Applications of the high-efficiency time-resolved XAFS facility at PNC-CAT

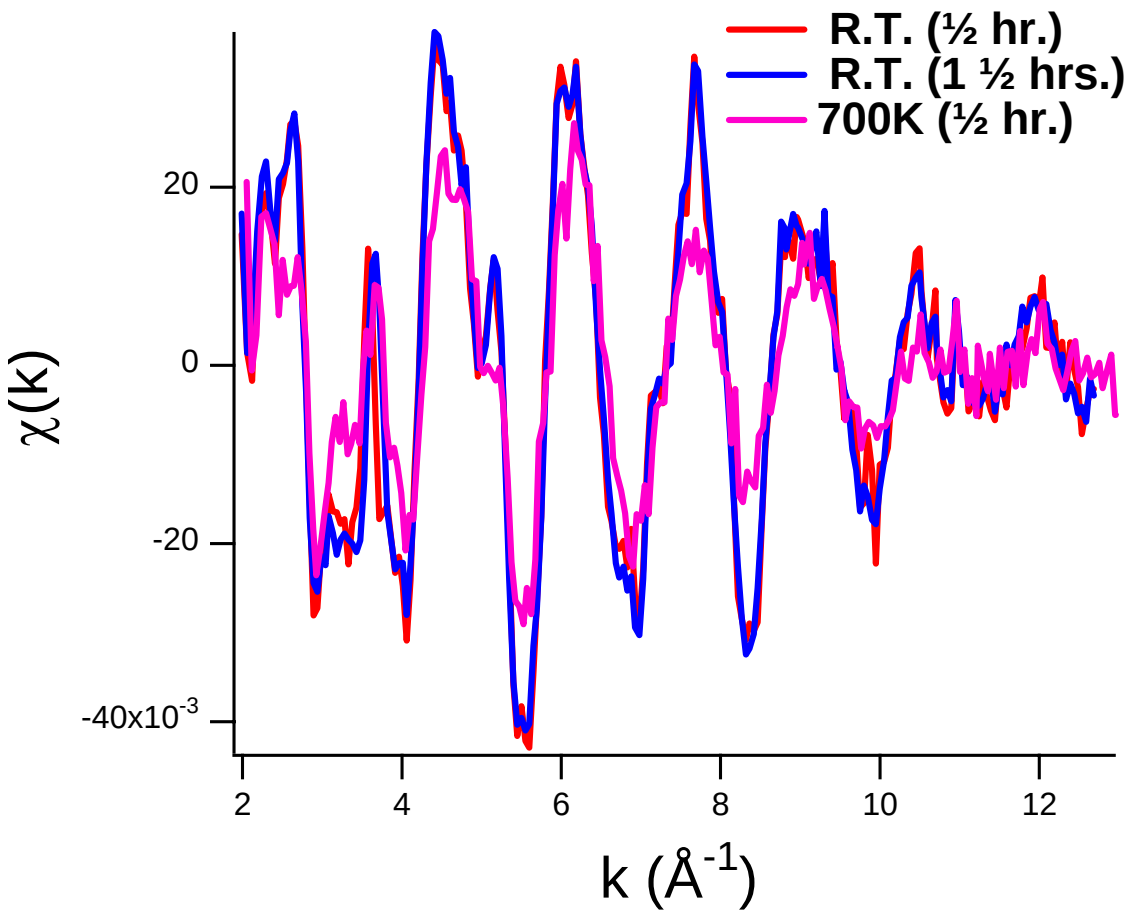
- Description of Facility
- Ongoing and planned experiments
  - Laser heating and melting of Ge
  - Spin dynamics in GdCo alloys
  - Phase change in DVD-RAM materials
- Future improvements

High-efficiency collection of time-resolved XAFS

- High-rep rate (272 kHz) Ti-sapphire laser allows use of all photons from a single bunch
- Micro-focused laser (50 μm) and x-rays (10μm) provide high flux density
- Time resolution limited by bunch length
- XAFS useful for disordered or dilute materials – can concentrate in the interesting site

Data collection time is similar to standard fluorescence XAFS

Ge EXAFS from 200 nm film



Laser heating and melting of Ge

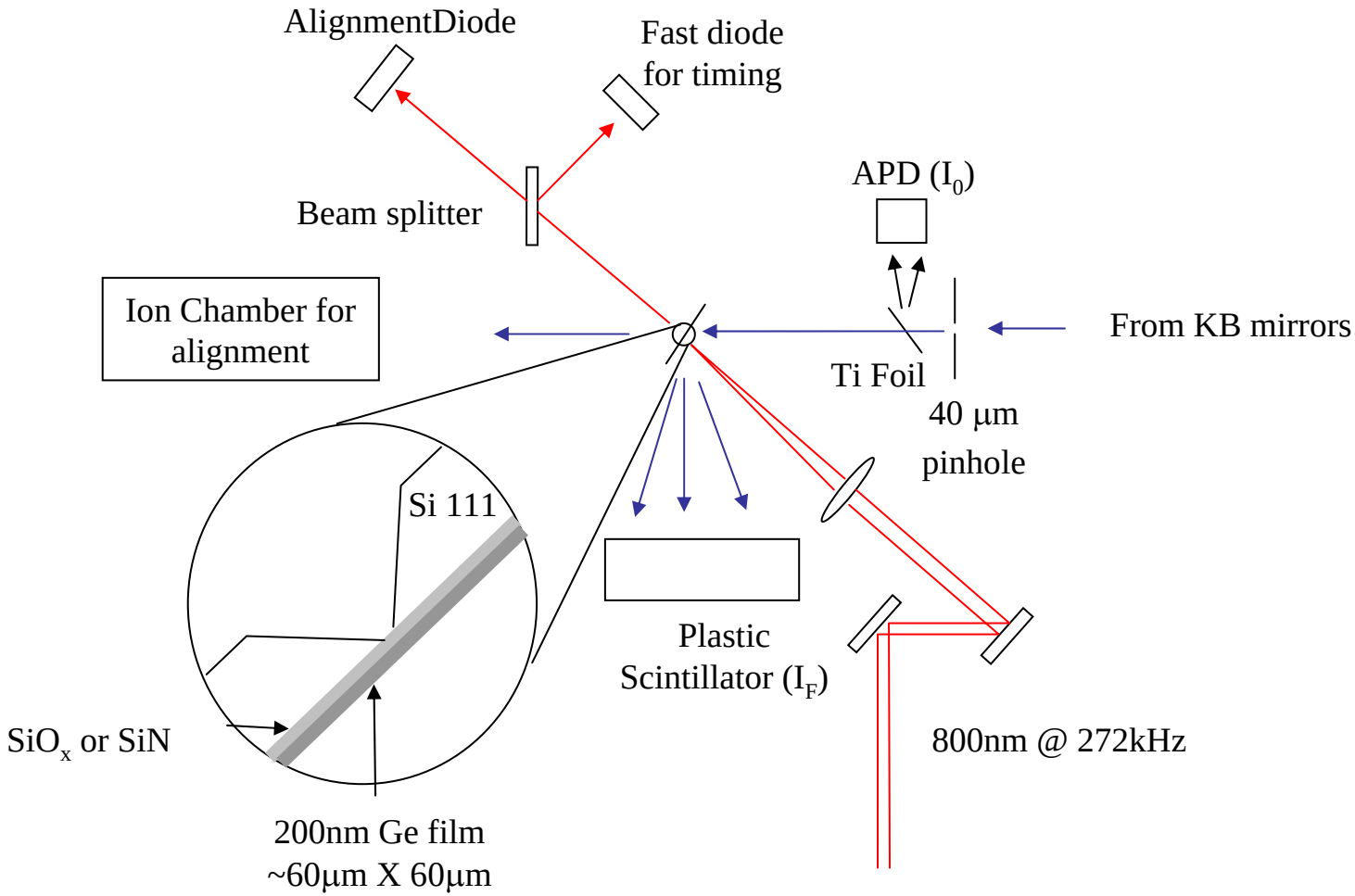
E. A. Stern and D. L. Brewe, Univ. of Washington

- Study the energy transfer between carriers and lattice
  - Laser excites carriers
  - EXAFS sensitive to lattice
- Good starting experiment
- Ultimate goal to look at melting
  - Solid tetrahedral bonding → liquid six-fold coordinated
  - Complex change with possible intermediate states

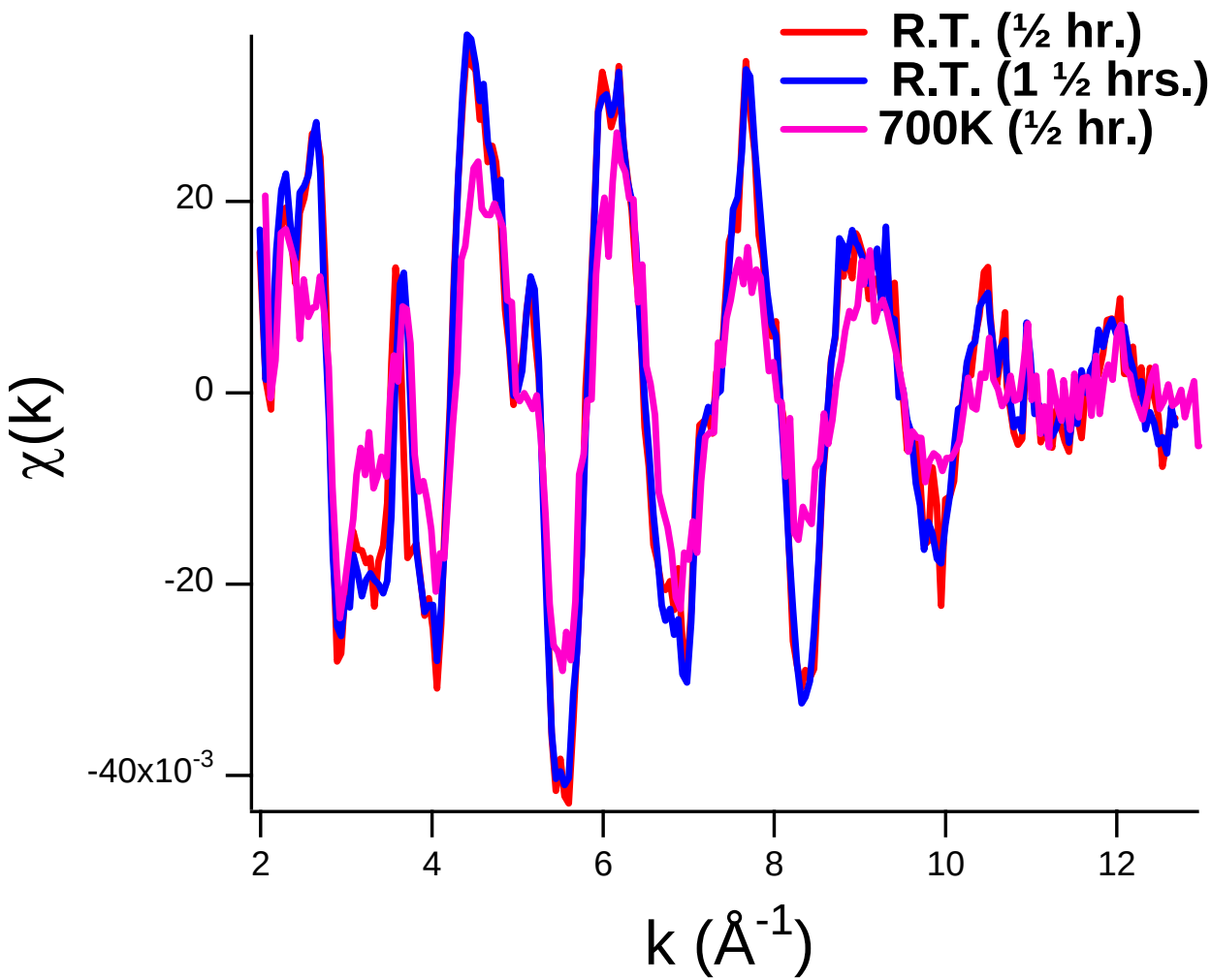
Laser excitation

- Laser at 1.55eV excites electrons into conduction band (indirect gap ~0.67eV, direct gap ~0.8eV)
- Carriers lose energy by phonon emission until states at the bottom of the conduction band are filled (fast, ~1/2 energy?)
- Phonons equilibrate via anharmonic coupling
- Remaining carriers recombine by Auger recombination – "Delayed Auger Heating" (Downer and Shank PRL 1986) (100's psec tail)
- Carriers excited by Auger process lose energy to phonons as above

Experiment Setup

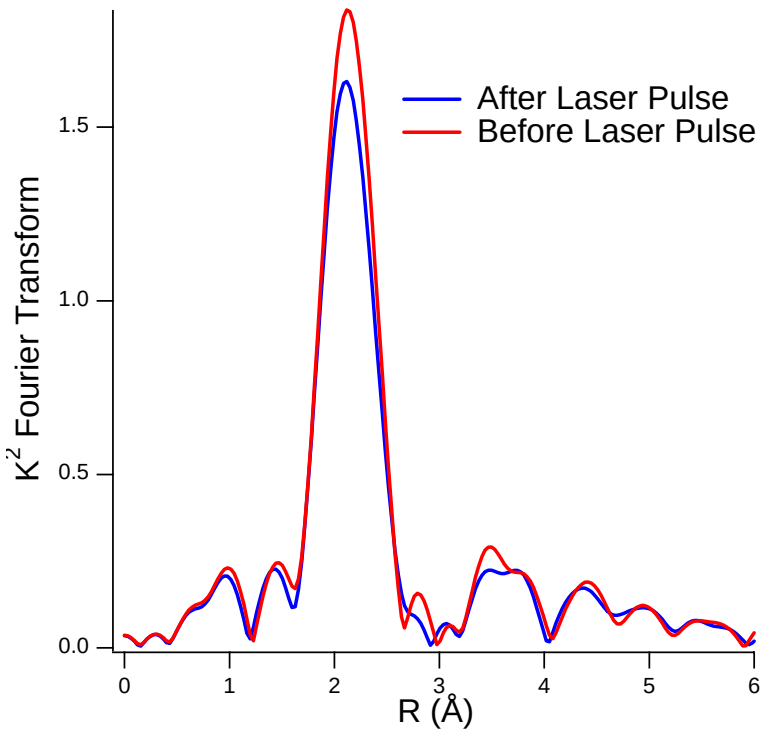


Ge EXAFS

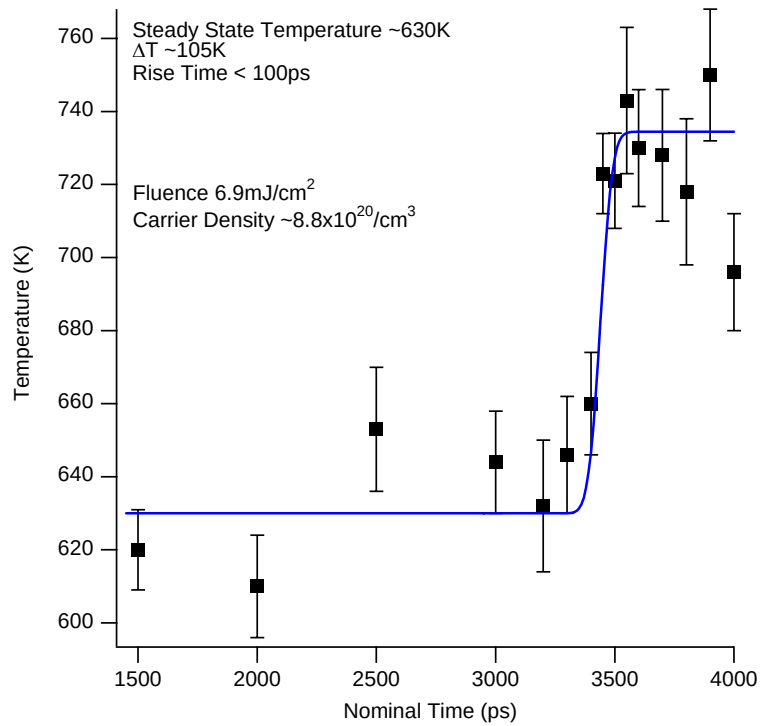


EXAFS Debye-Waller factor gives the lattice temperature in Ge following laser pulse

Fourier transform of EXAFS at different delay times:



Time dependent temperature:



Comparison to Delayed Auger Model

From delayed Auger model expect measurable tail – not observed

However:

- Precision of measurements need to be improved
- Auger recombination may be enhanced:
  - Films are polycrystalline – Most studies are done with single crystals.
  - Significant strains are present, caused by differential thermal contraction of film and substrate after deposition at high temperature.
  - Strained Germanium electronic properties are significantly different from perfect crystal (this property used in latest generation of semiconductor devices).

Spin dynamics in Gd_xCo_{1-x}

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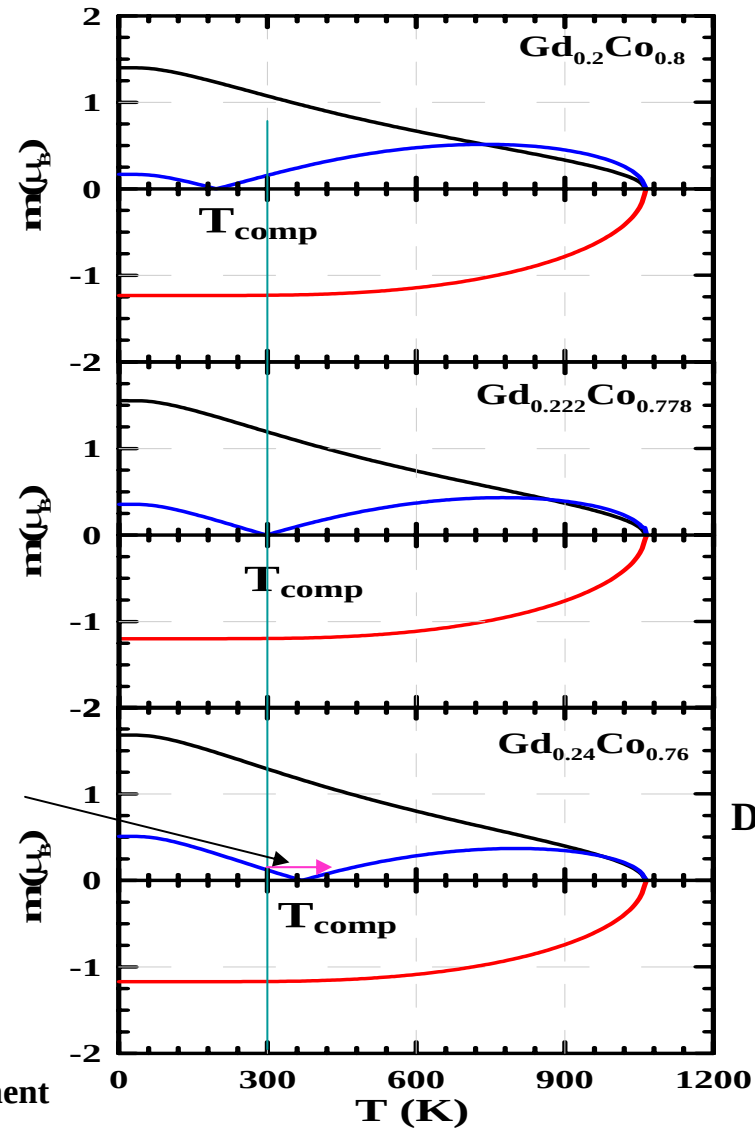
[1] Simon Fraser University, [2] PNC-CAT, APS, [3] University of Washington

- Spin dynamics important in many types of technological devices
- Typically psec spin flip times
- Ferrimagnetic materials near the compensation temperature can have much longer times
- XMCD can separate contributions from different components in compound

Amorphous Gd_xCo_{1-x} Ferrimagnetic Alloy Films



Increased Co Concentration
T_{comp} Decreased

m_{Co}
m_{Gd}
m_{Tot}

Laser heating

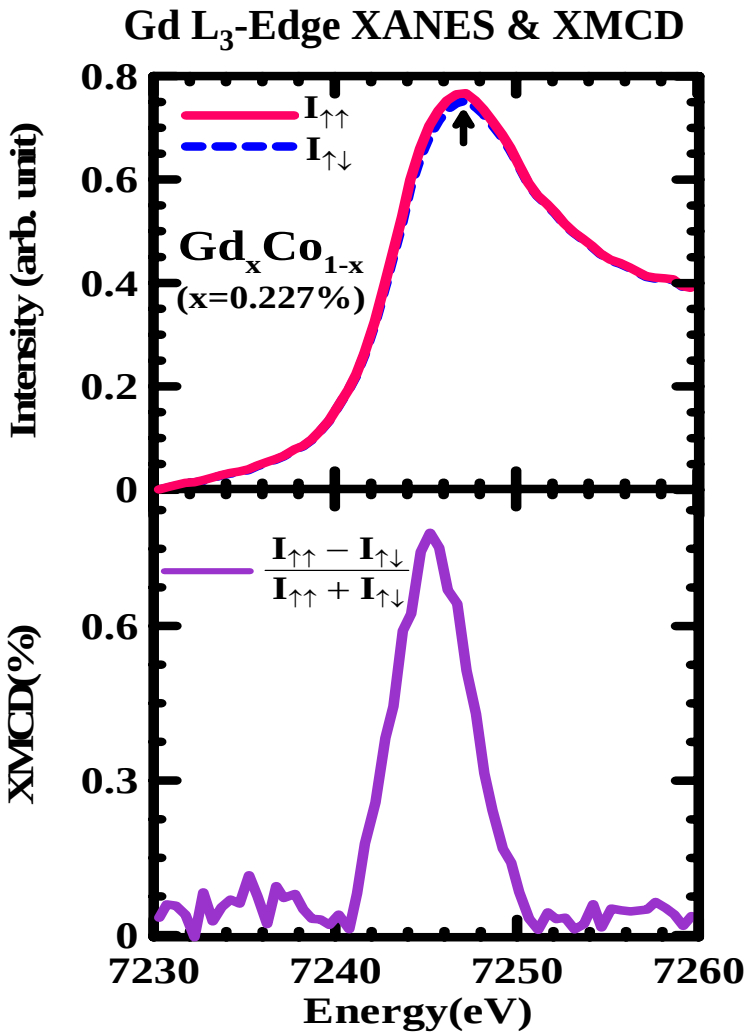
Decreased Co Concentration
T_{comp} Increased

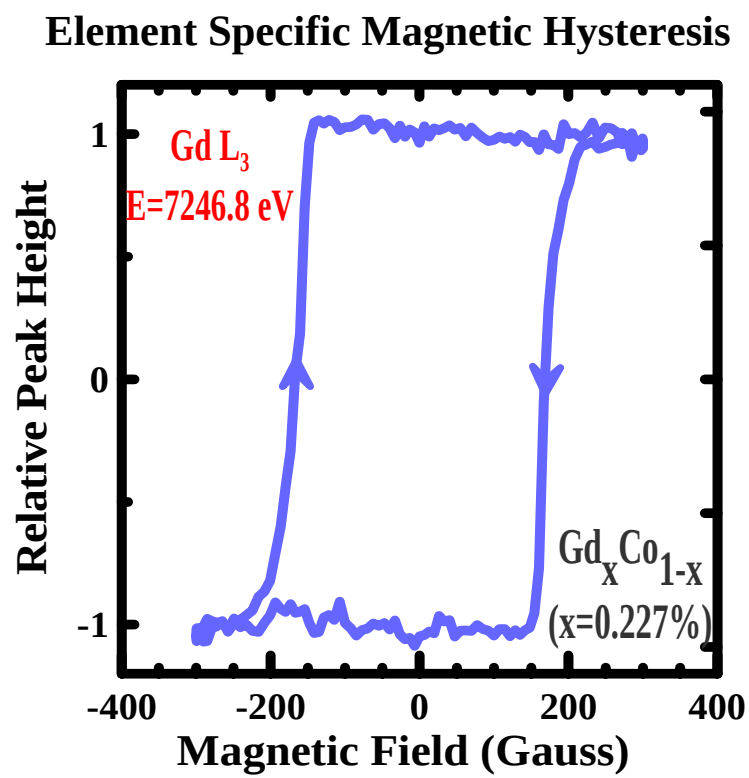
$H_{applied}$

Sublattices Alignment Below T_{comp}

Sublattices Alignment Above T_{comp}

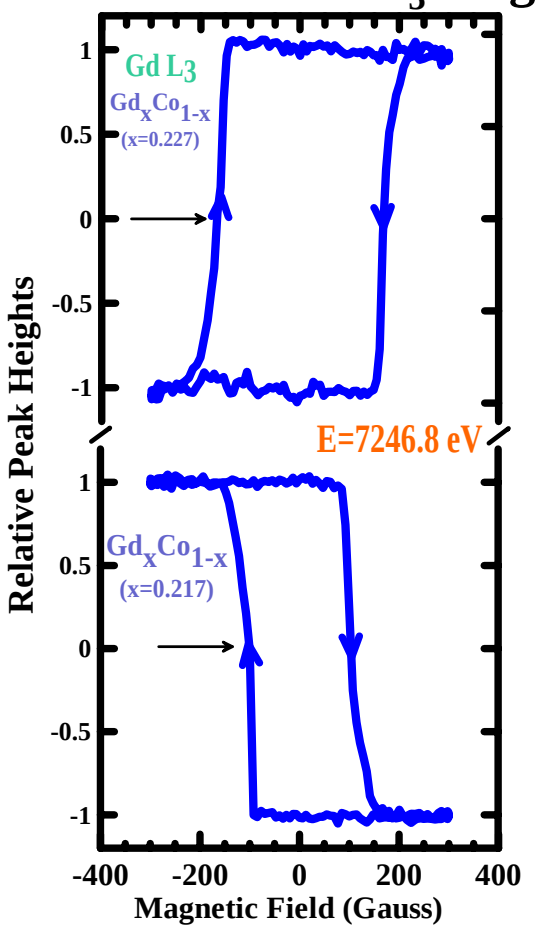
Element Specific Magnetic Hysteresis of Gd in Gd_xCo_{1-x} Ferrimagnetic Amorphous Alloy Films





ESMH of CO and Gd for Gd_xCo_{1-x} (x=0.227 and 0.217) Ferrimagnetic Amorphous Alloy Films

ESMH at Gd L_3-Edge

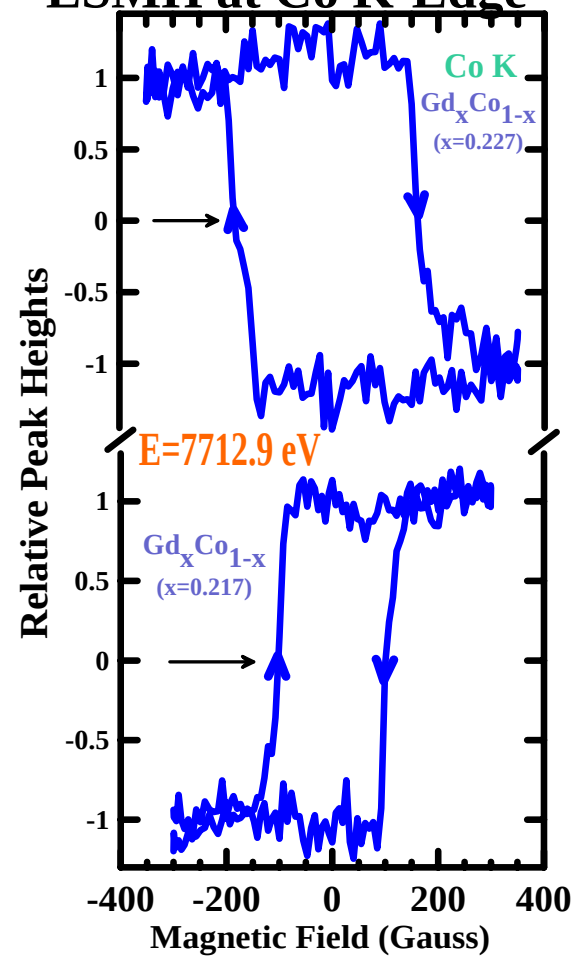


$T_{comp} > T_R$

$T_{comp} < T_R$

$H_c^{Gd} = H_c^{Co}$

ESMH at Co K-Edge

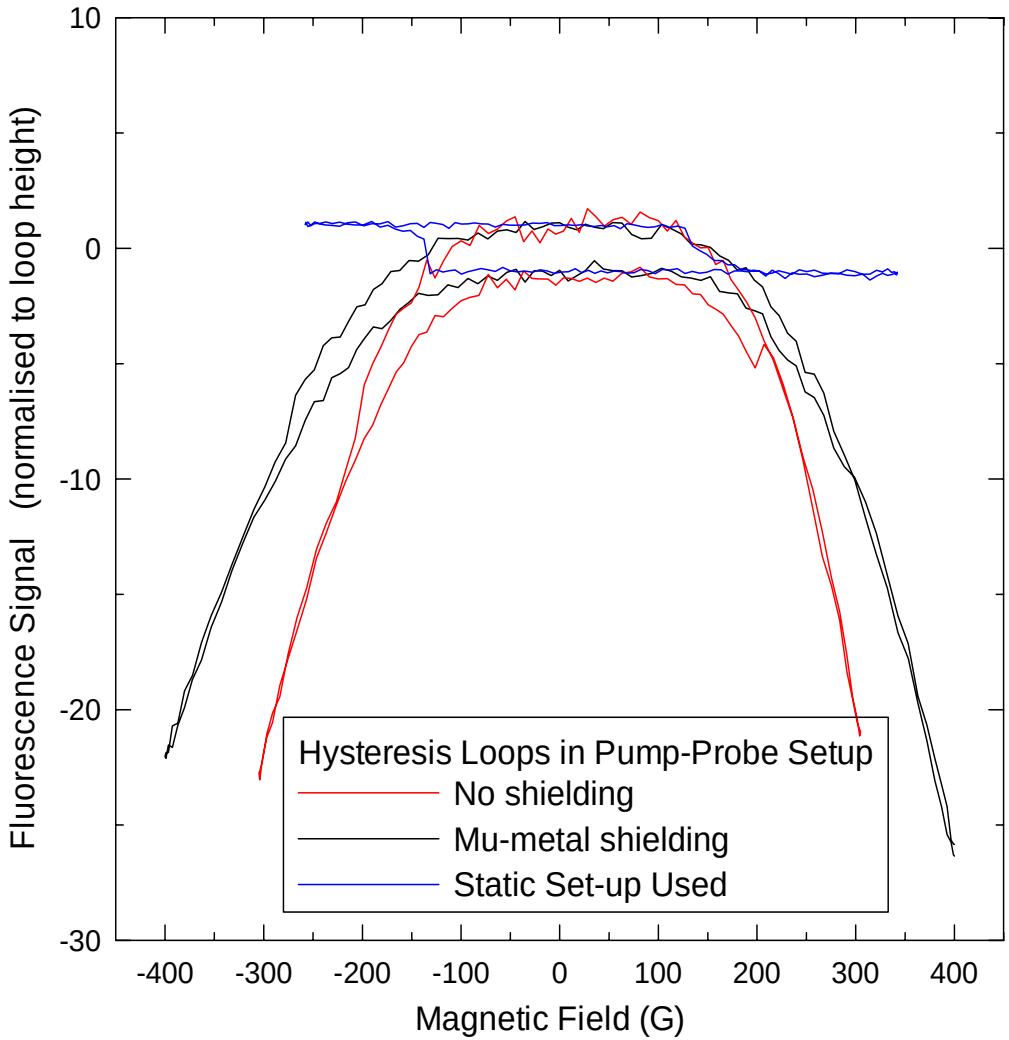


< Sense of Loop Reverses >

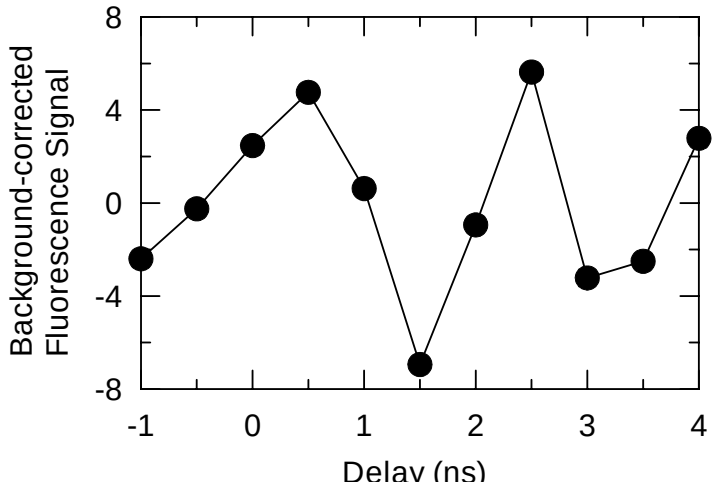
First timing result (detector problems)

Static – ion chamber: no **B** field dependence

Timing – plastic scintillator with PMT: strong **B** dependence



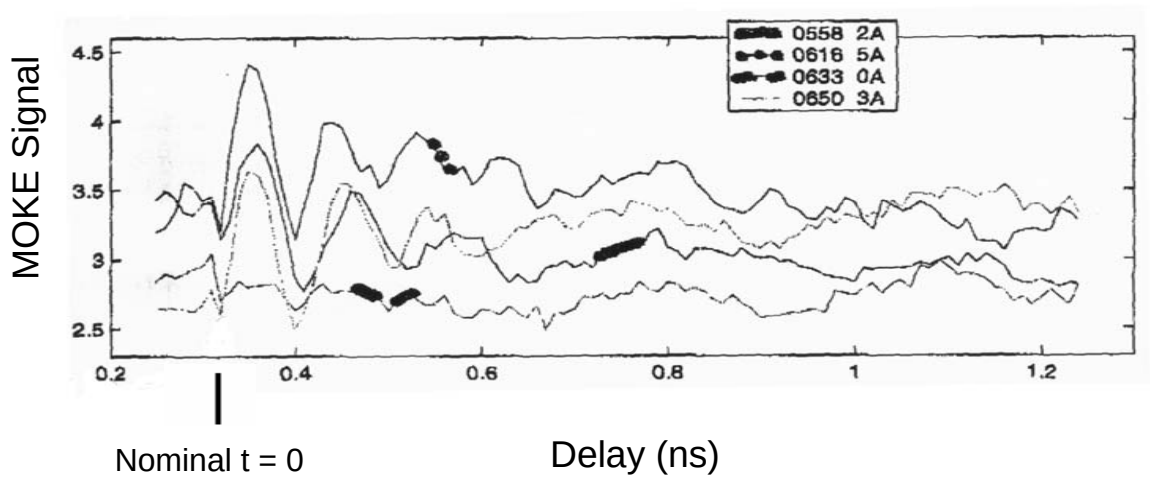
First Timing Results (initial data)



Gd Signal in $Gd_{0.227}Co_{0.773}$

Oscillations observed!!

Real or Artifact?



Optical: Red Pump,
Blue Probe with MOKE

Period from X-ray study: ~2ns Period from Laser study: ~100ps

Conclusions from Initial Spin dynamics study

- All elements of the experiments demonstrated to be feasible
- Possible oscillations observed but much longer than optical studies
  - X-rays sampling smaller volume in concentration gradient?
  - Closer to ideal concentration?
  - Effect due to beam or thermal damage to sample?
- psec pulses would open up much wider range of materials
- Better more efficient detectors needed
  - Less sensitivity to magnetic field
  - More efficient → less beam damage

Phase Change in DVD-RAM materials

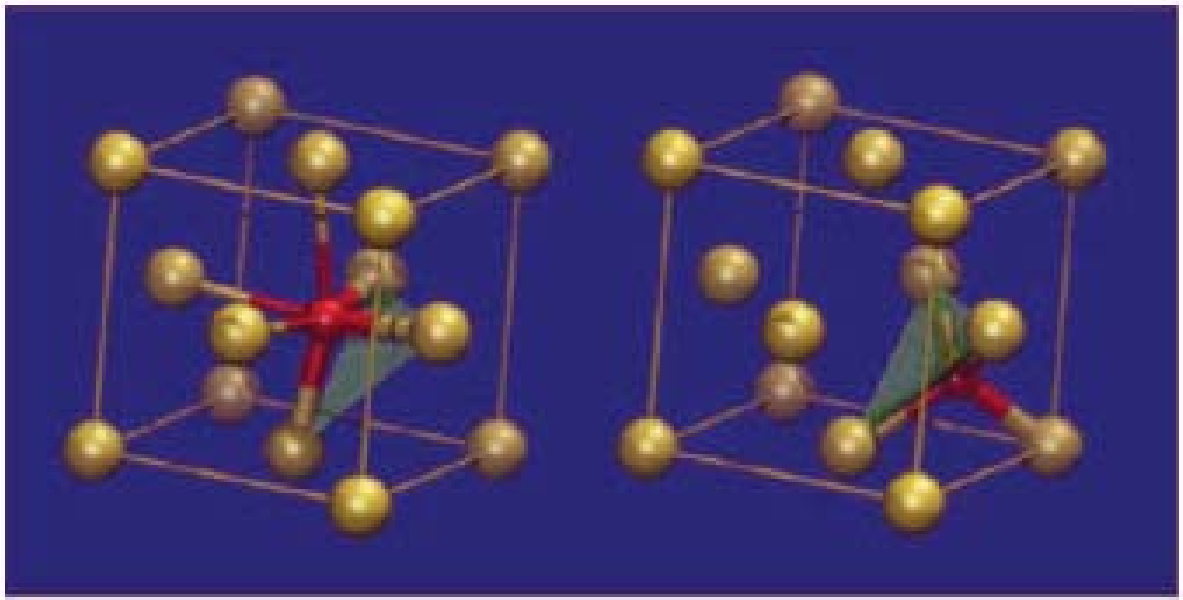
P. Fons, Center for near field optics research

- $Ge_2Sb_2Te_5$ reversible crystalline to amorphous phase change
  - Why are these materials fast?
  - Why are they reversible?
  - How can they be improved?
- EXAFS on end state materials suggests an “umbrella-flip” of the Ge atoms
- Time-resolved measurements can measure the intermediate “liquid” state
- First GU laser experiment

Crystalline to amorphous transformation

See Kolobov etal, Nature Mat. 3, 703 (2004)

Crystalline phase cubic from diffraction

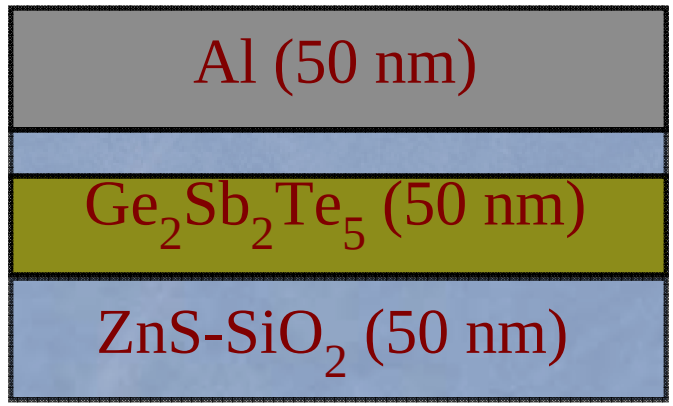


From EXAFS:
Ge 6 fold in crystal
4 fold in amorphous

Sb lattice relatively unchanged – may not actually melt

Planned experiment

EXAFS during the writing process-
capture the “liquid” phase in actual
device structure



Future: measure time dependent EXAFS in erasure (crystallization) process-
Will need to move disk for each shot

Future Directions

- Better detectors (APD arrays)
  - At least 10x more efficient
  - Less sensitive to magnetic fields
  - Project with the APS
- Frequency doubler or OPA for wider wavelength range
- Faster pulses?
  - Support the APS effort for specialized sector(s)
  - Meantime - special lower current/shorter bunch?

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