Diluted Magnetic Semiconductors - An Introduction -

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- 1. What you need to know about nonmagnetic semiconductors
 - Band structure
- 2. How you make a semiconductor magnetic
 - Doping and *sp-d* exchange
- 3. The consequences of exchange interaction
 - Spin-split bands and ferromagnetism
- 4. Making "use" of ferromagnetism in semiconductors
 - Electrical control of ferromagnetism
 - Current induced domain wall motion
 - Tunneling with magnetism

- 1. What you need to know about nonmagnetic semiconductors
 - Band structure



Electronic band structure From Wikipedia, the free encyclopedia



taken from Walter A. Harrison "Electronic Structure and the Properties of Solids -The Physics of Chemical Bond -, 1980

> Bottom of the conduction band: Ga 4s Top of the valence band: As 4p



J. R. Chelikowsky and M. L. Cohen, Phys. Rev. B14, 556 (1976)





2. How you make a semiconductor magnetic

• Doping and *sp-d* exchange

Making semiconductor magnetic



Magnetic ion: Mn



Hund rule: Distributing *n* electrons over 2(2l+1) degenerate atomic orbitals, the lowest energy state is the state that maximize the total spin angular momentum S

Mn: *l*=2, *n*=5 -> S=5/2

Magnetic ion: Mn

- intra site correlation energy $U = E_{n+1} E_n$ for n = 5, $U \approx 15$ eV
- intra-site exchange interaction: ferromagnetic Hund's rule: S the highest possible for n = 5, $E_{S=3/2} - E_{S=5/2} \approx 2 \text{ eV}$
- transition metal atoms, 3dn4s1, e.g., Mn: ferromagnetic

 $E_{S=2} - E_{S=3} \approx 1.2 \text{ eV} \Rightarrow J_{s-d} \approx 0.4 \text{ eV}$



II-VI Diluted Magnetic Semiconducotors



Lattice Constant (nm)

Density of states with transition metal impurity







Virtual crystal approximation

molecular-field approximation

mean-field approximation

$$H_{n} = (1-x)U_{0}(\mathbf{r} - \mathbf{R}_{n}) + xU(\mathbf{r} - \mathbf{R}_{n}) - xJ(\mathbf{r} - \mathbf{R}_{n})\mathbf{s} \cdot \mathbf{S}_{n}$$

$$\frac{x}{V}\sum_{n} \mathbf{S}_{n} \rightarrow xN_{0} \langle \mathbf{S}(\mathbf{r}) \rangle$$

$$\rightarrow -\frac{\mathbf{M}(\mathbf{r})}{g\mu_{B}}; \quad \mathbf{M}(\mathbf{r}) = -xN_{0}g\mu_{B} \langle \mathbf{S}(\mathbf{r}) \rangle$$

$$\rightarrow -\frac{\mathbf{M}}{g\mu_{B}}; \quad \mathbf{M} = \langle \mathbf{M}(\mathbf{r}) \rangle$$

III-V Diluted Magnetic Semiconductors



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(Ga,Mn)As, (In,Mn)As, (Ga,Mn)Sb
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Mn in GaAs

Mn on Ga site: 113 meV acceptor

- d⁵+hole (weak coupling) ←
- d⁴
- d⁵+hole (strong coupling)

from spectroscopy

$$H = T - \frac{e^2}{\varepsilon r} - V_0 \exp\left(-\left(\frac{r}{r_0}\right)^2\right) - xN_0\beta \mathbf{s} \cdot \mathbf{S} \qquad N_0\beta = -0.9 \text{ eV}$$

kinetic, Coulomb, central cell correction, *p-d* exchange interaction

theory: Bhattacharjee & C. Benoit a la Guillaume, Solid State Commun. **113**, 17 (2000) exp: M. Linnarsson et al. Phys. Rev. **B 55**, 6938 (1997)

3. The consequences of exchange interaction

• Spin-split bands and ferromagnetism



1.92 K 111 ZnMnSe x=0.033

60

80

101

Y. Shapira et al.

no spontaneous magnetization

Density of states with transition metal impurity



Faraday rotation

 A linearly polarized light arriving in the medium can be decomposed in two circularly polarized waves

- After travelling in the medium over a distance *d* the two components develop a phase shift $\delta = \frac{\omega}{c} (n_+ n_-)d$
- equivalent to a rotation of the polarization plane by the Faraday rotation angle

$$\vartheta = \frac{1}{2}\delta = \frac{\omega}{2c}(n_+ - n_-)d$$

$$\mathbf{B}$$

J. Gaj, Spintech III

Faraday effect



J. A. Gaj, R. R. Galazka, and M. Nawrocki, *Solid State Commun.*, vol. 25, p. 193 (1978).

Splitting of the bands



Measurements of the band splitting



magnetoreflectance: Aggarwal et al. Phys. Rev. B34, 5894 (1986)



circles: photoemission squares: optical spectroscopy

3. The consequences of exchange interaction

• Spin-split bands and ferromagnetism

GaAs (or InAs) + Mn

Material Science

Molecular beam epitaxy of (Ga,Mn)As









Molecular beam epitaxy of (Ga,Mn)As



Extended X-ray-Absorption Fine Structure (EXAFS)



- X-ray is absorbed by target atom \rightarrow creation of photoelectron
- Photoelectron is scattered by adjacent atoms
- Interference of photoelectron and reflected photoelectron
- · Oscillating structure in absorption spectra includes the information of

local structure around target atom

Local Structures - Extended X-ray-Absorption Fine Structure (EXAFS)



Most of Mn substitute III-cation site (Mn²⁺ \rightarrow acceptor)

Atom Probe Microscopy of (Ga,Mn)As



M. Kodzuka, T. Ohkubo, K. Hono (NIMS) F. Matsukura, and H. Ohno (Tohoku), Ultramicroscopy 2009

Magnetism, transport, and more materials science

Magnetization of (Ga,Mn)As

(Ga,Mn)As & (In,Mn)As: Mn acts as a source of spin and hole

Ferromagnetism in (In,Mn)As: H. Ohno *et al.*, *Phys Rev. Lett.* 1992 and in (Ga,Mn)As: H. Ohno *et al.*, *Appl. Phys. Lett.* 1996



Magnetization and transport of (Ga,Mn)As



Hall Effects



Low-temperature & high-field measurements



Low-temperature annealing



T. Hayashi et al., Appl. Phys. Lett. 78, 1691 (2001).

Low-temperature annealing 220-250°C

- · Decreases lattice constant
- · Increases electrical conductivity
 - Increases hole concentration
- Increases ferromagnetic transition temperature

Correlation between $T_{\rm C}$ and p

See also K. W. Edmonds et al., Appl. Phys. Lett. 81, 4991 (2002).

Mn_i

Ferromagnetism
Zener Model Description of Ferromagnetism in Zinc-Blende Magnetic Semiconductors

T. Dietl,^{1,2}* H. Ohno,¹* F. Matsukura,¹ J. Cibert,³ D. Ferrand³

Ferromagnetism in manganese compound semiconductors not only opens prospects for tailoring magnetic and spin-related phenomena in semiconductors with a precision specific to III-V compounds but also addresses a question about the origin of the magnetic interactions that lead to a Curie temperature (T_c) as high as 110 K for a manganese concentration of just 5%. Zener's model of ferromagnetism, originally proposed for transition metals in 1950, can explain T_c of Ga_{1-x}Mn_xAs and that of its II-VI counterpart Zn_{1-x}Mn_xTe and is used to predict materials with T_c exceeding room temperature, an important step toward semiconductor electronics that use both charge and spin.

SCIENCE VOL 287 11 FEBRUARY 2000

1019



Like other thermodynamic quantities, Fc[M] is expected to be weakly perturbed by static disorder

We suggest that the holes in the extended or weakly localized states mediate the long-range interactions between the localized spins on both sides of the MIT in the III-V and II-VI magnetic semiconductors.

Visualizing Critical Correlations Near the Metal-Insulator Transition in Ga_{1-x}Mn_xAs

Anthony Richardella,^{1,2*} Pedram Roushan,^{1*} Shawn Mack,³ Brian Zhou,¹ David A. Huse,¹ David D. Awschalom,³ Ali Yazdani¹†

www.sciencemag.org SCIENCE VOL 327 5 FEBRUARY 2010

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Fig. 5. The spatial variations of the LDOS at the Fermi level, with their histogram and multifractal spectrum. The LDOS mapping of a 700 Å by 700 Å area of (**A**) $Ga_{0.985}Mn_{0.015}As$, (**B**) $Ga_{0.97}Mn_{0.03}As$, and (**C**) $Ga_{0.985}Mn_{0.05}As$. (**D**) The normalized histogram of the maps presented in (A) to (C). The local values of the *dlldV* are normalized by the average value of each map. The inset shows the multifractal spectrum, $f(\alpha)$, near the value α_0 where the maximum value occurs. For comparison, the results of a similar analysis over a LDOS map at -100 mV (valence band states) for the 1.5% doped sample are also shown.



p-d Zener model of ferromagnetism

Ferromagnetic (Ga,Mn)As

Mn acts as a source of spin and hole

H. Ohno *et al. Appl. Phys. Lett.* 1996 H. Ohno *Science* 1998



T. Dietl, *et al. Science* **287**, 1019 (2000) and *PRB* **63**, 195205 (2001) also Koenig et al. *Phys. Rev. Lett.* 2000

T_C by the *p-d* Zener model



T_C by the *p-d* Zener model



Valence band structure (Γ_7 , Γ_8)

 $H_{kp}\Psi = E\Psi$ Basis functions

$$u_{1} = \frac{1}{\sqrt{2}}(X + iY) \uparrow, \quad u_{2} = i\frac{1}{\sqrt{6}}[(X + iY) \downarrow -2Z \uparrow], \quad u_{3} = \frac{1}{\sqrt{6}}[(X - iY) \uparrow +2Z \downarrow],$$
$$u_{4} = i\frac{1}{\sqrt{2}}(X - iY) \downarrow, \quad u_{5} = \frac{1}{\sqrt{3}}[(X + iY) \downarrow +Z \uparrow], \quad u_{6} = i\frac{1}{\sqrt{3}}[-(X - iY) \uparrow +Z \downarrow]$$

k·*p* matrix

$$H_{kp} = -\frac{\hbar^2}{2m_0} \begin{pmatrix} P+Q & L & M & 0 & iL/\sqrt{2} & -i\sqrt{2}M \\ L^{\dagger} & P-Q & 0 & M & -i\sqrt{2}Q & i\sqrt{3/2}L \\ M^{\dagger} & 0 & P-Q & -L & -i\sqrt{3/2}L^{\dagger} & -i\sqrt{2}Q \\ 0 & M^{\dagger} & -L^{\dagger} & P+Q & -i\sqrt{2}M^{\dagger} & -iL^{\dagger}/\sqrt{2} \\ -iL^{\dagger}/\sqrt{2} & i\sqrt{2}Q & i\sqrt{3/2}L & i\sqrt{2}M & P+\Delta & 0 \\ i\sqrt{2}M^{\dagger} & -i\sqrt{3/2}L^{\dagger} & i\sqrt{2}Q & iL/\sqrt{2} & 0 & P+\Delta \end{pmatrix}$$

$$P = \gamma_1 k^2, \ Q = \gamma_2 \left(k_x^2 + k_y^2 - 2k_z^2\right), \ L = -2\sqrt{3}\gamma_3 \left(k_x - ik_y\right)k_z, \ M = -\sqrt{3}\left[\gamma_2 \left(k_x^2 - k_y^2\right) - 2i\gamma_3 k_x k_y\right] \\ \gamma_1 = 6.85, \ \gamma_2 = 2.1, \ \gamma_3 = 2.58, \ \Delta = 0.34 \text{ eV for GaAs}$$

T. Dietl, H. Ohno and F. Matsukura, Phys. Rev. B63, 195205 (2001)

$$(H_{kp} + H_{pd})\Psi = E\Psi$$

p-d exchange interaction matrix

$$H_{pd} = B \cdot \begin{bmatrix} 3b_{x}w_{z} & i\sqrt{3}b_{x}w_{-} & 0 & 0 & \sqrt{6}b_{x}w_{-} & 0 \\ -i\sqrt{3}b_{x}w_{+} & (2b_{z}-b_{x})w_{z} & 2ib_{z}w_{-} & 0 & i\sqrt{2}(b_{x}+b_{z})w_{z} & \sqrt{2}b_{z}w_{-} \\ 0 & -2ib_{z}w_{+} & -(2b_{z}-b_{x})w_{z} & i\sqrt{3}b_{x}w_{-} & \sqrt{2}b_{z}w_{+} & -i\sqrt{2}(b_{x}+b_{z})w_{z} \\ 0 & 0 & -i\sqrt{3}b_{x}w_{+} & -3b_{x}w_{z} & 0 & -\sqrt{6}b_{x}w_{+} \\ \sqrt{6}b_{x}w_{+} - i\sqrt{2}(b_{x}+b_{z})w_{z} & \sqrt{2}b_{z}w_{-} & 0 & -(2b_{x}-b_{z})w_{z} & ib_{z}w_{-} \\ 0 & 0 & i\sqrt{2}(b_{x}+b_{z})w_{z} & -\sqrt{6}b_{x}w_{-} & -ib_{z}w_{+} & (2b_{x}-b_{z})w_{z} \end{bmatrix}$$

$$B = -\frac{1}{6} \times N_{0}\beta\langle S_{z}\rangle, \qquad \beta_{x} = b_{x}\beta, \qquad \beta_{z} = b_{z}\beta$$

$$w_{z} = \langle S_{z}\rangle/|\langle S\rangle|, \qquad w_{\pm} = (\langle S_{x}\rangle \pm i\langle S_{y}\rangle)/|\langle S\rangle|$$

$$|\langle S\rangle| = |\langle S_{x}\rangle^{2} + \langle S_{y}\rangle^{2} + \langle S_{z}\rangle^{2}|^{1/2}$$

T. Dietl, H. Ohno and F. Matsukura, Phys. Rev. B63, 195205 (2001)

Exchange energy: $N_0\beta$

Core level photoemission and modeling: $N_0\beta = -1.2 \pm 0.2 \text{ eV}$

Okabayashi et al., Phys. Rev. B58, 1998.



FIG. 1. Photoemission spectrum of the Mn 2p core level (dots) and its cluster-model analysis (solid curves) assuming the negatively ionized Mn²⁺ (a) and neutral Mn³⁺ (b) ground states. The vertical bars are unbroadened spectra. The calculated background is shown by dashed curves.

Comparison of exp. and calculated $T_{\rm C}$



Strain induced-anisotropy: Theory vs. Experiment



Hole bands are responsible for the anisotropy

axis

Acceptor doping and susceptibility of Zn_{1-x}Mn_xTe:P



Sawicki et al. (Warsaw) pss'02 Kępa et al. (Warsaw, Oregon) PRL'03

Comparison of Experimental and Calculated $T_{\rm C}$



exp.:(Ga,Mn)As:T. Omiya *et al.*, Physica E **7**, 976 (2000). (In,Mn)As: D. Chiba *et al.*, J. Supercond. and Novel Mag.16, 179 (2003). (Zn,Mn)Te: D. Ferrand *et al.*, Phys. Rev. B **63**, 085201 (2001).

p-d exchange energy



4. Making "use" of ferromagnetism in semiconductors

- Electrical control of ferromagnetism
- Current induced domain wall motion
- Tunneling with magnetism

commentary

A window on the future of spintronics

Hideo Ohno

4. Making "use" of ferromagnetism in semiconductors

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Electric-field control of ferromagnetism



(In,Mn)As

H. Ohno et al., Nature 408, 944 (2000).

Electric-field control: the case of (Ga,Mn)As



See also Y. Nishitani et al., Phys. Rev. B 81, 045208 (2010) and M. Sawicki, et al., Nature Physics, 6, 22 (2010)

Field-effect structures of (Ga,Mn)As



backgate structure



ferroelectric-gate structure

I. Stokichnov *et al.*, Nature Mater. **7**, 464 (2008).



electric double layer



M. Endo *et al.*, Appl. Phys. Lett. **96**, 022515 (2010).

K. Olejník *et al.*,
Phys. Rev. B **78**, 054403 (2008);
M. S. H. Owen *et al.*, New J. Phys. **11**, 023008 (2009).





M. Overby *et al.*, Appl. Phys. Lett **92**, 192501 (2008); A. W. Rushforth *et al.*, Phys. Rev. B **78**, 085314 (2008); S. T. B. Goennenwein et al., phys. stat. sol. (RRL) **2**, 96 (2008); C. Bihler *et al.*, Phys. Rev. B **78**, 045203 (2008).

Electric-field control of magnetization direction



Electric-field effects on metals



FePt, FePd: M. Weisheit et al., Science (2007).

а





Au/ ultrathin Fe: T. Maruyama *et al.*, Nature Nanotechnology (2009).

All at room temperature



CoFeB: M. Endo, S. Kanai, S. Ikeda, F. Matsukura, and H, Ohno, *Appl. Phys. Lett.* 2010

O-23 Kanai et al.

4. Making "use" of ferromagnetism in semiconductors

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Well defined domains: (Ga,Mn)As

1 mm





5 μm

Welp et al., PRL 2003

Shono et al., APL 2000

compressive strain in-plane easy axis 90° domains tensile strain perpendicular easy axis stripe domains

Current induced domain wall motion





Patterning of coercive force H_c by etching of (Ga,Mn)As

Preparation of domain wall

Current induced domain wall motion



M. Yamanouchi et al., Phys. Rev. Lett. 96, 096601 (2006)

Mechanism

Adiabatic spin transfer (conservation of angular momentum)



Domain wall motion: Theory

• Square root dependence (Spin transfer)

G. Tatara and H. Kohno, Phys. Rev. Lett. 92, 086601 (2004).

$$v = A \left(j^2 - j_C^2 \right)^{1/2}$$



Current induced domain wall motion



Transfer factor and threshold current density



M. Yamanouchi et al., Phys. Rev. Lett. 96, 096601 (2006)

$$j_C = \frac{2 e K \delta_w}{\pi \hbar P}$$

	(Ga,Mn)As (<i>T</i> / <i>T</i> _c ~ 0.9)	Metal (in plane)	Metal (perpendicular)
<i>K</i> (hard axis anisotropy, J/m ³)	60	4x10 ⁵	10 ⁵
δ_{w} (domain wall width, nm)	20	100	10
<i>P</i> (spin polarization, %)	20	60	60
j _C (A/m²)	6x10 ⁹	7x10 ¹³	2 x10 ¹²

Observation of the intrinsic pinning of a magnetic domain wall in a ferromagnetic nanowire

T. Koyama¹, D. Chiba^{1,2}, K. Ueda¹, K. Kondou¹, H. Tanigawa³, S. Fukami³, T. Suzuki³, N. Ohshima³, N. Ishiwata³, Y. Nakatani⁴, K. Kobayashi¹ and T. Ono^{1*}

The spin transfer torque is essential for electrical magnetization switching^{1,2}. When a magnetic domain wall is driven by an electric current through an adiabatic spin torque, the theory predicts a threshold current even for a perfect wire without any extrinsic pinning³. The experimental confirmation of this 'intrinsic pinning', however, has long been missing. Here, we give evidence that this intrinsic pinning determines the threshold, and thus that the adiabatic spin torque dominates the domain wall motion in a perpendicularly magnetized Co/Ni nanowire. The intrinsic nature manifests itself both in the field-independent threshold current and in the presence of its minimum on tuning the wire width. The demonstrated domain wall motion purely due to the adiabatic spin torque will serve to achieve robust operation and low energy consumption in spintronic devices⁵⁻⁸.



4. Making "use" of ferromagnetism in semiconductors

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D. Chiba et al. Physica E 2004

Resonant Tunneling Diode with a Ferromagnetic Emitter



Resonant Tunnel Diode with a Ferromagnetic Emitter

Sample structures

(Ga _{0.965} Mn _{0.035})As	200 nm
GaAs	15 nm
AlAs	5 nm
GaAs	$d_{\rm W}$ nm
AlAs	5 nm
GaAs	5 nm
GaAs:Be (5x10 ¹⁷ cm ⁻³)	150 nm
GaAs:Be (5x10 ¹⁸ cm ⁻³)	150 nm
p ⁺ GaAs sub.	





H. Ohno et al. Appl. Phys. Lett., 1998

Ultra-high doping of Mn in III-V compounds



Metallic - merged impurity and valence bands



T. Jungwirth et al., Phys. Rev. B76, 125206 (2007)


Reconciling results of tunnelling experiments on (Ga,Mn)As

T. Dietl^{1,2} and D. Sztenkiel¹ arXiv 1102.3267v2 (Spintech VI poster FP-42)

(Formation of 2D holes in GaAs:Be explains the resonant tunnel-like feature and the thickness dependence)



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