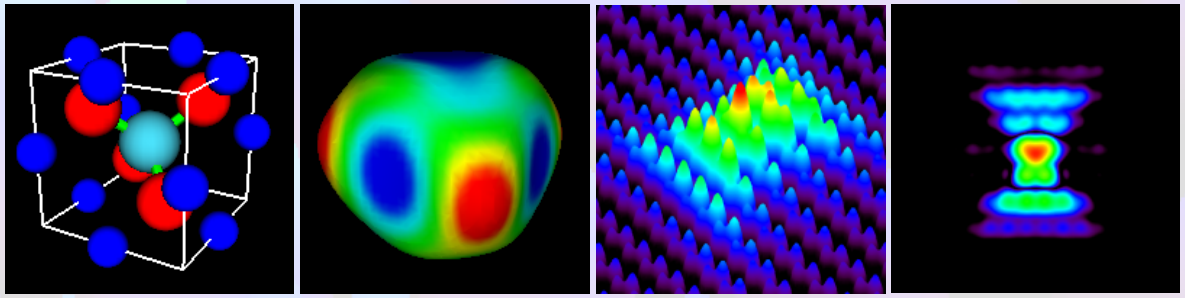
# Optical properties of diluted magnetic semiconductors and all-electrical control of individual dopant spins

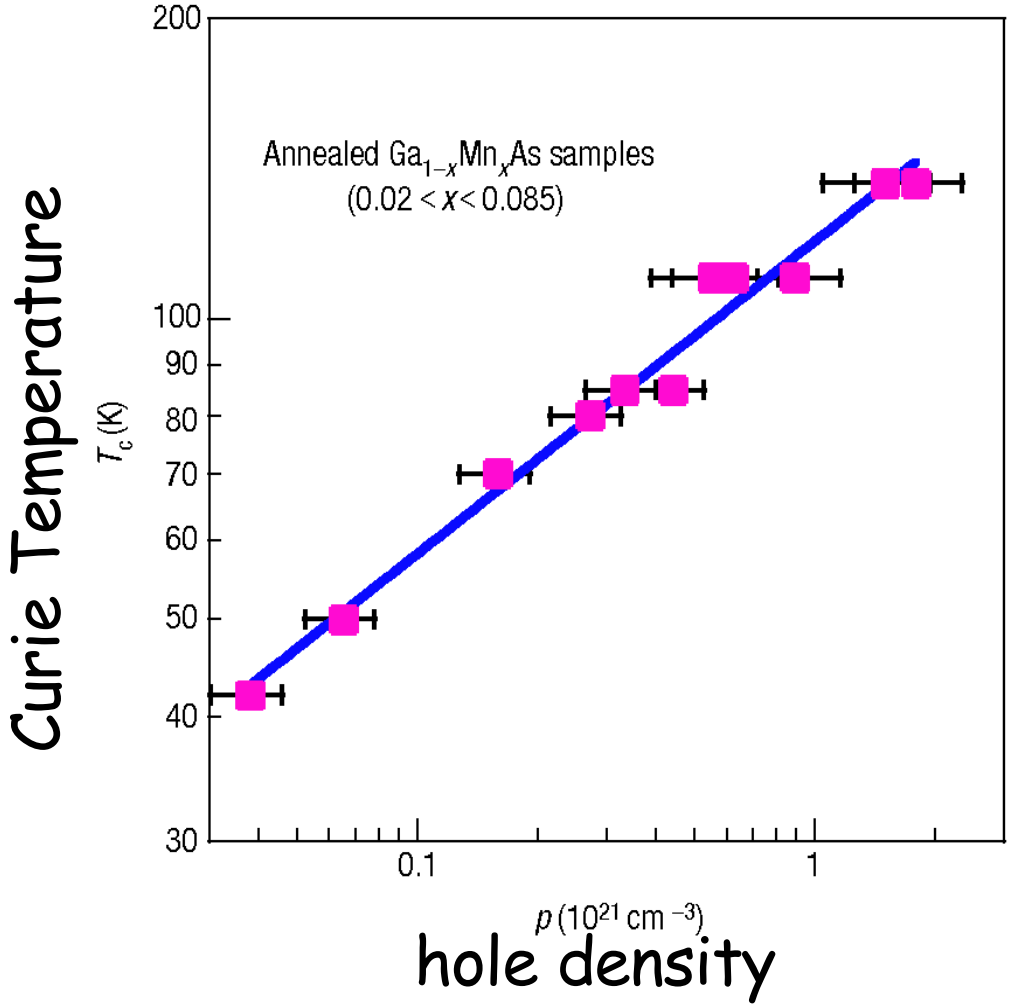
**Jian-Ming Tang**

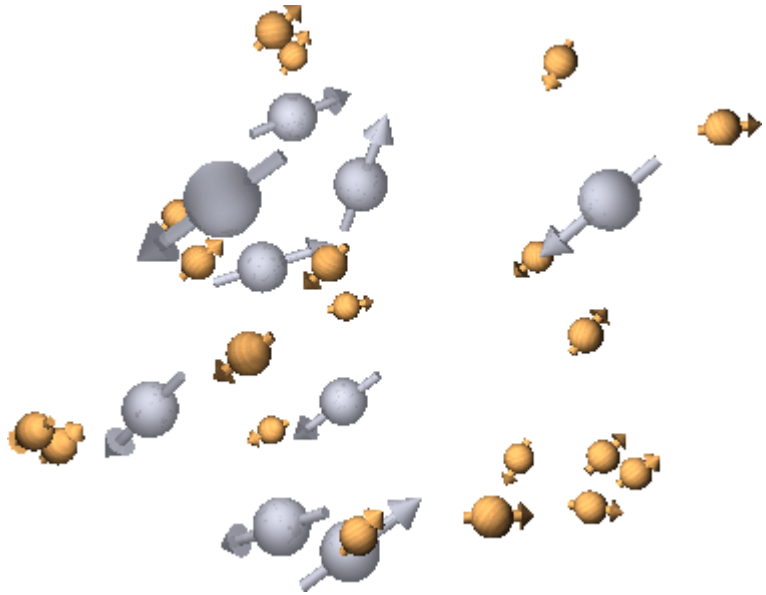
Department of Physics and Astronomy
Optical Science and Technology Center
University of Iowa

Supported by ARO



# Carrier-mediated ferromagnetism



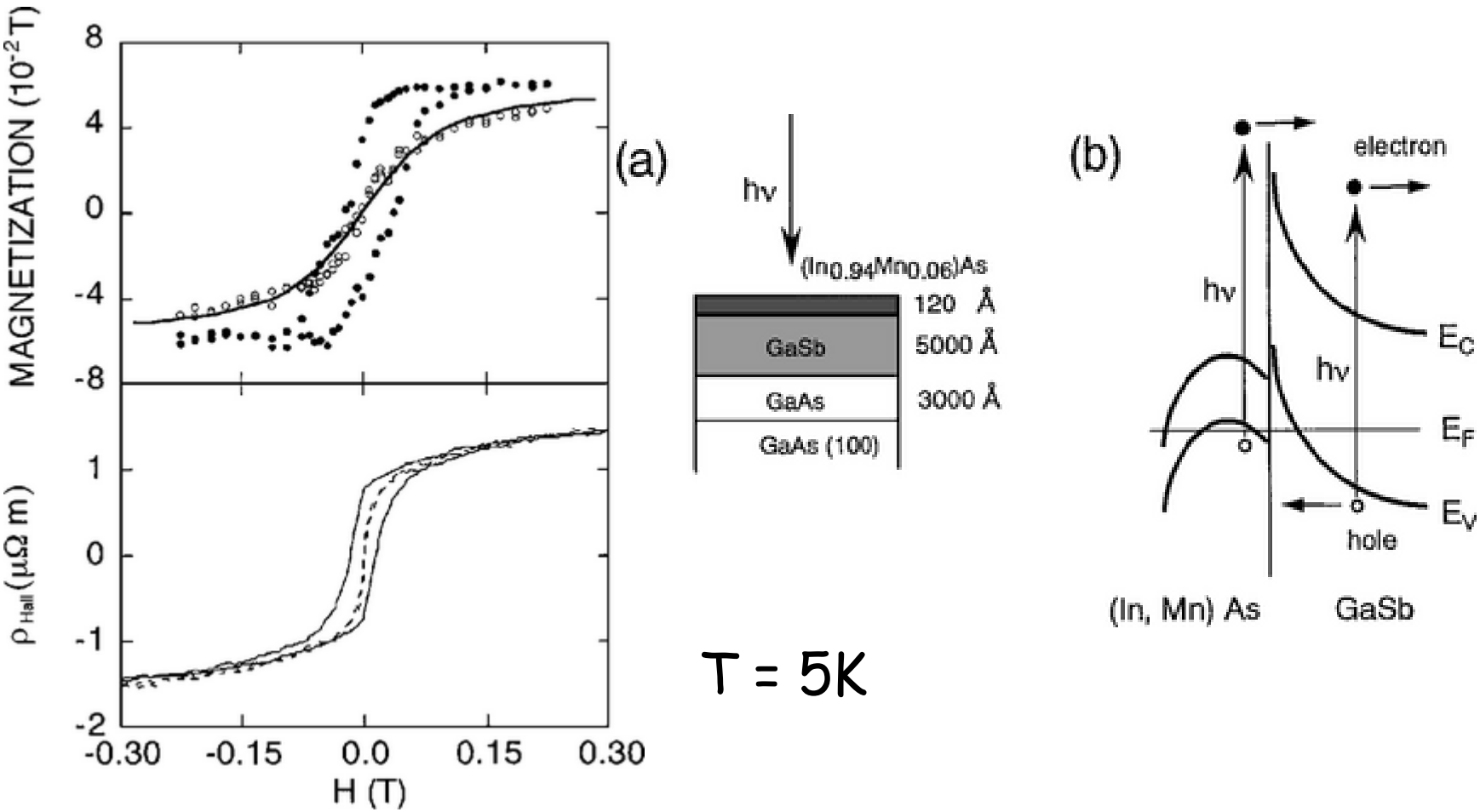
Mn: $3d^5$ $4s^2$

Ga: $4s^2$ $4p^1$

As: $4s^2$ $4p^3$

K.C. Ku et al., APL 82, 2302 (2003)
figure reprinted from A.H. MacDonald et al.
Nature Materials 4, 195 (2005)

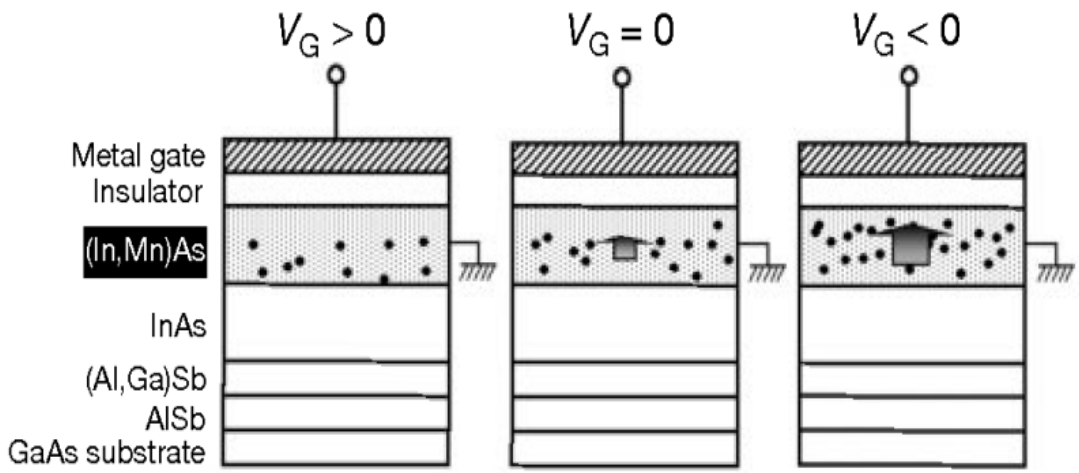
# Light-induced ferromagnetic order

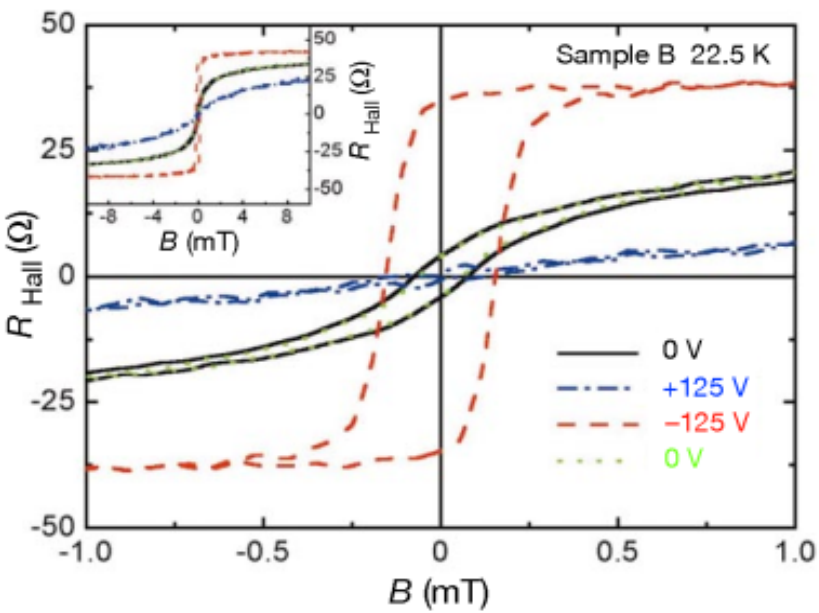


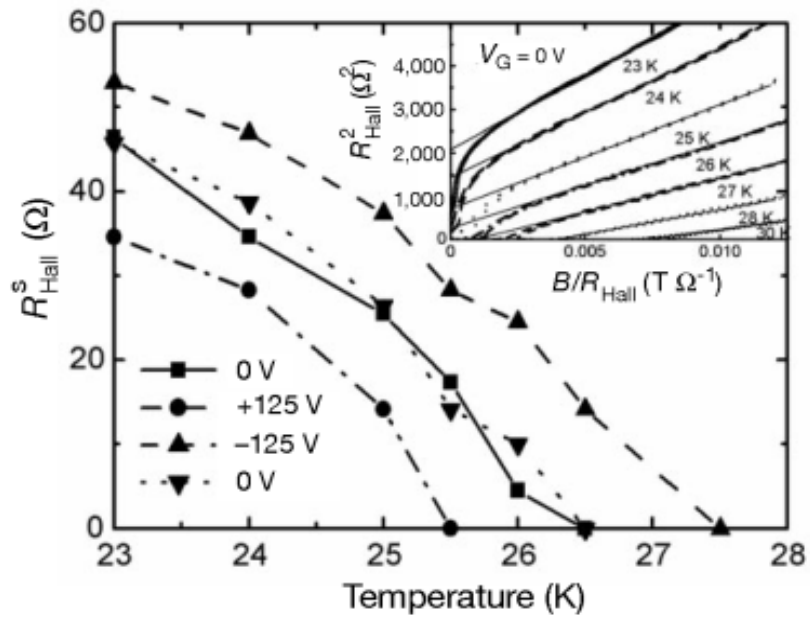

T = 5K

S. Koshihara et al., PRL 78, 4617 (1997)

# Electric-field control of ferromagnetism




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

# Outline

- Theory of single magnetic dopants
- magnetic circular dichroism in optical absorption
- electrical manipulation of single Mn spins

# Collaborators

- **Theory**
  - **Michael E. Flatté**

    University of Iowa, USA
  - **Jeremy Levy**

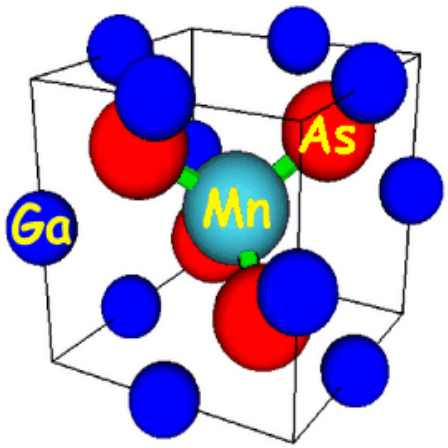
    University of Pittsburgh, USA
- **Experiment**
  - **Andrei M. Yakunin, Andrei Yu. Silov, Paul M. Koenraad, Joachim H. Wolter**

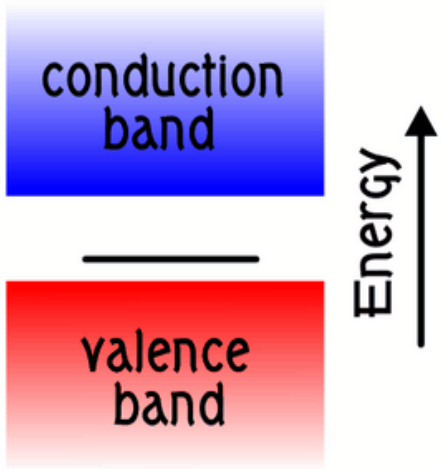
    Eindhoven University of Technology, The Netherlands
  - **Wim Van Roy, Joe De Boeck**

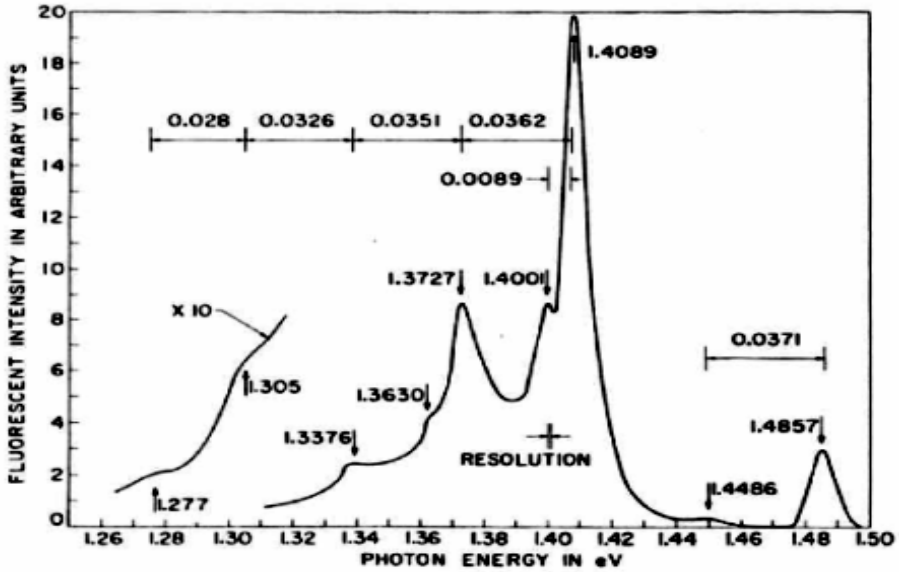
    Interuniversity Micro-Electronics Center, Belgium
  - **Dale S. Kitchen, Anthony Richardella, Ali Yazdani**

    Princeton University

# $Mn_{Ga}$ is an acceptor in GaAs



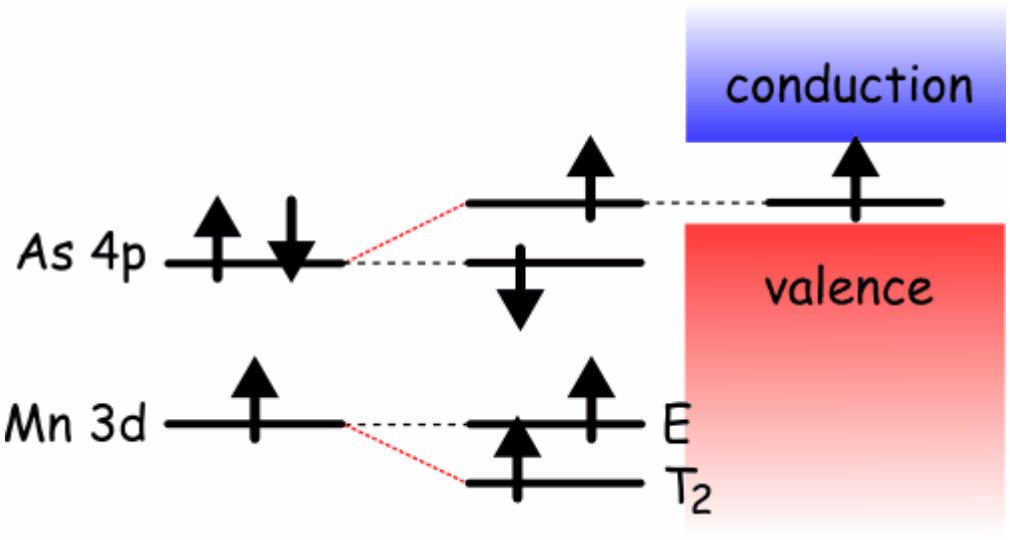




T.C. Lee and W.W. Anderson,
Solid State Commun. 2, 265 (1964)

| | | Cr | Mn | Fe | Co |
|---|---|---|---|---|---|
| **GaP** | (2.3) | 1 | 0.4 | 0.8 | 0.4 |
| **GaAs** | (1.5) | 0.6 | 0.1 | 0.5 | 0.1 |
| **InP** | (1.4) | 1 | 0.2 | 0.8 | 0.3 |
| **InAs** | (0.4) | | 0.03 | | |

(unit: eV)

# Antiferromagnetic p-d hybridization




P. Vogl and J.M. Baranowski,
Acta. Phys. Polo. A67, 133 (1985)

- "Hydrogenic" levels
  - 25.6 meV in GaAs
  - 16.6 meV in InAs

A. Baldereschi and N.O. Lipari
Phys. Rev. 8, 2697 (1973)

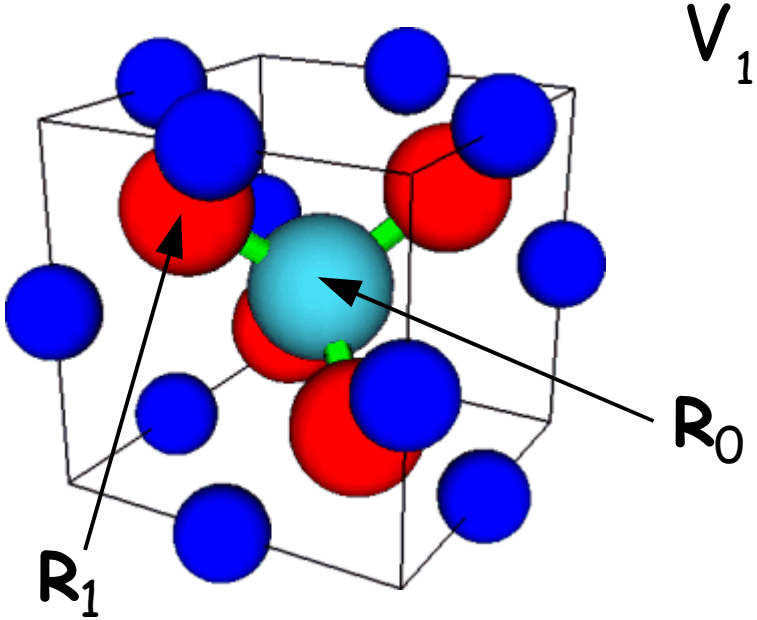
- Mn level:
  - 113 meV in GaAs
  - 28 meV in InAs

# Semiclassical exchange model

$$V(\mathbf{R},\mathbf{R}') = \left[ V_0 \sum_{\ell\sigma} c^{\dagger}_{\ell\sigma}(\mathbf{R_0}) c_{\ell\sigma}(\mathbf{R_0}) + V_1 \sum_{\ell j} c^{\dagger}_{\ell\uparrow}(\mathbf{R}_j) c_{\ell\uparrow}(\mathbf{R}_j) \right] \delta_{\mathbf{R},\mathbf{R}'}$$

$$V_0 = 1 \text{ eV}$$

$$V_1 = 3.59 \text{ eV}$$




$$|\psi(\mathbf{R}_0)|^2 \sim 0.1$$

$$|\psi(\mathbf{R}_1)|^2 \sim 0.05$$

- Coulomb potential (~1/εr)
  - 0.5 eV at $\mathbf{R}_1$
- effective mass model
  - acceptor level: 25.6 meV
- Mn level: 113 meV

# Koster-Slater approach

$$H = H_0 + V_{\text{impurity}}$$

$$\hat{G}_0(\mathbf{k};\omega) = \left[\omega - \hat{H}_0(\mathbf{k}) \pm i\varepsilon\right]^{-1}$$

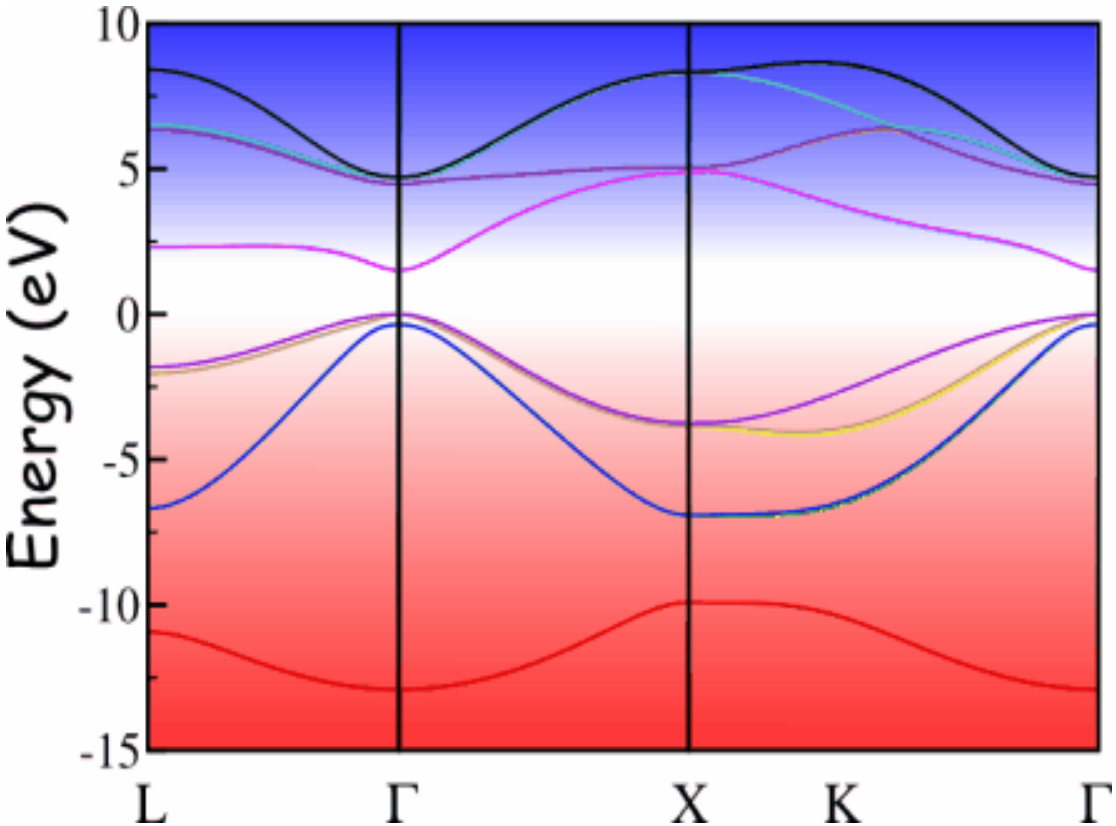
$$\hat{G}_0(\mathbf{R},\mathbf{R}';\omega) = \Omega_{\mathbf{k}}^{-1} \int d^3\mathbf{k}\, \hat{G}_0(\mathbf{k};\omega) e^{i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')}$$

$$\hat{G}(\mathbf{R},\mathbf{R}';\omega) = \hat{G}_0(\mathbf{R},\mathbf{R}';\omega) + \sum_{\mathbf{R}_a,\mathbf{R}_b} \hat{G}(\mathbf{R},\mathbf{R}_a;\omega)\hat{V}(\mathbf{R}_a,\mathbf{R}_b)\hat{G}_0(\mathbf{R}_b,\mathbf{R}';\omega)$$

$$\hat{\mathbf{G}}(\omega) = [\hat{\mathbf{1}} - \hat{\mathbf{G}}_0(\omega)\hat{\mathbf{V}}]^{-1}\hat{\mathbf{G}}_0$$

- G.F. Koster and J.C. Slater, PR 95, 1167 (1954)
- H.P. Hjalmarson, P. Vogl, D.J. Wolford and J.D. Dow, PRL 44, 810 (1980)
- J.-M. Tang and M.E. Flatté, PRL 92, 047201 (2004)

# $H_0(k)$ ($sp^3$ tight-binding model)



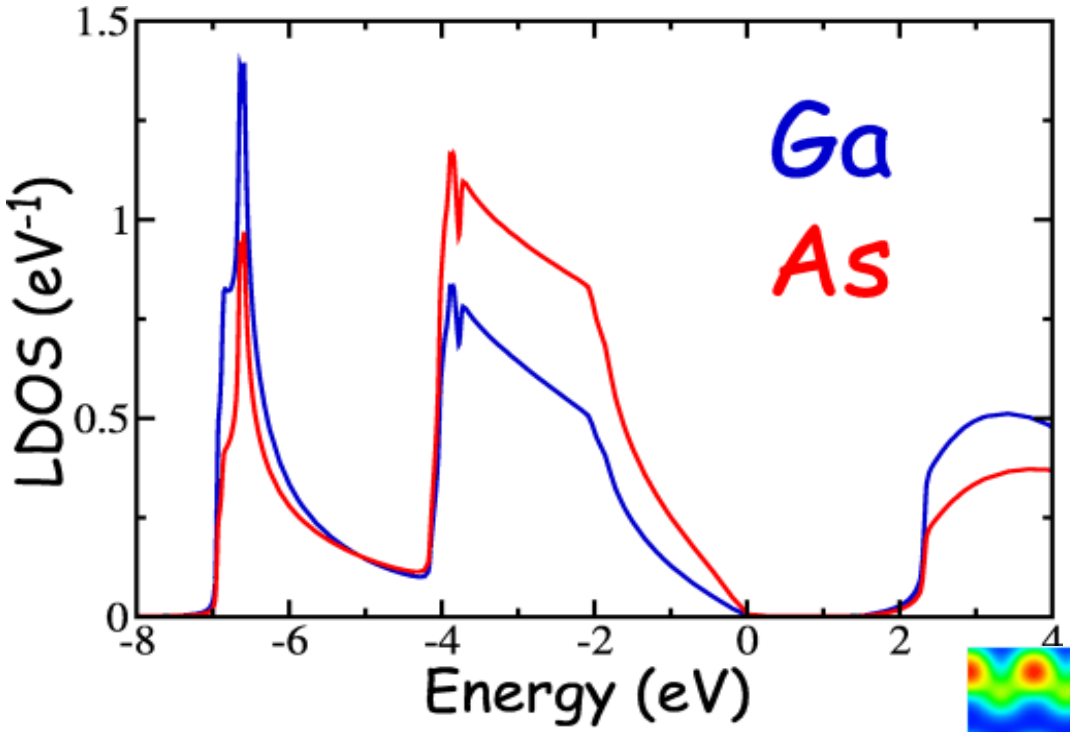

- full-zone band structure
  - spatial structure at atomic scales
  - crystal symmetry
- spin-orbit interaction

D. J. Chadi, PRB 16, 790 (1977)

- Atomic orbital basis: $\varphi_{\ell,s}(\mathbf{R})$
  - minimum set for zincblende lattice: 16 basis states

    2 (atomic sites) x 4 ($sp^3$ orbitals) x 2 (spins)
- nearest-neighbor interaction
- on-site spin-orbit interaction

# Local density of states (LDOS)



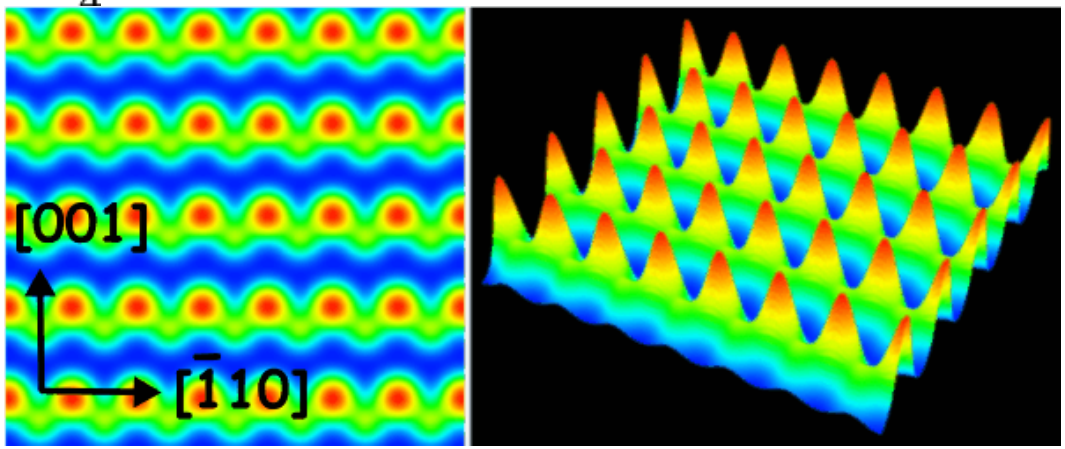

- density of states

$$D(E) = \sum_n \delta(E - E_n)$$

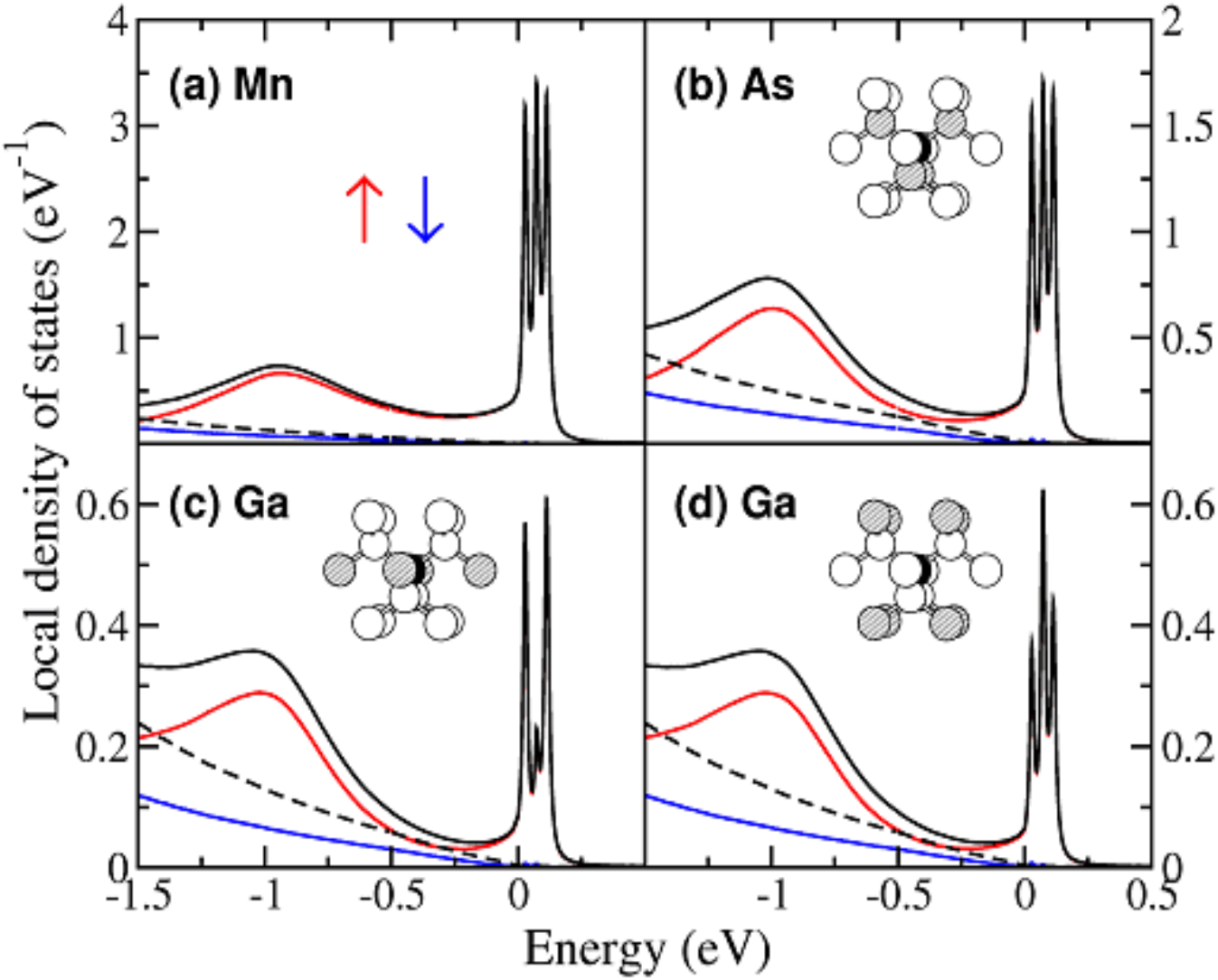
$$\mathrm{LDOS}(\mathbf{r}, E)$$

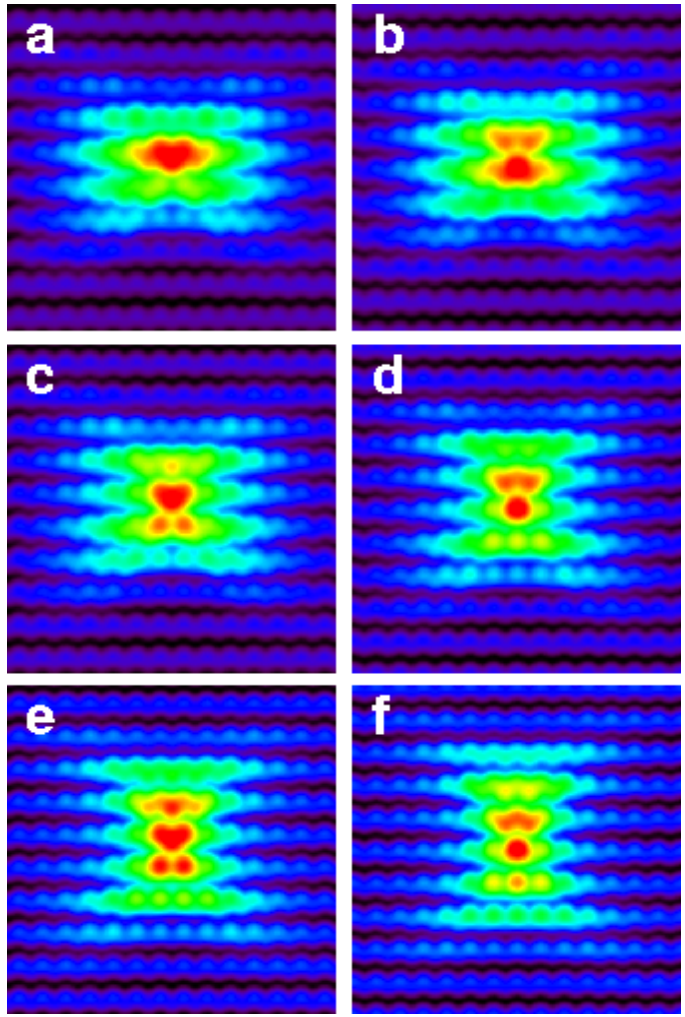
$$= \sum_n \left|\Psi_n(\mathbf{r})\right|^2 \delta(E - E_n)$$

$$\approx -\frac{1}{\pi} \Im \left[ \sum_{\mathbf{R},\alpha} \hat{G}_{\alpha,\alpha}(\mathbf{R}, \mathbf{R}; E) \left|\phi_\alpha(\mathbf{r} - \mathbf{R})\right|^2 \right]$$



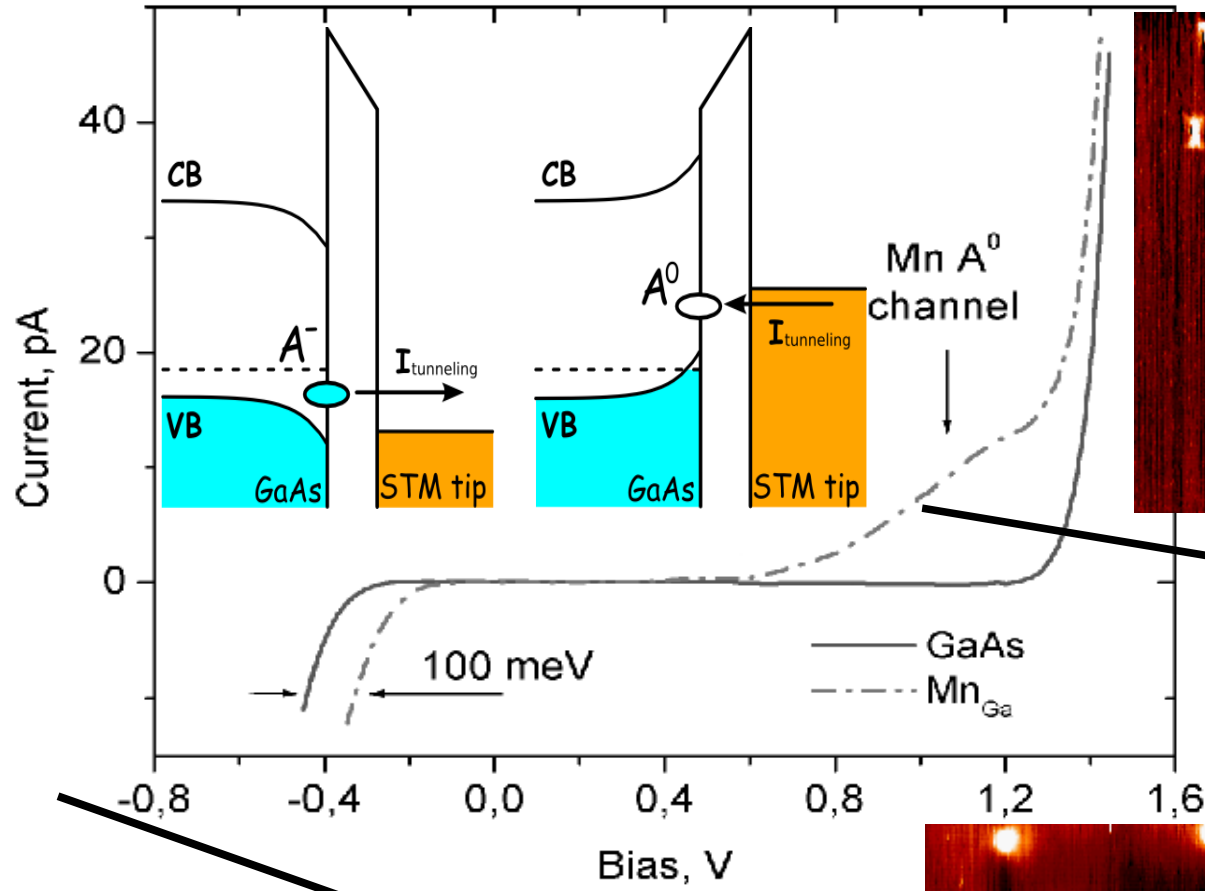
# LDOS near a Mn dopant

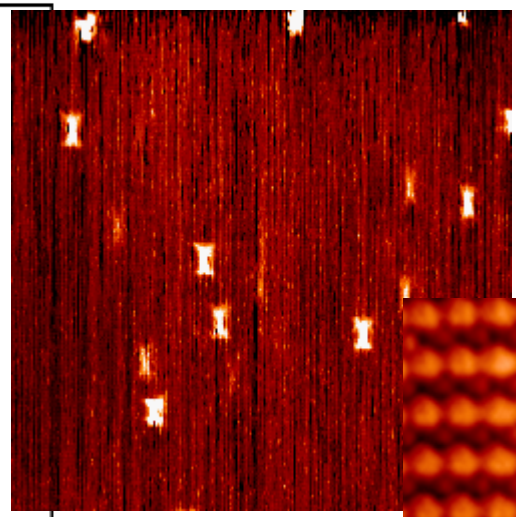


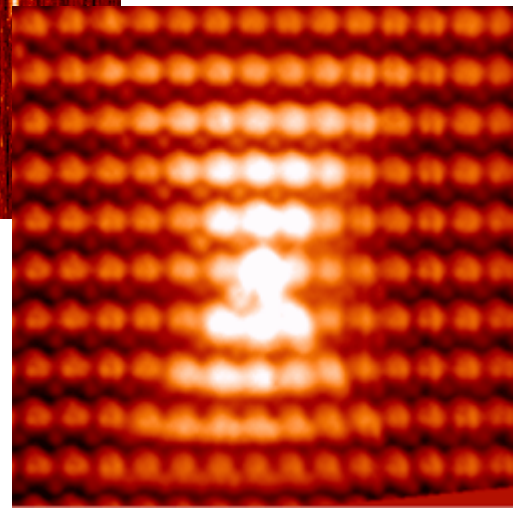



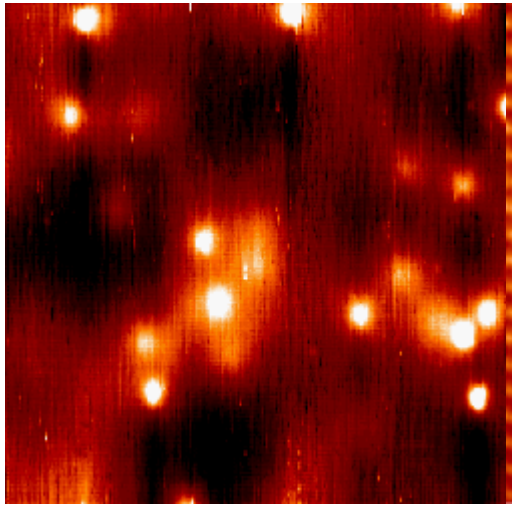

J.-M. Tang and M. E. Flatté, PRL 92, 047201 (2004)

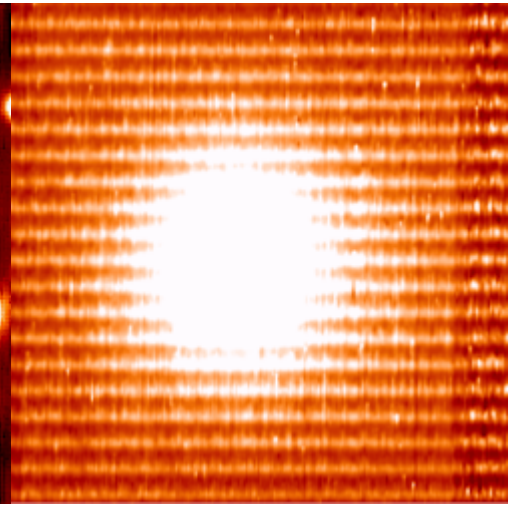
# Scanning Tunneling Microscopy



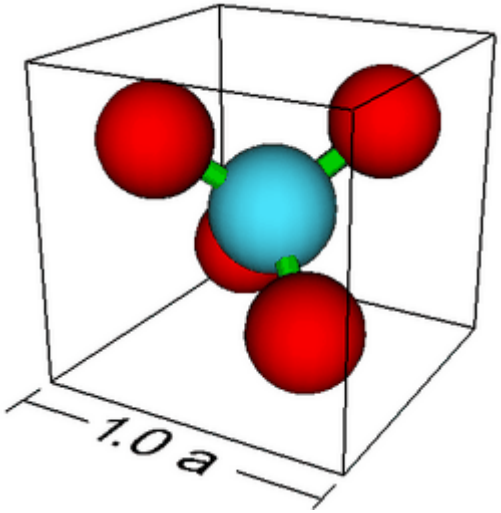


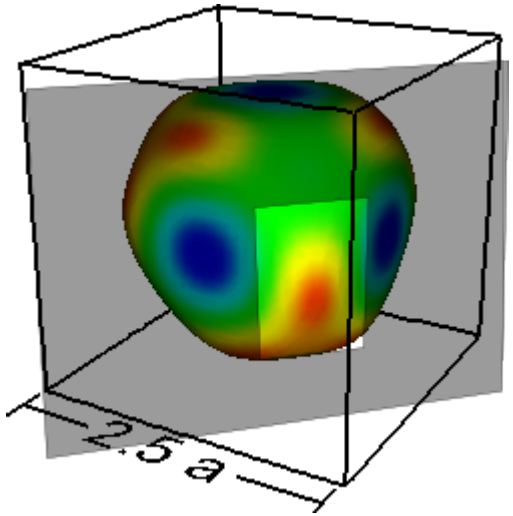


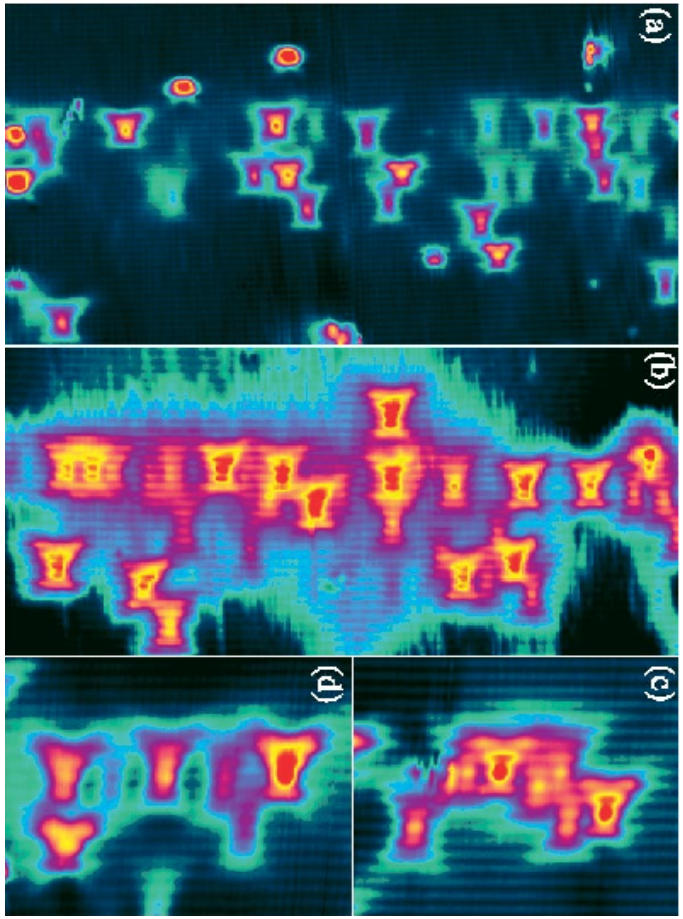


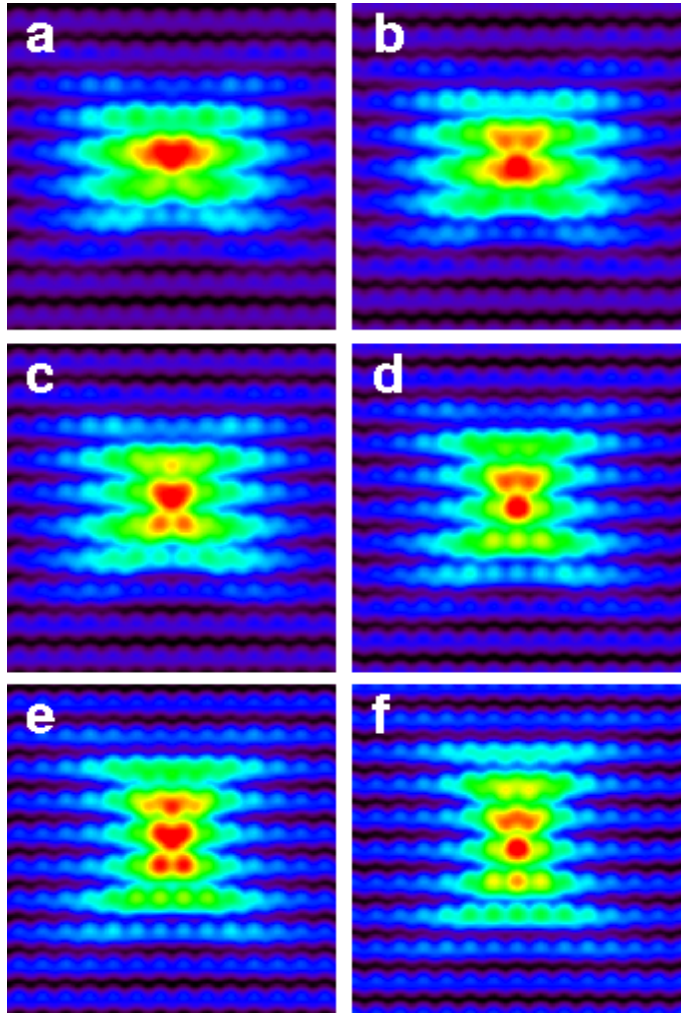
A.M. Yakunin et al.
Physica E 21, 947 (2004)

# Mn acceptor wavefunction



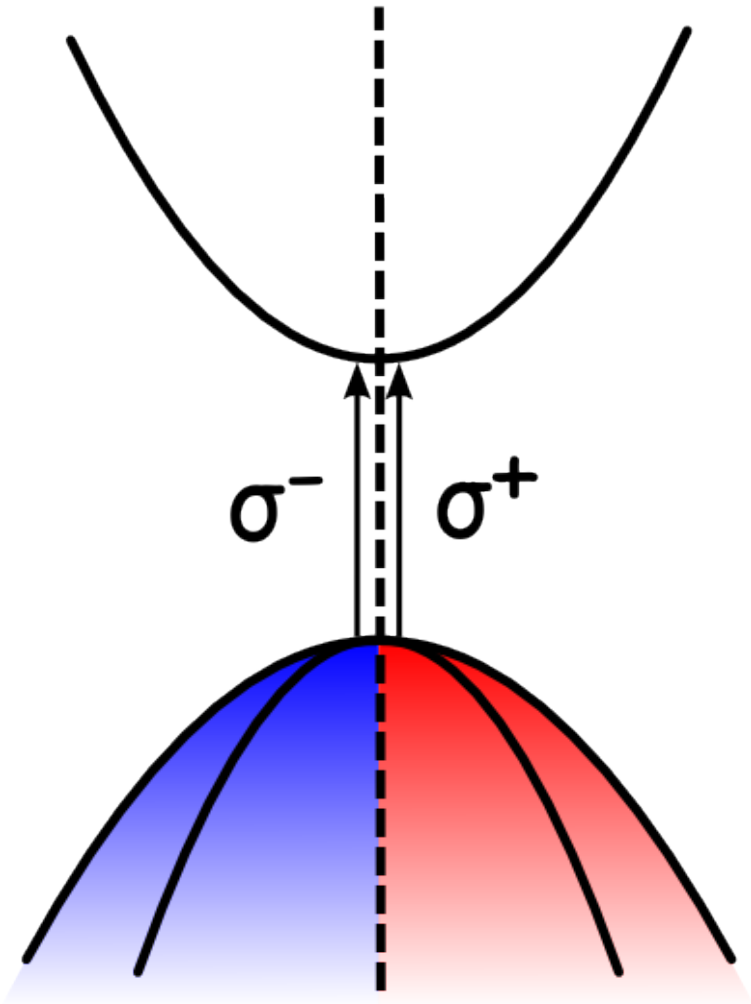



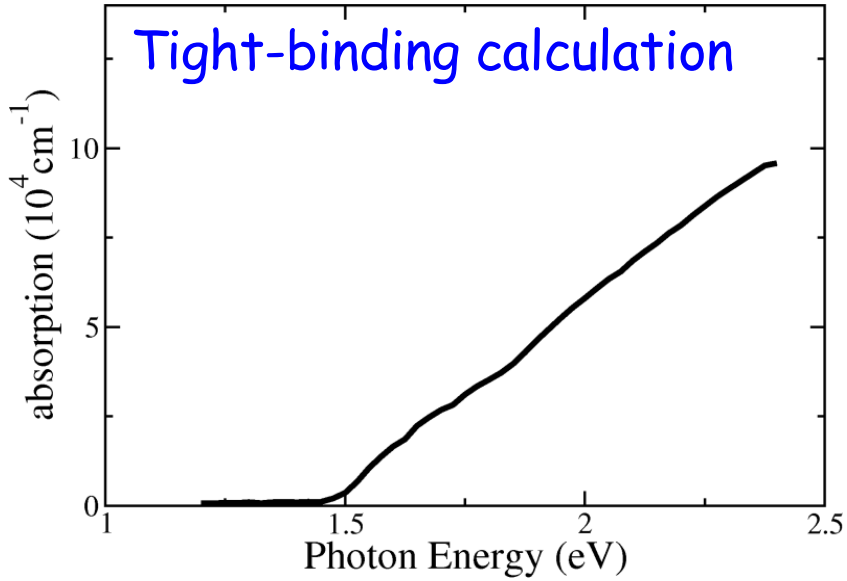

A.M. Yakunin et al.,
PRL 95, 256402 (2005)

# Magnetic circular dichroism in optical absorption

# Optical absorption in bulk semiconductors

- Momentum is a good quantum number
- Interband transitions



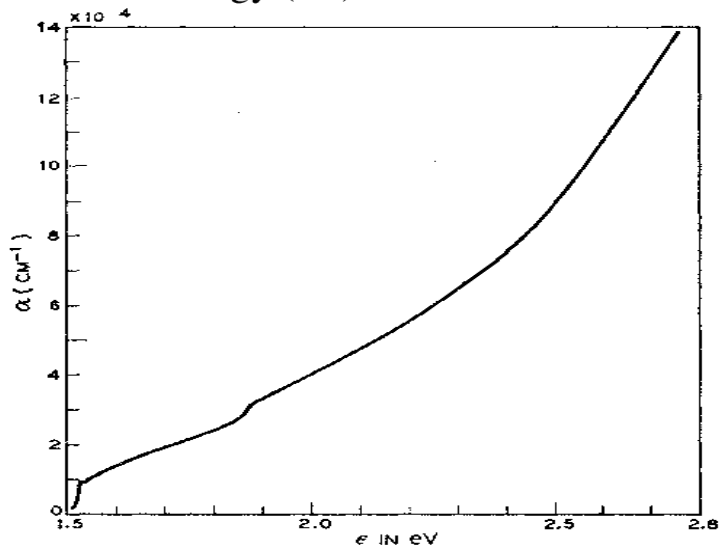
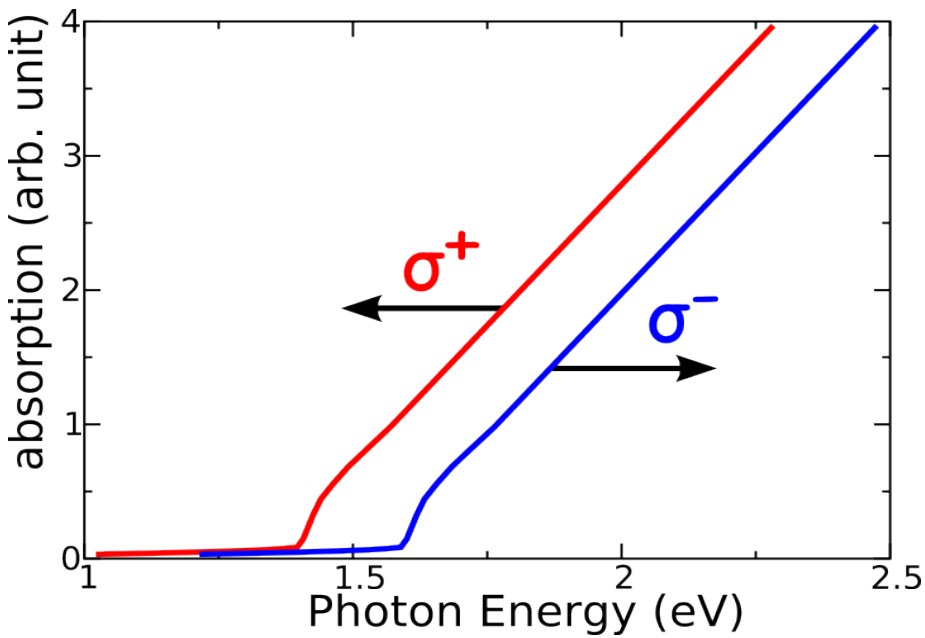
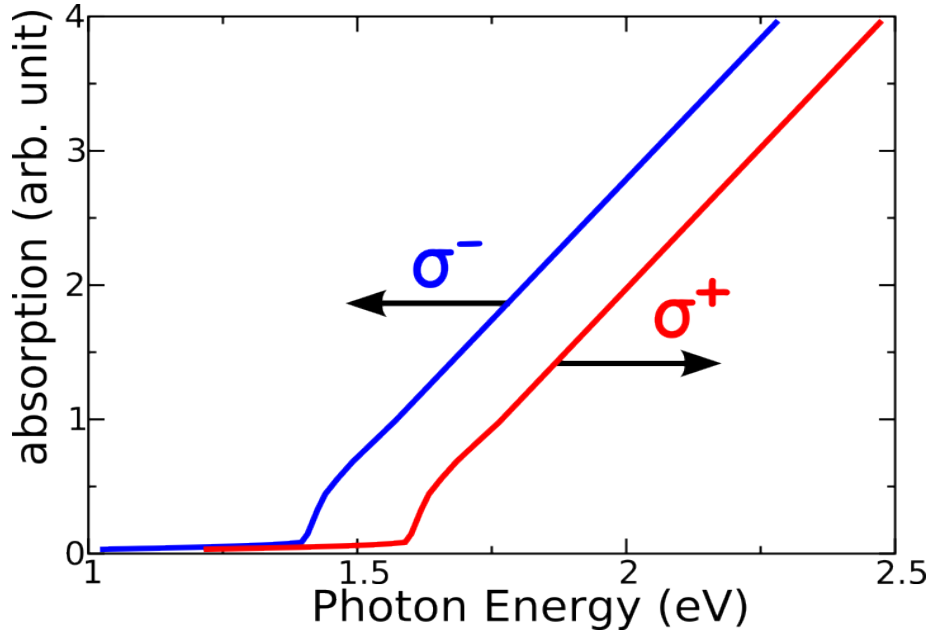



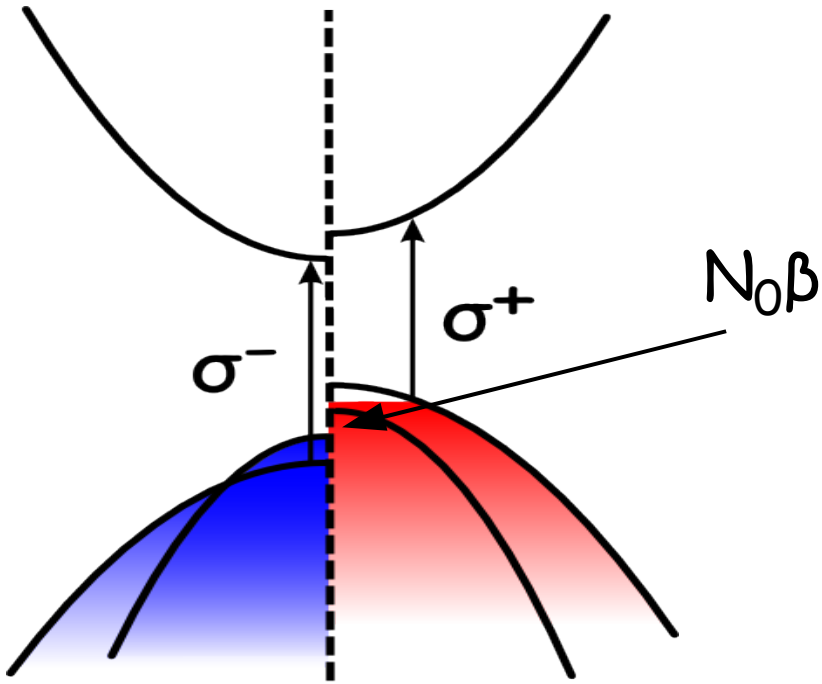

FIG. 6. Absorption of GaAs at 21°K.

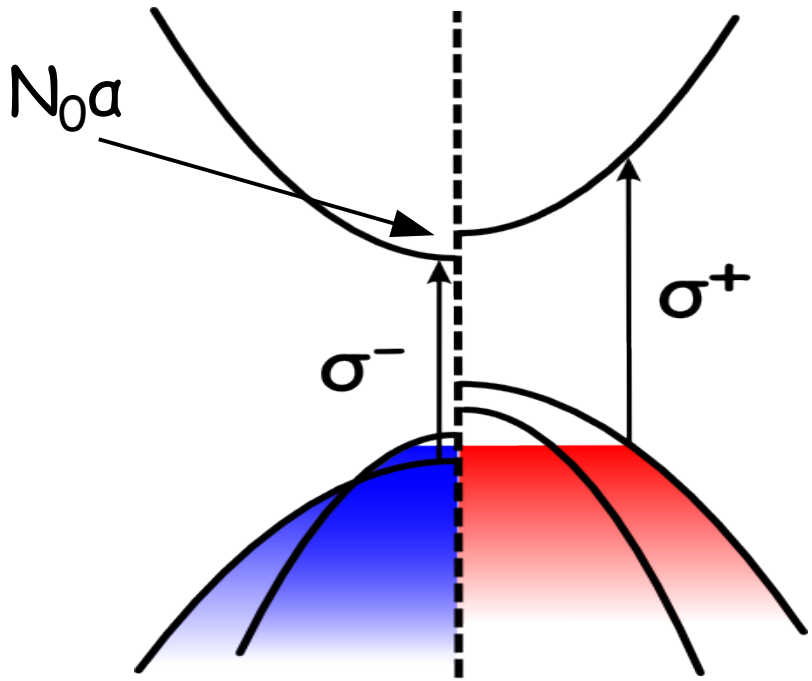
M.D. Sturge, Phys. Rev. 137, 768 (1962)

# Exchange splitting and Moss-Burnstein shift

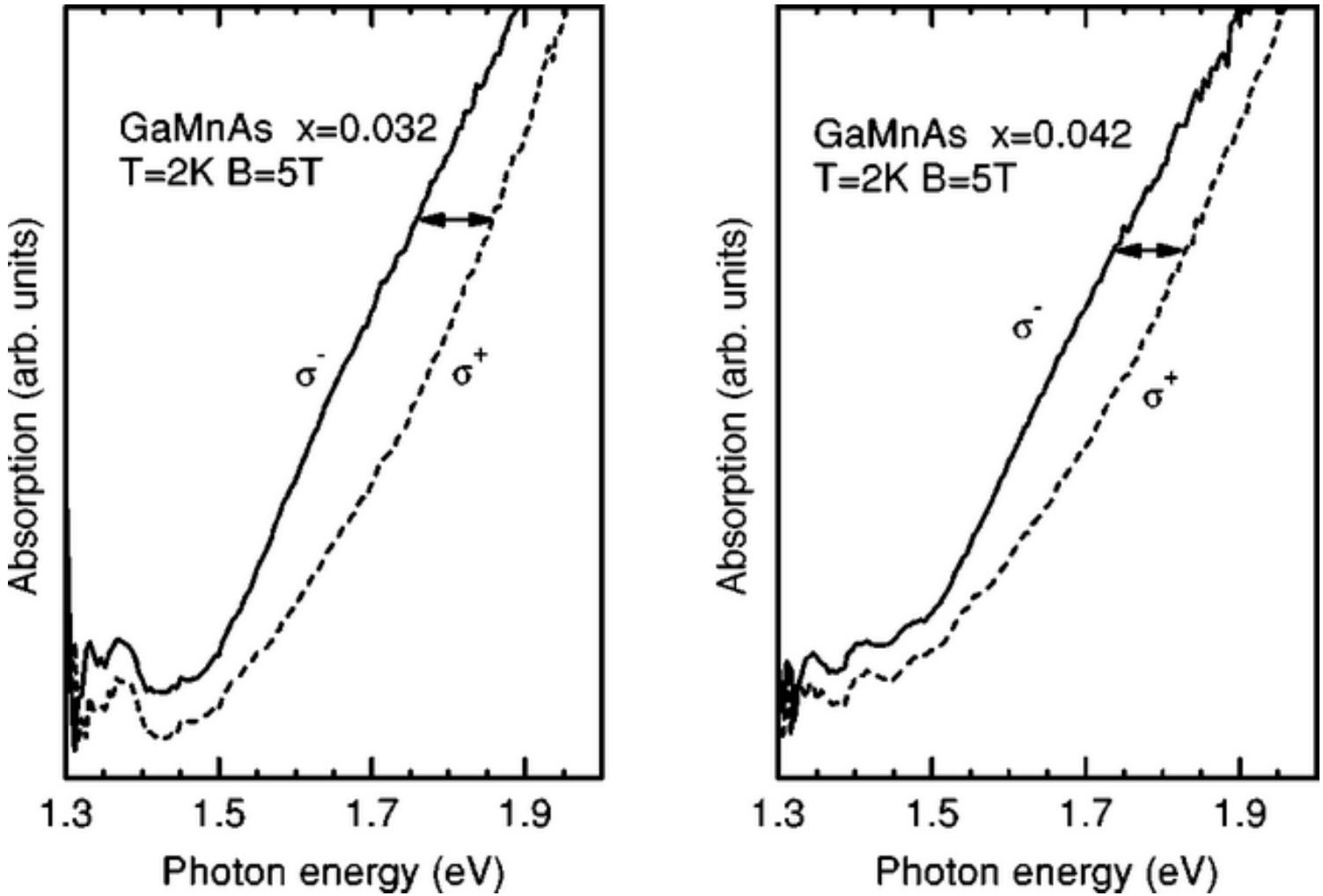


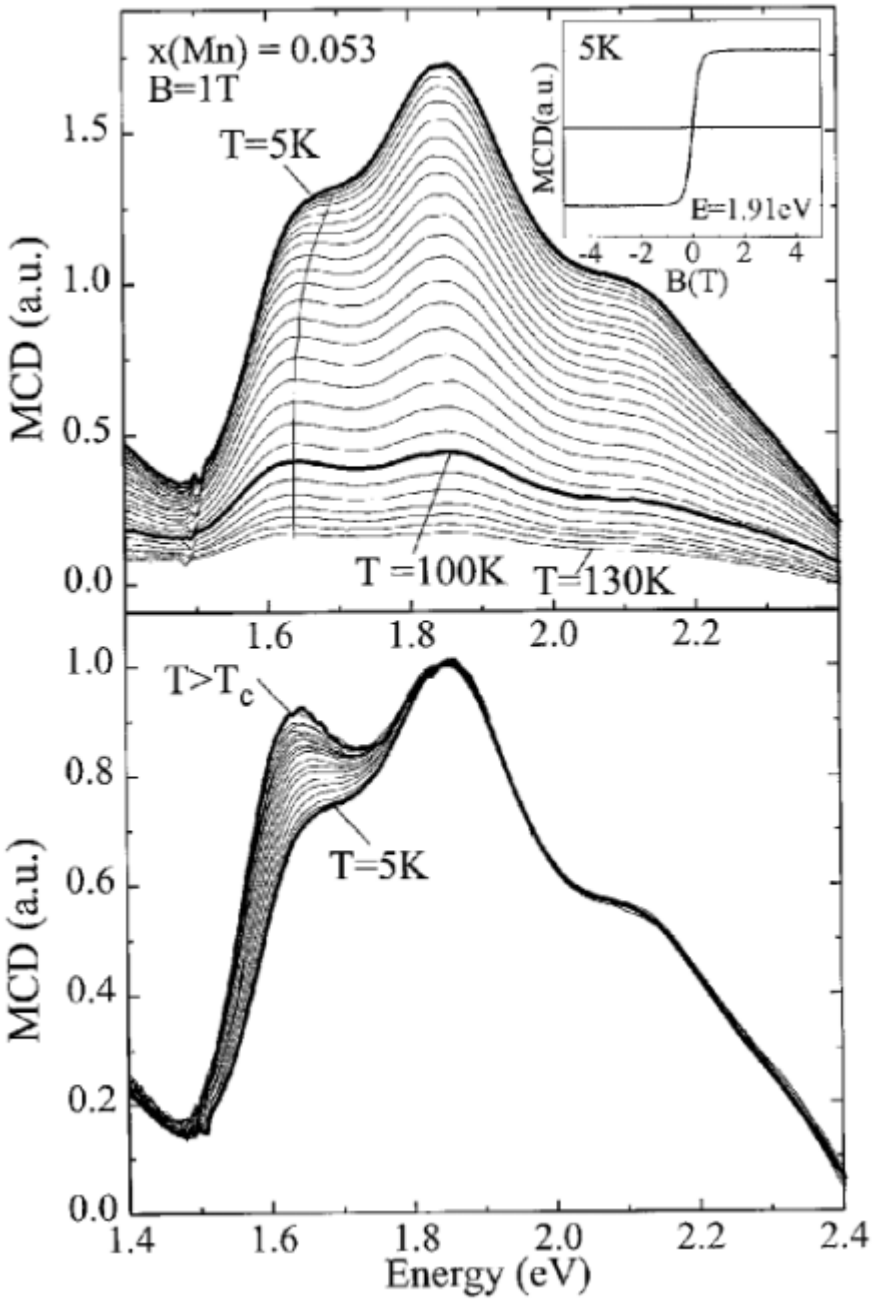



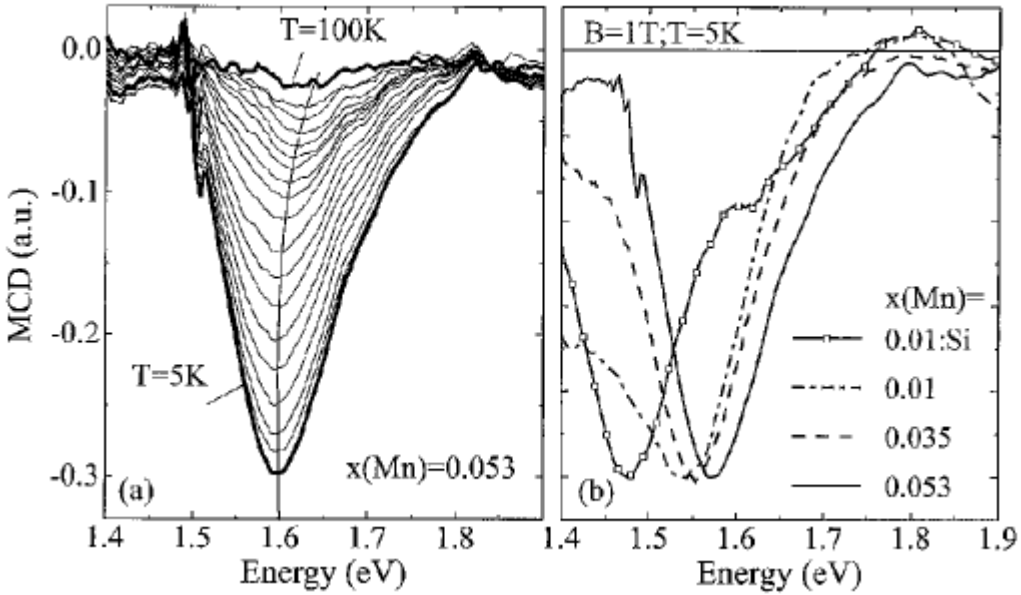
# Magneto-optical absorption




J. Szczytko et al., PRB 59, 12935 (1999)

# Magnetic circular dichroism

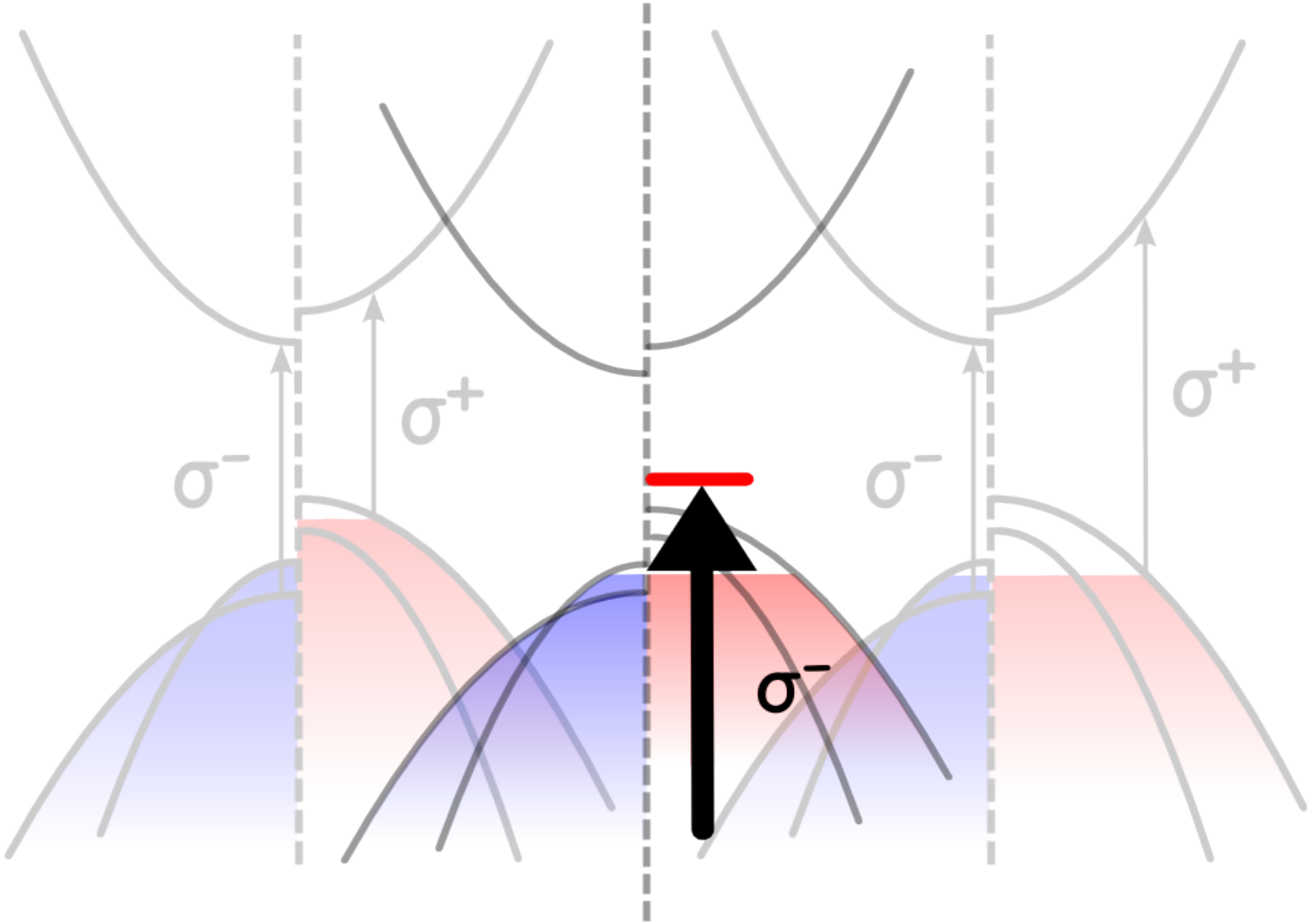


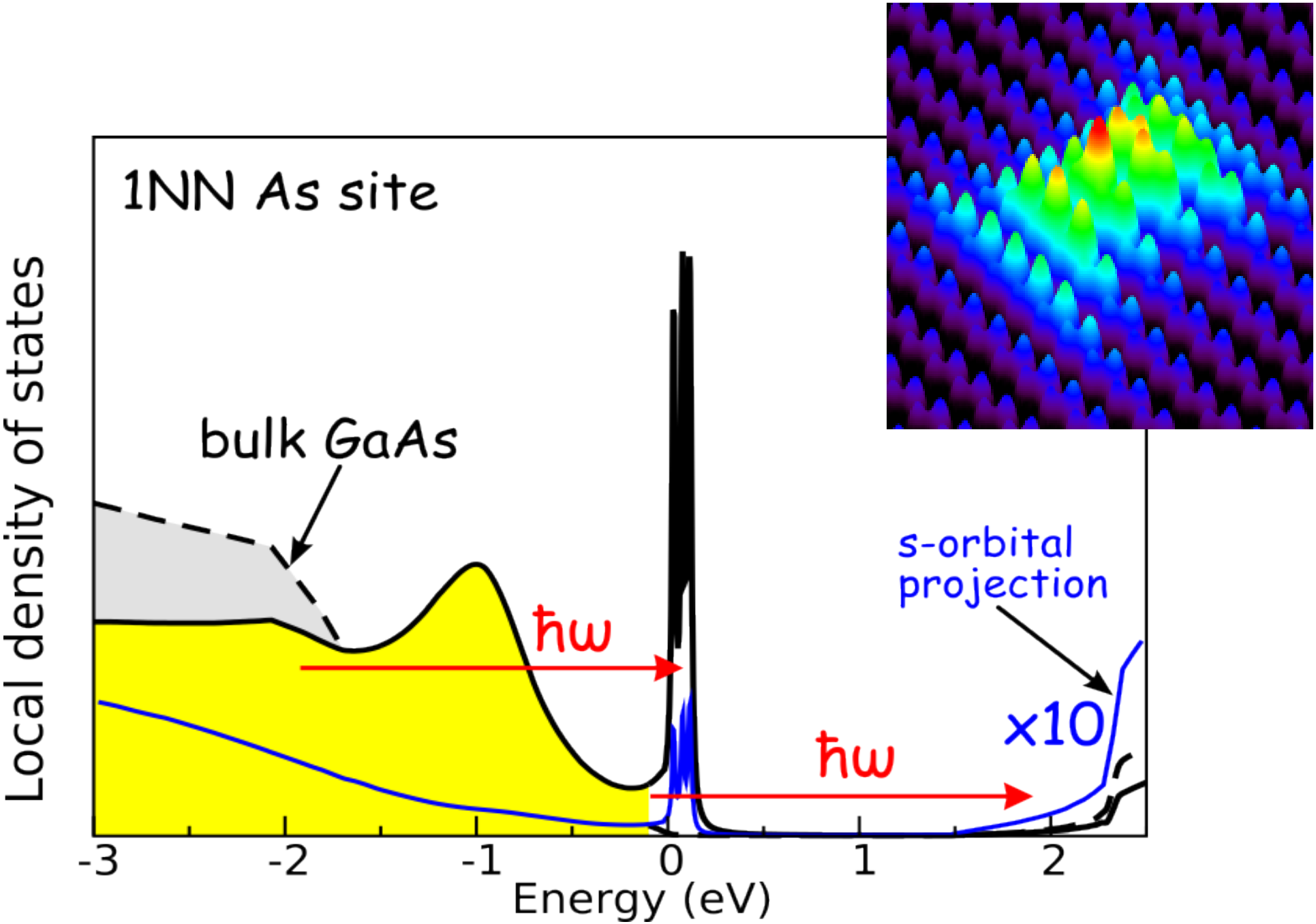




$$\mathrm{MCD} = \frac{\alpha^- - \alpha^+}{\alpha^- + \alpha^+}$$

B. Beschoten et al., PRL 83, 3073 (1999)

# Momentum is not a good quantum number




1NN As site
bulk GaAs
ħω
ħω
s-orbital
projection
x10
Local density of states
-3
-2
-1
0
1
2
Energy (eV)

# absorption coefficients

$$\hat{G}_0^{R,A}(\mathbf{k};\omega) = \left[\omega - \hat{H}_0(\mathbf{k}) \pm \varepsilon\right]^{-1}$$

$$\hat{G}(\mathbf{R},\mathbf{R}';\omega) = \hat{G}_0(\mathbf{R},\mathbf{R}';\omega) + \sum_{\mathbf{R}_a,\mathbf{R}_b} \hat{G}(\mathbf{R},\mathbf{R}_a;\omega)\hat{V}(\mathbf{R}_a,\mathbf{R}_b)\hat{G}_0(\mathbf{R}_b,\mathbf{R}';\omega)$$

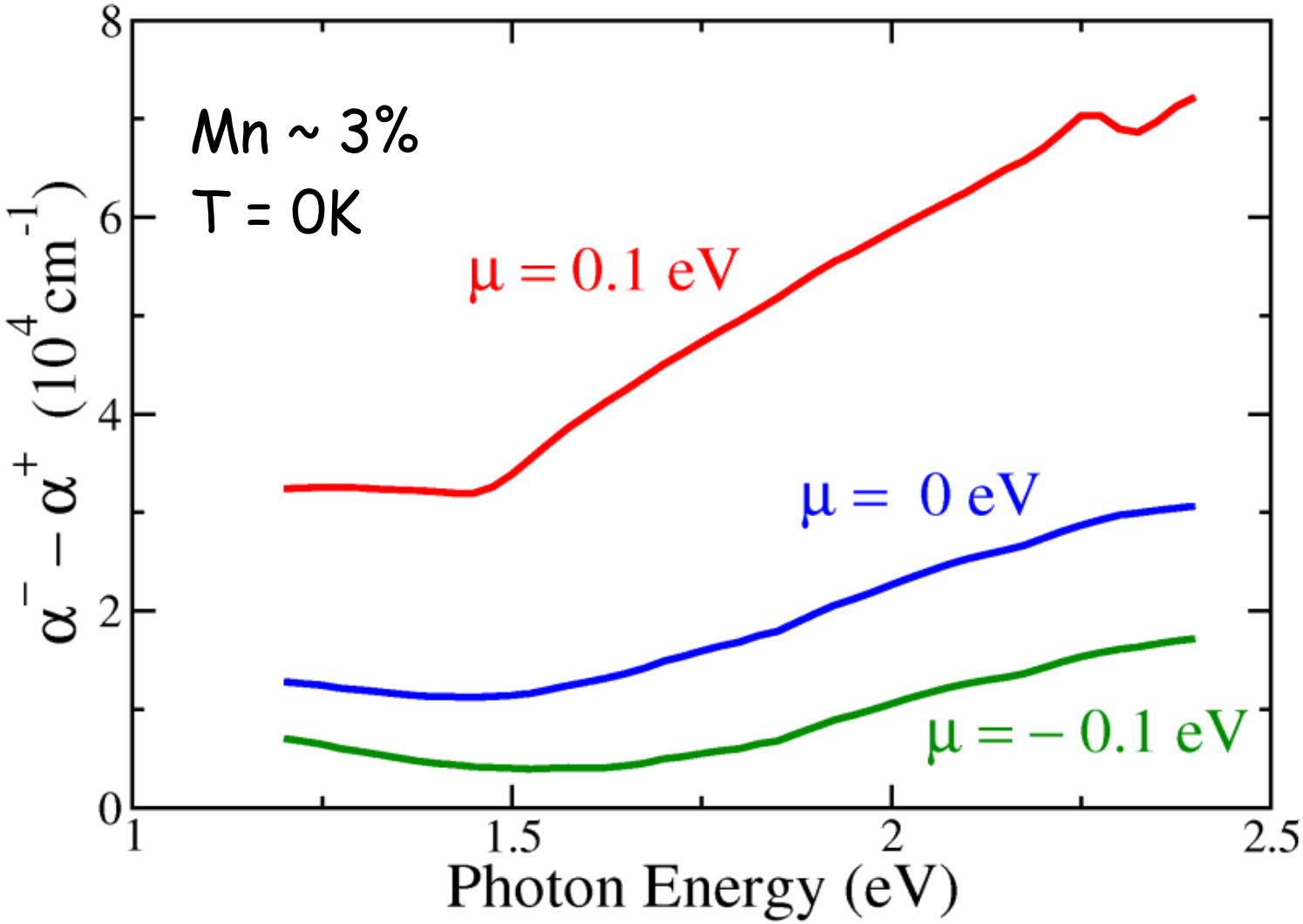
$$\alpha^{\pm} = \frac{\pi e^2}{(2\pi)^2 V n \varepsilon_0 m^2 \omega c} \int dE_f \int dE_i f(E_i)\left[1 - f(E_f)\right]\delta(E_f - E_i - \hbar\omega)$$

$$\times \mathrm{Tr} \sum_{\mathbf{R}_1,\mathbf{R}_2,\mathbf{R}_3,\mathbf{R}_4} \left[\hat{G}^R(\mathbf{R}_1,\mathbf{R}_2;E_i) - \hat{G}^A(\mathbf{R}_1,\mathbf{R}_2;E_i)\right]\left\langle \phi(\mathbf{R}_2)\middle|\hat{P}^{\pm\dagger}\middle|\phi(\mathbf{R}_3)\right\rangle$$

$$\times\left[\hat{G}^A(\mathbf{R}_3,\mathbf{R}_4;E_f) - \hat{G}^R(\mathbf{R}_3,\mathbf{R}_4;E_f)\right]\left\langle \phi(\mathbf{R}_4)\middle|\hat{P}^{\pm}\middle|\phi(\mathbf{R}_1)\right\rangle$$

approximation: only on-site momentum matrix element

# Magnetic circular dichroism



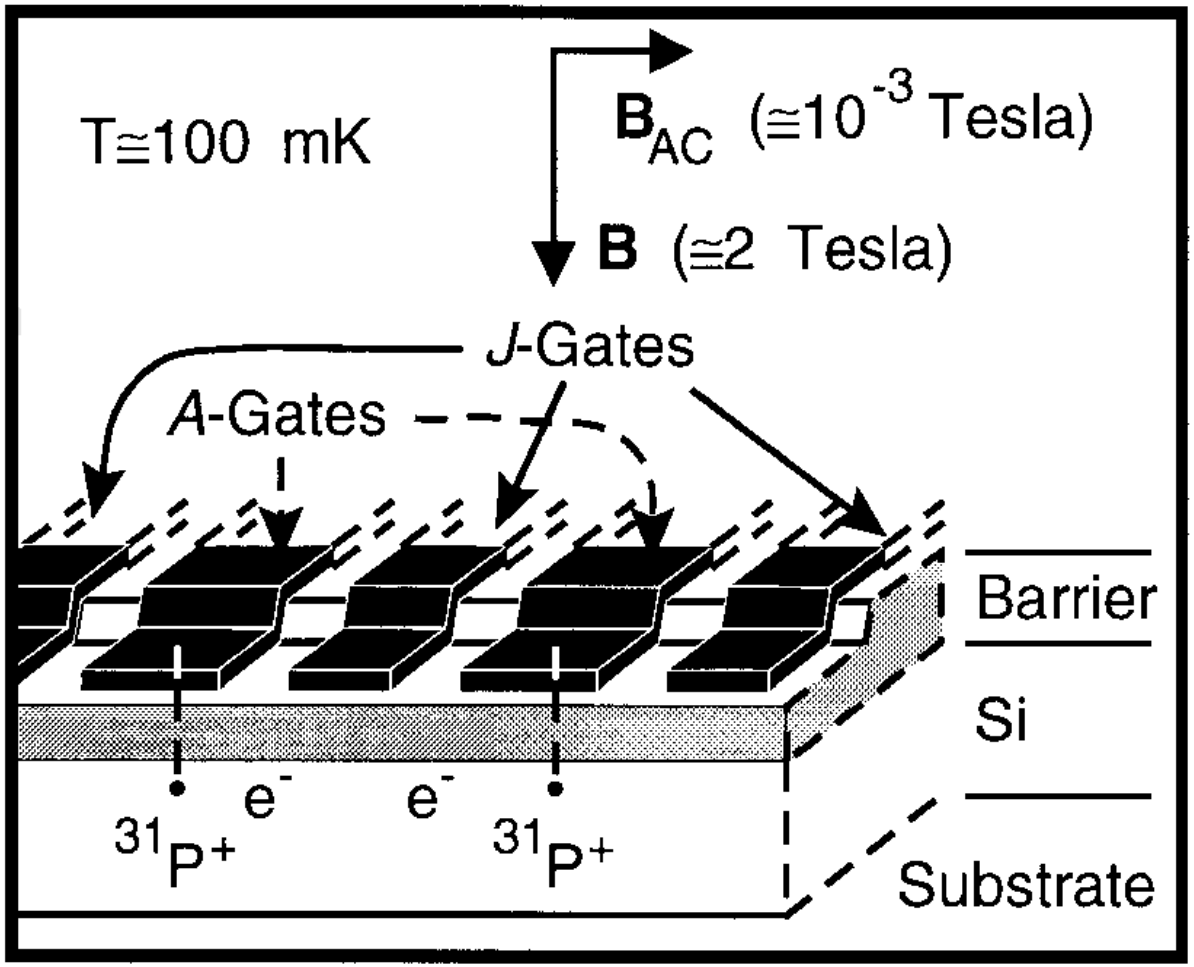
# Summary

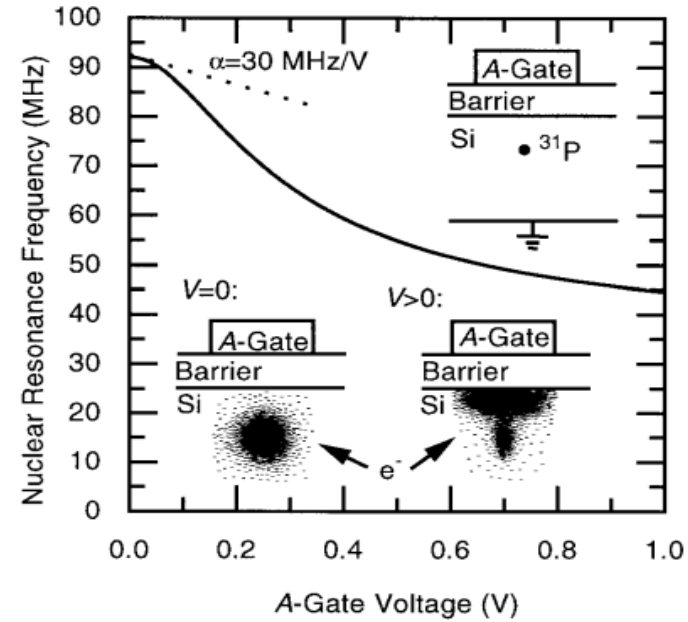
- A new theoretical tool for calculating optical processes in the presence of impurity band
- Optical transitions from the valence band to the impurity band are the main contribution to the magnetic circular dichroism in diluted magnetic semiconductors

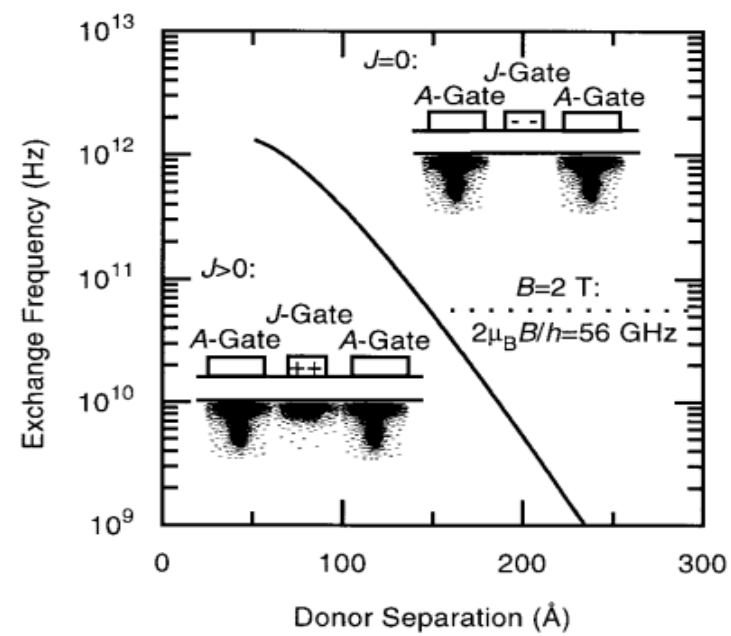
# Electrical manipulation of single Mn spins

J.-M. Tang, J. Levy, and M. E. Flatté
quant-ph/0605203

# Si:P quantum computer




B. E. Kane, Nature 393, 133 (1998)

# Electrical manipulation of electron spins

- Gigahertz electron spin manipulation using voltage-controlled g-tensor modulation

  Y. Kato et al., Science 299, 1201 (2003)

- Coherent manipulation of coupled electron spins in semiconductor quantum dots

  J. R. Petta et al., Science 309, 2180 (2005)

# Quantum computation

- qubit
- initialization
  - apply a dc magnetic field to split spin states
- control
  - apply an ac magnetic field to flip spin
  - two-qubit operation
- detection

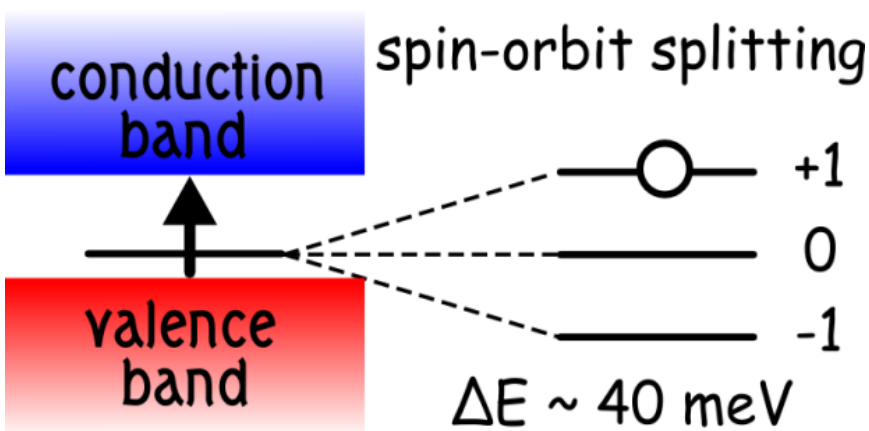
# Quantum spin states

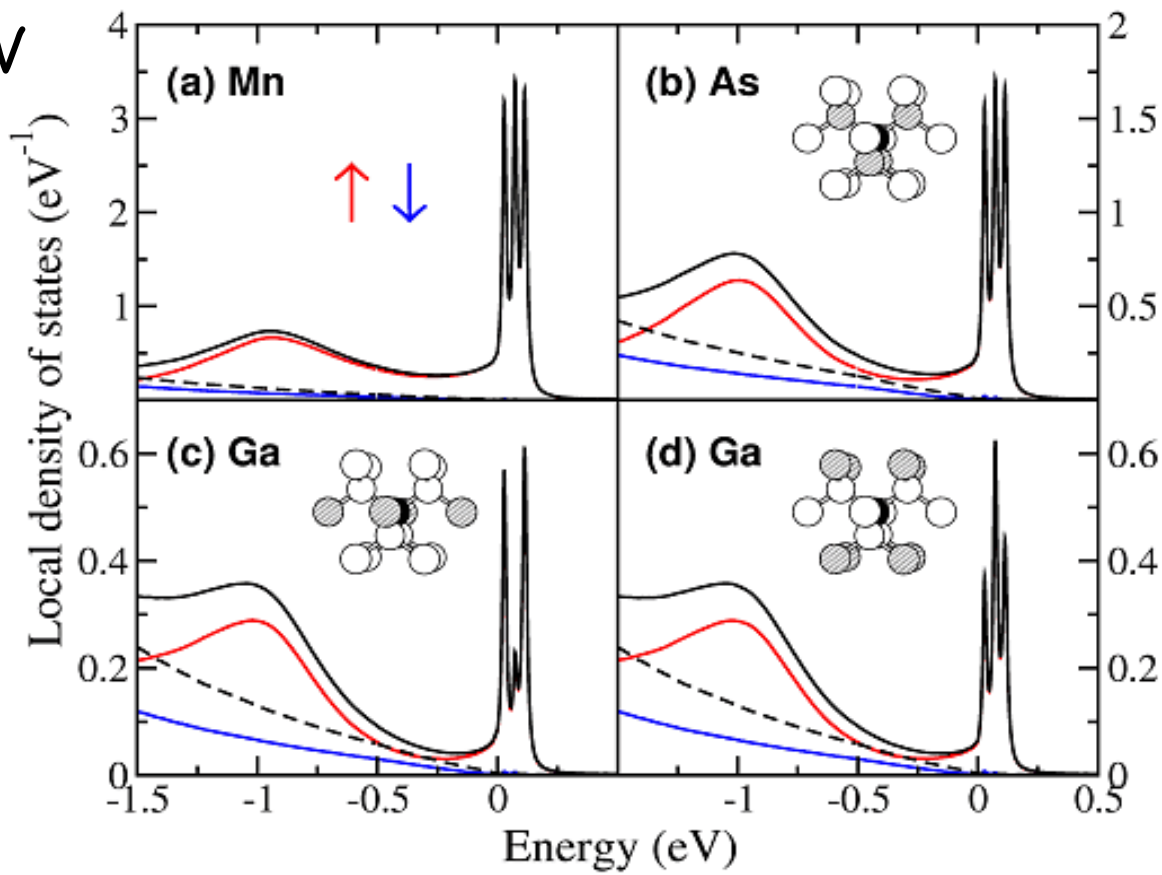
$$H_{eff} = A\, \mathbf{S}_{Mn} \cdot \mathbf{s}_h + B\, \mathbf{s}_h \cdot \boldsymbol{\ell}_h$$

$$S_{Mn} = 5/2,\ \ell_h = 1,\ s_h = \tfrac{1}{2}$$

$A \sim -300$ meV, $B \sim 80$ meV

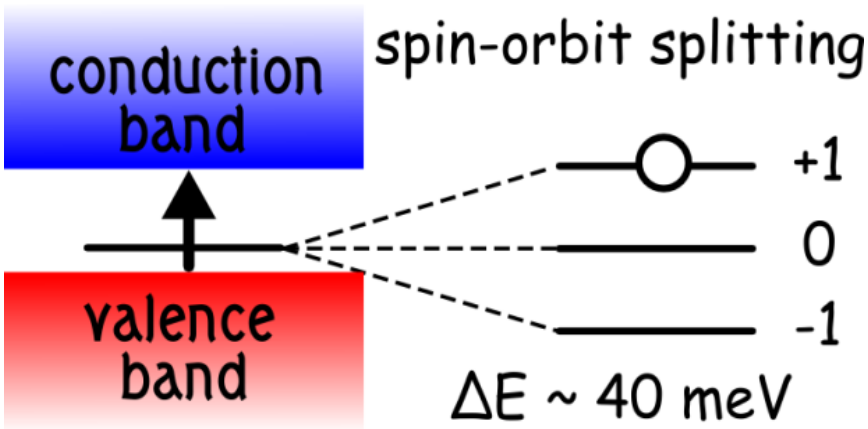
$A < 0$, $B > 0$, $|A| \gg |B|$

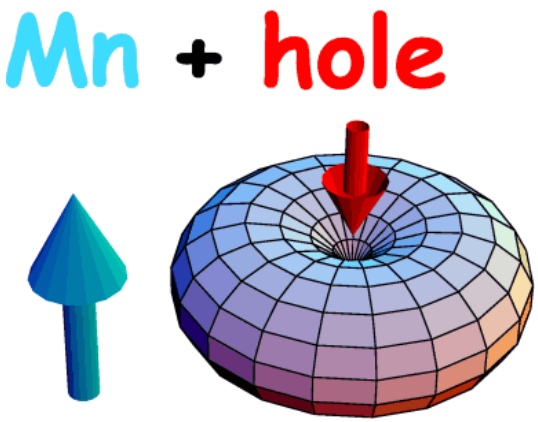


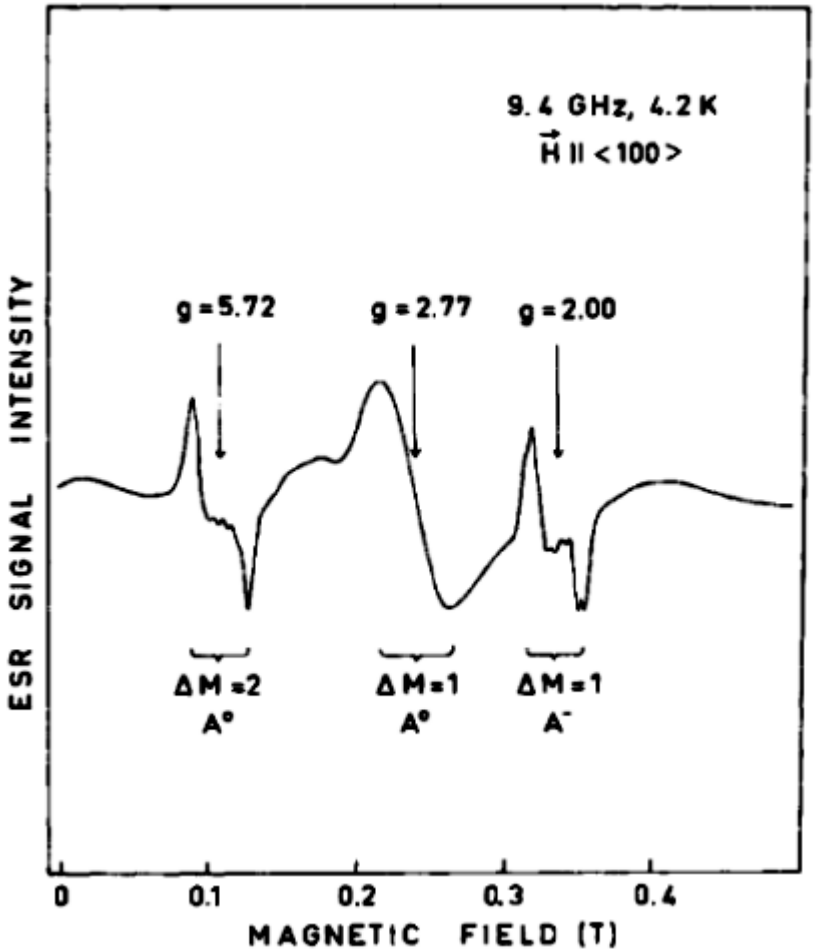



# (Mn+hole) is a J=1 complex

$$H_{eff} = A\, \mathbf{S}_{Mn} \cdot \mathbf{s}_h + B\, \mathbf{s}_h \cdot \boldsymbol{\ell}_h$$



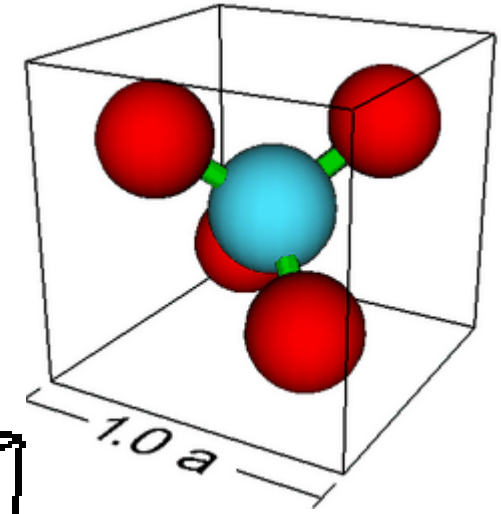



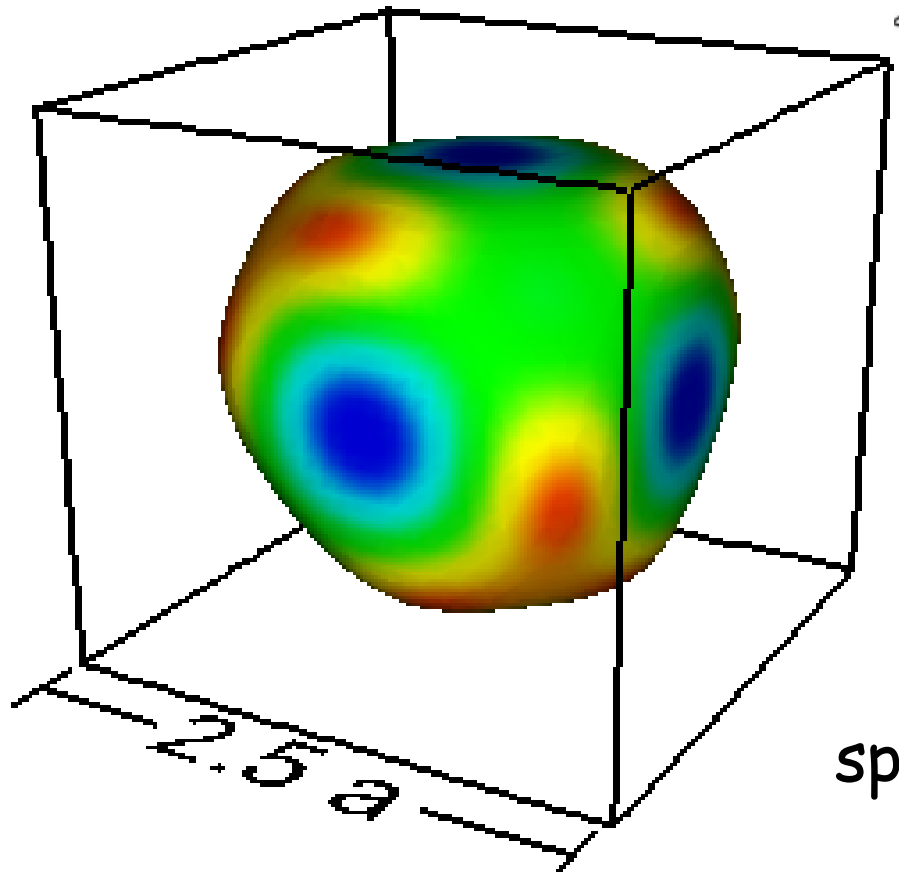




- J. Schneider et al., PRL 59, 240 (1987)

# Broken inversion symmetry

- Inversion asymmetry
  - bulk (Ga v.s. As)
  - impurity location
  - surface




spin averaged

# How to control a spin

- magnetic field

$$H_B = g\mu_0 \mathbf{J}\cdot\mathbf{B}$$

- strain

$$H_S = b\left[\epsilon_{xx}\left(\mathbf{J}_x^2 - \mathbf{J}^2/3\right) + \text{c.p.}\right] + d\left[\epsilon_{xy}\left(\mathbf{J}_x\mathbf{J}_y + \mathbf{J}_y\mathbf{J}_x\right) + \text{c.p.}\right]$$

- electric field

$$H_E = \gamma\left[\mathbf{E}_x\left(\mathbf{J}_x\mathbf{J}_y + \mathbf{J}_y\mathbf{J}_x\right) + \text{c.p.}\right]$$

# Energy scales

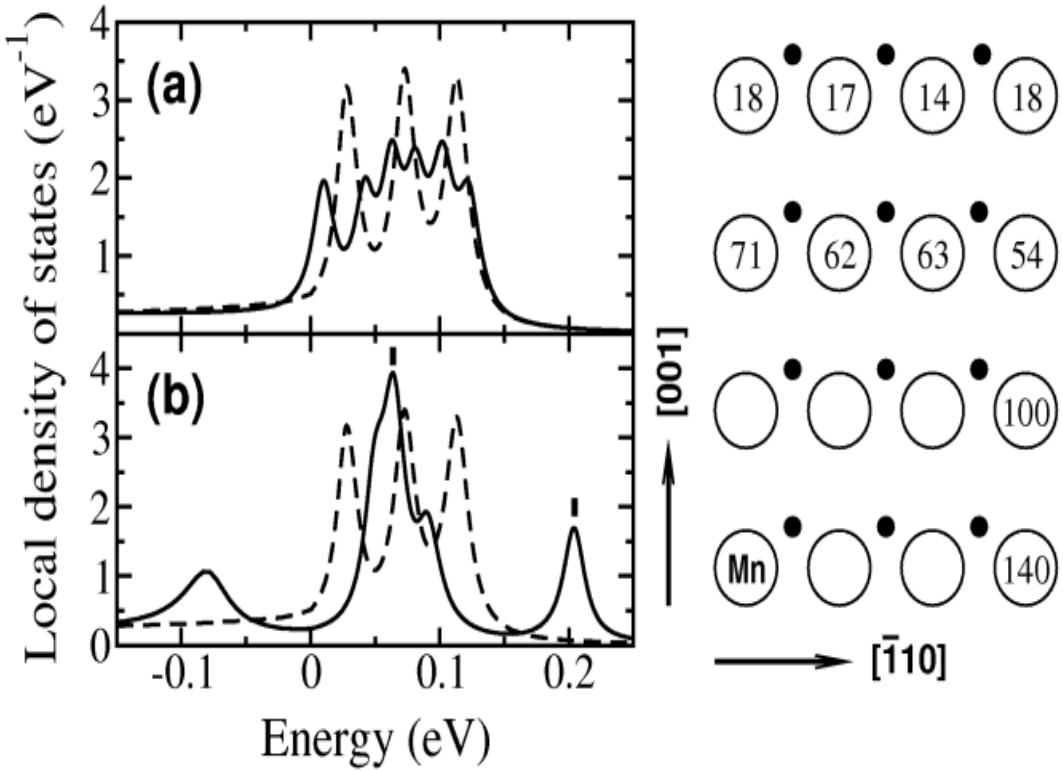
- coupling to E fields

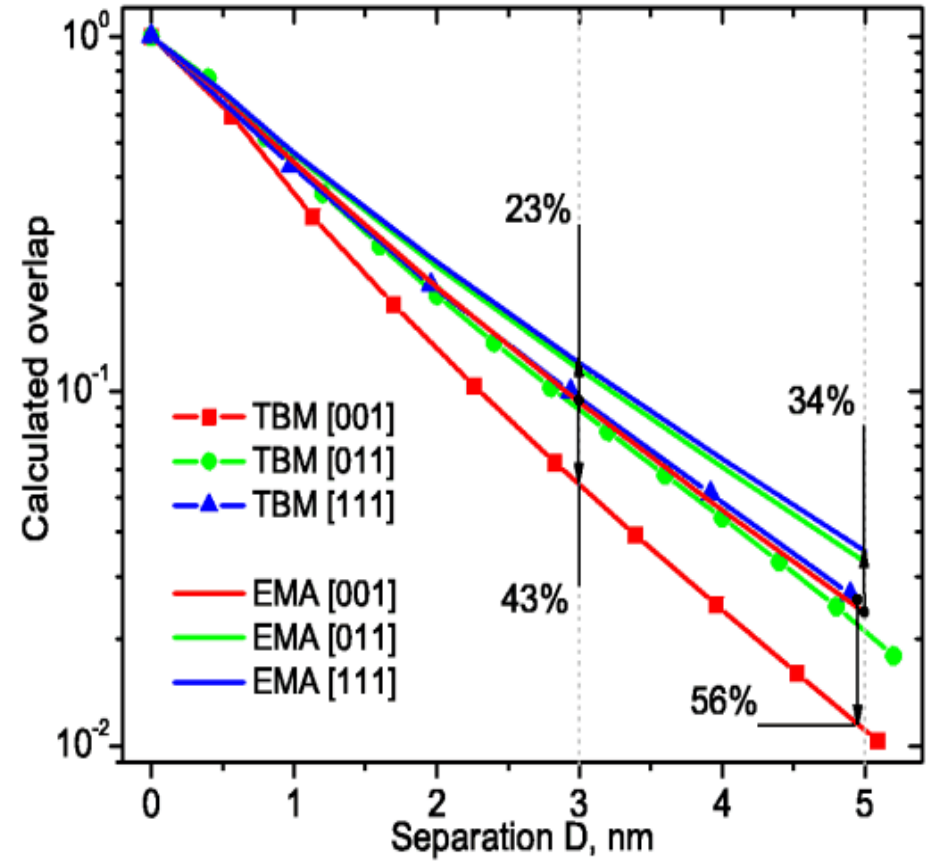
$$\left\langle \hat{\mathbf{n}},+1 \middle| \mathbf{r}\cdot\mathbf{E} \middle| \hat{\mathbf{n}},+1 \right\rangle \quad \Rightarrow \quad \gamma E = 160\,\mu eV$$

$$E = 40\,keV/cm \quad \Leftrightarrow \quad B = 1\,Tesla$$

- Mn-Mn coupling
  - ~10 – 100 $\mu$eV for 10 nm separation

# Mn-Mn overlap

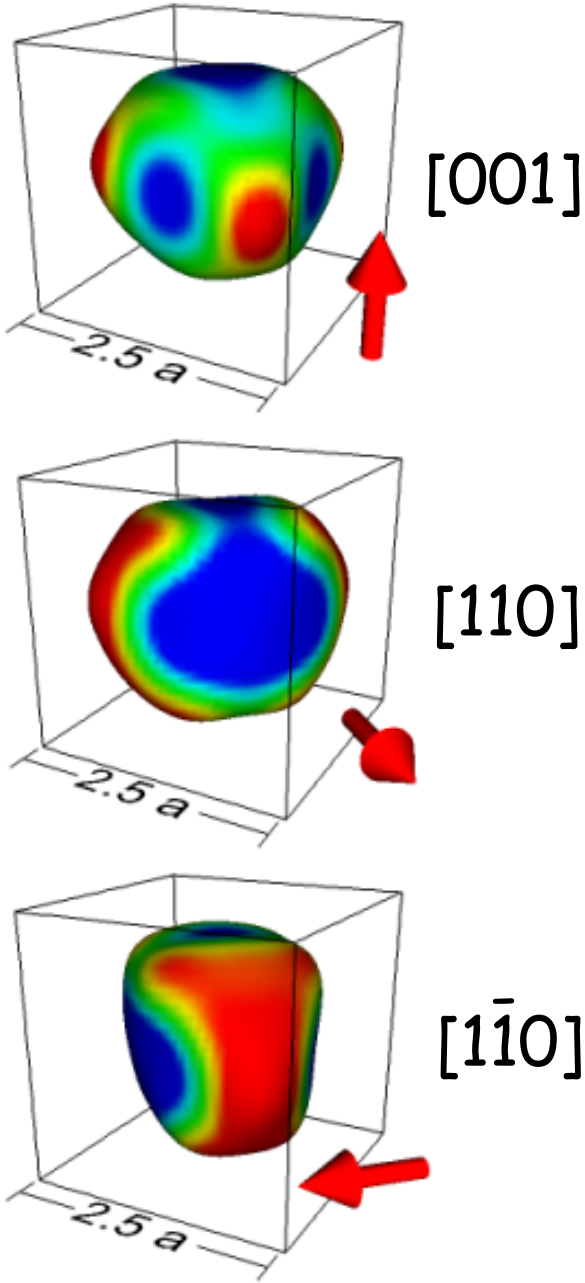


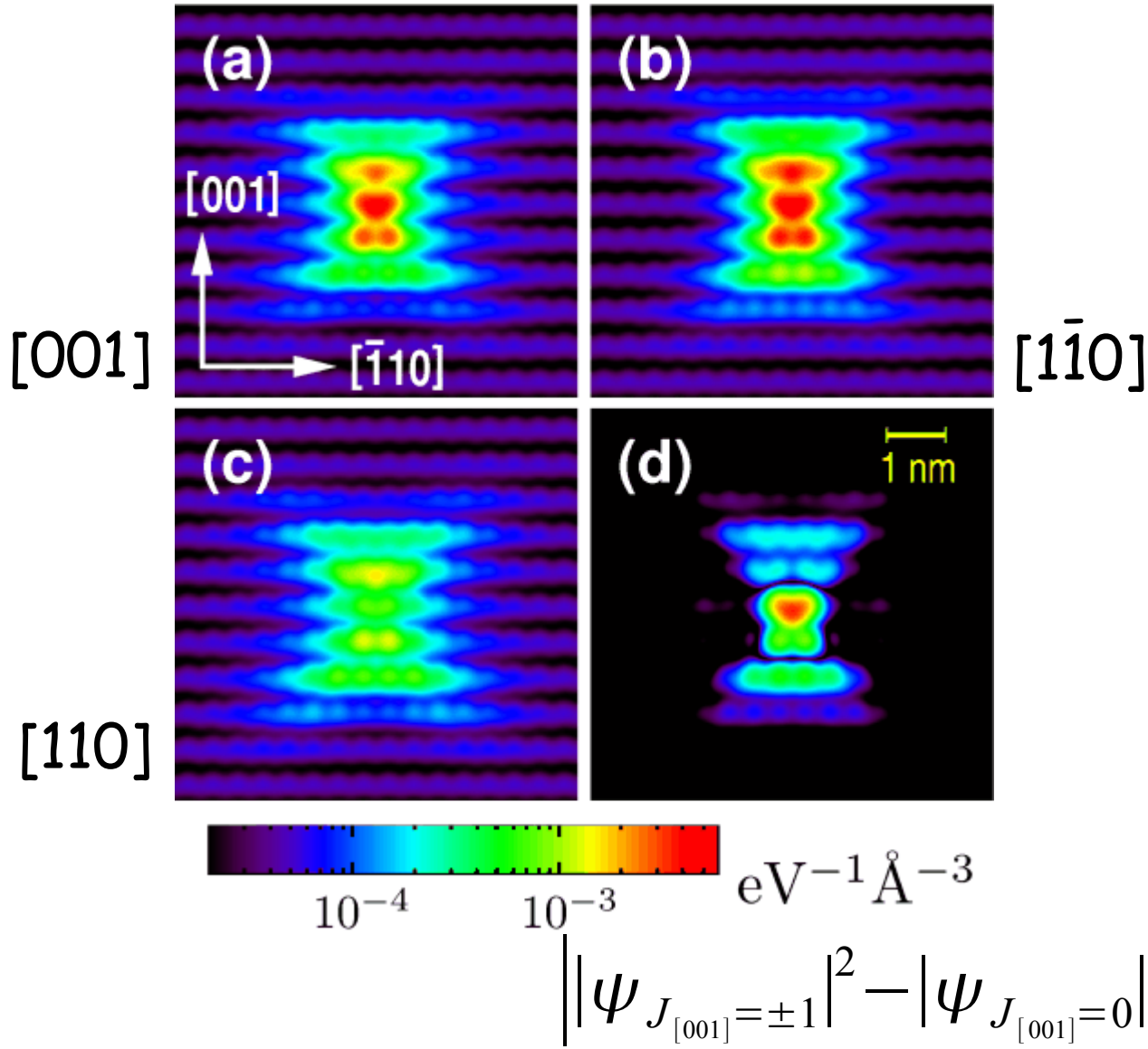




J.-M. Tang and M. E. Flatté,
PRL 92, 047201 (2004)

A.M. Yakunin, et al.,
PRL 95, 256402 (2005)

# Spin-orientation-dependent LDOS



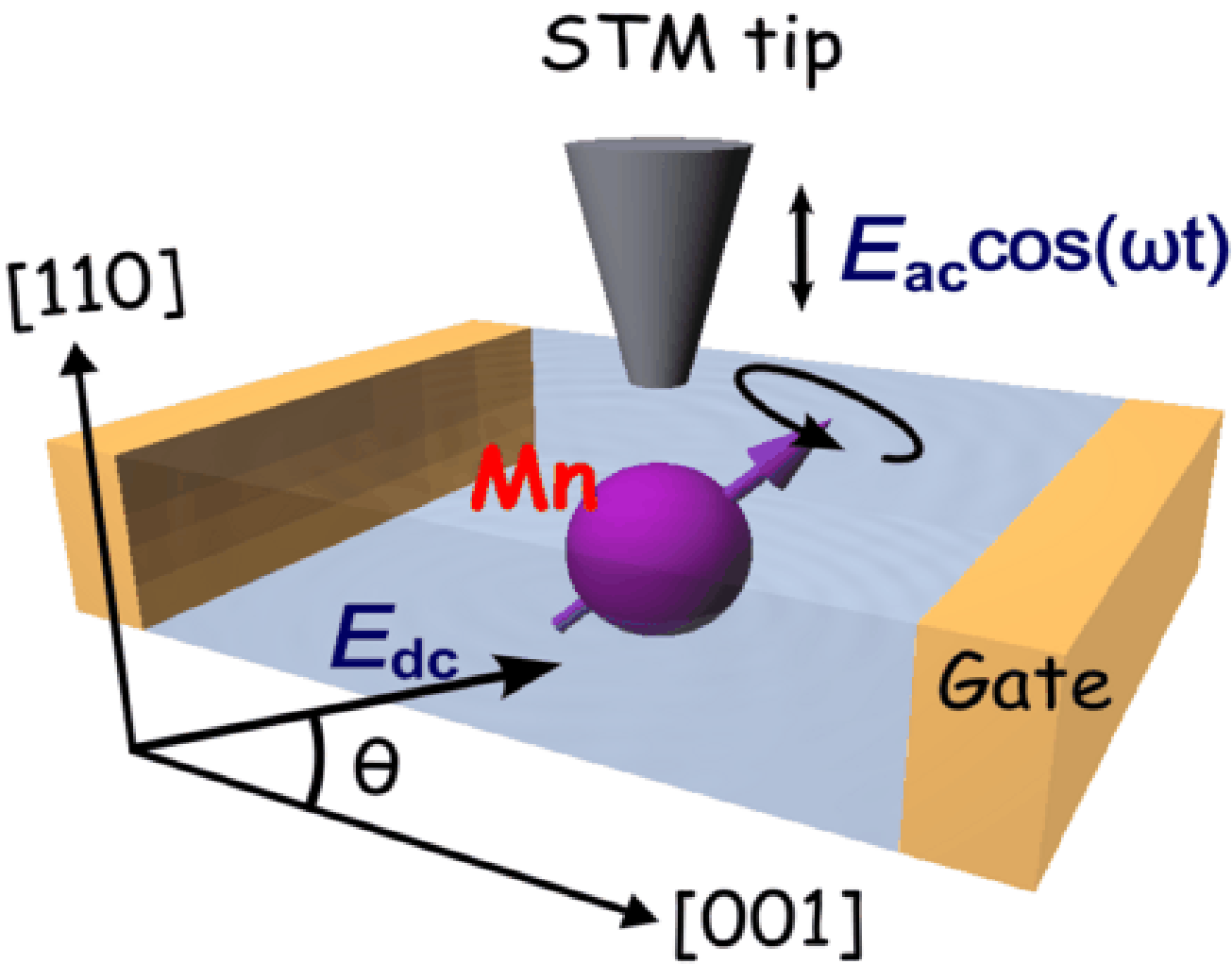




$$\left| \left|\psi_{J_{[001]}=\pm 1}\right|^2 - \left|\psi_{J_{[001]}=0}\right|^2 \right|$$

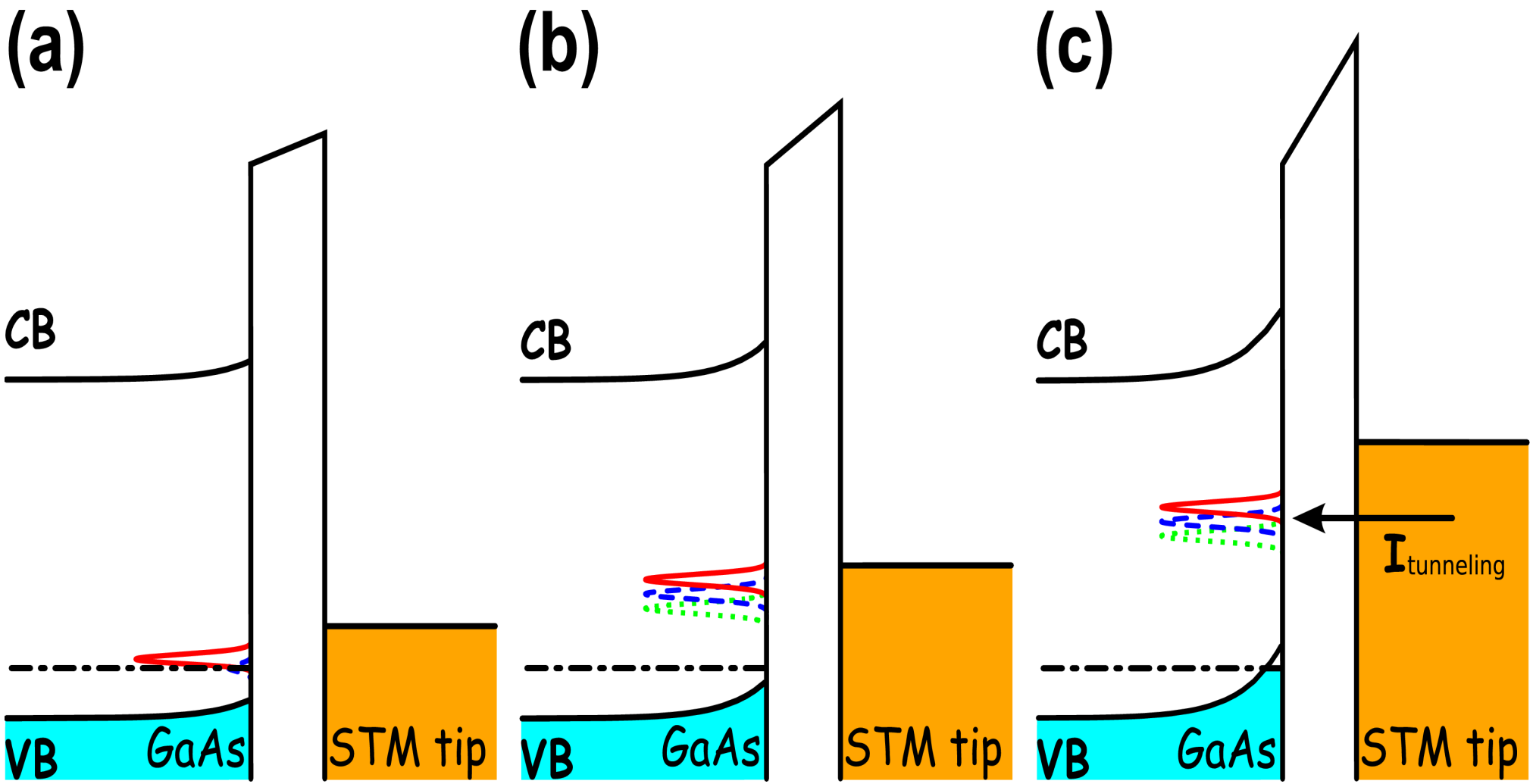
Showing the 4th layer away from the Mn layer

J.-M. Tang and M. E. Flatté, PRB 72, 161315 (2005)

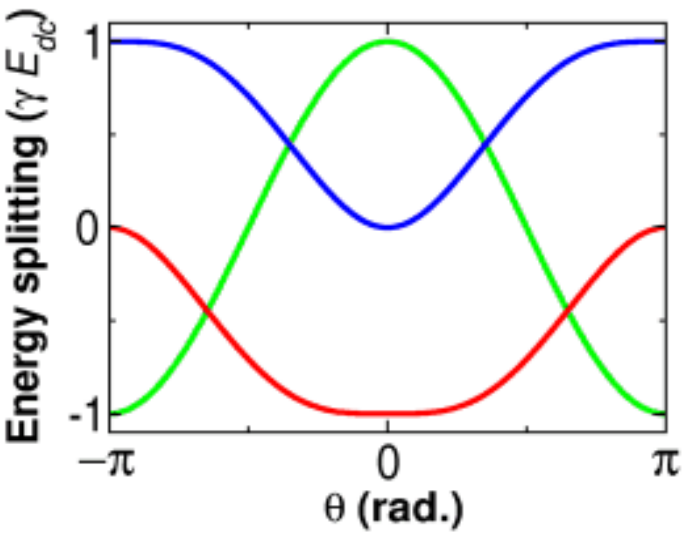
# Theorist's STM setup

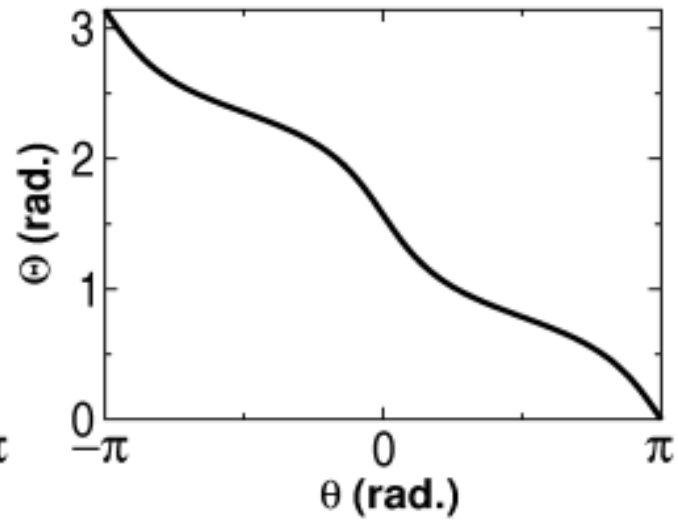


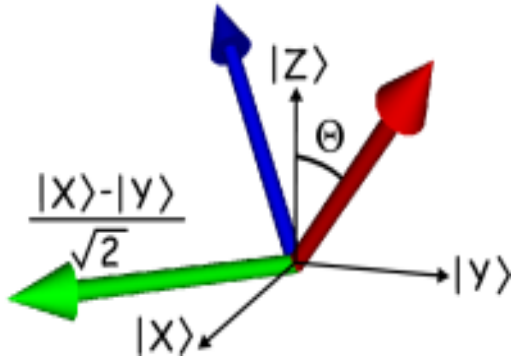
# Manipulation scheme

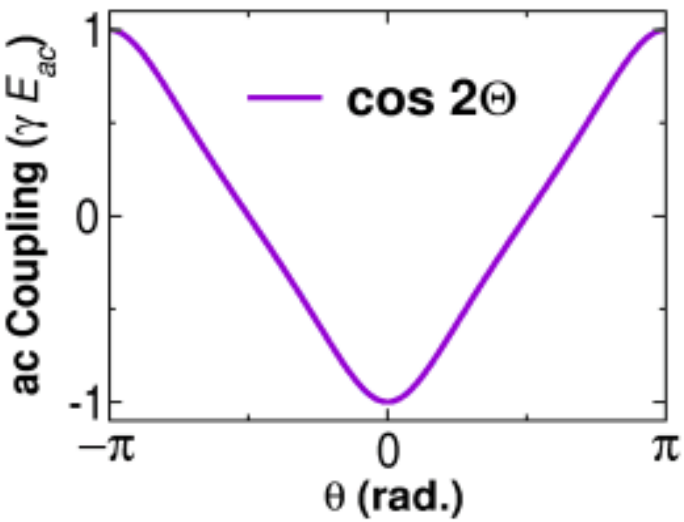


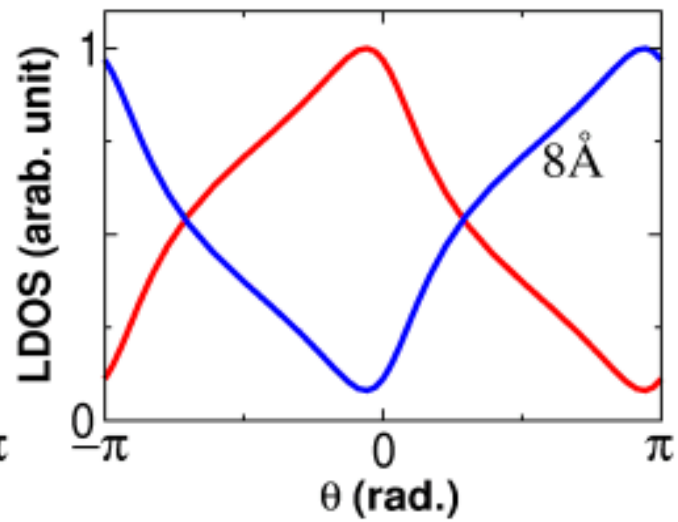
# Manipulation and detection

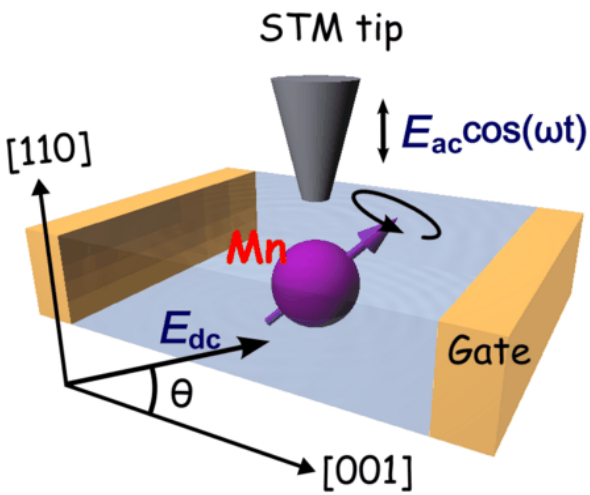


# Summary

- The principal determinant of the anisotropic shape of the Mn wavefunction is the cubic symmetry of the lattice.
- The cubic symmetry is lowered by the dopant substitution, spin-orbit interaction, and bulk inversion asymmetry.
- The combination of spin-orbit anisotropy and broken inversion symmetry leads to the possibility of controlling the spin state of an ionic spin in a solid using electric or strain fields.