# Light-induced charge and spin dynamics in nano and molecular structures

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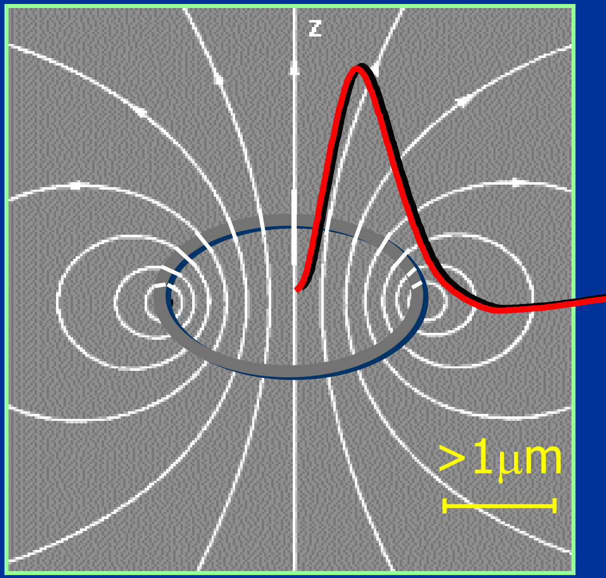
**goal**:

*1. generation of temporally and spatially controlled magnetic pulses by shaped light pulses*

*2. study & control of spin and charge dynamics*

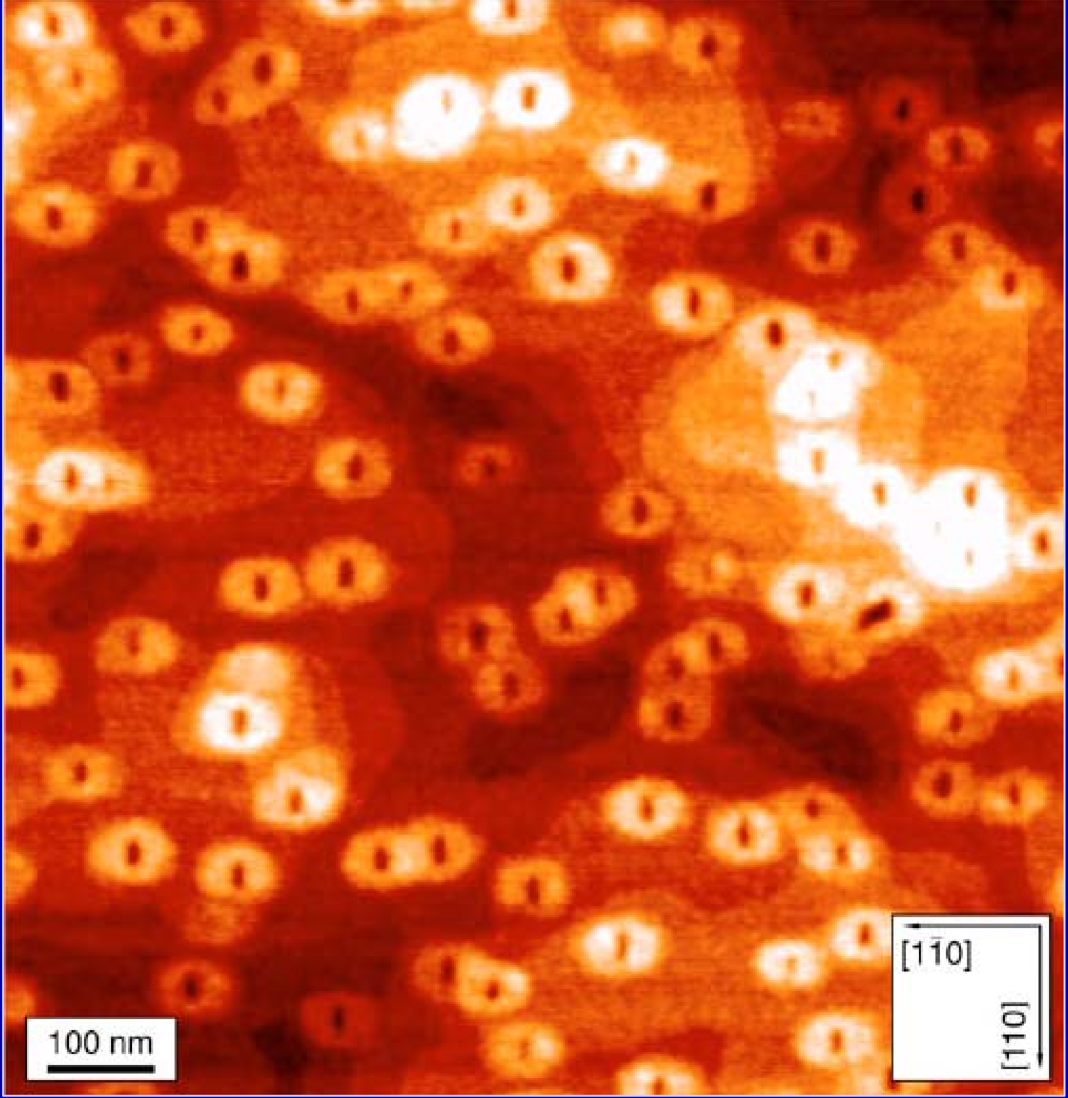



http://qft0.physik.uni-halle.de

# Overview

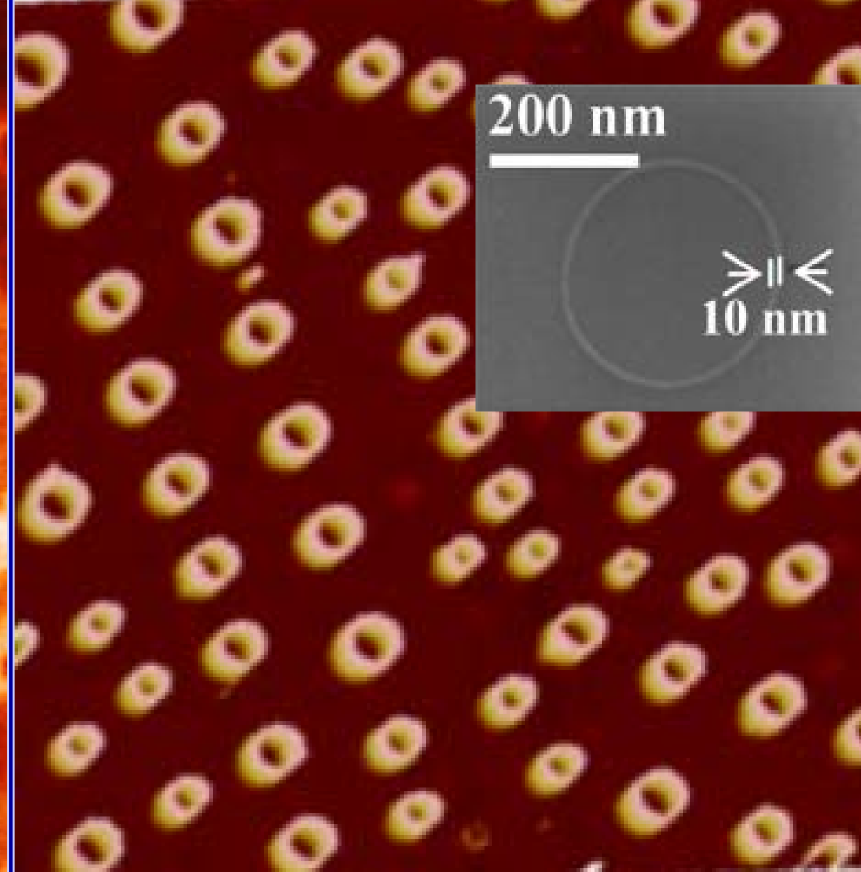
1. introduction: experimental status
2. Aharonov-Bohm effect & persistent currents
3. charge dynamics in meso- & nanoscopic rings
4. relaxation & decoherence
5. applications


*J. Berakdar, MLU , Halle*



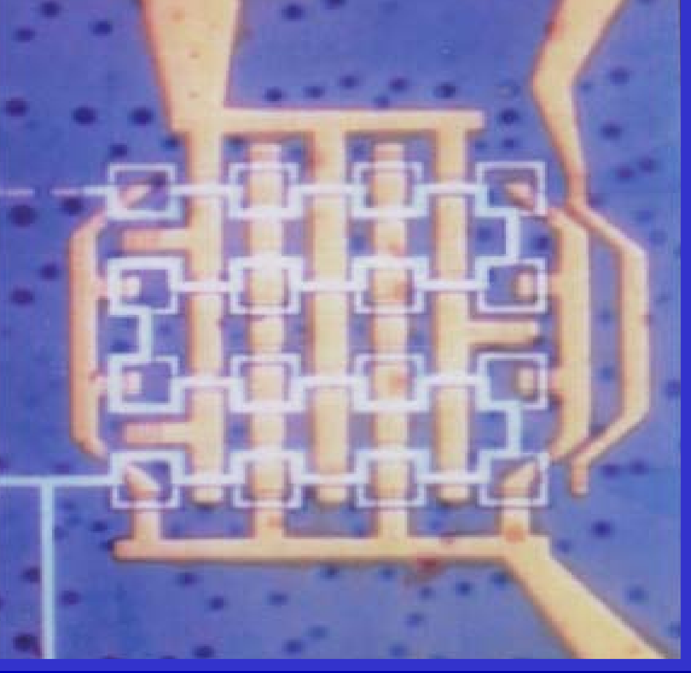

**atomic force microscopy (AFM)**
**of InAs/GaAs rings**

P. Offermans *et al.*
Appl. Phys. Lett. **87**, 131902 (2005)



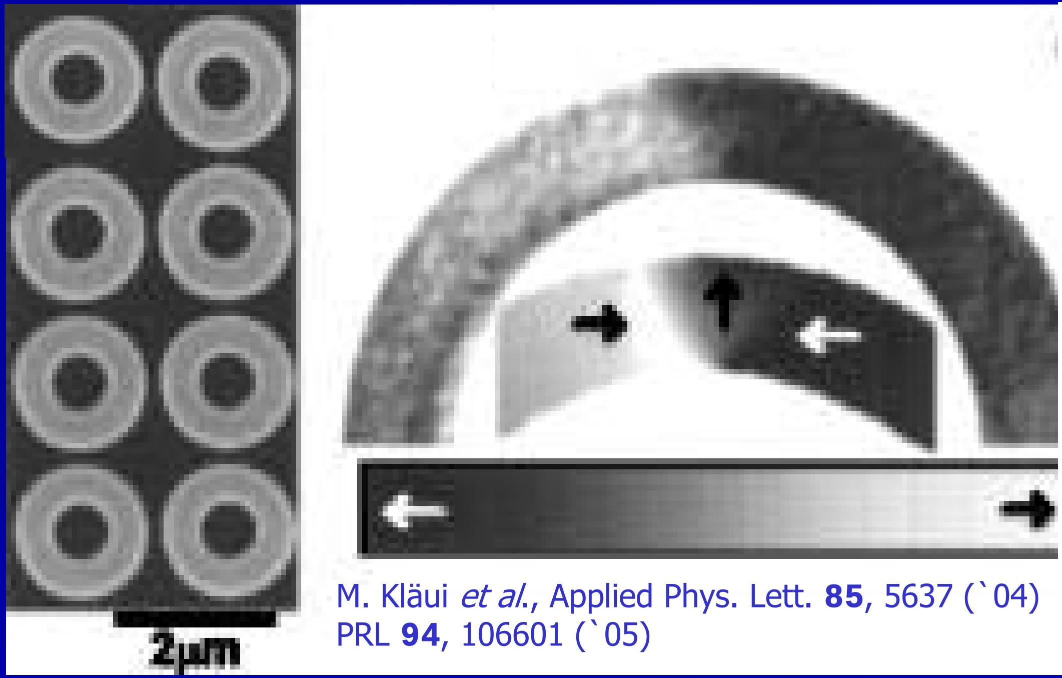

AFM of Si quantum ring array

You *et al.* PRL **98**, 166102 (2007)



***Connected (μm) rings***

W. Rabaud *et al.* , PRL **86** 3124 (2001)



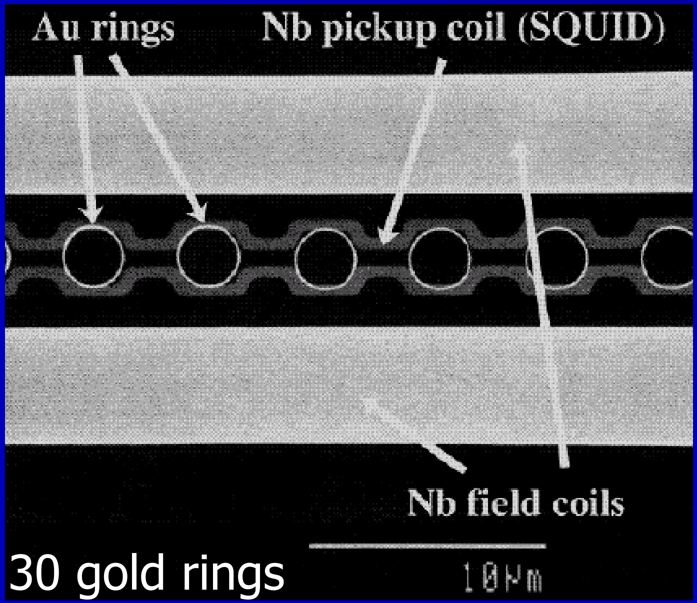

M. Kläui *et al.*, Applied Phys. Lett. **85**, 5637 (`04)
PRL **94**, 106601 (`05)

*polycrystalline Co rings*

outer diameter = 1.65 μm

width = 530 nm

thickness = 34 nm

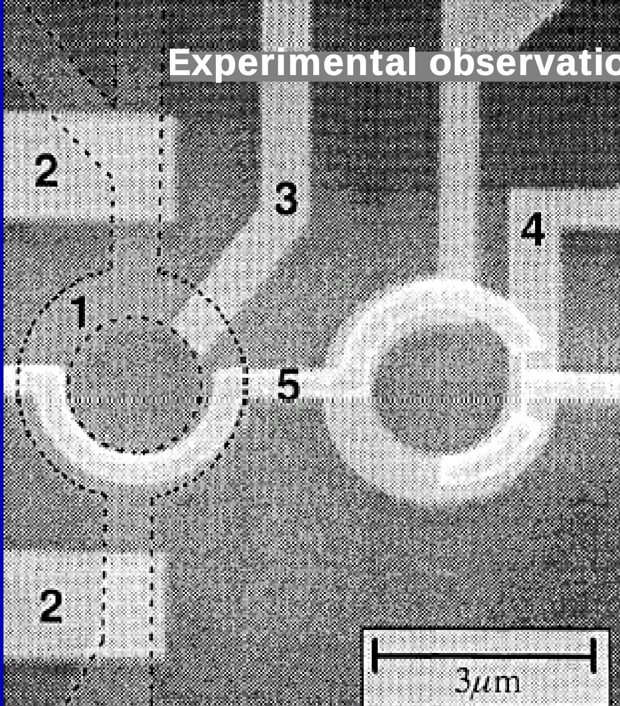



30 gold rings

*L ~ 8μm,* Mohanty, Ann. Phys. ('99)
Ariwala *et al.*, PRL ('01)

**Experimental observation of persistent currents in GaAs-AlGaAs single loop**

D. Mailly, C. Chapelier, and A. Benoit
PRL **70**, 2020 (1993).

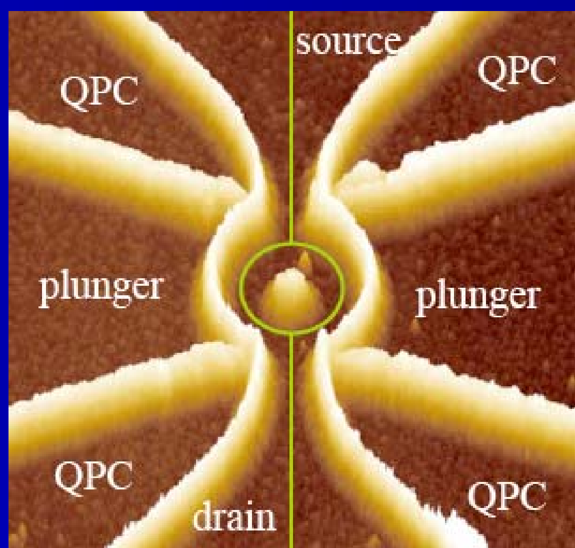


ring etched in GaAs 2DEG (labeled 1)

Aharonov & Bohm
Phys. Rev. **115**,485-491 (1959).
Büttiker, Imry & Landauer
Phys. Lett. **96A**, 365-367 (1983).

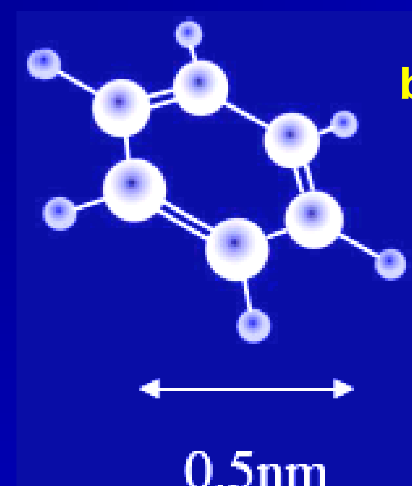
**atomic force microscopy defined quantum ring**

300 nm

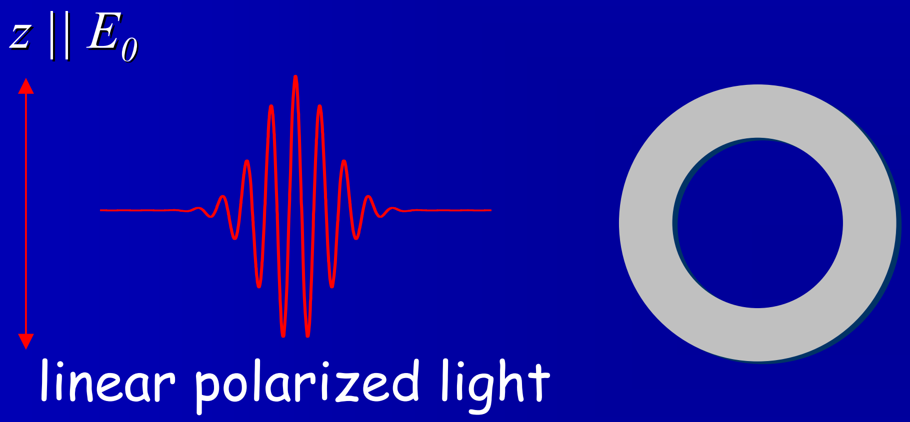



A. Fuhrer *et al.* Nature 413, 822 (2001)

**benzene ring**

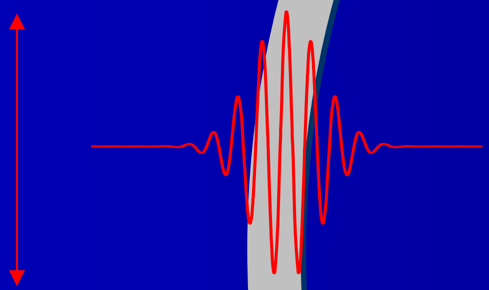


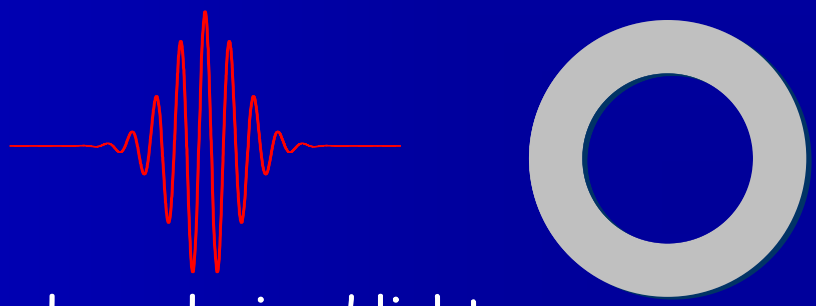

Kekulé Bull. Soc. Chim. Fr. **3**, 98 (1865)
F. Hund Ann. Phys. (Leipzig) **32**, 102 (1938)
F. London J. Phys. France **8**,379 (1937)


$z \parallel E_0$
linear polarized light

$$z(t=0)=0,\ \dot{z}(t=0)=\dot{z}_0,\quad e=1,\ T=\frac{2\pi}{\omega}$$

$$m\ddot{z}=-E_0\sin\omega t\ \Rightarrow \begin{cases}\dot{z}=\dfrac{E_0}{m\omega}\cos\omega t+\dot{z}_0\\[2ex] z=\dfrac{E_0}{m\omega^2}\sin\omega t\end{cases}$$





circular polarized light

E. Räsänen *et al.,* Phys. Rev. Lett. **98**, 157404 (2007)

Katsuyuki Nobusada *et al.*, Phys. Rev. A **75**, 032518 (2007)

Hiroshi Nakatsuji *et al.,* J. Chem. Phys. **126**, 084104 (2007)

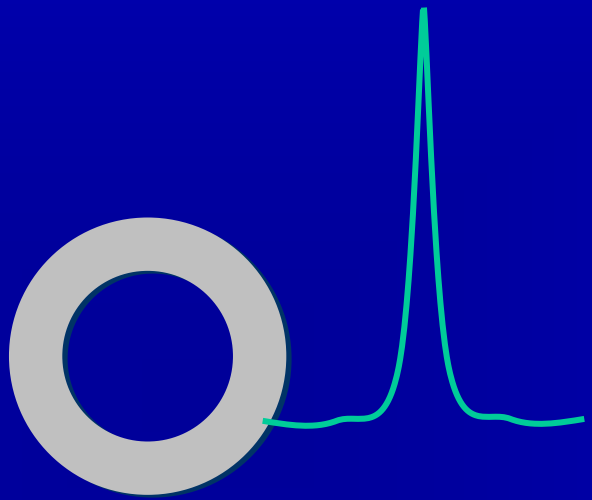
I. Barth *et al.,* J. Am. Chem. Soc. **128**, 7043 (2006);

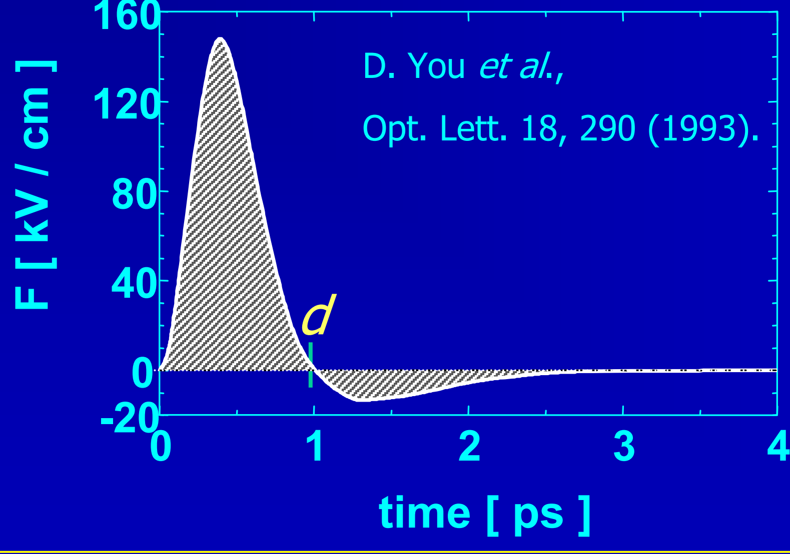
I. Barth *et al.,* Angew. Chem., Int. Ed. **45**, 2962 (2006)

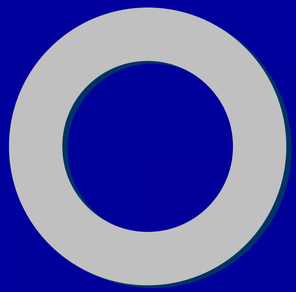
Y. V. Pershin *et al.,* Phys. Rev. B **72**, 125348 (2005)

$$\dot{z}(t=0)=\dot{z}_0$$

$$m\ddot{z}=-\bar{E}\delta(t) \;\Rightarrow\; \begin{cases} \dot{z}=\dfrac{-\bar{E}}{m}+\dot{z}_0 & \text{for } t>0 \\ \dot{z}=\dot{z}_0 & \text{for } t<0 \end{cases}$$







$$H = \frac{1}{2m}\left(\hat{p} - \frac{e}{c}A\right)^2$$

# Persistent currents in rings

stationary single particle states

$$\frac{\hbar^2}{2m^*}\left(-i\partial_s + \frac{2\pi}{L}\frac{\phi}{\phi_0}\right)^2 \psi(s) = E\psi(s), \quad \psi(s+L) = \psi(s)$$

magnetic flux

$$\phi = \int_C A \cdot dr$$

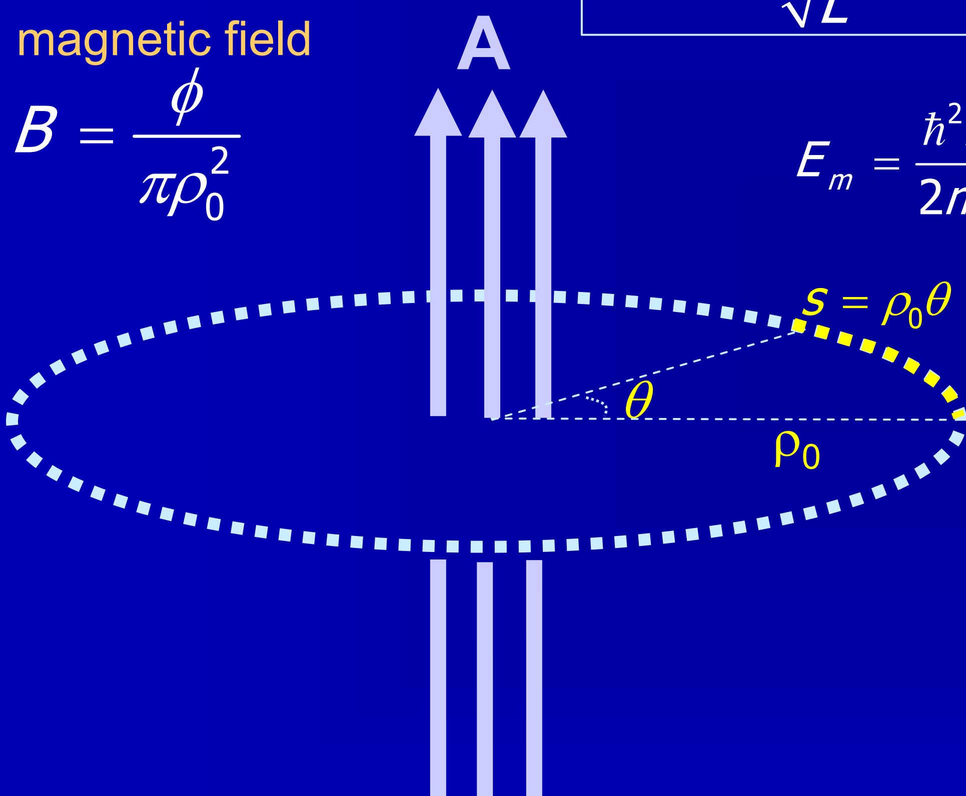
$$\psi(s) = \frac{1}{\sqrt{L}} e^{-i\theta\,\phi/\phi_0} \quad e^{ik_m s}$$

flux quantum $\phi_0 = \dfrac{hc}{e}$

$$\phi_0 \approx 2.07\ 10^{-15}\ Wb$$

magnetic field

$$B = \frac{\phi}{\pi\rho_0^2}$$




Aharonov-Bohm geometry

$$E_m = \frac{\hbar^2 k_m^2}{2m^*}, \quad k_m = \frac{2\pi}{L}\left(m + \frac{\phi}{\phi_0}\right), \quad m = 0, \pm 1, \cdots$$

$$I_m \approx \frac{ev_m}{L}; \quad v_n = \frac{1}{\hbar}\frac{\partial E_m}{\partial k_m} = \frac{\hbar}{m^*}\frac{2\pi}{L}(m + \phi/\phi_0)$$

$$E_m = E_{-m} \Rightarrow I_m + I_{-m} = 0$$

$$E_m \neq E_{-m} \Rightarrow \phi \approx \phi_0 \Rightarrow B \approx \frac{\phi_0}{\pi\rho_0^2}$$

→Benzene ring → B~5000 T

*Mailly et al. 1993* → **I ~ 4 nA**

# Persistent currents in rings

stationary single particle states

magnetic flux

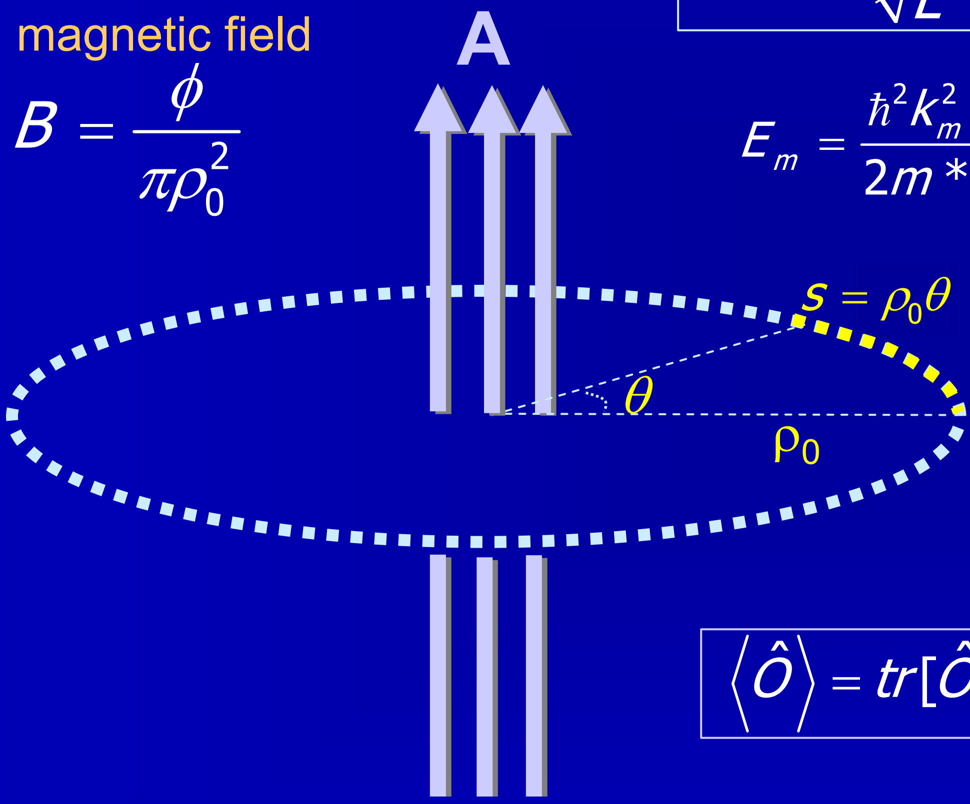
$$\phi = \int A \cdot dr$$

magnetic field

$$B = \frac{\phi}{\pi \rho_0^2}$$

$$\psi(\theta) = \frac{1}{\sqrt{L}} e^{-i\,\theta\,\phi/\phi_0} \quad e^{ik_m s}$$

$$E_m = \frac{\hbar^2 k_m^2}{2m^*}, \quad k_m = \frac{2\pi}{L}\left(m + \frac{\phi}{\phi_0}\right)$$




$$\langle \hat{O} \rangle = tr[\hat{O}\hat{\rho}(t)]$$

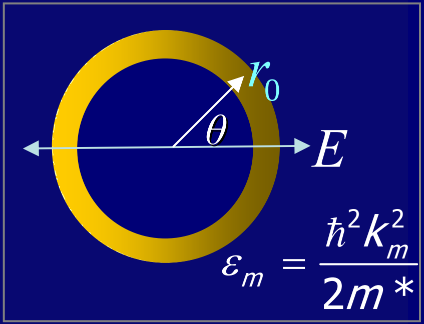
Aharonov-Bohm geometry

# density matrix formalism

Rossi & Kuhn, *Rev. Mod. Phys.* 74, 895 (2002)

Single-electron density matrix $\rho_{m,m'} = \langle m|\hat{\rho}|m'\rangle = \mathrm{Tr}[\hat{\Sigma}\hat{a}^\dagger_m \hat{a}_{m'}] \equiv \langle \hat{a}^\dagger_m \hat{a}_{m'}\rangle$

$$\hat{H}_{\mathrm{tot}} = \hat{H}_0^{\mathrm{carr}} + \hat{H}_0^{\mathrm{phon}} + \hat{H}_{\mathrm{C}} + \hat{H}_{\mathrm{P}} + \hat{V}$$




$$\hat{H}_0^{\mathrm{carr}} = \sum_m \varepsilon_m \hat{a}^\dagger_m \hat{a}_m \qquad \hat{H}_0^{\mathrm{phon}} = \sum_{\vec{q}} \hbar\omega_{\vec{q}} \left( b^\dagger_{\vec{q}} b_{\vec{q}} + \frac{1}{2} \right)$$

$$\hat{H}_{\mathrm{C}} = \frac{1}{2} \sum_{m_1,m_2,m} V_m \; \hat{a}^\dagger_{m_1} \hat{a}^\dagger_{m_2} \hat{a}_{m_2+m} \hat{a}_{m_1-m}$$

– electron-electron interaction

$$\hat{H}_P = \sum_{\vec{q},m,m'} G_{\vec{q}}^{m'} b_{\vec{q}} \, a^\dagger_m a_{m-m'} + \mathrm{h.c.}$$

– electron-phonon interaction

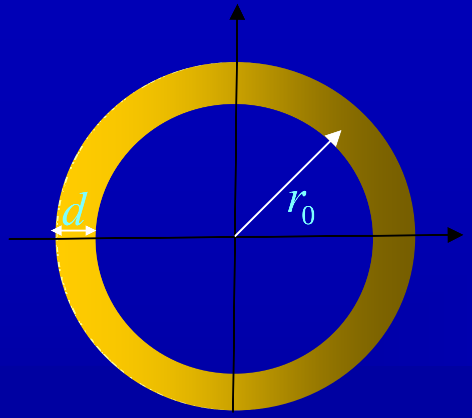
$$\hat{V} = -eE(t) r_0 \sum_{m,m'} \langle m| \cos\theta |m'\rangle a^\dagger_m a_{m'}$$

– interaction with light field

Heisenberg equations of motion ⇒ hierarchy problem ⇒ truncation scheme ⇒ closed system of ODE's

# Energy spectrum

$E_F$ = 4.23 meV,  field = 100 V/cm



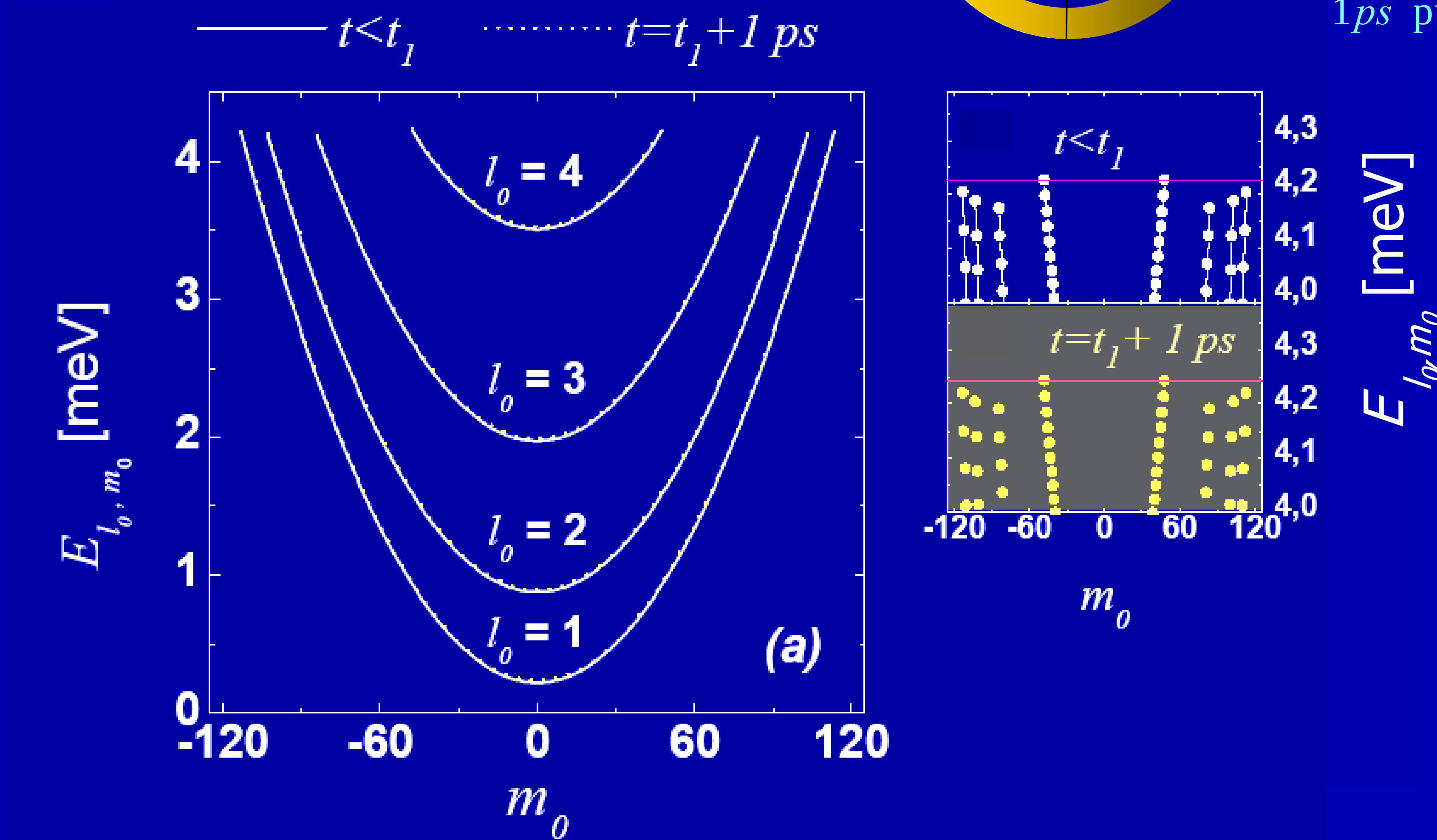

$r_0 = 1.35\ \mu m$

$m^* = 0.067 m_e$

$N = 1400$

$d = 160\ nm$

$1 ps$ pulses



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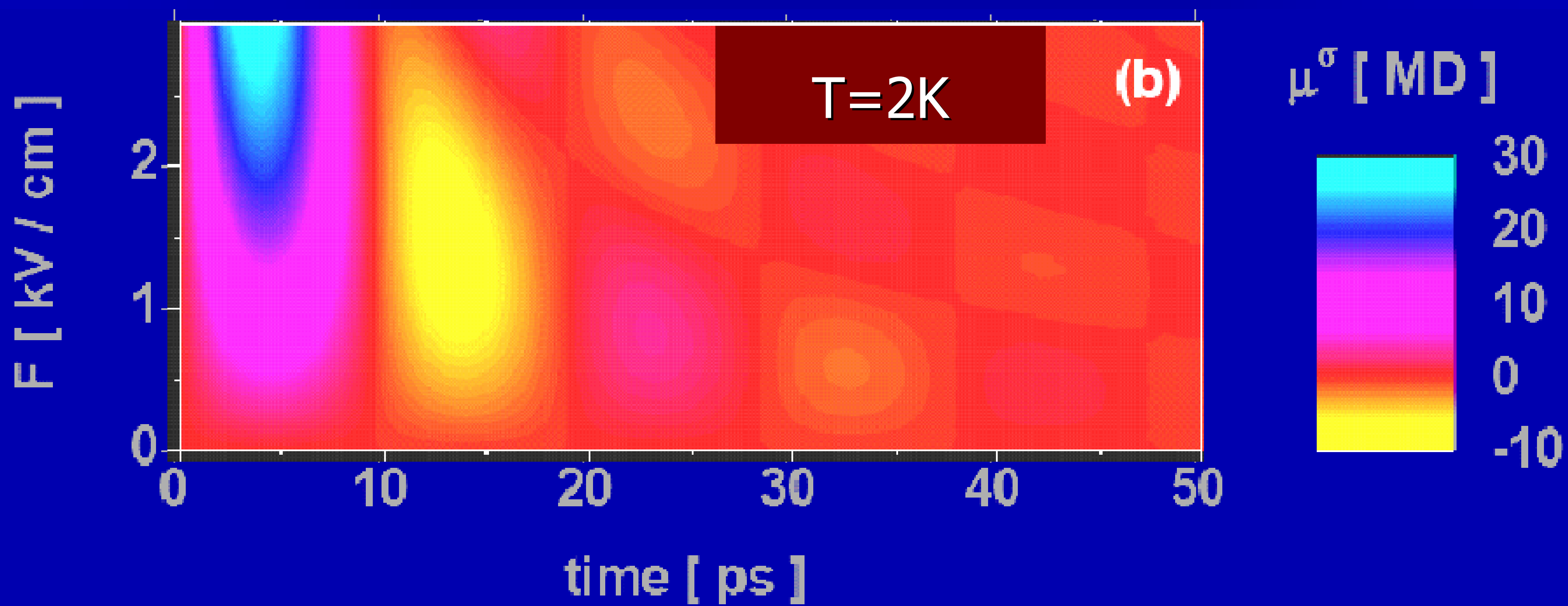
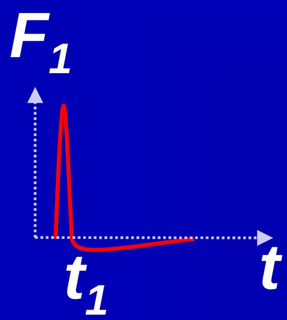
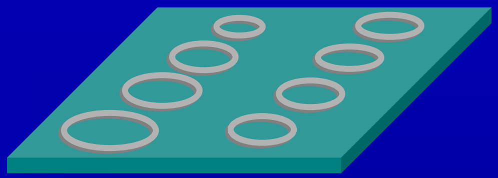
Time dependence of the total induced electric **Dipole moments** in units von $10^6$ D. F is the peak-field amplitude.

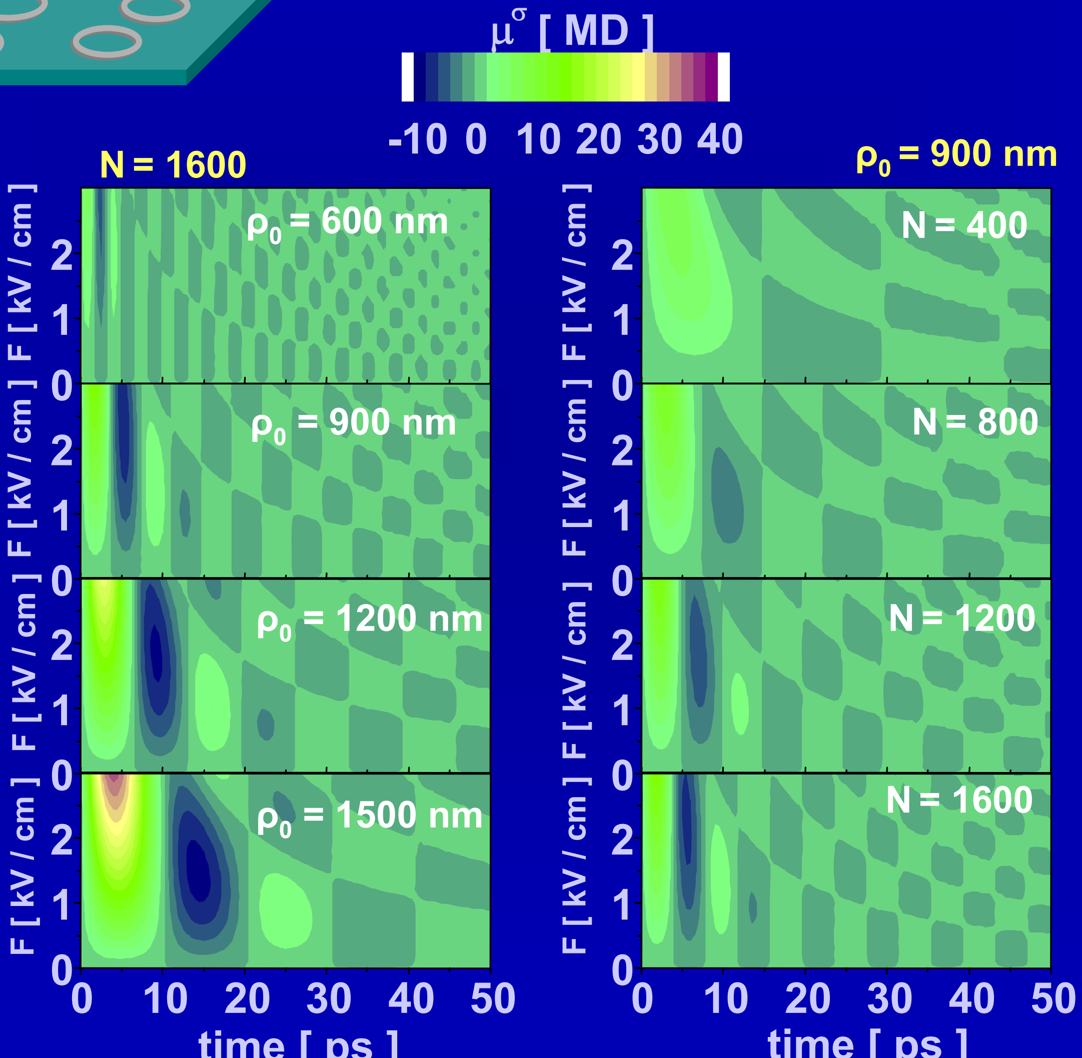



$$\vec{\mu} = tr[e\vec{r}\,\hat{\rho}(t)], \quad \mu_{\parallel} = er_0 \sum_m \mathrm{Re}[\rho_{m+1,m}], \quad \mu_{\perp} = er_0 \sum_m \mathrm{Im}[\rho_{m+1,m}]$$

# Dynamical electric dipole moment of ring structures







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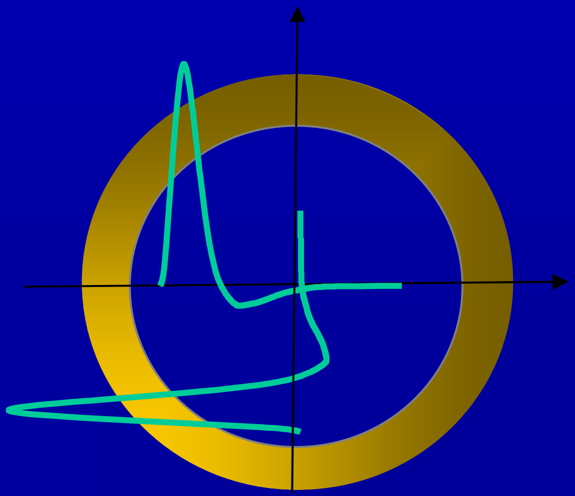
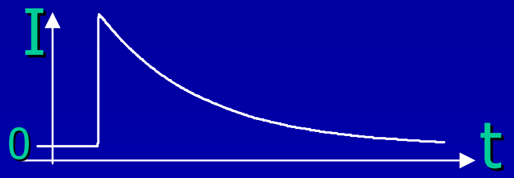
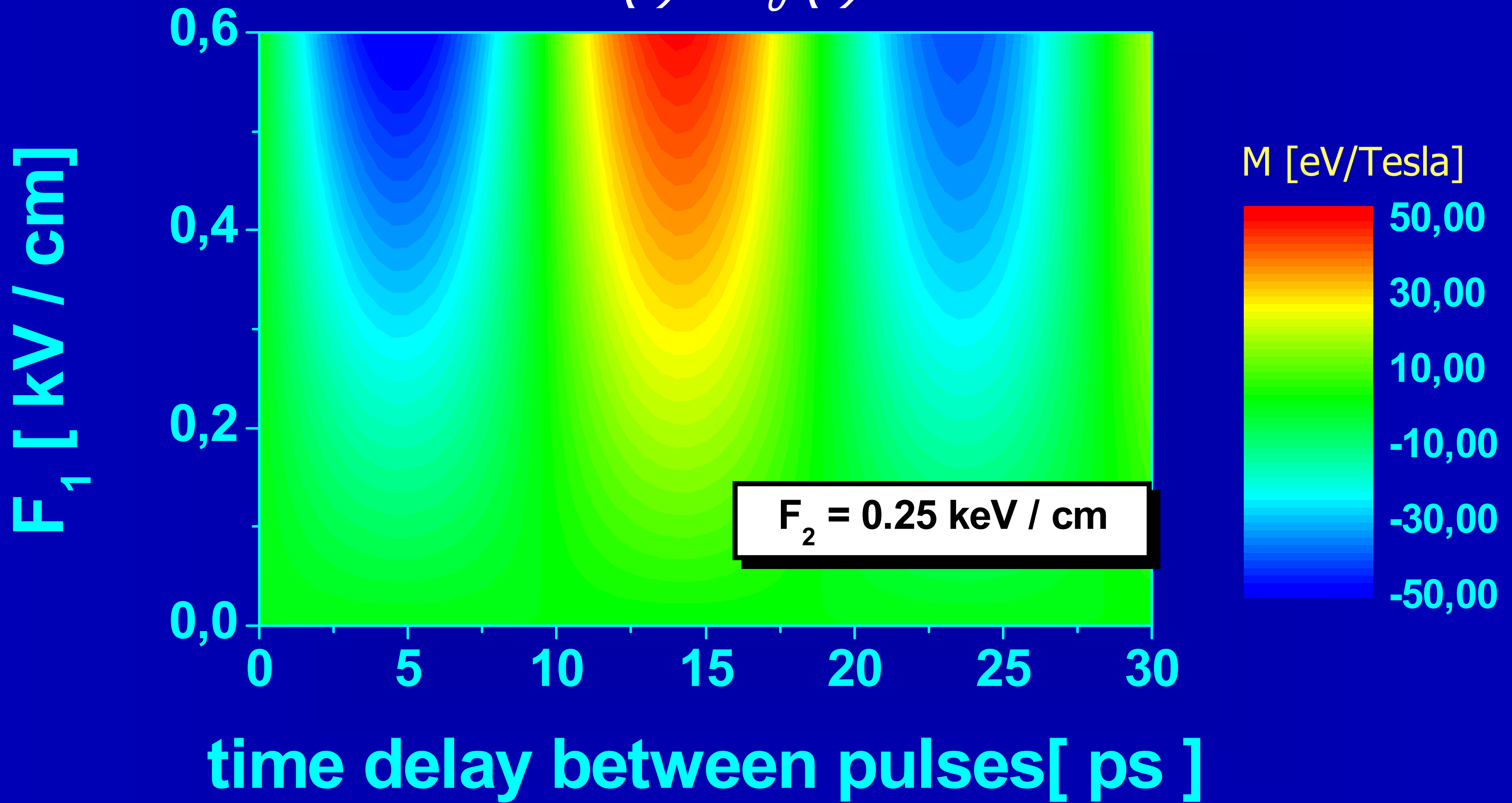
# charge current generation





# induced magnetization

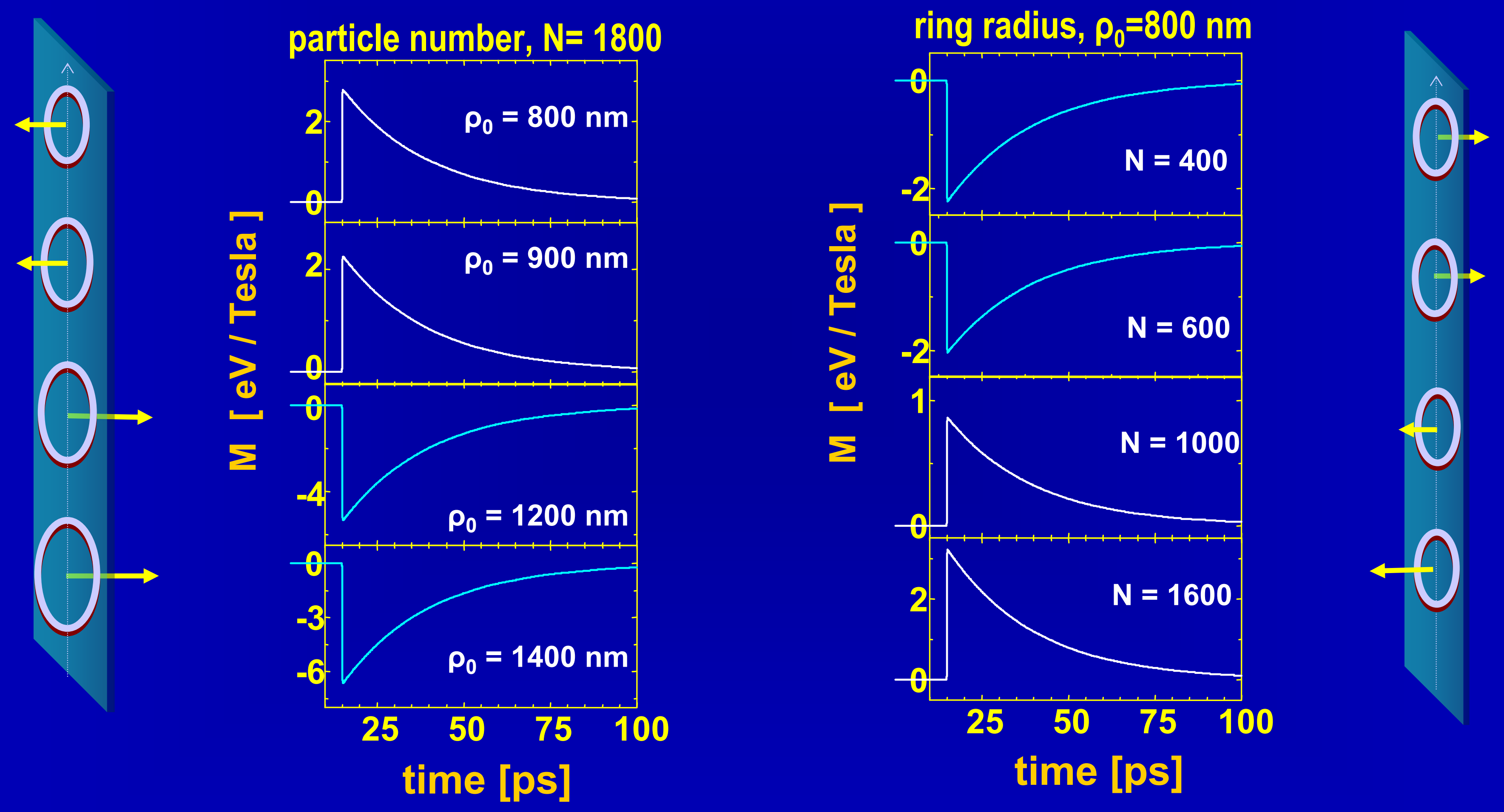
$M(t)=\pi r_0 I(t)$




1 Bohr magneton, $\mu_B = e\hbar/2m_e \sim 7 \cdot 10^{-5}$ eV/T

I=1µA → M~112 eV/Tesla

# Induced magnetization in 1D ring chain




Phys. Rev. Lett. **94**, 166801 (`05)

# Relaxation & revivals

Dipole moment || pulse polarization axis

$$\mu(t) = e r_0 \sum_m \mathrm{Re}[\rho_{m+1,m}]$$

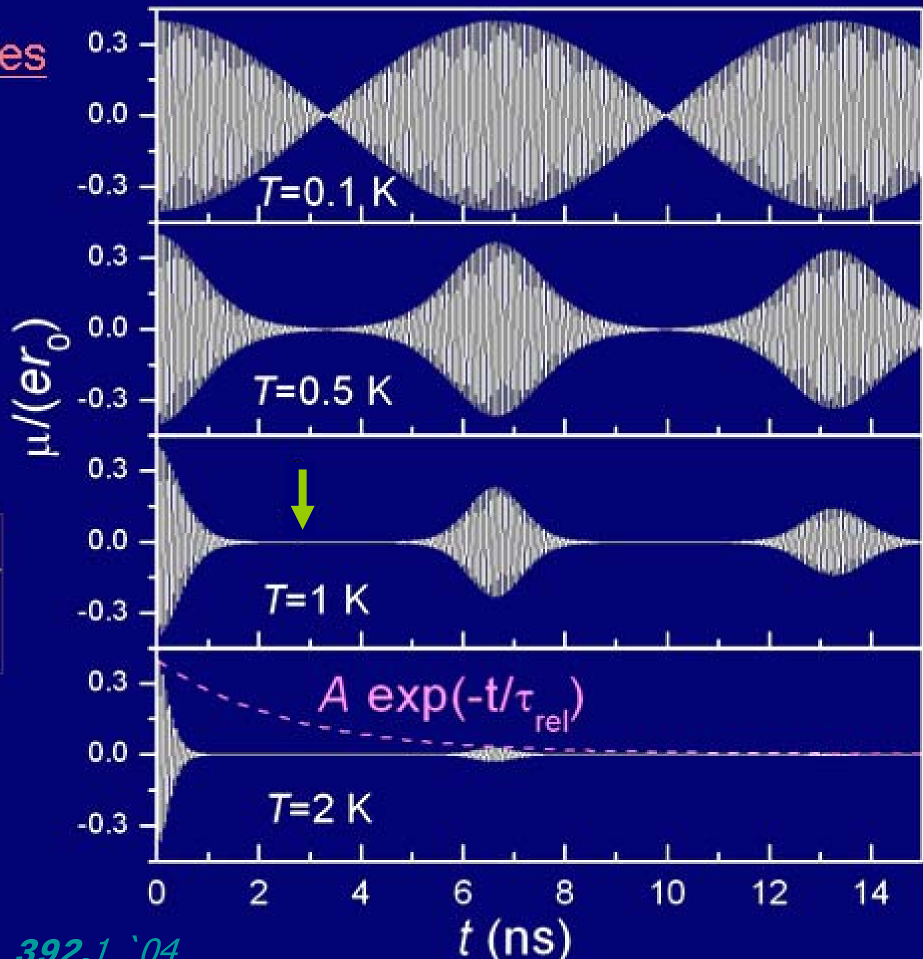
two time scales

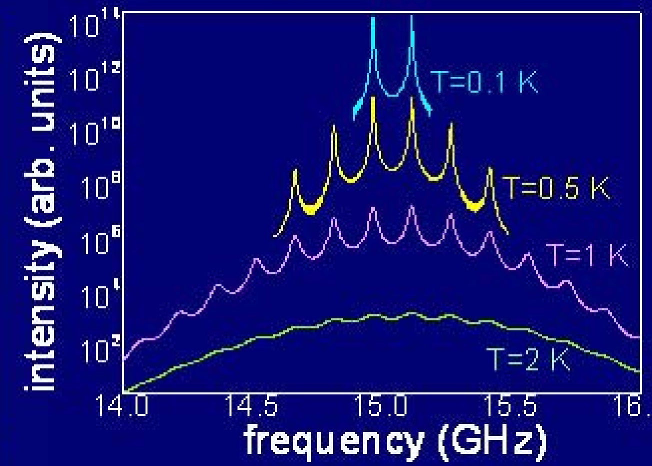
$$T_{cl} = \frac{2\pi\hbar}{\left|\frac{\partial \varepsilon_m}{\partial m}\right|}$$

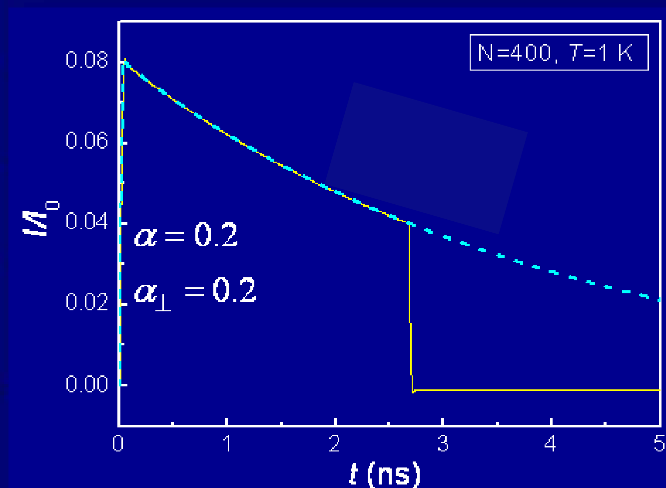
$(T_{cl} \approx \tau_F)$

$$T_{rev} = \frac{4\pi\hbar}{\left|\frac{\partial^2 \varepsilon_m}{\partial m^2}\right|}$$

$m$ at the Fermi level



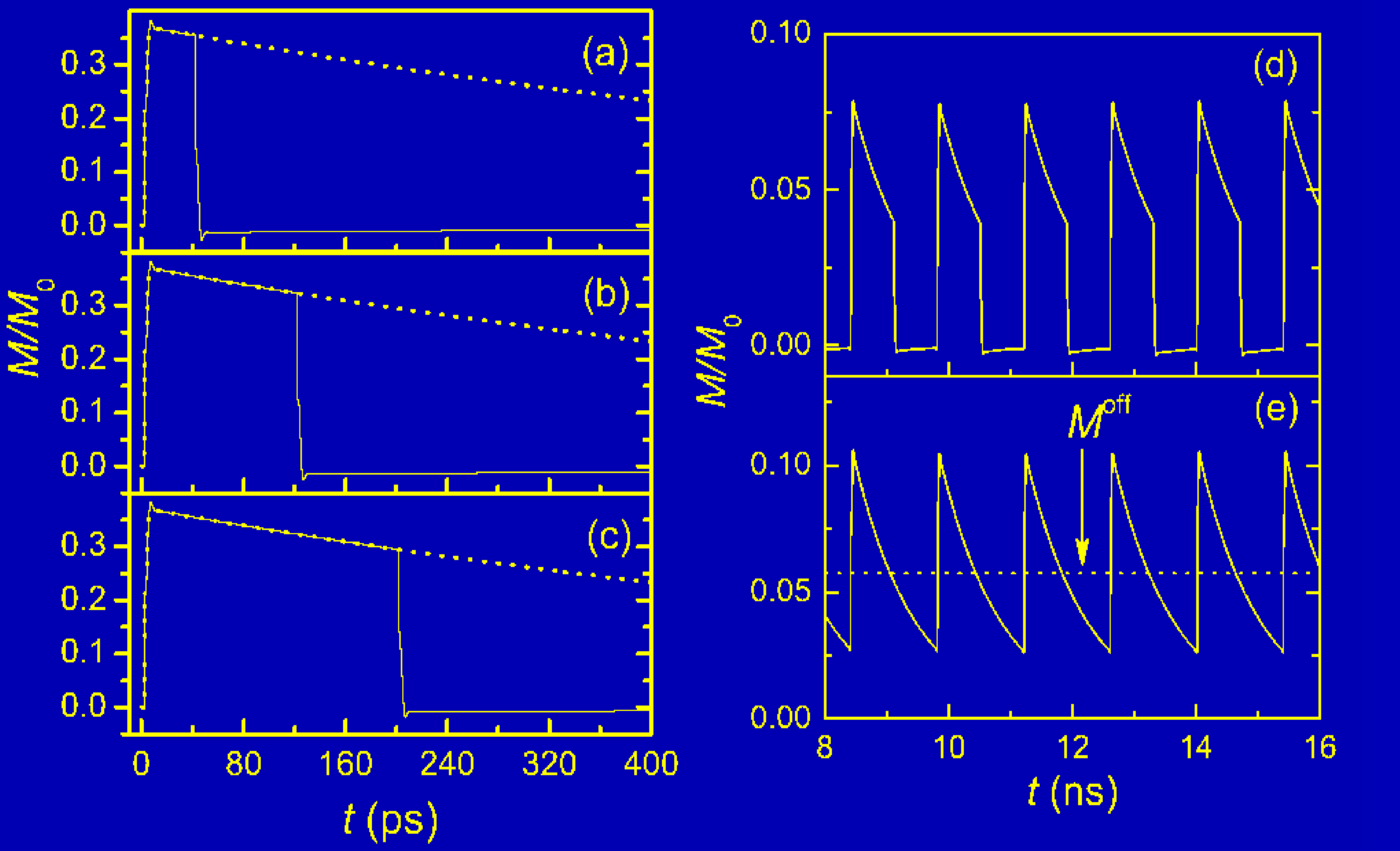




*Robinett, Phys.Rep.* ***392****,1, `04*

GaAs parameters, $N = 400, r_0 = 1.35\ \mu\text{m}, d = 50\ \text{nm}$

PRB **70**, B 74, 161303 **(R)** (2006)
Europhys.Lett. **78,** 57001 (2007)

# Magnetic pulses

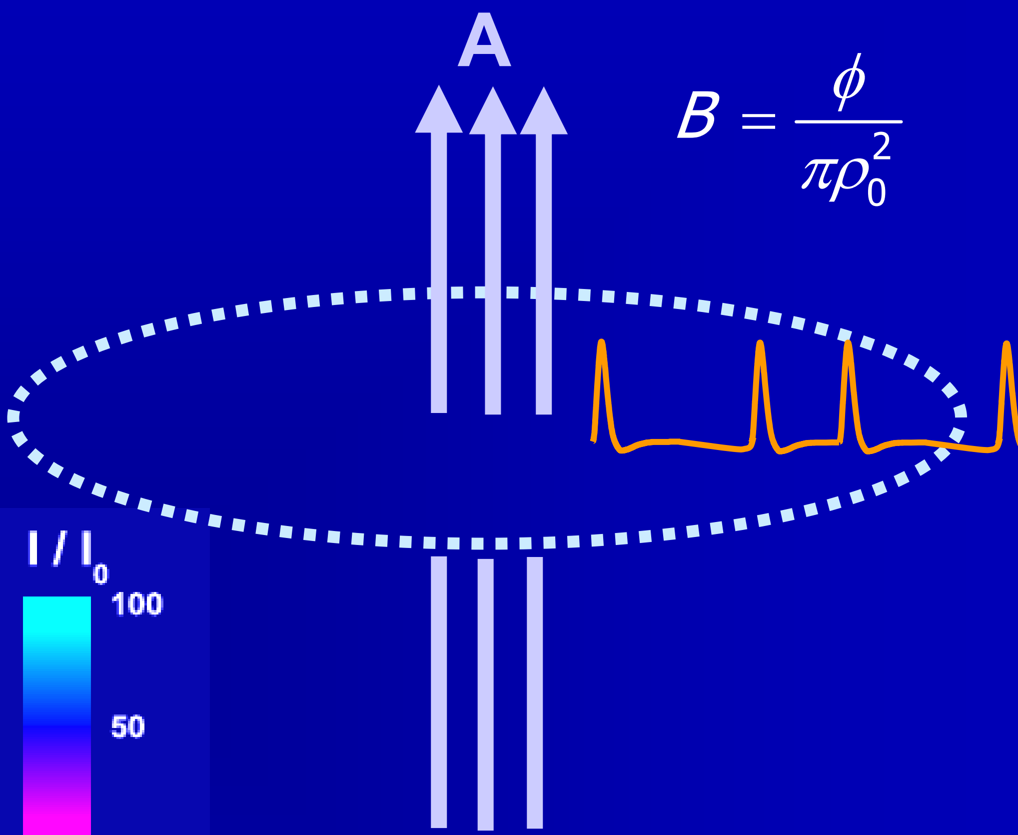



$M_0 = \pi r_0 I_0$

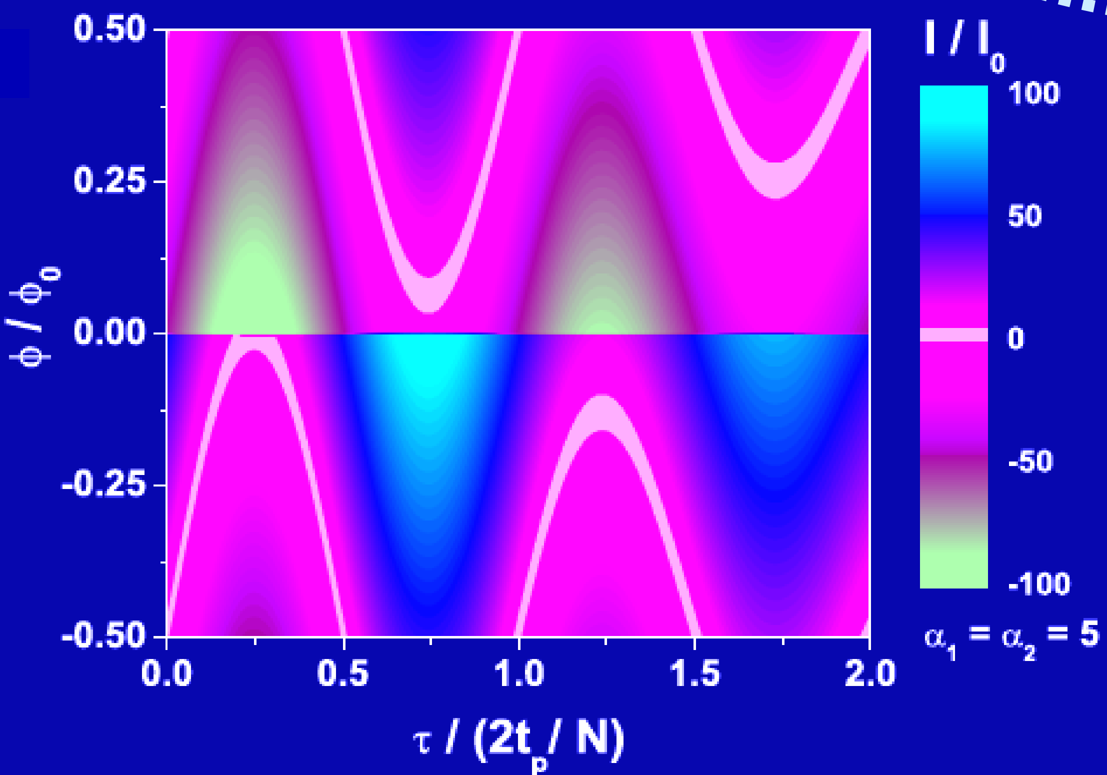
PRB **70**, B 74, 161303 **(R)** (2006)

# Persistent current control

$$I(t) = I_{pers} + \Theta(t-\tau) I_{dyn}(t)$$



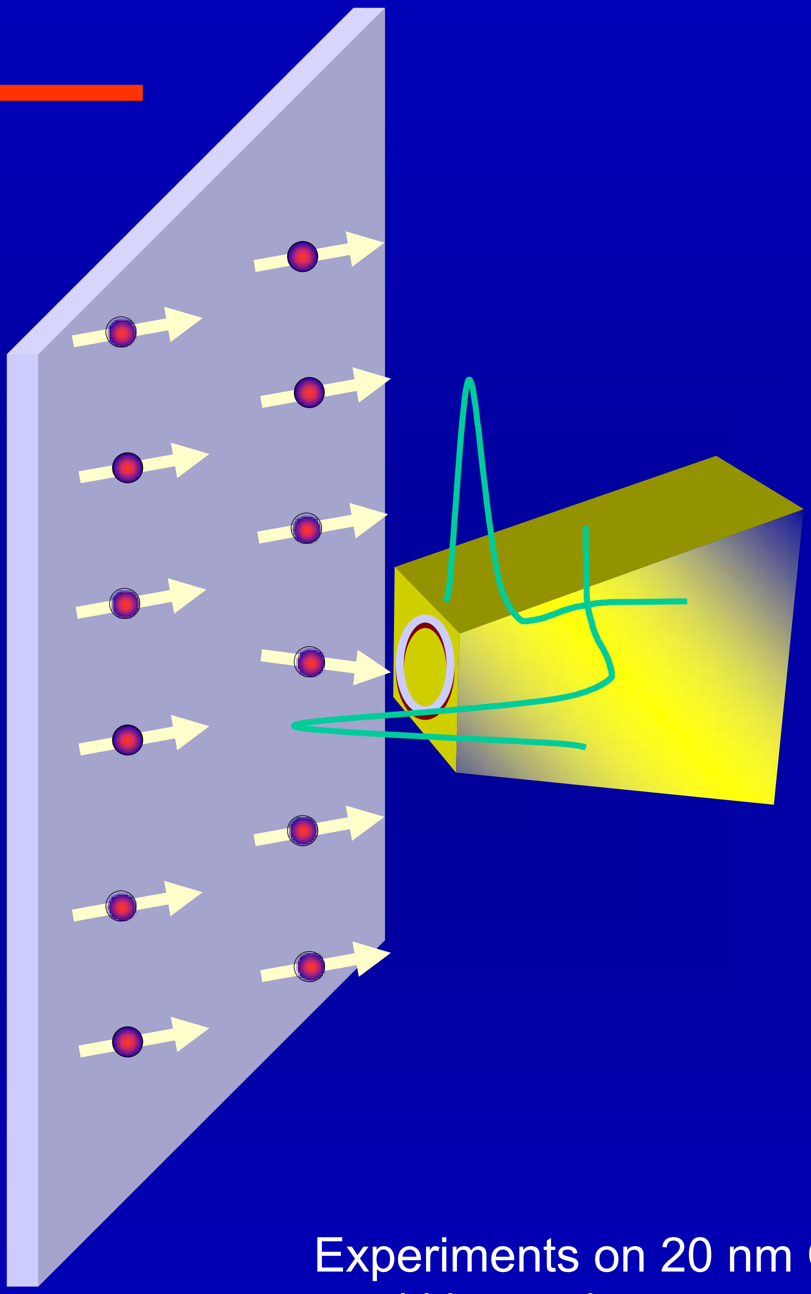

$$B = \frac{\phi}{\pi \rho_0^2}$$




Aharonov-Bohm geometry

Europhys.Lett. **78,** 57001 (2007)

# Applications...

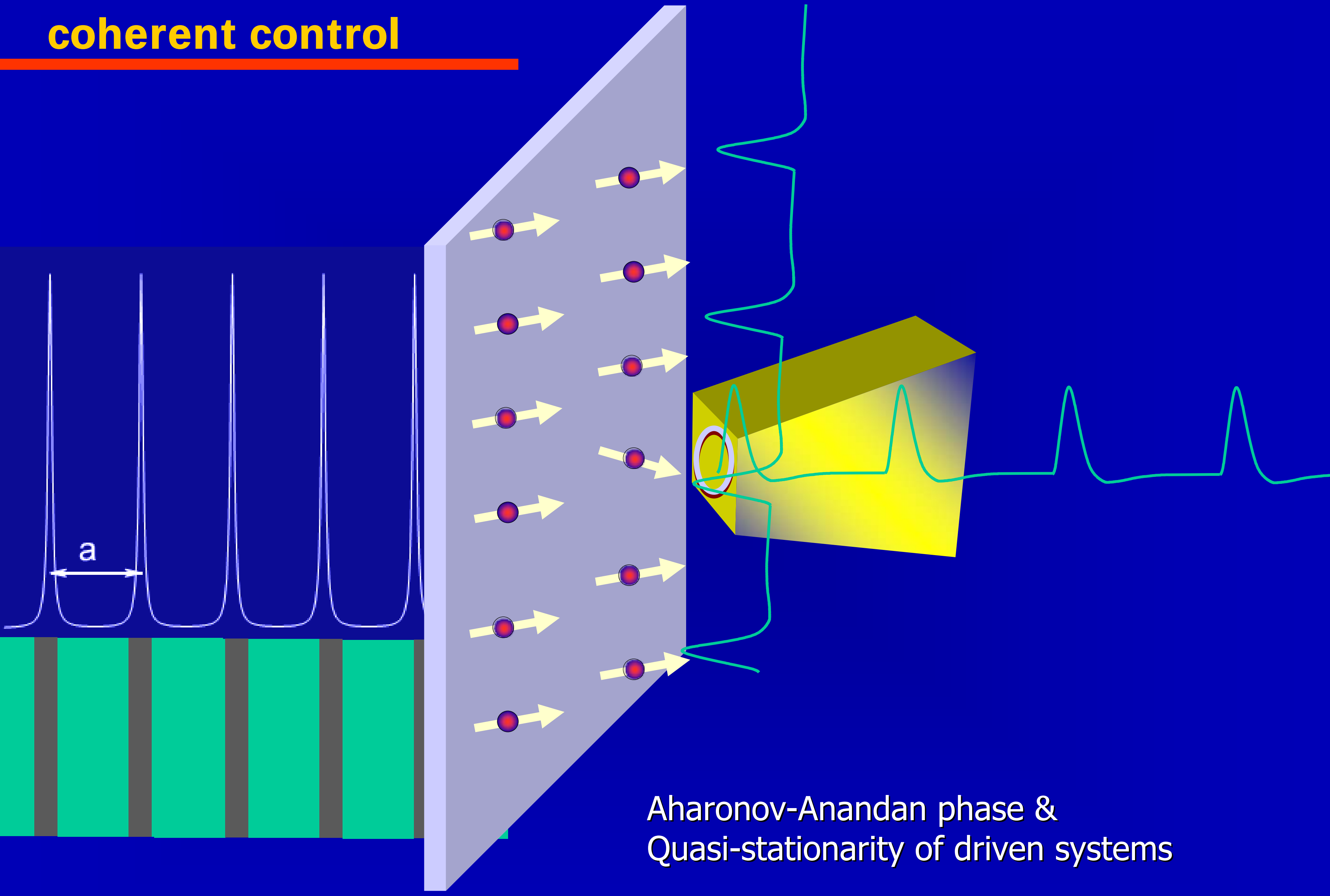


Experiments on 20 nm Co – nanoparticles.

*C. Thirion et al., Nature Mater.* ***2****, 524 (2003)*

*J. Phys.: Cond.Mat.* ***20****, 125226 (2008)*

# coherent control




Aharonov-Anandan phase &
Quasi-stationarity of driven systems

*Phys.Rev. A **73**, 024102 (2006);*
*Europhys. Letters **71**, 705711 (2005)*

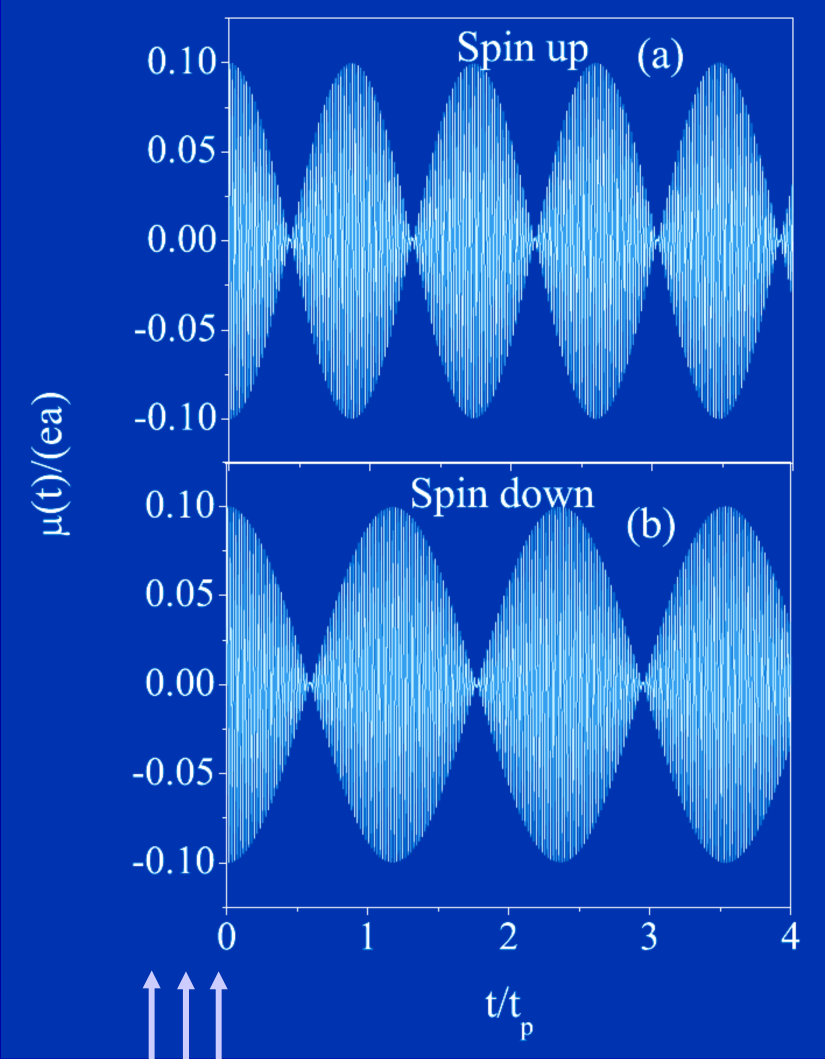
# Work in progress...

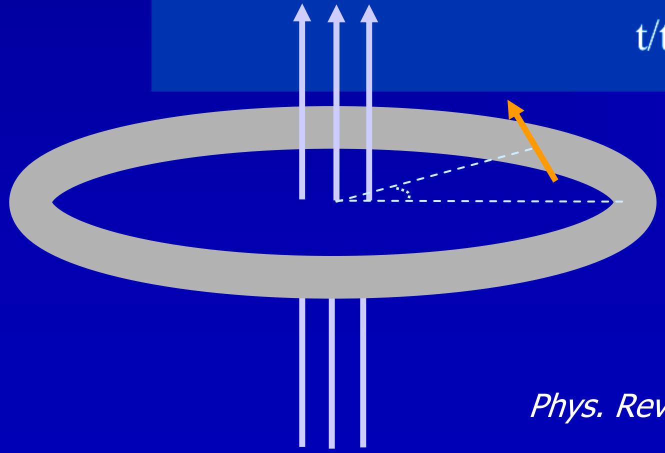
a) pulse-induced spin currents

b) currents in superconducting rings

c) pulse driven transport

d) ab-initio current calculations
in molecular structures.



*Phys. Rev. B at press*

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