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Hickey, M.C.; Damsgaard, Christian Danvad; Farrer, I; Holmes, S N; Husmann, A; Hansen, Jørn Bindslev; Jacobsen, Claus Schelde; Ritchie, D A; Pepper, M

Published in:
Applied Physics Letters

Link to article, DOI:
[10.1063/1.1949722](https://doi.org/10.1063/1.1949722)

Publication date:
2005

Document Version
Publisher's PDF, also known as Version of record

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Citation (APA):
Hickey, M. C., Damsgaard, C. D., Farrer, I., Holmes, S. N., Husmann, A., Hansen, J. B., ... Pepper, M. (2005). Spin injection between epitaxial Co_{2.4}Mn_{1.6}Ga and an InGaAs quantum well. Applied Physics Letters, 86(25), 252106. DOI: 10.1063/1.1949722

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Spin injection between epitaxial $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ and an InGaAs quantum well

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(Received 7 February 2005; accepted 12 May 2005; published online 14 June 2005)

Electrical spin injection in a narrow [100] $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ quantum well in a GaAs *p-i-n* optical device is reported. The quantum well is located 300 nm from an AlGaAs Schottky barrier and this system is used to compare the efficiencies and temperature dependences of spin injection from Fe and the Heusler alloy $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ grown by molecular-beam epitaxy. At 5 K, the injected electron spin polarizations for Fe and $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ injectors are 31% and 13%, respectively. Optical detection is carried out in the oblique Hanle geometry. A dynamic nuclear polarization effect below 10 K enhances the magnetic field seen by the injected spins in both devices. The $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ thin films are found to have a transport spin polarization of $\sim 50\%$ by point contact Andreev reflection conductivity measurements. © 2005 American Institute of Physics. [DOI: 10.1063/1.1949722]

The spin light-emitting diode (spin-LED) has become a self-contained experimental platform in which to explore spin dynamics and spin transport in semiconductors as well as the interplay between spin and charge degrees of freedom. The requirements for spin qubit-based quantum computing¹ in the solid state has motivated intense research to create the ideal materials system and device design for probing spin states and generating completely spin-polarized currents in semiconductors. The search for such ideal spin-injection devices based on semiconductors has divided into three research avenues: (1) use of a tunnel barrier oxide or semiconductor Schottky barrier with ferromagnetic metallic thin film as spin injectors, (2) the use of dilute magnetic semiconductors (DMS) as spin aligning contacts, and (3) the use of the theoretically predicted half-metallic ferromagnetic materials as 100% spin polarized contacts. Jiang *et al.*² have recently demonstrated injected spin polarization of 60% from an Fe/MgO contact at 100 K and spin polarizations of 80% have been recently seen in a DMS spin-LED-based on GaMnAs.³ For half-metals, the conductivity mismatch between the metal and semiconductor may not be an issue for

spin injection and high T_C ferromagnetic metals offer the possibility of spin-injection devices operating at room temperature. The purpose of this work is to examine the feasibility of injecting spins from the half-metallic^{4,5} ferromagnetic ternary alloy $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ in the device architecture of an InGaAs quantum well (QW) spin-LED. III-V layers are grown by molecular-beam epitaxy (MBE) to form a *p-i-n* doping structure with a surface *n*-AlGaAs barrier and a 5 nm wide InGaAs QW, 300 nm from the ferromagnetic metal-semiconductor (FM/SC) interface. The device consists of 15 nm *n*- $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ ($3 \times 10^{18} \text{ cm}^{-3}$), 15 nm *n*- $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ ($1 \times 10^{18} \text{ cm}^{-3}$), 100 nm *n*-GaAs ($1 \times 10^{18} \text{ cm}^{-3}$), 200 nm GaAs, 5 nm $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$, 500 nm GaAs, and 500 nm *p*-GaAs ($1 \times 10^{18} \text{ cm}^{-3}$). The AlGaAs Schottky barrier is designed to enable tunneling of spin-polarized carriers with a spin-dependent resistance at the FM/SC interface and satisfies the Rowell criteria for single-step tunneling.⁶ The spin-LED wafer was arsenic capped and then transferred to a ternary metal MBE chamber where the arsenic layer was thermally desorbed. A 5 nm thick Co-Ga buffer layer is grown followed by a 10 nm $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ film (at a growth rate of 0.75 \AA s^{-1}) and then a 5 nm capping layer of Au. The Co-Ga buffer layer was grown as an inter-

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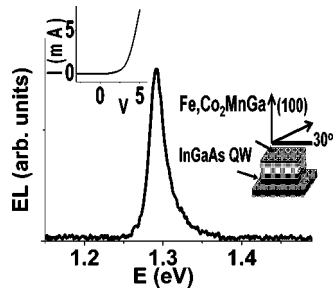


FIG. 1. A schematic of the spin-LED device with a Schottky I - V characteristic at 300 K and an EL emission spectrum from a 9 mA excitation current.

rupt layer so as to retain the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ spin-transport properties at the AlGaAs interface. The growth temperature of the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ layer was 200 °C. Stoichiometric analysis by inductively coupled plasma optical emission spectroscopy of the wafer indicates a composition of $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ rather than the ideal Heusler structure with a composition of Co_2MnGa . The Fe wafers were deposited by MBE onto an identical LED wafer in order to compare spin-injection effects from both metals. The device structure is depicted in Fig. 1.

Room-temperature emission from the InGaAs quantum well is observed at 1.30 eV. We use the emission arising from the QW electron ground state to heavy-hole valence band transition to determine the optical polarization efficiency (P_{EL}), as defined by

$$P_{\text{EL}} = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)}, \quad (1)$$

where $I(\sigma^+)$ is the intensity of positive helicity luminescence and $I(\sigma^-)$ is the intensity of negative helicity luminescence. The oblique Hanle effect (OHE) geometry is used with the magnetic field applied at an angle of 30° to the plane of the device mesa as outlined by Motsnyi *et al.*⁷ This geometry requires small magnetic fields ($B < 1$ T) to saturate the spin-polarized signal. A LED with an epitaxial Au contact shows no polarization properties with $P_{\text{EL}} < 0.01\%$ at 0.8 T. Using the OHE geometry we can estimate the effective spin lifetime (T_s^*) in the intrinsic region of the device, where the effective spin lifetime is related to the Hanle curve half-width (ΔB) by

$$\Delta B = \left(\frac{g\mu_B T_s^*}{\hbar} \right)^{-1}, \quad (2)$$

where g is the electron Landé g factor and μ_B is the Bohr magneton. The electroluminescence (EL) emission is collected along the (100) direction (see Fig. 1). The $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ device shows a much more abrupt EL polarization switching with magnetic field [see Fig. 2(a)], compared with the Fe spin-LED [see Fig. 2(c)]. The polarization of the spins injected from the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ film retains the easy-axis characteristic of the in-plane component of the magnetization, which saturates at lower fields than the out-of-plane component. In the 5 nm Fe film, surface anisotropy is more dominant, and the magnetization tilts more easily out of plane with an applied field. As a result, the measured polarization reflects the hard-axis characteristic of the (100) direction. Injection from Fe gives an optical polarization from the InGaAs QW emission at 5 K of $5.0 \pm 0.5\%$ and $1.3 \pm 0.1\%$ at 300 K. The $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ spin-LED exhibits an

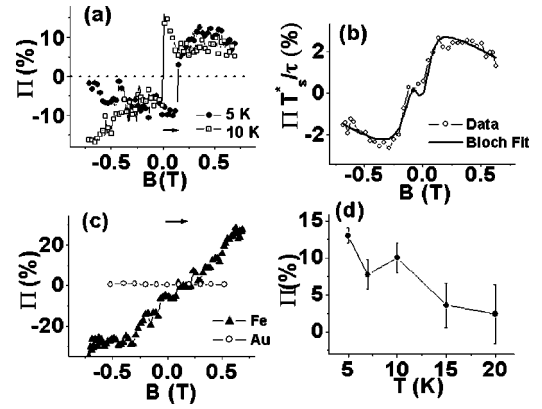


FIG. 2. (a) The injected spin polarization from the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ spin-LED, showing a DNP offset in the total magnetic field at a current density of 3 A cm^{-2} . (b) The Hanle data from a $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ device (2.8 A cm^{-2}) and a fit to Bloch theory. From this fit, the effective spin lifetime was calculated. The data show a small MCD linear component ($< 0.2\%$). (c) The injected spin polarization from the Fe device at 8 K measured in the OHE geometry showing a hard-axis magnetic field dependence. The DNP offset is marked with an arrow. (d) The temperature dependence of the injected spin polarization in the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ device.

optical polarization of 2.4% at 5 K with a magnetic circular dichroism (MCD) background of $< 0.2\%$. The optical polarization is related to the injected spin polarization via

$$\frac{P_{\text{EL}}}{\cos(\phi)\sin(\phi)} = \Pi \frac{T_s^*}{\tau}, \quad (3)$$

where ϕ is the Hanle angle, Π is the spin-injection efficiency, and the effective spin lifetime is related to τ_s (the spin scattering time), and τ (the radiative lifetime) by $1/T_s^* = 1/\tau_s + 1/\tau$. The ratio (T_s^*/τ) represents the spin detection efficiency of the QW. T_s^* is estimated to be 140 ± 30 ps at 5 K from fits to steady-state Bloch equations⁷ for the device operating at a current density of 2.8 A cm^{-2} [see Fig. 2(b)]. The radiative lifetime is estimated to be 400 ± 10 ps from standard time-resolved photoluminescence measurements on undoped reference QW samples, which gives a spin-detection efficiency ratio of $35 \pm 5\%$ at 5 K. As a result, the injected carrier polarization can be estimated to be $31 \pm 2\%$ for Fe [see Fig. 2(c)] and $13 \pm 3\%$ for $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ at 5 K. At 300 K, Π for Fe is $8 \pm 2\%$ while the spin injection from $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ vanishes at 20 K [see Fig. 2(a)]. The saturated magnetization of the Fe and $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ thin films are measured by a superconducting quantum interference device magnetometer to be $1.8 \pm 0.3 \mu_B$ per atom and $3.5 \pm 0.2 \mu_B$ per formula unit, respectively. The saturated magnetization of bulk Fe and Co_2MnGa are $2.1 \mu_B$ per atom⁸ and $4.1 \mu_B$ per formula unit,⁹ respectively. The in-plane switching fields for the magnetic thin films are 5 and 20 mT for Fe and $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$, respectively, whereas the out-of-plane magnetization saturates in fields of 2 T. In the Fe spin-LED, the maximum injected spin polarization of 31% occurs at a bias of 3.2 V (2.5 A cm^{-2}), followed by a gradual decrease to 0 at a bias of 4.7 V (4.3 A cm^{-2}) indicating a transition from tunneling transport to thermionic emission as noted in previous work.^{7,10}

The magnetic field dependence of the injected polarization in the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ device [see Fig. 2(a)] shows a departure from Bloch theory at 3 A cm^{-2} . This data was taken with a slower magnetic field sweep rate ($\sim 0.002 \text{ T s}^{-1}$ compared with 0.01 T s^{-1}) and would indicate a coupling to phe-

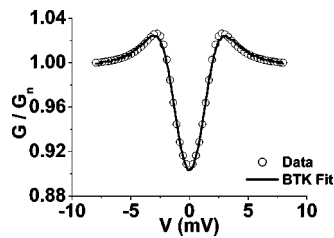


FIG. 3. The normalized conductance as a function of applied voltage for a Nb- $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ point contact and a numerical fit to BTK theory. The fitting parameters are the transport spin polarization ($P=49.7\%$), oxide barrier strength ($Z=0.23$), Nb superconducting energy gap ($\Delta=1.4$ meV), and temperature ($T=6$ K).

nomena with long relaxation times such as the orientation of nuclear spins. The use of the OHE geometry provides sensitivity to the onset of dynamic nuclear spin polarization (DNP) as previously observed as a current density-dependent effect¹¹ generating effective nuclear fields of up to 0.9 T. Injected spin-polarized electrons interact with the nuclear spins via hyperfine coupling and the dynamically polarized nuclear field enhances the applied field seen by the injected electron spins according to the relation $\mathbf{B}_{\text{tot}}=\mathbf{B}_a + C\mathbf{B}_a(\langle S \rangle \cdot \mathbf{B}_a)/|\mathbf{B}_a|^2$ as suggested by previous work,¹² where \mathbf{B}_a is the applied field, $\langle S \rangle$ is the time-averaged electron spin and C depends upon the overlap between the electron and nuclear wave functions. In our experimental geometry, the DNP-enhanced magnetic field is observed simply as an offset to the applied field in the polarization in both the Fe (which vanishes at 15 K) and $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ (which vanishes at 10 K) devices at current densities of 2.4 A cm^{-2} (for Fe) and 3 A cm^{-2} for the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ spin-LED. The observed offsets are 0.14 and 0.2 T for the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ and Fe devices, respectively [see Figs. 2(a) and 2(c)]. This suggests that the DNP effect is dependent upon the polarization of the injected spins. This voltage tunability of DNP has been demonstrated recently in an Fe/GaAs Schottky diode.¹³ Although we have demonstrated injection from Fe up to 300 K in this spin-LED structure, the injected polarization from the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ layer disappears at ~ 20 K on the same device substrate [see Fig. 2(d)]. Temperature-dependent antisite defects¹⁴ as well as moderate tetrahedral lattice strain¹⁵ at interfaces have been known to heavily reduce the Fermi level spin polarization of Heusler alloys to $\sim 55\%$.

Using point contact Andreev reflection (PCAR) conductance measurements¹⁶ at low temperature, we have measured independently the transport spin polarization of the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ wafer. In Fig. 3 we show the conductance (ratio to the normal conductance) as a function of applied bias in a Nb- $\text{Co}_{2.6}\text{Mn}_{1.4}\text{Ga}$ point contact at 6 K. Numerical fits of this data to Blonder-Tinkham-Klapwijk (BTK) theory¹⁷ provide a spin polarization of $50 \pm 1\%$ and an interface impedance (Z) of 0.23. For a perfect ballistic contact $Z=0$, and for a tunneling contact $Z \rightarrow \infty$. The reduced values of spin-polarization efficiency determined optically and electrically are in agreement with a half-metallic alloy possessing Co antisite defects and could be due to the imperfect thin-film stoichiometry. PCAR measurements¹⁸ on bulk samples of Co_2MnGe have shown significantly lower spin polarizations than 100%.

In summary, we have compared spin injection from $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ and Fe into a [100] InGaAs QW LED structure. We have measured the transport spin polarization of the

$\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ injector and compared this measurement with the estimated injected spin polarization injected across 300 nm of GaAs into the QW. We have outlined discrepancies between measured transport spin polarization and spin-injection efficiency from the $\text{Co}_{2.4}\text{Mn}_{1.6}\text{Ga}$ film, and we attribute its poor comparison with Fe to interfacial disorder. While the aforementioned effects are observed in our films, a comparison with a perfectly stoichiometric Heusler thin film is desirable. Recent work¹⁹ has demonstrated that the Heusler alloy Co_2MnGe shows an injected spin polarization of 27% with a GaAs/AlGaAs QW detector. We found that optical detection in the OHE geometry is sensitive to magnetic anisotropy, dynamic nuclear spin polarization, and the effective spin lifetime. The spin-LED device demonstrates the possibility of using dynamic nuclear polarization to manipulate nuclear spins in semiconductor spintronic devices with an optical readout mechanism.

The authors thank K. Cooper and T. Meng for processing and MBE growth, and D. Gevaux, M.B. Ward, and A. J. Shields for help with optical setup. One of the authors (M.C.H.) acknowledges financial support from EPSRC and the Cambridge European Trust. Other authors (C.D.D., J.B.H., and C.S.J.) acknowledge support from the Danish Technical Research Council and the Danish Natural Sciences Research Council.

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