Electron Spin Resonance (ESR) with Terahertz Waves and Pulses

Laszlo Mihaly *Stony Brook University*

Past: ESR at 9.5GHz, cavity resonance, field sweep

Present:

Pulsed ESR at fixed frequency (90GHz)

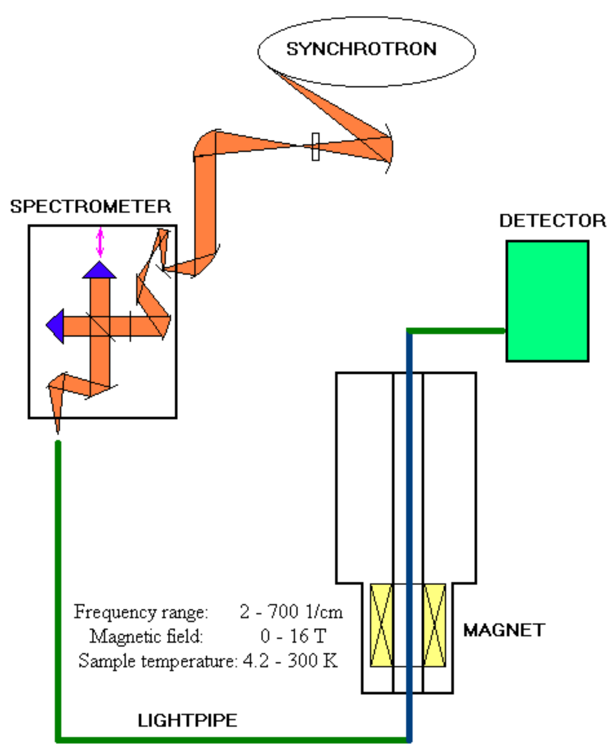
High field ESR, Gunn diode/IR laser, field sweep

ESR with white THz radiation; complete map in terms of frequency ($4cm^{-1}$ - $40cm^{-1}$) and field (0T-16T)

Future:

"Non resonant" ESR with short pulses, H_1 comparable to H_0

Instrument Layout

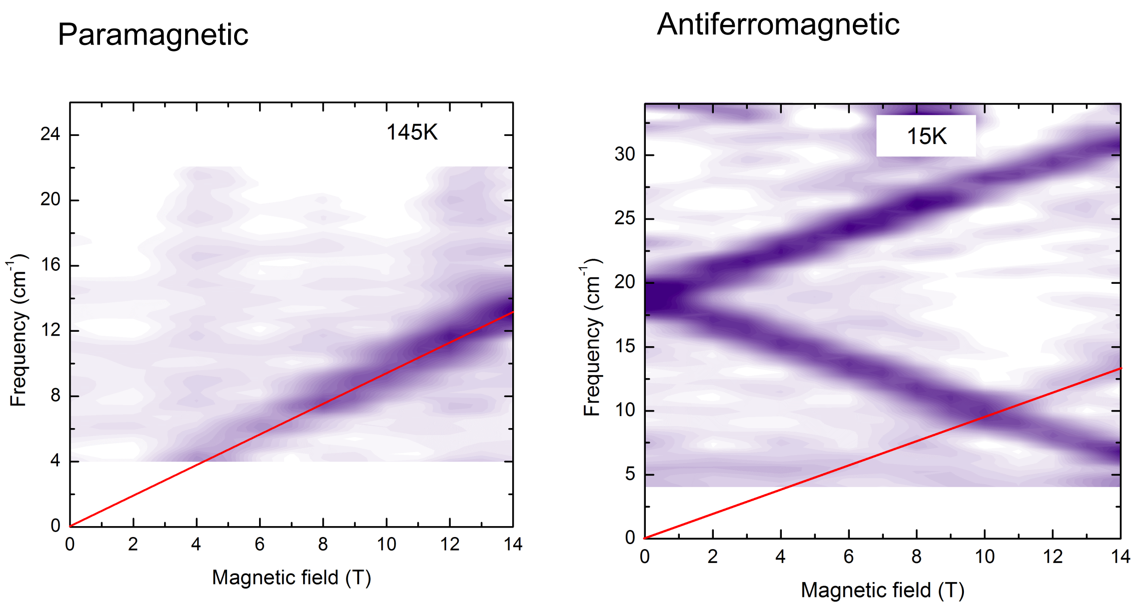


U12IR beamline

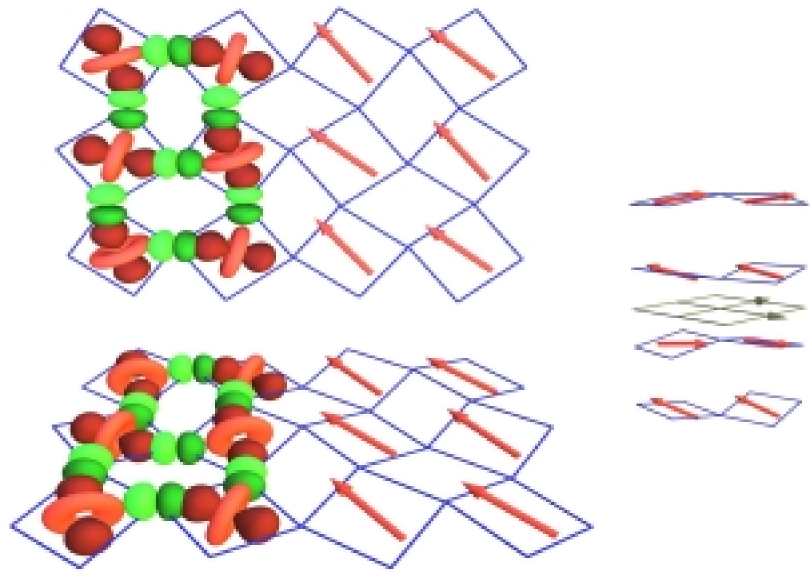
National Synchrotron Light Source

Brookhaven National Laboratory

Electron spin resonance in $LaMnO_3$



L. Mihaly, D. Talbayev, et al. Phys.Rev. B **64** 024414 (2004)



D.Talbayev L. Mihaly J. Zhou, Phys.Rev. Letters, **93** 017202 (2004)

Bloch equations *vs*. single pulse

Resonant excitation

- Magnetization under the influence of external field

  $d\mathbf{M}(t)/dt = \mathbf{M}(t) \times \gamma\mathbf{H}(t)$
- Static and oscillating fields: $\mathbf{H}=\mathbf{H}_0+\mathbf{H}_1\cos(\omega t)$; $H_1 << H_0$
- Select $\mathbf{H}_1$ perp. to $\mathbf{H}_0$ and $\omega_0=\gamma H_0$,
- In rotating frame of reference: $d\mathbf{M}(t)/dt = \mathbf{M}(t) \times \gamma\mathbf{H}_1$
- Spin precesses around $\mathbf{H}_1$ with Rabi frequency $\omega_1=\gamma H_1$

Single pulse excitation: Spin precesses around $\mathbf{H}_0+\mathbf{H}_1$

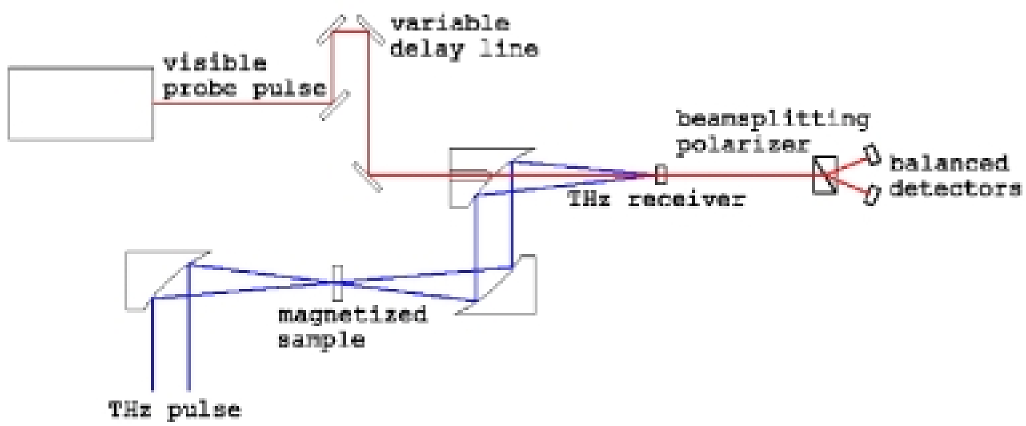
After excitation is over, precession around $\mathbf{H}_0$

Spin excitation with short THz pulses

- Half cycle THz pulse: Assume
  - $1mm^2$ cross section
  - 1 μJ total energy
  - 0.5ps pulse length
  - ~1Tesla magnetic field

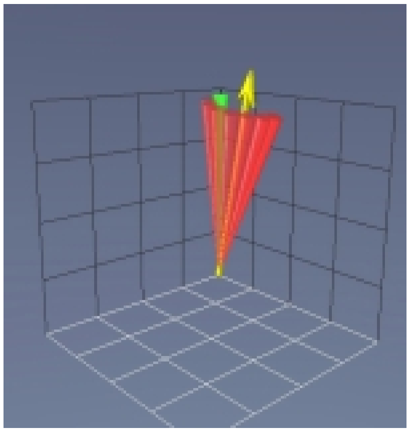
- NMR: pulsed methods making it tremendously powerful
- ESR: short lifetime (T_1) is a problem. Excitation field of about H_1=1Gauss, at f=95GHz is state of art. Yields τ=70ns pulse length for 90^o spin rotation
- $H_1\tau$=const. Optimum pulse length 7ps.
- Even a half cycle pulse can excite spins - NO RESONANCE
- Sample close to metal surface: $H'=2H$, E=0, reduced heating

Schematic layout of single-pulse spin resonance device

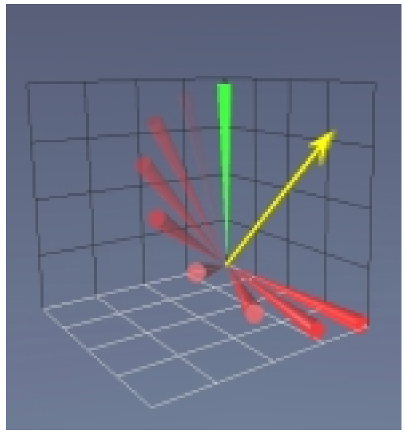


The visible and the THz pulse are generated simultaneously. The THz pulse passes through the sample, the visible probe pulse is delayed and used to detect the oscillating field emitted by the sample.

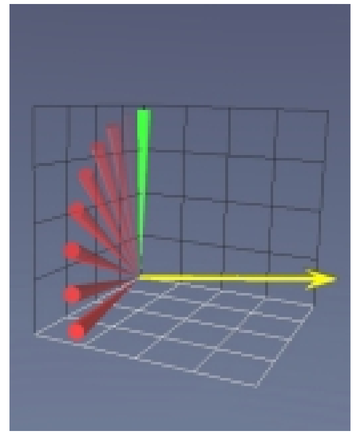
Spin response to half cycle pulse



$H_1 < H_0$



$H_1 = H_0$



$H_1 > H_0$

Free induction decay after half cycle pulse

H_0=1T, Gaussian line of width σ

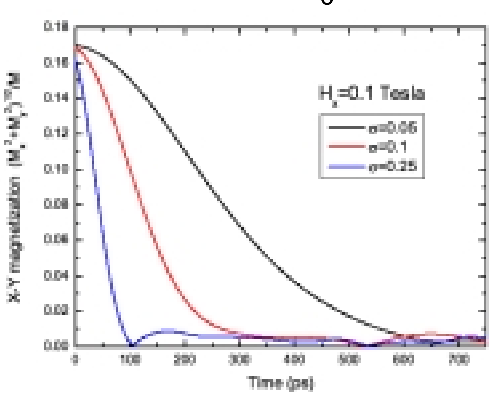


FIG. 5: Envelopes of the FID signal generated by model spin systems in response to a pulse of 1 T field. The resonance linewidth is simulated by the spins' g-factors normally distributed aroun $g = 2$ with the standard deviation σ.

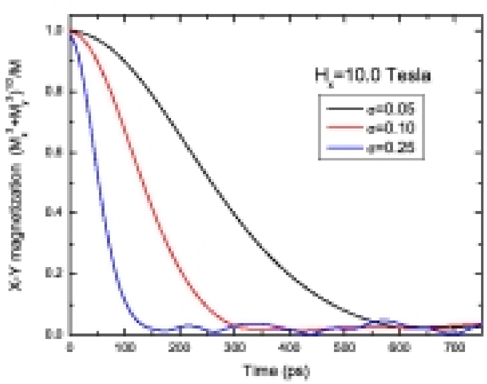


FIG. 3: Envelopes of the FID signal generated by model spin systems in response to a pulse of 10 T field. The resonance linewidth is simulated by the spins' g-factors normally distributed around $g = 2$ with the standard deviation σ.

Spin echo with half cycle pulses

H_0=1T, Gaussian line of width $\sigma = 0.05$

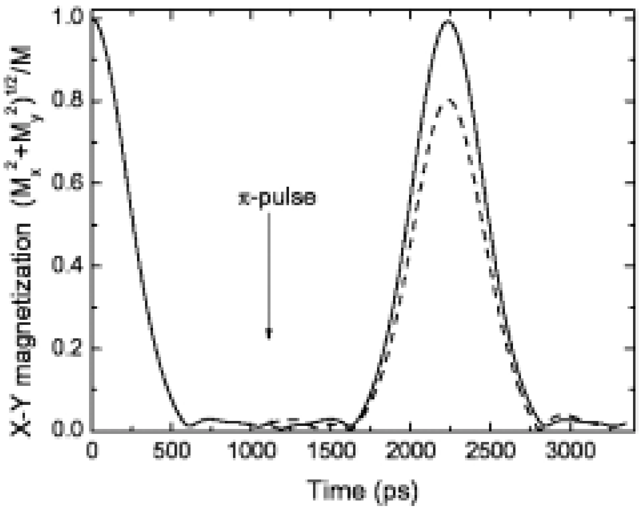


FIG. 8: Free induction decay following a 10 T $\pi/2$-pulse, $\sigma = 0.05$. The application of a π-pulse, $H_1 = 20$ T, $t_p = 1.4$ ps, at time τ generates an echo of the same amplitude at time 2τ. The application of longer π-pulse, $H_1 = 2.5$ T, $t_p = 11.4$ ps, produces an echo with reduced amplitude.

Applications

- Nanoparticles (inhomogeneous broadening)
  - on metal surface
  - inside semiconductors
- Systems with strong spin-spin interaction (homogeneous broadenening)
  - antiferromagnets
  - frustrated magnetic strucures
  - separation of spin and charge excitations
  - high T_c cuprates
- Systems with strong spin lattice interaction (lifetime broadening)
  - spin orbit interaction
  - anisotropy (soft/hard magnets)
  - non-conventional metals

Thanks

András Jánossy (Budapest)
Diyar Talbayev (College of William and Mary)
Larry Carr (BNL, NSLS)
Tom Weinacht (Stony Brook)