# Spin-polarized injection into a p-type GaAs layer from a $Co_2MnAl$ injector*

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Electric luminescence and its circular polarization in a $Co_2MnAl$ injector-based light emitting diode (LED) has been detected at the transition of e–$A_C^0$, where injected spin-polarized electrons recombine with bound holes at carbon acceptors. A spin polarization degree of 24.6% is obtained at 77 K after spin-polarized electrons traverse a distance of 300 nm before they recombine with holes bound at neutral carbon acceptors in a $p^+$-GaAs layer. The large volume of the $p^+$-GaAs layer can facilitate the detection of weak electric luminescence (EL) from e–$A_C^0$ emission without being quenched at higher bias as in quantum wells. Moreover, unlike the interband electric luminescence in the $p^+$-GaAs layer, where the spin polarization of injected electrons is destroyed by a very effective electron–hole exchange scattering (BAP mechanism), the spin polarization of injected electrons seems to survive during their recombination with holes bound at carbon acceptors.

**Keywords:** spin injection, $Co_2MnAl$ Heusler alloy, electric luminescence

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Semiconductor devices combining the spin degree-of-freedom of carriers with traditional band gap engineering are expected to offer spin field-effect transistors (spin FET) and the like with new functionality and performance. To approach such a goal, an efficient spin injector is the first priority. Single element ferromagnetic transition metals such as iron (Fe) are attractive spin injectors as they possess a high Curie temperature and exhibit well understood thin film magnetism. There is also a strong driving force for investigating epitaxial single-crystal magnetic material/semiconductor heterostructures as spin injectors. Among them, Heusler alloy/compound semiconductor structures have drawn special attention. That is because many Heusler alloys have crystal structures (fcc) and lattice parameters similar to many compound semiconductors, while exhibiting high spin polarization at the Fermi level, and high Curie temperatures.[1,2] The efficiency of electron spin-polarization transmission across the Heusler/semiconductor interface was determined from spin-light emitting diode (spin-LED) structures incorporating $Co_2FeSi$,[3] $Co_2MnGa$,[4,5] and $Co_2MnGe$[6] spin injectors, showing the degrees of circular electric luminescence (EL) polarization $P_{EL}$ of 15% (at 20 K), 0.6% (at 5 K), and 14% (at 2 K), respectively. These $P_{EL}$ are far smaller than the values expected from nearly 100% spin polarization at the Fermi level of half metallic Heusler alloys. The low $P_{EL}$ was attributed to various lattice defects, chemical interaction, and inter-diffusion at the interface, and the band alignment across the interface, which may create a magnetic dead layer and cause spin flip scattering at the interface.[1]

In the present study we give the first report, to the best of our knowledge, on EL circular polarization in a $Co_2MnAl$ injector-based LED. A spin polarization degree of 24.6%, deduced from EL circular polarization, is obtained at 77 K after spin-polarized electrons traverse over a distance of 300 nm before they recombine with the holes bound at neutral carbon acceptors in the $p^+$-GaAs layer.

Our devices were grown by molecular beam epitaxy (MBE) on insulating (i) (001) GaAs substrates in the following layer sequence: 500-nm i-GaAs buffer layer, 100-nm $p^+$-GaAs ($p = 1\times10^{18}$ cm$^{-3}$), 100-nm $p^+$-$Al_{0.1}Ga_{0.9}As$ ($p = 1\times10^{18}$ cm$^{-3}$), 100-nm i-$Al_{0.1}Ga_{0.9}As$, 20-nm i-GaAs quantum well (QW), 25-nm i-$Al_{0.1}Ga_{0.9}As$, 12-nm i-GaAs QW, 25-nm i-$Al_{0.1}Ga_{0.9}As$, 8-nm i-GaAs QW, 10-nm i-$Al_{0.1}Ga_{0.9}As$, 5-nm $n^+$-$Al_{0.1}Ga_{0.9}As$ ($n = 5\times10^{18}$ cm$^{-3}$), 3-nm i-AlAs, and 12-nm $Co_2MnAl$. Among them, 3-nm thick i-AlAs acts as tunneling barrier, three quantum wells (QW) of different widths are designed to probe EL and its polarization at different depths. In addition, the EL across the intrinsic band gap and possible EL due to the recombination of injected electrons with neutral acceptors (like carbon centers) in the 100-nm $p^+$-GaAs layer are also used for checking EL circular polarization. The layer structure and equilibrium band diagram of our sample are schematically drawn in Figs. 1(a) and 1(c). The sample was mounted inside a long horizontal cooled finger of a nitrogen cryostat, which was inserted into the clear bore of a permanent magnet. The latter provides a 1.3-T magnetic field perpendicular to the sample surface. The EL emission

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was collected by a specially designed optical fiber system, and then dispersed by a spectrometer.

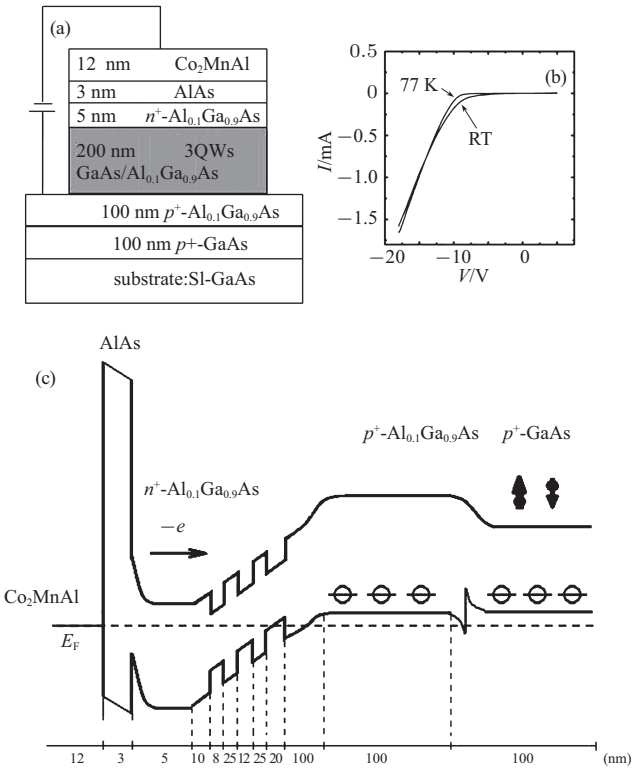



**Fig. 1.** (a) Schematic layer structure of $Co_2MnAl$ emitter-based LED. QW region is hatched. (b) *I–V* characteristics measured at 77 K and 300 K. (c) Schematic band diagram of whole $Co_2MnAl$ emitter-based LED at zero bias. Discrete levels with open circles on in the $p^+$-$Al_{0.1}Ga_{0.9}As$ and $p^+$-GaAs layers denote holes caught on neutral carbon acceptors. Longer and shorter up- and down-arrows represent spin-up and spin-down electrons injected.

The *I–V* characteristics of the sample were measured at both 77 K and 300 K in Fig. 1(b), showing a normal characteristic. The rapid increase of current at the bias beyond –10 V ensures strong injection for EL measurements. The hysteresis loop of $Co_2MnAl$ layer was obtained in a perpendicular magnetic field scan as shown in Fig. 2. Saturated magnetization is reached at the magnetic field beyond 2 T, after the magnetization turns from its original in-plane easy axis to the vertical. The photoluminescence spectrum (PL) at zero bias shows three peaks at the wavelengths of 802, 811, and 819.3 nm, corresponding the PLs from 8-nm, 12-nm, and 20-nm wide QWs, as shown in Fig. 3(a). A shoulder at 823.4 nm comes from PL in the bulk $p^+$-GaAs layer. A weak peak at 837 nm beyond the absorption edge is assigned to the recombination luminescence of electrons with neutral carbon acceptors (e–$A_C^0$), because the neutral carbon acceptors, $A_C^0$, are routinely present in the bulk $p^+$-GaAs. By biasing the top electrode negatively (i.e., increasing the negative current), three QW PL peaks are almost quenched, while the PL of bulk p-type GaAs and e–$A_C^0$ peak are somewhat enhanced. The PL spectra above indicate that QWs are not very effective for collecting photo-excited electrons and holes at high bias, as they bypass QWs due to the overshoot, as easily understood from the band diagram in Fig. 1(c).

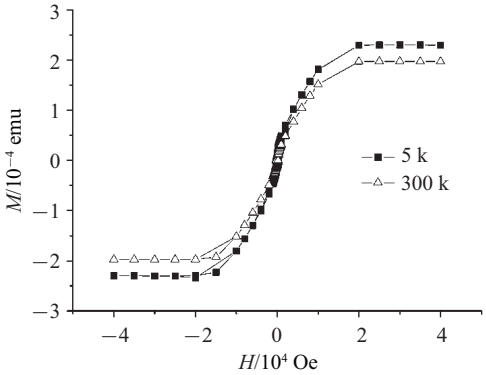



**Fig. 2.** Hysteresis loops measured by superconducting quantum interference device (SQUID) in a perpendicular magnetic field between ±4 T at 5 K and 300 K, 1 Oe = 79.5775 A/m.

The highest field of our permanent magnet is about 1.3 T, which is still lower than saturated magnetization. Figure 3(b) gives the EL spectra measured at four different biasing currents of 0.5, 0.7, 1.0, and 1.5 mA, respectively. In accordance with the PL spectra under the biasing (Fig. 3(a)), only two EL peaks from bulk $p^+$-GaAs and e–$A_C^0$ recombination emissions appear at $\lambda = 823.4$ nm and $\lambda = 837.4$ nm, respectively. With increasing biasing current, the peak intensities of the two EL peaks increase. As seen from Fig. 3(b), only the ELs from the e–$A_C^0$ transition show pronounced circular polarization. The degrees of circular polarization, defined by $P_{EL} = (I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$, are obtained to be 12.3%, 10%, 10.3%, and 9.5% for the biasing currents of 0.5, 0.7, 1.0, and 1.5 mA, respectively. $P_{EL}$ has its highest value at the lowest biasing current due to avoiding joule heating, as long as the EL has enough intensity for detection.

Before we analyze the physical mechanism of the observed circular polarization at e–$A_C^0$ transition, the background polarization induced by Zeeman splitting should be estimated first. Under the assumption that both electrons and holes are in their own thermodynamic equilibrium, respectively, the contributions of electrons and holes to the photoluminescence polarization, excited by linearly polarized light, are additive in the linear term of magnetic field $B$. It turns out[7] that the average angular momentums of electrons in the conduction band, of holes in the valence band or in acceptors can be estimated by $\langle S_z^c \rangle = \mu_B g_e B/4kT$, $\langle J_z^v \rangle = 3\mu_B g_h B/2kT$, and $\langle J_z^A \rangle = 5\mu_B g_A B/4kT$, where $g_e$, $g_h$, and $g_A$ are the effective Lande $g$-factors of electrons, free holes, and holes bound to acceptors. By putting the corresponding number ($g_e = -0.41$) in them, for example, $\langle S_z^c \rangle$ is equal to $3.3 \times 10^{-3}$. As a result, the background spin polarization attributed to Zeeman splitting should be negligible.

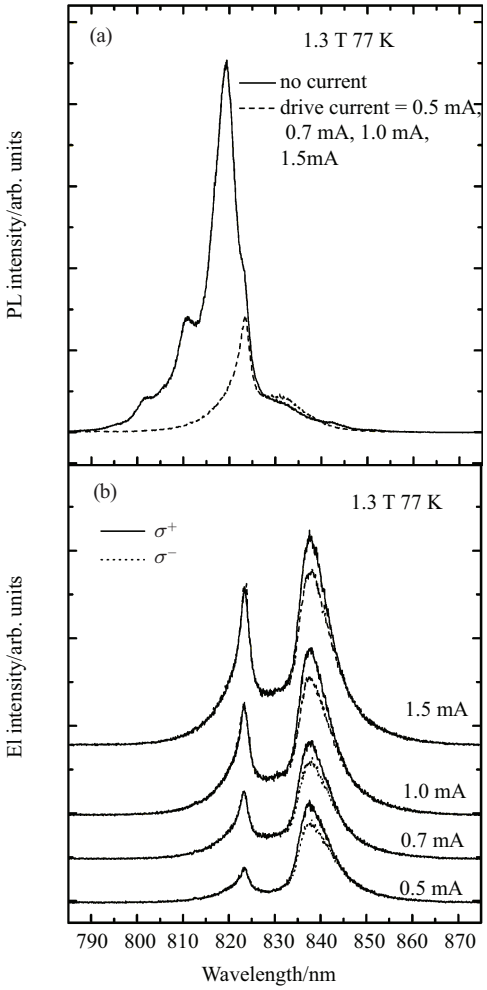



**Fig. 3.** (a) PL spectra measured at 77 K in the absence of and in the presence of biasing. (b) EL spectra measured at 77 K and 1.3 T under different biasing. Solid and dashed curves are for $\sigma^+$- and $\sigma^-$-polarized electric luminescences.

Next we turn to discuss experimental results. It is easy to explain why no circular polarization is detected for the EL peak at 823.4 nm. It is due to interband recombination of injected electrons with free holes in the valence band. It is well established that the exchange interaction between electrons and holes leads to electron spin relaxation, known as the Bir–Aronov–Pikus (BAP) mechanism.[8,9] BAP plays a major role in the spin relaxation, especially in p-type doped GaAs materials. That should effectively quench the spin polarization of the injected electrons. This observation also implies that the background circular polarization, induced by Zeeman splitting, is indeed not detectable at 77 K. Thus, one can be convinced, from the experimental point of view, that the observed EL circular polarization at the e–$A_C^0$ transition stems from injected spin-polarized electrons. However, the circular polarization of the EL peak at 837.4 nm is somehow peculiar from the physical point of view, and is worthy of discussion.

From the BAP mechanism[8,9] it follows that exchange scattering rates, induced by either free holes in the valence band or bound holes in acceptors, are proportional, respectively, to the free hole concentration $N_p$ or the bound hole concentration $N_A - N_p$. The contributions of incompletely ionized p-type GaAs materials to the spin scattering rate was simply considered to be additive with the same factor of $1/\tau_0$ in the original BAP theory given by[8,9]

$$\frac{1}{\tau_s} = \frac{2}{\tau_0}\left(N_A a_B^3\right)\frac{v_k}{v_B}\left[\frac{N_p}{N_A} + \frac{5}{3}\frac{N_A - N_p}{N_A}\right], \qquad (1)$$

where the parameters in Eq. (1) are defined by $E_B = \hbar^2/(2m_e a_B^2)$, $v_B = \hbar/(m_e a_B)$, $a_B = \hbar^2\varepsilon_0/(e^2 m_e)$ with $a_B$ being the Bohr radius, $m_e$ the electron's effective mass, and $v_k$ the electron velocity. $N_A$ is the concentration of p-type dopants. In our case, the second term in Eq. (1) should be replaced by $(10/3\tau_0)\left(N_C a_B^3\right)(v_k/v_B)$, with $N_C$ being the concentration of carbon residue impurities. From Eq. (1) it would seem that spin scattering rates, contributed by free holes and bound holes, are on an equal footing except for $N_C \ll N_P$. However, one has to check the important term $1/\tau_0$ in Eq. (1). It is this term that relates to the effective constant of spin–spin interaction $\overline{D_s^2}$ by

$$\frac{1}{\tau_0} = \frac{\pi}{2\hbar}\frac{\overline{D_s^2}}{E_B}\left|\phi_{3D}(0)\right|^4.$$ [8,9]

Here, $|\phi_{3D}(0)|^2$ is the so-called Sommerfeld factor, describing an enhancement of the electron–hole exchange interaction induced by the attraction between electrons and holes. But at rather high hole concentrations, $|\phi_{3D}(0)|^2$ reduces to one, owing to a screening effect. In the case where an electron is scattered by a hole bound on an acceptor, $|\phi_{3D}(0)|^2$ also becomes one, because the acceptor as a whole is neutral.

As pointed out by BAP theory,[8,9] the exchange interaction between electrons and holes consists of two parts: a short-range part and a long-range part. The former induces the exchange splitting $\Delta_{ex}^{SR}$ of the exciton ground state, which is the same for both heavy and light holes. The latter leads to the longitudinal–transverse splitting $\Delta_{ex}^{LR}$ of the exciton state, which is effective only for light holes. Usually, the direction-averaged, short-range exchange constant, $\overline{D_{s,SR}^2} = (5/32)\left(\Delta_{ex}^{SR}\right)^2$ is much larger than the long range one, $\overline{D_{s,LR}^2} = (5/288)\left(\Delta_{ex}^{LR}\right)^2$ (bound holes). In the present case, the condition of $N_C \ll N_P$ means that a tiny quantity of neutral carbon acceptors are immersed in the hole sea. Therefore, we conject, it may be possible that the hole sea may block the short range exchange interaction between the injected electrons and bound holes at the carbon acceptors, leaving only the long range term $\overline{D_{s,LR}^2}$. As a result, the spin polarization of injected electrons may survive until they recombine with the holes bound at carbon acceptors by reduced spin exchange scattering. This might explain why we have still observed a rather high EL circular polarization at e–$A_C^0$ transition of 837.4 nm. However, this point should be checked by theoretical work, which is beyond the scope of the present work.

Finally, to determine the spin polarization of injection from the EL circular polarization, we have to know the optical selection rule suitable to e–$A_C^0$ transition. In the frame of the spherical model of a shallow acceptor center, the ground and excited states of the carbon acceptor can be described by the BL equations.[10,11] It turns out that the wave functions of the carbon acceptor are transformed according to the $\Gamma_8^+$ irreducible representations of the $O_h$ group. Since the $\Gamma_8^+$ carbon acceptor state is the degenerate state of the total angular momentum $J = 3/2$, which is the same as the degenerate valence band of GaAs at the $\Gamma$ critical point; therefore, the selection rule of the recombination of electrons with carbon acceptors is the same as the interband transitions in bulk GaAs materials. As a result, by using the relation

$$P_{\mathrm{EL}} = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)} = \frac{1}{2}\frac{n_\uparrow - n_\downarrow}{n_\uparrow + n_\downarrow} = \frac{1}{2}P_{\mathrm{spin}},$$

a highest spin polarization degree $P_{\mathrm{spin}}$ of 24.6% is obtained for the electrons injected from $Co_2MnAl$ Heusler alloy.

In summary, the present paper is the first report, to the best of our knowledge, on the EL circular polarization in a $Co_2MnAl$ injector-based LED. A spin polarization degree $P_{\mathrm{spin}}$ of 24.6% is obtained at 77 K after spin-polarized electrons traverse over a distance of 300 nm before they recombine with the holes bound at neutral carbon acceptors in the $p^+$-GaAs layer. Compared to the previously published results employing $Co_2FeSi$, $Co_2MnGa$, and $Co_2MnGe$ as spin injectors, the $P_{\mathrm{spin}}$ value obtained in the present work is rather high. The use of the e–$A_C^0$ transition for detecting EL polarization also has advantages over the inner QW transitions. The large volume of the $p^+$-GaAs layer (compared to single QW) can facilitate the detection of weak EL from e–$A_C^0$ emission without the problem of being quenched at higher bias. Moreover, in contrast to EL in the bulk $p^+$-GaAs layer, where the spin polarization of injected electrons is destroyed by very effective electron–hole exchange scattering (BAP mechanism), the spin polarization of injected electrons seems to survive during their recombination with holes bound at carbon acceptors. In forthcoming work, we shall replace the layer structure of three quantum well structures (100-nm i-$Al_{0.1}Ga_{0.9}As$, 20-nm i-GaAs QW, 25-nm i-$Al_{0.1}Ga_{0.9}As$, 12-nm i-GaAs QW, 25-nm i-$Al_{0.1}Ga_{0.9}As$, 8-nm i-GaAs QW, 10-nm i-$Al_{0.1}Ga_{0.9}As$) by an i-GaAs layer of proper thickness to shorten the flying distance of injected electrons. In such a simplified n–i–p structure it is expected that even higher spin polarization will be observed, detected by the e–$A_C^0$ transition, due to reduced spin flip scattering of injected electrons during their travel.

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