DCミュオンの科学
ーμSR物性研究におけるdcビーム利用の意義ー

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Plan
1) Introduction: μSR as a probe of matter
2) Pulse vs dc muon beam

1. μSR as a probe of matter

--Target fields for μSR--

What is observed by μSR?

Local electronic states (**q**=0) :

Local magnetic fields ($\omega = \gamma_\mu B$) exerted by electrons, and their fluctuation (with a correlation time τ_c) is observed.

Observables = Spectral density $P(\omega)$ [and its modulation due to fluctuation]

↕ Complementary

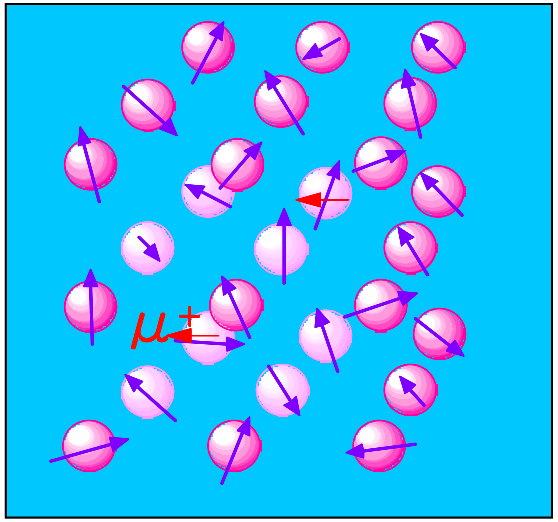
What is observed by X-ray (SR), neutron diffraction:

Charge/spin structure: an avelage over entire volume

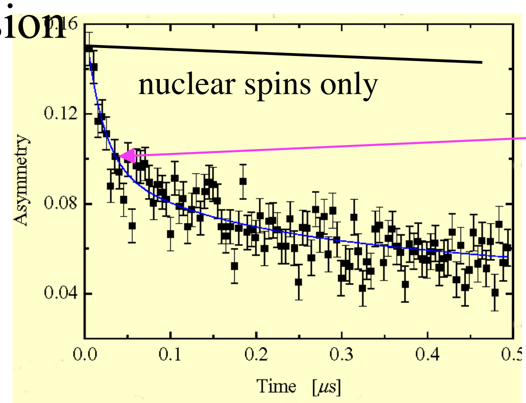
Observables = e.g., structure function $S(\mathbf{q},\omega)$

1) Magnetism probed by μSR

Paramagnetic

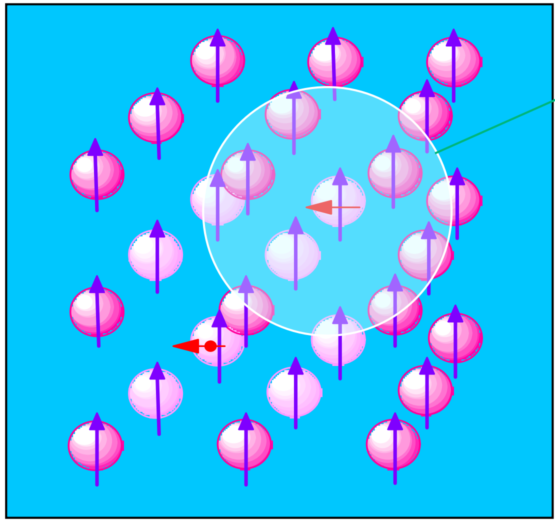


Magnitude of local field varies site to site, leading to loss of phase coherence for muon precession



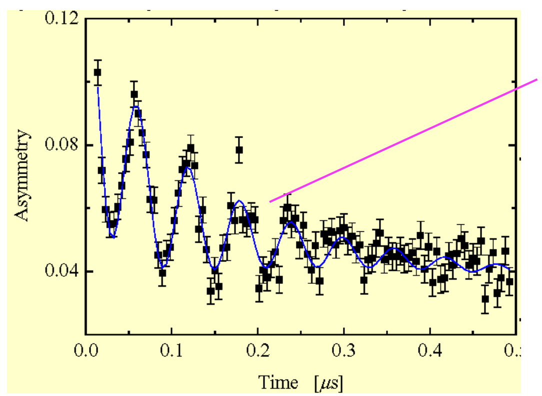
Moment size can be guessed from the magnitude of relaxation

Ordered (ferromagnetic)



sensitive only to the nn atomic moments

Magnitude of local field is common to all muon sites, leading to coherent muon precession.

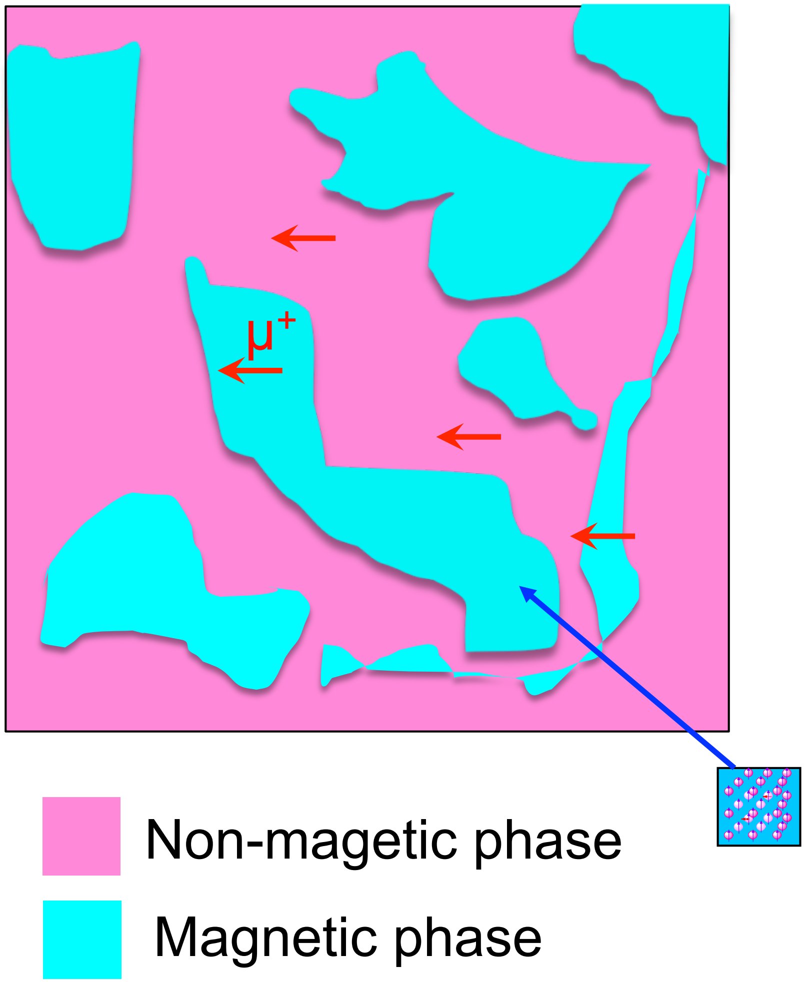


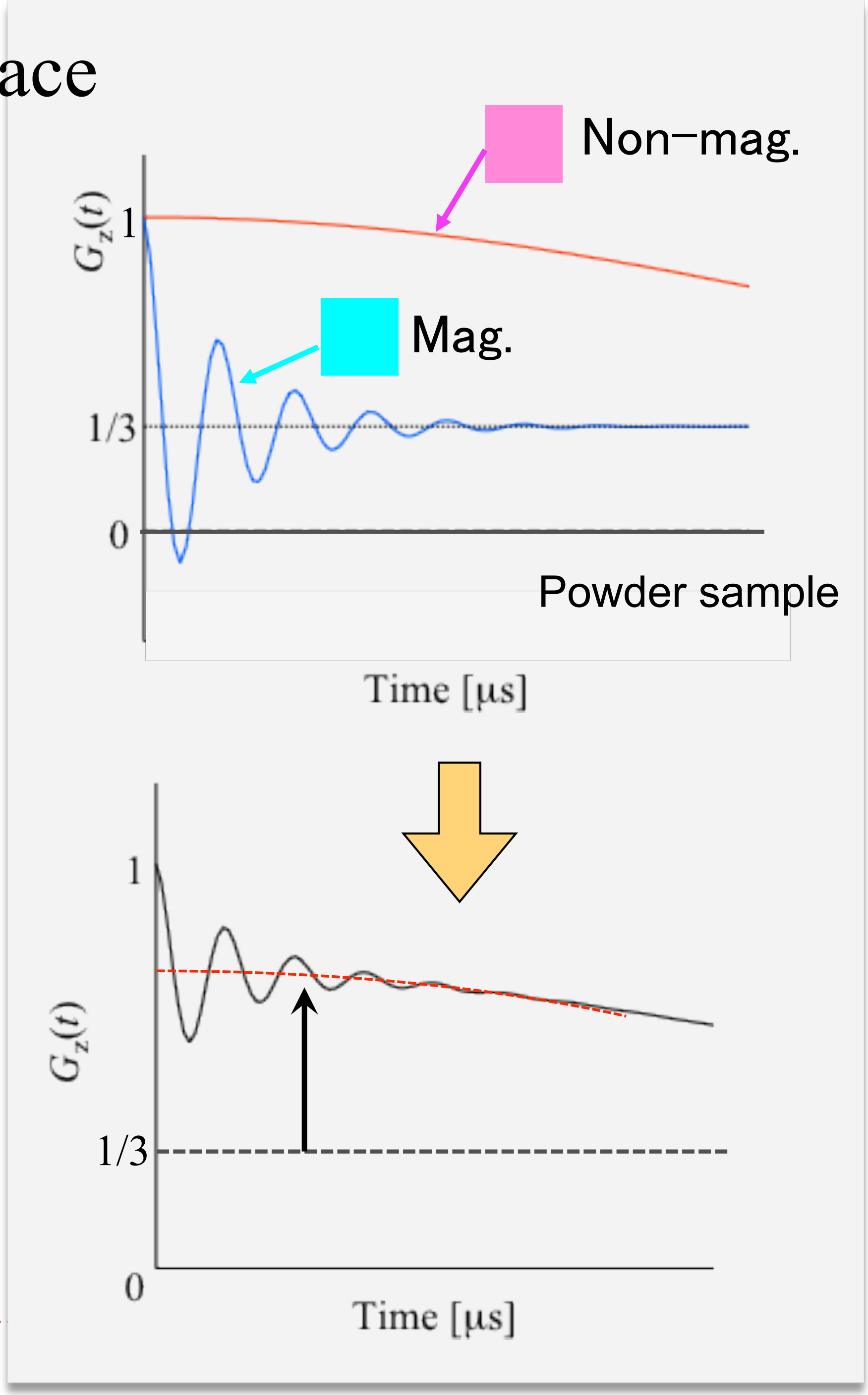
Oscillating signal
- frequency
- relaxation rate
- amplitude

However, it is not sensitive to the long-range structure.

Phase separation in the real space

Muon takes random sampling of internal field



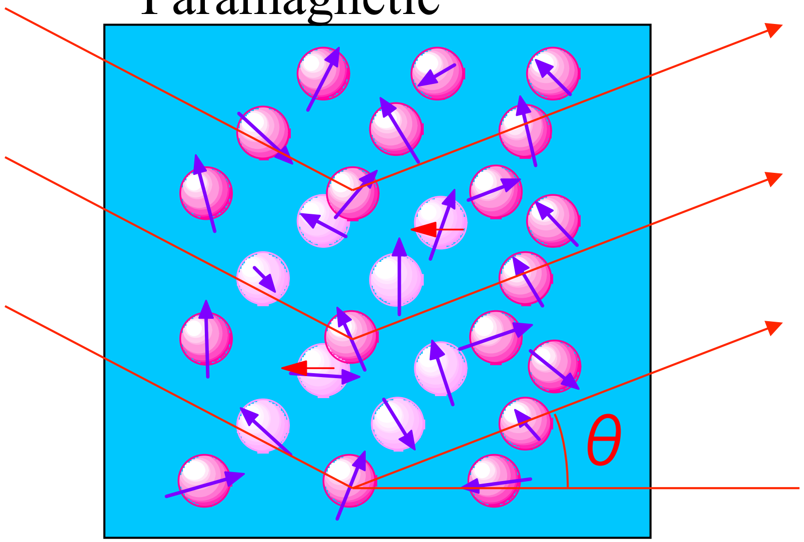


▶ *cf.* multiple sites in the unit cell

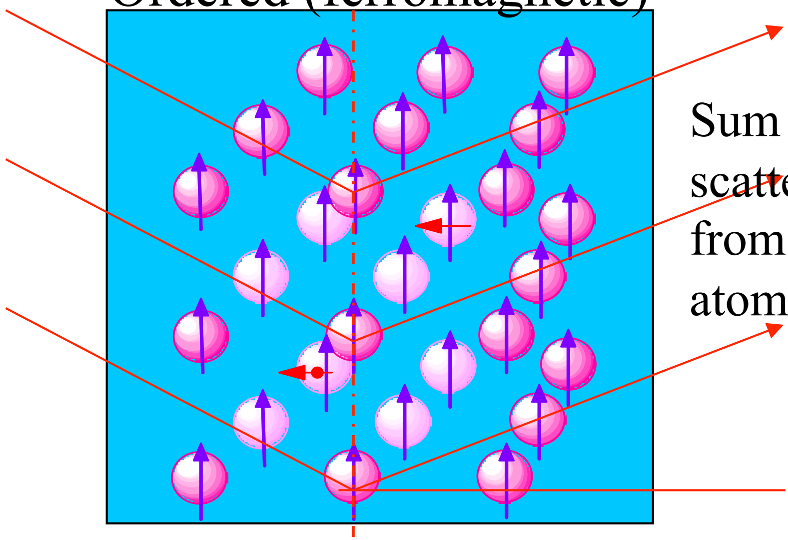
cf. Neutron diffraction

Diffraction intensity is proportional to the degree of long-range order

Paramagnetic

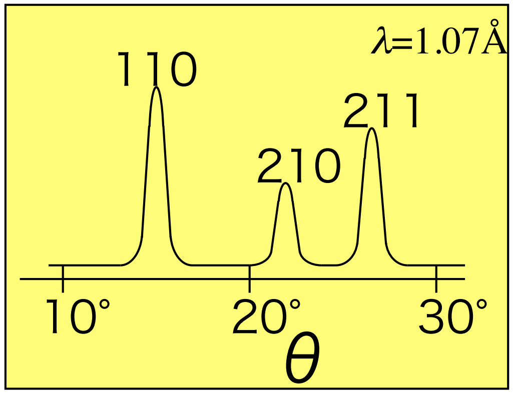


Ordered (ferromagnetic)



Sum of scattering from all atoms

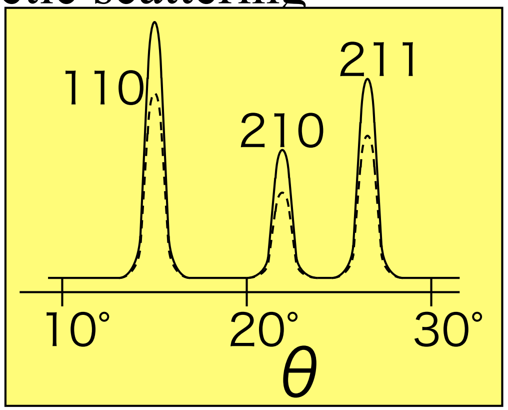
Diffraction due to nuclear scattering



$2d\sin\theta = \lambda$

Bragg's formula

Increase of diffraction signal due to magnetic scattering

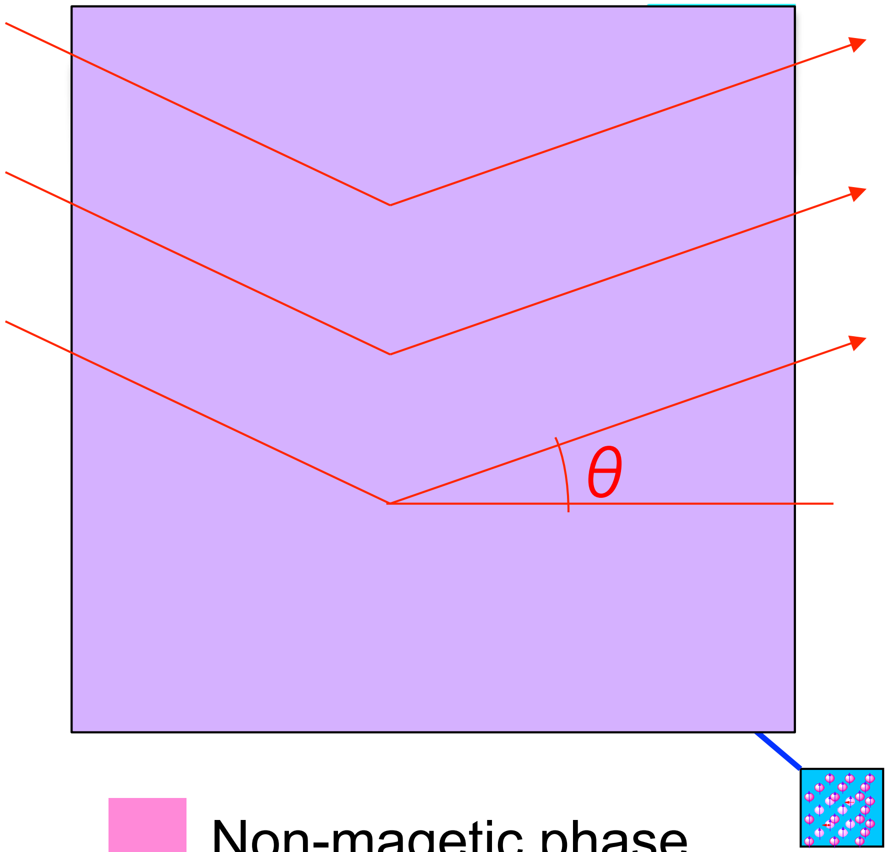


Volume-averaged size of magnetic moment can be evaluated from the intensity.

Diffraction pattern is sensitive to the structure.

Phase separation in the real space

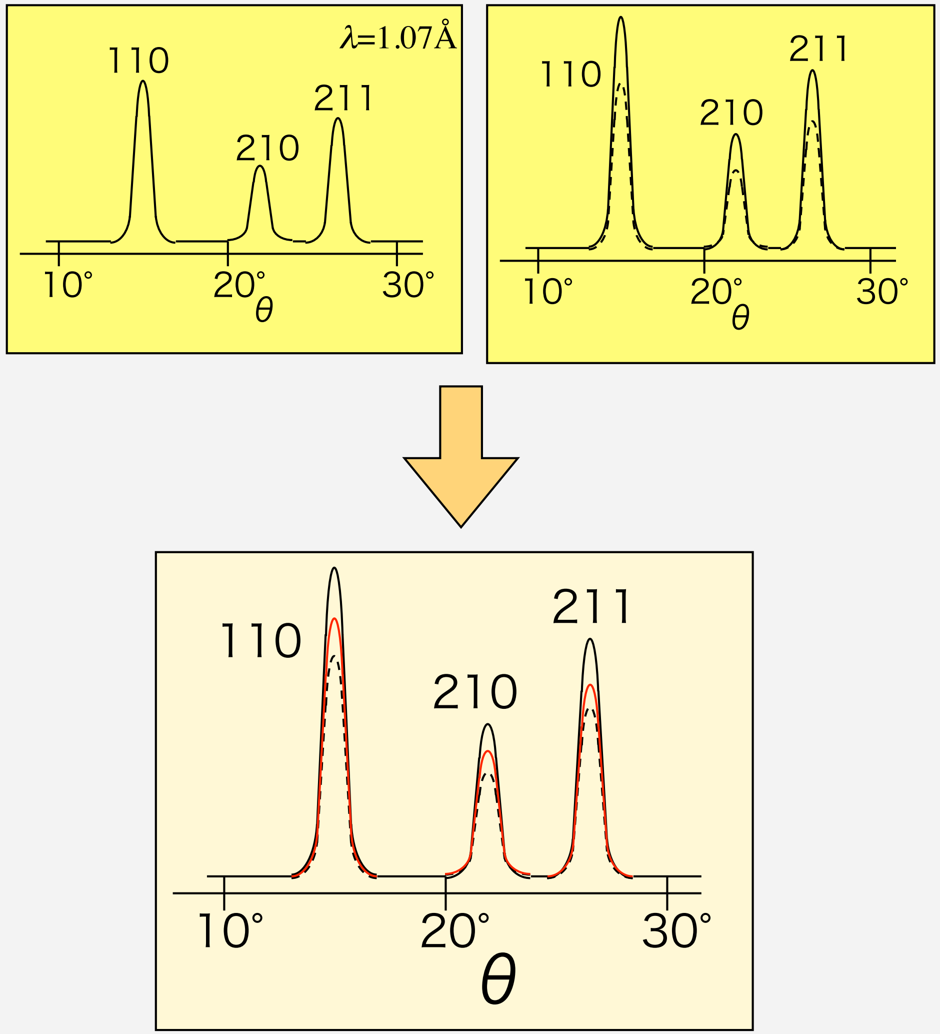
Neutron/X-ray takes volume averaging of order parameters



Non-magetic phase

Magnetic phase

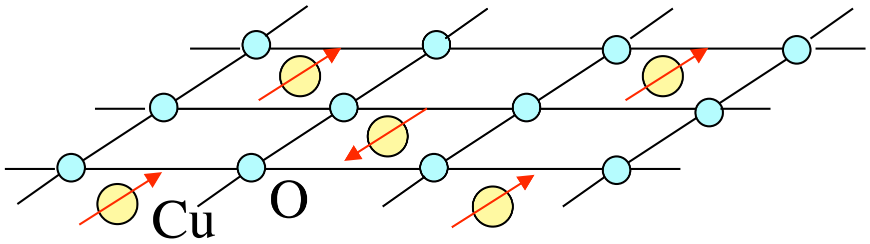
Non−mag.   Mag.

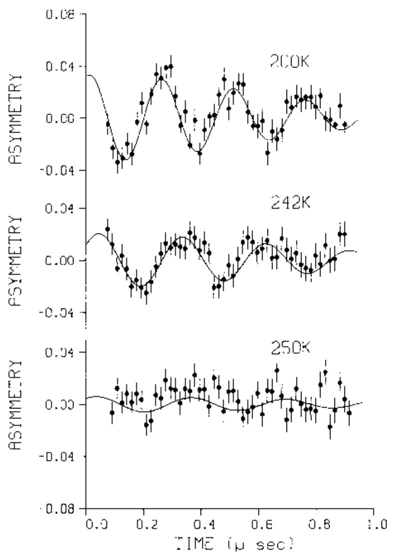


…difficult to distinguish two phases

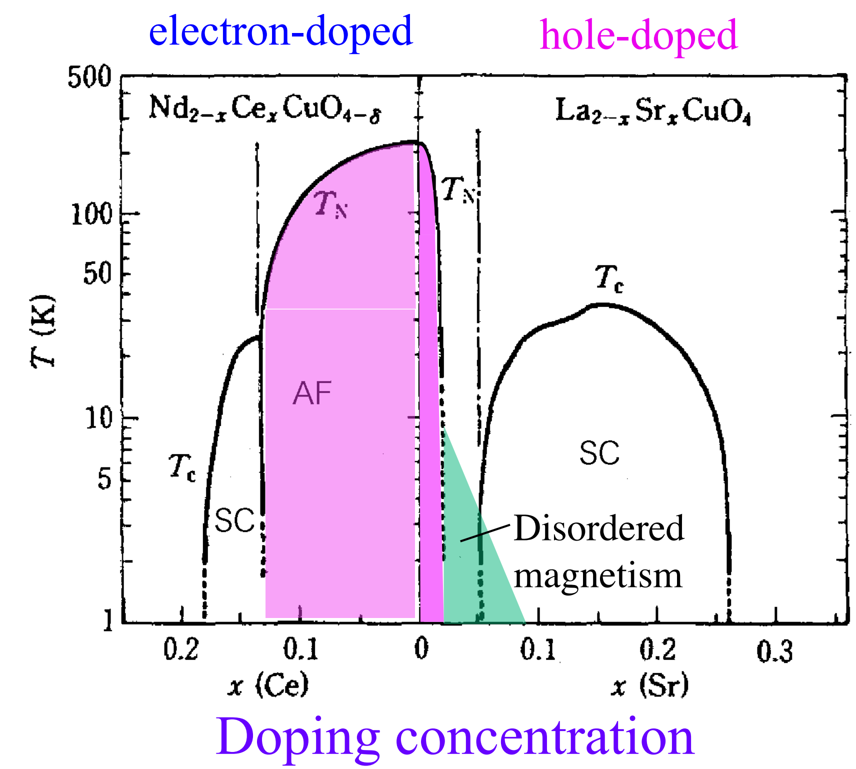
Magnetism of high T_c cuprates

It is µSR measurements that first demonstrated antiferro-magnetism in the parent compounds of high T_c cuprates.



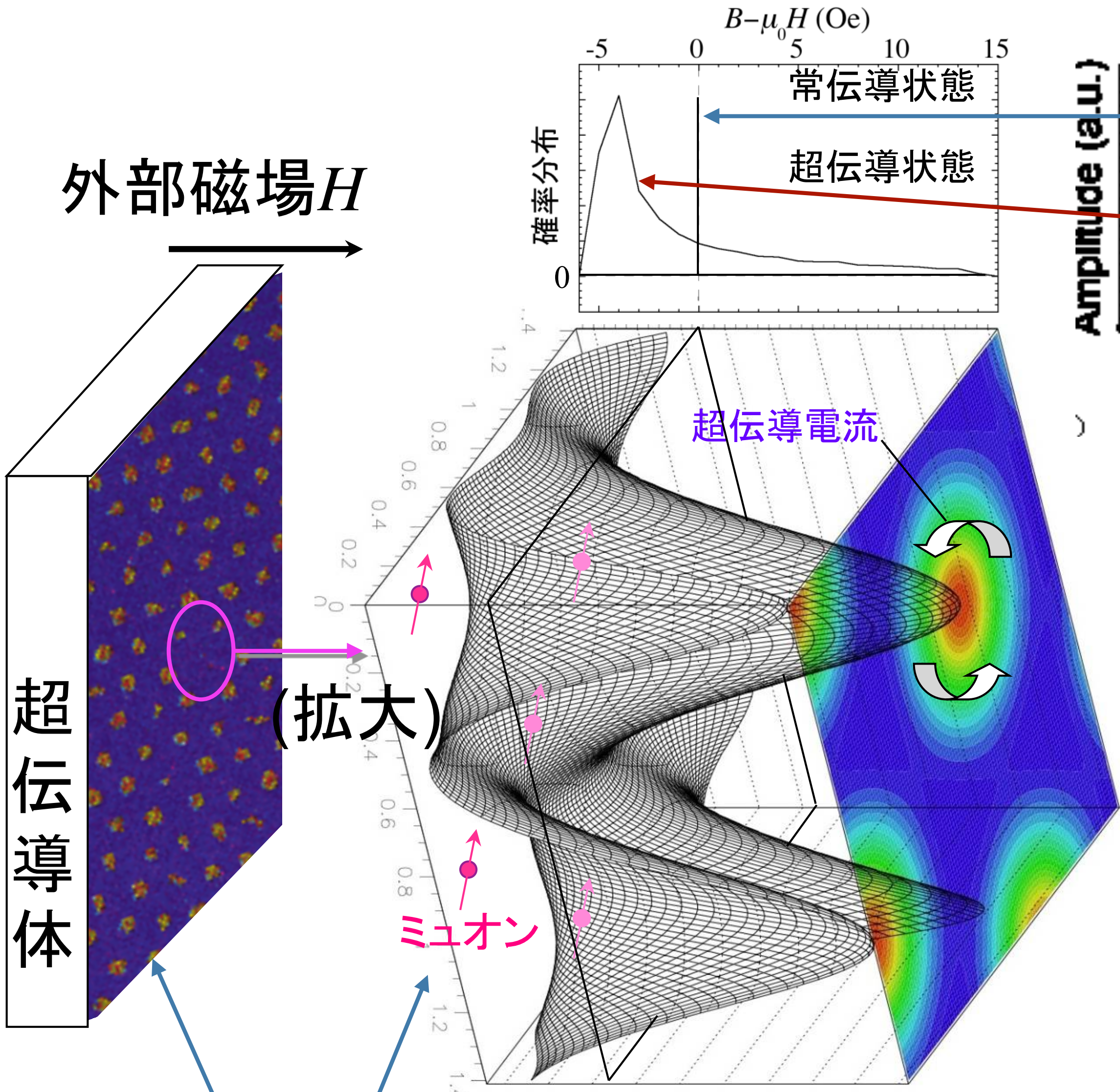


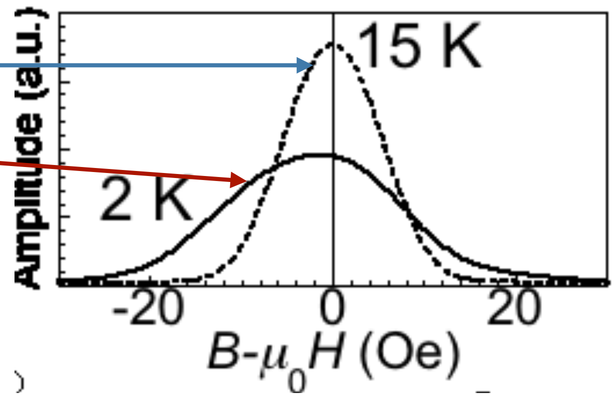
←ZF-µSR time spectra in La_2CuO_4:
A spontaneous oscilla-tion of muon spins are clearly visible.



µSR measurements can be performed with a relatively small amount of samples:
→Powerful technique for the evaluation of new materials.

2) Superconductivity probed by μSR





磁束格子　超伝導体内部の磁場分布

↑ミュオンで見た超伝導電流による磁場分布の変化：（左は測定の分解能を考慮しない場合の計算値で、これに実際の分解能を考慮すると右図の測定結果に対応する。）

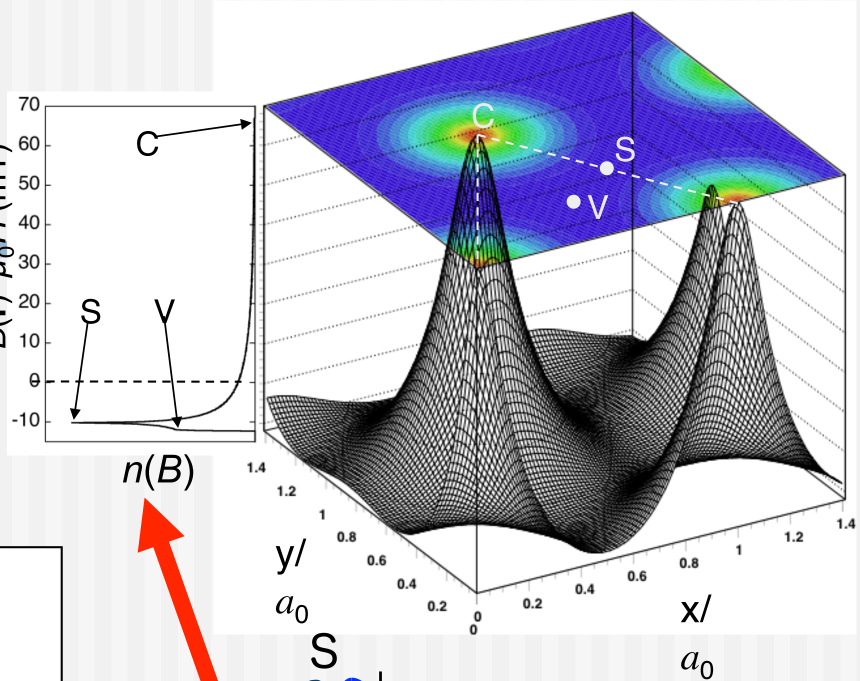
超伝導状態では磁束格子の形成により、磁場に分布ができ、分布幅の増大は超伝導電流の強さで決まる。

ミュオンは磁場分布をランダムにサンプリングする。

TF-μSR signal provides a density distribution $n(B)$ [i.e., random sampling] of the internal field profile, $B(\mathbf{r})$.

$$P(t) = \int_{-\infty}^{\infty} \exp(i\omega t) n(B) dt$$

$$n(B) = \langle \delta(B(r) - B) \rangle_r$$

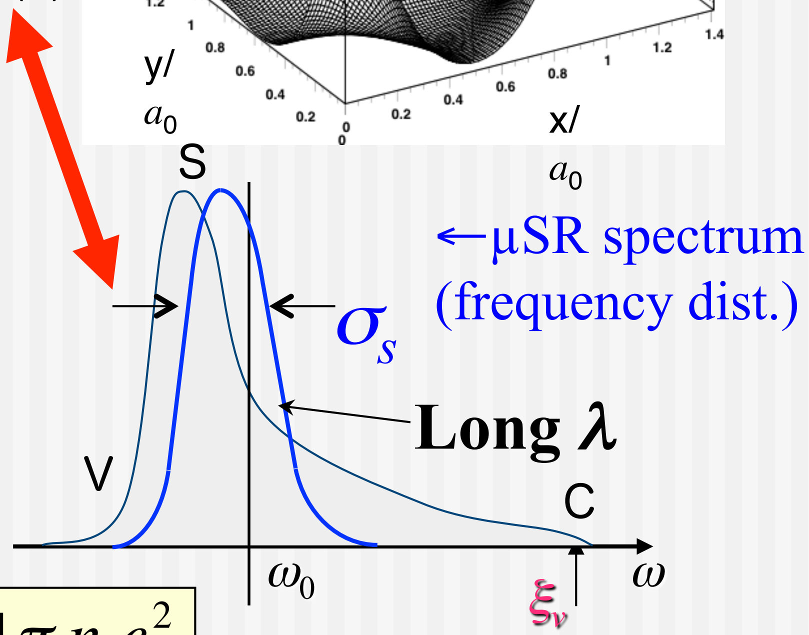


NB: λ is the *effective* penetration depth

$$\frac{1}{\lambda^2} = \frac{4\pi n_s e^2}{m^* c^2} \quad n_s(T,H) = n_s(0,0)[1-g(T,H)]$$

$g(T,H)$: quasiparticle density

…reflecting superfluid density



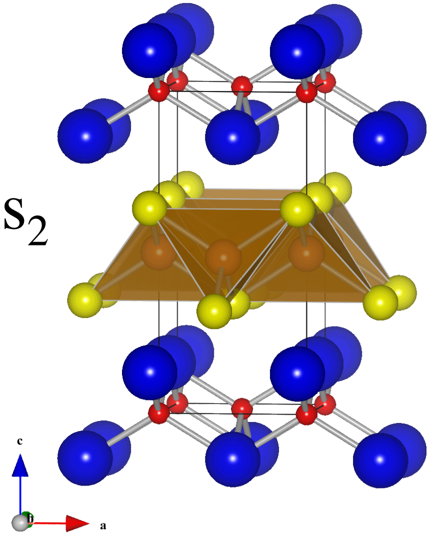
Long λ → Gaussian approx. →

$$\text{...Gaussian linewidth } \sigma_s \propto \frac{1}{\lambda^2} = \frac{4\pi n_s e^2}{m^* c^2}$$

Recent example:

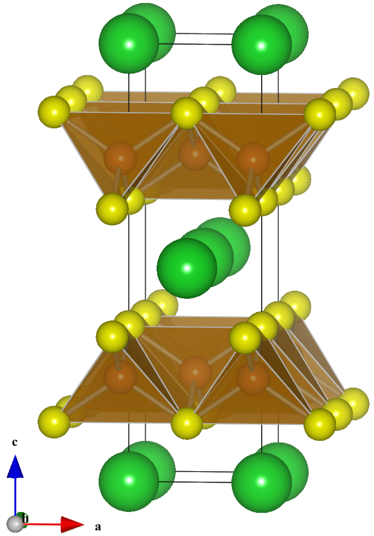
High-T_c Superconductivity in Iron Pnictides

Fe_2As_2 layer



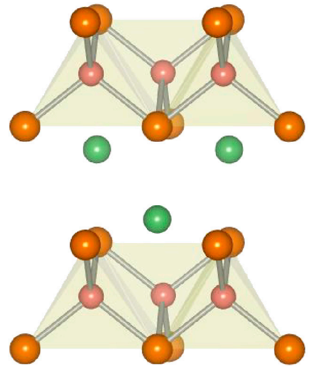
1111 family

($LaFeAsO$)



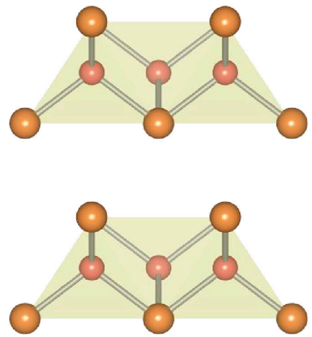
122 family

($BaFe_2As_2$)



111 family

($LiFeAs$)



11 family

($FeSe$)

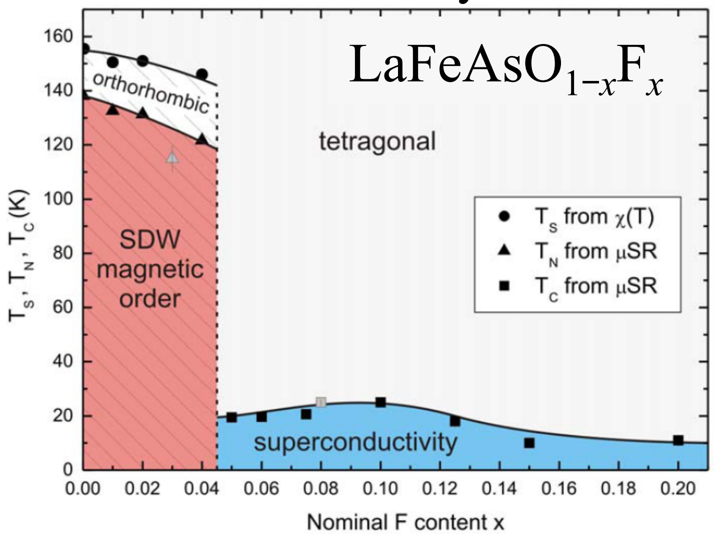
…more

- High-T_c realized on materials containing iron.
- Number of similarities with cuprates.

  Carrier doping of 2D-Fe_2As_2 layers, AF(SDW)-SC phase diagram with optimal doping for high-T_c, etc…

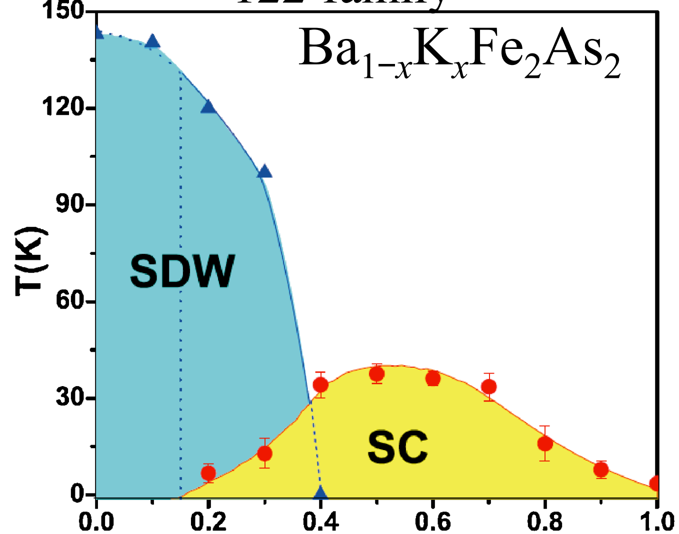
Doping-phase diagram (by various μSR groups)

1111-family

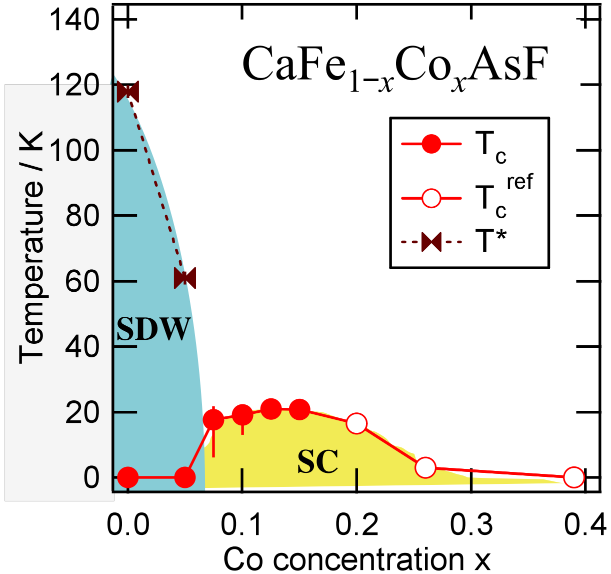


x: Electron-doping

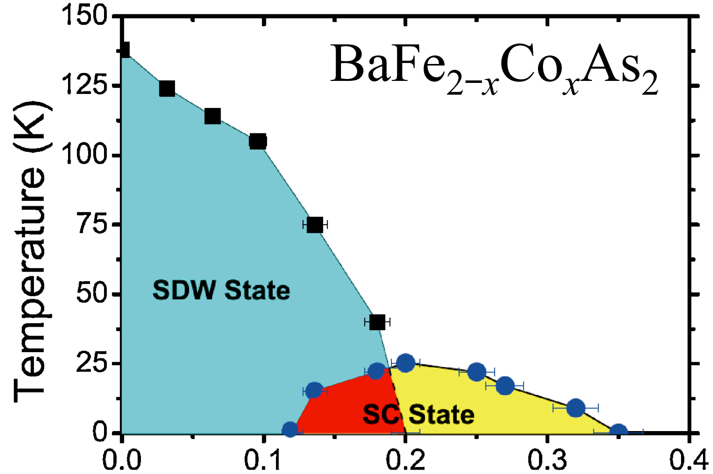
122-family



x: Hole-doping



x: Electron-doping

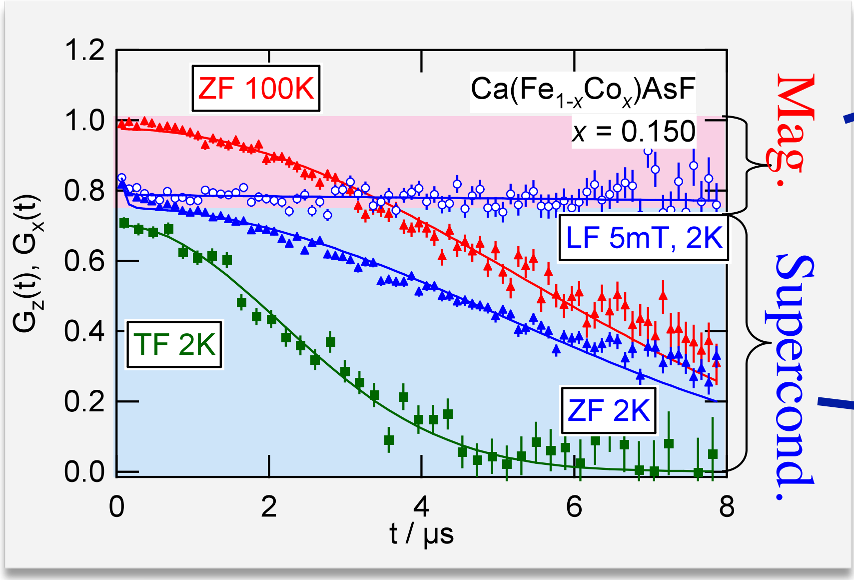


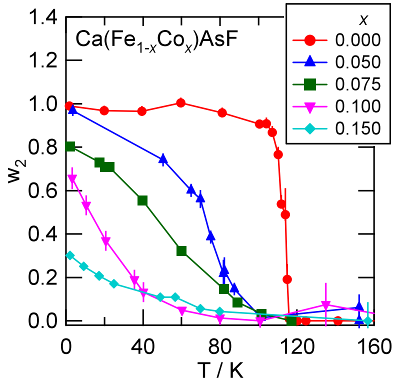
x: Electron-doping

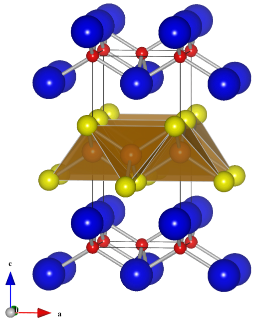
(…hole-doping is difficult)

Coexistence of superconducting and magnetic phases in $CaFe_{1-x}Co_xAsF$

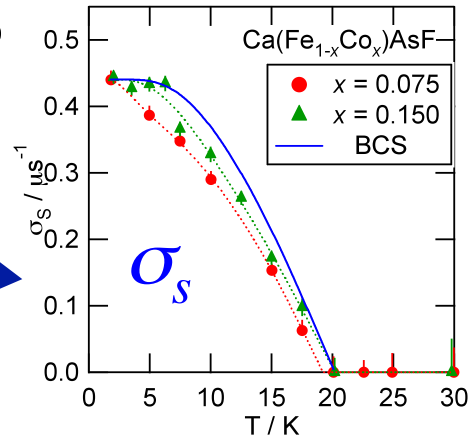
*μ*SR is sensitive to phase separation.

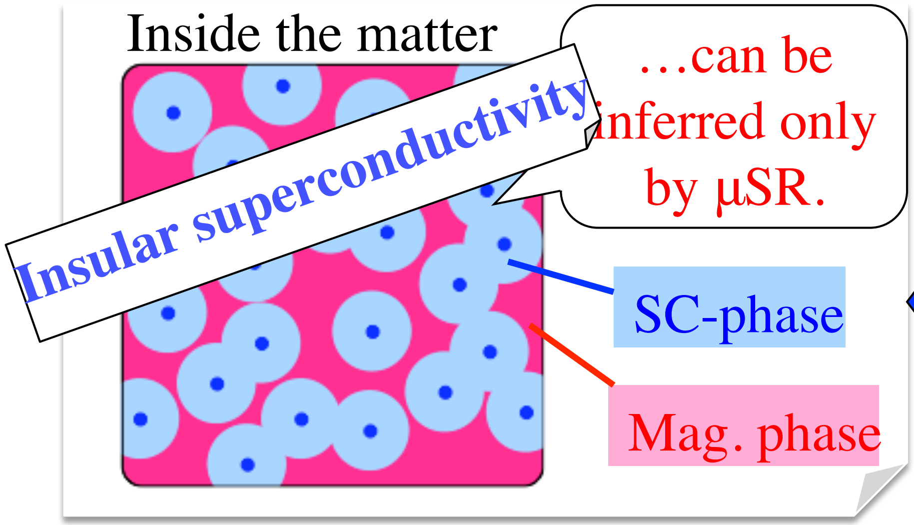


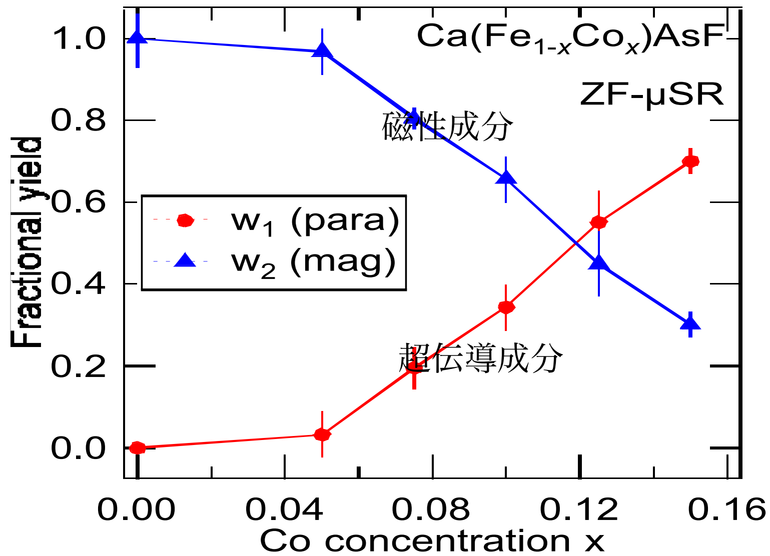




One can extract information from respective parts.

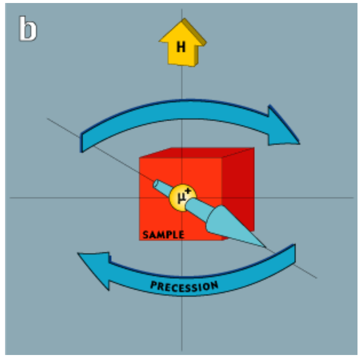






S. Takeshita *et al*., Phys. Rev. Lett. **103**, 027002 (2009)

Muons (μ^+'s) in Matter



Positive muon (μ^+) : a light radioisotope of proton

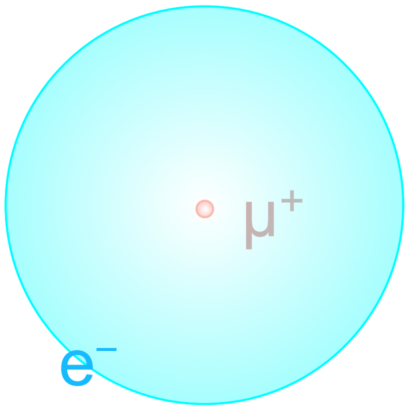
Electronic property: [mass=m_p/9, charge=e^+, spin=1/2]

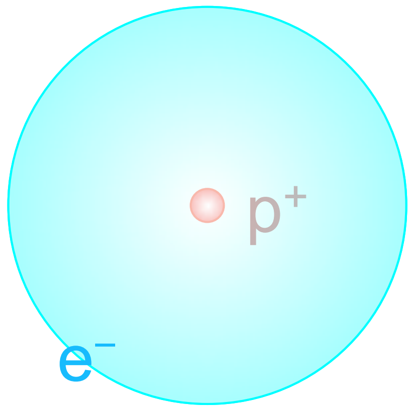
Atom	Muonium	Hydrogen
Reduced mass (m_e)	0.995187	0.999456
Bohr radius (Å)	0.531736	0.529465
Ground state energy (eV)	-13.5403	-13.5984

Difference in electronic structure : =0.4%

Mu ≈ H

[...not necessarily true for dynamical property]





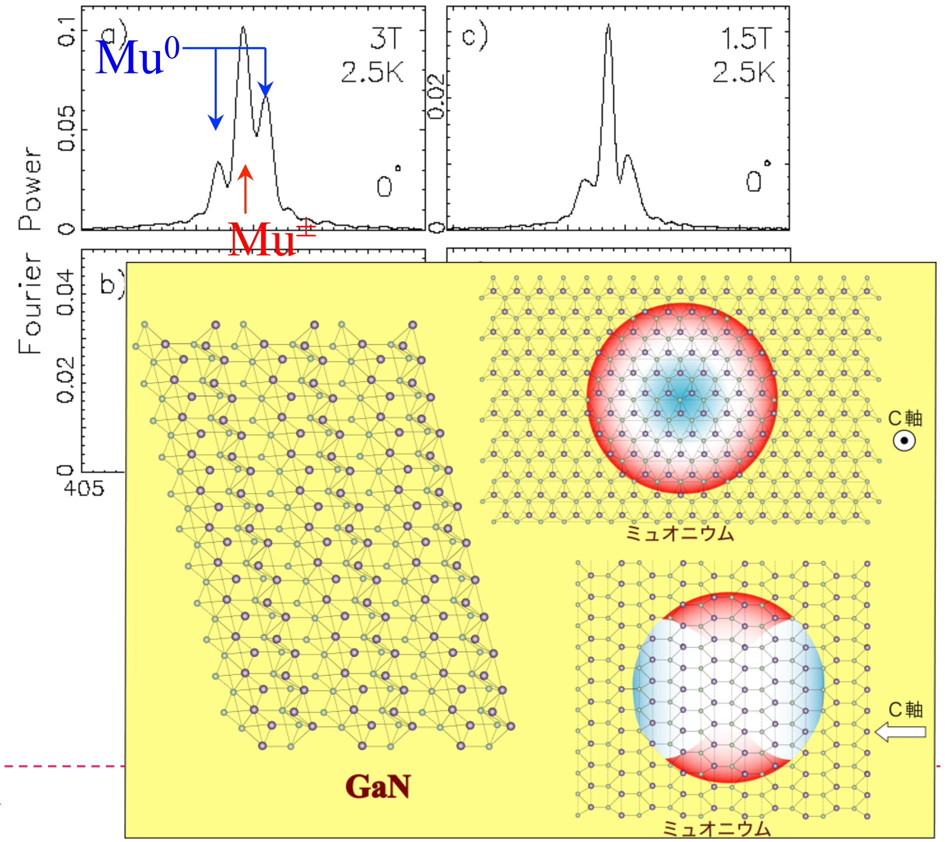
3) Muon as H-simulator in matter

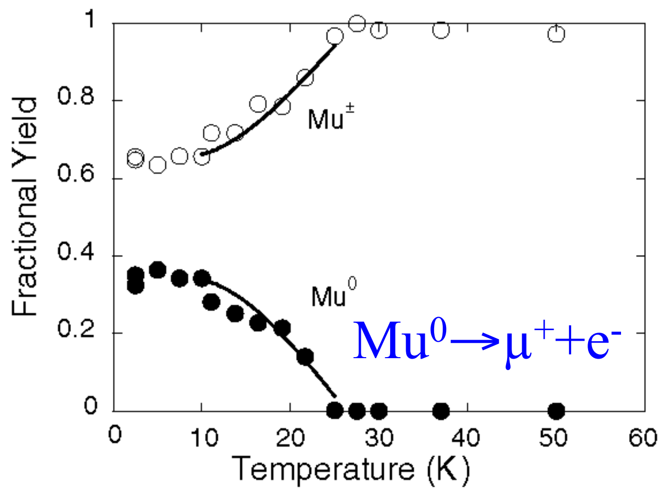
Typical example 1: Muonium in semiconductors

Hydrogen (H, often located at interstitial sites) is the key player in semiconductors...their electrical activity is strongly altered by H.←It is difficult to obtain information on *isolated* interstitial hydrogen (at the dilute limit).

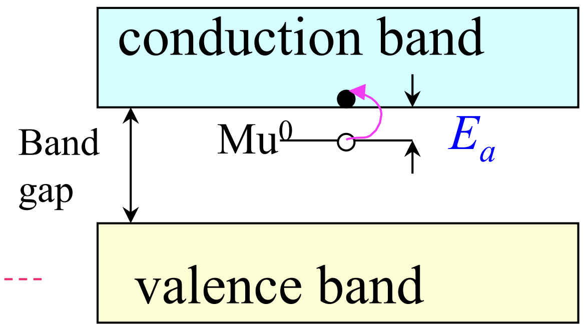
Muonium simulates the state of interstitial H in matter!

Recent example: Muonium (Mu^0) states in GaN





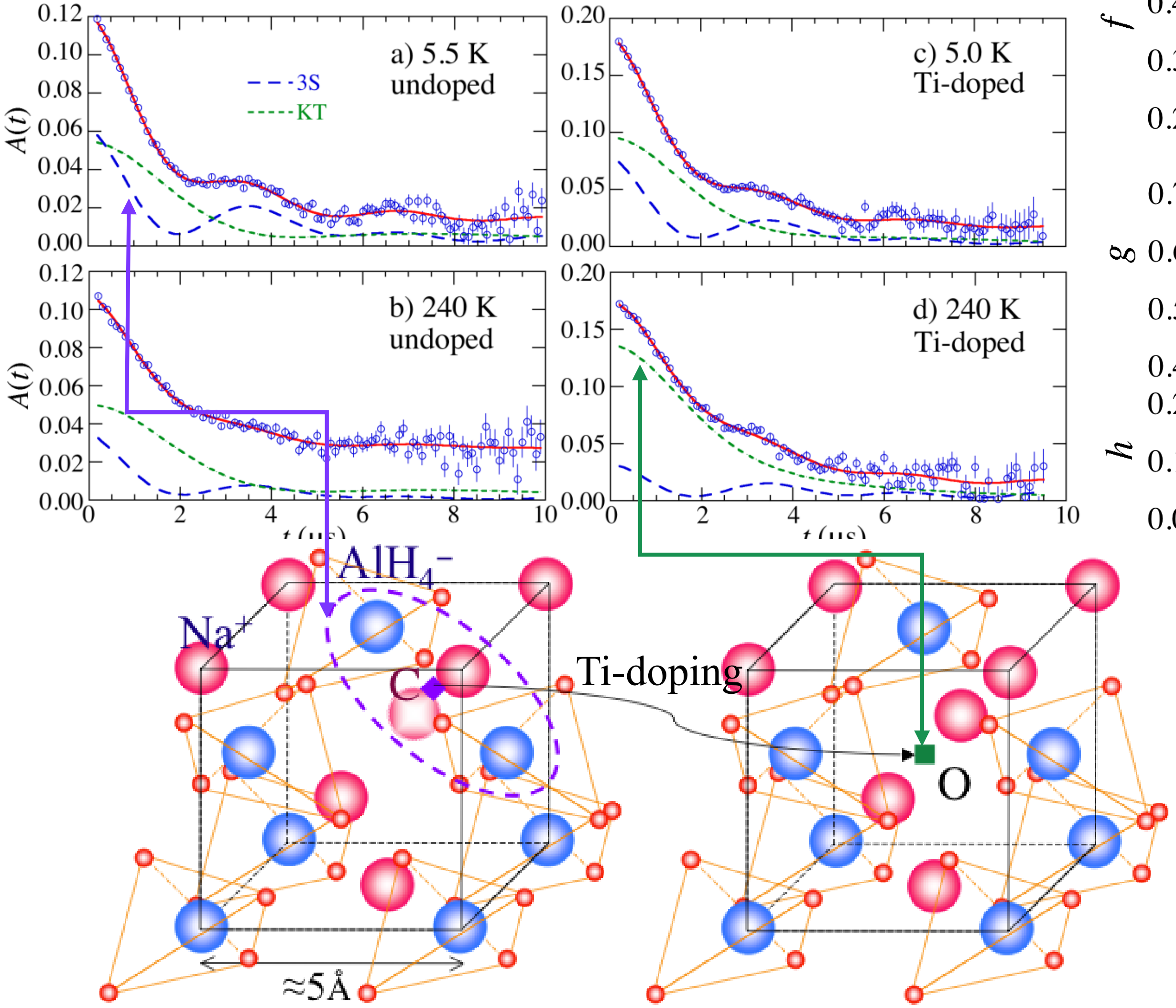
Small activation energy E_a = shallow doner state→GaN may exhibit n-type conductivity by H doping

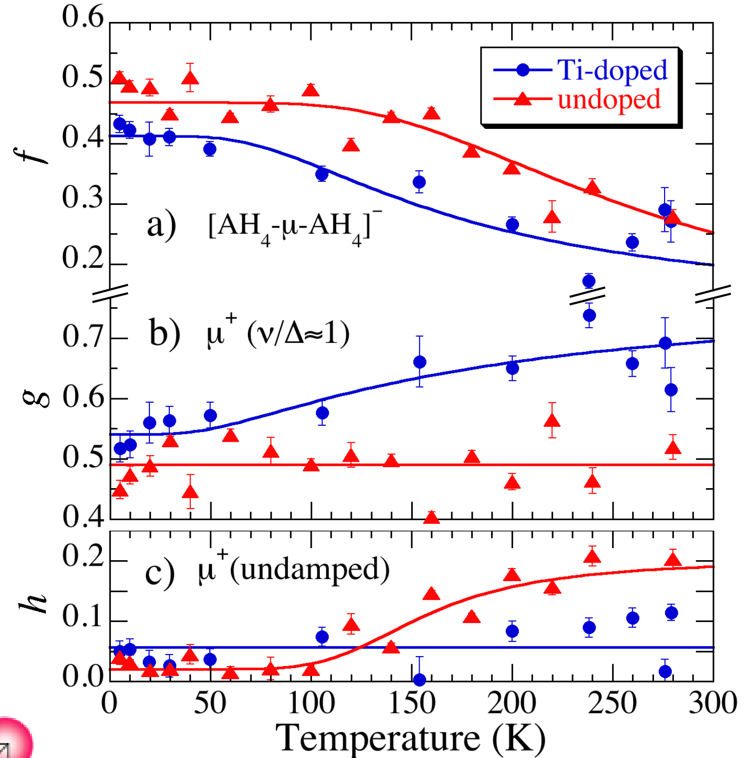


Shimomura, Kadono *et al*. PRL **92** (2004) 135505

Typical example 2:

μ^+ in H-storage materials, $NaAlH_4$





Behavior of μ^+ in $NaAlH_4$ changes drastically by Ti doping.→the same must be true for H.

μ^+ in $NaAlH_4$ demonstrated the formation of a dialanate complex $[AlH_4^- \text{-} \mu^+ \text{-} AlH_4^-]$ by hydrogen bonding.

Kadono, Shimomura *et al.* PRL **100** (2008) 026401

μSR as a specific probe of matter - unique "niche"

Local magnetic probe in the atomic scale:

Compared with neutron diffraction:

1) Magnetism can be evaluated irrespective of long-range order.

...tolerates small magnetic moments (S)/large unit volume v_0

cf. neutron diffraction intensity $\propto V \cdot S^2/v_0^2$ ↔ μSR frequency $\propto S$

Asymmetry $\propto$ volume of magnetic phase

2) It requires small quantity of samples.

3) μSR responds linearly to the magnetic order (...no "extinction")

Compared with nuclear magnetic resonance (NMR):

1) It can be applied to any matter irrespective of nuclear spins.

2) No complexity due to electric quadrupolar interaction because of pure magnetic probe (muon spin=1/2).

Compared with both neutron & NMR:

μSR is sensitive to the time window inaccessible to those two probes.

2. Pulse vs dc muon beam

--Towards the era of J-PARC MUSE

Muon beams @J-PARC (provisional) …Intensity frontier

	Intensity (@1MW)	Polarization	Pulse width	Energy
D-line	10^7 μ^+/s (~10^6 μ^+/s/cm^2)	~100%	~90-140 ns	10^0-10^1MeV
U-line	10^6 μ^+/s (~10^6 μ^+/s/cm^2)	~50%	~1 ns	10^2-10^4eV
S-line	10^7 μ^+/s (~10^6 μ^+/s/cm^2)	~100%	~90-140 ns	4 MeV
H-line	10^7 μ^+/s (~10^6 μ^+/s/cm^2) 10^6 μ^+/s (~10^6 μ^+/s/cm^2)	~100% ~50%	~90-140 ns ~1 ns	4 MeV 10^{-2}eV

パルスビームの弱点

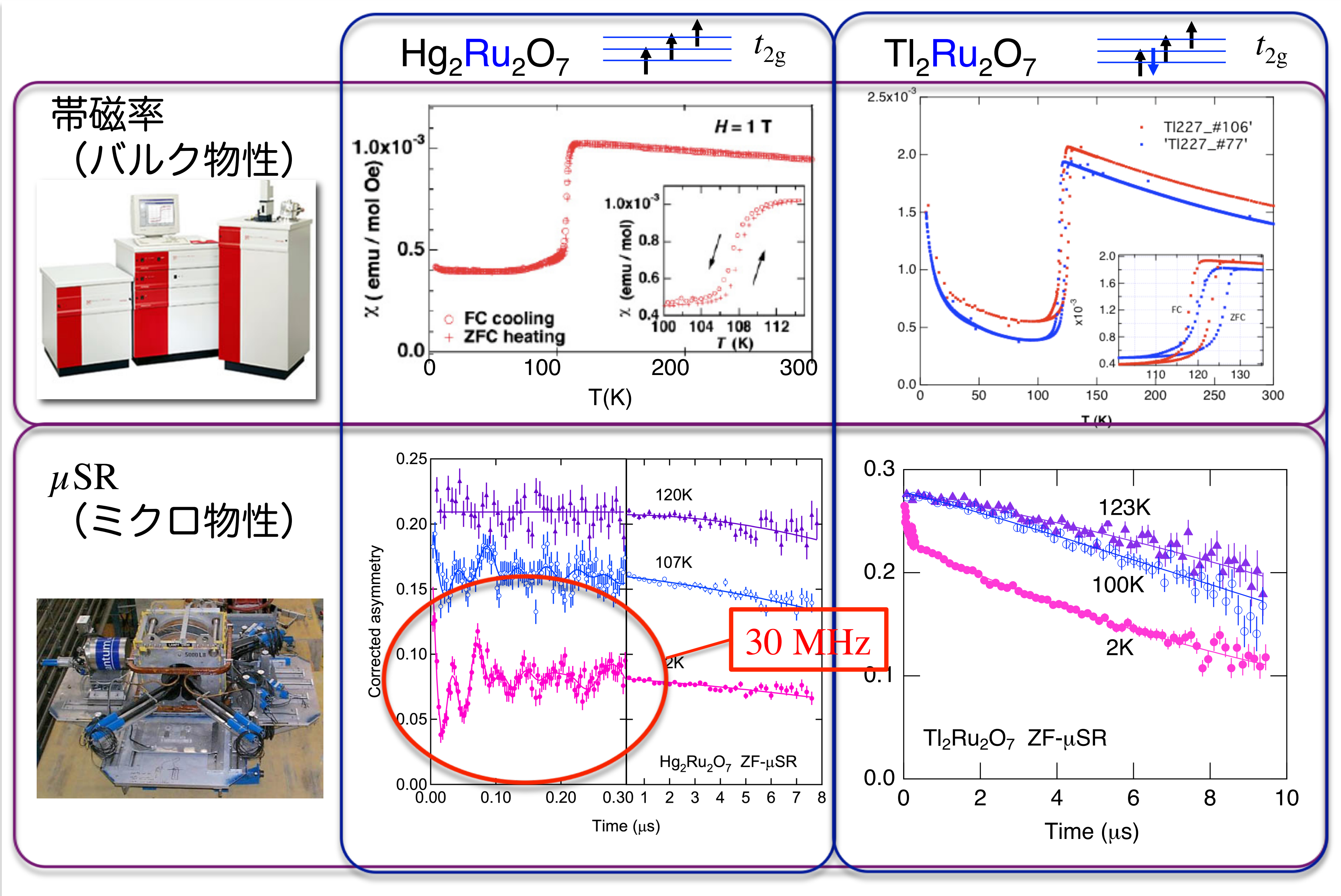
1）時間分解能がビームパルス幅に制約される：
・ゼロ磁場測定では絶対的な制約（磁性体等の内部磁場観測困難）。
・磁場中測定ではπ/2-RFパルス法により克服可能だが、実験装置が大掛かりになる。
2）入射ミュオンを一つずつ同定できない：
・試料以外に止まったミュオンからの信号を排除することが困難。
（＝試料サイズの減少とともにS/Nが悪化。）

∴dcビーム利用が有利な実験

＝時間分解能が必要な実験：
ゼロ磁場μSRによる（特に微小試料）評価(希土類磁性研究)[a]
高横磁場μSRによるミュオン・ナイトシフト測定(磁性研究)[b]、
第二種超伝導体の磁束格子状態観察（超伝導研究)[b]、
物質中のミュオニウムスピン回転分光（水素同位体研究）[b]、等

a）超低速ミュオンビームが実現すると優位性は失われる。
b）ビームラインにスピンローテーターが必要。

Zero-field μSR with high-time resolution



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a）超低速ミュオンビームが実現すると優位性は失われる。

b）ビームラインにスピンローテーターが必要。

Muon Knight shift ＝原子スケールでの局所帯磁率

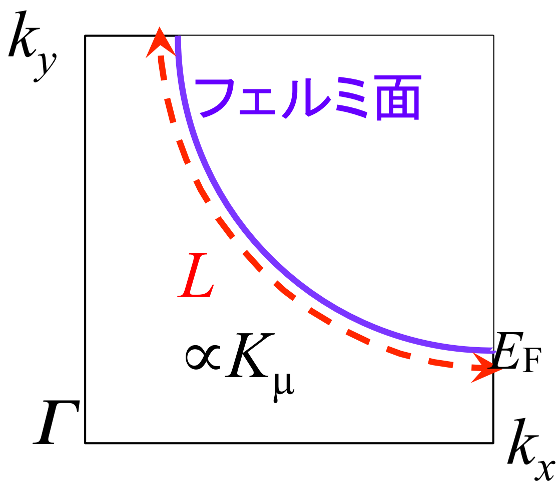
フェルミ面の状態密度 $D(E_F)$ ［＝ナイトシフトに比例］は電子物性の基本情報

$$K_\mu = K_0 + \left[A_c + \left(B_{dip}\right)_z\right]\frac{\chi_{//}}{N_A\mu_B}$$

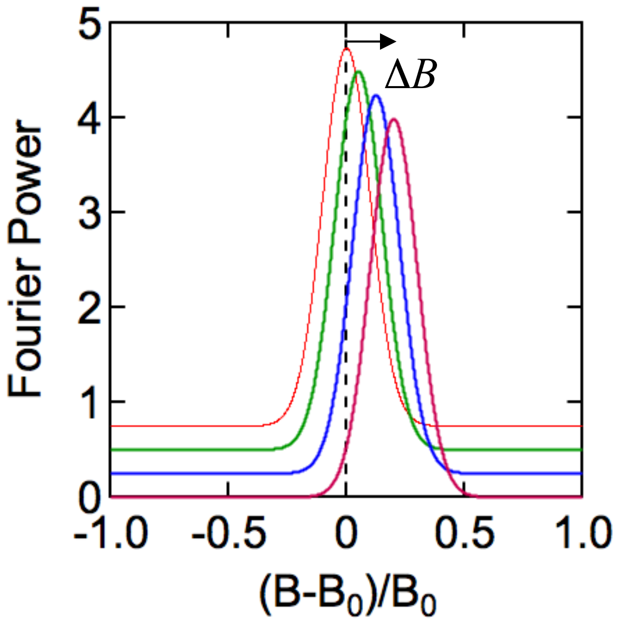
$\chi_{//}$: 局所帯磁率∝$D(E_F)$
B_{dip} : 超微細相互作用
K_0: Pauli常磁性項
N_A: Avogadro数
μ_B: Bohr磁子
A_c: Fermi接触項 (~0 for muon)

Experimental observable

$$K_\mu = -\frac{\Delta B}{B_{ext}} = -\frac{f_s - f_{Ag}}{f_{Ag}}$$



ナイトシフトはフェルミ面の面積に比例（左図：二次元電子系のフェルミ面）

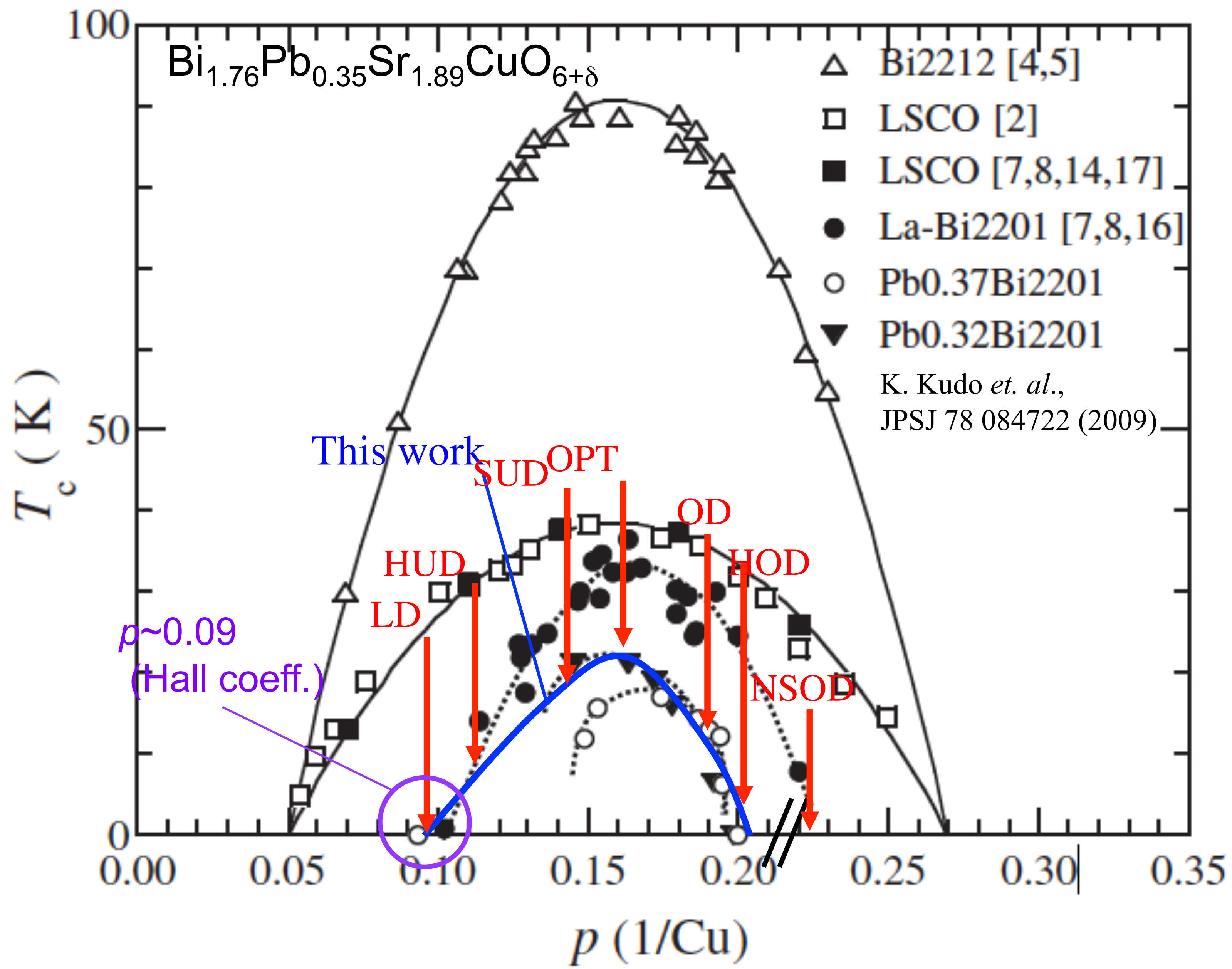


- "HiTime" spectrometer @TRIUMF, M15

Measure. condition：H//c, H=7 T, 2K~300K

現在世界で唯一の実験装置
（PSIで10 Tの装置が2012年秋より稼働予定）

Example of muon Knight shift study: “Pseudogap” in cuprates



p was estimated from T_c using ref. data (Kudo et al.).

Analysis

Assuming…

・Two muon sites --> ratio of initial asymmetries fixed to the value at 250K, where they are clearly discerned.

Each frequency corresponds to …

1. Far from CuO_2 plane : $K_{\mu 1} \rightarrow (A_1, f_1)$
2. Near the CuO_2 plane site: $K_{\mu 2} \rightarrow (A_2, f_2)$

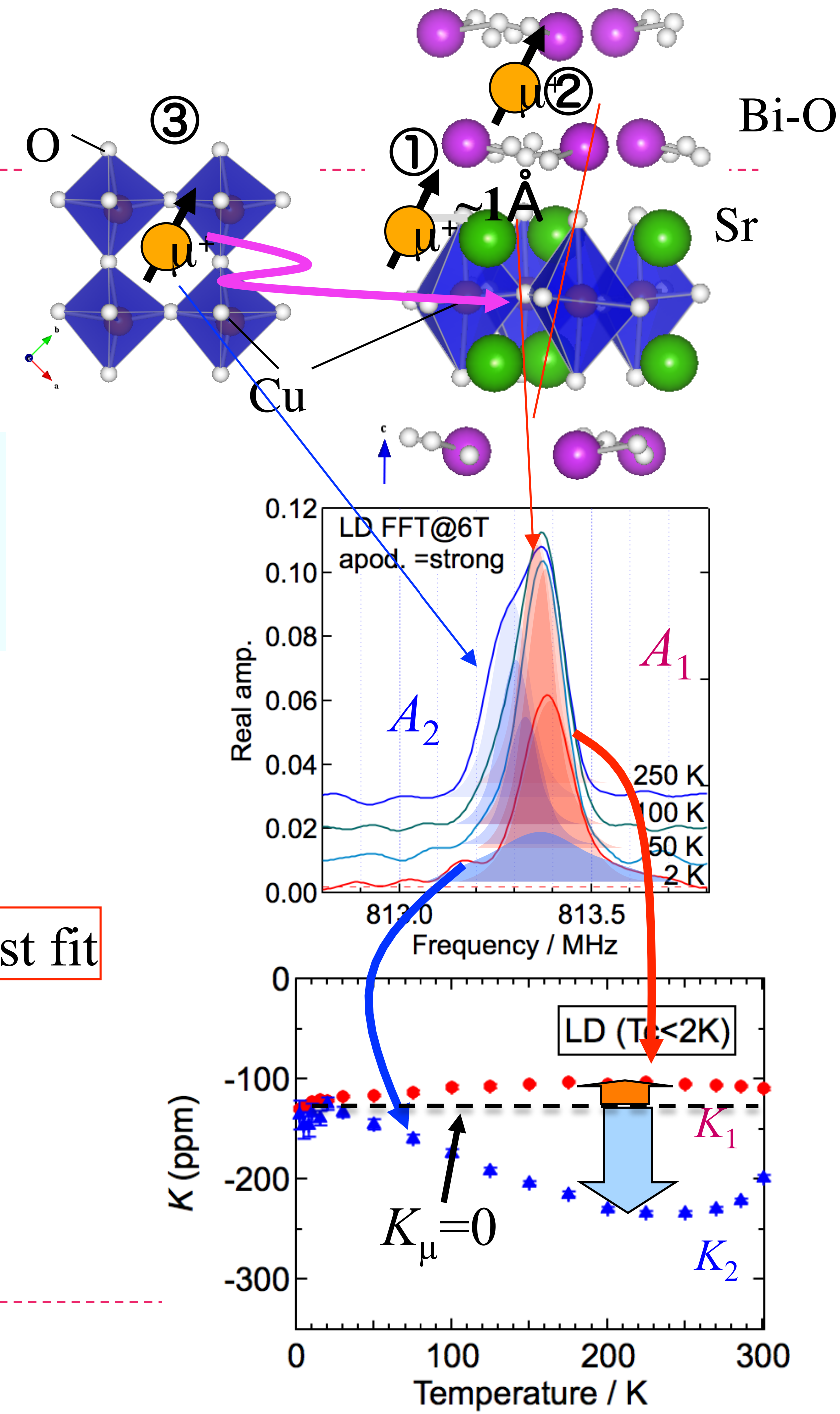
Three patterns of fitting A_1+A_2 with …

1. Gaussian + Gaussian
2. Gaussian + Exponential
3. Exponential + Gaussian <---Best fit

Analysis: fits by

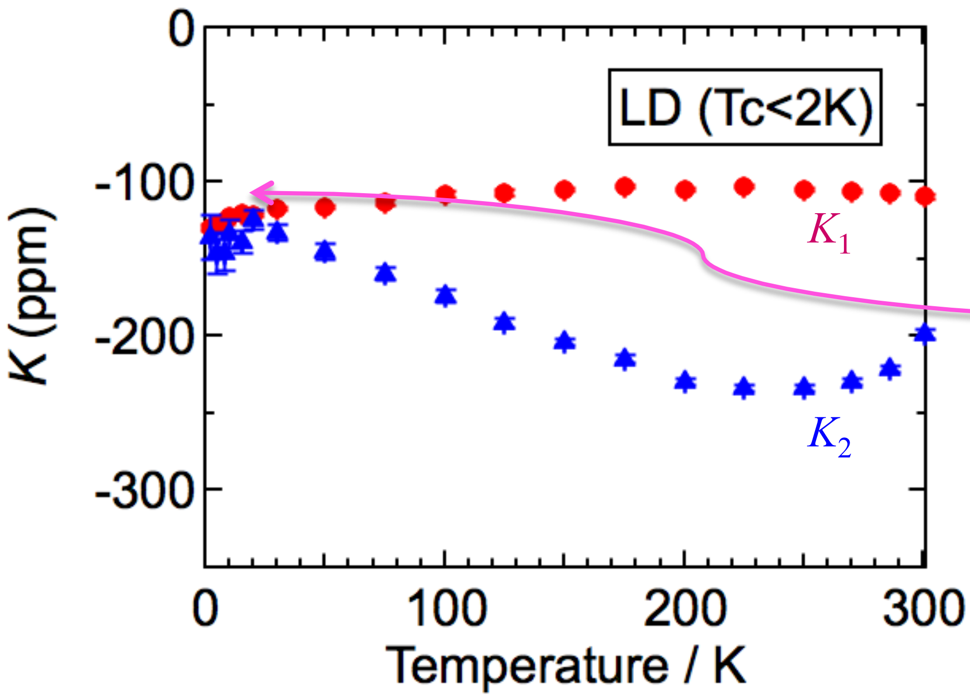
$$P(t)=A_1\exp(-\lambda_1 t)\cos(2\pi f_1 t+\phi)$$
$$+A_2\exp(-\sigma_2^2 t^2)\cos(2\pi f_2 t+\phi)$$

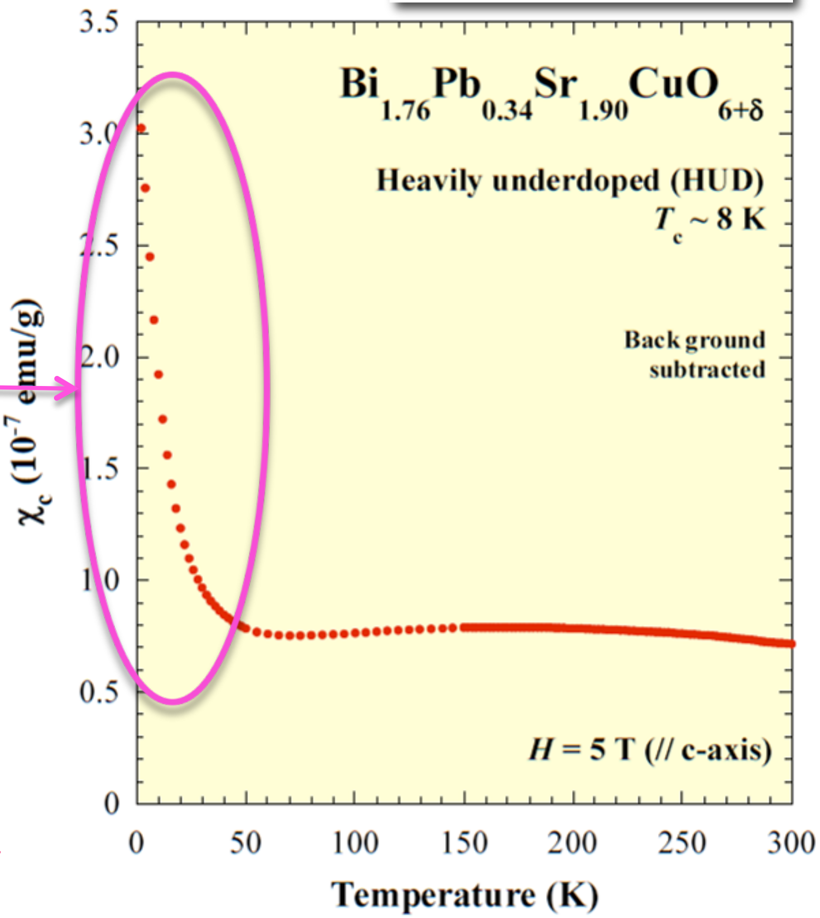
※Ratio of A_1:A_2 is fixed at 250K



Analysis

何故μSRでナイトシフト（局所帯磁率）を測るのか？
…バルク帯磁率は不純物に敏感





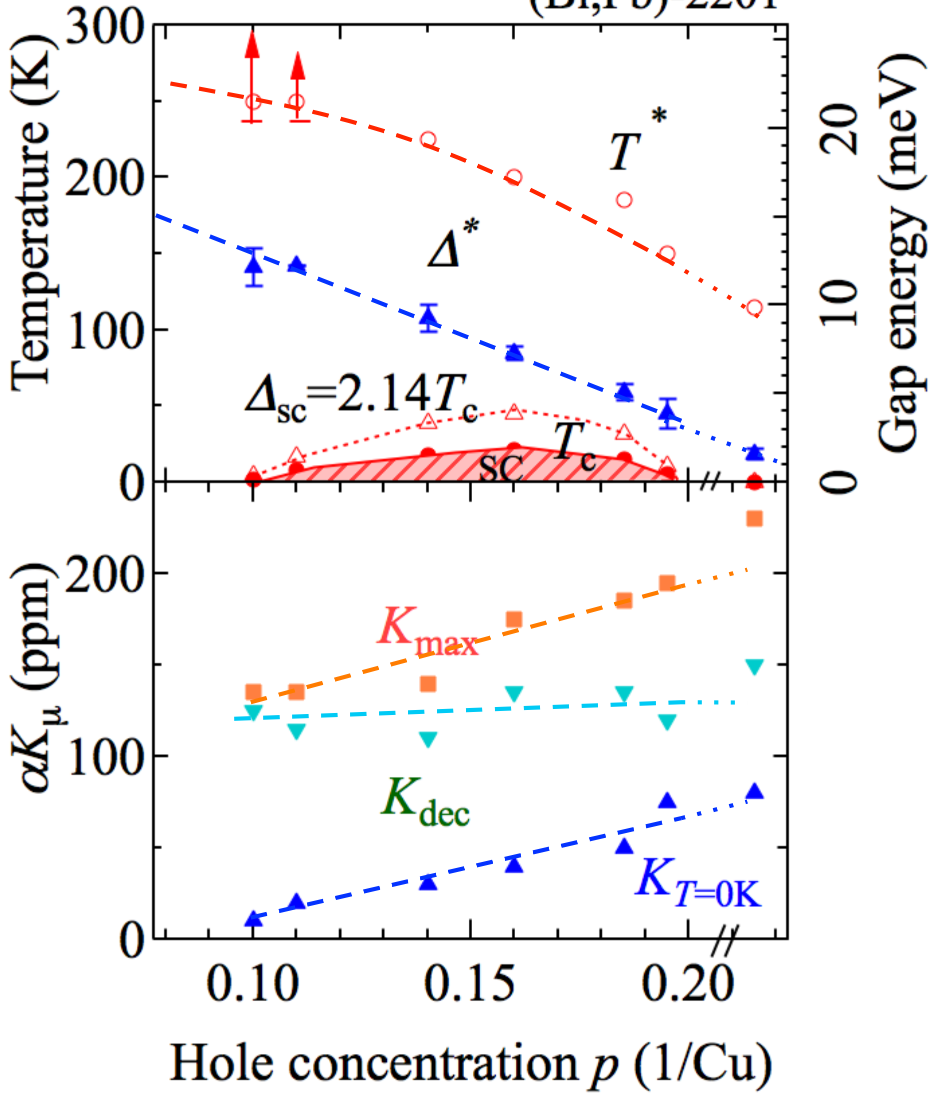
Summary of μSR result

<u>We define PG by Δ^*</u>

Δ^* is the more physical quantity than T^*

$Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$

(Bi,Pb)-2201



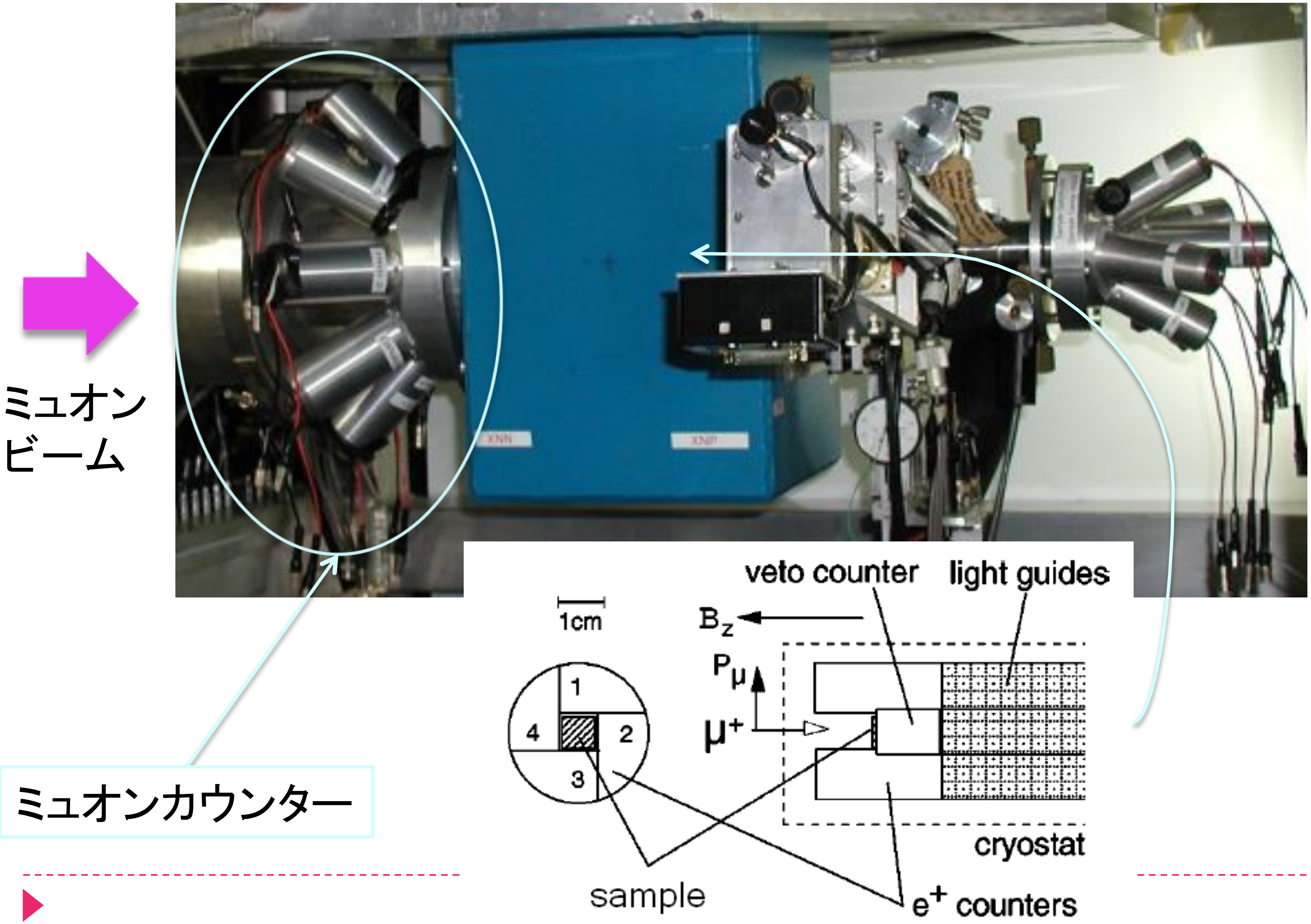
1. Magnitude of Δ^* and its weak correlation with Δ_{sc} over the entire p range seems to *disfavor the "positive" relation of PG to SC"*.
2. $1-N(0)$ [$\propto K_{dec}$] does not seem much to depend on p.
   ⇒ DOS amplitude of PG is independent of p.
3. $N(0)$ [$\propto K_{T=0K}$] increases with p, suggesting phase separation between *anomalous metallic* phase and *normal metalic* phase for $T>T_c$.

   ⇒ A part of $N(0)$ exhibits SC for $T<T_c$.

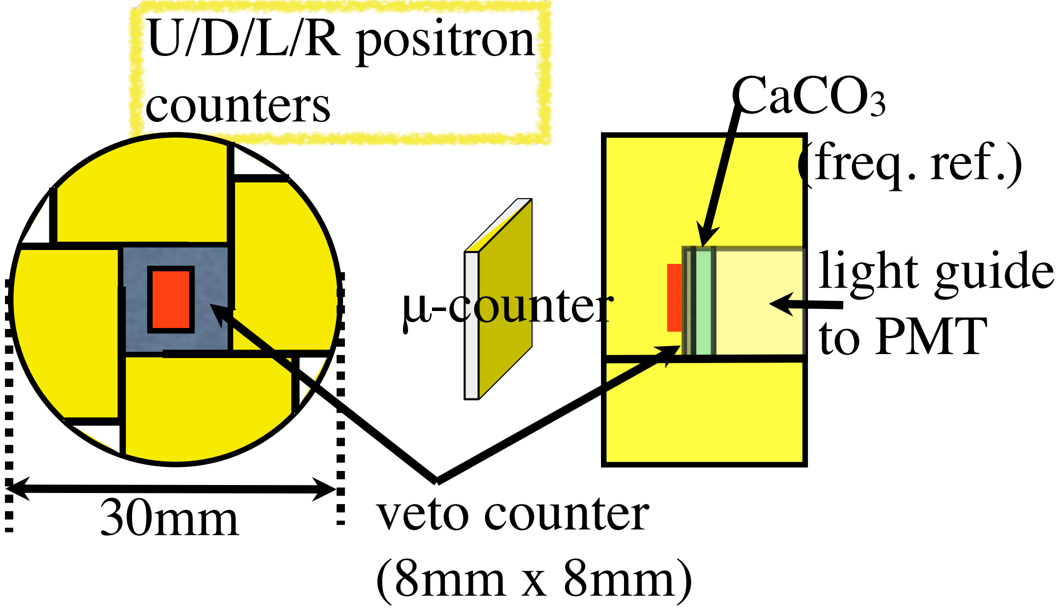
高時間分解能μSR装置「HiTime」@TRIUMF

…コンパクトでシンプルな実験装置

7テスラの磁場(ビーム軸に平行)中にカウンター5個



(by K.M. Kojima, Mar. 2012)



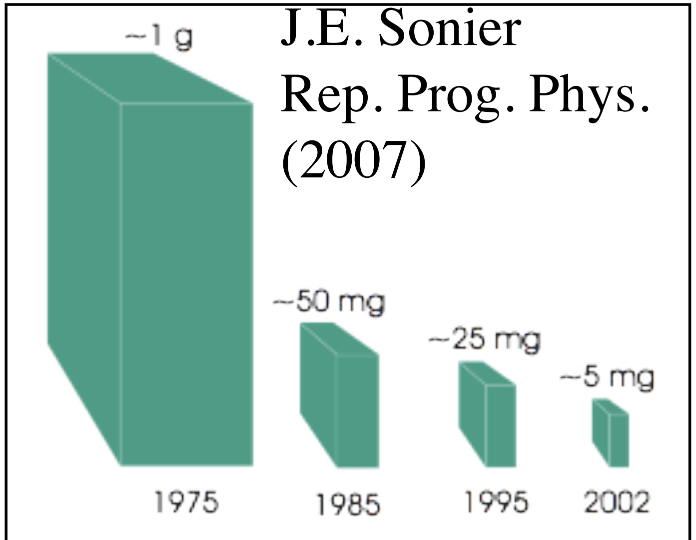
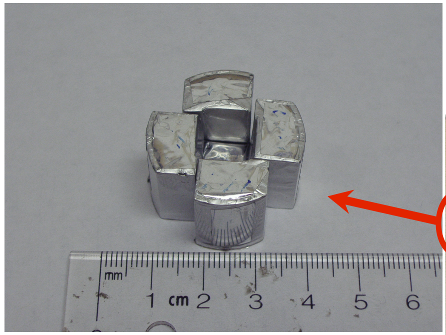
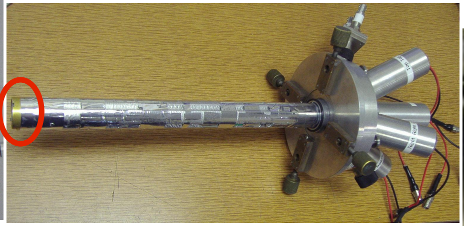
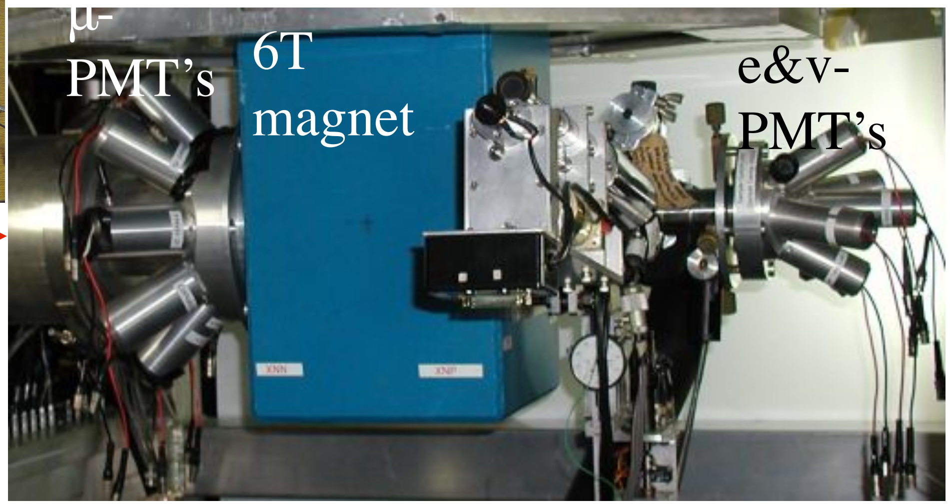


FIG. 3: Evolution of the minimum sample size required for μSR studies of the vortex state.

smaller, the better







beam

HiTime@
TRIUMF

Sample size
>3mm x 3mm
still large, but OK

Summary & Conclusion

1) μSRにとっては「時間スペクトル」がすべての情報で、ビームの時間構造は死活的な重要度を持つ。

2) 1) の観点から見て、dc beamとpulued beamとは基本的に相補的で、win-winの関係になり得る。（中性子利用における定常原子炉中性子源とパルス中性子源の関係に類似。中性子コミュニティも両方の存続を強く希望。）

3) 凝集系物理の分野でμSRが最も大きなインパクトを持っているのは磁性・超伝導などの電子物性物理。

4) 時間分解能を必要とするする高横磁場μSR（ナイトシフト測定等）は「スピンローテータ（兼静電分離器）＋μSR分光器」という比較的小さな投資でスタート可能。→RCNPでミュオン物性研究の立ち上げ初期にインパクトのある成果創出に有利。

Fin