Beyond the Kubo-Toyabe and stretched exponential functions: how μSR can reveal spatial magnetic correlations

P. Dalmas de Réotier, A. Yaouanc, and A. Maisuradze[1]

Institut Nanosciences et Cryogénie
Université Grenoble Alpes & CEA Grenoble, France
[1]Department of Physics, Tbilissi State University, Georgia

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Experimental ZF-μSR spectra

Phenomenological polarization functions

Evidencing spatial correlations

Extension of the KT model

Model-free analysis

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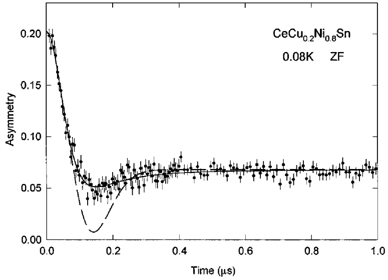
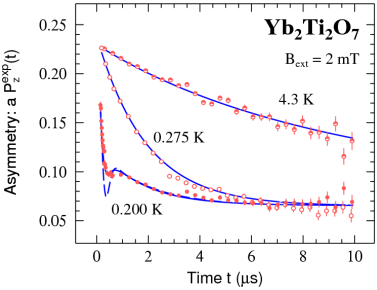
ZF-μSR spectra with unconventional shape

From Kubo-Toyabe-like shape



Non-dilute highly disordered intermetallic alloy.

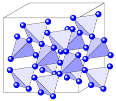
Noakes and Kalvius, PRB **56**, 2352 (1997).



$Yb_2Ti_2O_7$: frustrated magnet on the pyrochlore lattice, quantum spin-ice candidate; splayed FM order below T_c = 0.24 K.

Hodges *et al.*, PRL **88**, 077204 (2002).

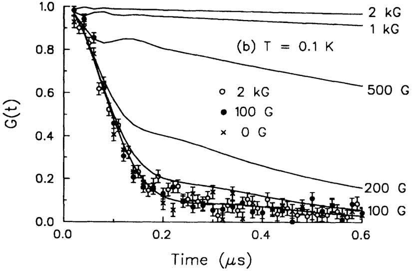
Spectral shape reminiscent of the Kubo-Toyabe function with Gaussian decay at short times, but weaker dip than predicted by Kubo-Toyabe model.



Pyrochlore lattice

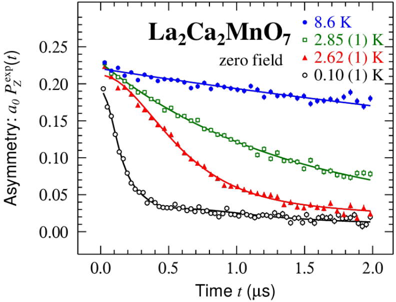
ZF-μSR spectra with unconventional shape (2)

From Kubo-Toyabe-like shape



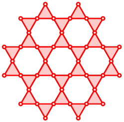
$SrCr_8Ga_4O_{19}$: frustrated magnet on Kagome lattice; $T_g = 3.5$ K.
(Undecouplable Gaussian.)

Uemura *et al.*, PRL **73**, 3306 (1994)

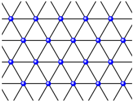


$La_2Ca_2MnO_7$: frustrated magnet on triangular lattice; $\sqrt{3} \times \sqrt{3}$ AFM order below $T_N = 2.8$ K.

Dalmas de Réotier *et al.*, SPIN **5**, 1540001 (2015).



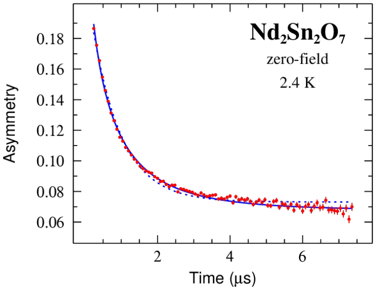
Kagome lattice



Triangular lattice

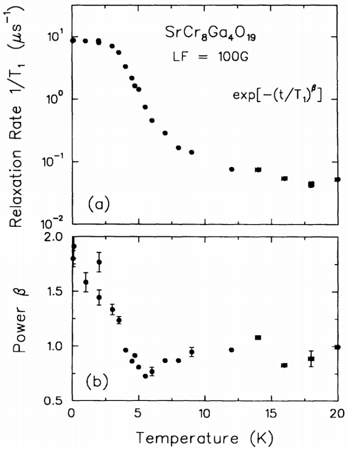
ZF-μSR spectra with unconventional shape (3)

Towards exponential-like shape



$Nd_2Sn_2O_7$: frustrated magnet on the pyrochlore lattice; all-in–all-out AFM order below $T_c = 0.91$ K.

$P_Z(t) = \exp[-(\lambda_Z t)^\beta]$.



$SrCr_8Ga_4O_{19}$

Uemura *et al.*, PRL **73**, 3306 (1994)

$$P_Z(t) = \exp[-(t/T_1)^\beta],$$

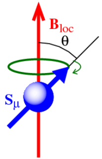
with $1/T_1 \equiv \lambda_Z$.

Outline

The Kubo-Toyabe polarization function

Polarization function from Larmor equation solution:

$$P_Z^{\mathrm{stat}}(t) = \int \left[\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)\right] D_{\mathrm{v}}(\mathbf{B})\mathrm{d}^3\mathbf{B}.$$



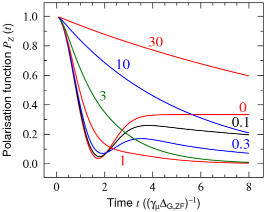
Assume

$$D_{\mathrm{v}}(\mathbf{B}) = D_{\mathrm{c}}(B_X)\,D_{\mathrm{c}}(B_Y)\,D_{\mathrm{c}}(B_Z),$$

with

$$D_{\mathrm{c}}(B_X) = D_{\mathrm{c}}(B_Y) = D_{\mathrm{c}}(B_Z) \propto \exp\left[\frac{-(B_Z)^2}{2\Delta^2}\right],$$

$$P_Z^{\mathrm{KT}}(\Delta, t) = \frac{1}{3} + \frac{2}{3}\left(1 - \gamma_\mu^2\Delta^2 t^2\right)\exp\left(-\frac{\gamma_\mu^2\Delta^2 t^2}{2}\right).$$



$P_Z(t)$.

Dynamics is accounted for by the so-called strong collision model:

$$P_Z(t) = P_Z^{\mathrm{KT}}(t)\exp(-\nu_c t) + \nu_c\int_0^t P_Z(t-t')P_Z^{\mathrm{KT}}(t')\exp(-\nu_c t')\mathrm{d}t'$$

When $\nu_c/\gamma_\mu\Delta \gg 1$, $P_Z(t) \to \exp(-\lambda_Z t)$ with $\lambda_Z \equiv 2\gamma_\mu^2\Delta^2/\nu_c$.

Hayano *et al.*, PRB **20**, 850 (1979).

Phenomenological fit functions (1)

The exponential-power polarization function

$$P_Z(t) = \exp[-(\lambda_Z t)^\beta]$$

- $0 < \beta < 1$: stretched-exponential function (or Kohlrausch function, 1854).

A distribution of exponential relaxation functions:

$$\exp[-(\lambda_Z t)^\beta] = \int_0^\infty \exp(-s\lambda_Z t) P(s,\beta) \mathrm{d}s.$$

Physical ground for distribution $P(s,\beta)$?

- $\beta > 1$: compressed-exponential function.

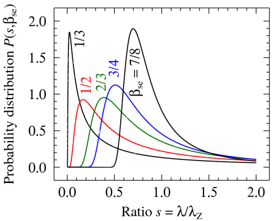
Rarely appearing in physics except for $\beta = 2$.
No physical backing in ZF-μSR, even for $\beta = 2$.

- $\beta = 1/2$: singular case.

$$P_Z(t) = \exp\left(-\sqrt{\lambda_Z t}\right),$$

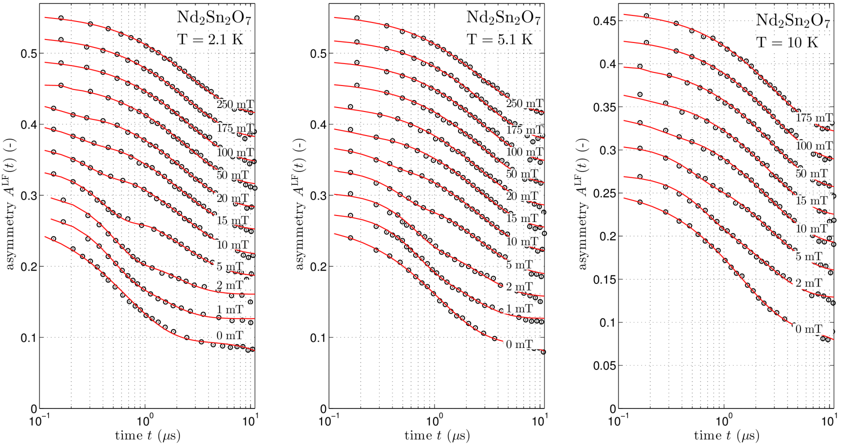
is derived for *diluted* magnetic systems in the extreme motional narrowing limit.

Experimental confirmation by Tse and Hartmann, PRL **21**, 511 (1968), Uemura *et al.*, PRB **31**, 546 (1985)...



$P(s,\beta)$ versus s.
Johnston, PRB **74**, 184430 (2006).

A complete set of high statistics data can reveal the fate of stretched-exponential spectra:



Simultaneous fit of data with dynamical Kubo-Toyabe model: slow spin tunnelling in the paramagnetic phase of $Nd_2Sn_2O_7$.

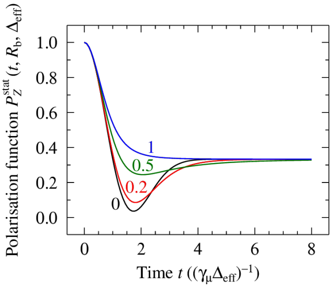
Dalmas de Réotier *et al.*, PRB **95**, 134420 (2017).

Phenomenological fit functions (2)

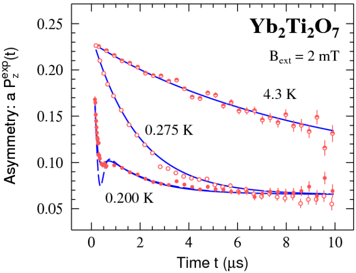
The Gaussian-broadened Gaussian polarization function. [Noakes and Kalvius, PRB **56**, 2352 (1997)]

Average of Kubo-Toyabe polarization functions with Gaussian-distributed field widths:

$$P_Z^{\mathrm{GbG}}(t) = \frac{1}{\sqrt{2\pi}\Delta_{\mathrm{GbG}}} \int_{-\infty}^{\infty} P_Z^{\mathrm{KT}}(\Delta, t) \exp\left(-\frac{(\Delta-\Delta_0)^2}{2\Delta_{\mathrm{GbG}}^2}\right) \mathrm{d}\Delta.$$



$P_Z^{\mathrm{GbG}}(t)$ *as a function of* $R \equiv \Delta_{\mathrm{GbG}}/\Delta_0$, *with* $\Delta_{\mathrm{eff}}^2 \equiv \Delta_0^2 + \Delta_{\mathrm{GbG}}^2$.



Full line at 0.200 K: $P_Z^{\mathrm{GbG}}(t)$.

Hodges *et al.*, PRL **88**, 077204 (2002).

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$$P_Z^{\rm stat}(t) = \int \left[\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)\right] D_{\rm v}(\mathbf{B}){\rm d}^3\mathbf{B}$$

Assume

$$D_{\rm v}(\mathbf{B}) = D_{\rm c}(B_X)\, D_{\rm c}(B_Y)\, D_{\rm c}(B_Z)$$

and

$$D_{\rm c}(B_X) = D_{\rm c}(B_Y) = D_{\rm c}(B_Z).$$

No linear relation between field distribution and asymmetry spectrum!

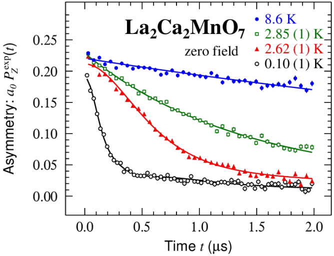
Direct search for $D_{\rm c}(B_Z)$ from the data not possible.

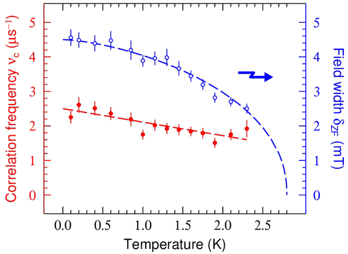
Two routes for $D_{\rm c}(B_Z)$ determination:

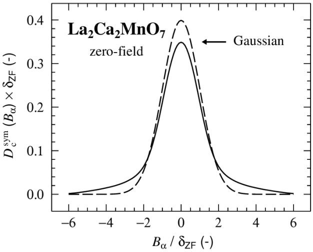
- $D_{\rm c}(B^Z) \propto \exp\left(\frac{-B_Z^2}{2\Delta^2}\right) \longrightarrow D_{\rm c}(B_Z) \propto \exp\left[-g\left(\frac{B_Z}{\delta}\right)\right]$ with $g(x) = \frac{1}{2}x^2 + \frac{1}{3}(\eta_3 x)^3 + \frac{1}{4}(\eta_4 x)^4$.
- Direct search for distribution consistent with the data. Use of Maximum Entropy supplemented Reverse Monte Carlo (ME-RMC) algorithm.

Application to the triangular magnet $La_2Ca_2MnO_7$

Magnetically ordered phase







Model function simultaneously fitted to 14 spectra recorded in the ordered phase.

A single value for $\eta_3 = 0.74\,(2)$ and $\eta_4 = 0.47\,(2)$.

Tails in the distribution.

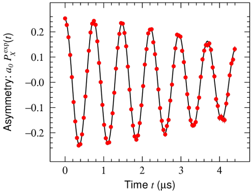
Dalmas de Réotier *et al.*, SPIN **5**, 1540001 (2015).

Outline

Field distribution in transverse field experiments

- Traditional method: Fourier transform of the asymmetry spectrum

  Caveats:
  - noise in asymmetry data (finite μ^+ lifetime) is not accounted for:
    apodization $\longrightarrow$ broadening of distribution
  - no error bars on the resulting distribution



- A better approach: inverse problem

$$P_X^{\mathrm{stat}} = \int \cos(\omega_\mu t) D_{\mathrm{c}}(B_Z) \mathrm{d}B_Z$$

  - find the distributions which provide the best fit to the data
  - among the solutions, choose that with maximum entropy (ME) S

$$S = -\sum_i D_{\mathrm{c}}(B_{Z,i}) \delta B_Z \log \left[\frac{D_{\mathrm{c}}(B_{Z,i})}{d_i} \right] \quad \text{(information theory)}$$

δB_Z: step in $D_c(B_{Z,i})$; d_i prior estimate

Rainford and Daniell, Hyperfine Interact. **87** 1129 (1994)
Riseman and Forgan, Physica **B 289-290** 718 (2000)

Case of zero-field asymmetry spectra

$$P_Z^{\mathrm{stat}}(t) = \int \left[\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)\right] D_{\mathrm{c}}(B_X)\, D_{\mathrm{c}}(B_Y)\, D_{\mathrm{c}}(B_Z)\mathrm{d}B_X\mathrm{d}B_Y\mathrm{d}B_Z$$

- direct search for distribution which best fits the data
- minimization of

$$F = \chi^2 - \lambda S$$

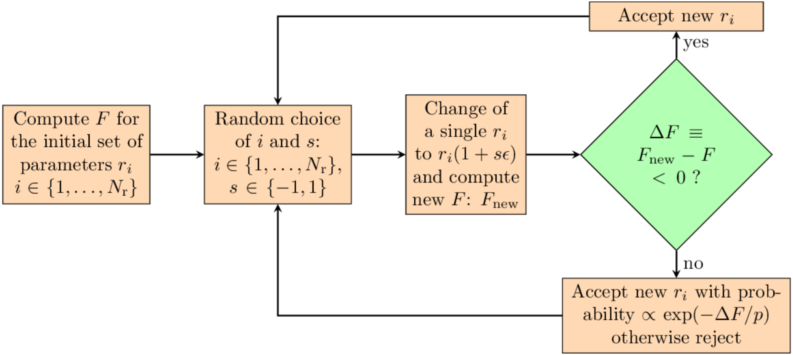
where

$$\chi^2 = \sum_i^{N_i} \frac{\left[A_i - a_0 P_Z(t_i)\right]^2}{\sigma_i^2},$$

and λ is a Lagrange parameter.

Minimization with a Reverse Monte Carlo (RMC) algorithm.

Reverse Monte Carlo algorithm



with $0.003 \lesssim \varepsilon \lesssim 0.03$, and $0.003 \lesssim p \lesssim 0.03$.
Convergence is typically reached after ≈100 loops per degree of freedom.

Estimate of error bars

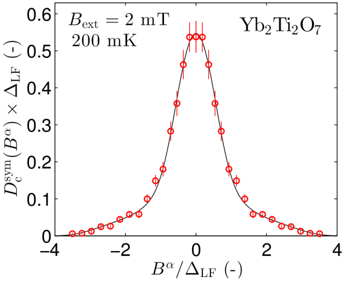
$$\begin{aligned}\delta F(\mathbf{r}) &\simeq \sum_i \frac{\partial F}{\partial r_i}\delta r_i + \frac{1}{2}\sum_{i,j}\frac{\partial^2 F}{\partial r_i \partial r_j}\delta r_i \delta r_j \\ &\simeq \frac{1}{2}\sum_{i,j}\frac{\partial^2 F}{\partial r_i \partial r_j}\delta r_i \delta r_j \equiv \sum_{i,j}\frac{1}{2}\mathcal{H}_{i,j}\delta r_i \delta r_j.\end{aligned}$$

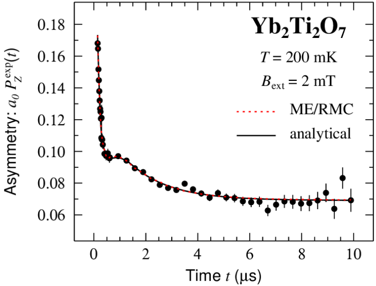
where $\mathbf{r}$ is the vector formed by the free parameters $[D_{\mathrm{c}}(B_i)$, a_0, ν_c, $a_{\mathrm{bg}}, \ldots]$ of the fit and $\mathcal{H}$ is the so-called Hessian matrix.
The error bars σ_{r_i} are given by:

$$\sigma_{r_i}^2 = \left[\mathcal{H}^{-1}\right]_{ii}.$$

Outline

Comparison of analytical model and ME-RMC analysis



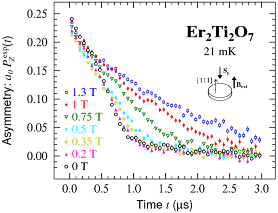


- Full line: fit to the asymmetry data with $D_c(B_Z) \propto \exp\left[-g\left(\frac{B_Z}{\delta}\right)\right]$ and $g(x) = \frac{1}{2}x^2 + \frac{1}{3}(\eta_3 x)^3 + \frac{1}{4}(\eta_4 x)^4$. $\eta_3 = 0.73\,(2)$, $\eta_4 = 0.46\,(2)$.
- Red circles: ME-RMC fit to the asymmetry data.

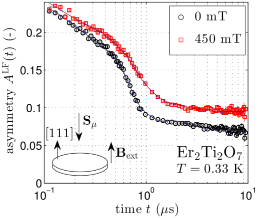
Comparison in time domain.

Application of the ME-RMC algorithm

$Er_2Ti_2O_7$: a XY pyrochlore antiferromagnet with $T_N = 1.25$ K



Dalmas de Réotier *et al*,
PRB **86**, 104424 (2012)



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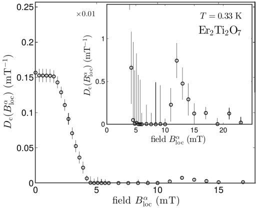

To be published

- thanks to the ME-RMC algorithm, and the availability of a high statistic spectrum, evidence for a weak contribution centered at 12 mT
- allows for a reliable fit in *time domain* of the asymmetry spectrum

Evidence for short-range correlations

- Non-Gaussian distribution $\longrightarrow$ short-range magnetic correlations
  Contrapositive statement of the Central Limit Theorem
- Coexistence of short-range correlations with long-range order for $La_2Ca_2MnO_7$, $Yb_2Ti_2O_7$, $Yb_2Sn_2O_7$, and $Er_2Ti_2O_7$
  Possible relation with magnetic moment fragmentation
- What next?
  - Quantitative information about the correlation length (Monte Carlo simulations)
  - Quantitative information in terms of physical parameters entering a Hamiltonian
    See, e.g. Bramwell *et al.*, PRE **63**, 041106 (2001), who calculated the magnetic moment distribution for the classical XY Hamiltonian on the 2D square lattice (BKT transition).
    Required:
    - extension to $\mathbf{B}_{\rm loc}$ at the muon
    - extension to other Hamiltonians.

Outline

Summary and Conclusions

- Framework for the interpretation of magnetic materials spectra with unconventional shape
  - An analytical model
  - A model-free analysis using the ME-RMC algorithm
  - Both focussed for ZF data and isotropic distributions
    Generalization to LF data straightforward
    Generalization to anisotropic distributions possible
- μSR is primarily sensitive to time correlations, but a detailed analysis of large statistics data can unravel spatial correlations
- Despite the muons are a local probe, they can evidence spatial correlations

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