Spin Injection From EuS/Co Multilayers Into GaAs Detected by Polarized Electroluminescence

A. Goschew[1], R. C. Roca[2], N. Nishizawa[2], H. Munekata[2], A. Delimitis[3,4], and P. Fumagalli[1]

[1]Department of Experimental Physics, Freie Universität Berlin, 14195 Berlin, Germany
[2]Imaging Science and Engineering Laboratory, Tokyo Institute of Technology, Yokohama 226-8503, Japan
[3]Chemical Process and Energy Resources Institute, Centre for Research and Technology Hellas, 57001 Thessaloniki, Greece
[4]Department of Mechanical and Structural Engineering and Materials Science, University of Stavanger, N-4036 Stavanger, Norway

We report on the successful spin injection from EuS/Co multilayers into (100) GaAs at low temperatures. The spin injection was verified by means of polarized electroluminescence (EL) emitted from AlGaAs/GaAs-based spin-light-emitting diodes in zero external magnetic field. Spin-polarized electrons were injected from prototype EuS/Co spin injector multilayers. The use of semiconducting and ferromagnetic EuS circumvents the impedance mismatch. The EL was measured in side emission with and without an external magnetic field. A circular polarization of 5% at 8 K and 0 T was observed. In view of the rather rough interface between the GaAs substrate and first EuS layer, improvement of the interface quality is expected to considerably enhance the injected electron spin polarization.

***Index Terms*—Electroluminescence (EL), EuS, magnetic semiconductor, spin-LED.**

I. INTRODUCTION

A MAJOR working field in spintronics covers efficient spin injection and transport in spintronic devices [1]–[6]. Usually, spin injection means the injection of spin-polarized electrons from an injector site (usually a ferromagnetic metal) into a semiconductor (Si or GaAs). The straightforward approach of growing a magnetic layer on top of a semiconductor yields only an insignificantly small degree of spin polarization in the injected current [7], [8]. The reason for this was found in the mismatch of conductivities between metals and semiconductors which differ by a few orders of magnitude [9], [10]. This so-called impedance mismatch prevents spin injection for directly connected devices. Rashba [11] proposed tunnel contacts as a means to circumvent this problem. This has indeed proven to be a successful means of efficiently injecting spin-polarized electrons into semiconductors [12]–[16].

Instead of using ferromagnetic metals as an injection source, ferromagnetic semiconductors like EuS might also present suitable candidates for spintronic devices. Using a semiconductor would considerably reduce the impedance mismatch.

EuS is an intrinsic ferromagnetic semiconductor with a bandgap of 1.6 eV, a magnetic moment of 7 μ_B, and a Curie temperature (T_C) of 16.5 K [17]. This is a considerable drawback for this material as it would otherwise be an ideal candidate for creating spin-polarized currents. The energy splitting in the conduction band is about 0.36 eV below T_C [18]. This makes EuS in its ferromagnetic state a good candidate for efficient spin filtering [19]–[22].

Recent experiments have shown that the EuS bandgap can be tuned using quantum confinement effects which could simplify the use of EuS-based devices together with other semiconductors [23]. In addition, it was found that its Curie temperature could be raised considerably by embedding EuS nanoparticles in a Co matrix [24] and even further when using multilayers of thin EuS sandwiched between Co or Ni. Element-specific X-ray circular magnetic dichroism measurements have confirmed that EuS/Co and EuS/Ni multilayers still show a spin-polarized Eu signal at room temperature (RT) [25]–[27]. The effect is larger when using Co compared to Ni but comes at the cost of more interfacial roughness between EuS and Co compared to EuS and Ni. In such multilayer systems, EuS couples antiferromagnetically to Co or Ni, which leads to an increase in the ordering temperature of EuS. The enhancement of the EuS ordering temperature is attributed to a "magnetic proximity effect." An explanation of the effect can be found in [28].

The idea of this paper is to use commercial spin-LED substrates to grow one or more bilayers of EuS/Co on top. The degree of optically measured circular polarization, P_c, is connected to the degree of spin polarization in the injection site, P_{el}, by the simple formula

$$P_c = \frac{I^+ - I^-}{I^+ + I^-} = \frac{1}{2}\frac{n_\uparrow - n_\downarrow}{n_\uparrow + n_\downarrow} = \frac{1}{2}P_{el} \tag{1}$$

where I^+ and I^- are the intensities of right and left circularly polarized light, while $n_\uparrow$ and $n_\downarrow$ are the densities of spin-up and spin-down electrons, respectively.

This formula is valid for bulk semiconductor LEDs without quantum wells, where the degeneracy between light and heavy holes is not lifted. In LEDs using narrow quantum wells, the easy axis of the angular momentum of the heavy holes is perpendicular to the sample surface and the energetic degeneracy of light and heavy holes is lifted [29], yielding $P_c = P_{\mathrm{el}}$. Thus, by applying a magnetic field perpendicular to

---

Manuscript received November 5, 2018; revised January 5, 2019 and January 20, 2019; accepted January 21, 2019. Corresponding author: A. Goschew (e-mail: alexander.goschew@fu-berlin.de).

This paper has supplementary downloadable material available at http://ieeexplore.ieee.org, provided by the author.

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TMAG.2019.2895907

TABLE I
SAMPLE COMPOSITION

Sample	**Structure** (values given in nm)
A	Au(70)/EuS(2)/Co(6)/EuS(2)/LED
B	Au(70)/[EuS(2)/Co(6)]$_2$/EuS(2)/LED

the surface, the magnetic and quantization axes coincide and are used in surface emitting LED experiments. The drawback of surface emitting experiments is the need for large external magnetic fields in order to saturate the sample magnetization, which is usually confined in the sample plane due to its anisotropy. Hence, measuring in remanence requires the use of magnetic systems with perpendicular magnetic anisotropy [30], [31]. In addition, light emitted parallel to the sample surface, i.e., from the sample side (side emission) does not show any polarized electroluminescence (EL) [32].

However, increasing the size of the quantum wells enables observation of EL in side emission [33]. The easy axis of magnetization in EuS/Co and EuS/Ni systems is known to be located in the sample plane [34]. Thus, measuring the degree of circular polarization of the emitted EL of such a system would enable one to further study EuS/Co systems and would give insights on the degree of electron spin polarization on the injector site and thus the EuS/Co interface. The main objective using this system is to achieve efficient spin injection at RT.

II. EXPERIMENTAL DETAILS

High-quality p-GaAs (001) custom LED [35] wafers were used as substrates. Their structure is: n-GaAs (15 nm, Si: 5×10^{18} cm^{-3})/n-Al$_{0.3}$Ga$_{0.7}$As (500 nm, Si: 10^{17} cm^{-3})/ i-Al$_{0.3}$Ga$_{0.7}$As (15 nm)/p-GaAs (500 nm, C: 10^{18} cm^{-3})/ p-Al$_{0.3}$Ga$_{0.7}$As (500 nm, C: 10^{18} cm^{-3})/p-GaAs (500 nm, C: 10^{18}cm^{-3})/p-GaAs (600 μm and Zn: 1.3–2.0 × 10^{19} cm^{-3}). A thick Arsenic capping layer of about 1 μm was used for passivation, which was later removed by heating [36]. The substrates were cleaved into 1 × 1 cm^2 pieces and put into a Balzers UMS 630 evaporation chamber, where the additional EuS/Co was evaporated by electron beam evaporation (e-beam) from tungsten crucibles. E-beam evaporation is suitable for materials with very high melting points, such as EuS ($>$2200 °C). EuS powder and Co pellets were used as target materials. The base pressure of the chamber was 10^{-8} mbar. Before deposition, the arsenic capping layer was removed by indirectly heating the samples to 450 °C for 2 h with an inbuilt heating device. Afterward, the temperature was lowered to 100–150 °C for deposition. Two samples, A and B, were prepared with their details given in Table I.

In order to evaporate the gold capping layer as an electric contact, the samples had to be taken out of the vacuum chamber. However, this does not harm the magnetic properties as EuS is very stable against oxidation and acts itself as a protection layer for the underlying Co.

The samples were then cleaved into smaller pieces (1×1 mm^2) along their (110) direction and contacted between a copper block and a copper finger on a custom-built sample holder for electrical connection. The sample holder was placed inside a cryostat (Suzuki Shokan 4K GM) with the (110) edge facing the window of the cryostat. A superconducting magnet with a maximum magnetic field of 2 T was used to magnetize the sample along its (110) direction, i.e., parallel to the magnetic moment of the sample. To verify that the GaAs was not damaged during the sample fabrication, photoluminescence (PL) measurements were carried out by exciting the sample side edge with a 4 mW He–Ne laser. The emitted light was observed by a CCD camera. Fig. 1 shows the observed PL for sample B.

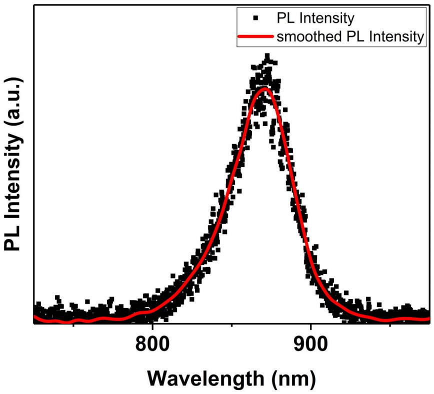

Fig. 1. PL spectrum of sample *B* at RT. A 4 mW **He–Ne** laser was used to excite the sample. Due to a low signal-to-noise ratio, the data were smoothed (red curve). The peak intensity found at 872 nm corresponds well to the GaAs bandgap.

The EL was observed with the same CCD camera while applying a voltage in forward bias. The light was guided through a set of lenses, a quarter-wave plate and a polarizer (Glan-Thompson), and detected by a photomultiplier tube (PMT). The signal was measured using a lock-in technique for which a mechanical chopper was placed in front of the PMT.

III. EXPERIMENTAL RESULTS

All samples were tested for EL at low temperature (8 K) and prior to each measurement, their I–V curves were recorded. Some I–V curves showed leak currents, whereas others showed typical LED current–voltage characteristics. Most of the EL measurements show a very low signal-to-noise ratio. The results of sample A (not shown here) do not show conclusive results as to a clear circular polarization at low temperatures (or RT). Sample B, however, shows a clear degree of circular polarization at 8 K shown in Fig. 2.

The observed degree of circular polarization amounts to $P_C = -5\%$ and $+5\%$ for the remanent state after initially applying a positive magnetic field of $+1$ T and a negative magnetic field of -1 T, respectively.

The observed change of sign of P_c for both remanent states indicates that the observed degree of polarization is indeed due to the spin-polarized current from the injection side and not due to artifacts.

However, the size of P_c is rather low. Given that all of the EuS should be ferromagnetic at 8 K, a higher degree of spin-polarized current injection could be expected.

The polarization of the injected electron current calculated by $P_{el} = 2 \times P_c$ is 10% for sample B. This value corresponds

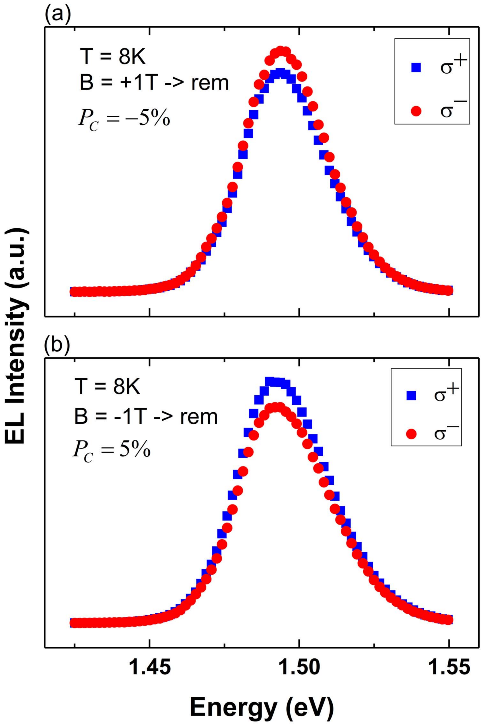

Fig. 2. EL spectrum of sample *B* at 8 K in the remanent state. Remanent state after applying (a) positive (+1 T) and (b) negative initial field (−1 T).

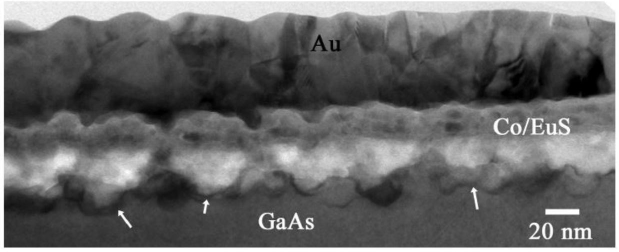

Fig. 3. TEM characterization of sample *B*. The substrate-sample interface is rather rough and shows a number of defected areas as indicated by white arrows. The Co/EuS layers as well as the Au capping layer are readily observed.

to the still measurable P_c recorded in the active layer of the LED. Estimating a typical spin decay length of 1 μm in GaAs [37], [38] at 5 K and considering an average distance traveled by the electrons of 750 nm until the center of the active region in our LEDs, one would lose about $e^{0.75} \sim 50\%$ of the initially injected spin polarization at the EuS/LED interface, giving $P_{\mathrm{c,initial}} = 20\%$.

In addition, EL measurements were performed in remanence at RT. Because the use of the superconducting magnet was restricted to low temperatures, no magnetic field could be applied at this temperature. The remanent state was obtained at low temperature by applying a negative magnetic field of −1 T. Then, it was switched off and the temperature was increased to RT. The observed intensity of the emitted EL at RT was less compared to the low-temperature measurement and the data only indicate a difference between right circularly polarized light and left circularly polarized light at RT. Therefore, the data are not shown here but are available as supplementary material at http://ieeexplore.ieee.org. Further measurements at RT will be carried out to get a conclusive result.

The quality of the wafer/sample interface was investigated by means of cross-sectional transmission electron microscopy. The results of sample B are shown in Fig. 3, where the EuS/Co and the thick Au capping layers are indicated. The surface of the substrate shows considerable roughness and defected areas indicated by white arrows. In addition, some parts of the surface are oxidized, as implied by the fainter contrast band between GaAs and EuS/Co in Fig. 3 and derived by energy dispersive X-ray spectroscopy measurements in the TEM, too. Sample A showed a similar substrate/sample interface as sample B. The reason for these defects is still under investigation but is most likely connected to the wafer processing steps during sample preparation.

The fact that the interface quality of the samples is poor is not discouraging but rather encouraging from an application point of view: Generally, the spin injection efficiency will strongly depend on the quality of the interface and will be lower for rough interfaces [39], [40]. For our samples, we find a value of $P_{\mathrm{C}} = 5\%$ which is comparable to the values given in [31], [41], and [42]. The fact that in our samples spins injected through a rough interface still lead to a measurable circular polarization encourage further investigations of EuS-based layers as spin-injection sources. An improvement of the interface quality is expected to lead to a considerable increase in injection efficiency.

IV. CONCLUSION

In summary, EL measurements were conducted at low temperature and RT in side-wall emission. Spins were injected from EuS/Co layers into GaAs-based LEDs yielding a degree of circular polarization of 5% at 8 K at zero external magnetic field. At low temperature, we could show that the sign of P_{c} reverses when the initial magnetic field was reversed while the EL intensity at RT was inconclusive and further measurements are needed to get a conclusive result. The interface quality of the substrate sample interface is far from perfect and future work will focus on improving the interface quality and should result in an enhanced injection efficiency.

REFERENCES

[1] S. A. Wolf *et al.*, "Spintronics: A spin-based electronics vision for the future," *Science*, vol. 294, no. 5546, pp. 1488–1495, 2001.
[2] I. Žutić, J. Fabian, and S. Das Sarma, "Spintronics: Fundamentals and applications," *Rev. Mod. Phys.*, vol. 76, pp. 323–410, Apr. 2004.
[3] G. Schmidt, "Concepts for spin injection into semiconductors—A review," *J. Phys. D, Appl. Phys.*, vol. 38, no. 7, pp. R107–R122, 2005.
[4] J. Fabian, A. Matos-Abiague, C. Ertler, I. Žutić, and I. Zutic, "Semiconductor spintronics," *Acta Phys. Slovaca*, vol. 57, pp. 565–907, Aug. 2007.
[5] S. D. Bader and S. S. P. Parkin, "Spintronics," *Annu. Rev. Condens. Matter Phys.*, vol. 1, no. 1, pp. 71–88, 2010.
[6] W. Liu, P. K. J. Wong, and Y. Xu, "Hybrid spintronic materials: Growth, structure and properties," *Prog. Mater. Sci.*, vol. 99, pp. 27–105, Jan. 2019.
[7] G. Vereecke, E. Röhr, and M. M. Heyns, "Laser-assisted removal of particles on silicon wafers," *J. Appl. Phys.*, vol. 85, no. 7, p. 3837, 1999.
[8] P. R. Hammar, B. R. Bennett, M. J. Yang, and M. Johnson, "Observation of Spin Injection at a Ferromagnet-Semiconductor Interface," *Phys. Rev. Lett.*, vol. 83, pp. 203–206, Jul. 1999.

[9] G. Schmidt, D. Ferrand, L. W. Molenkamp, A. T. Filip, and B. J. van Wees, "Fundamental obstacle for electrical spin injection from a ferromagnetic metal into a diffusive semiconductor," *Phys. Rev. B, Condens. Matter*, vol. 62, pp. R4790–R4793, Aug. 2000.

[10] A. Fert and H. Jaffrès, "Conditions for efficient spin injection from a ferromagnetic metal into a semiconductor," *Phys. Rev. B, Condens. Matter*, vol. 64, p. 184420, Oct. 2001.

[11] E. I. Rashba, "Theory of electrical spin injection: Tunnel contacts as a solution of the conductivity mismatch problem," *Phys. Rev. B, Condens. Matter*, vol. 62, no. 24, p. R16267, 2000.

[12] V. F. Motsnyi *et al.*, "Optical investigation of electrical spin injection into semiconductors," *Phys. Rev. B, Condens. Matter*, vol. 68, p. 245319, Dec. 2003.

[13] X. Jiang *et al.*, "Highly spin-polarized room-temperature tunnel injector for semiconductor spintronics using MgO(100)," *Phys. Rev. Lett.*, vol. 94, Feb. 2005, Art. no. 056601.

[14] T. Sasaki, T. Oikawa, T. Suzuki, M. Shiraishi, Y. Suzuki, and K. Tagami, "Electrical spin injection into silicon using MgO tunnel barrier," *Appl. Phys. Express*, vol. 2, no. 5, p. 053003, 2009.

[15] R. Benabderrahmane, M. Kanoun, N. Bruyant, C. Baraduc, A. Bsiesy, and H. Achard, "$\mathrm{Al_2O_3}$ tunnel barrier as a good candidate for spin injection into silicon," *Solid-State Electron.*, vol. 54, no. 8, pp. 741–744, 2010.

[16] N. Nishizawa and H. Munekata, "Efficient spin injection through a crystalline AlO_x tunnel barrier prepared by the oxidation of an ultra-thin Al epitaxial layer on GaAs," *J. Appl. Phys.*, vol. 114, no. 3, p. 033507, 2013.

[17] P. Wachter, "Electronic structure, magnetic exchange, and electrical transport properties of the magnetic compounds EuS, GdS and GdP," *Phys. Rep.*, vol. 44, pp. 159–186, Aug. 1978.

[18] P. Wachter, "Europium chalcogenides: EuO, EuS, EuSe and EuTe," in *Handbook on the Physics and Chemistry of Rare Earths*, vol. 2. Amsterdam, The Netherlands: North-Holland, 1979, pp. 507–574.

[19] N. Müller, W. Eckstein, W. Heiland, and W. Zinn, "Electron spin polarization in field emission from EuS-coated tungsten tips," *Phys. Rev. Lett.*, vol. 29, pp. 1651–1654, Dec. 1972.

[20] X. Hao, J. S. Moodera, and R. Meservey, "Spin-filter effect of ferromagnetic europium sulfide tunnel barriers," *Phys. Rev. B, Condens. Matter*, vol. 42, p. 8235, Nov. 1990.

[21] G.-X. Miao, M. Müller, and J. S. Moodera, "Magnetoresistance in double spin filter tunnel junctions with nonmagnetic electrodes and its unconventional bias dependence," *Phys. Rev. Lett.*, vol. 102, p. 076601, Feb. 2009.

[22] M. Müller, R. Schreiber, and C. M. Schneider, "Controlling magnetic properties of eus-based spin valve structures on Si(001)," *IEEE Trans. Magn.*, vol. 47, no. 6, pp. 1635–1638, Jun. 2011.

[23] P. Poulopoulos *et al.*, "Band-gap tuning at the strong quantum confinement regime in magnetic semiconductor EuS thin films," *Appl. Phys. Lett.*, vol. 100, no. 21, p. 211910, 2012.

[24] P. Fumagalli, A. Schirmeisen, and R. J. Gambino, "Exchange-induced enhancement of T_C in $\mathrm{Co}_{1-x}(\mathrm{EuS})_x$ macroscopic ferrimagnets," *Phys. Rev. B, Condens. Matter*, vol. 57, pp. 14294–14298, Jun. 1998.

[25] S. D. Pappas *et al.*, "Direct evidence for significant spin-polarization of EuS in Co/EuS multilayers at room temperature," *Sci. Rep.*, vol. 3, Feb. 2013, Art. no. 1333.

[26] P. Poulopoulos *et al.*, "Induced spin-polarization of EuS at room temperature in Ni/EuS multilayers," *Appl. Phys. Lett.*, vol. 104, no. 11, p. 112411, 2014.

[27] A. Goschew, M. Scott, and P. Fumagalli, "Verification of antiferromagnetic exchange coupling at room temperature using polar magneto-optic Kerr effect in thin EuS/Co multilayers with perpendicular magnetic anisotropy," *Appl. Phys. Lett.*, vol. 109, no. 6, p. 062401, 2016.

[28] C. Müller, H. Lippitz, J. J. Paggel, and P. Fumagalli, "Evidence of exchange-induced spin polarization in the semiconductor EuS at 300 K," *J. Appl. Phys.*, vol. 99, p. 073904, Feb. 2006.

[29] M. I. Dyakonov and V. I. Perel, *Optical Orientation*. Amsterdam, The Netherlands: Elsevier, 1984, pp. 11–71.

[30] S. H. Liang *et al.*, "Large and robust electrical spin injection into GaAs at zero magnetic field using an ultrathin CoFeB/MgO injector," *Phys. Rev. B, Condens. Matter*, vol. 90, no. 8, p. 085310, 2014.

[31] B. S. Tao *et al.*, "Electrical spin injection into GaAs based light emitting diodes using perpendicular magnetic tunnel junction-type spin injector," *Appl. Phys. Lett.*, vol. 108, no. 15, p. 152404, 2016.

[32] R. Fiederling, P. Grabs, W. Ossau, G. Schmidt, and L. W. Molenkamp, "Detection of electrical spin injection by light-emitting diodes in top- and side-emission configurations," *Appl. Phys. Lett.*, vol. 82, no. 13, pp. 2160–2162, 2003.

[33] O. M. J. van't Erve, G. Kioseoglou, A. T. Hanbicki, C. H. Li, and B. T. Jonker, "Remanent electrical spin injection from Fe into AlGaAs/GaAs light emitting diodes," *Appl. Phys. Lett.*, vol. 89, no. 7, p. 072505, 2006.

[34] B. Lewitz *et al.*, "Proximity effects and curie temperature enhancement in Co/EuS and Fe/EuS multilayers," *Spin*, vol. 2, no. 4, p. 1250016, 2012.

[35] Optowell Co., Ltd. Accessed: Oct. 15, 2018. [Online]. Available: www.optowell.com

[36] K. Knorr, M. Pristovsek, U. Resch-Esser, N. Esser, M. Zorn, and W. Richter, "*In situ* surface passivation of III–V semiconductors in MOVPE by amorphous As and P layers," *J. Cryst. Growth*, vol. 170, nos. 1–4, pp. 230–236, 1997.

[37] J. M. Kikkawa and D. D. Awschalom, "Lateral drag of spin coherence in gallium arsenide," *Nature*, vol. 397, pp. 139–141, Jan. 1999.

[38] N. Nishizawa, K. Nishibayashi, and H. Munekata, "A spin light emitting diode incorporating ability of electrical helicity switching," *Appl. Phys. Lett.*, vol. 104, p. 111102, Mar. 2014.

[39] C. H. Li *et al.*, "Spin injection across (110) interfaces: Fe/GaAs(110) spin-light-emitting diodes," *Appl. Phys. Lett.*, vol. 85, no. 9, pp. 1544–1546, 2004.

[40] S. F. Masoudi and Z. Ebrahiminejad, "Effects of interface structure on spin filter tunnel junctions," *Surface Sci.*, vol. 630, pp. 85–88, Dec. 2014.

[41] N. Yokota, Y. Aoshima, K. Ikeda, N. Nishizawa, H. Munekata, and H. Kawaguchi, "Room temperature spin injection into (110) GaAs quantum wells using Fe/x-AlO_x contacts in the regime of current density comparable to laser oscillation," *J. Appl. Phys.*, vol. 118, no. 16, p. 163905, 2015.

[42] R. Mansell *et al.*, "InGaAs spin light emitting diodes measured in the Faraday and oblique Hanle geometries," *J. Phys. D, Appl. Phys.*, vol. 49, no. 16, p. 165103, 2016.