Spin filter in spin-dependent recombination and nuclear effects

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Motivation

Attractive ideas:
Spin of carriers and nuclei in semiconductor is supposed to be used for quantum computation and spintronics.

Realization of these ideas at room temperature would certainly benefit their practical implementation.

We propose using spin-dependent Shockley-Read recombination via deep paramagnetic centers in order to achieve very high (up to 100%) both electron and nuclear spin polarization in nonmagnetic semiconductors at room temperature and zero and weak magnetic fields.
Problem:
Spin relaxation time of conduction electrons in nonmagnetic direct-gap semiconductors
\[ \tau_s < 100 \text{ ps at } T=300 \text{ K} \]

Striking result:
We have found at \( T=300 \text{ K} \)
in GaAsN alloys \([N]=0.6\div3.5\%\)
spin polarization of free electrons \( P \sim 90 \% \)
spin lifetime \( T_s > 1 \text{ ns} \)
Optical orientation in semiconductors

Interband excitation by circular polarized light; \( \Delta m = + \hbar \) for \( \sigma^+ \) light

Luminescence: \( \rho \propto P \)

\[ P = \frac{n_+-n_-}{n_++n_-} \]

\[ \rho = \frac{J^+ - J^-}{J^+ + J^-} \]

Optical orientation, eds. F. Meier and B. Zakharchenya (North-Holland, Amsterdam, 1984)


GaAs\(_{1-x}\)N\(_x\), \( x = 0.6\div3.5\% \)

Ga(In)As\(_{1-x}\)N\(_x\) films 0.1\( \mu \)m

\( T = 300 \)K

\( \rho \propto P \)

GaAs\(_{0.98}\)N\(_{0.02}\)
PL circular polarization in GaAsN
continuous-wave pumping

$P = 2\rho \sim 60\% > P_{\text{theor}} = 50\%$

$P = \frac{n_+ - n_-}{n_+ + n_-}$

$\rho = \frac{J^+ - J^-}{J^+ + J^-}$


Spin dynamics in GaAs$_{0.98}$N$_{0.02}$ under pulse (1.5 ps) pumping

Time (ps)

PL intensity (a. u.)

PL circular polarization (%)

$P \sim 90\%$

electron spin polarization

$P = \frac{n_+ - n_-}{n_+ + n_-}$

$\rho = \frac{J^+ - J^-}{J^+ + J^-}$

Spin-dependent recombination in GaAs$_{0.98}$N$_{0.02}$

continuous wave pumping

For a review on GaNAs properties, see e.g., *Dilute III-V Nitride Semiconductors and Material Systems*, Springer Series in Material Science Vol. 105, ed. A. Erol (Springer, Berlin, 2008)
Outline

• Introduction
• Spin-dependent recombination (SDR) as an origin of a giant electron spin-polarization:
  (a) continuous-wave pumping
  (b) pulsed pumping
• SDR model and comparison with experiment
• Spin-dependent photoconductivity and ODEMR
• Enhancement of electron spin-polarization by a longitudinal magnetic field
• Dynamic nuclear spin-polarization and hyperfine nuclear magnetic fields
• Summary
The source of the SDR is the Pauli exclusion principle:
new electron cannot be captured on the energy state if the state is already
occupied by the other electron with the same spin orientation.

When electrons are polarized
electron-capture cross-section depends on their polarizations: \( \sigma_e = \sigma_0(1-PP_c) \)

- **S. Geschwind et al., PRL (1959):**
  ODEPR in fluorescence of Cr\(^{3+}\) excited states in Al\(_2\)O\(_3\).

- **D. Lepine, PRB (1972):**
  EPR of paramagnetic centers on the surface of crystalline silicon;
  observed in photoconductivity.

- **T. Wosinski et al., phys. status solidi (1976):** SDR in dislocated silicon.

Model of Spin-Dependent Recombination

\[ P_i = 50\% \]

1) Deep centers are paramagnetic and unpolarized without light.
2) Recombination of free electrons with deep centers is dominant.
3) A capture of electron with parallel spin is forbidden.
4) Holes are unpolarized and their capture is spin-independent.
5) \( \tau_{sc} \gg \tau_s \)

- Weisbuch and Lampel, SSC 14, 141 (1974)
  AlGaAs epilayer, 77K
- Miller et al., PRB 21, 1569 (1980)
  AlGaAs/GaAs SL, 4K
- Paget, PRB 30, 931 (1984)
  GaAs bulk, 4K
1) deep centers are paramagnetic and unpolarized without light
2) recombination of free electrons with deep centers is dominant
3) a capture of electron with parallel spin is forbidden
4) holes are unpolarized and their capture is spin-independent
5) $\tau_{sc} \gg \tau_s$

- Weisbuch and Lampel, SSC 14, 141 (1974)
Dynamic polarization of electron spins

1) Increase of $\rho$ and $J$ with increasing pump power

2) Decrease of $J$:
   (a) $\text{circ} \rightarrow \text{lin}$, $B=0$
   (b) $\text{circ}$, $B_{\perp} \neq 0$
Spin-filtering effect vs excitation power

$P = 2\rho \sim 60\%$

$Ivchenko, Kalevich, Shiryaev, Afanasiev, Masumoto,$


$\tau_{SC} \gg \tau_S$
Spin-dependent recombination in GaAs$_{0.98}$N$_{0.02}$

continuous wave pumping

1.05 1.10 1.15 1.20 1.25

PL intensity (rel. units)

Energy (eV)

1 - circ , $B=0$
2 - lin , $B=0$
3 - circ , $B = 1$ kG

$B \perp z$

T=300K

$W = 150$ mW

MBE grown at Ioffe Institute

Kalevich, Ivchenko, Afanasyev, Shiryaev, Egorov, Ustinov, Pal and Masumoto, JETP Lett. 82, 455 (2005)

$\tau_{SC} \gg \tau_{S}$
Hanle effect under SDR

\[ B \perp z, \quad \Omega = g \mu_B B / \hbar \]

\[ \rho(B) = \frac{1}{1 + (\Omega T_s)^2} = \frac{1}{1 + (B / B_{1/2})^2} \]

\[ B_{1/2} = \frac{\hbar}{g \mu_B T_s} \]

\[ \frac{1}{T_s} = \frac{1}{\tau_s} + \frac{1}{\tau} \]

\[ \tau_{sc} \gg \tau_s \]

\[ \tau_c \gg \tau \]

\[ T_{sc} \gg T_s \Rightarrow B_{1/2}^c \ll B_{1/2} \]

\[ B_{1/2}^c = 100 \text{G} \]

\[ B_{1/2} = 25000 \text{G} \]

\[ g_c = +2 \]

\[ g = +1 \]

\[ T_{sc} \sim 1 \text{ ns} \]

\[ T_s \sim 1 \text{ ps} \]

\[ \tau_{sc} \sim 1 \text{ ns} \]

\[ \tau \sim 1 \text{ ps} \]

Spin stability of the coupled system of spin-polarized free and bound electrons is controlled by the long spin relaxation time of bound electrons \( \tau_{sc} \approx 1\,\text{ns} \gg \tau_s \approx 100\,\text{ps} \).
Hanle effect in intensity

continuous wave pumping

![Graph showing the Hanle effect in intensity for free and bound electrons under continuous wave pumping.](image)
Spin dynamics in GaAs$_{0.98}$N$_{0.02}$ under pulse (1.5 ps) pumping

$P \sim 90\%$

$T=300K$
Spin dynamics in $\text{GaAs}_{0.98}\text{N}_{0.02}$ under pulse (1.5 ps) pumping

$T=300\text{K}$
$130\text{mW}$
$B=0$

Intensity (a.u.)
Time (ps)
Circular polarization (%)
PL kinetics under strong SDR

pulse pumping: \( n, p >> N_c \), \( n, p \approx 30N_c \)

Fast decay: \( \tau_h \)

\[
\begin{align*}
n_+ \quad & \quad \downarrow \uparrow \\
\rightarrow \quad & \quad N_2 \approx N_c \\
p_+ \quad & \quad \uparrow \downarrow \\
p_+ \quad & \quad \uparrow \downarrow \\
p_+ \quad & \quad \uparrow \downarrow \\
p_+ \quad & \quad \uparrow \downarrow
\end{align*}
\]

\( n, p \propto \exp(-t/\tau_h) \)

\( J \propto np \propto \exp(-2t/\tau_h) \)

Slow decay: \( \tau_s \)

\[
\begin{align*}
n_+ \quad & \quad \uparrow \\
\rightarrow \quad & \quad N_+ \approx N_c \\
p_+ \quad & \quad \uparrow \\
p_+ \quad & \quad \uparrow \\
p_+ \quad & \quad \uparrow
\end{align*}
\]

\( n, p \propto \exp(-t/\tau_s) \)

\( J \propto np \propto \exp(-2t/\tau_s) \)
Measurement of $\tau_s$ from PL decay

$\tau_{h\min} = (30\pm1) \text{ ps}$ \quad \Rightarrow \quad R_{hN_c} = 1/\tau_{h\min} = 0.033 \text{ ps}^{-1}$

$\tau_s > 1 \text{ ns}$

$\tau_s = (140\pm4) \text{ ps}$

$[N] = 2\%$

$T=300\text{K}$

$130\text{mW}$

$B=0$

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SDR model for the arbitrary excitation intensity and magnetic field

\[
\frac{dn_{\pm}}{dt} = G_{\pm} - R_e n_{\pm} N_\mp - \frac{n_{\pm} - n_{\mp}}{2\tau_s} + \gamma n_{\pm} p
\]

\[
\frac{dN_{\pm}}{dt} = \frac{R_h}{2} pN_2 - R_e n_{\mp} N_\pm - \frac{N_{\pm} - N_\mp}{2\tau_{sc}}
\]

\[
p = n + N_2
\]

\[
n = n_+ + n_- , \quad N_1 = N_+ + N_- , \quad N_c = N_1 + N_2
\]

- \( n, S \) are the total free-electron density and spin, \( S = (n_+ - n_-)/2 \)
- \( G \) is the photogeneration rate of free electrons; \( G = G_+ + G_- \)
- \( p \) is the free-hole density,
- \( N_c \) is the total density of deep centers,
- \( N_1, S_c \) are the density and total spin of centers with 1 electrons, \( S_c = (N_+ - N_-)/2 \)
- \( R_e, R_h \) are the recombination constants of free electrons and holes,
- \( \tau_s, \tau_{sc} \) are the spin relaxation times of free and bound electrons
SDR model for the arbitrary excitation intensity and magnetic field

\[
\frac{dn}{dt} = G - \frac{R_e}{2} (nN_1 - 4\vec{S}\vec{S}_c)
\]

\[
\frac{dp}{dt} = G - R_h (N_c - N_1) p
\]

\[p = n + N_c - N_1\]

\[
\frac{d\vec{S}}{dt} = \vec{S}_i + \frac{R_e}{2} (\vec{S}_c n - \vec{S}N_1) - \frac{\vec{S}}{\tau_s} + (\vec{\Omega} \times \vec{S})
\]

\[
\frac{d\vec{S}_c}{dt} = \frac{R_e}{2} (\vec{S}N_1 - \vec{S}_c n) - \frac{\vec{S}_c}{\tau_{sc}} + (\vec{\Omega}_c \times \vec{S}_c)
\]

\[\Omega = g\mu_B B / \hbar\]

\[\Omega_c = g_c\mu_B B / \hbar\]

\[\vec{S}_i = \frac{nP_i}{2} \vec{\delta}_z\]

Polarization oscillations in a transverse magnetic field

pulse (1.5 ps) pumping

$|g| \approx 1$

$B = 7.8 \text{ kG}$

$\mu \approx 1$

$[N]=2\%$

$T=300\text{K}$

$I_-\text{PL circular polarization}$

Kalevich, Ivchenko, Shiryaev, Egorov, Lombez, Lagarde, Marie and Amand, JETP Lett. 85, 174 (2007)
Measured and estimated parameters

g = +1 (from polarization oscillations; sign from the Hanle effect)

gc = +2 (measured by ODMR; sign from the Hanle effect)

\( \tau_s = 140 \text{ ps} \) (from PL kinetics)

\( \tau_{sc} = 700 \text{ ps} \) (from the Hanle effect)

\( R_e N_c = 2/\tau_{\text{min}} = 1 \text{ ps}^{-1}, \quad \tau_{\text{min}} = 2 \text{ ps} \) (from the Hanle effect)

\( R_h N_c = 1/\tau_{h\text{ min}} = 0.033 \text{ ps}^{-1}, \quad \tau_{h\text{ min}} = 30 \text{ ps} \) (from PL kinetics)

\( N_c = ? \)

\( P_i = ? \)

Estimation of $N_c$ and $P_i$

(a) $\frac{J(\text{circ})}{J(\text{lin})}$

$P_i = 0.24$

(b) $\rho$ (%)

$B = 0$

$N_c = 3 \cdot 10^{15} \text{ cm}^{-3}$

$P_i = 24\%$

Calculated Hanle effect

**Computation**

- **Bound electrons (a)**
  - $W (\text{mW})$: 220 (red), 100 (green), 50 (blue), 25 (black)
  - $\tau_s = 140$ ps
  - $\tau_{sc} = 700$ ps
  - $N_c = 2 \times 10^{15} \text{ cm}^{-1}$
  - $R_e N_c = 1 \text{ ps}^{-1}$
  - $R_h N_c = 0.033 \text{ ps}^{-1}$
  - $g = +1$, $g_c = +2$
  - $P_i = 24\%$

- **Free electrons (b)**

**Experiment**

- **Bound electrons (a)**
  - $W (\text{mW})$: 220 (red), 100 (green), 50 (blue), 25 (black)

- **Free electrons (b)**
Kinetics of $J$ and $\rho$

$[N] = 2\%$

$T = 300$ K

$\tau_s = (140\pm4)$ ps

Kalevich, Ivchenko, Shiryaev, Egorov, Lombez, Lagarde, Marie and Amand,
JETP Lett. 85, 174 (2007)
Spin-dependent photoconductivity in GaAsN

\[ \text{[N]} = 2.1\% , \text{ silicon doped } 2 \times 10^{18}\text{cm}^{-3} \]

\[ T = 300\text{K} \]

\[ B = 0 \]

Photocurrent (rel. u.)

\[ 1.3 \]
\[ 1.2 \]
\[ 1.1 \]
\[ 1.0 \]

Time (arb. u.)

\[ \text{lin} \]
\[ \text{lin} \]
\[ \text{lin} \]

\[ \text{circ} \]
\[ \text{circ} \]

\[ \text{B} (\text{Tesla}) \]

Photocurrent (r. u.)

\[ 130 \]
\[ 120 \]
\[ 110 \]

\[ P = 80\text{mW} \]

\[ P = 50\text{mW} \]

\[ (\text{pulsed}) \]

\[ (\text{CW}) \]


What is the nature of paramagnetic centers?

Optically detected electron magnetic resonance (ODEMR) of self-interstitial defect Ga$_{2}^{+}$

- strong under circular pumping / as grown
- absent under linear pumping / after RTA

\[ H = \mu_B \mathbf{B} \cdot \mathbf{S} + AS \cdot \mathbf{I} \]

\[ \Psi_e(r): \quad s - \text{shape} \]
\[ \sim 20\% \text{ localized on nucleus of center} \]

Linköping University, Sweden

Effect of rapid thermal annealing on spin dynamics in Ga(In)AsN/GaAs

Post-growth rapid thermal annealing reduces strongly the density of paramagnetic centers

$L.\ Lombez,\ P.-F.\ Braun,\ H.\ Carrère,\ B.\ Urbaszek,\ P.\ Renucci,\ T.\ Amand,\ X.\ Marie,\ J.C.\ Harmand\ and\ V.K.\ Kalevich,\ APL\ 87,\ 252115\ (2005)$
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Enhancement of spin-filtering by magnetic field
Faraday geometry, $B \parallel z$

1) Increase of $\rho$ and $J$

Kalevich, Afanasiev, Shiryaev, Egorov,
Enhancement of spin-filtering by magnetic field
Faraday geometry, $B || z$

1) Increase of $\rho$ and $J$

2) Shift $|B_{\text{eff}}| \sim 200$ G
   reverses sign with $\sigma^+ \rightarrow \sigma^-$

Suppression of spin relaxation
of bound electrons
by a longitudinal magnetic field

$$\frac{1}{\tau_{sc}(B)} = \frac{2 \omega J^2 \tau_{cor}}{1 + \omega^2 \tau_{cor}^2} = \frac{1}{\tau_{sc}(0)} \frac{1}{1 + (B/B_{1/2})^2}$$

Dyakonov and Perel, JETP 38, 177 (1974)

Suppression of the nuclear field fluctuations

$$y(B) = y_{\text{max}} + (y_{\text{min}} - y_{\text{max}}) / [1 + (B - B_{\text{eff}})^2/B_{1/2}^2]$$

$$y_{\text{min}} = y(B = B_{\text{eff}}), \quad y_{\text{max}} = y(B \rightarrow \infty)$$

$\rho$ = 75 mW

GaAs$_{0.98}$N$_{0.02}$
Spin-filter amplification vs pump power

\[
\frac{1}{\tau_{sc}(B)} = \frac{1}{\tau_{sc}^*} + \frac{1}{\tau_{sc}^{(1)}} \frac{1}{1 + (B/B_{1/2})^2}
\]

(a) 250 mW
150 mW
75 mW
25 mW
8 mW

(b) 250 mW
150 mW
75 mW
25 mW
8 mW

Pump power (mW)

Longitudinal magnetic field (Gauss)

PL intensity (arb. u.)

\[ \rho \]

\[ B_{max} = 6.5kG \]

\[ B = B_{max} = 6.5kG \]

\[ B = 0 \]

\[ \frac{\mathcal{J}(B_{max})}{\mathcal{J}(B=0)} \]

\[ B_{max} = 6.5kG \]

Pump power (mW)
Amplification of spin-filtering by magnetic field

\[ \frac{1}{\tau_{sc}(B)} = \frac{1}{\tau_{sc}^*} + \frac{1}{\tau_{sc}^{(1)}} \frac{1}{1 + (B/B_{1/2})^2} \]

**experiment**

**computation**

<table>
<thead>
<tr>
<th>Outline</th>
</tr>
</thead>
<tbody>
<tr>
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</tr>
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</tr>
<tr>
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</tr>
</tbody>
</table>
Nonlinear strongly coupled electron-nuclear spin-system

\[ \sigma \text{- excitation} \quad \text{free electrons} \quad \text{luminescence} \]

\[ \text{bound electrons} \quad \text{SDR} \]

\[ \sum_{n} \sum_{n} + - + + = \cdots \]

\[ \sum_{n} \sum_{n} n + n_{hf} I_s I_z + \frac{1}{2} (s + I_- + s - I_+) \]

\[ a_n = A v_0 |\Psi (\vec{r}_n)|^2, \quad A \approx 100 \mu \text{eV} \]
Nuclear spin fields acting on a localized electron

\[ \hat{H}_{hf} = \sum_n a_n (\vec{s} \cdot \vec{I}_n) = \sum_n a_n \left[ s_z I_z + \frac{1}{2} (s_+ I_- + s_- I_+) \right] \]

\[ a_n = A v_0 \left| \Psi (\vec{r}_n) \right|^2 , \quad A \approx 100 \ \mu\text{eV} \]

a) Polarized nuclear spins

\[ \vec{B}_N = \frac{A}{\mu_B g_e} \langle \vec{I} \rangle \]

\[ B_{N_{\text{max}}}^{\text{GaAs}} \approx 53 \ \text{kG} \]

\[ \vec{B}_{\text{total}} = (\vec{B} + \vec{B}_N) \]

b) Disordered nuclear spins

\[ \left\langle B_{f}^{2} \right\rangle = \frac{v_0^2 \sum_n \left| \Psi (\vec{r}_N) \right|^4 A_n^2 I_n^2 (I_n + 1)^2}{(\mu_B g_e)^2} \approx \frac{B_{N_{\text{max}}}^{2}}{N} \]
Electron spin relaxation on nuclei

Merkulov, Efros, Rosen, PRB 65, 205309 (2002)

Nuclear spins

\[ \langle S \rangle = \frac{\langle S_0 \rangle}{3} \]

\[ T_2 \sim 10^{-4} \text{s} \]

GaAs

\[ N \sim 10^5 \]

\[ B_f \sim 50 \text{ G} \]

\[ B_{N, \text{max}}^{\text{GaAs}} \approx 53 \text{kG} \]

\[ B_f \approx \frac{B_{N, \text{max}}}{N} \cdot \sqrt{N} = \frac{B_{N, \text{max}}}{\sqrt{N}} \]

\[ A \langle I \rangle S = - \mu_e B_N, \quad \mu_e = g_e \mu_B S \]

\[ B_N = A \langle I \rangle / g_e \mu_B, \quad A \sim 100 \mu\text{eV} \]

\[ g_e = 2, \quad B_{N, \text{max}}^{\text{GaAs}} \approx 13 \text{kG} \]

\[ B_f \sim B_{1/2}^{\parallel} \sim 1000 \text{ G} \quad \rightarrow \quad N < 100 \]

\[ a_B < 7 \text{ Å} \]

Longitudinal magnetic field (Gauss)

PL intensity (arb. u.)
Extremely fast polarization of nuclei by electrons

The time required to polarize nuclei by electrons \( \equiv \) Nuclei spin-relaxation time by electrons \( T_{1e} \leq 15 \mu s \)
Enhancement of spin-filtering by magnetic field
Faraday geometry, \( B \parallel z \)

\[ y(B) = y_{\max}^+ (y_{\min} - y_{\max}) / \left[1 + (B - B_{\text{eff}})^2 / B_{\text{eff}}^2 \right], \]

\[ y_{\min} = y(B = B_{\text{eff}}), \quad y_{\max} = y(B \to \infty) \]
Hyperfine vs Exchange

Hyperfine: \[ H_{hf} = A s_c I \rightarrow B_N \propto \langle I \rangle \propto P_c = const \]

Exchange: \[ H_{ex} = Q s_c S \rightarrow B_{ex} \propto S = Pn/2 \propto W \]

The field \( B_{eff} \) is created by nuclear polarization

\[ B_{eff} \equiv B_N \]
Degree of nuclear polarization?
One electron – one nucleus

\[ \hat{H} = A S \cdot I + g_c \mu_B S \cdot B \]

1) B = 0: \[ \tilde{F} = \tilde{S} + \tilde{I}, \quad S = I = 1/2, \quad F = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad E = \begin{pmatrix} A/4 \\ -3A/4 \end{pmatrix} \]

 triplet: \[ F_z = \begin{pmatrix} +1 \\ -1 \end{pmatrix} = \begin{pmatrix} \uparrow \uparrow \\ \downarrow \downarrow \end{pmatrix} \]

 singlet: \[ F_z = (0) = \begin{pmatrix} \uparrow \downarrow - \downarrow \uparrow \end{pmatrix} \]

2) \( g_c \mu_B m_s B \gg A \)

\[ E = g_c \mu_B m_s B + A m_s m_1, \quad m_s = \pm 1/2, \quad m_1 = \pm 1/2 \]

Mixing time \( \hbar / A \sim 1\text{ps} \ll \tau_c \quad (\tau_c > 50\text{ps}) \)

\[ \langle S \rangle / \langle S_0 \rangle = 1/2, \ 3/8, \ 1/3 \quad \text{for} \quad I = 1/2, \ 3/2, \ I >> 1 \]

\[ \frac{\langle S \rangle}{\langle S_0 \rangle} = 1 - \frac{0.5}{1 + (g_c \mu_B B / A)^2}, \quad g_c \mu_B B \sim \delta, \quad \delta_{\exp} \sim 10\mu\text{eV} \]

Nuclear polarization

\[ \langle I \rangle = \langle S_0 \rangle n_{\text{e}} / (n_{\text{e}} + n_1), \quad n_{\text{e}} = f / 2 \]

\[ n_{\text{e}} = (f / 2) / [1 + (g_c \mu_0 B / A)^2] \]
SUMMARY

✓ Spin-dependent recombination via deep paramagnetic centers is an effective mechanism to create very high (up to 100%) spin polarization of free electrons in conduction band, paramagnetic centers and nuclei in a nonmagnetic semiconductor at room temperature and zero or weak magnetic field.
Collaboration with

Ioffe Institute, Saint-Peterburg

Tsukuba University, Japan
M. Ikezawa, B. Pal, and Y. Masumoto

INSA, Toulouse, France, *) LPN, Marcoussis, France
H. Carrère, L. Lombez, D. Lagarde, F. Zhao, A. Balocchi, B. Urbaszek, T. Amand, X. Marie, and J.C. Harmand*)

Linköping University, Sweden
X.J. Wang, I. Buyanova, and W. Chen