Hellman Lab

Magnetic States in Disordered Magnetically Doped Semiconductors Near the 3D Metal-Insulator Transition: from "Kondo-like" Systems to Local Moments

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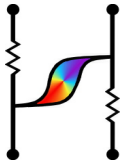
LBL Materials Sciences Division



Thanks to

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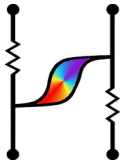


Magnetically-doped amorphous semiconductors

Amorphous (*a*-) X-Si, X-Ge, X-C alloys:

X= Gd, Y, Tb, Mn

Gd: $4f^7 5d^1 6s^2$: trivalent ion, S=7/2 μ_B, L=0

Y: $4d^1 5s^2$: trivalent ion (same size as Gd), S=0

Both Gd and Y dope Si, Ge, C (3 electrons donated per ion)

Samples prepared by e-beam co-evaporation in UHV conditions
(some also by magnetron co-sputtering)

Structural: TEM, X-ray, EXAFS, RBS

Specific heat, thermal conductivity, (limited) thermopower

Conductivity, magneto-conductivity

Magnetization, magnetic susceptibility

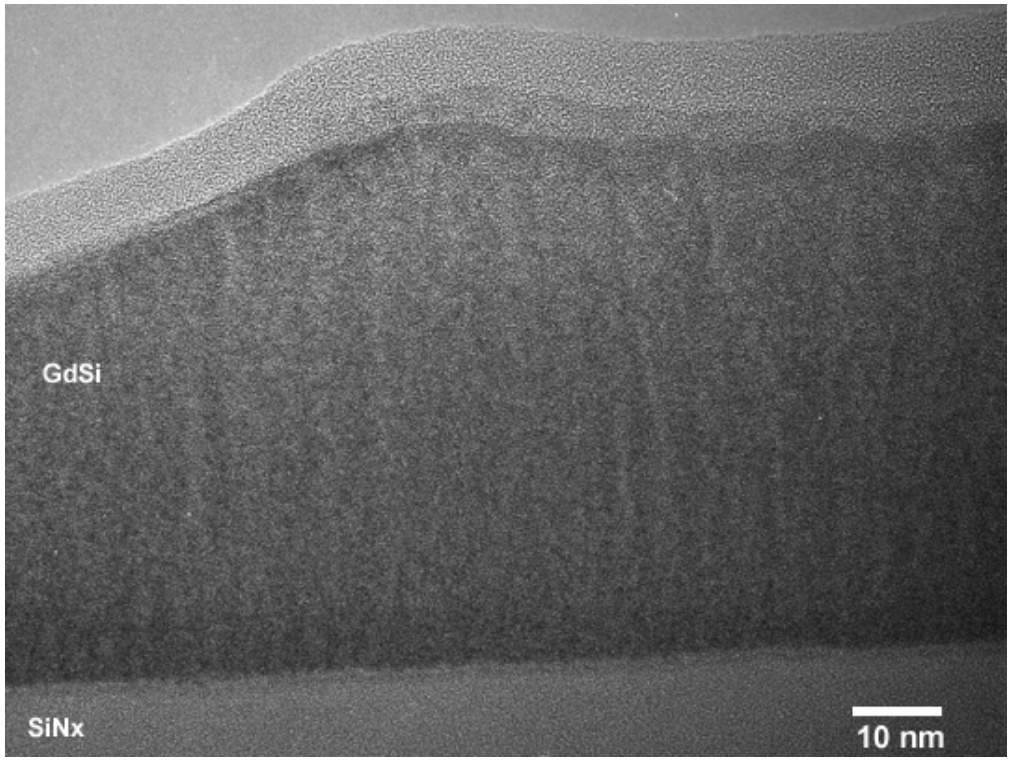
XAS (x-ray absorption spectroscopy); electron spin resonance

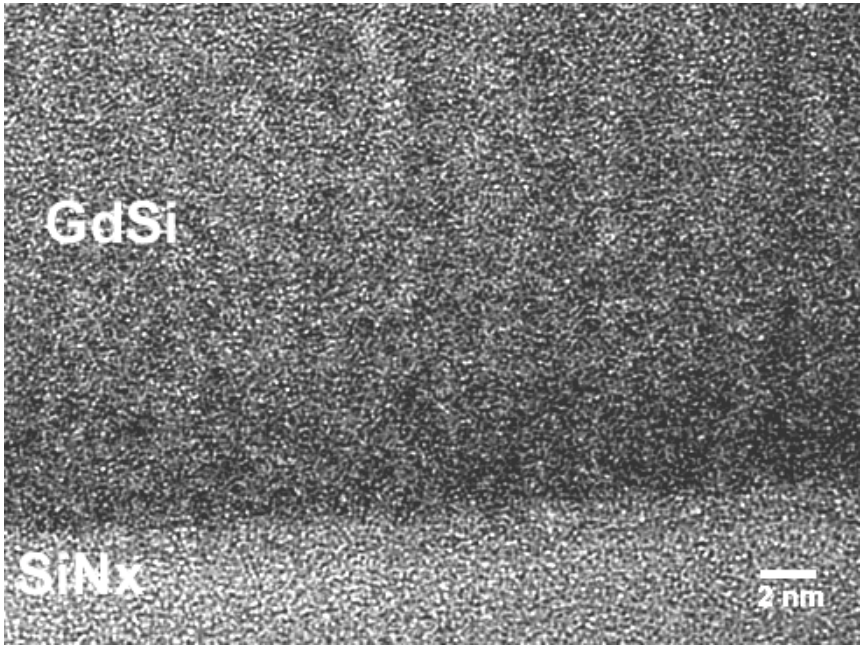
Tunneling spectroscopy

IR absorption spectroscopy

Theoretical analysis: LDA (Y-Si); FLAPW calculations (Gd-Si); magnetic disorder in Anderson type model

Amorphous Magnetic Semiconductors: Structure

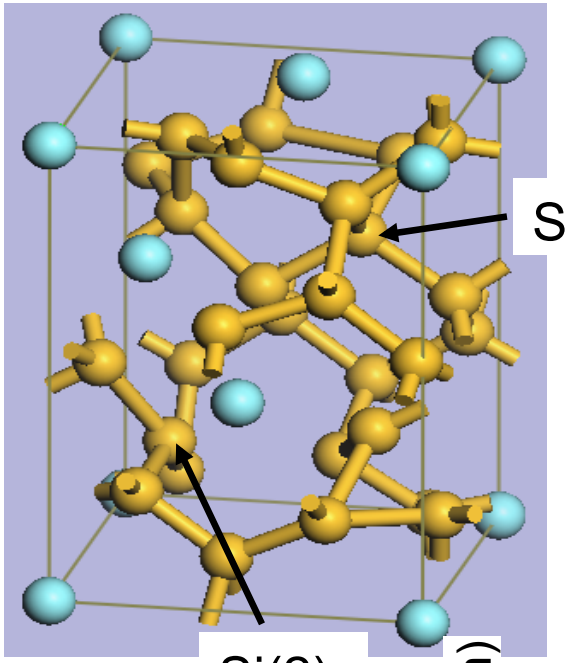


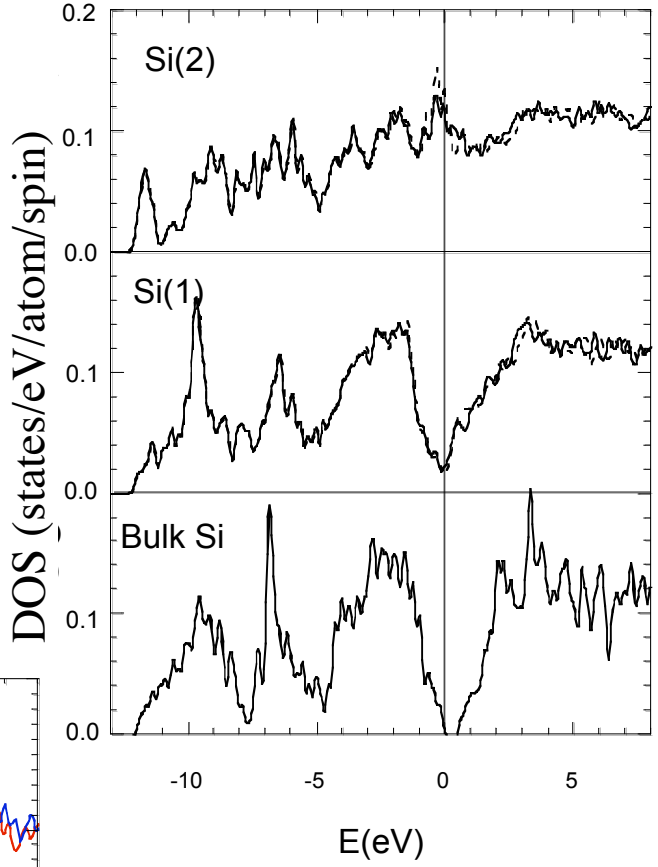


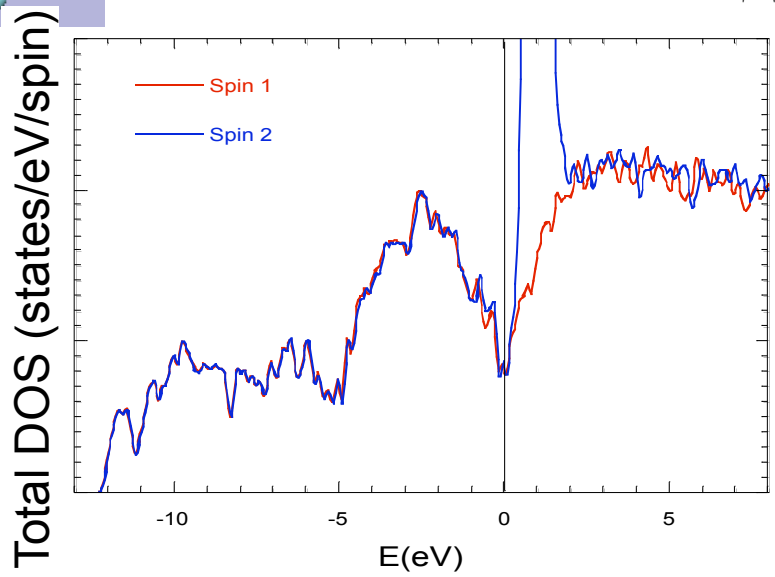
High Resolution TEM: Low magnification cross section TEM shows slight columnar growth morphology; high resolution TEM shows films are amorphous
Samples also prepared by magnetron co-sputtering: no columns, compressive instead of tensile strain, similar atomic density, same magnetic and magneto-transport properties

EXAFS: no clustering of Gd
Both Gd and Si surrounded by Si
Si tetrahedral bonding recovers quickly (nnn) from perturbation of Gd ions

FLAPW calculations for a-Gd_4Si_{28} ($Gd_{13}Si_{87}$)
Ruqian Wu, UCIrvine

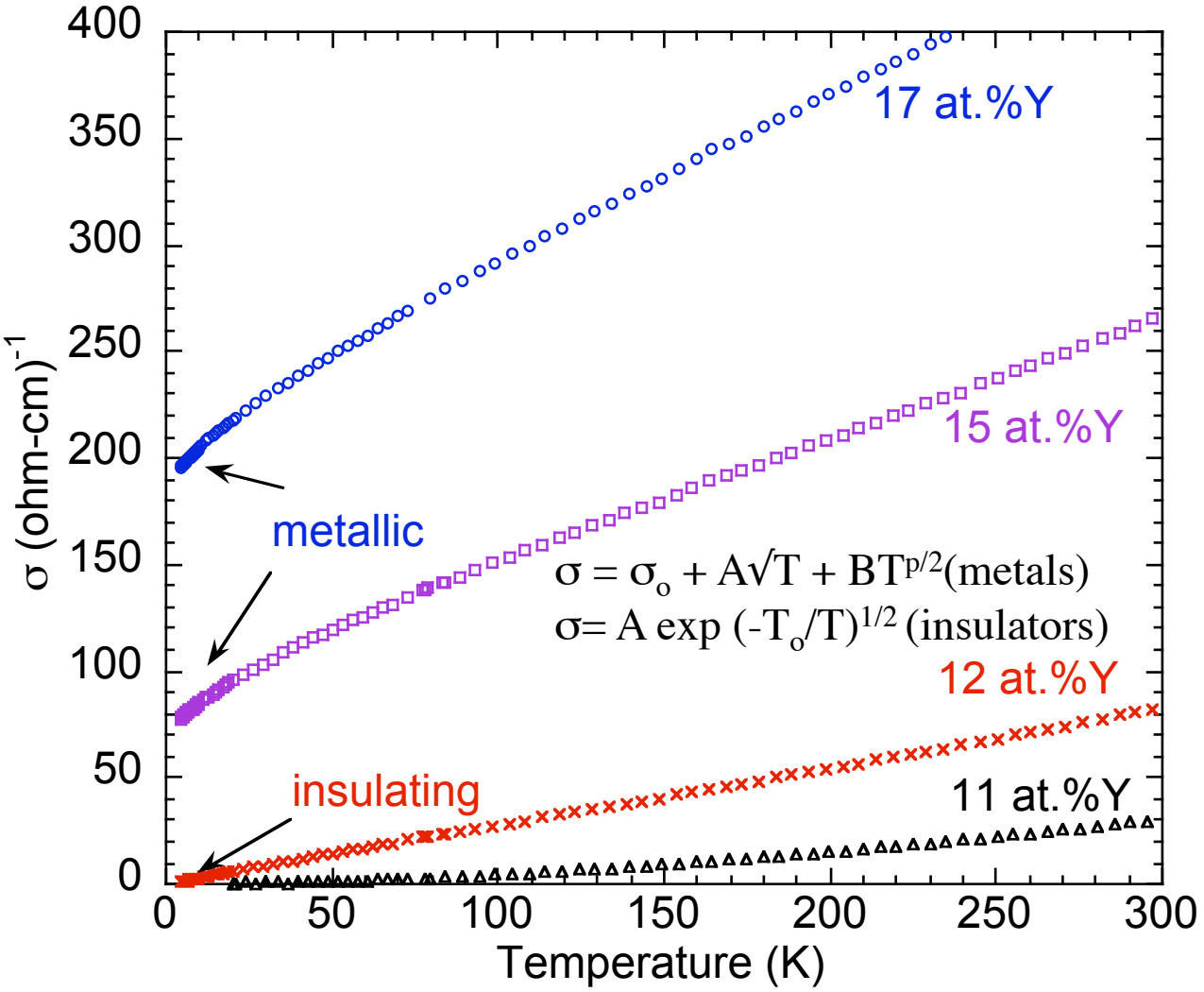






Electrical conductivity of non-magnetic ions (Y) in amorphous Si

Y: $4d^15s^2$: non-magnetic trivalent ion (same size as Gd). Expect 3 electrons donated per ion

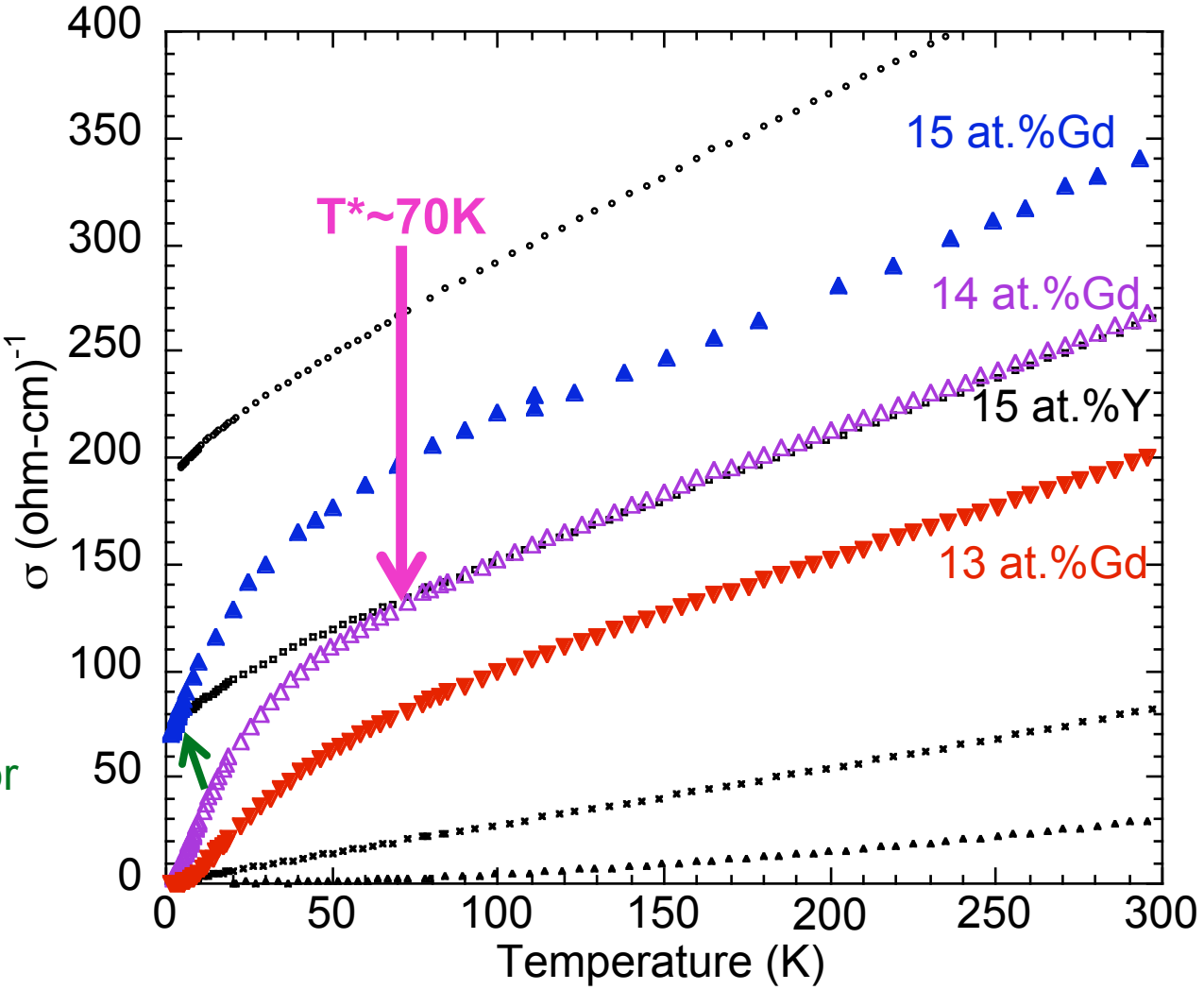


Metal-Insulator transition between 12 and 15 at.%Y (14 at.%Y)

Electrical conductivity of magnetic ions (Gd) in amorphous Si

Gd: $4f^7 5d^1 6s^2$: trivalent ion, S=7/2 μ_B, L=0
Also 3 electrons donated per ion

Magnetic field increases σ for Gd-Si



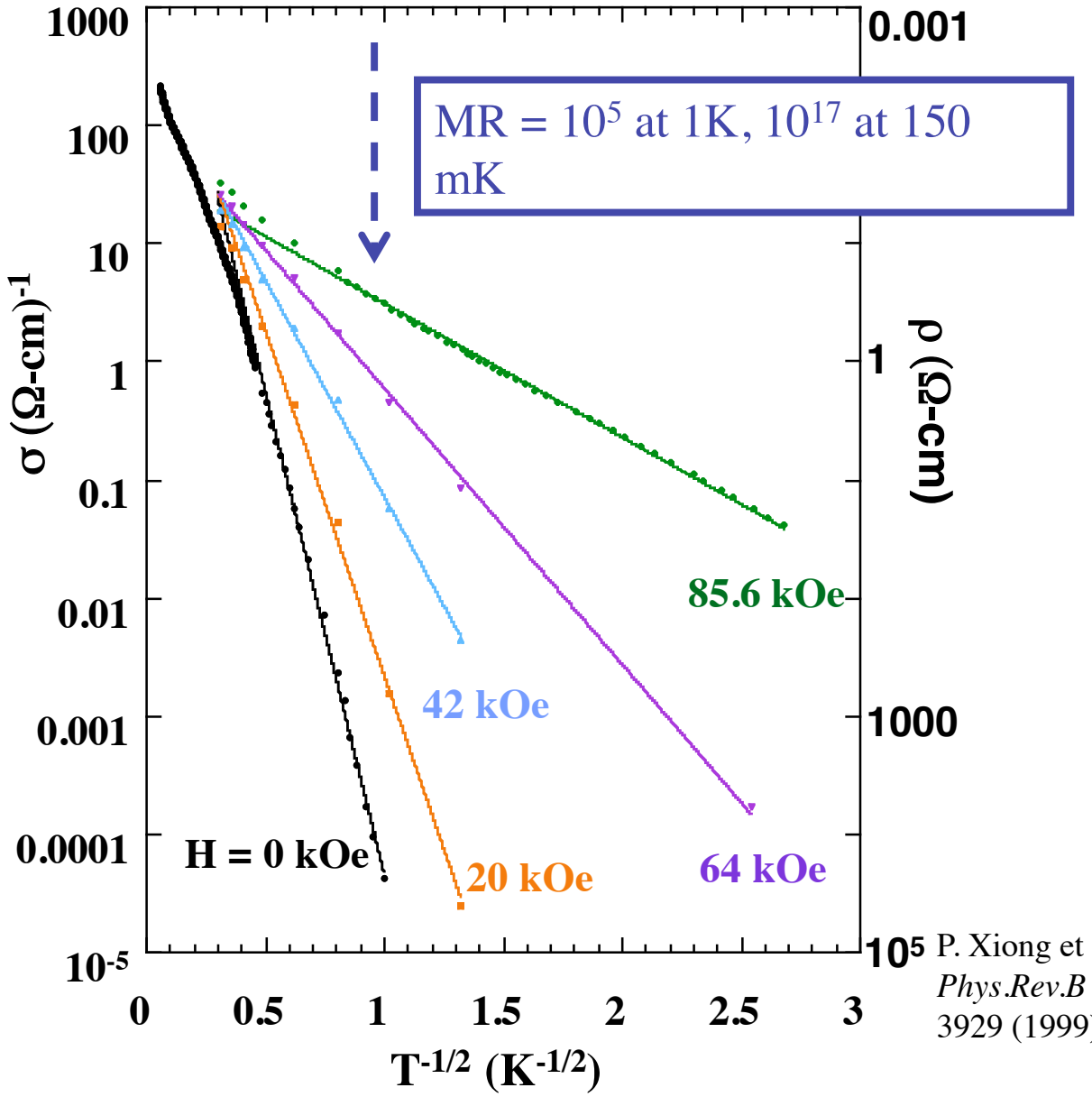
Magnetic ions (Gd) enormously reduce σ for T<T*~70K

F. Hellman et al, Phys. Rev. Lett. 85, 4652 (1996).

Effect of magnetic field is enormous (negative magnetoresistance) for Gd-Si (negligible for Y-Si)

e.g. 13 at.%Gd
(an insulator at H=0;
nearly metallic at high H)

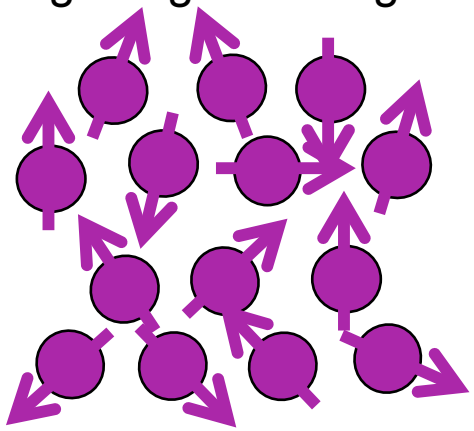
$\sigma = A \exp(-T_o/T)^{1/2}$
T_o depends on H
A independent of H



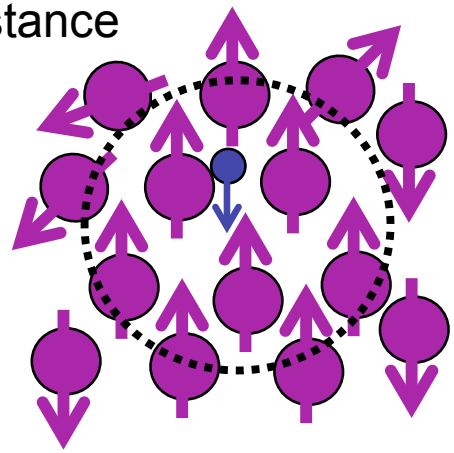
P. Xiong et al, *Phys.Rev.B* **59**, 3929 (1999).

Magnetic polaron model
Magnetic disorder and shift of mobility edge

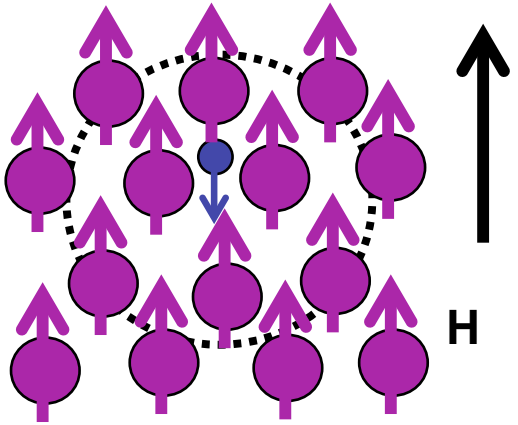
Crystalline magnetic semiconductors, e.g. doped Gd_3S_4, CdMnSe
Large negative magnetoresistance



Undoped: moments random



Doped: moments aligned inside Bohr radius. Hopping frustrated; high ρ



Magnetic field aligns moments everywhere Hopping easier; low ρ

However, in *a*-Gd_xSi_{1-x},
$n_{el} \sim 10^{22}$ cm^{-3}
n_{el}/n_{Gd} ~1-3
Orders of magnitude larger than polaron model (also a problem with metallic samples)
Instead, magnetic disorder due to carriers interacting with randomly oriented moments (J_{sf}) shifts mobility edge higher in band at zero field, lower when moments align in H
Qualitatively makes sense, but this material is already in strong disorder limit, and the scale of the effect is not predicted

Metallic samples (≥ 14 at.%Gd)

Samples 2-4 slightly different compositions
I-M transition at 50 kOe for sample 3

Perfect overlay of data with different H, x

x changes E_F

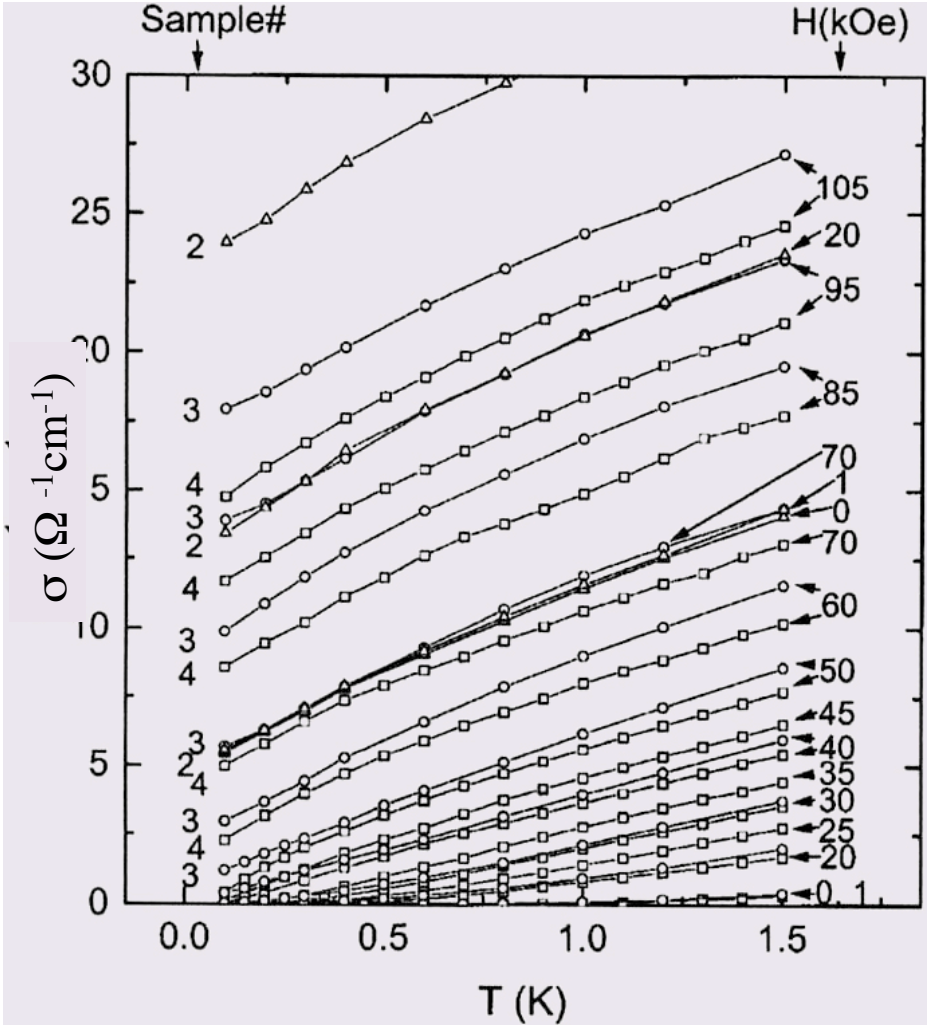
H changes E_c

Only E_c-E_F is important

$\sigma = \sigma_o + \sigma_1\sqrt{T} + \sigma_2 T^{p/2}$

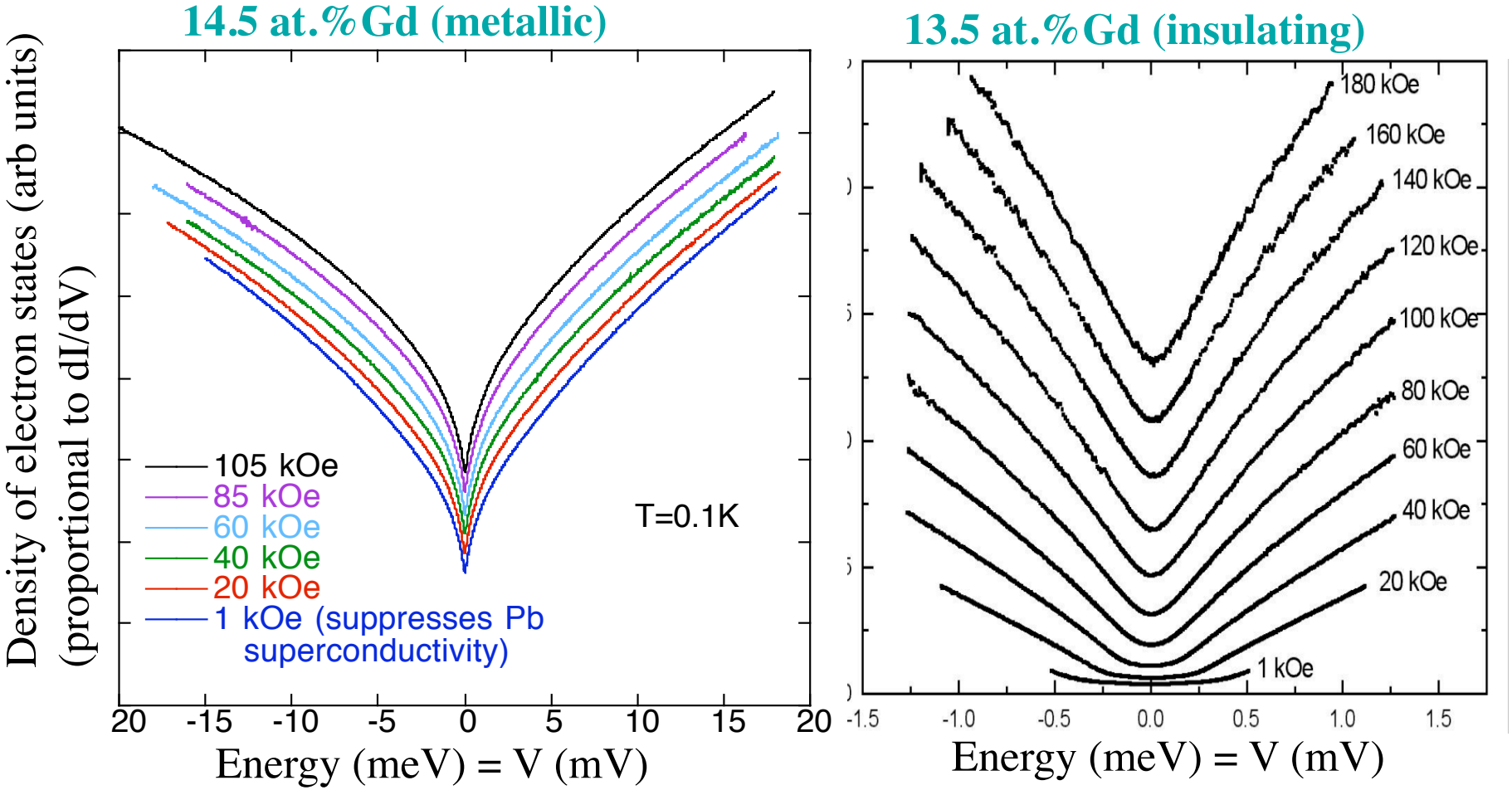
$\sigma_o = \sigma_o(H,x)$

σ_1, σ_2 independent of H, x



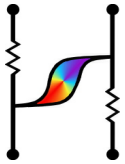
W. Teizer et al, *Phys. Rev. Lett.* **85**, 848 (2000).

To measure electron density of states: Tunnel junctions: Gd-Si/oxide/Pb or Al



W. Teizer, F. Hellman, R.C. Dynes, Phys. Rev. Lett. 85, 848 (2000)

Tunneling conductance (proportional to density of electron states) depends strongly on magnetic field



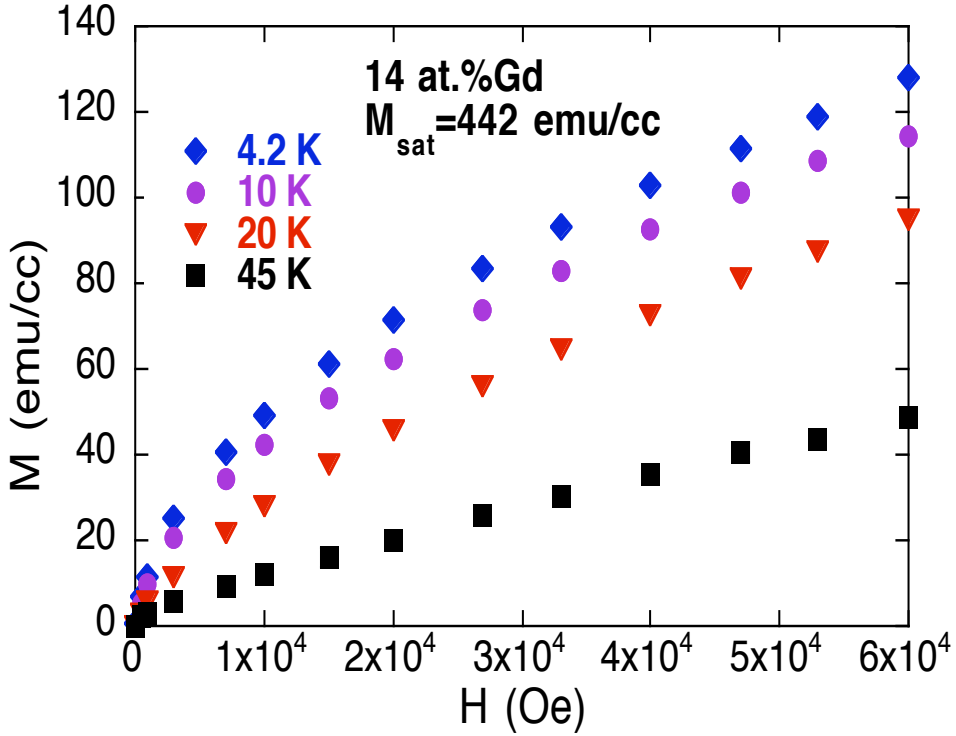
Effect of magnetic moments on the 3D metal-insulator transition

Some parameters become field dependent

	Metal	Insulator
Conductivity	$\sigma(T) = \sigma_0 + \sigma_1 T^{1/2} + \sigma_2 T^{p/2}$	$\sigma(T) = Ae^{-\left(\frac{T_0}{T}\right)^{1/2}}$
Density of States	$N(\varepsilon) = N_0 + N_1 \varepsilon^{1/2}$	$N(\varepsilon) = N_2 \varepsilon^2$

Magnetic disorder: increases localization which shifts E_c; this in turn affects Coulomb gap by decreasing screening which increases correlation effects

Magnetic properties of amorphous Gd-Si
Gd f shells have no direct exchange, only indirect



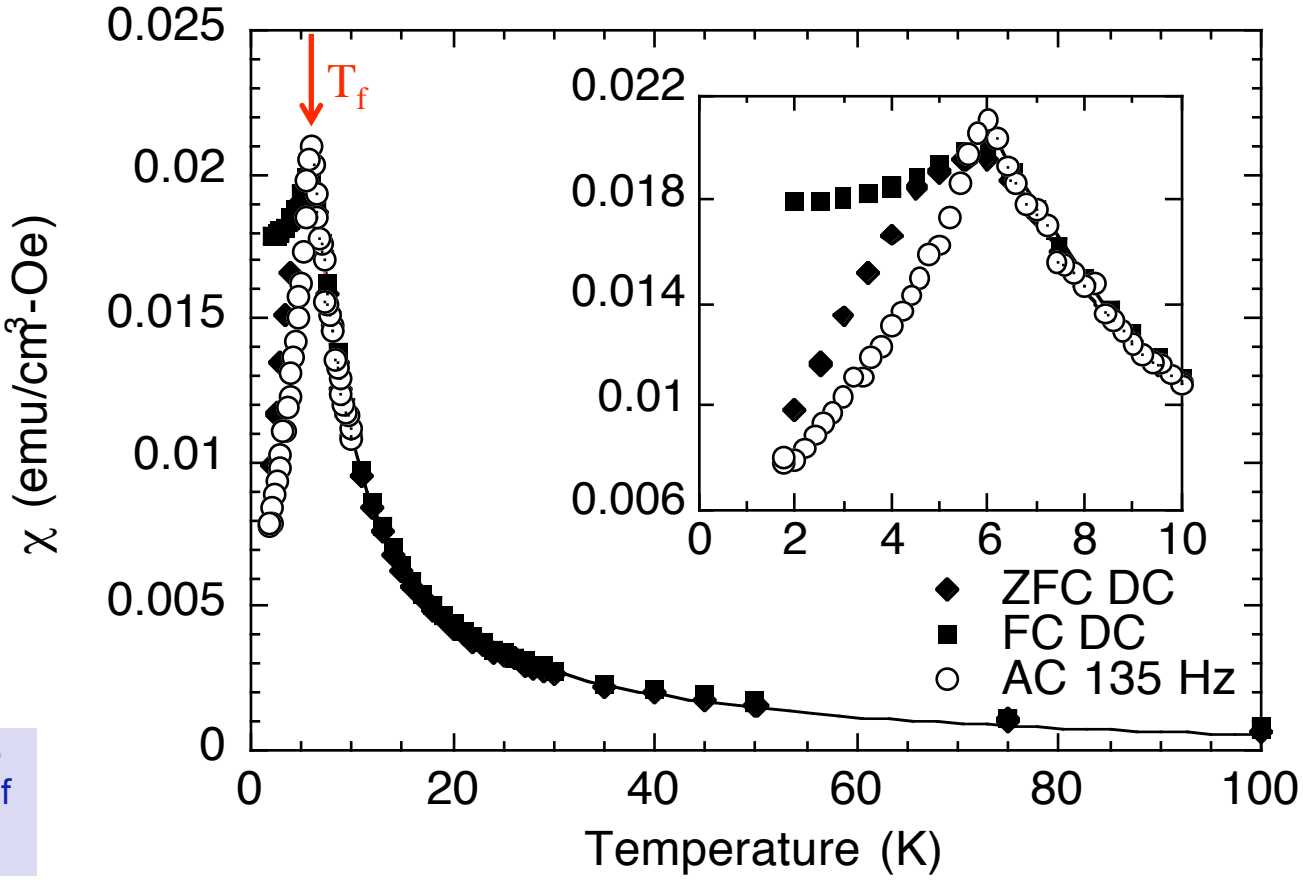
F.Hellman, D.R.Queen, R.M.Potok, B.L.Zink, Phys. Rev. Lett. 84, 5411 (2000)

M(H,T) significantly below Brillouin function even at *25T*; doesn't scale with H/T

Strong (>25T, 125K) interactions: ferro-, anti-ferromagnetic

Gd-Gd indirect exchange (RKKY-like) despite being insulating!

Magnetic susceptibility χ shows classic spin glass freezing

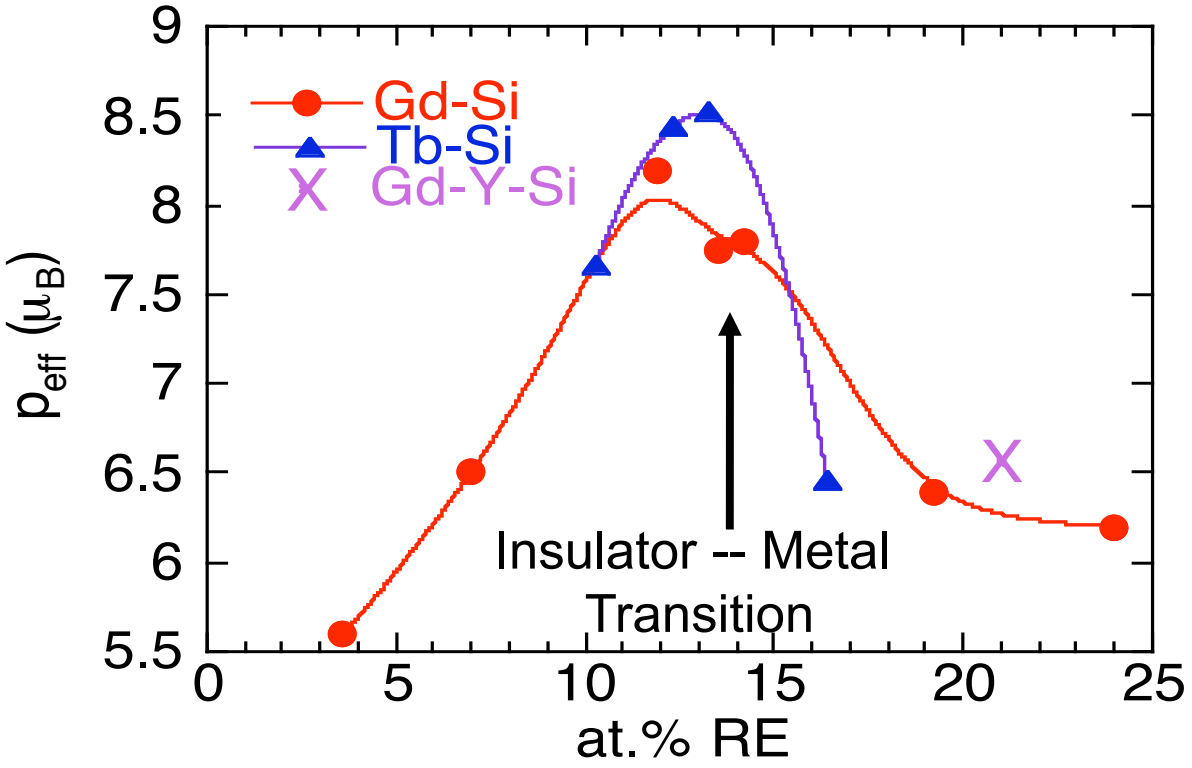


In PM state above T_f
$\chi = A/(T-\theta)$

θ small (<2.5K, $<<T_f$). Close to non-interacting Curie law, suggests nearly *perfectly balanced* FM/AFM interactions – very unusual for Gd - due to strong disorder (?)

$A = n_{Gd}p^2\mu_B^2/3k_B$; effective moment $p = (g^2J(J+1))^{1/2} = 7.9$ for Gd^{3+} ions

Effective moment

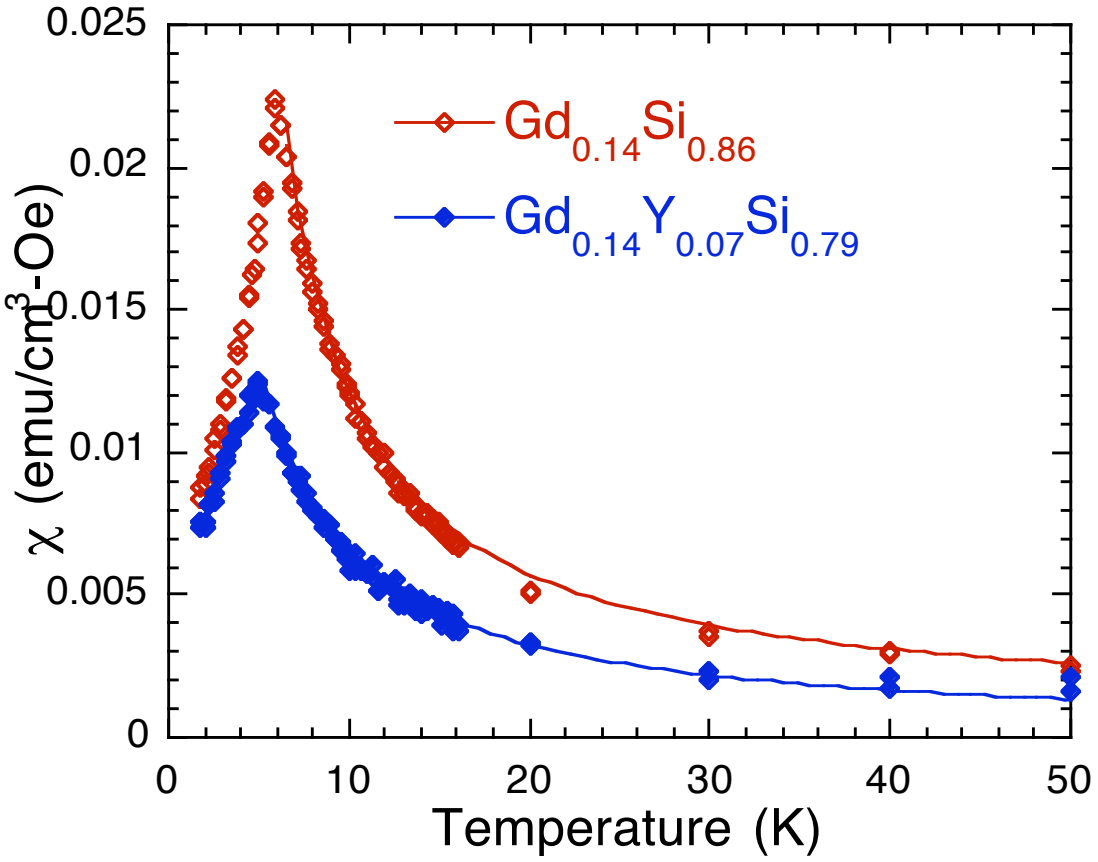


Effective moment $p_{eff} = (g^2J(J+1))^{1/2} = 7.9$ for Gd^{3+} ions

Here, p_{eff} of Gd (and Tb) depends on at.%RE, largest at metal-insulator transition (*not seen* in other metallic or insulating Gd alloys)

Magnetic susceptibility (low field)

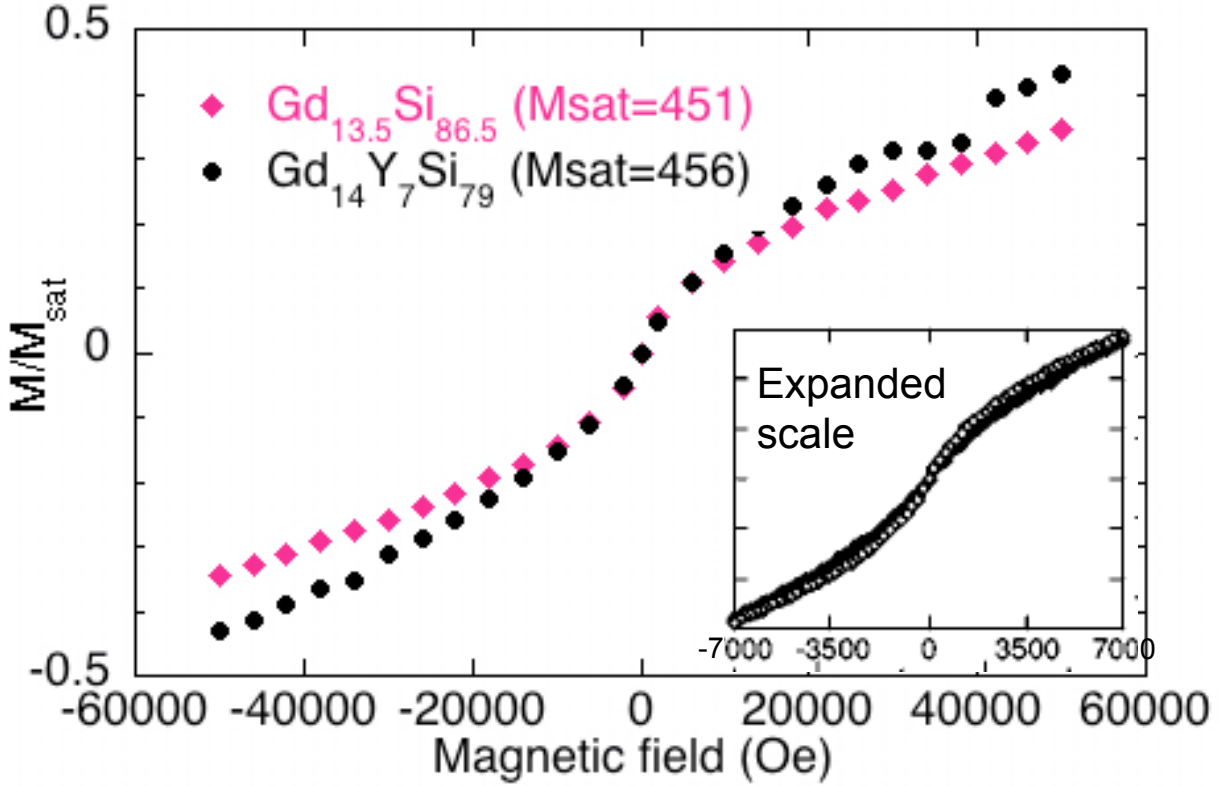
(above spin glass freezing temperature; no hysteresis)



AC χ(T)
4 Oe, 400 Hz

χ not monotonic with composition- twice as large at M-I transition!
Heat capacity also shows effect of M-I transition – excess entropy

High field M(H) for a-Gd-Y-Si alloy constant Gd at.%, varying Y adds carriers

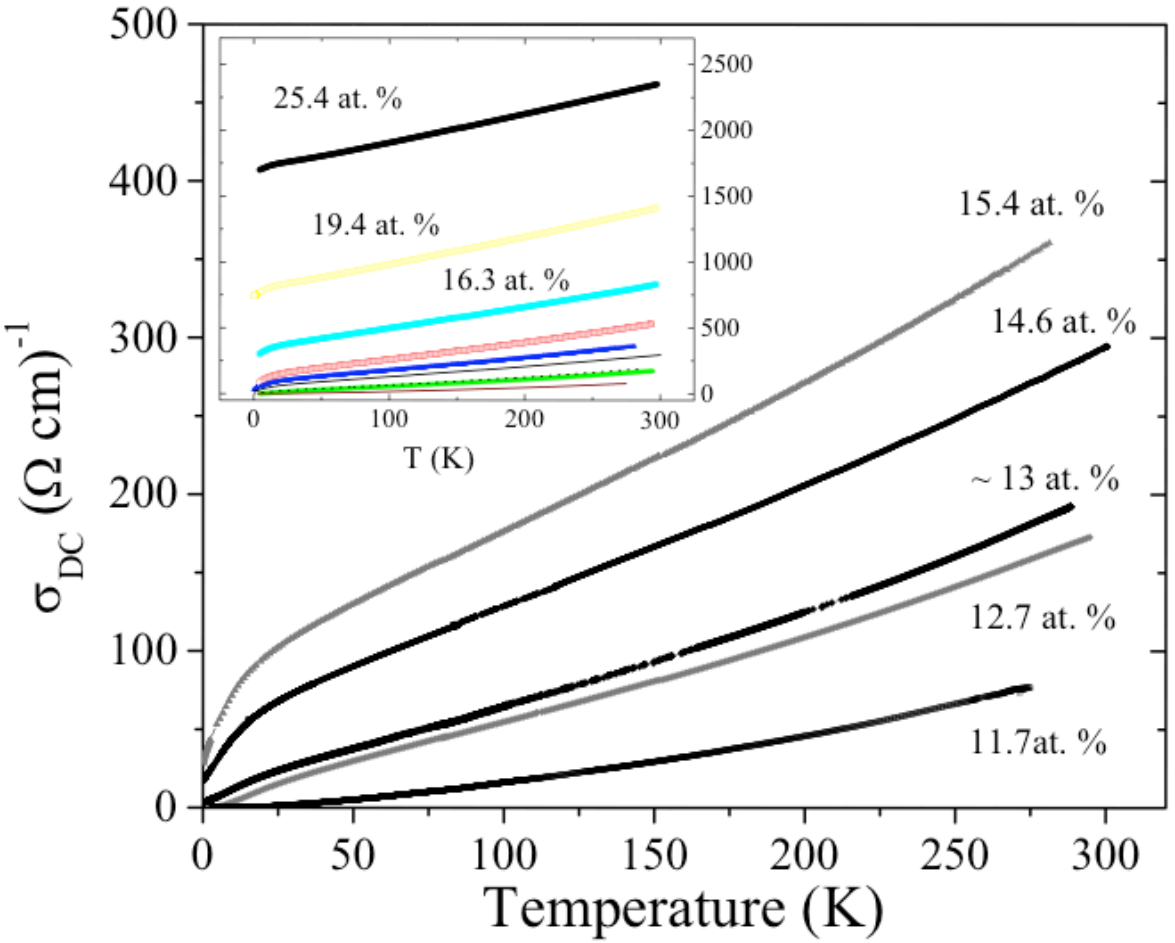


Adding carriers (Y) with constant number of moments (Gd) weakens Gd-Gd interactions – T_f drops (6K to 5K), M(H) closer to non-interacting (Brillouin function) limit, and already small θ drops to zero.

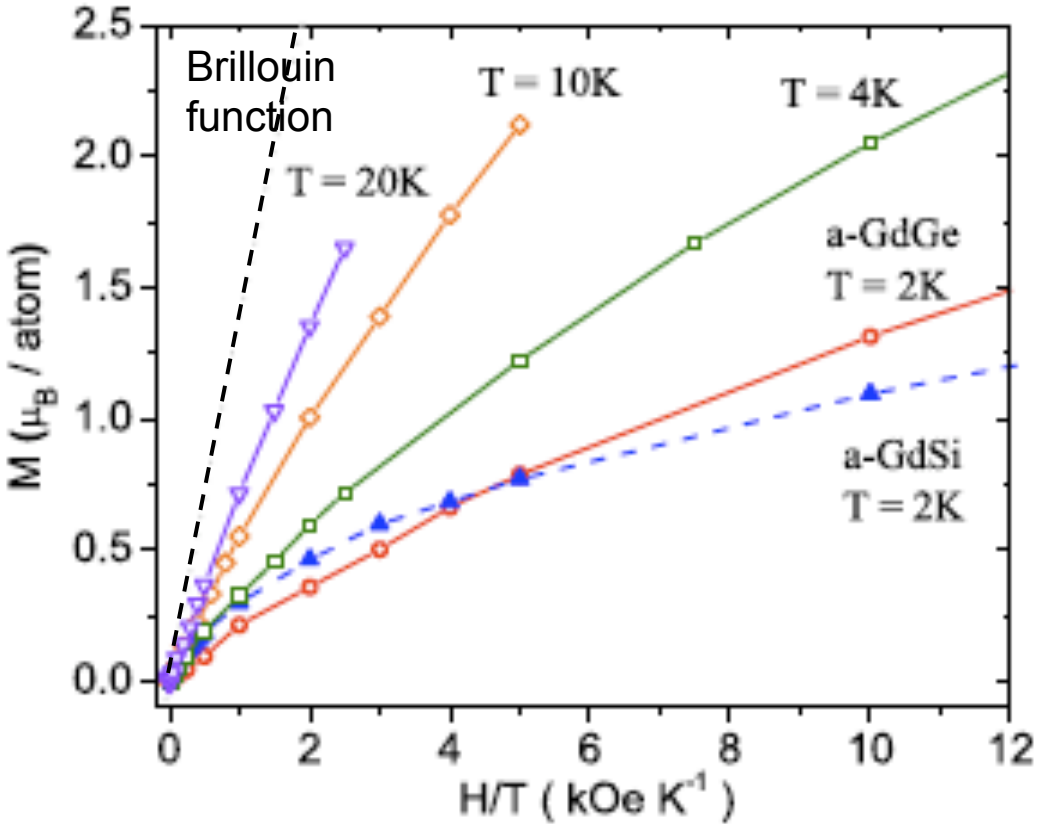
Increased *n* increases k_F while r remains constant: RKKY strength is reduced

$$J_{\text{Gd-Gd}}(r) = 6\pi Z J_{sf}^2 N(E_F)\left(\frac{\sin(2k_F r)}{(2k_F r)^4} - \frac{\cos(2k_F r)}{(2k_F r)^3}\right)$$

a-Gd-Ge: M-I transition still at 14 at.% Gd



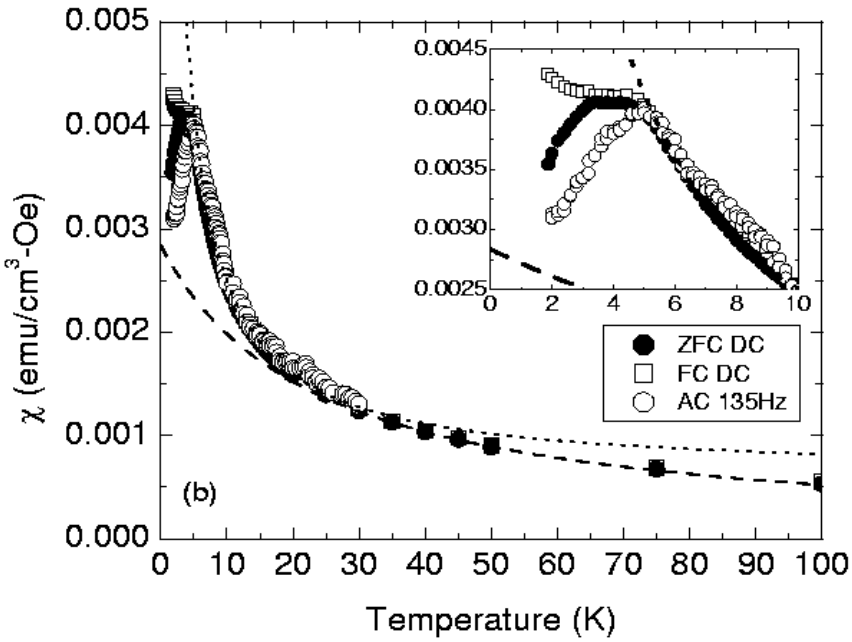
M(H) for a-Gd-Ge still well below non-interacting Brillouin function, but closer than a-Gd-Si

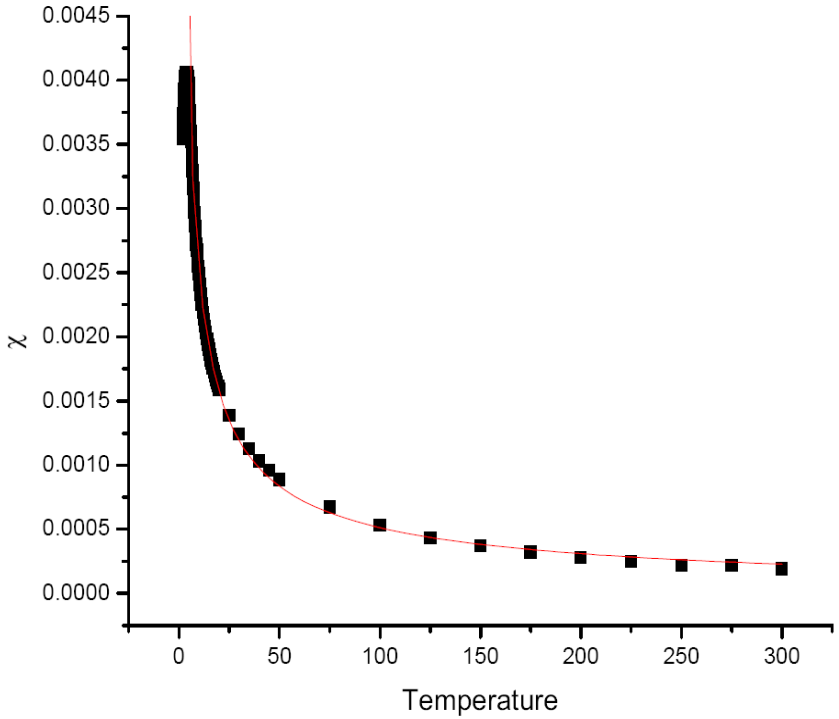


a-Gd-Si and *a*-Gd-Ge
Both 14 at.%Gd

Low field χ smaller for a-Gd-Ge than a-Gd-Si; high field M(H) is higher

Low field magnetic susceptibility: Gd-Ge quite different than Gd-Si





a-Gd-Ge (14.6 at.%)

VERY poor fit to Curie-Weiss law

Strongly suppressed χ (4x less than non-interacting Curie Law)

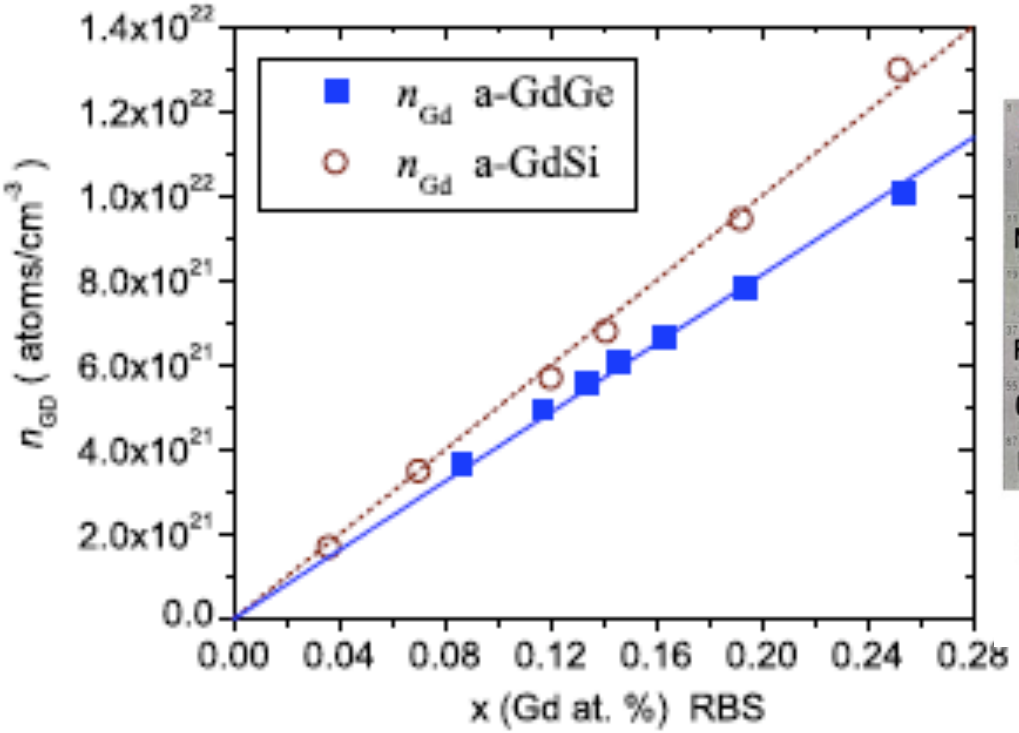
χ monotonic in at.%; no MIT peak

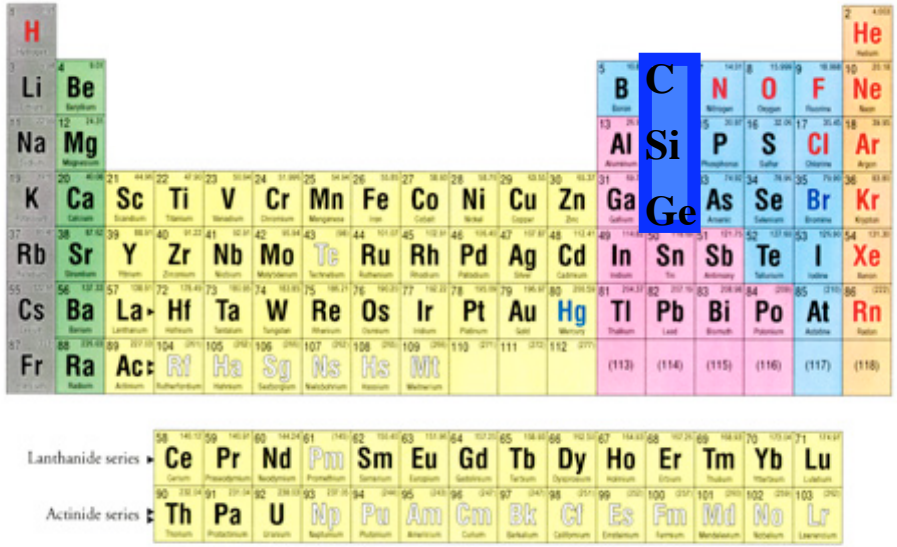
Fits Bhatt-Lee A/T^{α}

Random AFM spin singlets

α = 0.67 and A monotonic in x for all Gd_xGe_{1-x} near MIT

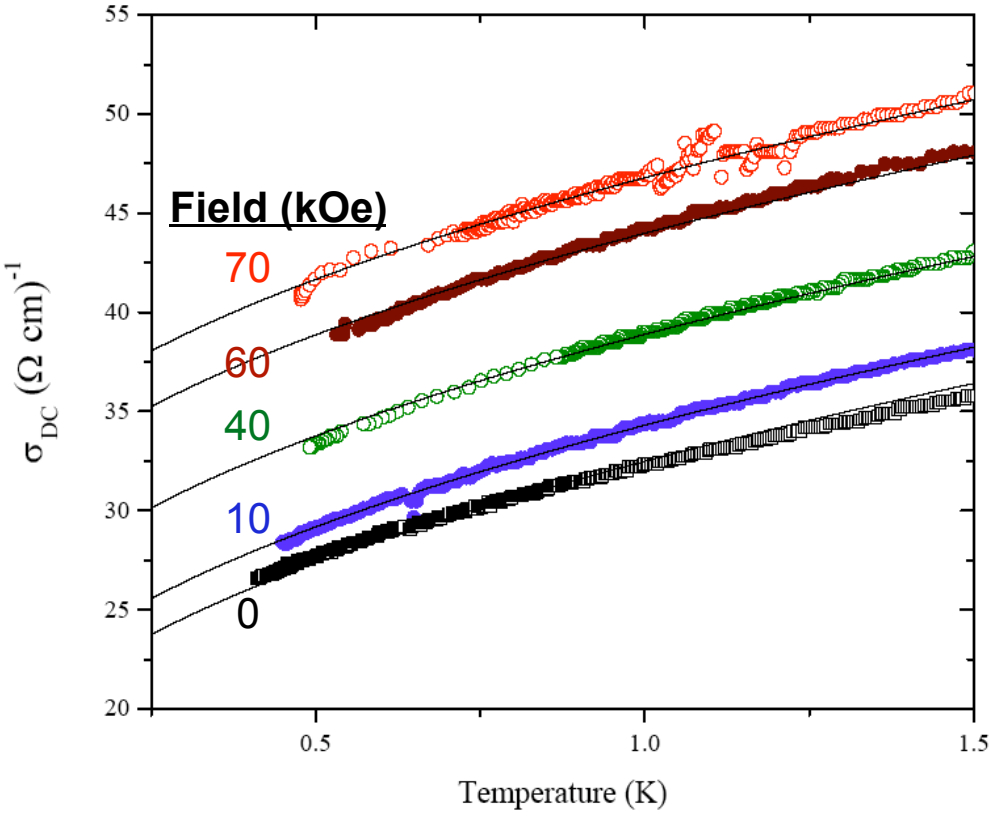
Atomic density of Gd-Si (and electron concentration) is lower in a-Ge than a-Si

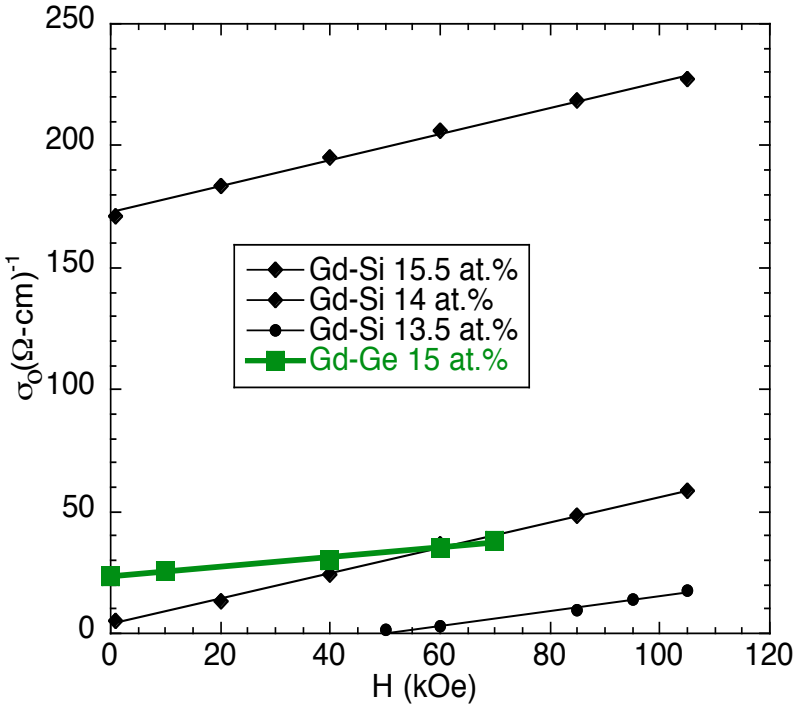




For each, total atomic density (atoms/cm^3) remains constant and equal to that of Si and Ge respectively (like a substitutional doping would do)

Temperature and field dependence of conductivity for metallic a-Gd-Ge (15 at.%)

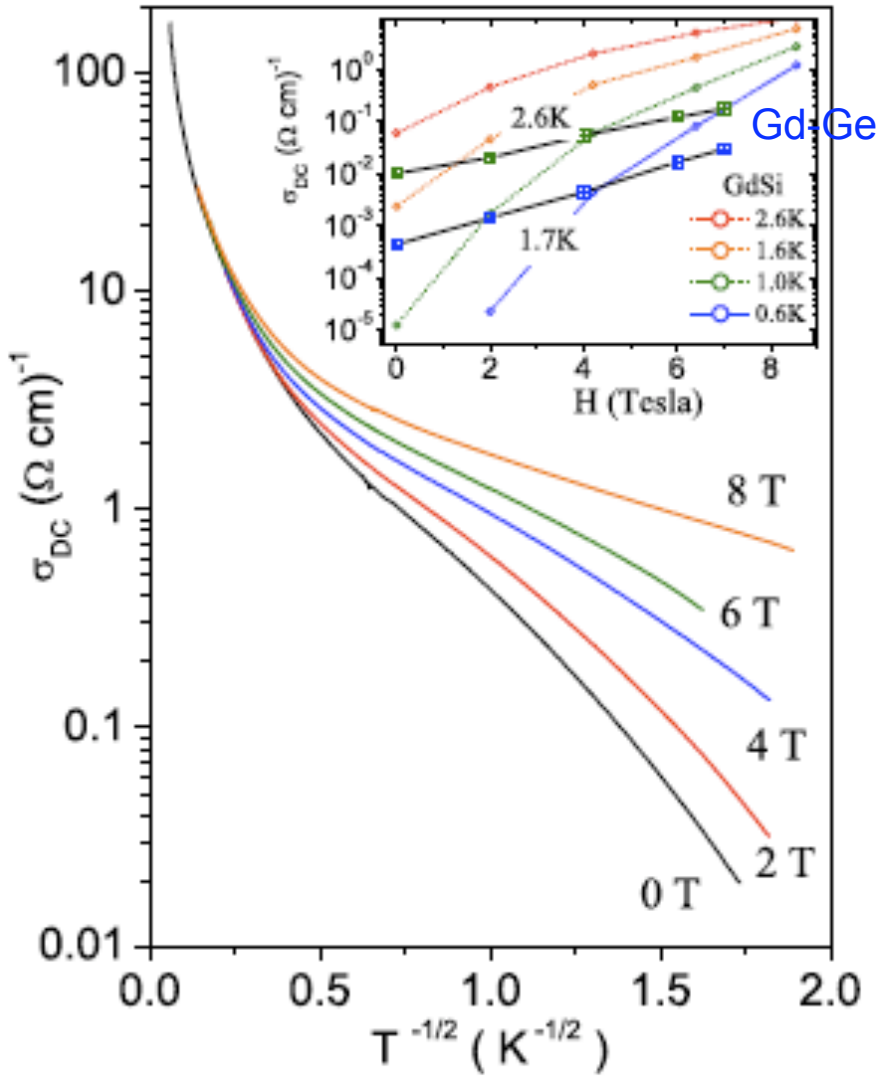




Temperature and composition dependence of σ similar to *a*-Gd-Si

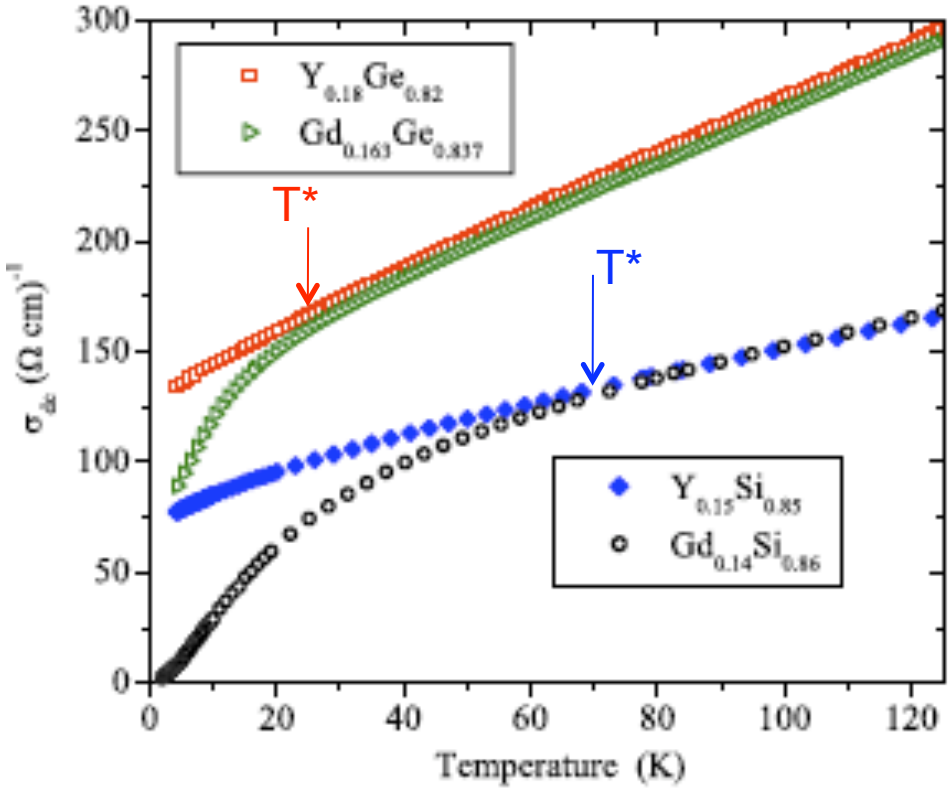
Field dependence much less

Field dependence of σ(T) is less in Gd-Ge at all T, x even though M(H) is higher



Also, temp dependence of insulating *a*-Gd-Ge less cleanly A exp $(-T_o/T)^{1/2}$

Onset of effects of magnetic moments occurs at lower T in Gd-Ge than Gd-Si*

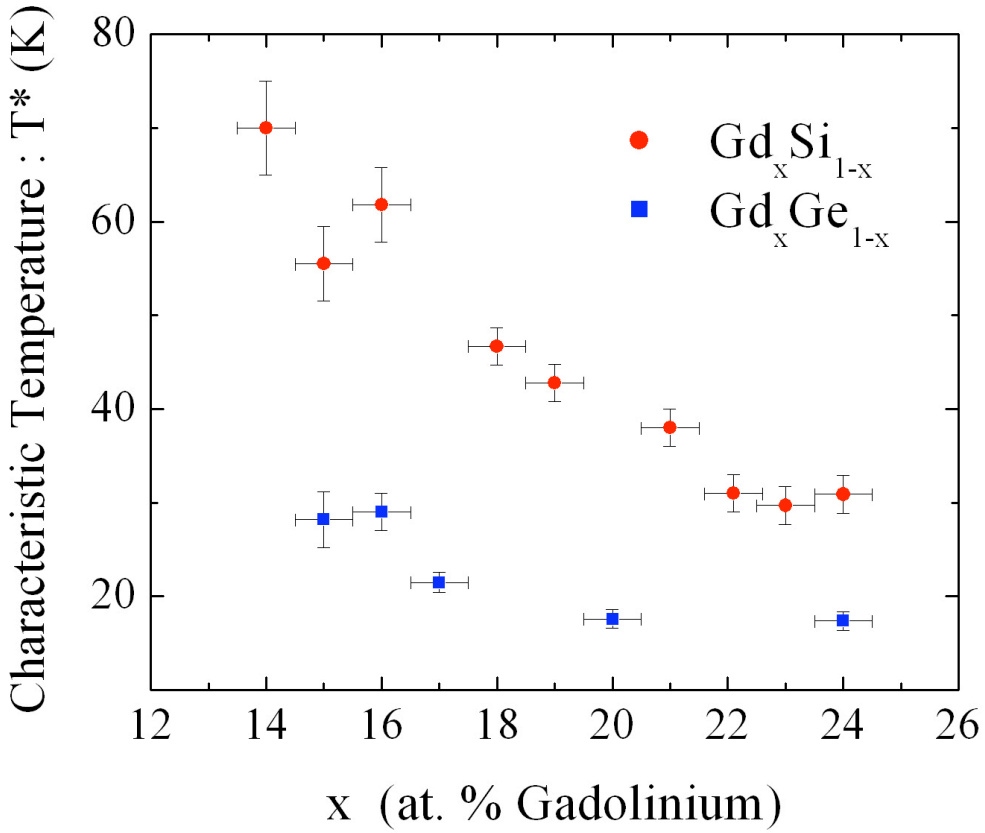


T* vs x (a-Gd-Si and a-Gd-Ge)

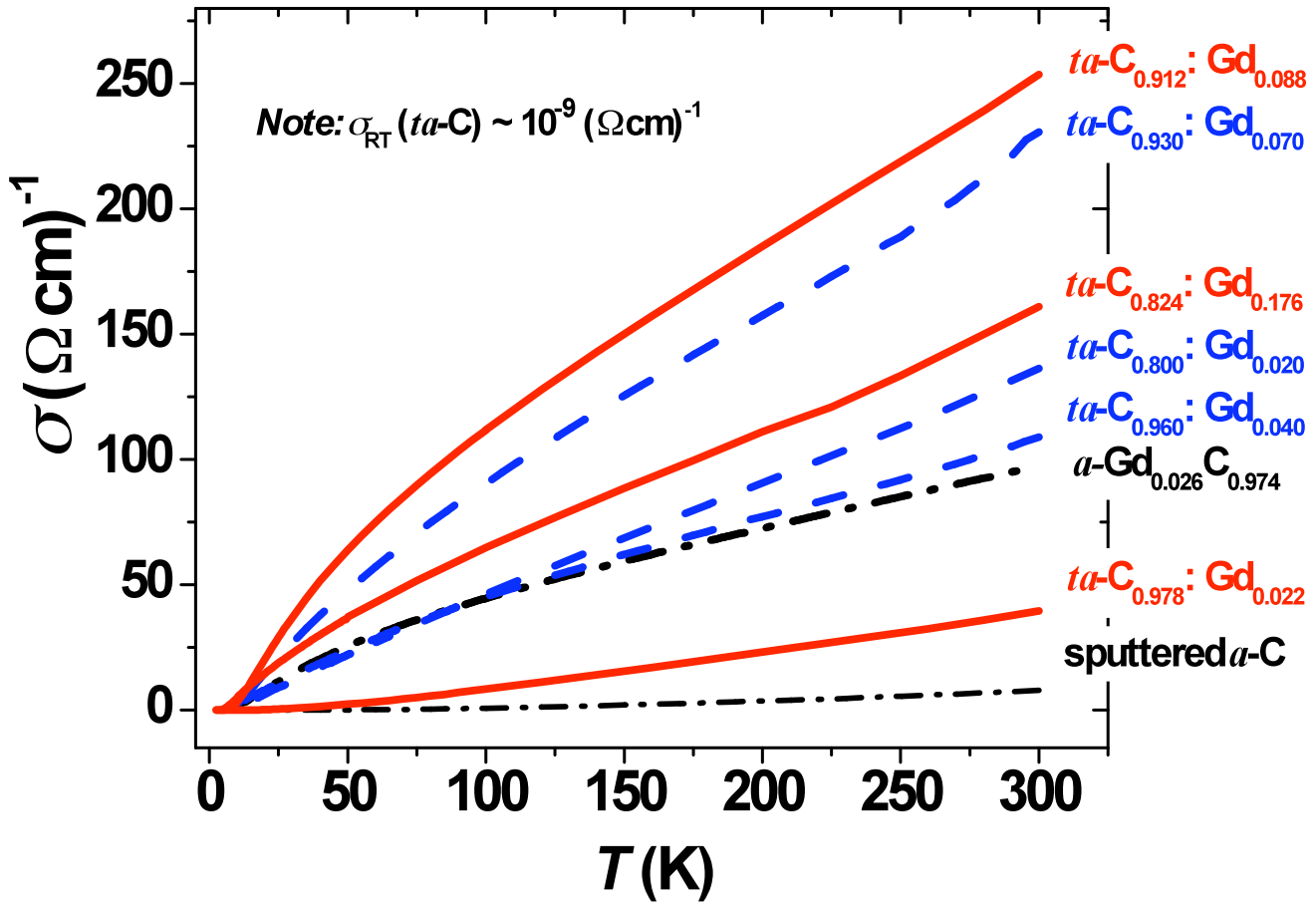
T* decreases with increasing Gd at.%, or decreasing band gap (Ge) (consistent with dependence of MR)

Also, decreases with constant Gd at.% and increasing Y at.%

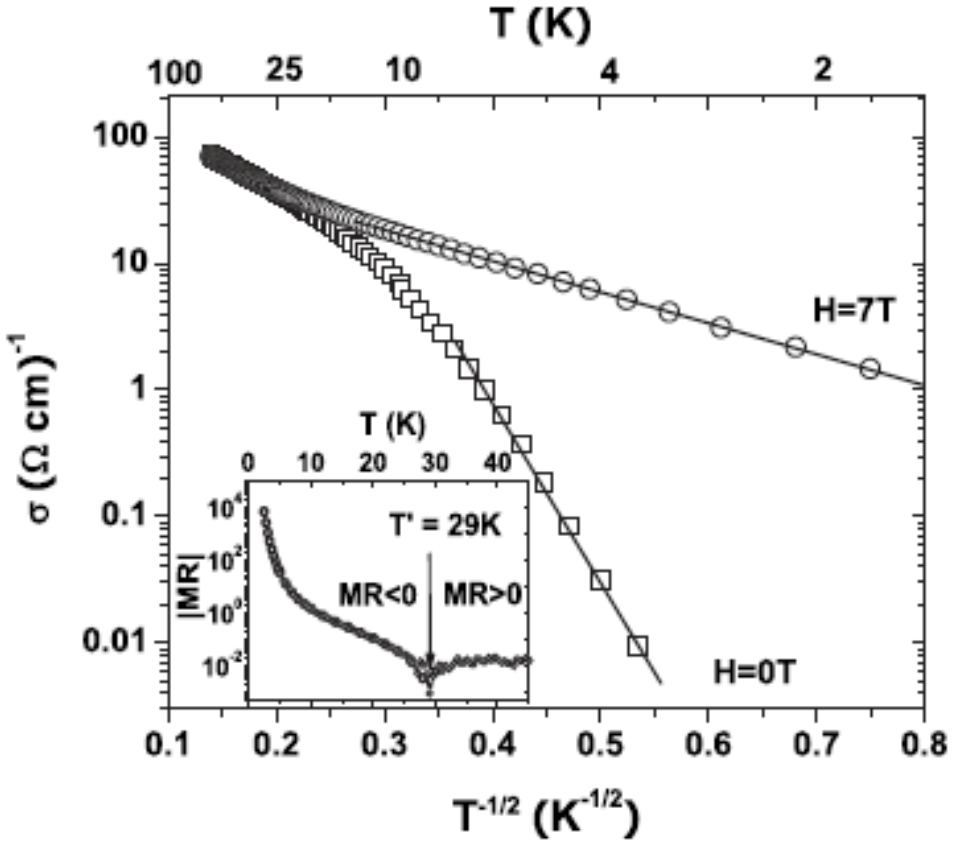
Seems to be an effect of band gap/screening – look at *a*-C



$\sigma(T)$ of various forms of a-Gd-C



Magnetoconductivity of a-Gd-C



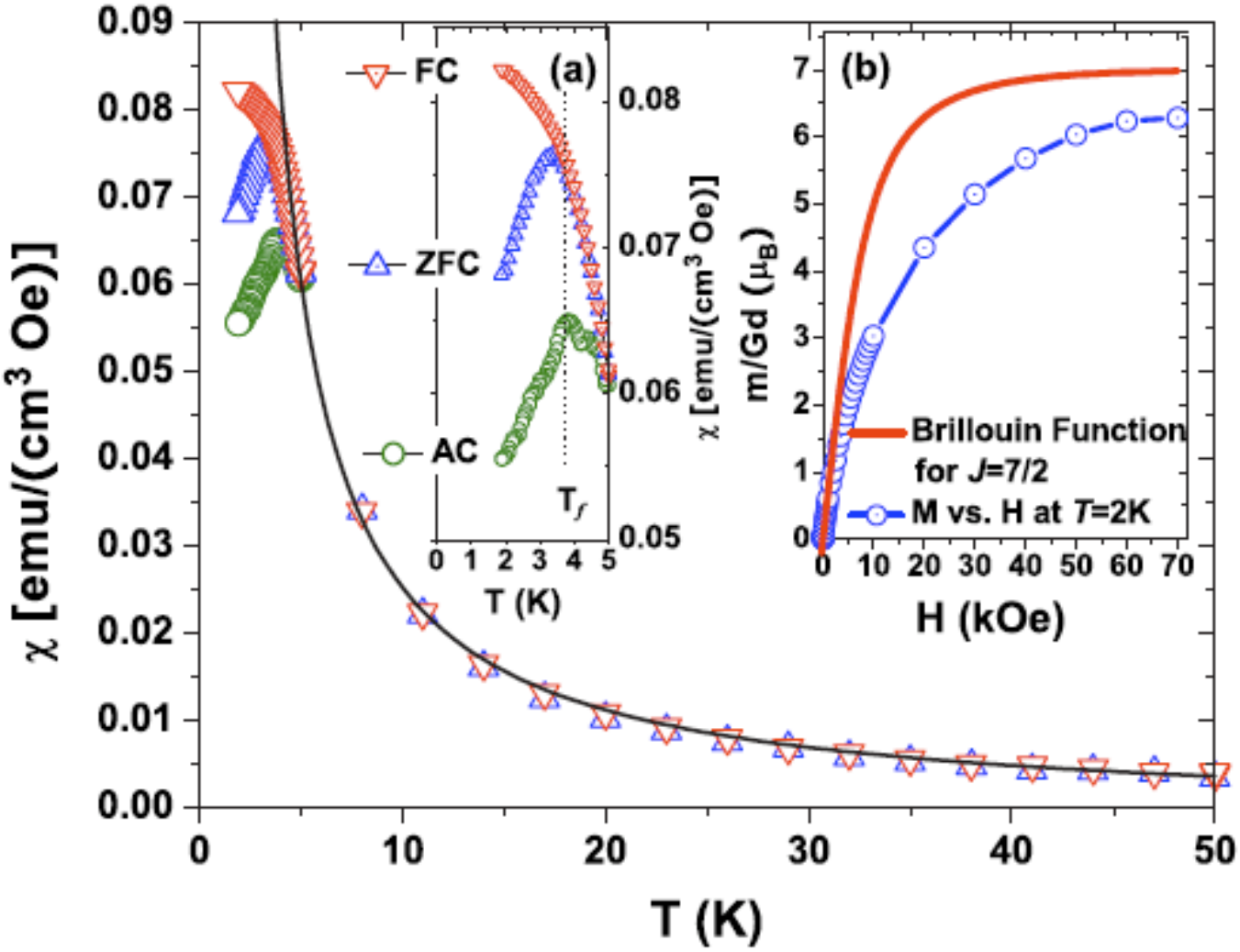
a-Gd-C
11 at.%Gd

Low T σ fits well to A exp $(-T_o/T)^{1/2}$

Magnetoconductance of a-Gd-C larger than a-Gd-Si at low T, but drops off faster with increasing T (is small and positive at high T)

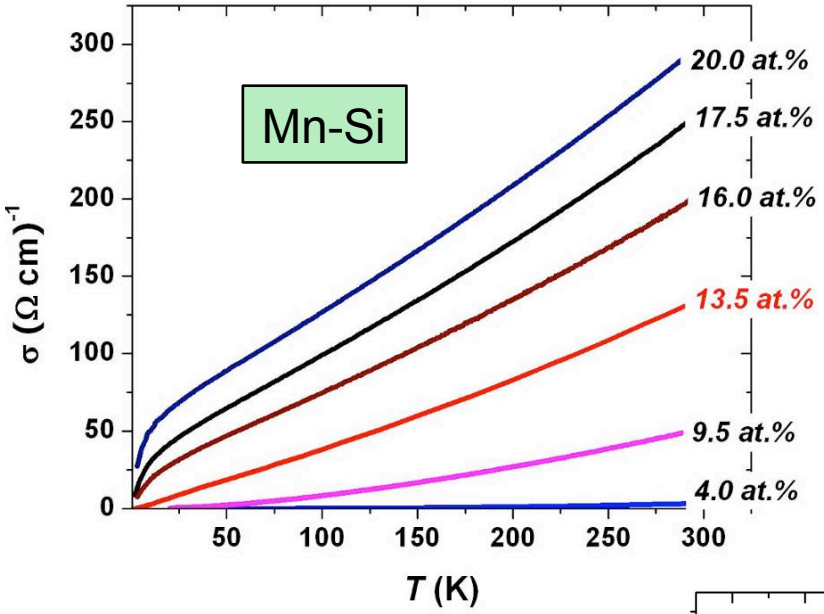
Also, low T large MG independent of x, unlike Gd-Si

Magnetization of a-Gd-C

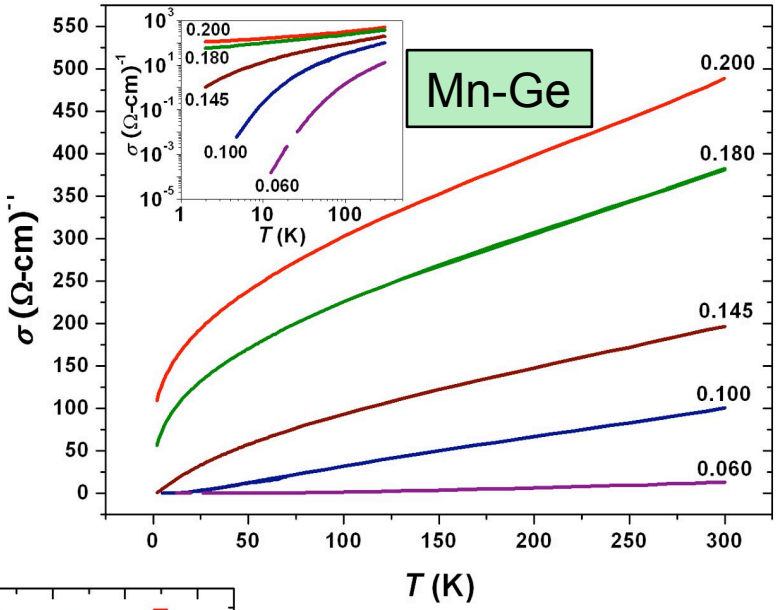


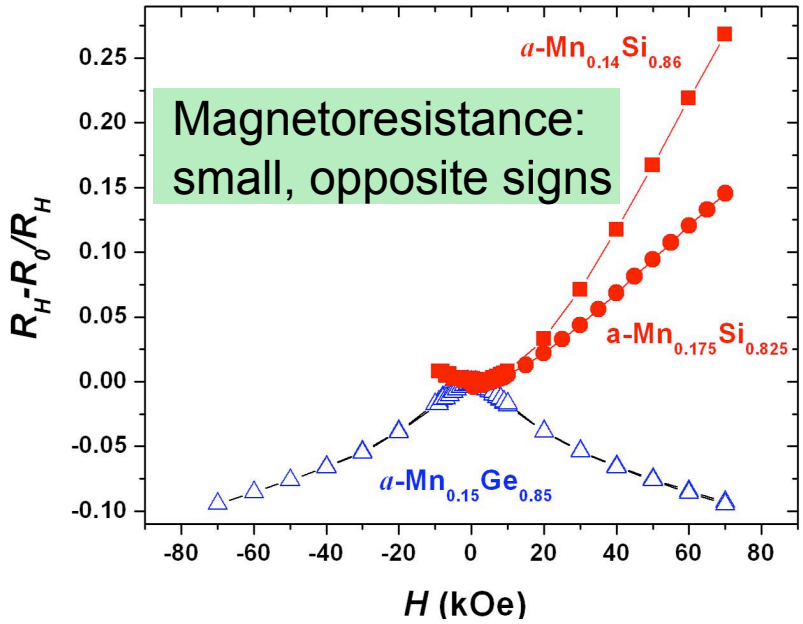
For all x and for all types of a-C :
Spin glass, with weaker Gd-Gd interactions and simple p_{eff} ~8.9μB and small θ

Amorphous Mn-Ge and Mn-Si



conductivity vs x,T similar



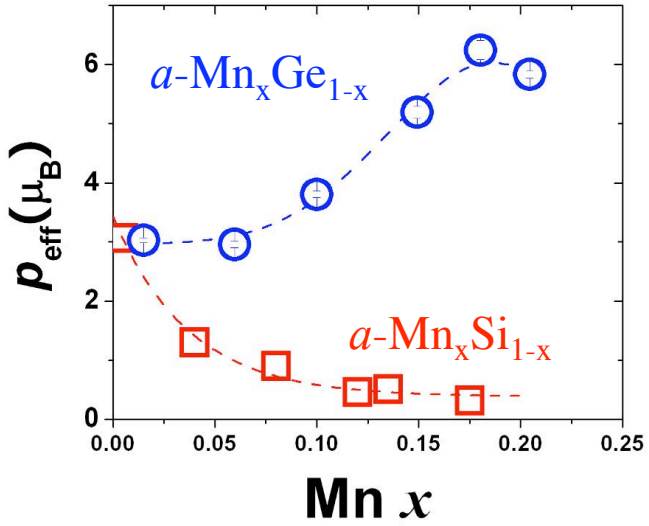


Summary of Amorphous Mn-Ge and Mn-Si

a-Mn-Si: σ(T) depends on Mn at.% (like Gd-Si) – means Mn acts as a dopant)

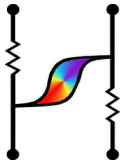
- nearly zero MR (like non-magnetic *a*-X-Si)
- *nearly zero magnetic moment*, no spin glass freezing: small paramagnetic susceptibility with 3 μ_B for <0.05 at.% Mn, down to <0.1 μ_B for >10 at.% Mn
- ESR shows g=2
- XAS shows broad metallic-like d-bands, not atomic multiplet states
- Negative Hall coefficient, density suggests interstitial-like Mn local environment

Perhaps a helical locally metallic AFM?



a-Mn-Ge*: Large moment! Spin glass freezing.*

- Strong AFM interactions – more than Gd-Si
- Small negative MR because M(H) is small
- XAS shows clear atomic multiplets
- Positive Hall coefficient, density suggests substitutional-like Mn local environment
- Why the *x*-dependence in p_{eff}? – perhaps different types of sites?



Summary – amorphous magnetically doped semiconductors

Transport: All amorphous Gd-Si, Ge, C and Mn-Ge alloys show enormous (orders of magnitude) negative magnetoresistance (MR) below a characteristic T*

Field-dependent correlated electron properties (Coulomb gap, optical $\sigma(\omega)$)

Low temp transport can be explained with field-dependent parameters including correlation and disorder effects, but no scaling with M/M_s, no quantitative model

Enormous effect of magnetic field H allows studies of M-I transition on a single sample as a function of x,T,H,ω (scaling analysis)

What controls the microscopic non-universal parameters (N_0, N_1, σ_0, σ_1)?

T largest for Gd-Si, decreases with increasing metallicity (Ge, increasing Gd or Y)*

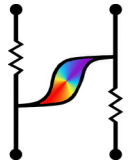
(T for a-Gd-C smaller because not really sp^3 bonded – higher $\sigma(T)$ than Gd-Si)*

What controls T, dependence on metallicity, band gap. Can we increase it?*

Magnetic properties: All (except *a*-Mn-Si) show strong frustrated magnetic interactions, with magnetization M(H) far below non-interacting Brillouin function, and spin glass freezing with T_f increasing with x. Low field χ(T) has unexplained properties.

- *Gd-C the most simple – p_{eff} as expected for Gd^{3+}; M(H,T) close to Brillouin function. T_f, θ increase with x*
- *Gd-Si shows exotic magnetic behavior at low H: large increase in χ at M-I transition*
- *Gd-Ge the most exotic at low H; $\chi \sim 1/T^{\alpha}$?*
- *Mn in a-Si has no moment – due to delocalized d-band. Perhaps a local helical magnet*

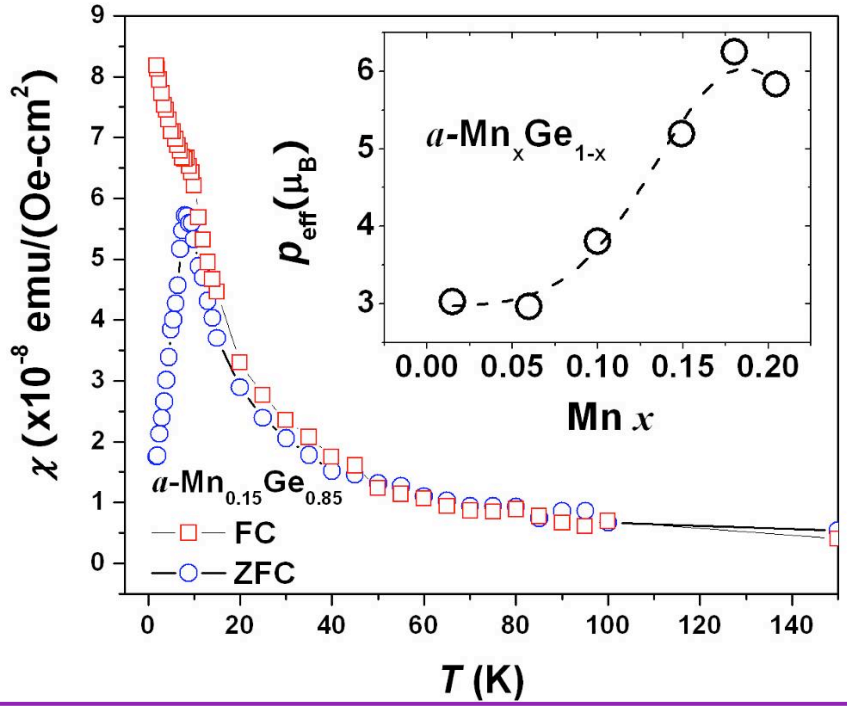
What happens to the RKKY-like interaction in the strongly disordered limit?

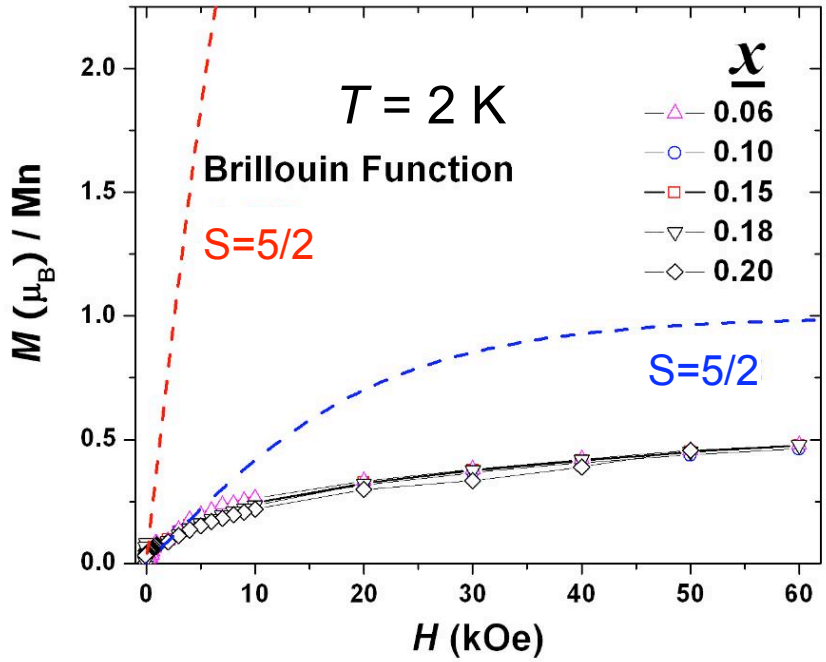


END OF TALK
Extra slides follow

Magnetization of a-Mn-Ge: large moment totally different than a-Mn-Si

S=1 to 5/2, strongly AFM/FM → spin glass

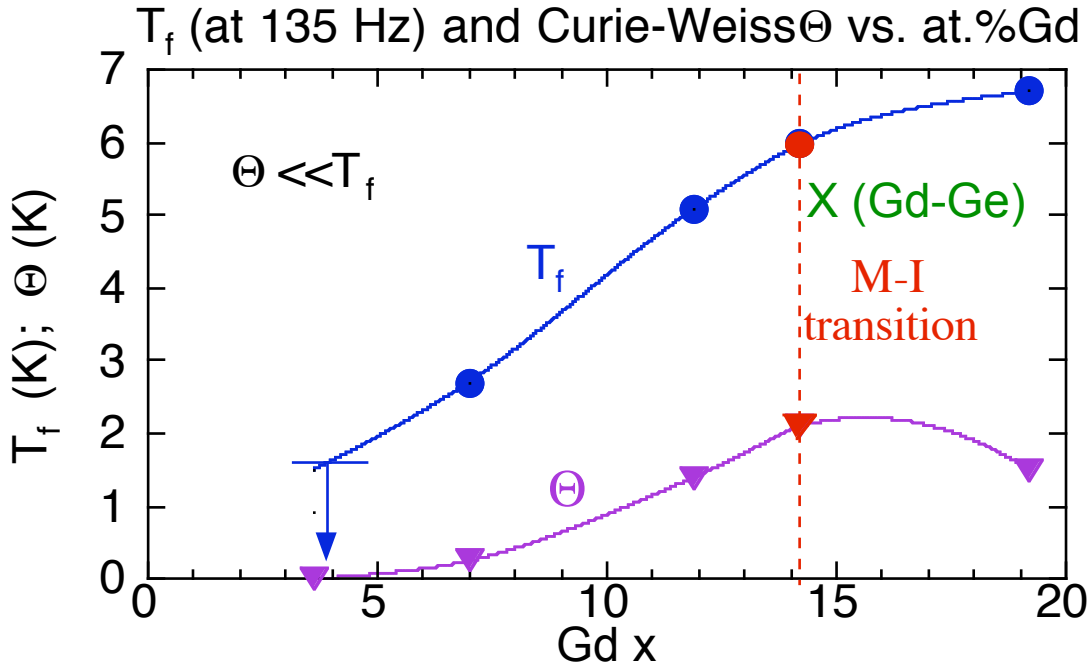


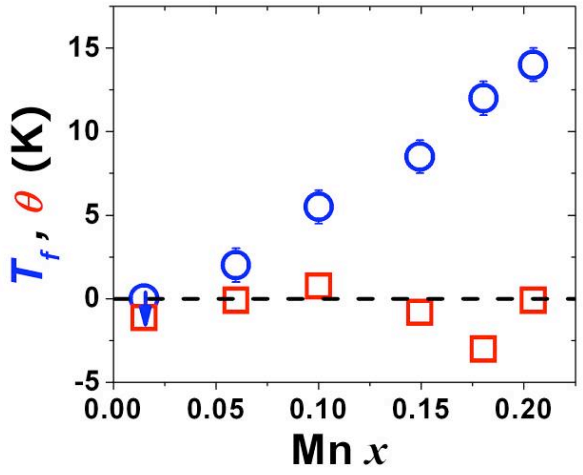


Spin-Glass freezing, large χ
Curie-Weiss fit above T_f
θ near 0 (as in *a*-Si), T_f increases with x
But, p_{eff} *large*, depends on x
S=1 gives p_{eff}=2.8
S=5/2 gives p_{eff}=5.9

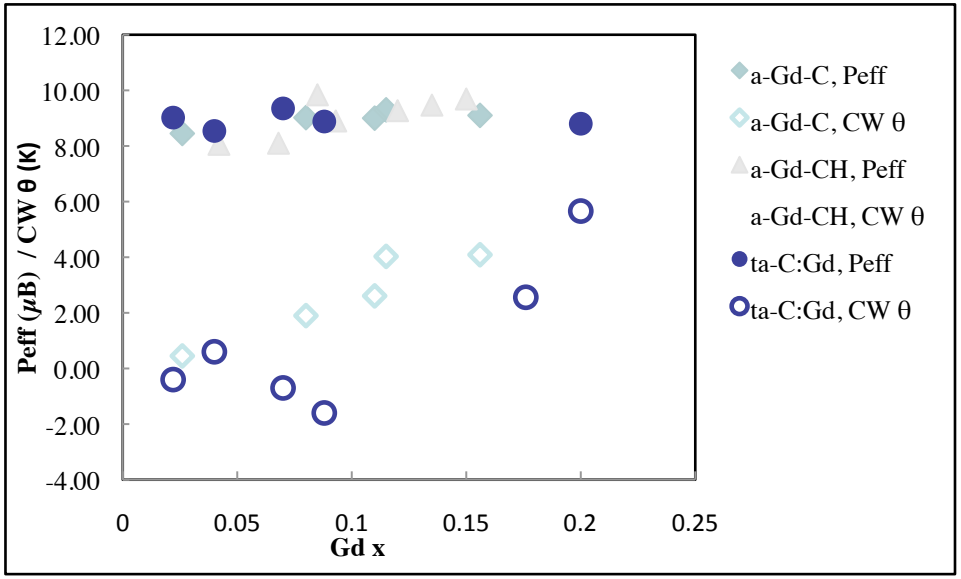
M vs *H* at *T* = 2 K not saturated at 6T
Shows strong AFM/FM interactions

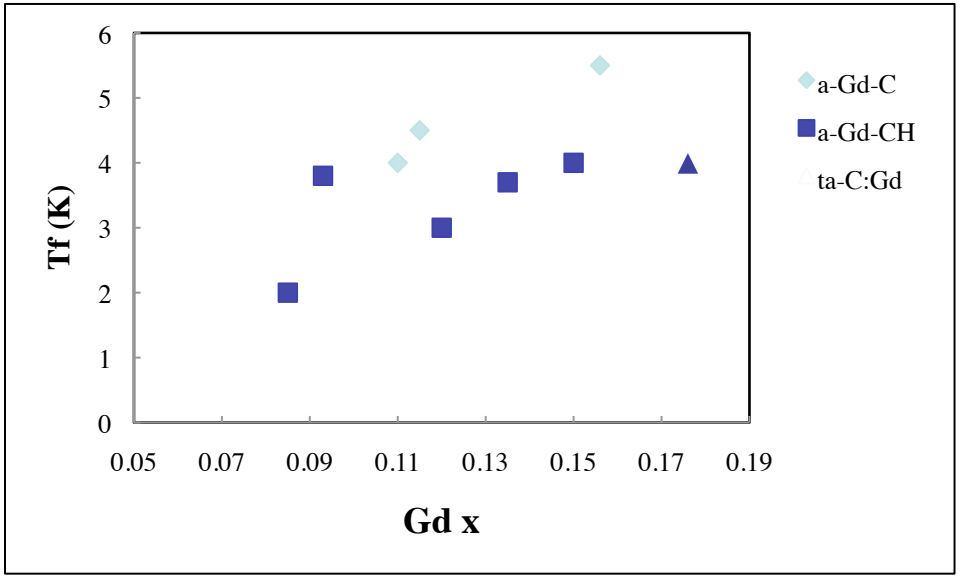
T_f and θ vs x for a-Gd-Si, a-Mn-Ge



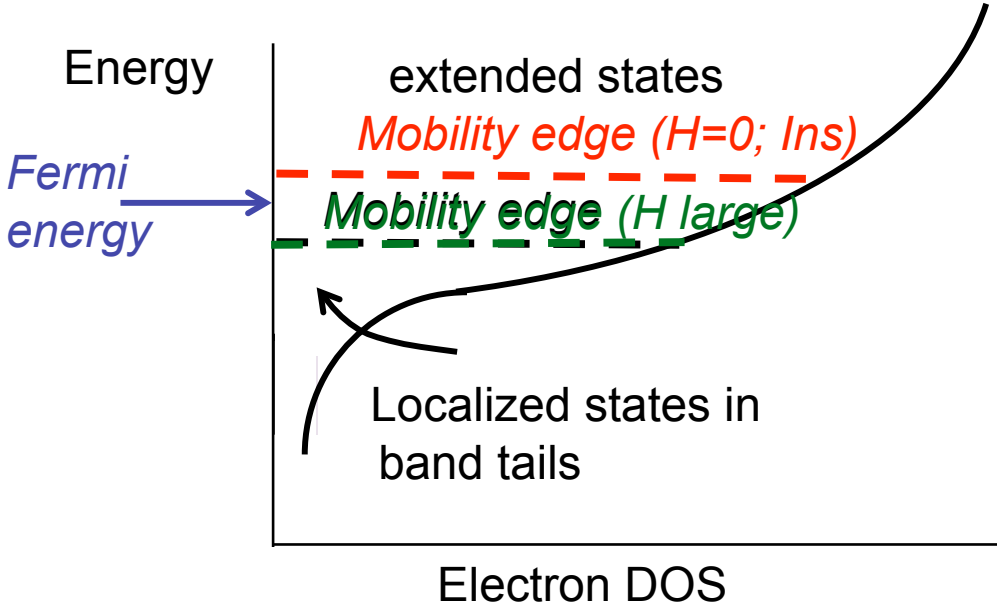


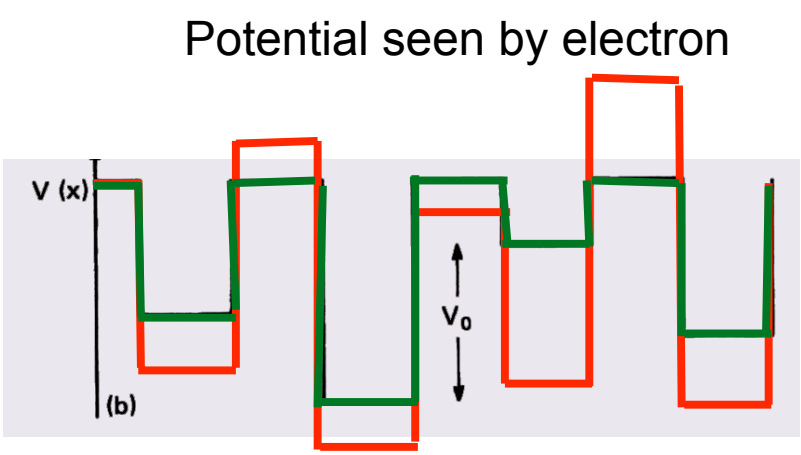
T_f (dc split only), p_{eff} and θ vs x for various a-Gd-C





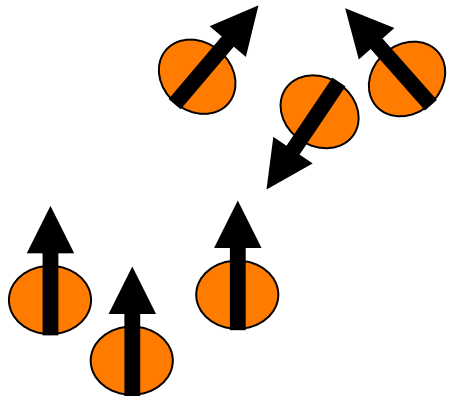
Magnetic Disorder Potential





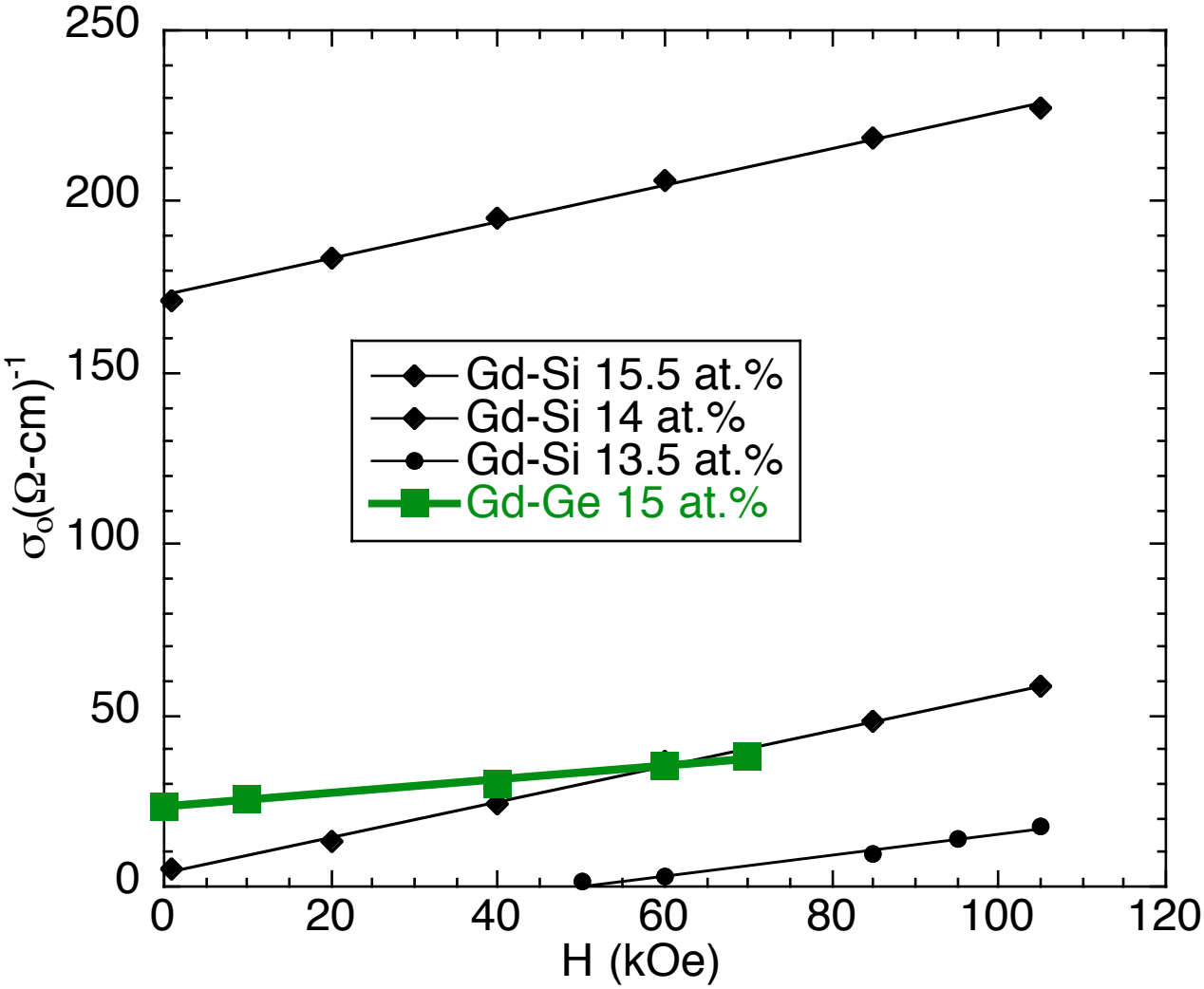
H=0: Gd spins randomly oriented
magnetic +structural disorder; $E_c > E_F$: insulating

H ⇒large: Gd spins aligned
structural disorder only; $E_F > E_c$: metallic

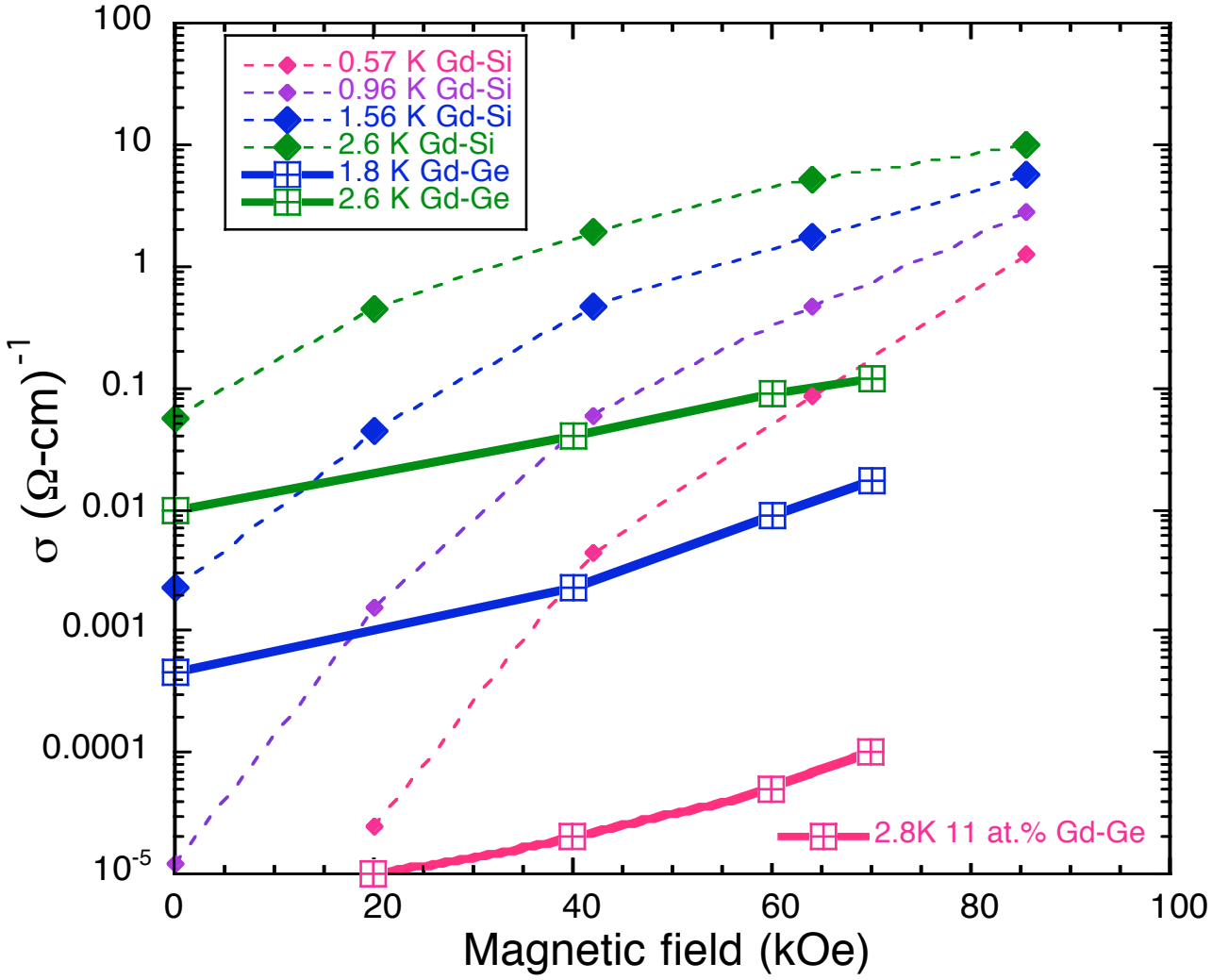


BUT- data does not show scaling with M/M_s = alignment angle of Gd moments. Likely because it doesn't include correlation effects

Field dependence of conductivity for metallic Gd-Ge << Gd-Si

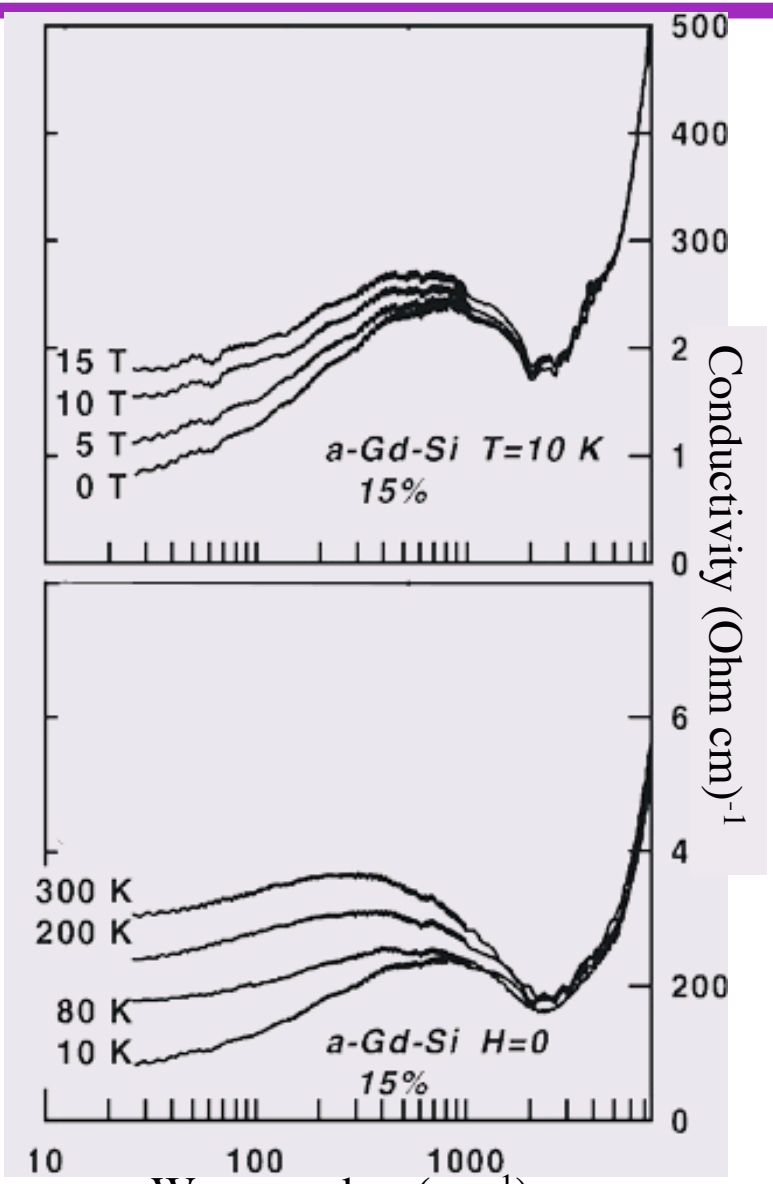


Field dependence of conductivity for insulating Gd-Ge << Gd-Si (as for metallic Gd-Ge)



So, MR is less for a-Gd-Ge than a-Gd-Si (even though M(H) is higher)

Optical/IR conductivity: sum rule "violations"



Si band edge absorption as expected
Very non-Drude form to sub-band gap absorption

Integral not conserved for *a*-Gd-Si out to Si band edge (1 eV)!
Depends on T and H
Spectral weight proportional to $\sigma(H,T)$

Y-Si alloys show similar effects with T but no field dependence

3D M-I transition physics

Use enormous magnetoresistance to study transport, Hall effect, electron density of states continuously through the M-I transition (hard to do in 3D usually)

- **Optical absorption**: Use Kramers-Kronig to get

  $\sigma(\omega)=\sigma_1(\omega)+i\sigma_2(\omega)$: spectral weight $\int_0^W \sigma_1(\omega)d\omega = \frac{\pi n_b e^2}{2m^*}$

  Here, non-conserving spectral weight to 1 eV with H or T

- **Scaling of conductivity and electron density of states**

  $\sigma \propto H-H_c$

  $N(0) \propto (H-H_c)^2$

  $\sigma \rightarrow 0$ as $N(0) \rightarrow 0$

  $\sigma^2 \propto N(0) \propto dI/dV(0)$

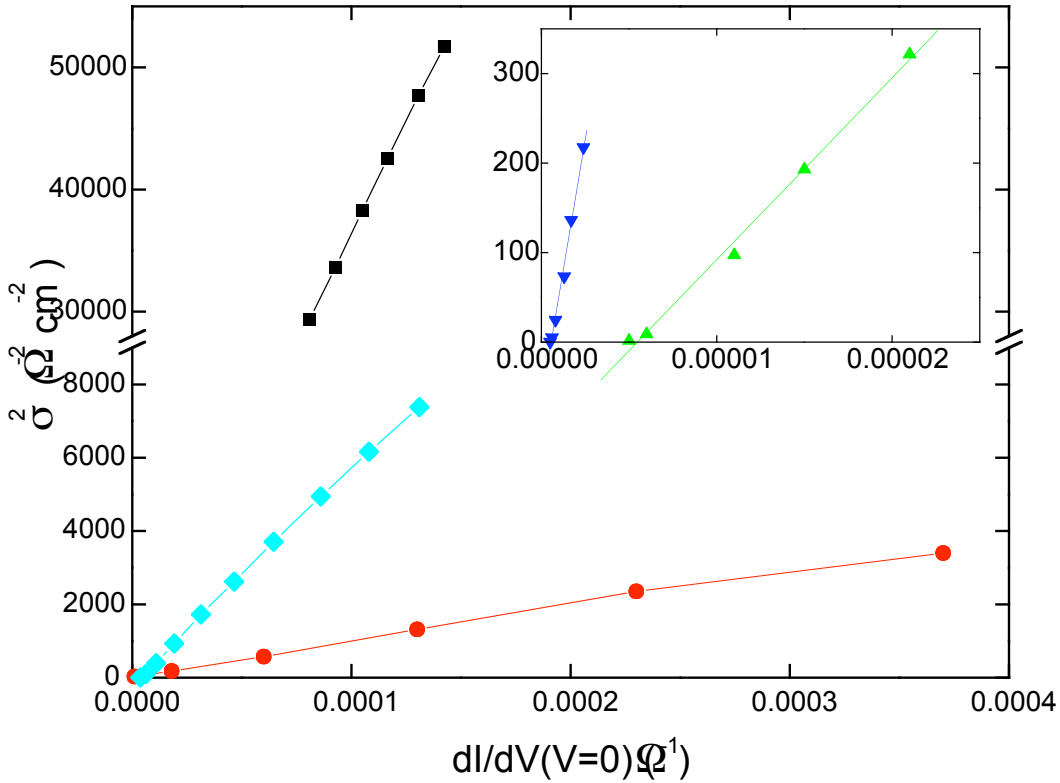
  (data taken at 100 mK)

- **Hall effect**:

  $R_0 \propto (H-H_c)^{-1}$;

  # mobile carriers $n \propto (H-H_c)$ (vanishes at M-I transition)

Scaling



T=100mK:

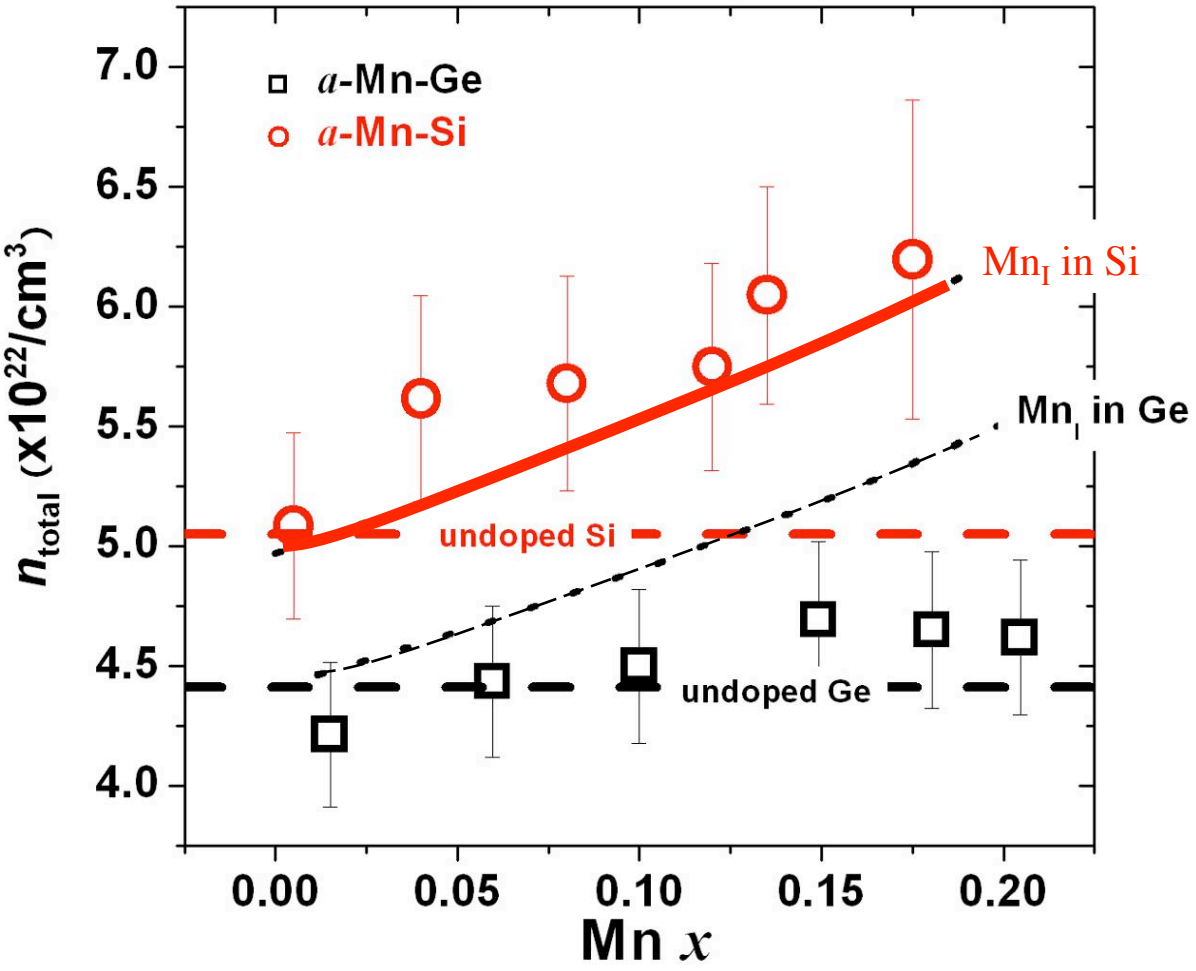
$\sigma \propto H-H_c$

$N(0) \propto (H-H_c)^2$

$\sigma \rightarrow 0$ as $N(0) \rightarrow 0$

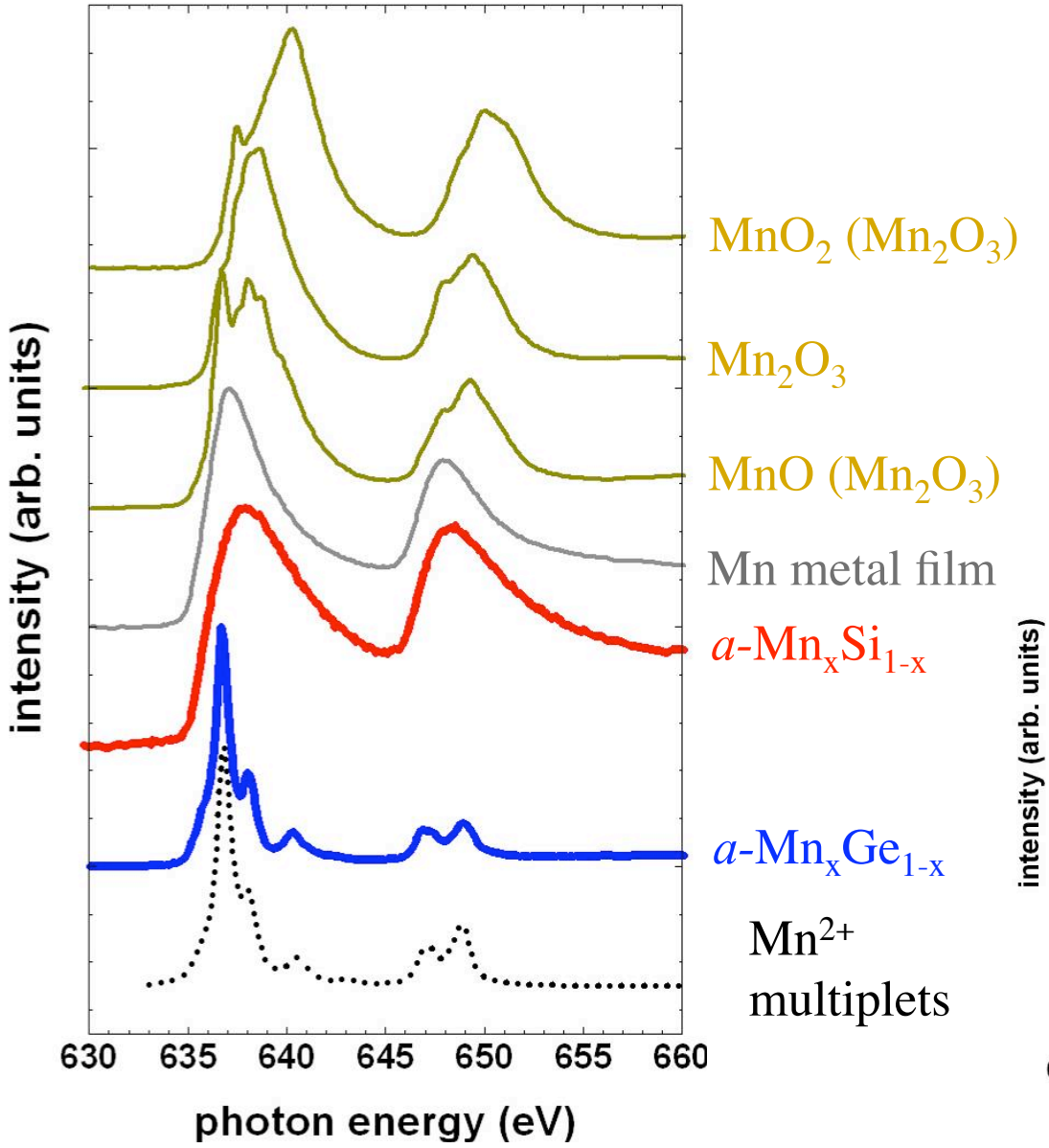
$\sigma^2 \propto N(0) \propto dI/dV(0)$

Total atomic density vs Mn at.% in a-Si and a-Ge



Mn in *a*-Si : Interstitial-like site (number density adds)
Mn in *a*-Ge : Substitutional-like site (number density constant)

X-ray absorption spectroscopy (XAS) on Mn $L_{3,2}$ edges

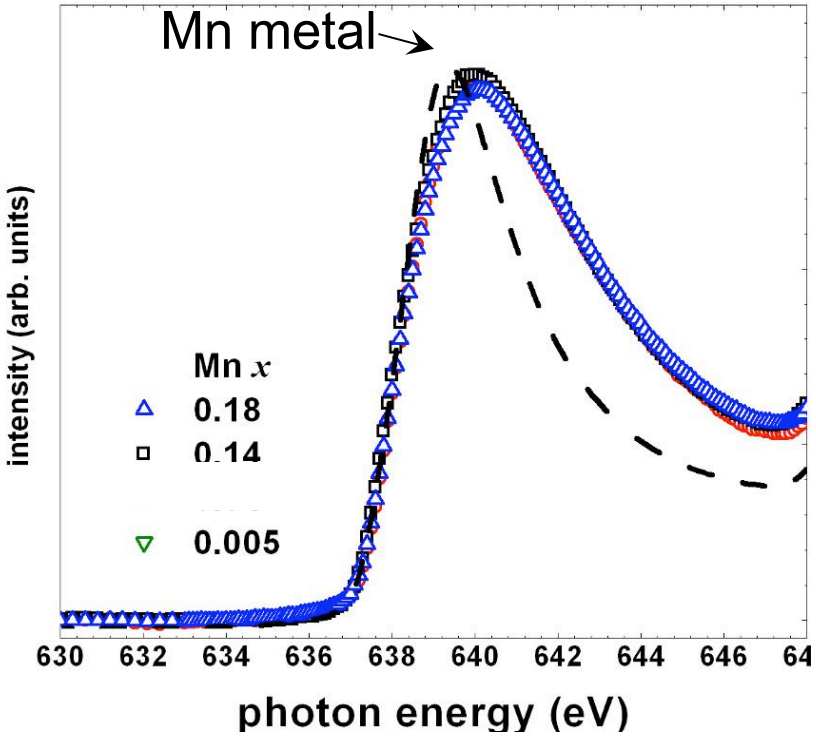


$L_3 : 2p_{3/2} \rightarrow 3d$
$L_2 : 2p_{1/2} \rightarrow 3d$

XAS probes element-specific electronic structures

L spectral shapes → 3*d* states

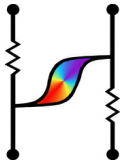
(Charge states → Spin states)



Ludwig-Woodbury Model for Mn in Si or Ge

	Substitutional (Mn in Ge)		Interstitial (Mn in Si)			
Ion configuration	Mn^{+} $3d^2$	Mn^{2-} $3d^5$	Mn^{2+} $3d^5$	Mn^{+} $3d^6$	Mn^{0} $3d^7$	Mn^{-} $3d^8$
Filling of 3*d* orbitals	t_2 ≡ e ::	t_2 ≡ :: e ::	e :: t_2 ≡ ::	e :: t_2 ≡ :: .	e :: t_2 ≡ :: ::	e :: t_2 ≡ :: :: ::
S	1	5/2	5/2	2	3/2	1
L	0	0	0	1	1	0
J	1	5/2	5/2	1, 2, 3	1/2, 3/2, 5/2	1

E. R. Weber: Appl. Phys A 30, 1-22 (1983)
G.W. Ludwig, H. H. Woodbury: Solid State Phys. 13 223 (1962)



Common Mn compounds

Compounds:

Mn_3Ge, fct, Ferri, T_C ~ 920 K

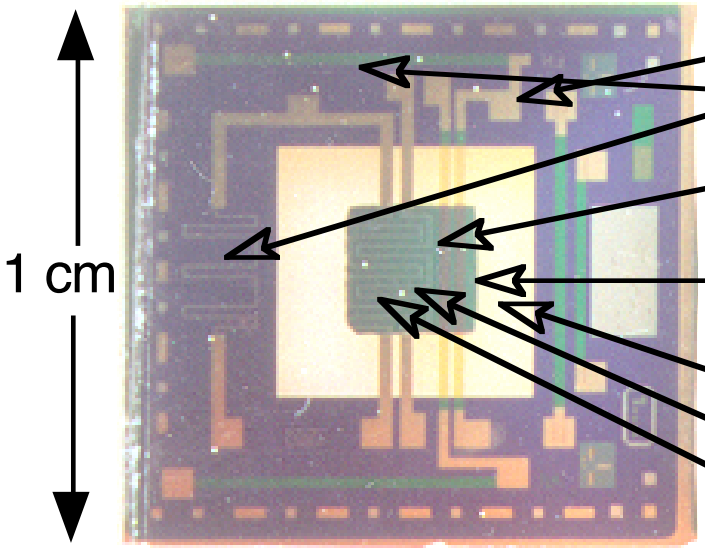
Mn_5Ge_2, Ferri, T_C ~ 710 K

Mn_5Ge_3, FM, T_C ~ 304 K

$MnSi_{1.7}$, tetragonal, weak itinerant FM, T_C~47 K, paramagnetic at RT

MnSi, Helical AFM

Micro/nanocalorimetry: overview (fabricated in the UCB Microlab)



Pt contact pads for electrical bonding
matching thermometers on Si frame
low temp thermometers (a-Nb-Si or B:poly Si)
sample and thermal conductivity layer (on back of membrane)
α-SiN membrane 180 nm thick (new: 30 nm)
heater (Pt)
high temp thermometer (Pt)

Allows us to make unique measurements: heat capacity of

- μg and sub-μg (films ~100 nm thick)
- Evaporated/sputtered films; powders; tiny crystals;
- Wide temperature range 1-500K (to date)
- Magnetic field (0-8T to date)
- *in situ* measurements

Related measurements: Thermal conductivity, thermopower

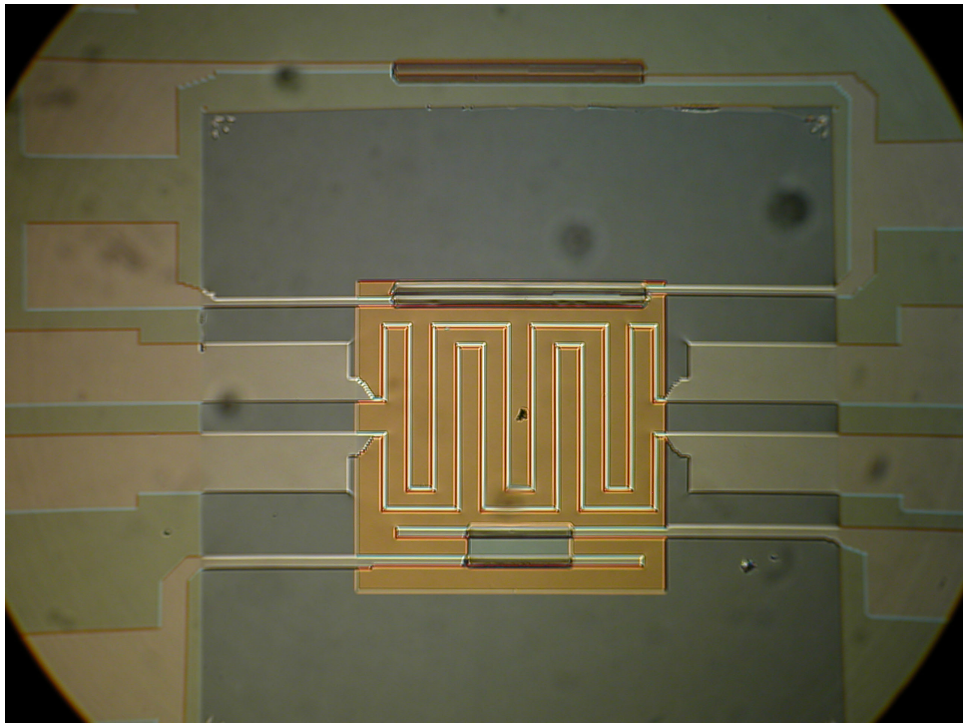
APS-Keithley Instrumentation Award 2006

D. W. Denlinger, E. N. Abarra, K. Allen, P. W. Rooney, S. K. Watson, F. Hellman , "*Thin film micro-calorimeter for heat capacity measurements from 1.5 K to 800 K*", Rev. Sci. Instr. **65**, 946 (1994);

D.R. Queen and F. Hellman, "*Thin film nanocalorimeter for heat capacity measurements of 30 nm films*", Rev.Sci.Instr. **80**, 63901 (2009).

Nanocalorimeters

Photo of nanocalorimeter: 30nm thick Cu on the back



Scaled down 5x in all important dimensions; membrane thickness 30 nm
Sample can be 30 nm and still get ~2% accuracy
(or thinner with less accuracy)

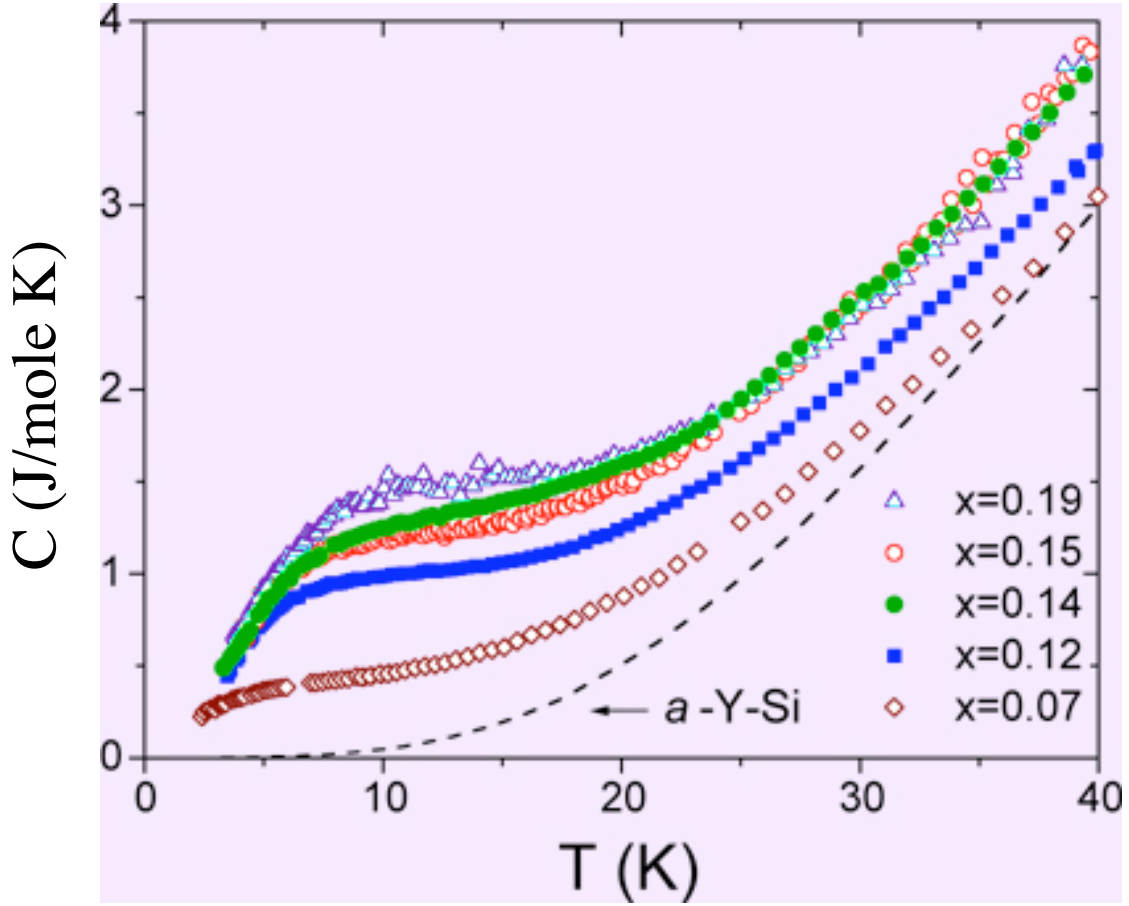
Specific heat of a-Gd-Si

Spin glass freezing gives large signature

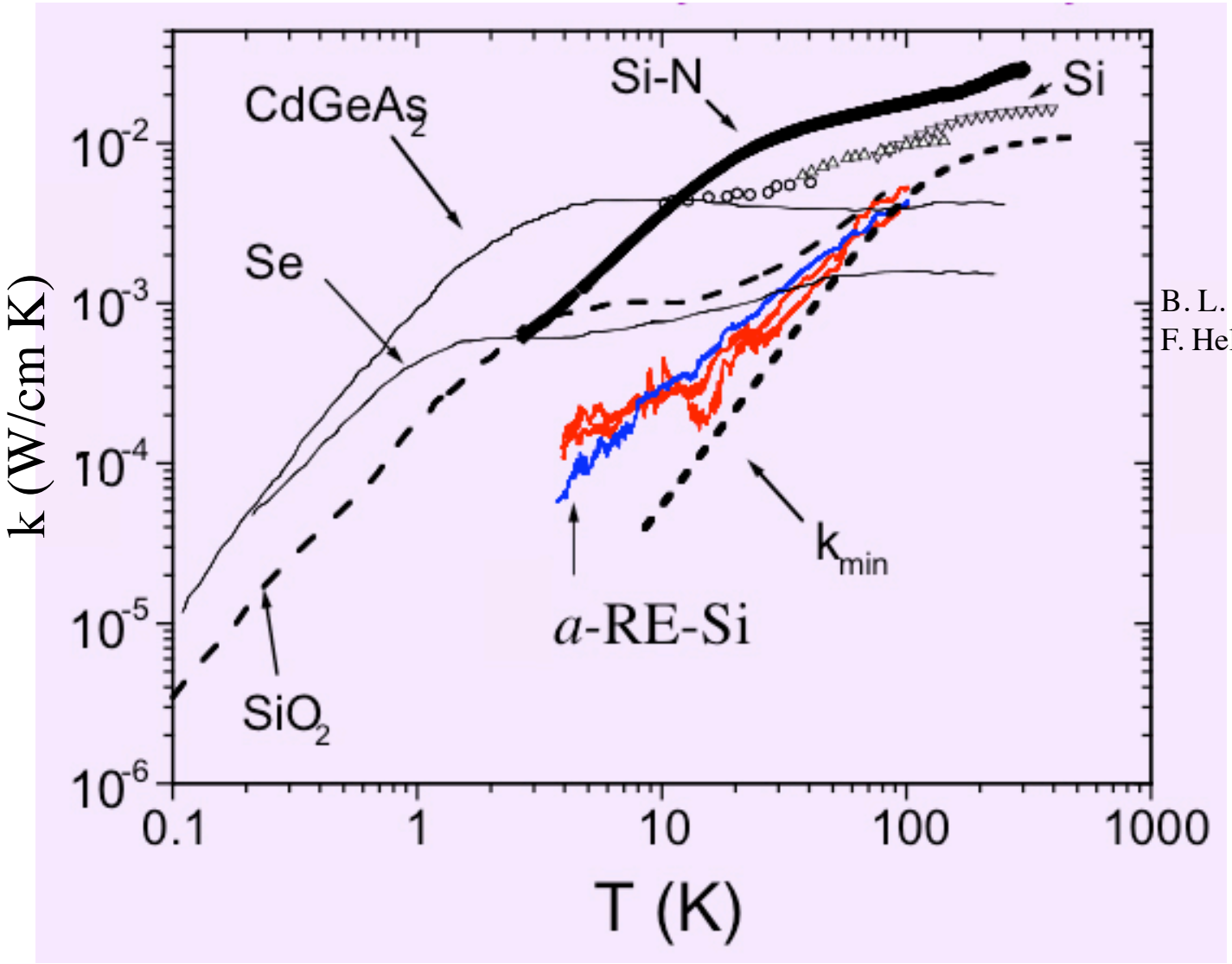
Has more entropy than Rln2J+1= Rln8

Due to conduction electron spins??

Excess heat capacity (Gd-Si minus Y-Si) persists to high T like MR



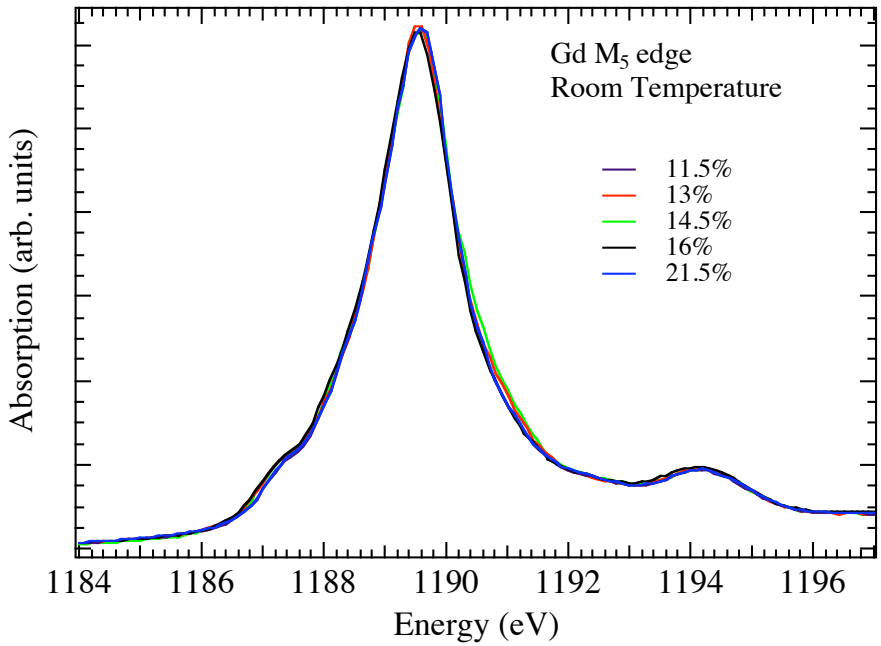
Thermal Conductivity of a-RE-Si alloys (sample covers whole membrane area: k from κ)



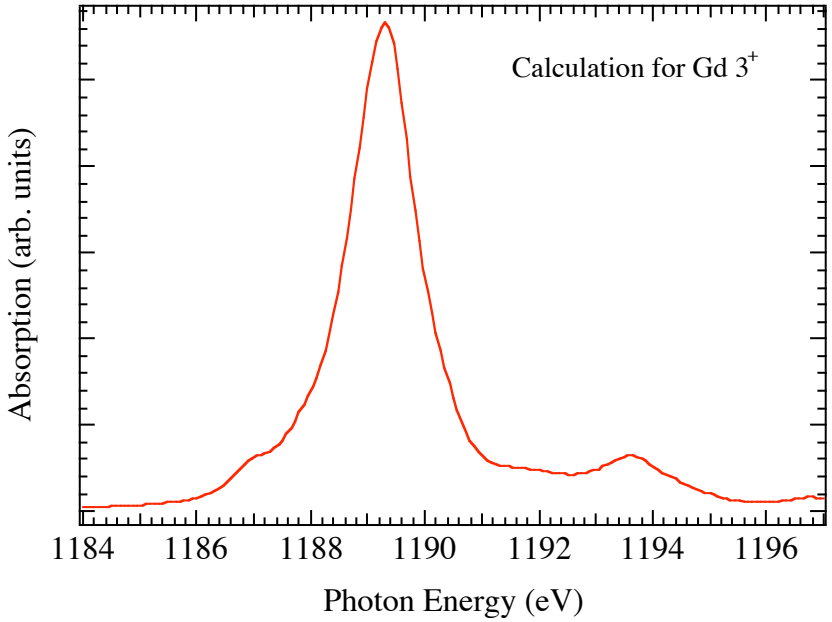
B. L. Zink, B. Revaz, F. Hellman, PRB

In analogy to filled skutterudites, RE “rattles” in Si cage, reducing k

Is Gd really in 3+ state?
Yes - XAS M edge measured at 300K

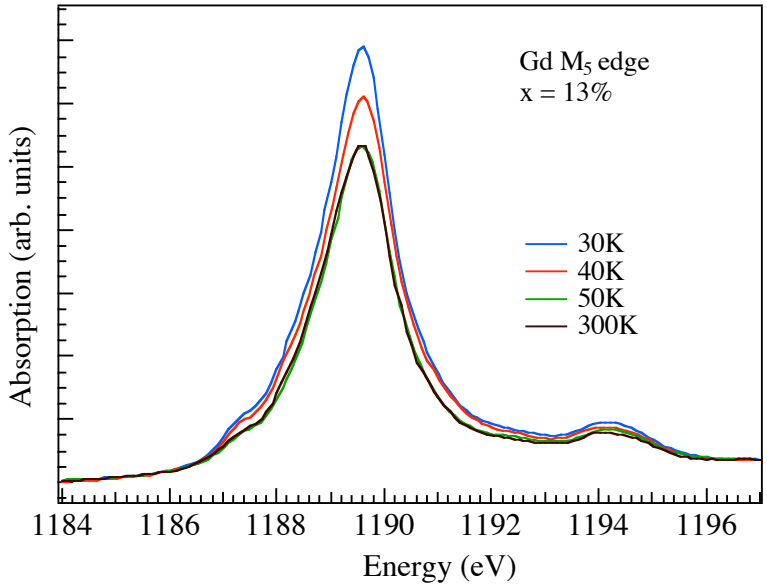


Gd M_5 x-ray absorption at 300K for various compositions. All spectra identical and consistent with Gd in 3+ valence state.

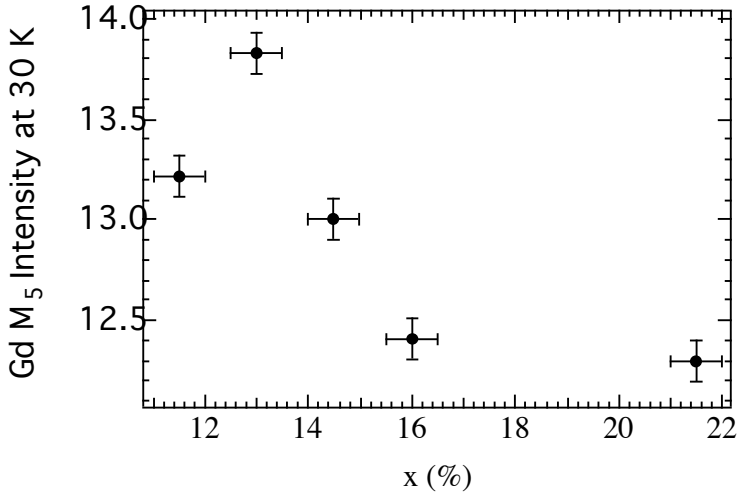


Calculated spectrum for Gd^{3+}. Atomic level Hartee-Fock calculation includes Coulomb interactions in 4f shell and between 4f shell and 3d core hole; spin-orbit coupling of 4f and 3d levels. Agreement with data is excellent.

Temperature dependence of XAS M edge



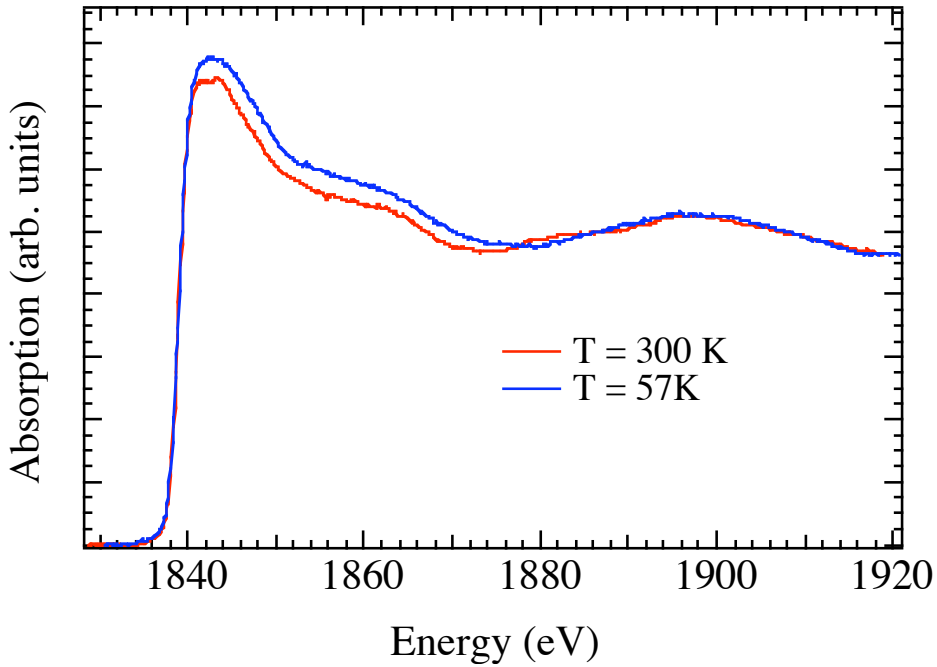
Gd M_5 x-ray absorption for x=0.13 for various temperatures, normalized to background far from peak. Data taken with left and right circularly polarized light are identical indicating no magnetic component at zero field.



Relative M_5 peak height at 30K as a function of Gd composition, x. Note peak at 14 at. % Gd, the Metal-Insulator transition

3d-4f absorption is due to atomic transitions, and cannot have temperature dependence without a shift in peak shape. Suggest instead BACKGROUND is temperature dependent, due to small but non-zero contribution of 3d-6p transition.

Temperature dependence of XAS Si K edge

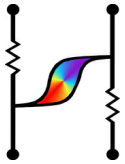


Si K edge x-ray absorption for x=0.13 for two block temperature

Si K-edge data suggests temperature dependence of 1s to unoccupied 3p states.

This is consistent with the suggested shift of background in the M edge absorption.

Interpretation: localization produces a shift in occupation from Si p-states to Gd p-states with decreasing temperature



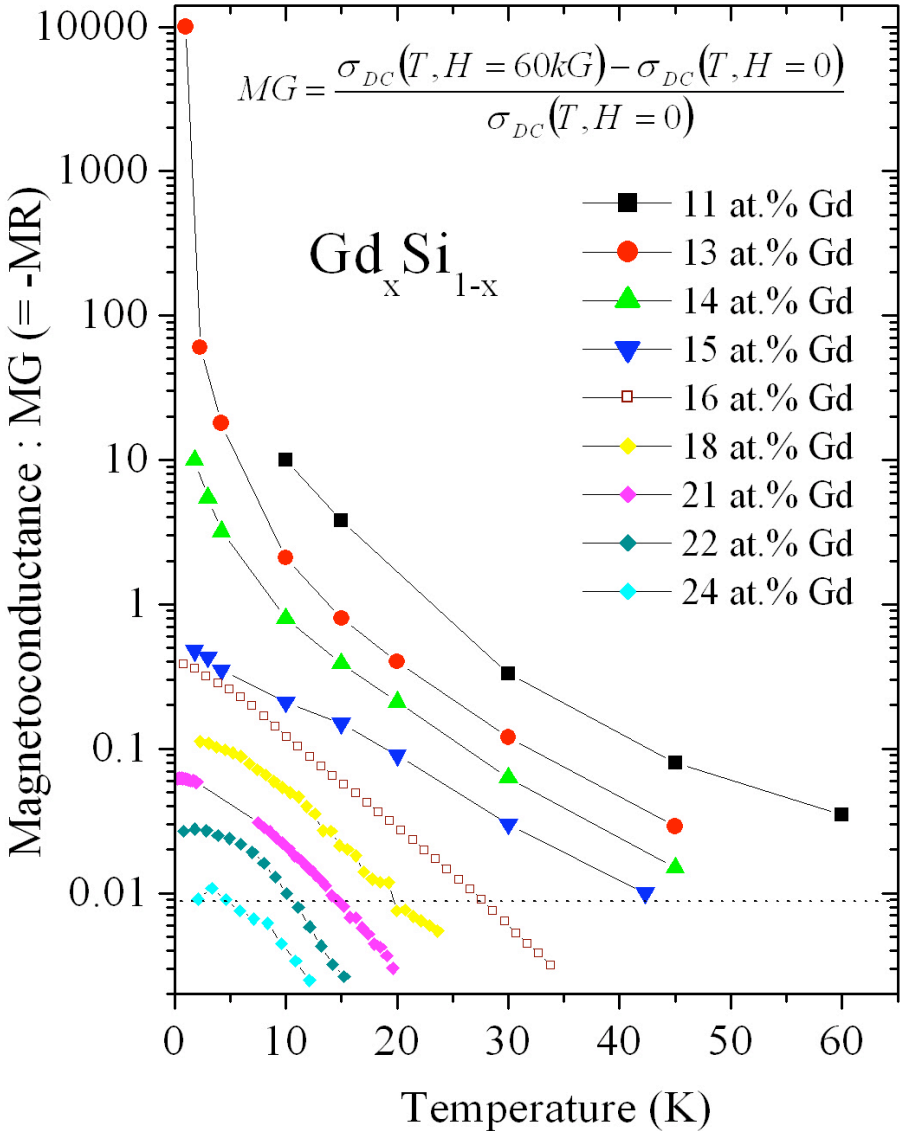
What controls the energy scale associated with T?*

- If T* were associated with a single electron-local moment interaction ($\mathcal{J}_{sf}$), it would be independent of Gd at.%
- If T* were associated with the Gd-Gd magnetic interactions, it should increase with increasing Gd
- Unlikely to be Kondo effect physics: J=7/2 and T_K should be very low and should increase with increasing Gd at.%
- T* *decreases* with increasing metallicity or decreasing band gap
- Suggestive of critical role of electron-electron interactions and screening
- As temperature is reduced, Coulomb gap develops which reduces the screening of the local moments, causing them to interact more strongly with the carriers. More metallic, lower band gap, more screening, so T* is reduced.

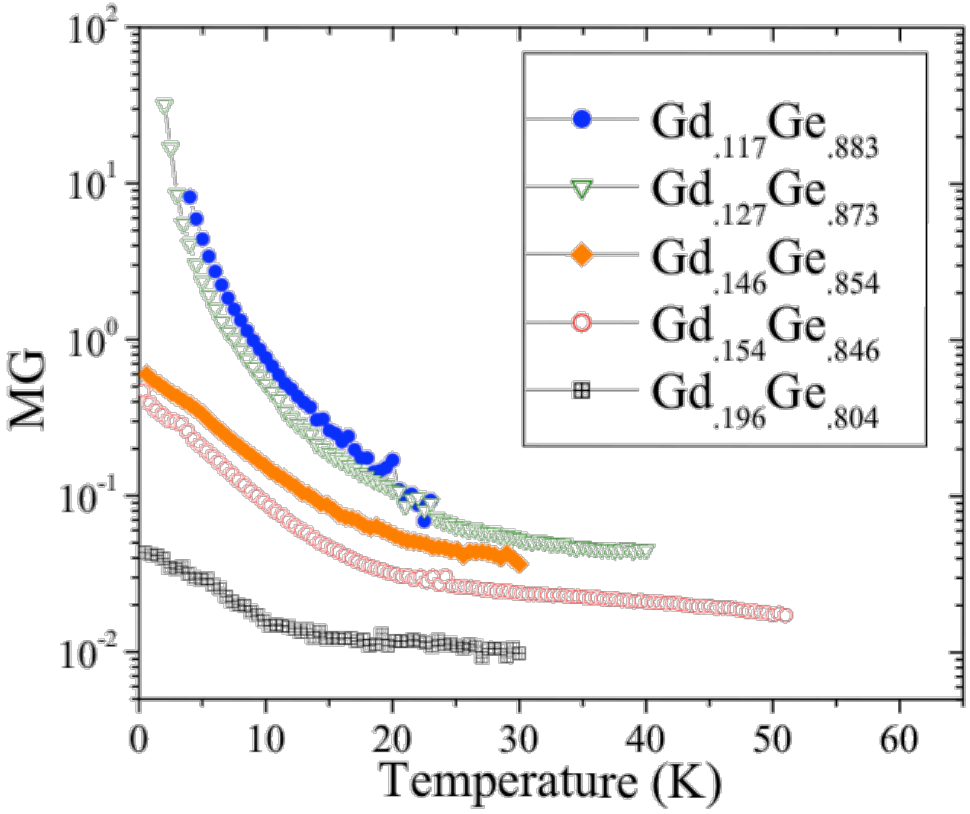
Amorphous vs crystalline magnetic semiconductors

- Amorphous alloys easy to make as films (co-deposition)
  - Easy to incorporate ions with large magnetic moments
- Much stronger disorder
  - But disorder crucial in all doped semiconductors
- Much higher electron concentration at Metal-Insulator transition
  - Same physics seen at and near the transition
  - *All* energies in problem correspondingly larger
  - "Low temperature" properties persist to higher temperature
- Gd-Gd interactions are mixed ferro/antiferromagnetic (RKKY)
  - Spin glass freezing instead of ferromagnetism
  - Magnetic and electrical transport properties show large effects of local moments on carriers and vice versa

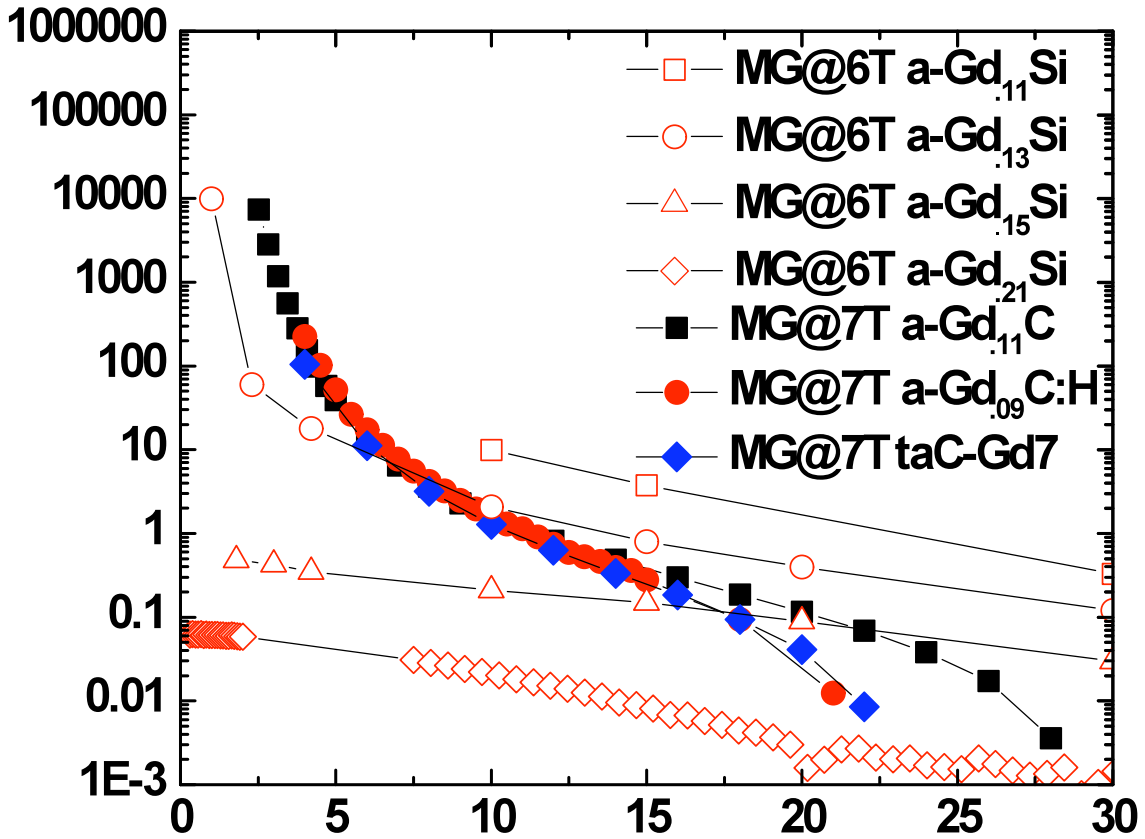
Magnetoresistance Gd-Si compared to Gd-Ge



E. Helgren, J. Cherry, L. Zeng, and F. Hellman, Phys. Rev. B (2005) and (2007)

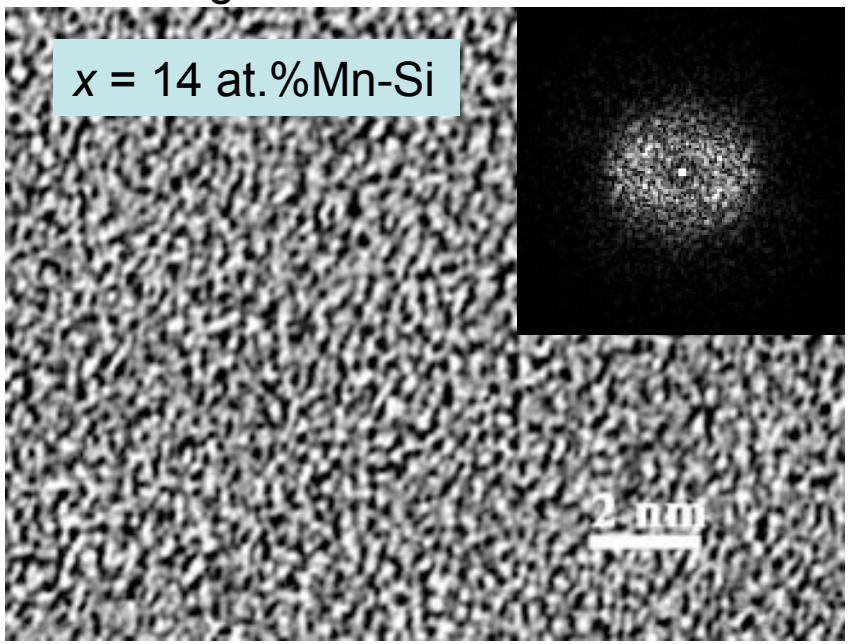


Comparison of MG for a-Gd-C and a-Gd-Si

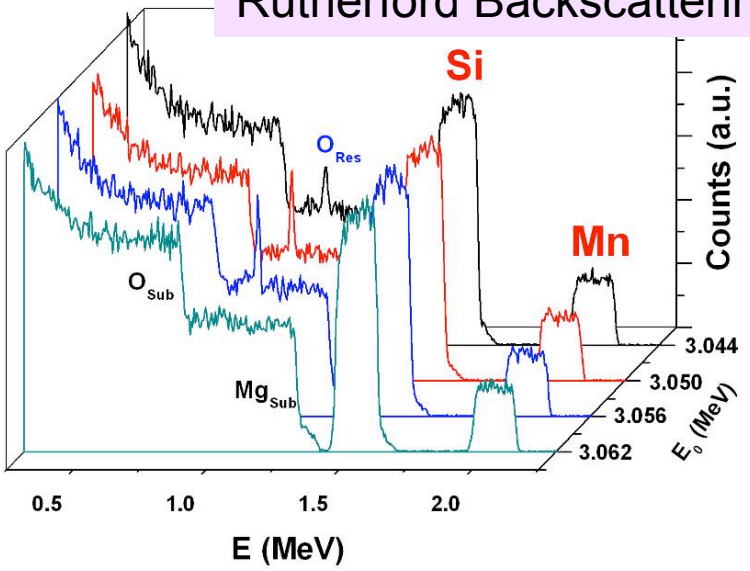


Amorphous Mn-Si, Mn-Ge

High resolution TEM



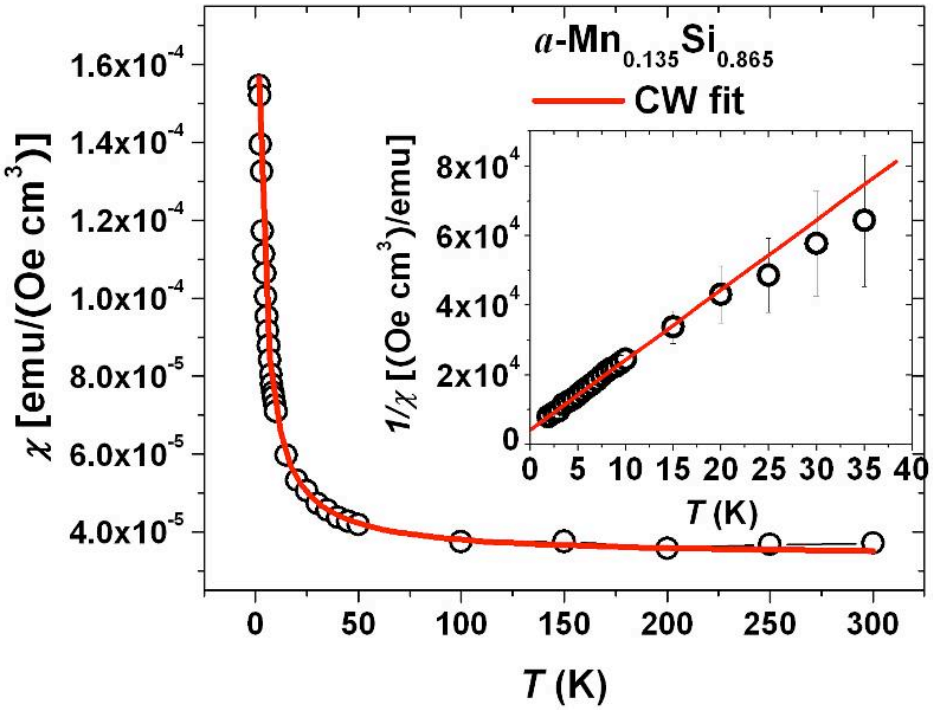
Oxygen resonant Rutherford Backscattering



Amorphous, no sign of clustering, inhomogeneity or oxygen contamination

Magnetization of a-Mn-Si

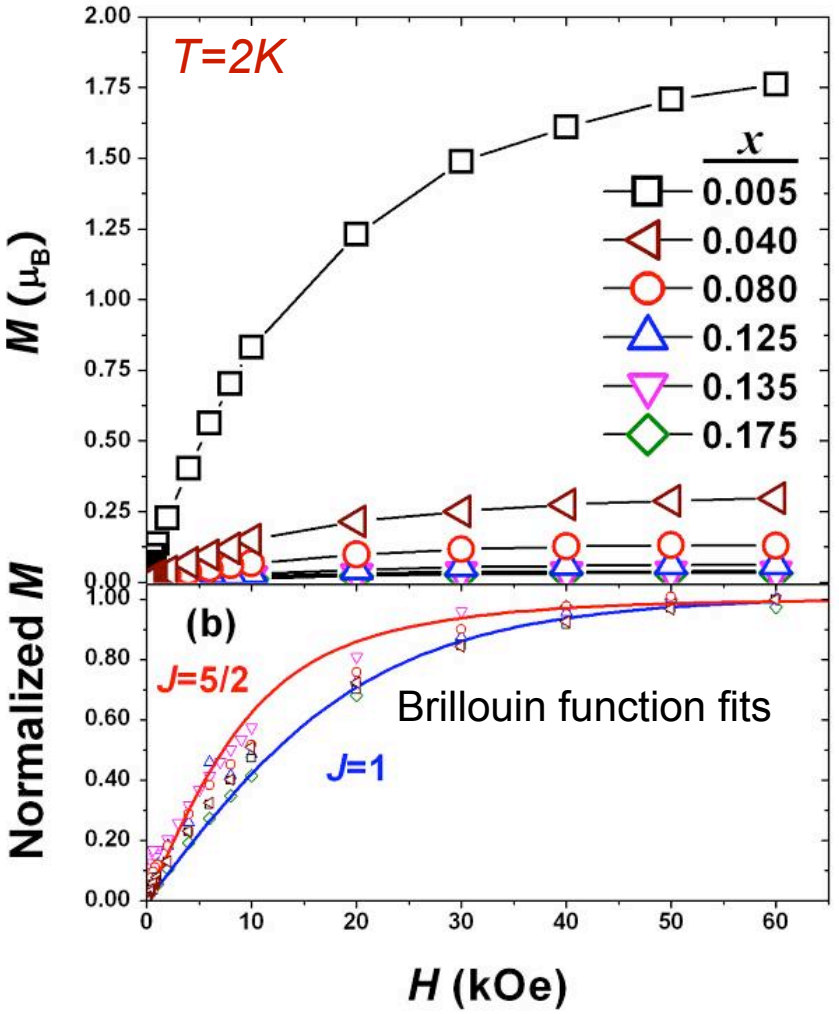
Mn moment is quenched!



Curie-Weiss law fit: $\chi = A/(T-\theta)$; θ near 0K

$A = n_{Mn} p_{eff}^2 \mu_B^2 / 3k_B$; effective moment $p_{eff} = (g^2 J(J+1))^{1/2} = 5.9$ for usual Mn^{2+} ions

Here, $p_{eff} = 0.5\ \mu_B$

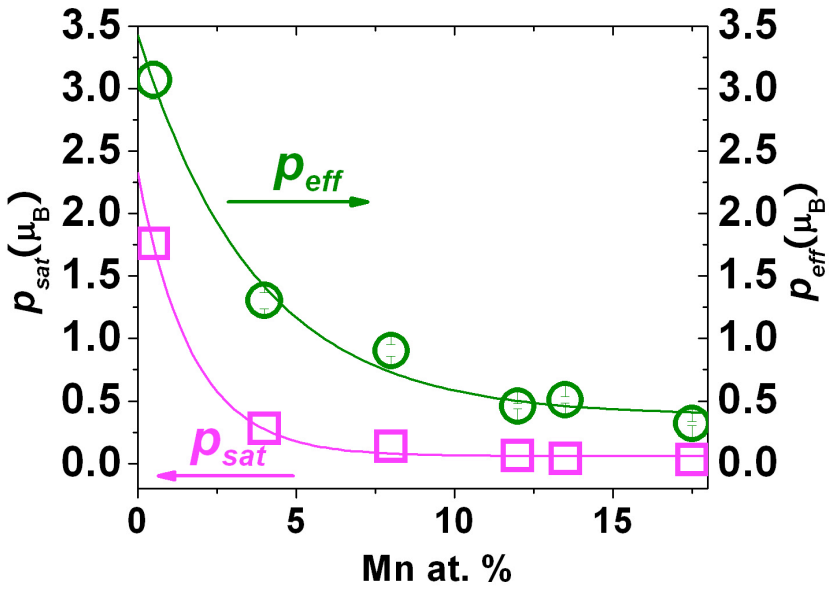


Similarly for low T (2K) M(H)

$p_{sat} = 0.1\ \mu_B$ for x=0.135

Li Zeng *et al.*
PRB 77, 073306 (2008)

Very low magnetization of a-Mn-Si



Above assumes all Mn contribute equally

Instead we suggest there is a tiny fraction (<5% for x>4 at.%Mn) that is magnetic (best fit to Brillouin function with J=5/2), and the rest are in non-magnetic states

DC Transport

a-Gd_xC_{1-x}(:H)

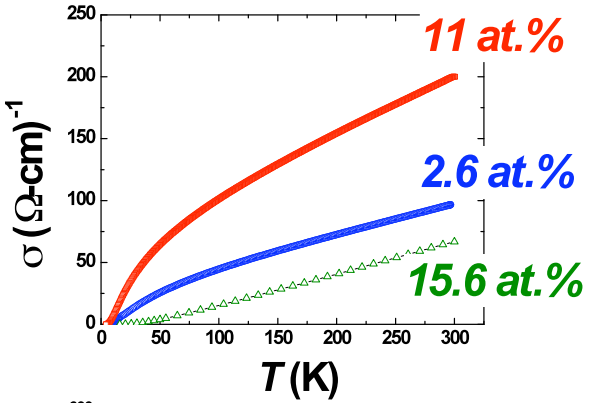
σ increases with x monotonically up to 11 %
and then decreases.
two-channel conductivity: change of a-C matrix

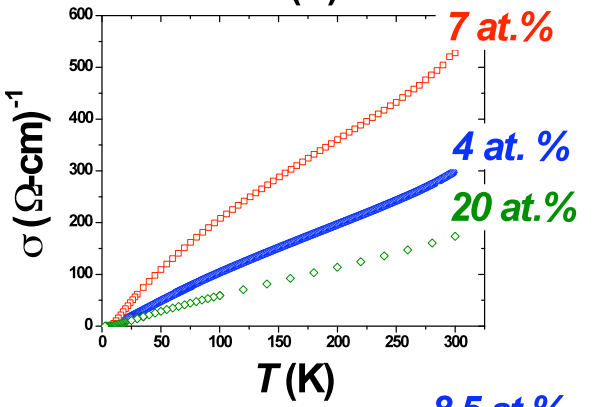
ta-C:Gd_x

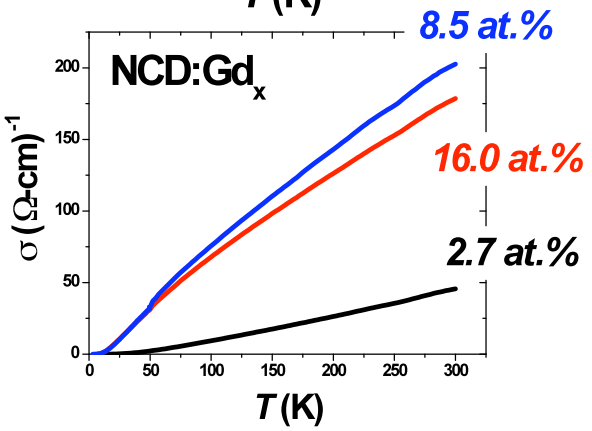
σ increases with x monotonically up to 14 %
and then decreases.
two-channel conductivity: change of *a*-C matrix

NCD:Gd_x and MCD:Gd_x

σ increases with x monotonically up to 8.5 %
and then also decrease
two-channel conductivity: change of *a*-C matrix



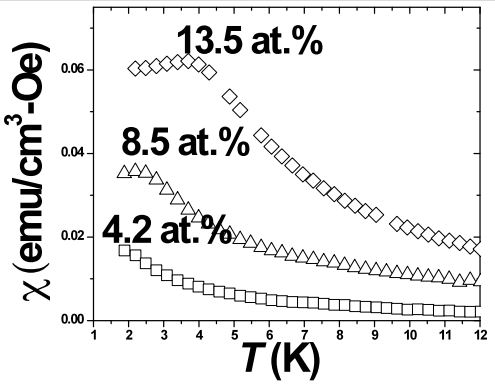




Magnetization

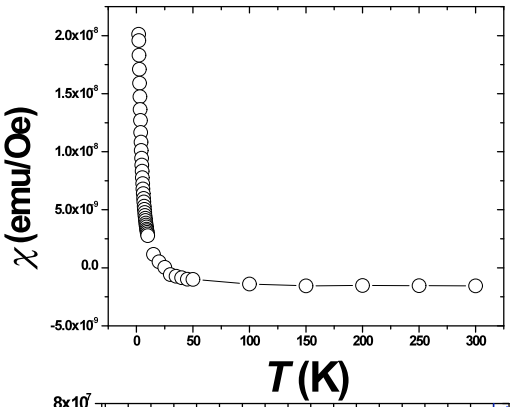
a-Gd_xC_{1-x}(:H)

Spin glass (SG) – like *a*-Gd_xSi_{1-x} and *a*-Gd_xGe_{1-x}
T_f increases with Gd at.%
Follows Curie-Weiss (CW) law, p_{eff} ~*8* μ_B



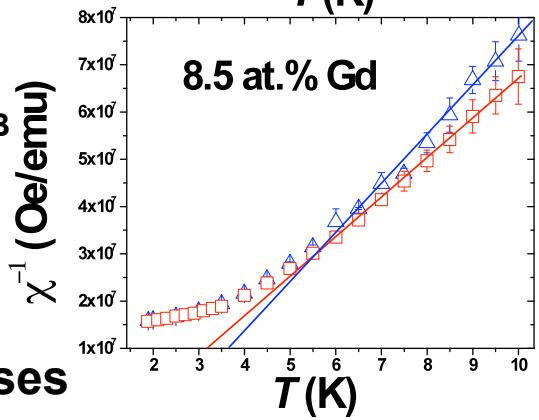
ta-C:Gd_x

Paramagnetic down to 2 K, for x ≤ 8.8 at.%
SG, for x ≥ 17.6 at.%
Follows Curie-Weiss (CW) law, p_{eff} ~*8* μ_B



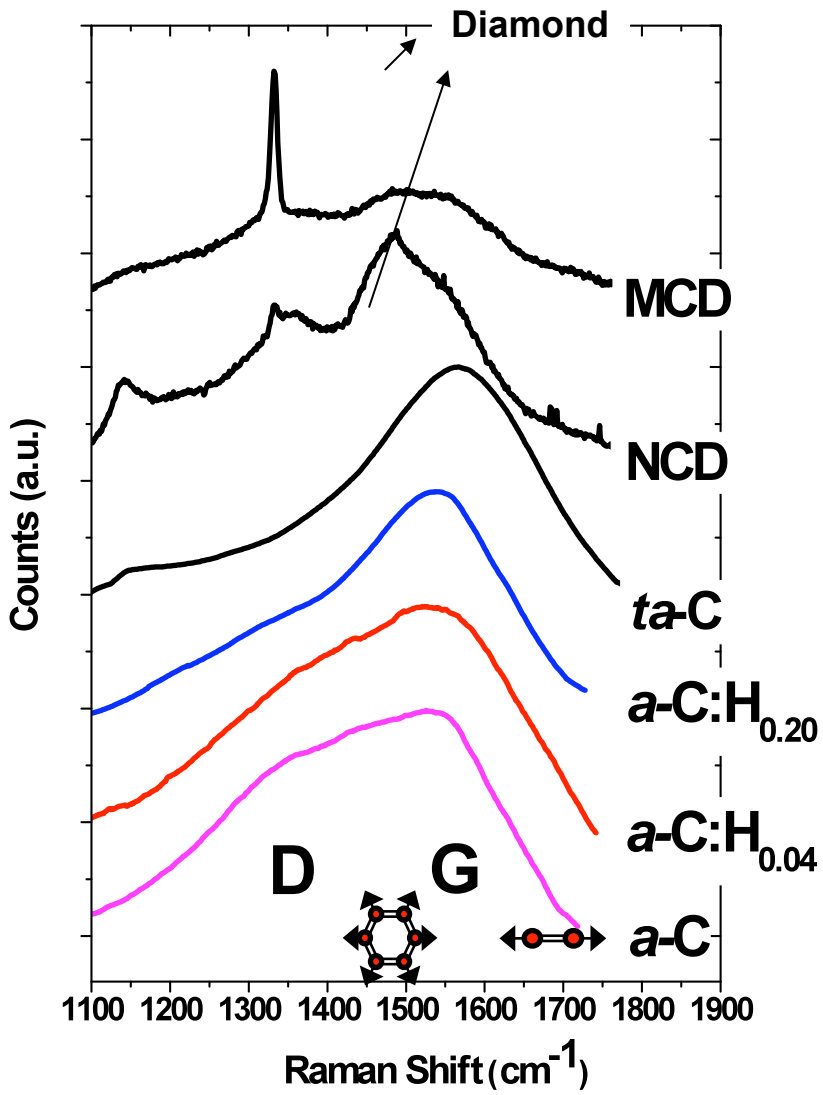
NCD:Gd_x and MCD:Gd_x

⩽ 3.2 at.%: Paramagnetic, follows CW law, p_{eff} ~*8* μ_B
5.3 at.%: deviated from CW law, but no SG splitting.



Overall: Gd behaves like *S*=7/2 local moment in *a*-C
Less interaction between Gd when sp^3 increases

Raman Spectroscopy



NCD:Gd_x and MCD:Gd_x

ta-C:Gd_x

a-Gd_xC_{1-x}(:H)