

Optical detection of electrical spin-polarized charge injection in a $\text{V}[\text{TCNE}]_{x \sim 2}$ -based $\text{AlGaAs}/\text{GaAs}$ hybrid LED structure

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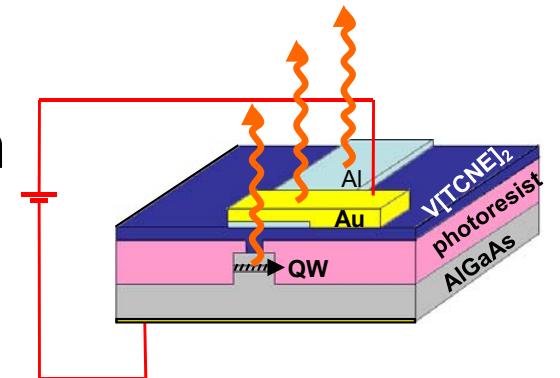
Supported in part by



Motivation

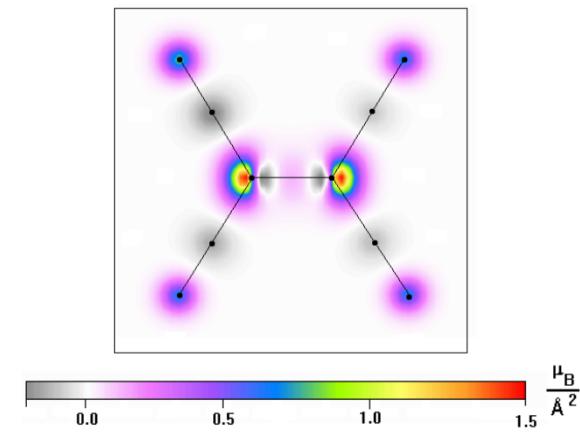
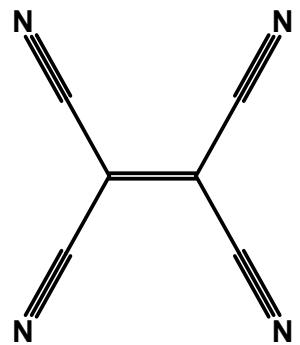
- Demonstration of spin-polarized charge injection from an organic magnet into an **inorganic semiconductors**.

Electrical spin injection —→ Optical detection



- Low cost , low temperature deposition
- Light weight: can be grown on various materials without any damage
- Flexible
- Chemically tunable electrical and magnetic properties
 - * Room temperature magnetism
 - * Semiconducting transport
- Tunable magnetic properties via external stimuli (optical, pressure)

V[TCNE]_{x~2}

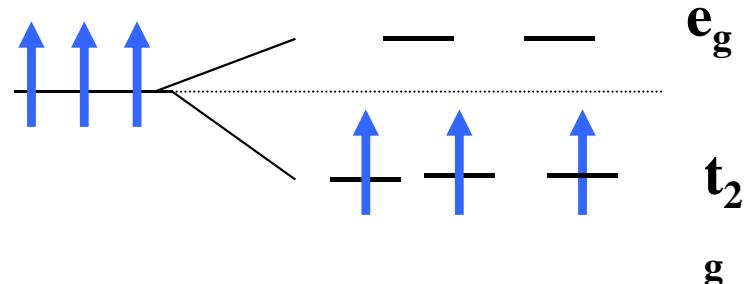


Chemical structure
of (TCNE)

S = 1/2 in p* orbital of [TCNE]⁻

Octahedral coordination of
V with **Ns**

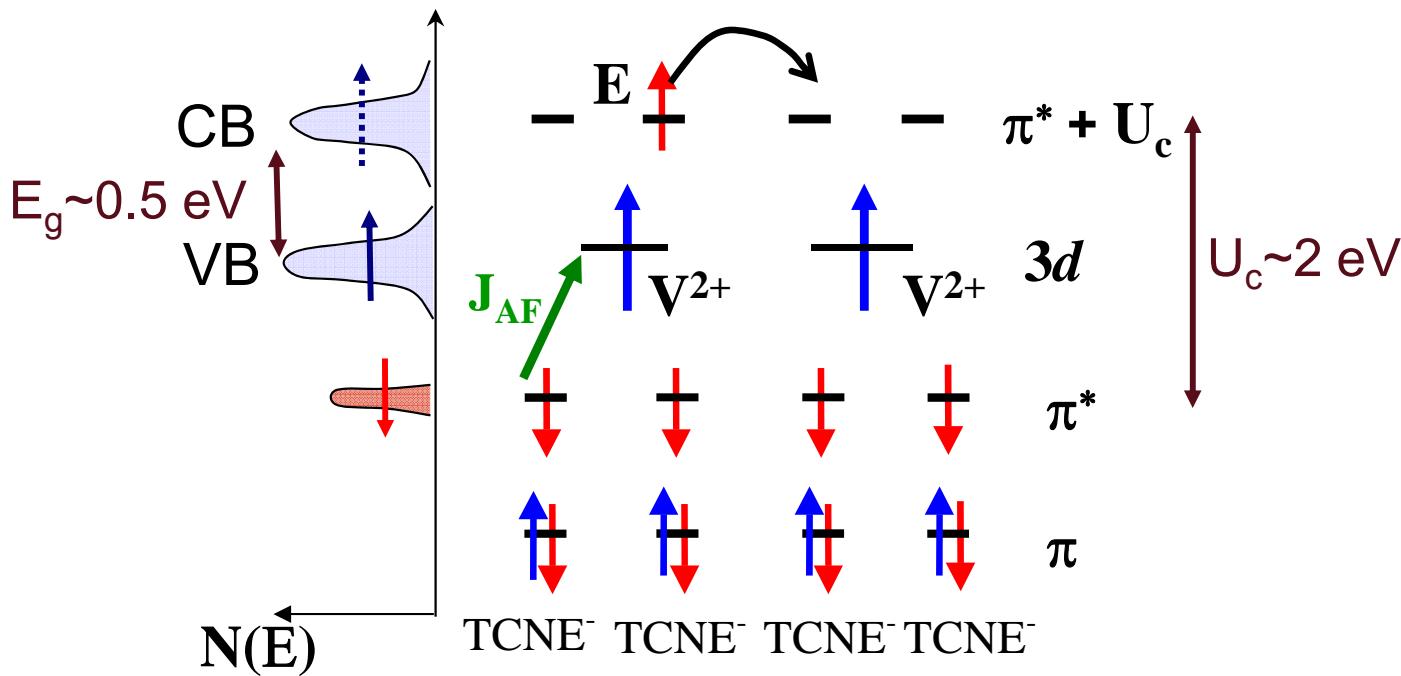
3d



V²⁺: S = 3/2 in t_{2g} orbitals

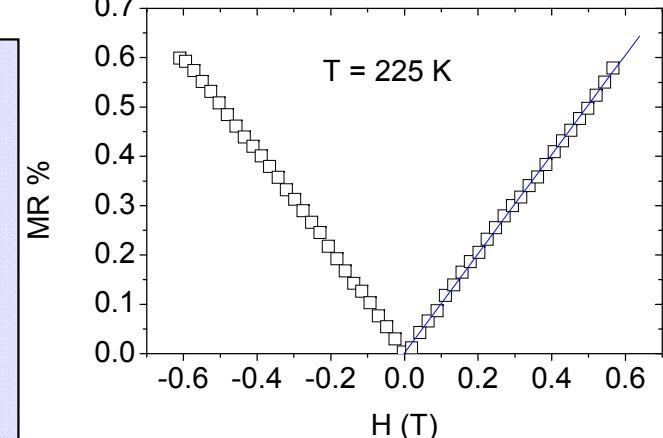
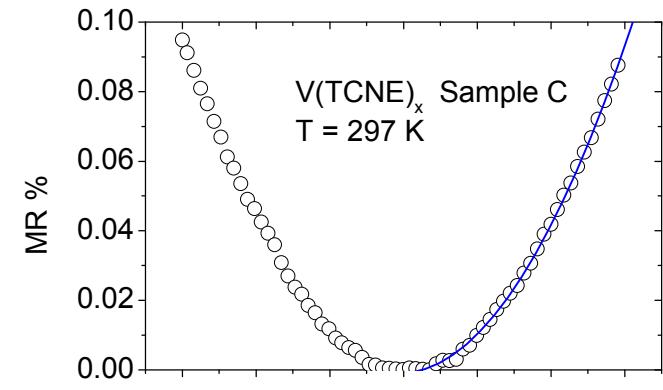
- Strong V-N bonding
- Direct exchange interaction **J_{AF}**
- Room temperature ordering ($T_c \sim 400$ K)
- Three dimensional network
- Disordered structure leads to random magnetic anisotropy (RMA)

Organic-Based Magnetic Semiconductor $V(TCNE)_{x \sim 2}$



$$R = R_0 \exp\left(\frac{E_g}{2k_B T}\right)$$

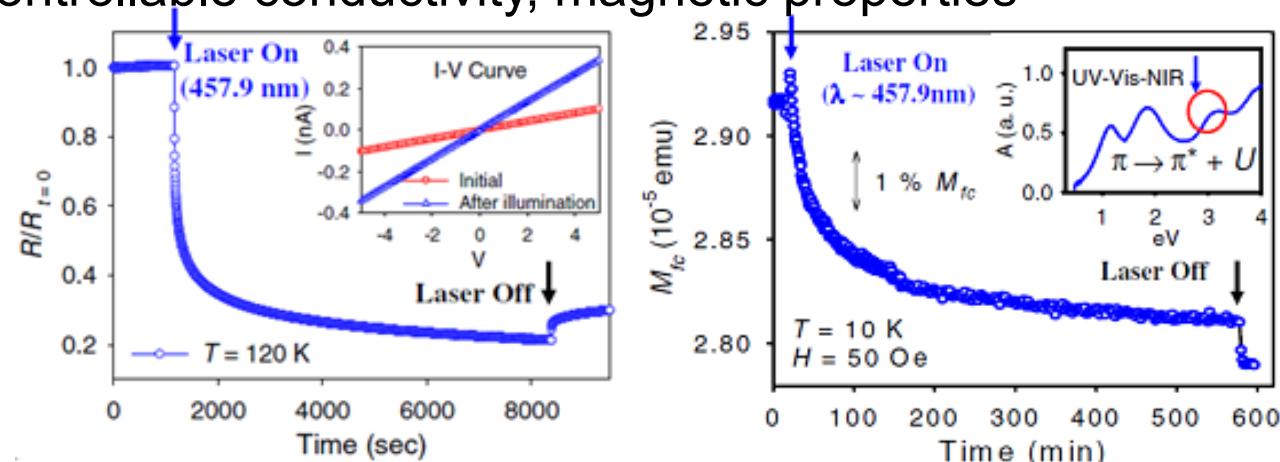
$$E_g' = E_g - J \langle S \rangle \langle \sigma \rangle$$



- Strong direct exchange J (antiferromagnetic) due to hybridization of V^{2+} ($S = 3/2$) and $[TCNE]^-$ ($S = 1/2$)
- Parallel alignment of spins in $\pi^* + U$ and V^{2+} bands
- Conduction and valance bands are spin polarized
- Anomalous positive MR

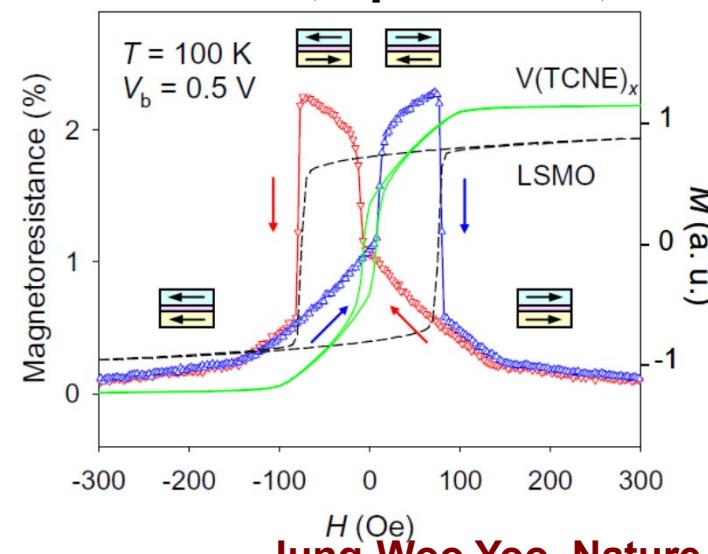
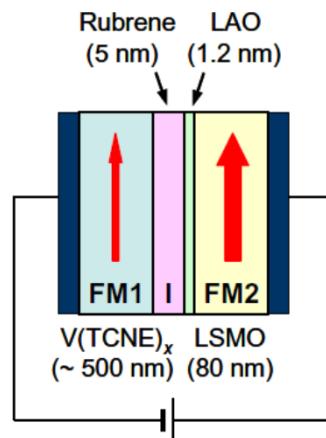
Multifunctionality of V[TCNE]_{x~2}

- Magneto-optic devices (memory devices, long distance switches)
 - Optically controllable conductivity, magnetic properties



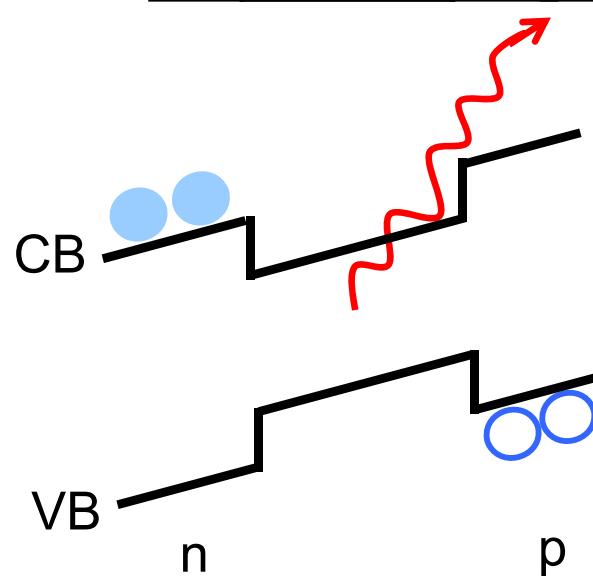
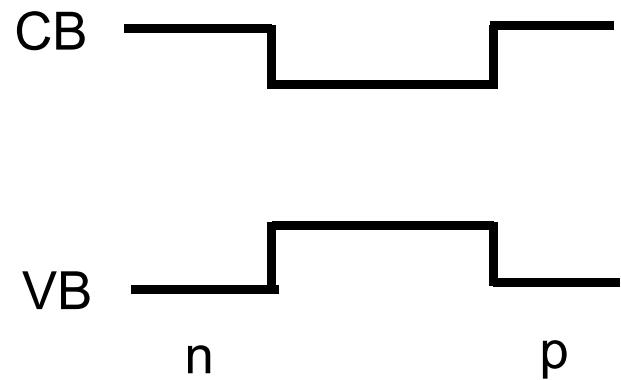
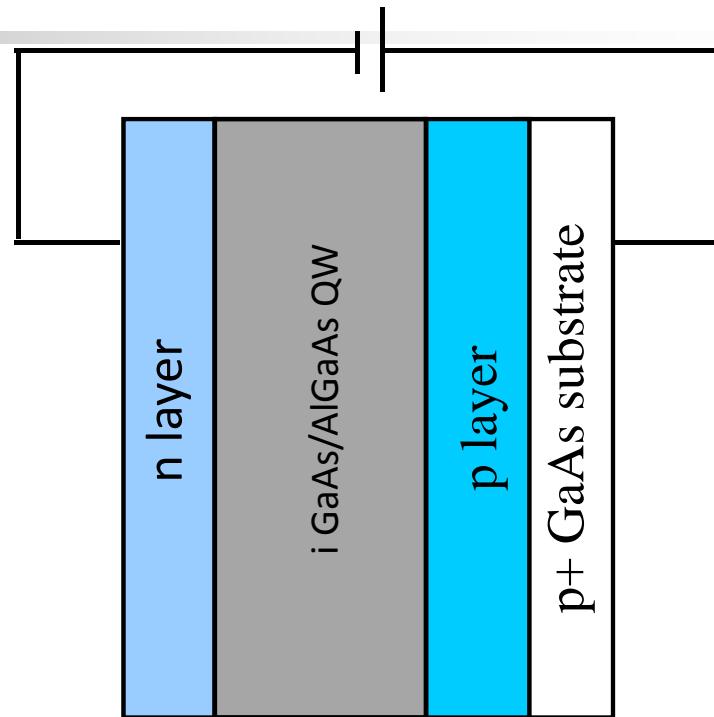
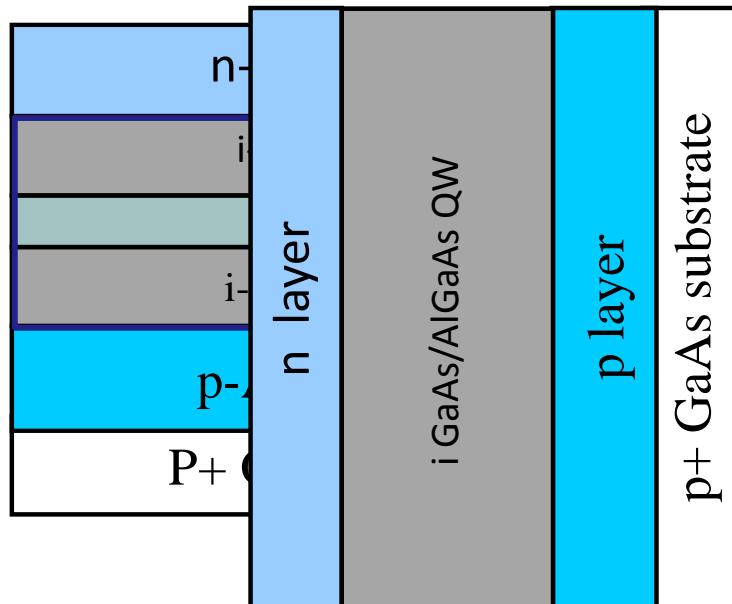
Jung Woo Yoo, PRL, 247205, 99 (2007)- PRL, 157205, 97 (2006)

- Spintronics (e.g. spin valves, spin transistors, spin LEDs, etc.)
 - Semiconductor
 - Near full spin polarization

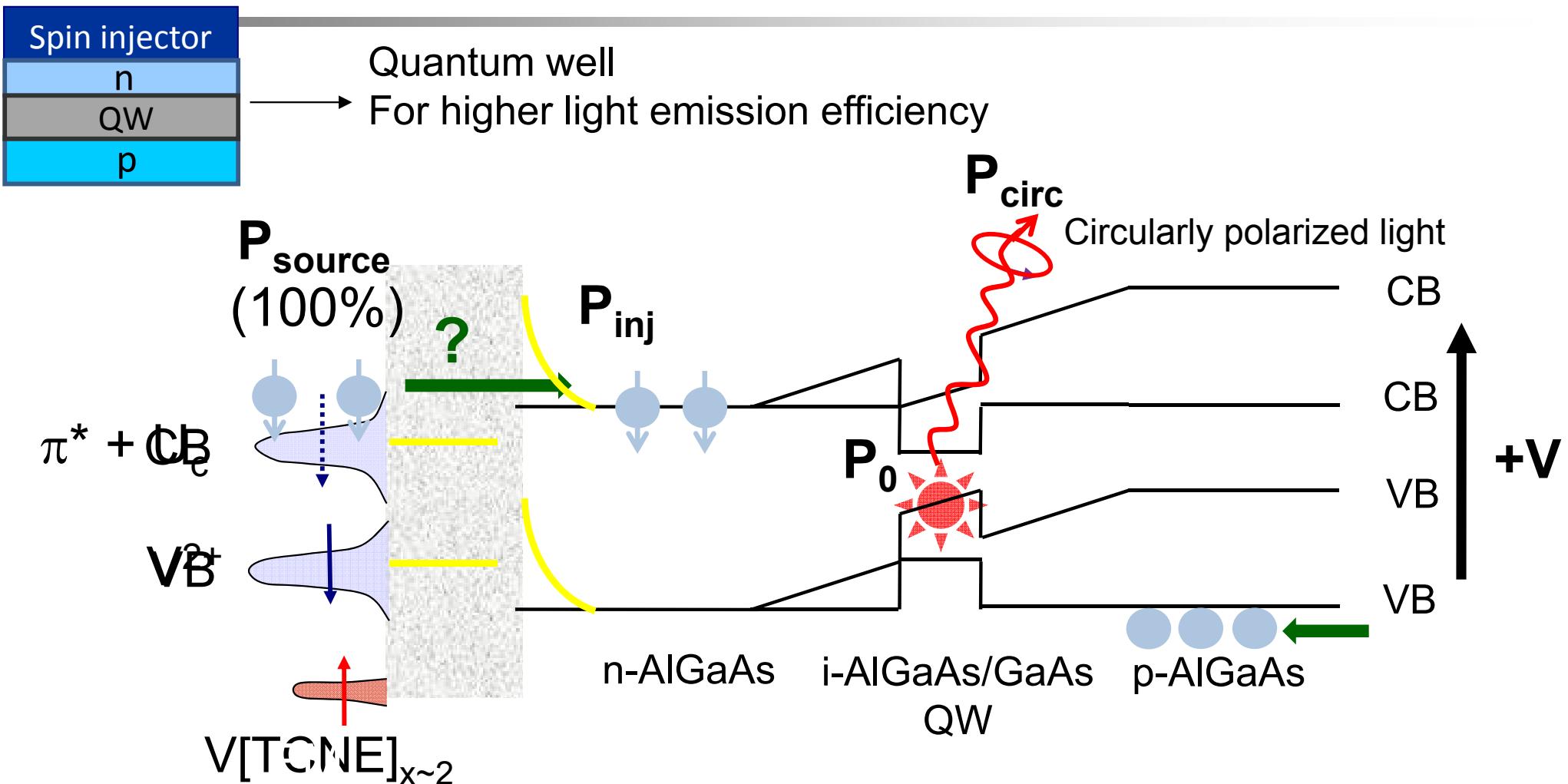


Jung-Woo Yoo, Nature Materials, 9, 638 (2010)

Light Emitting Diode (LED)



Spin Light Emitting Diode (spin-LED)



Inorganic ferromagnetic metals and semiconductors are commonly used as spin injector

- High temperature, high Vacuum
- Expensive

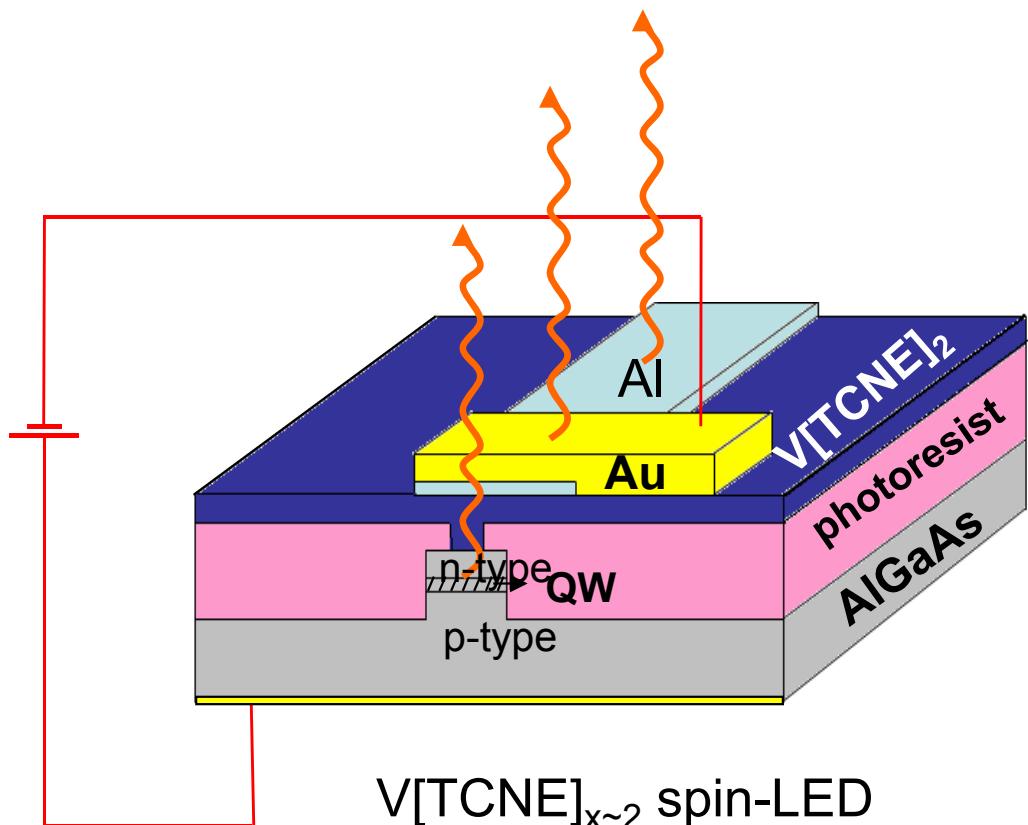
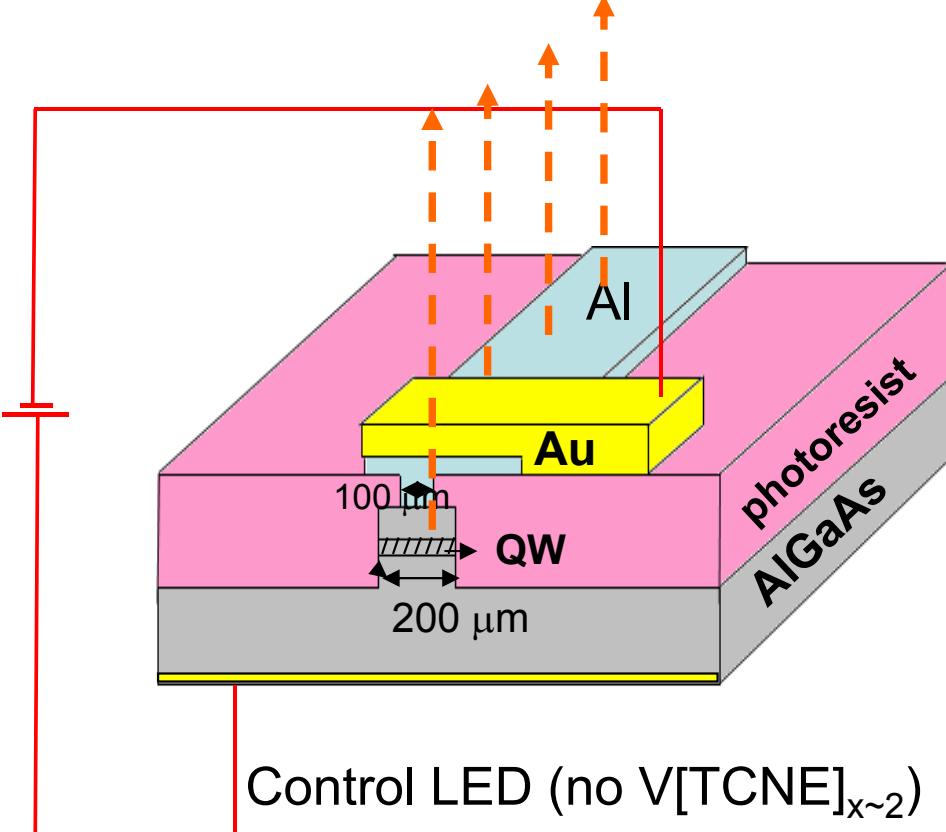
Spin-LED Device Structure and Fabrication



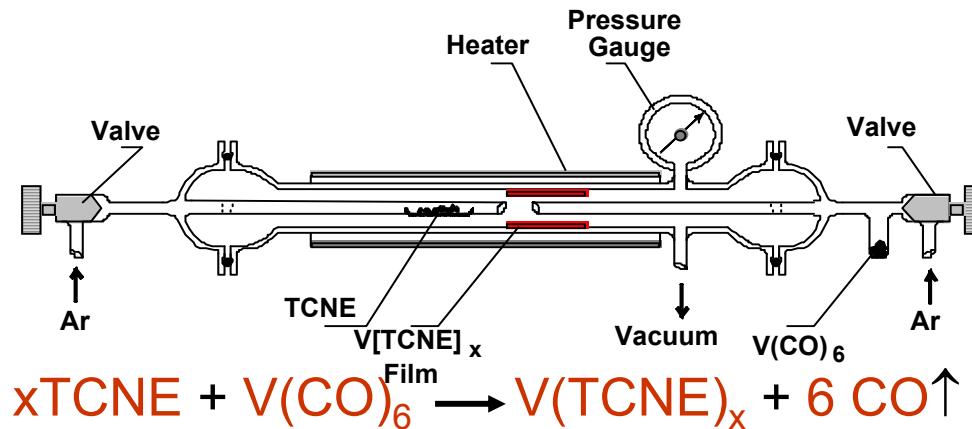
GaAs wafer (from top)

- 15 nm schottky contact
- 15 nm grading layer
- 100 nm drift layer
- 25 nm barrier
- 10 nm QW
- 25 nm barrier
- 200 nm p contact
- 300 nm buffer

Prof. Steve Ringel @ OSU



Spin-LED Device Fabrication



K.I. Pokhodnya, et al. *Adv. Mater.* 12, 410 (2000)



CVD deposition chamber
in an Argon glovebox



Metal deposition chamber
in a N_2 glovebox

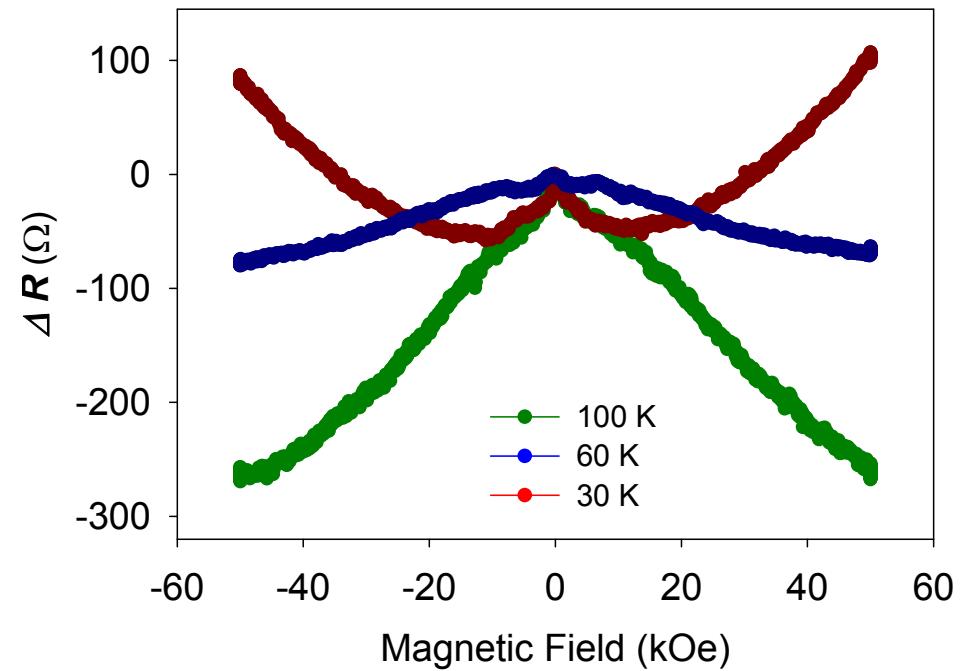
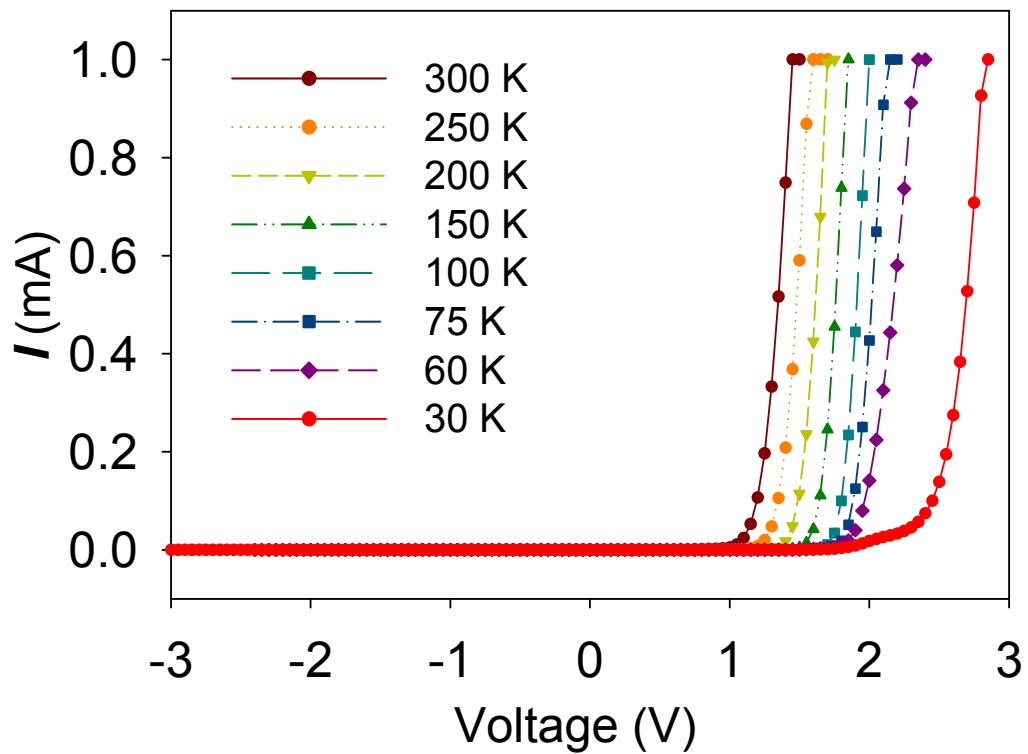


Air-free sample holder



→ Measurement
Chambers

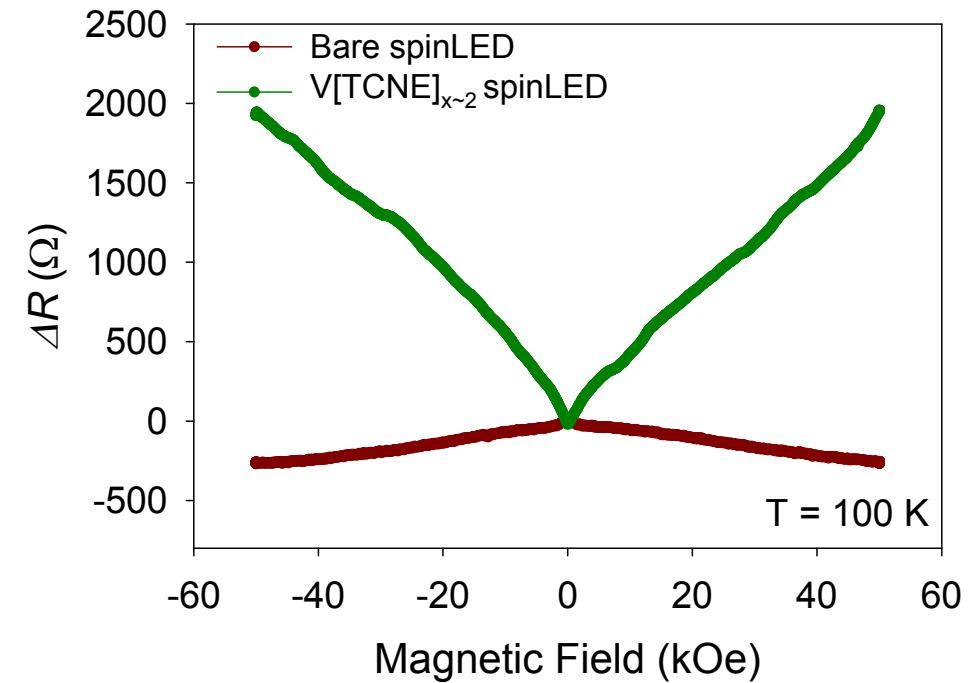
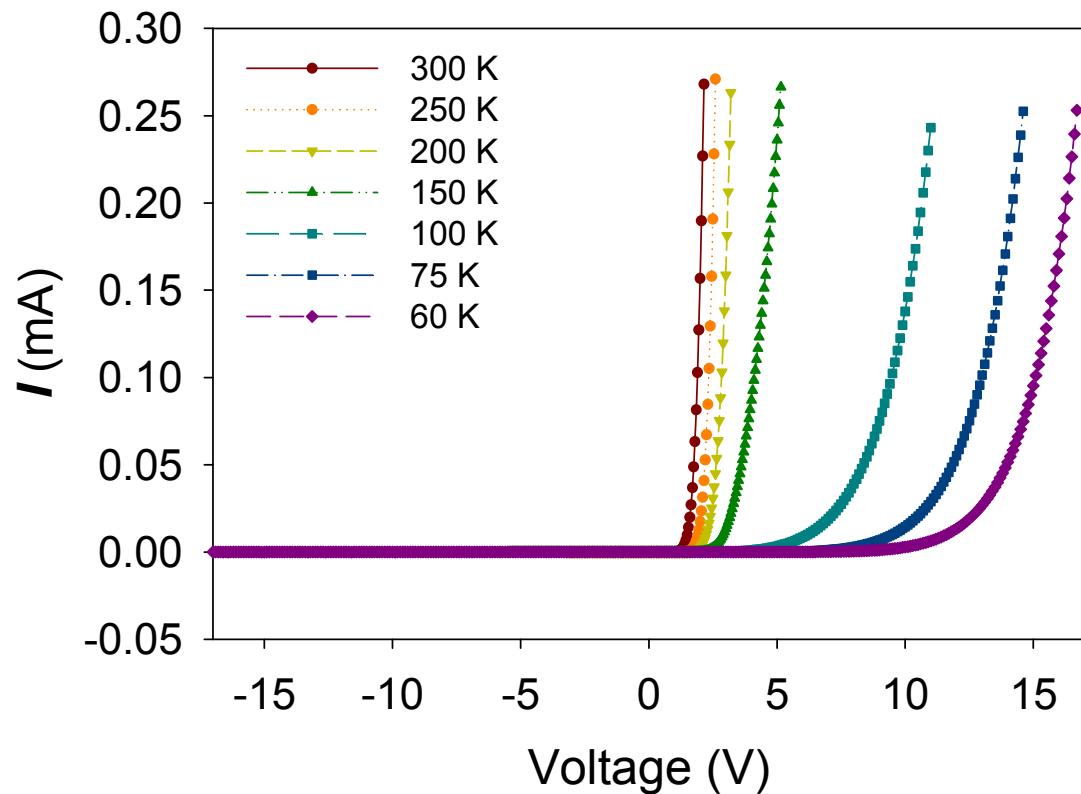
Bare LED Device (no V[TCNE]x~2)



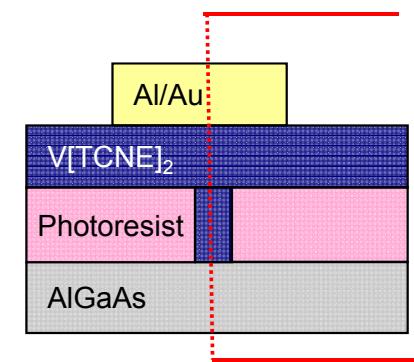
- Good diode behavior
- Low turn on voltage
- Weak temperature dependence

- Positive MR at low temperature
- Negative MR at higher temperatures
- Expected MR behavior for inorganic semiconductors

$V[TCNE]_{x \sim 2}$ Spin-LED Device



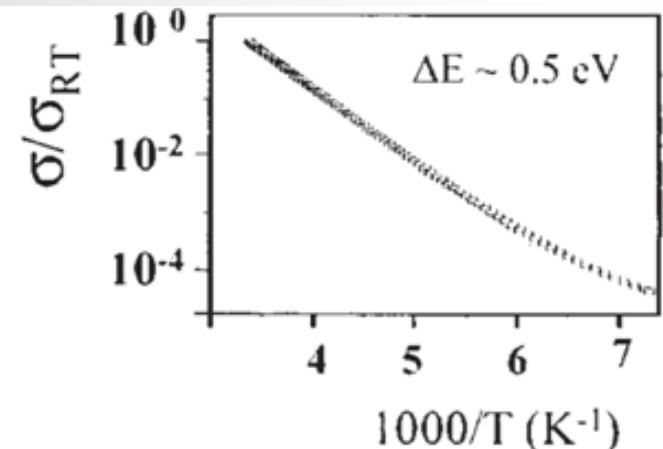
- Good diode behavior
- Strong temperature dependence
- Linear positive MR ($\sim 1\%$ at 5 T):
 - Transport dominated by $V[TCNE]_{x \sim 2}$
 - Consistent w/ transport through $V[TCNE]_x$



Constrained temperature window

High temperature

- Higher conductivity for $V[TCNE]_{x \sim 2}$

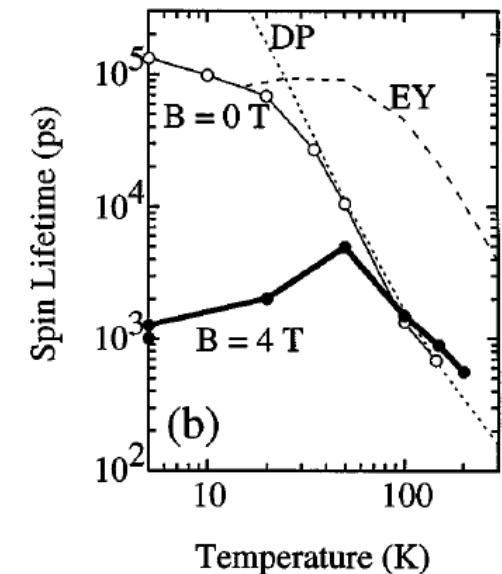
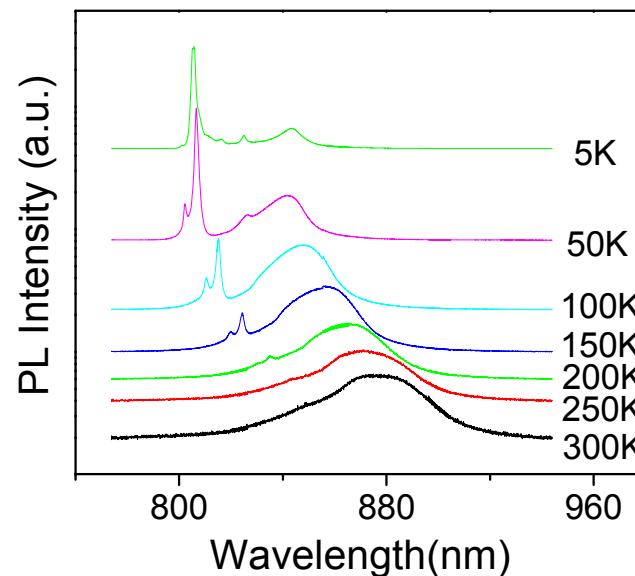


Prigodin V.N, et.al
Adv.Mater. 14, 1230 (2002)

Need to satisfy all constraints: $T \sim 60$ K

Low temperature:

- Bright QW
- Long spin lifetime of LED



J.M. Kikkawa and D.D Awschalom
Phy. Rev. Lett. 80, 4313 (1998)

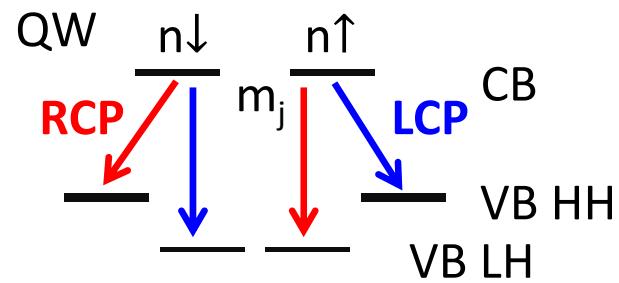
IV and EL results

Electrical Transport:

- Good $p-n$ junction in III-V
- $V[TCNE]_{x \sim 2}$ adds significant impedance

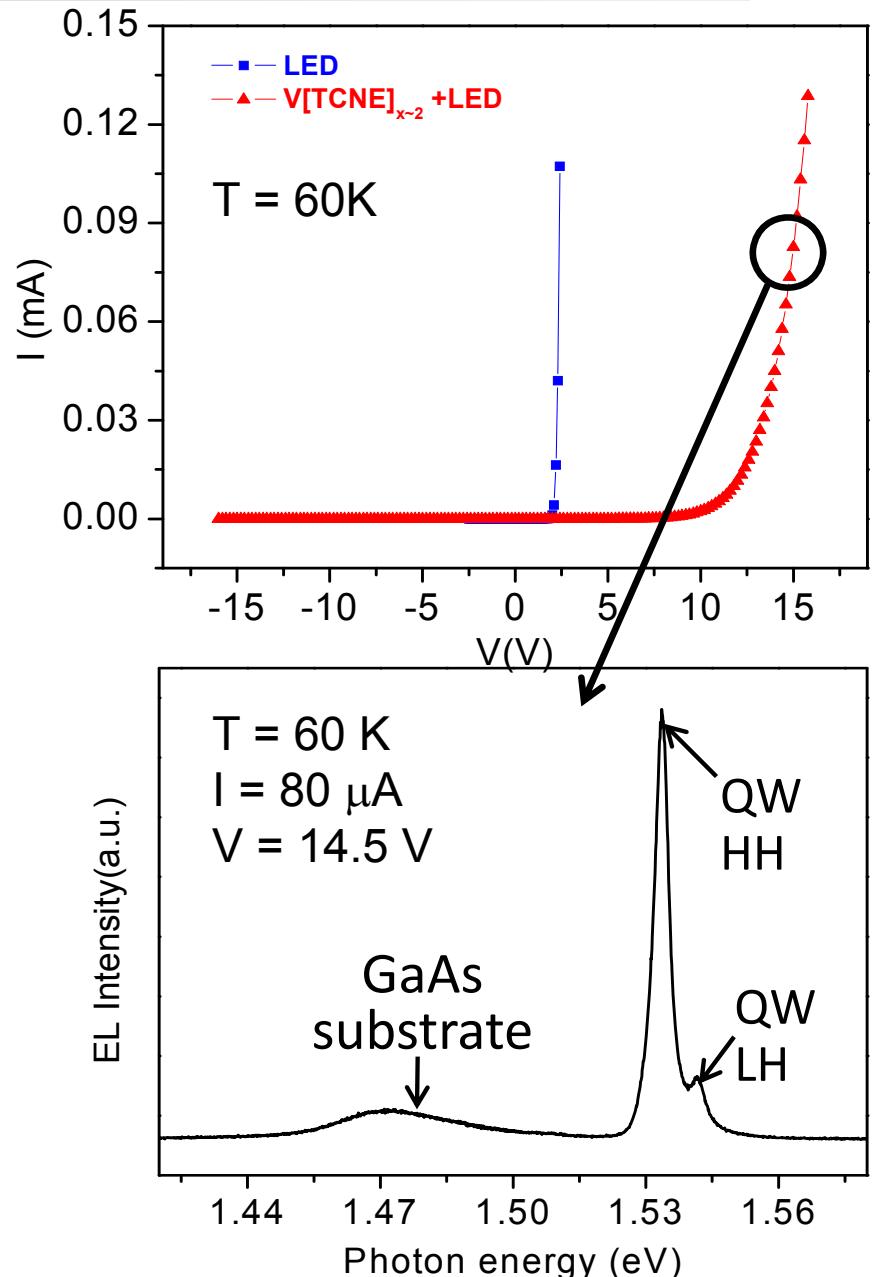
Electroluminescence:

- Three peaks from different transitions

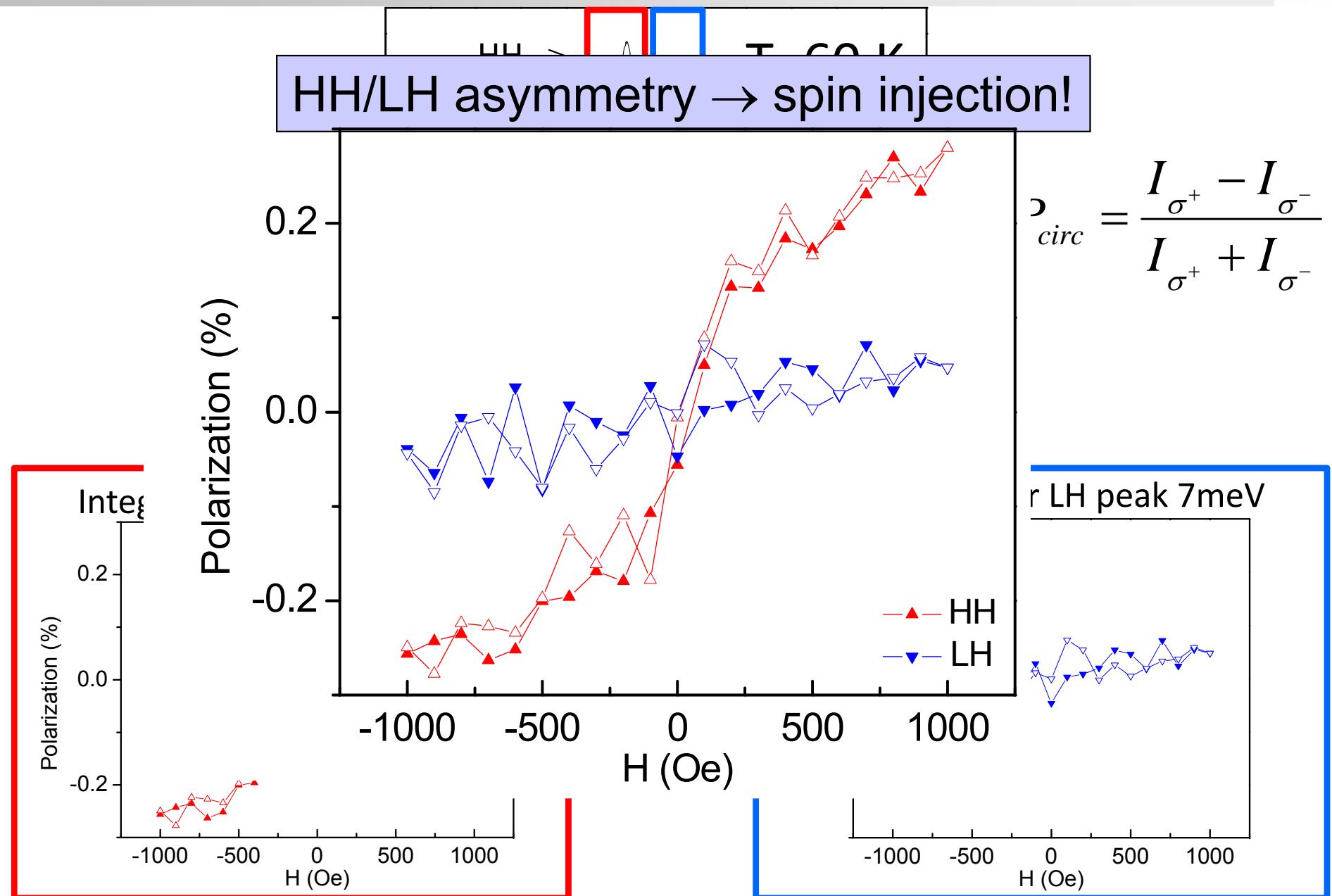


For HH transitions →

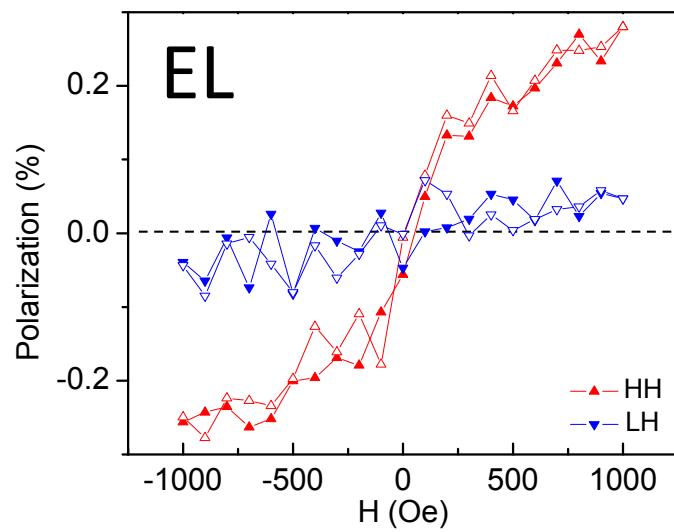
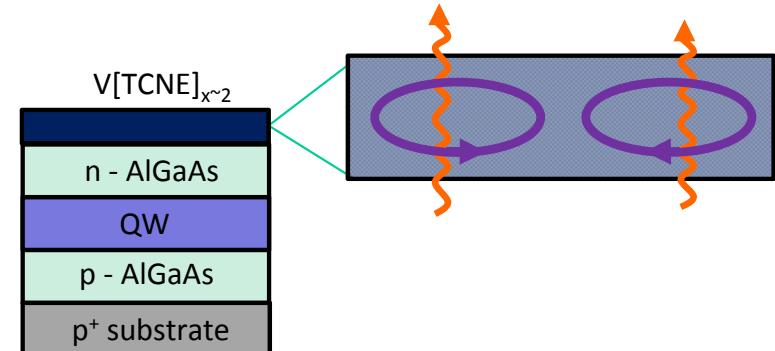
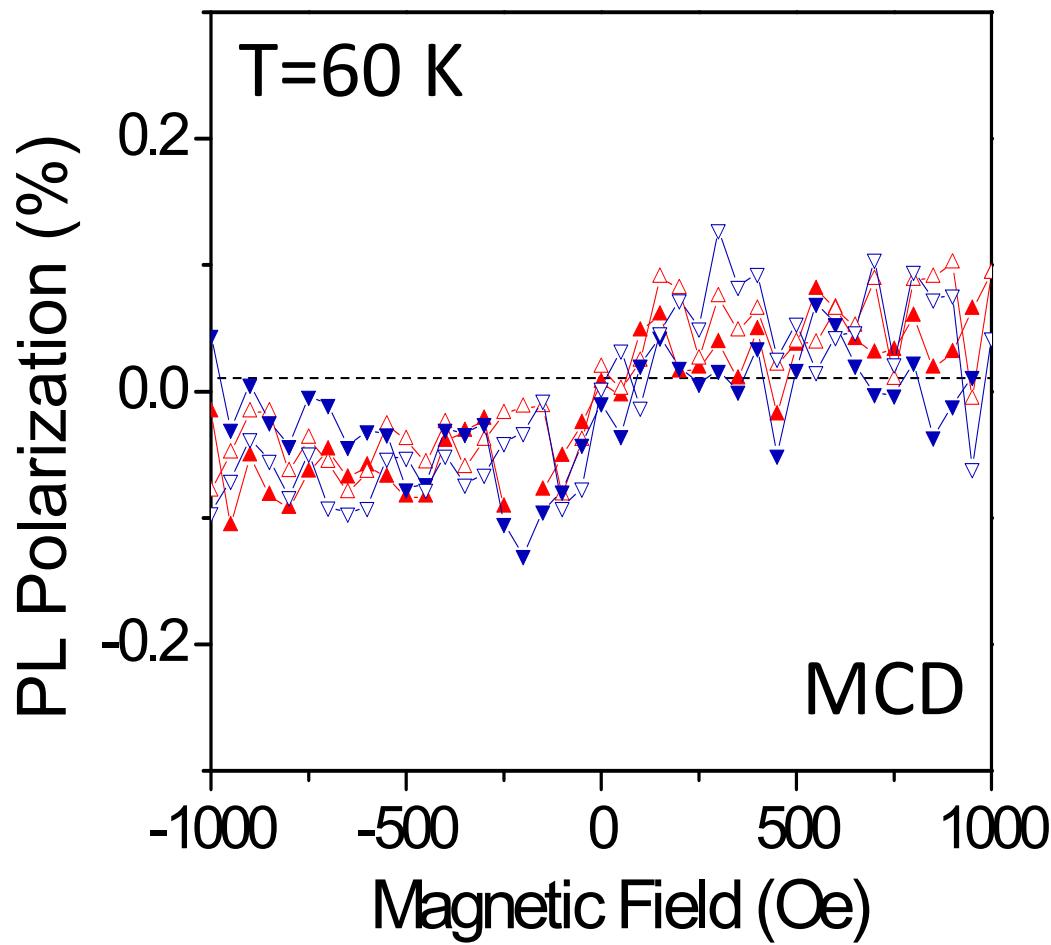
$$P_{circ} = \frac{3n_{\downarrow} - 3n_{\uparrow}}{3n_{\downarrow} + 3n_{\uparrow}} = -P_0$$



Polarization Analysis



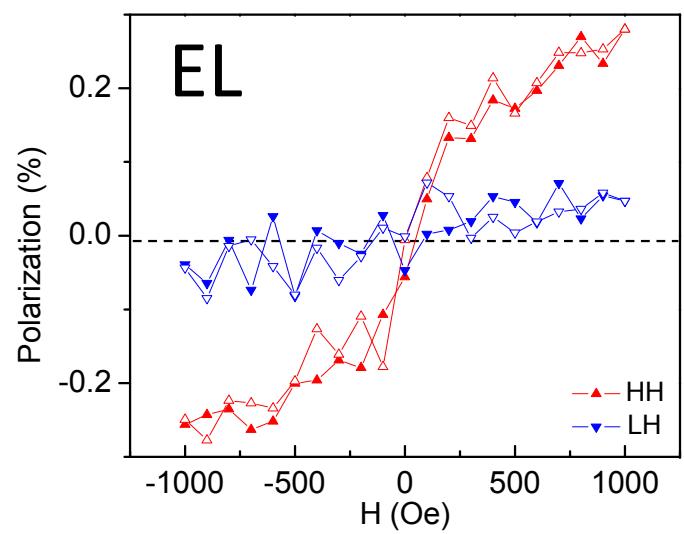
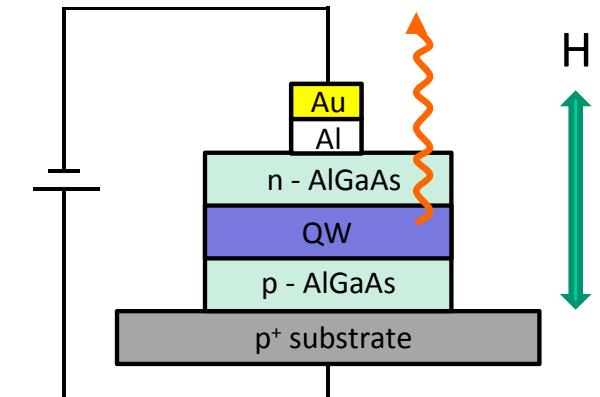
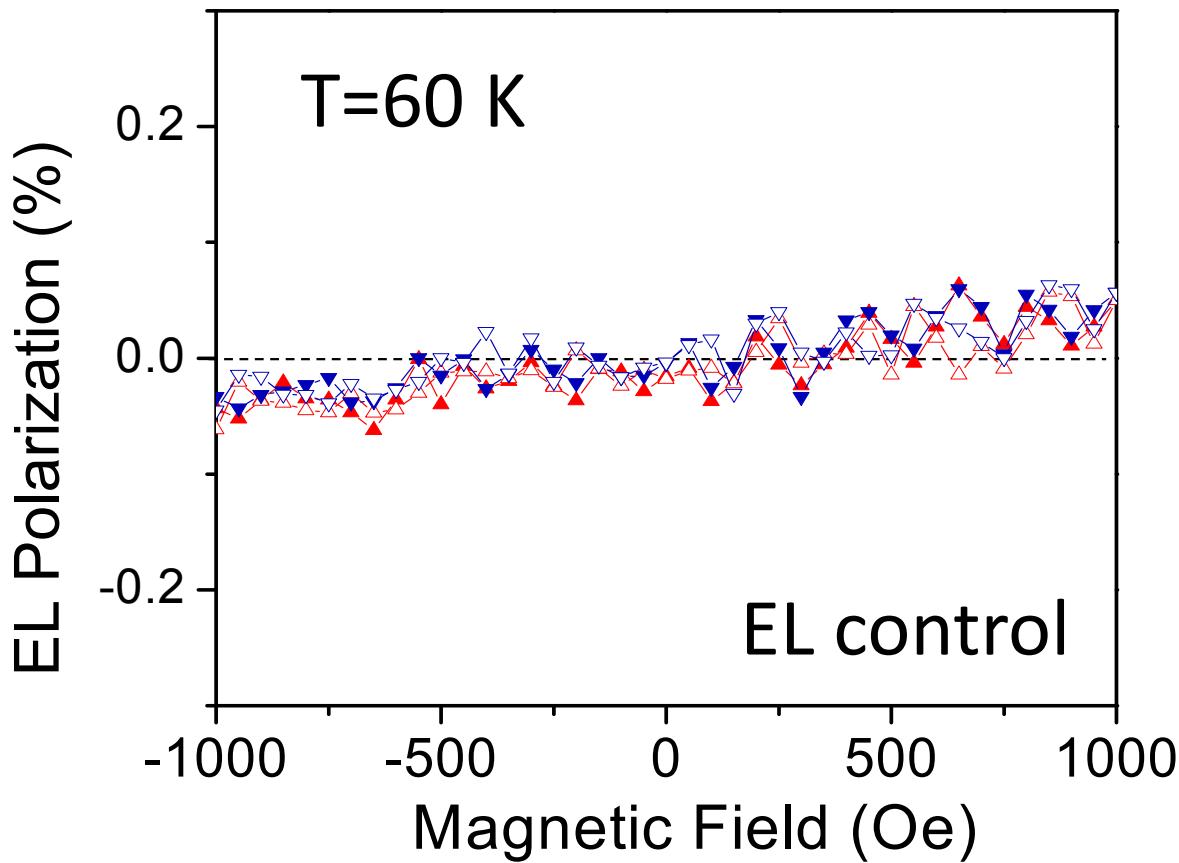
Control measurement (I): Magnetic Circular Dichroism



PL filtered through V[TCNE]_x shows

- weak dichroism
- symmetric HH/LH response

Control measurement (II): Bare LED field dependence

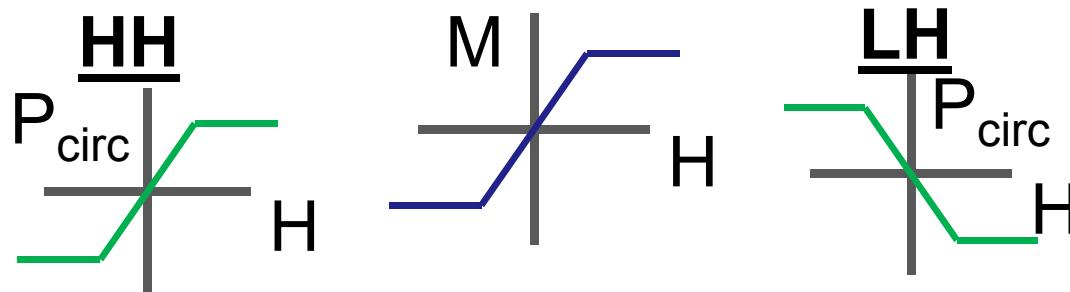


Bare EL shows small linear field dependence due to combination

- zeeman splitting in the quantum well
- dichroism from the cryostat windows

Understanding HH/ LH asymmetry

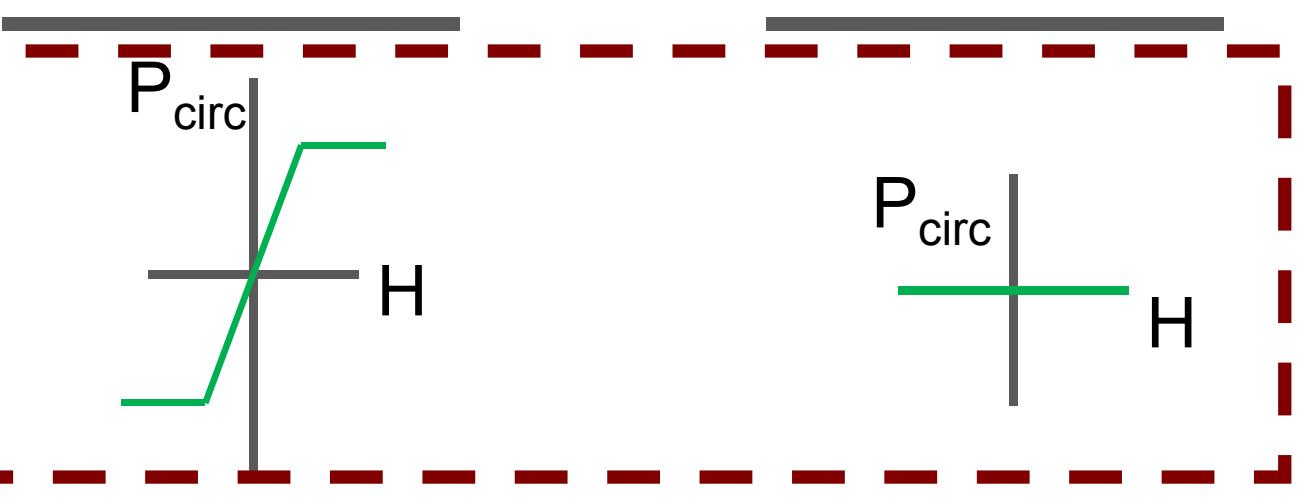
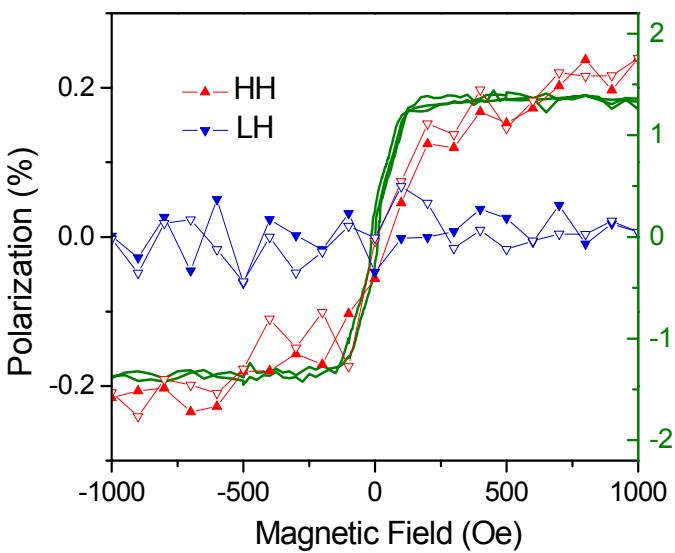
Spin injection



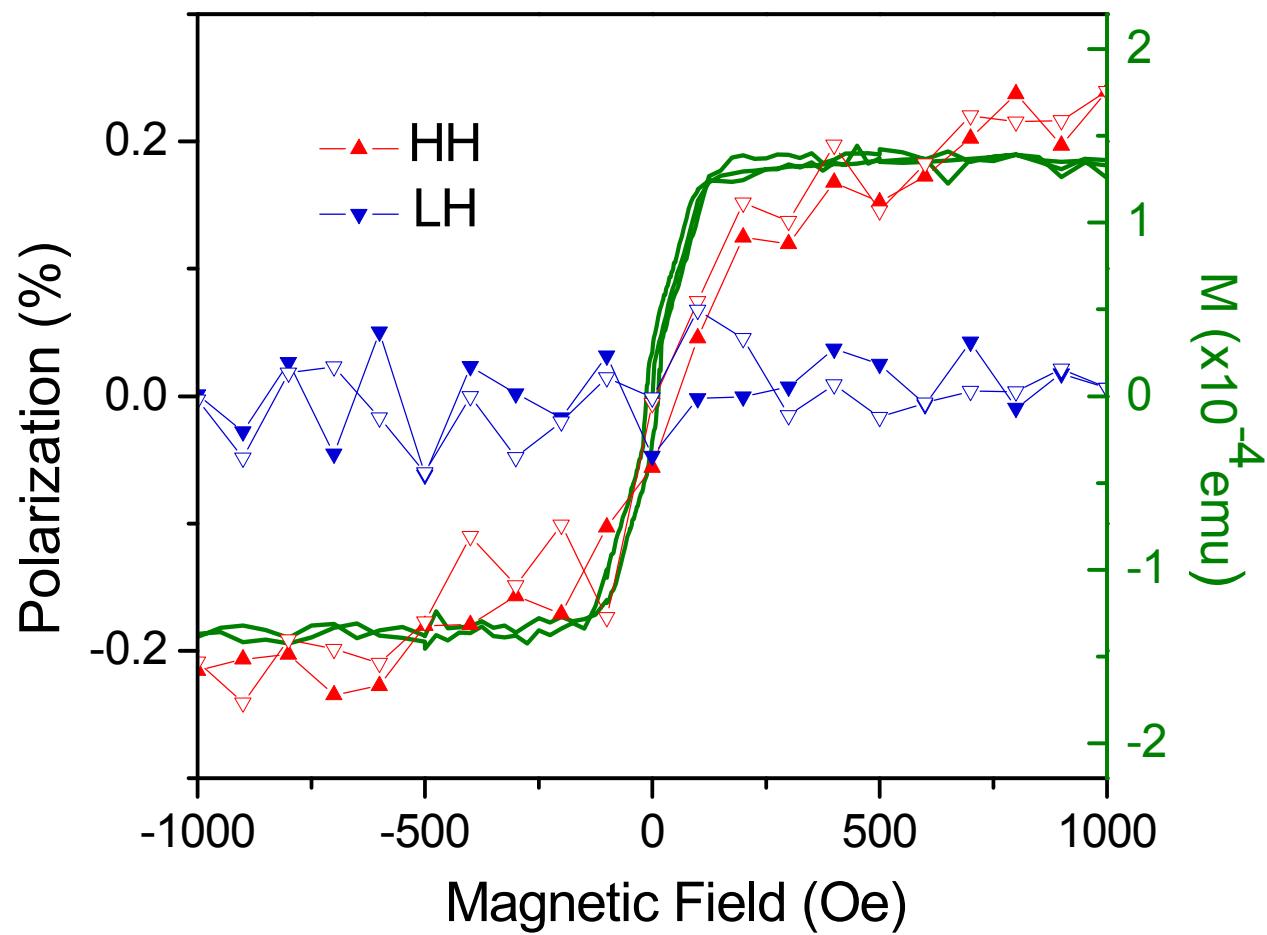
MCD



Measurement



Circularly Polarized EL in $V[TCNE]_2$ spin-LED



- Circular Polarization (CP) of electroluminescence follow hysteresis curve
- Control measurements show lower polarization signal ($P_c < 0.1 \%$)
- Heavy-hole/light-hole asymmetry in CP
- Small signal ($P_{\text{spin}} \sim 0.2 \%$) but can be improved

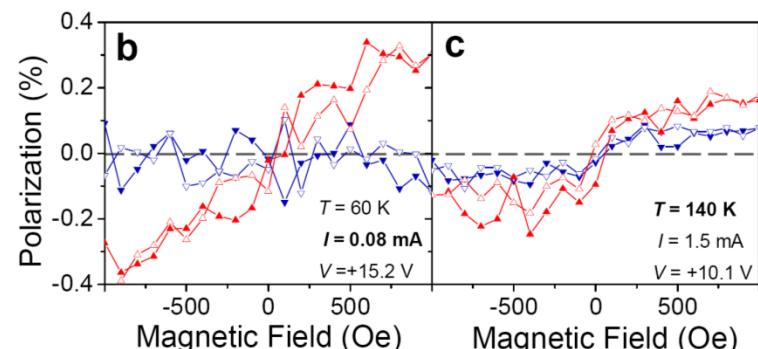
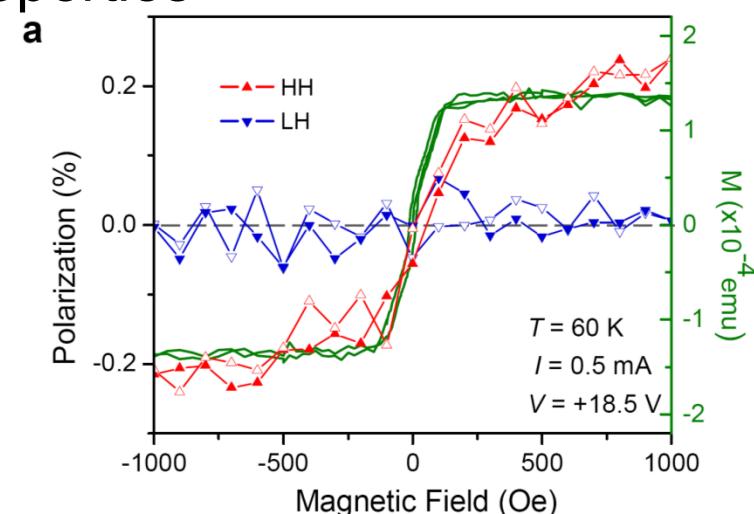
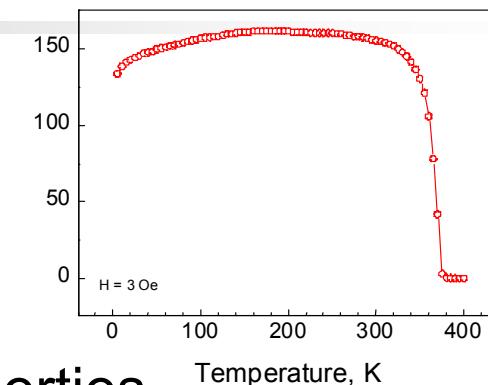
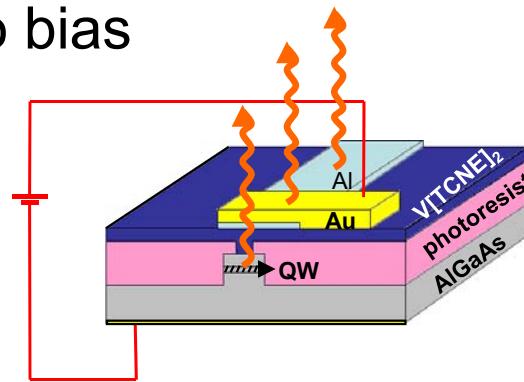
Summary

$V[TCNE]_{x \sim 2}$ is an exciting system

- Room temperature magnetic ordering
- Spin-polarized electronic structure
- Semiconducting transport behavior
- Optical control of magnetic and electronic properties

Possible candidate for spintronic applications

- Demonstrate spin-polarized charge injection from $V[TCNE]_{x \sim 2}$ into inorganic semiconductor through spin LED device structure
- Signal persists up to at least 140 K
- Not sensitive to bias

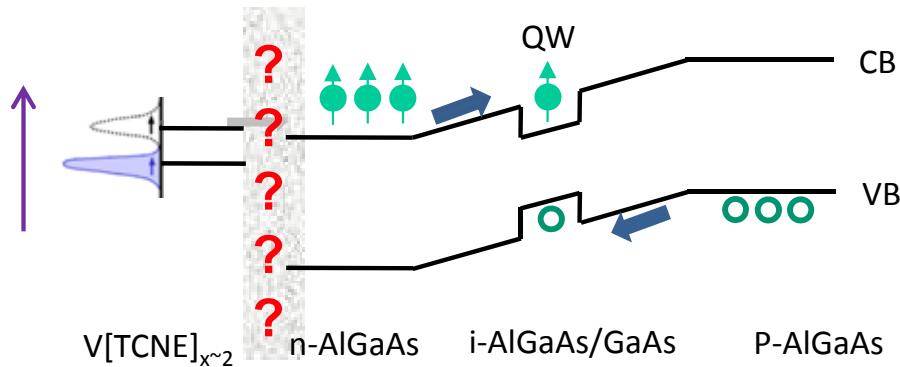


Thank you



Future Work

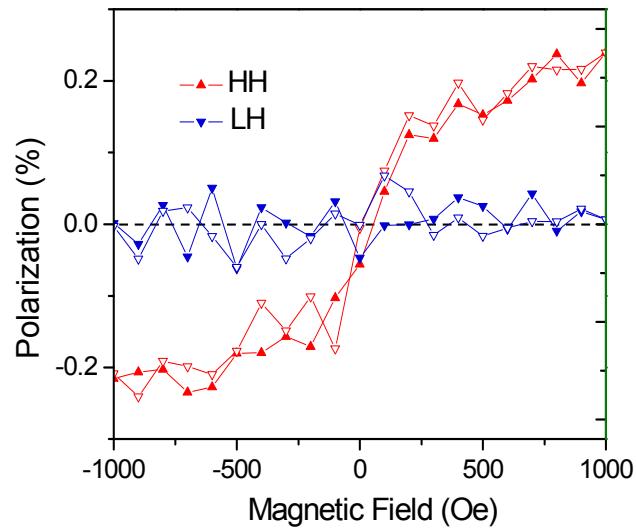
- More experimental & theoretical studies are under study to understand the $V[TCNE]_{x \sim 2}/AlGaAs$ interface



- Using other detector systems (e.g. GaN) in order to achieve room-temperature polarized spin injection

Extracting P_0

$T = 60 \text{ K}$



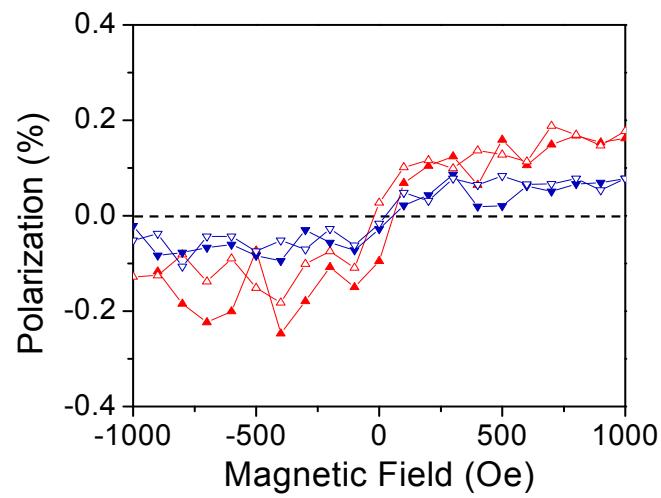
$$P_{circ} = 0.092\%$$

$$P_0 = P_{circ} \times \left(1 + \frac{\tau_r}{\tau_s}\right)$$

$$1 + \frac{\tau_r}{\tau_s} = \frac{1}{5.89\%} = 16.95$$

$$P_0 = 0.092\% \times 16.95 = 1.65 \pm 0.07\%$$

$T = 140 \text{ K}$



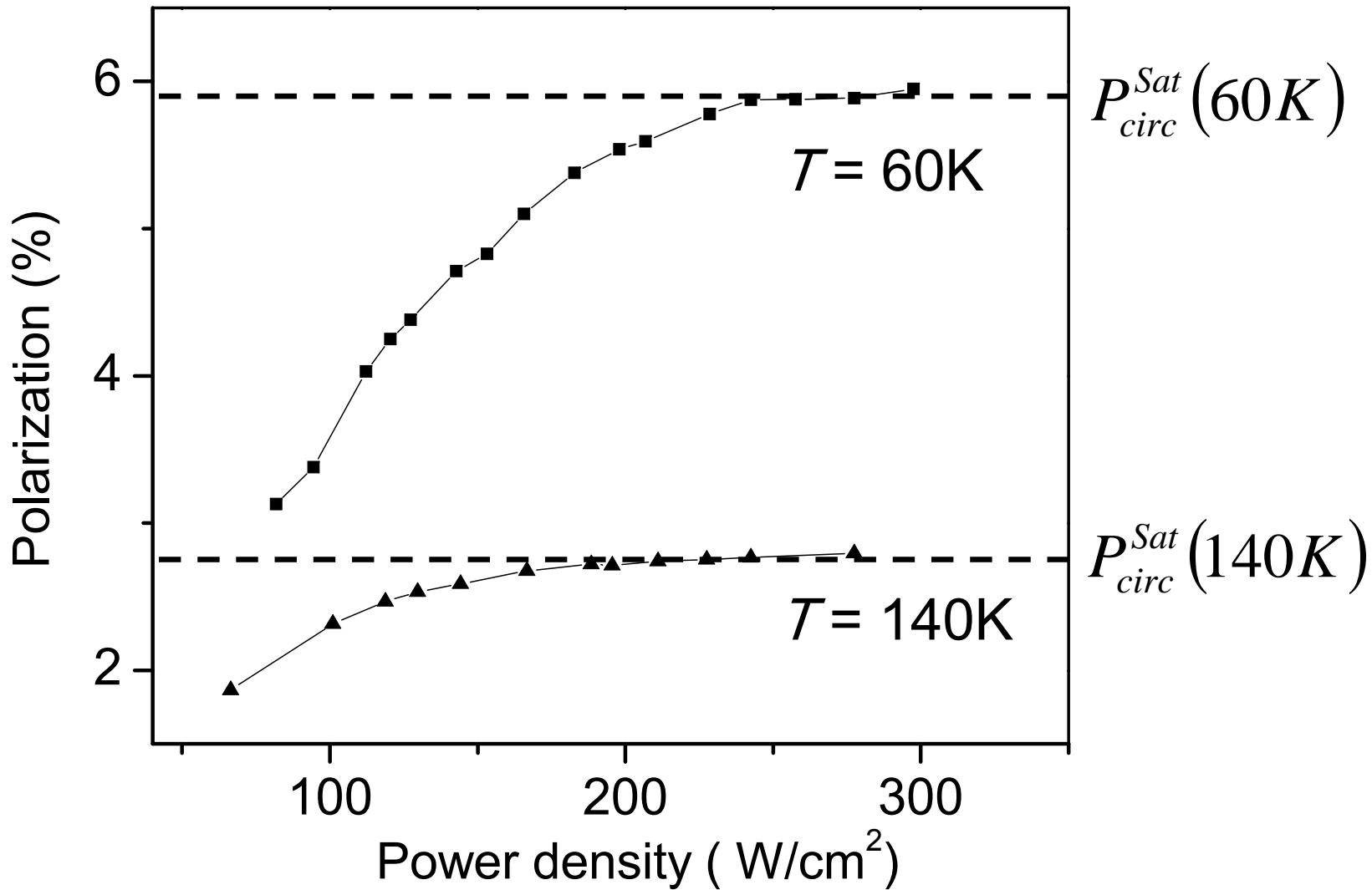
$$P_{circ} = 0.034\%$$

$$P_0 = P_{circ} \times \left(1 + \frac{\tau_r}{\tau_s}\right)$$

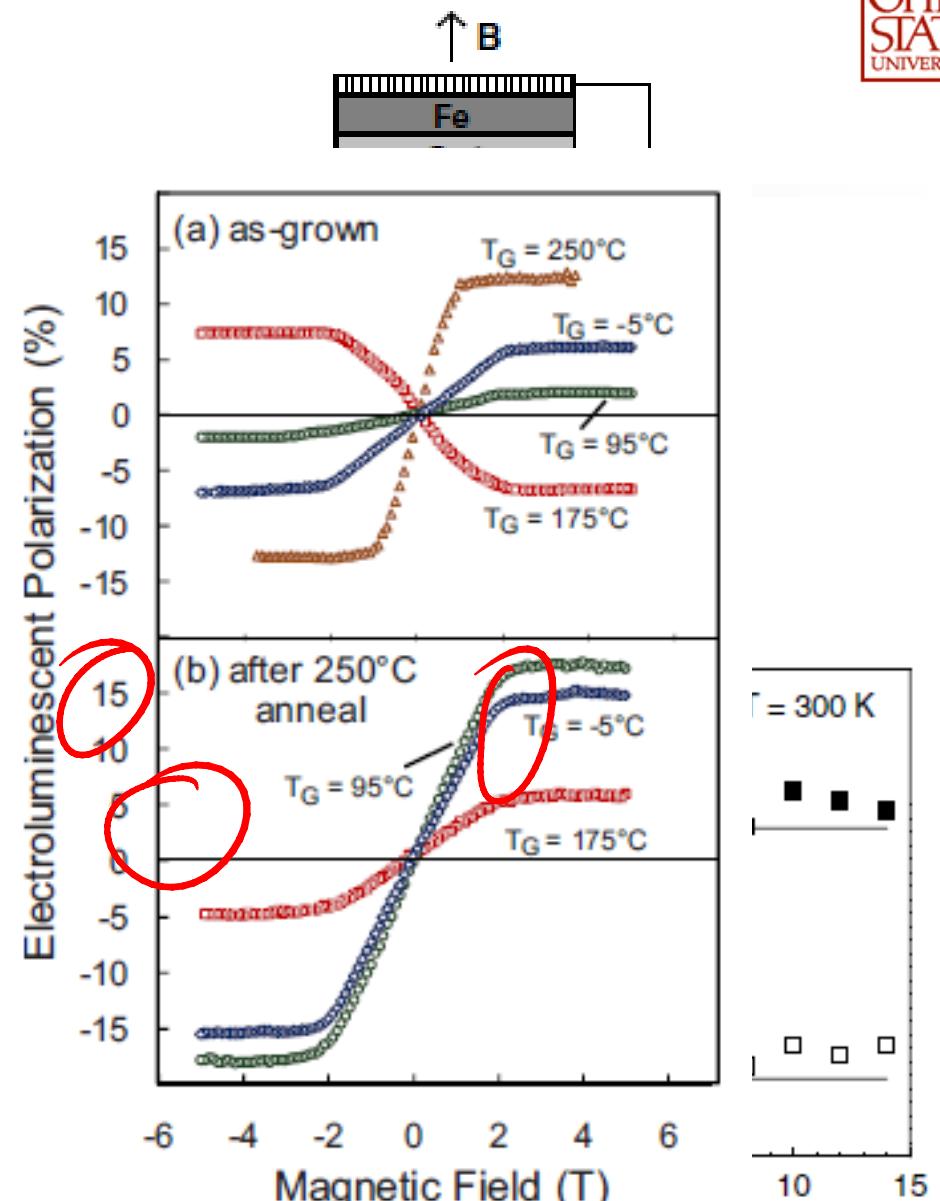
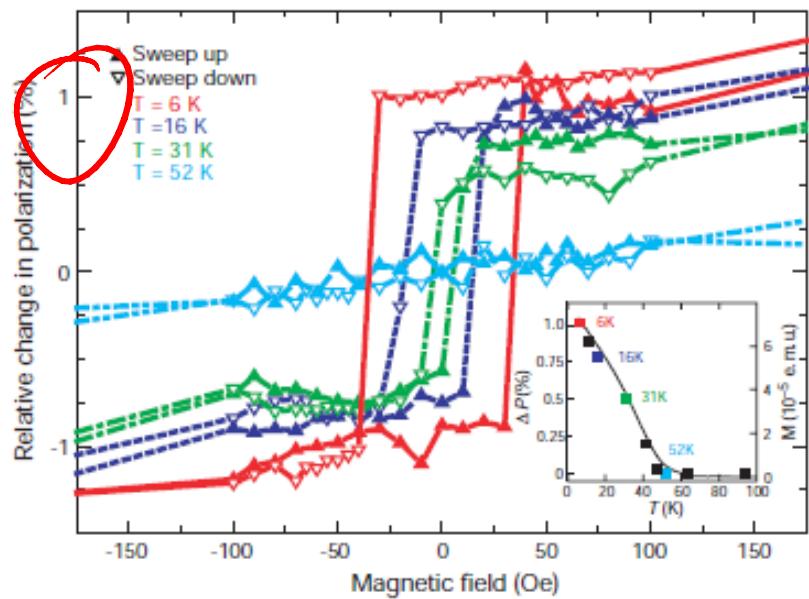
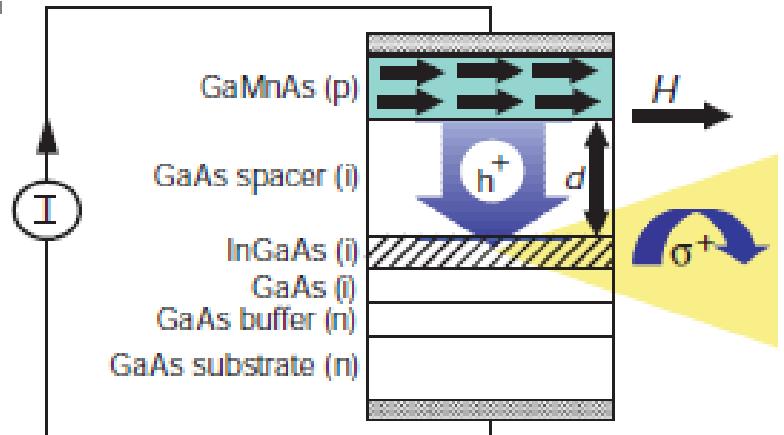
$$1 + \frac{\tau_r}{\tau_s} = \frac{1}{2.76\%} = 36.18$$

$$P_0 = 0.034\% \times 36.18 = 1.32 \pm 0.16\%$$

Extracting P_0 (II)



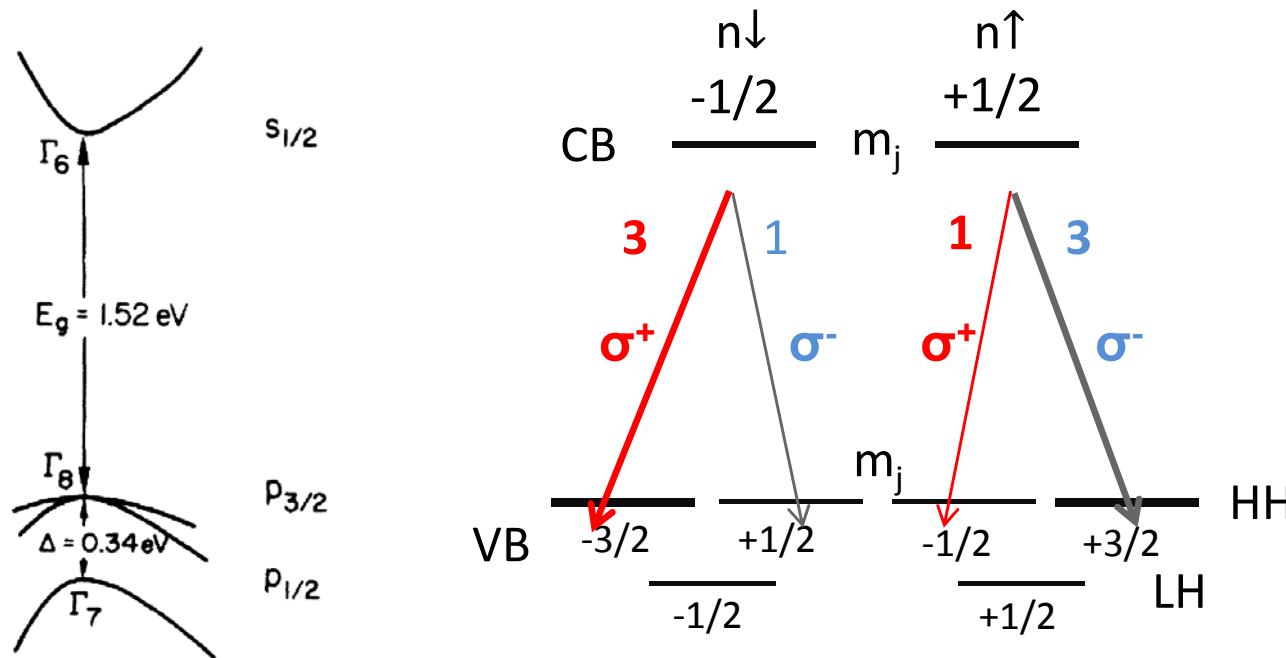
All-inorganic devices



Schultz *et al.*, PRB RC **80**, 201309 (2009)
 Zhu *et al.*, PRL **87**, 16601 (2001)

Quantifying spin injection in LED (I)

Band gap of bulk GaAs



$L=0 \ s=1/2, j=1/2$

$L=1 \ s=1/2, j=3/2, 1/2$

Selection rule: only $\Delta m_j = \pm 1$ transition is allowed

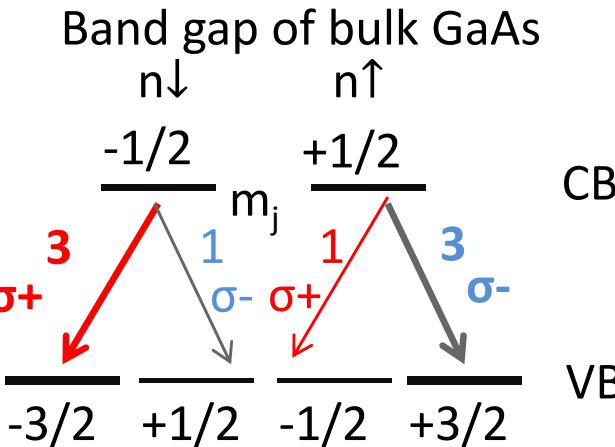
- $\Delta m_j = -1$, σ^+ is emitted
- $\Delta m_j = +1$, σ^- is emitted

Transition possibility

$$\frac{\left| \left\langle \frac{3}{2}, -\frac{3}{2} \left| Y_{1,1} \right| \frac{1}{2}, -\frac{1}{2} \right\rangle \right|^2}{\left| \left\langle \frac{3}{2}, -\frac{1}{2} \left| Y_{1,1} \right| \frac{1}{2}, \frac{1}{2} \right\rangle \right|^2} = 3^{25}$$

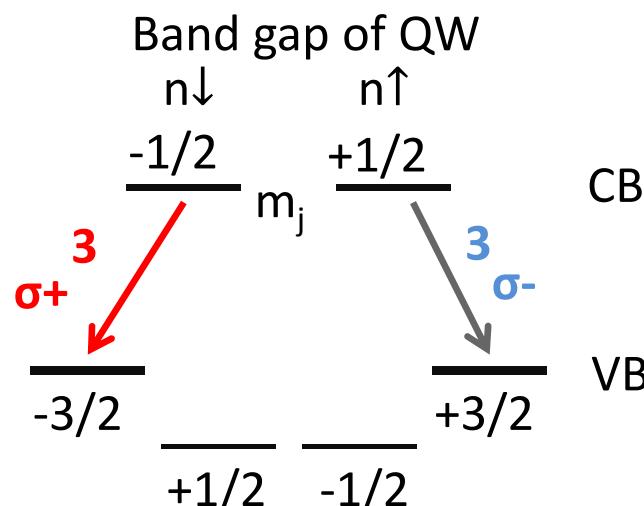
Quantifying spin injection in LED (II)

$$P_{circ} = \frac{I_{\sigma^+} - I_{\sigma^-}}{I_{\sigma^+} + I_{\sigma^-}}$$



- Bulk, degenerate hole bands

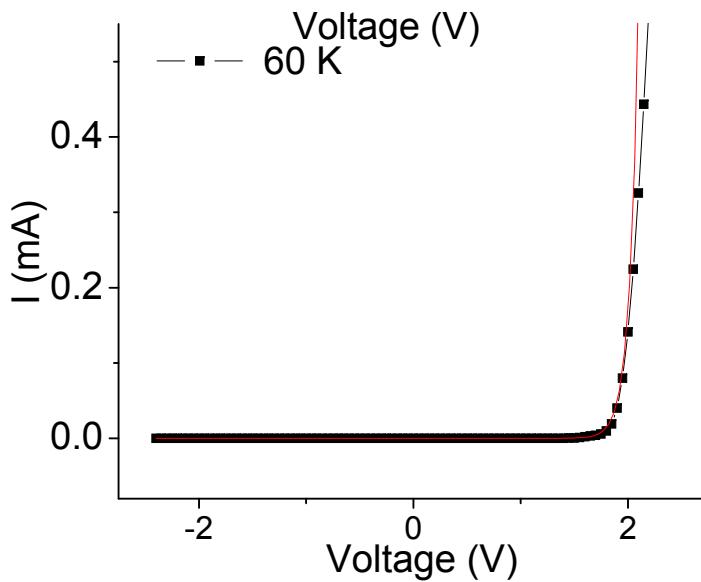
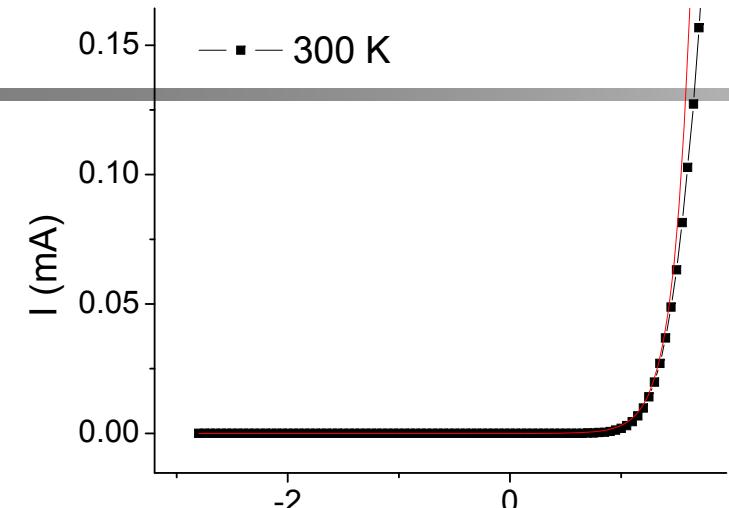
$$\begin{aligned} P_{circ} &= \frac{(3n_{\downarrow} + n_{\uparrow}) - (n_{\downarrow} + 3n_{\uparrow})}{(3n_{\downarrow} + n_{\uparrow}) + (n_{\downarrow} + 3n_{\uparrow})} \\ &= \frac{1}{2} \frac{n_{\downarrow} - n_{\uparrow}}{n_{\downarrow} + n_{\uparrow}} = -0.5P_0 \end{aligned}$$



- QW, no degeneracy

$$\begin{aligned} P_{circ} &= \frac{3n_{\downarrow} - 3n_{\uparrow}}{3n_{\downarrow} + 3n_{\uparrow}} \\ &= \frac{n_{\downarrow} - n_{\uparrow}}{n_{\downarrow} + n_{\uparrow}} = -P_0 \end{aligned}$$

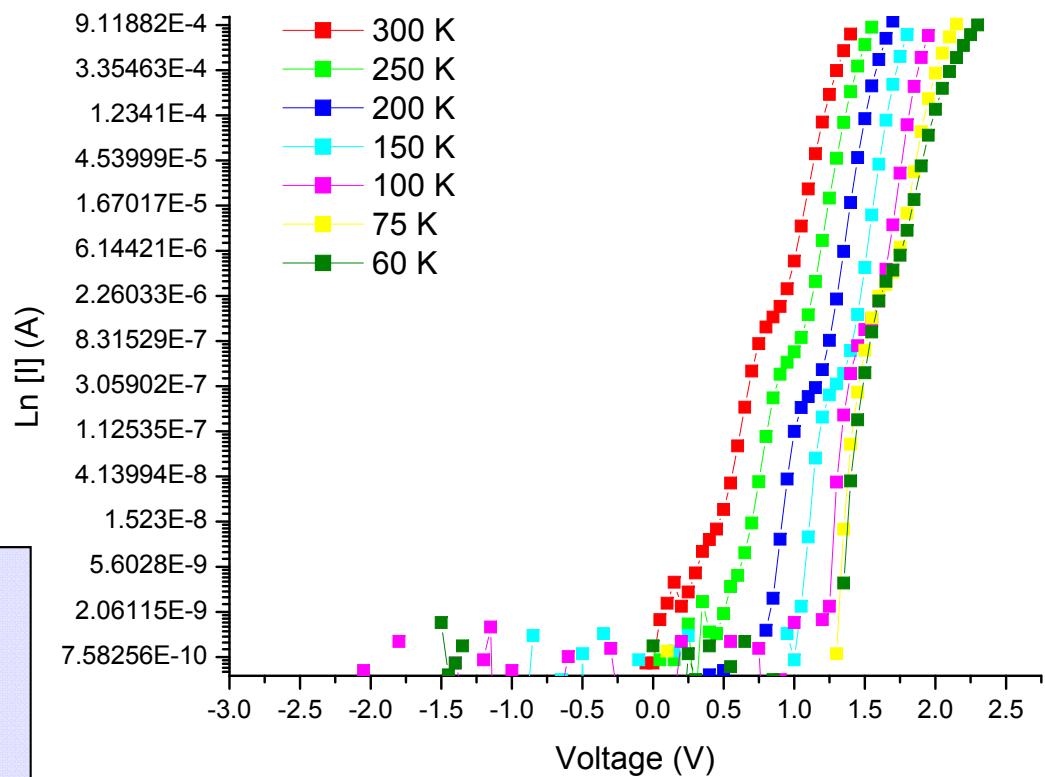
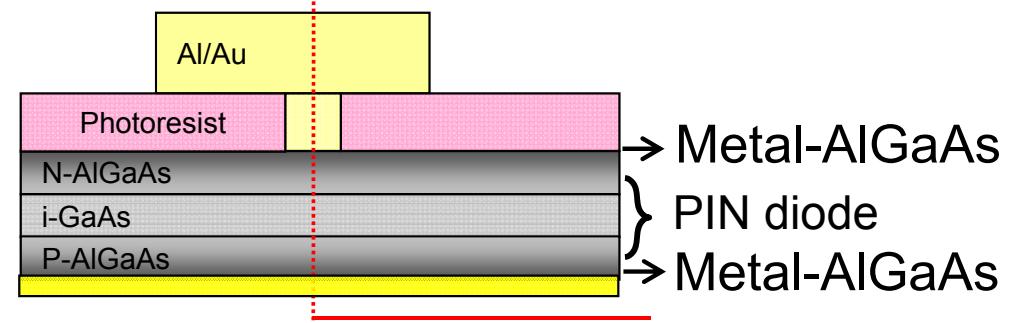
Understanding Interfaces



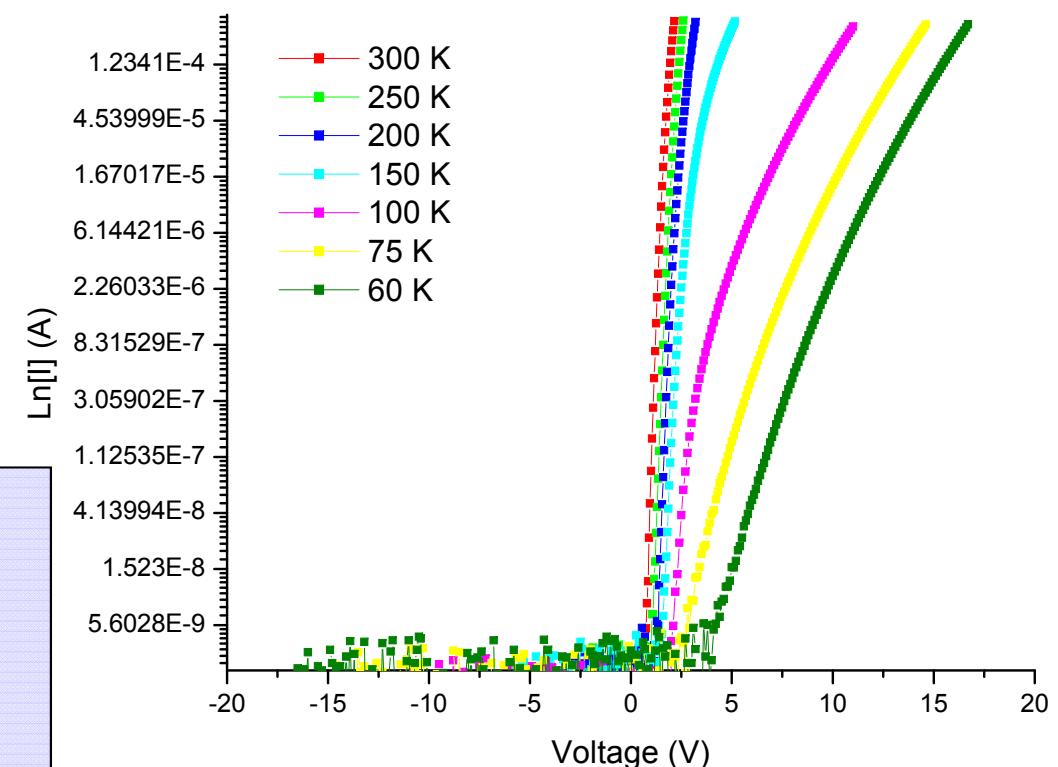
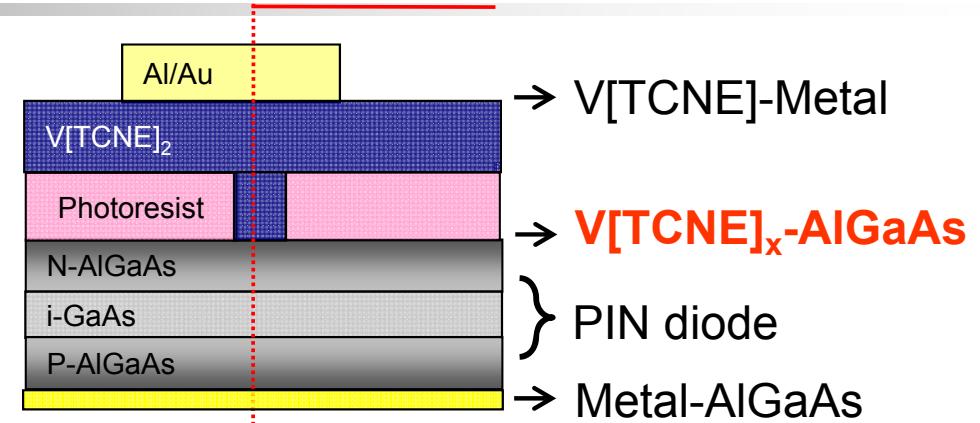
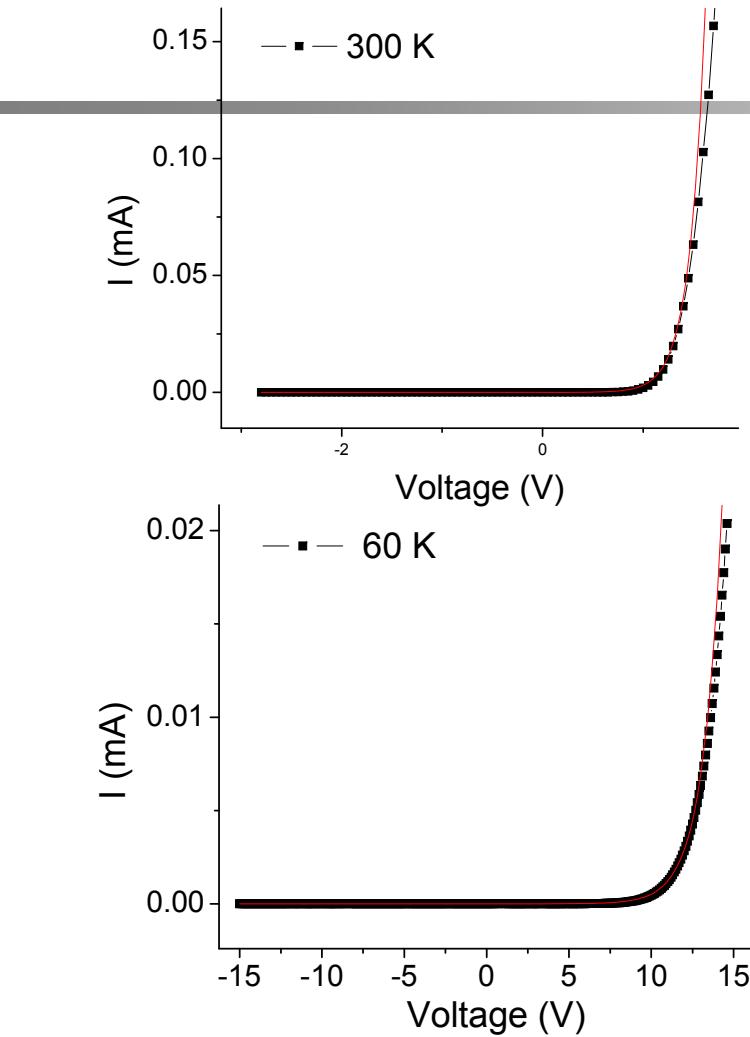
$$I = A^* T^2 \exp[-q\Phi_b/kT] (\exp[q(V-IR)/nkT] - 1)$$

From fitting of diode eqn:

$$\Phi_b \sim 420 \text{ mV}$$



Understanding Interfaces



- Barrier height could not be determined
- Bending of the IV curves; $V[TCNE]_x$ layer additional resistance in the system