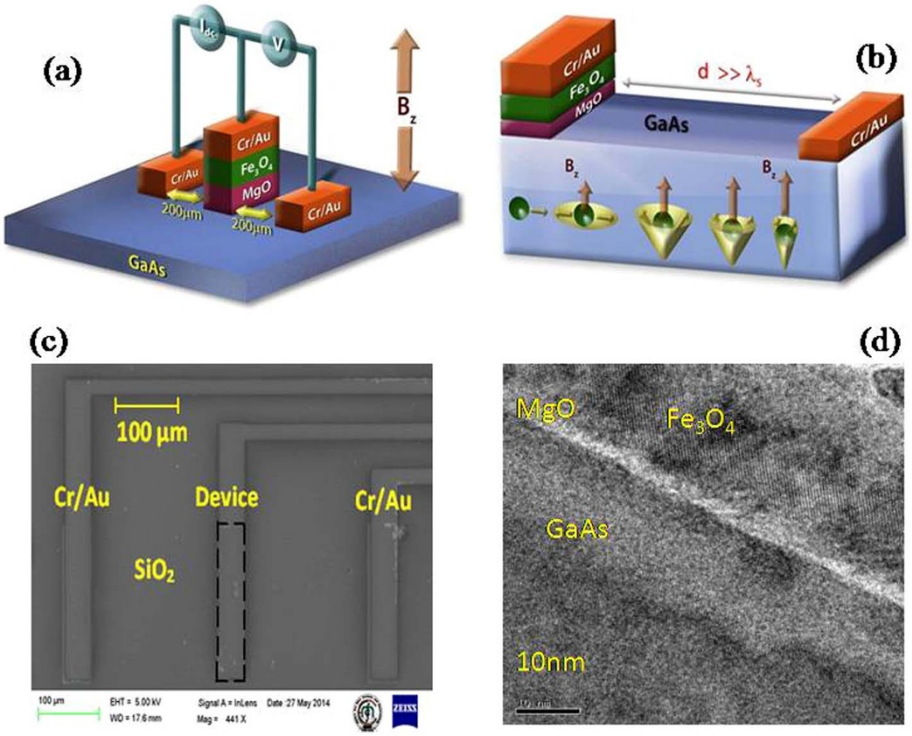

Figure 1 | Schematics of 3-terminal Hanle measurement geometry. (a) The structured devices of dimension 50 × 200 μm^2 are separated by d = 200 μm distance between the contact pads with $d \gg \lambda_s$. (b) With constant DC current I_{dc} flowing, accumulated $\Delta\mu$ precesses about B_z. As the strength of the applied field increases, the accumulated spins get dephased and relax close to the direction of the applied magnetic field. (c) Typical SEM image of the Hanle structure. (d) TEM micrograph of the Fe_3O_4/MgO/GaAs film.

netization as a function of temperature (Figure S3(a) of supplemental material) of the film shows a very sharp Verwey transition and also the log R vs. T plot as shown in Figure S4 confirms the superior quality of Fe_3O_4 film.

Spin injection into n-GaAs using Fe_3O_4. Investigation of spin injection and accumulation into n-GaAs using Fe_3O_4/MgO injection system is discussed in detail at first. Figure 2 shows the transport measurements carried out on the photlithographically structured 50 × 200 μm² devices of Fe_3O_4/MgO/n-type GaAs and the Hanle measurements results are plotted in Figure 2(a), (b) and (c). We have observed ΔV of ~0.35 mV for an injection current of 1 μA. From Figure 2(a), it is seen that the accumulated spins decay as the strength of B_z increases. Lorentzian fit to the curve yields a τ value of ~0.9 ns. This is a lower bound value of the spin relaxation time due to the broadening of the Hanle curve, as discussed by Dash S. P. *et al*[22]. At higher fields, the magnitude of ΔV increases again because of the rotation of the magnetization axis to out of plane for Fe_3O_4 layer. The spin resistance ΔR for the corresponding ΔV is calculated as ΔR = ΔV/I. The spin resistance-area product $\Delta R \times A$ in kΩ μm² vs. B_z for different injection currents are plotted in Figure 2(b) so that the magnitude of spin signals can be compared clearly. The FWHM of the curves at different injection currents remain same which clearly indicate the genuineness of the τ value extracted. Hanle measurements were repeated at 80 K and the data is shown in Figure 2(c). It is seen that the τ value has increased to ~1.6 ns with a huge ΔV of ~18 mV. A systematic study of τ extracted as a function of temperature is plotted in Figure 2(d). Since the resistivity of Fe_3O_4 becomes very high at lower temperatures (T<70 K), measurements are carried out at temperatures T ≥ 80 K. As expected, the value of τ decays from 1.6 ns to 0.9 ns from 80 K to 300 K due to the increased lattice scattering.

Spin injection into p-GaAs using Fe_3O_4. Figure 3(a) shows the plot of ΔV vs. B_z of Fe_3O_4/MgO/p-GaAs device of area 50 × 200 μm² at 300 K. I-V measurements on the sample at two different temperatures are plotted in Figure S5 of supplementary material. We have observed the accumulation of spins even in p-GaAs, and this is evident from a ΔV as high as 5 μV for 5 μA extraction current at room temperature in GaAs as shown in the inset of Figure 3(a). It can be seen that though the accumulation of spin are about 2 order lower than that of n-type, still a detectable injection of spins and the decay of spin is observed. Figure 3(b) is the plot of ΔR vs. B_z for different extraction currents. As B_z increases, dephasing results in the decay of the accumulated spins under the Fe_3O_4/MgO layer. The value of the hole spin relaxation time from the Lorentzian fit at room temperature is found to be ~0.32 ns; (assuming that the g value for holes to be 0.44, same as for electrons). Also the measurements at 80 K yield a systematic variation of the Hanle data for the junction. A spin relaxation time of ~0.6 ns was obtained from the Lorentzian equation mentioned above which is almost double of the value of τ at room temperature. In Figure 3(d), the $\Delta R \times A$ product of the junction at 2 different temperatures is compared. As the temperature is increased from 80 K to room temperature, there is a clear diminution in spin relaxation time to about 0.6 ns from 0.32 ns. It is quite understandable that the spins get relaxed more because of the increased lattice scattering due to temperature factor. A plot of the relaxation time as a function of extraction current at 300 and 80 K is given in the Figure S6 of the supplementary material.

It has to be noted that the obtained value of ΔV is weak for p-GaAs when compared with n-GaAs. Hence, in order to check the authenticity of the values obtained, we have done further measurements on different samples by varying the MgO barrier thickness. Figure 4 shows Hanle data for Fe_3O_4/MgO (x= 1, 1.5, 3 nm)/p-GaAs at room

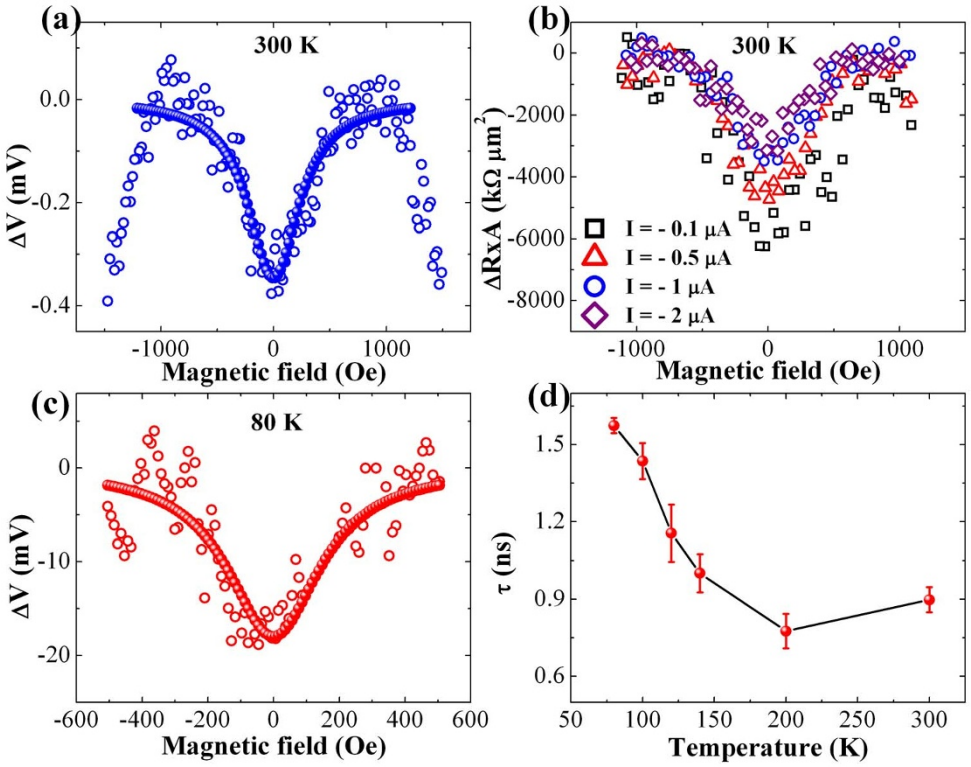

Figure 2 | Electrical Hanle measurements on Fe_3O_4/MgO/n-GaAs. (a) Hanle data of the film (circular open symbols) for 1 μA electron spin injection current at 300 K. Plot of ΔV vs. B_z shows a spin accumulation of ~0.35 mV. (b) $\Delta R \times A$ (kΩμm²) vs. B_z for different electron spin injection currents for 50 × 200 μm² area devices. (c) ΔV vs. B_z plot at 80 K is observed to be having lesser FWHM in comparison to 300 K data. (d) Spin relaxation time τ of n-GaAs as a function of temperature (circular solid symbols). The line drawn is just guide to eye.

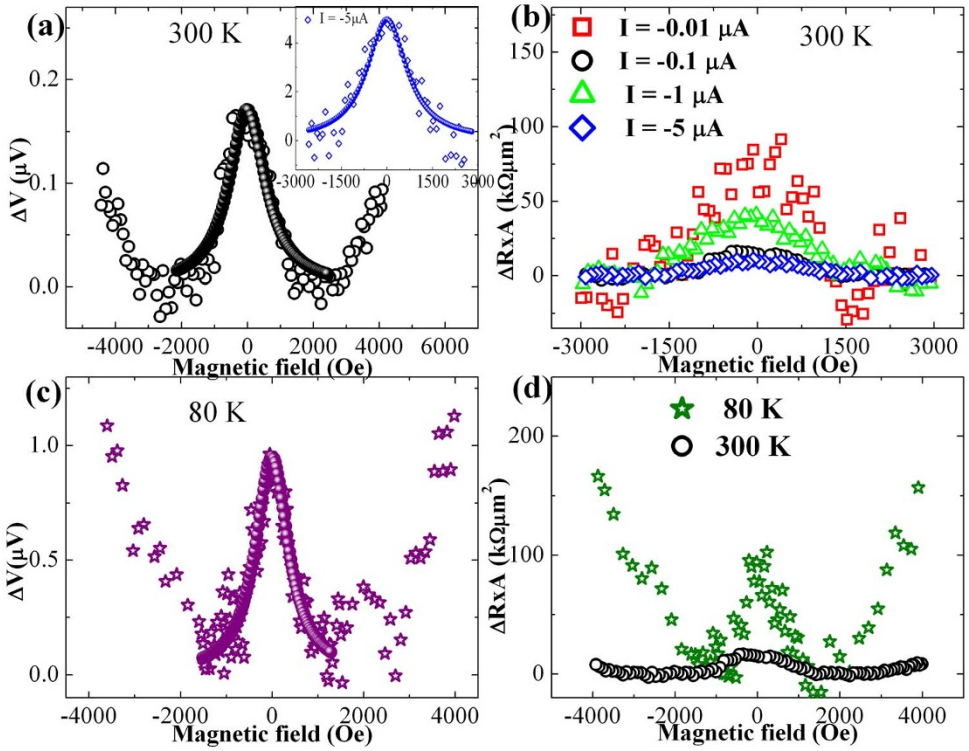

Figure 3 | Electrical Hanle measurement on Fe_3O_4/MgO/p-GaAs devices. (a) Plot of ΔV vs. B_z at 300 K at 0.1 μA (circular open symbols) with Hanle fit (circular solid symbols). Inset shows a maximum spin accumulation of 5 μV for an electron injection current of 5 μA (diamond open symbols). (b) Plot of $\Delta R \times A$ product (kΩμm²) vs. B_z at 300 K at various hole extraction currents for the devices with 50 × 200 μm² area. (d) Plot of ΔV vs. B_z at 80 K at 0.01 μA (star open symbols). (e) Comparison of $\Delta R \times A$ product (kΩμm²) at 80 and 300 K (star open symbols and circular open symbols respectively) at extraction current of 0.1 μA with different FWHM for the curves.

temperature. It is seen from the Figure 4 that the FWHM for different thickness of MgO remains almost similar indicating that there is no change in τ values extracted individually for these different samples. In all the cases, the average value of τ is ~0.29 ± 0.03 ns. Hence, the spin relaxation time obtained for p-GaAs is free of any extrinsic effects of the junction.

Discussion

From the demonstration of the spin injection in GaAs at room temperature as well as at low temperatures, we can see that the Fe_3O_4/MgO oxide system is highly efficient for the spin injection studies. We obtain a ΔV of ~0.3 mV at 300 K which is in fact comparable to ΔV obtained using 3TH measurements in Si[5,6,29] than ΔV in GaAs[15,30,31] and at low temperatures, ΔV as high as ~18 mV is observed. Apart from having a high spin polarization from Fe_3O_4, the spin injection and detection at room temperature as well as at low temperatures in GaAs might be attributed to the coherent spin tunneling due to the presence of symmetry states in MgO[32]. The Δ symmetry bands of MgO is known to be preserving the spins from scattering within MgO, and we believe that this could be the prime factor for observing a spin signal of ~mVs in GaAs in spite of its strong spin scattering near room temperature. One of the recent reports also support this fact where in similar scenario of MgO is discussed with the perpendicularly magnetized metallic layer[15]. This shows that the presence of MgO in this spin injection system is necessary to inject as well as detect the spins in GaAs at room temperature. In p-GaAs it is seen that the magnitude of ΔV observed increases from ~0.15 μV to ~0.8 mV with the barrier thickness. Hence it is clear that the increase in junction resistance improves the value of accumulated spin. Increase in the symmetry filtering effect introduced by MgO for increased thickness of MgO cannot be fully ruled out in this context.

In summary, we demonstrate the spin injection into the conventional semiconductor GaAs using an oxide magnetic material, Fe_3O_4, with the help of a tunnel barrier. The spin relaxation time for n-GaAs is found to be ~0.9 ns at room temperature. The temperature

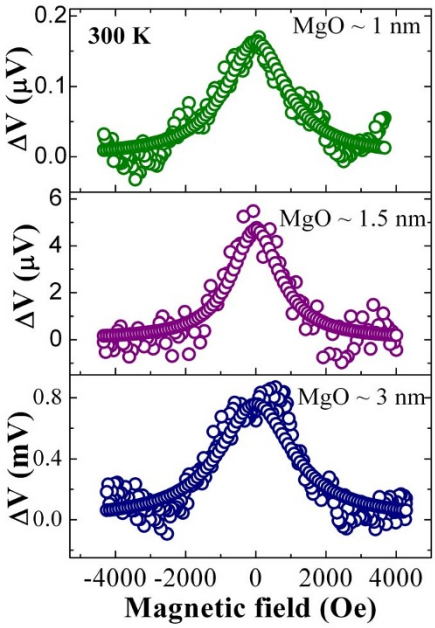

Figure 4 | Electrical Hanle measurement on Fe_3O_4/MgO (x = 1, 1.5, 3 nm)/p-GaAs devices. With different MgO barrier thickness, the FWHM of the curves obtained for different devices are of the same order of extracted spin relaxation time. The value of τ averages out to be 0.29 ± 0.03 ns.

dependent study of Hanle signal indicates the increment in τ value for decreasing the temperature as expected. Also, the demonstration of spin injection in p-GaAs strengthens the fact of high injection efficiency of Fe_3O_4/MgO combination. A spin relaxation time of ~0.32 ns is obtained for p-GaAs at room temperature, whereas there is a clear enhancement in the τ value to ~0.6 ns at 80 K. Hanle measurements for different thickness of MgO barrier on p-GaAs at room temperature also indicates the same relaxation time of ~0.29 ± 0.03 ns, ruling out the extrinsic effect of observing the signal in p-GaAs. All these experiments establish Fe_3O_4/MgO combination as a very good spin injection system. We thus conclude that even oxide magnetic materials are also equally attractive, promising, and capable of establishing a field of spin injection and detection in the area of oxides.

Methods

Sample preparation. Pulsed laser ablation technique is used to deposit the film stack of Fe_3O_4/MgO on GaAs. Substrate is Zn- doped p-type GaAs with a carrier concentration specified from the manufacturer to be $1 \times 10^{19}/cm^3$. Prior to the deposition, substrate was cleaned with acetone and IPA. Substrate was subsequently dipped in 1% HF in order to remove the native oxide on GaAs. Then the substrate was loaded into the deposition chamber and annealed at 500°C to get uniform and clean surface of GaAs. Different thicknesses of MgO (for different samples) were deposited at 500°C followed by a deposition of 30 nm of Fe_30_4 at 450°C. The post annealing of the film was carried out at 450°C for an hour in vacuum of the order 10^{-5} m bar.

Sample fabrication. In order to get 3-terminal Hanle geometry for the transport measurement, these continuous films were structured using photolithography followed by Ar ion beam etching. About 100 nm of SiO_2 layer was deposited for the isolation of the defined device area. Then a 2nd level of photolithography and deposition were carried out to define the contact pad, Cr (10 nm)/Au (100 nm). Al-wedge bonding was used to connect the electrical leads to these devices.

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Author contributions

S.G.B. performed the experiments and the subsequent measurements. P.S.A.K. supervised and coordinated the project and contributed towards the insights and ideas of the project. S.G.B. prepared the manuscript with inputs from P.S.A.K.

Additional information

Supplementary information accompanies this paper at http://www.nature.com/scientificreports

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CORRIGENDUM: Room temperature electrical spin injection into GaAs by an oxide spin injector

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The following sentence was inaccurate and has been removed from the abstract: "So far there is no success in using a magnetic oxide material for spin injection, which is very important for the development of oxide based spintronics devices."

The following sentence in the introduction was inaccurate: "From earlier reports it is seen that spin injection using in-plane magnetized ferromagnetic systems in GaAs is observed only up to ~120 K[2–4, 22, 23]." It has been changed to: "From earlier reports it is seen that all electrical spin injection and detection using in-plane magnetized ferromagnetic systems in GaAs is observed only up to ~120 K[2–4, 22, 23]."