Relaxation functions

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Outline

Foreword

- Polarisation function vs relaxation function
- Statistics, probability and stochastic processes theory
- Most methodologies apply to transverse and longitudinal polarisation functions
- Background for the lecture is in the framework of magnetism or sometimes the diffusion of a light interstitial in a crystal
- Reference: chapters 6 and 7 (marginally 5 and 10) of *Muon Spin Rotation, Relaxation and Resonance: Applications to Condensed Matter*, (Oxford University Press, Oxford, 2011) by A. Yaouanc and P. Dalmas de Réotier

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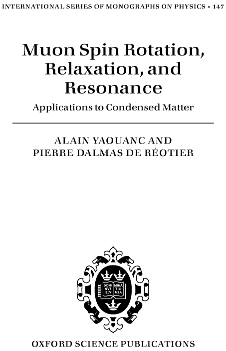
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Muon Spin Rotation, Relaxation, and Resonance

Applications to Condensed Matter

ALAIN YAOUANC AND PIERRE DALMAS DE RÉOTIER

OXFORD SCIENCE PUBLICATIONS

Outline

The evolution of the muon spin $\mathbf{S}_\mu(t)$

The Larmor equation

Basic principle of mechanics:
Time derivative of angular momentum is equal to the sum of the torques:

$$\frac{\mathrm{d}\hbar\mathbf{S}_\mu(t)}{\mathrm{d}t} = \mathfrak{m}_\mu(t) \times \mathbf{B}_{\rm loc}(t).$$

Since

$$\mathfrak{m}_\mu = \gamma_\mu \hbar \mathbf{S}_\mu,$$

by definition of the gyromagnetic ratio, we have

$$\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t} = \gamma_\mu \, \mathbf{S}_\mu(t) \times \mathbf{B}_{\rm loc}(t).$$

$\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1}.

Basics of motion properties deriving from the Larmor equation

From

$$\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t} = \gamma_\mu \, \mathbf{S}_\mu(t) \times \mathbf{B}_{\mathrm{loc}}(t)$$

we deduce:

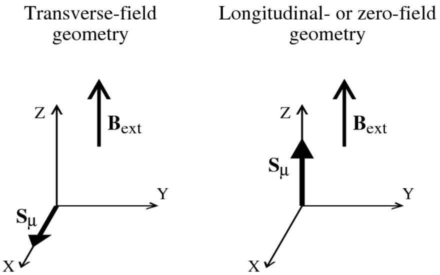
- $\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t} \cdot \mathbf{S}_\mu(t) = 0$:
  $S_\mu(t)$ is a constant of the motion, *i.e.* $S_\mu(t) = S_\mu(0)$
- $\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t} \cdot \mathbf{B}_{\mathrm{loc}}(t) = 0$:
  this implies $\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t}$ is perpendicular to $\mathbf{B}_{\mathrm{loc}}(t)$.

The transverse and longitudinal polarisation functions

- The polarisation function $P_\alpha(t)$ is the evolution of the projection of the muon ensemble polarisation along axis α:

$$P_\alpha(t) = \left\langle \frac{S_{\mu,\alpha}(t)}{S_\mu} \right\rangle .$$

- $\mathbf{S}_\mu \equiv \mathbf{S}_\mu(t=0)$: initial muon beam polarisation



Convention for the axes:
$\mathbf{B}_{\mathrm{ext}}$ *is always parallel to* $\mathbf{Z}$.

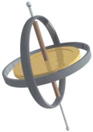
- in transverse field experiment: $\mathbf{S}_\mu \parallel \mathbf{X} \rightarrow P_X(t)\,\mathrm{or}\,P_Y(t)$.
- in zero-field and longitudinal-field experiment: $\mathbf{S}_\mu \parallel \mathbf{Z} \rightarrow P_Z(t)$.

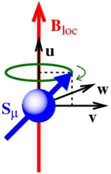
The muon spin evolution in a static field

Recall the Larmor equation,

$$\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t} = \gamma_\mu \, \mathbf{S}_\mu(t) \times \mathbf{B}_{\mathrm{loc}}(t).$$

Assuming $\mathbf{B}_{\mathrm{loc}}(t) = \mathbf{B}_{\mathrm{loc}}$, the solution is a precession motion:





$$\mathbf{S}_\mu(t) = S_\mu^{\parallel}(0)\,\mathbf{u} + S_\mu^{\perp}(0)[\cos(\omega_\mu t)\,\mathbf{v} - \sin(\omega_\mu t)\,\mathbf{w}],$$

with $\omega_\mu = \gamma_\mu B_{\mathrm{loc}}$.

The precession frequency only depends on B_{loc}, not on the angle between $\mathbf{S}_\mu$ and $\mathbf{B}_{\mathrm{loc}}$!

Outline

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Outline

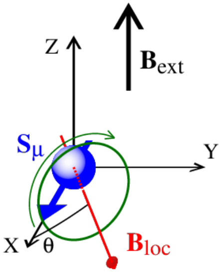
Transverse-field polarisation function

Per definition, $\mathbf{S}_\mu \equiv \mathbf{S}_\mu(t=0) \parallel \mathbf{X}$.
From the solution of the Larmor equation,

$$S_\mu^X(t) = S_\mu \left\{ \left(\frac{B_{\rm loc}^X}{B_{\rm loc}}\right)^2 + \left[1 - \left(\frac{B_{\rm loc}^X}{B_{\rm loc}}\right)^2\right] \cos(\omega_\mu t) \right\},$$

$$S_\mu^X(t) = S_\mu \left[\cos^2\theta + \sin^2\theta \cos(\omega_\mu t) \right],$$

with $B_{\rm loc}^2 = \left(B_{\rm loc}^X\right)^2 + \left(B_{\rm loc}^Y\right)^2 + \left(B_{\rm loc}^Z\right)^2$, and $\omega_\mu = \gamma_\mu B_{\rm loc}$.



Let $D_v(\mathbf{B}_{\rm loc})$ be the distribution of static fields probed by the muons,

$$P_X^{\rm stat}(t) = \left\langle \frac{S_\mu^X(t)}{S_\mu} \right\rangle = \int [\cos^2\theta + \sin^2\theta \cos(\omega_\mu t)] D_v(\mathbf{B}_{\rm loc})\, \mathrm{d}^3\mathbf{B}_{\rm loc}.$$

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Transverse-field polarisation function

Example: single field

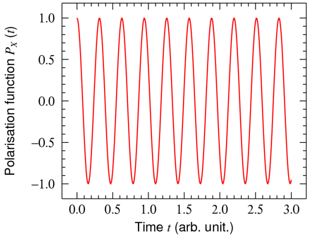
Recall,

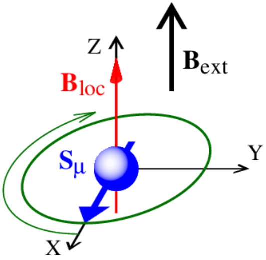
$$P_X^{\mathrm{stat}}(t) = \int [\cos^2\theta + \sin^2\theta \cos(\omega_\mu t)] D_v(\mathbf{B}_{\mathrm{loc}})\, \mathrm{d}^3\mathbf{B}_{\mathrm{loc}}.$$

Assume all the muons to be submitted to $\mathbf{B}_{\mathrm{loc}} = \mathbf{B}_0 \parallel \mathbf{Z}$, i.e. $\theta = \pi/2$,

$$P_X^{\mathrm{stat}}(t) = \cos(\omega_0 t)$$

with $\omega_0 = \gamma_\mu B_0$.





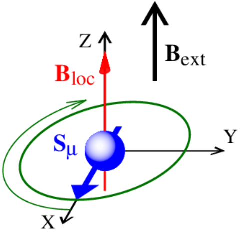
Transverse-field polarisation function

Large transverse field

Recall,

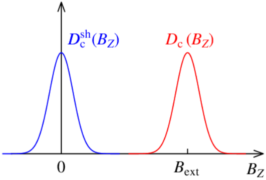
$$P_X^{\mathrm{stat}}(t) = \int [\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)] D_v(\mathbf{B}_{\mathrm{loc}})\, \mathrm{d}^3\mathbf{B}_{\mathrm{loc}}.$$

Suppose $\mathbf{B}_{\mathrm{loc}}$ to be dominated by $\mathbf{B}_{\mathrm{ext}}$, i.e. $\theta \approx \pi/2$, $B_{\mathrm{loc}} \approx \left|B_{\mathrm{loc}}^Z\right|$,



$$\begin{aligned} P_X^{\mathrm{stat}}(t) &= \int \cos(\omega_\mu t) D_c(B_{\mathrm{loc}}^Z)\, \mathrm{d}B_{\mathrm{loc}}^Z, \\ &= \underbrace{\left[\int D_c^{\mathrm{sh}}(x)\cos(\gamma_\mu t x)\, \mathrm{d}x\right]}_{\text{characteristic function}} \cos(\gamma_\mu B_{\mathrm{ext}} t). \end{aligned}$$

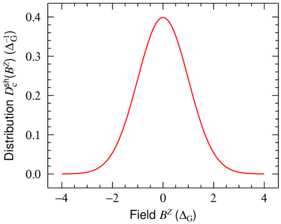
The last line is obtained after the substitution $B_{\mathrm{loc}}^Z = B_{\mathrm{ext}} + x$. $D_c^{\mathrm{sh}}(x)$ is assumed to be an even function, otherwise a phase shift is present.



Transverse-field polarisation function

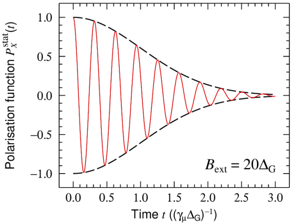
Example: typical distributions and associated polarisation functions

Gaussian distribution:



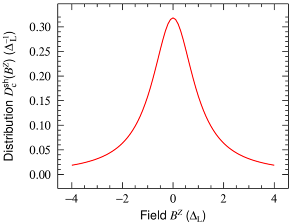
$$D_c^{\mathrm{sh}}(B) = \frac{1}{\sqrt{2\pi}\Delta_{\mathrm{G}}} \exp\left(\frac{-B^2}{2\Delta_{\mathrm{G}}^2}\right)$$

$$P_X^{\mathrm{stat}}(t) = \exp\left(\frac{-\gamma_\mu^2 \Delta_{\mathrm{G}}^2 t^2}{2}\right) \times \cos(\gamma_\mu B_{\mathrm{ext}} t)$$



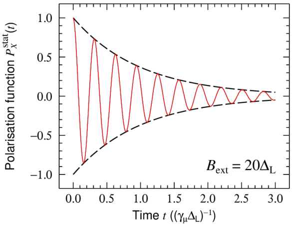
Example: nuclear dipoles

Lorentzian distribution:



$$D_c^{\mathrm{sh}}(B) = \frac{1}{\pi} \frac{\Delta_{\mathrm{L}}}{\Delta_{\mathrm{L}}^2 + B^2}$$

$$P_X^{\mathrm{stat}}(t) = \exp\left(-\gamma_\mu \Delta_{\mathrm{L}} t\right) \times \cos(\gamma_\mu B_{\mathrm{ext}} t)$$

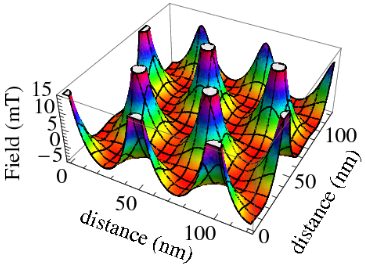


Example: diluted magnetic systems

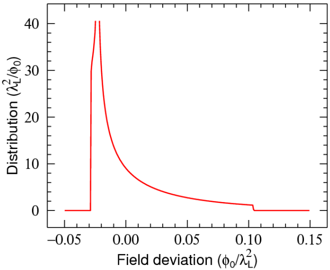
Transverse-field polarisation function

Example: Mixed phase of superconductors

Type II superconductors submitted to a magnetic field:



Field (deviation) profile in the flux-line lattice phase.



Associated field distribution.

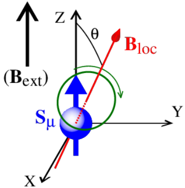
Outline

Zero- or longitudinal-field polarisation function

Per definition, $\mathbf{S}_\mu \equiv \mathbf{S}_\mu(t=0) \parallel \mathbf{Z}$.

From the solution of the Larmor equation,

$$
\begin{aligned}
S_\mu^Z(t) &= S_\mu \left\{ \left(\frac{B_{\rm loc}^Z}{B_{\rm loc}} \right)^2 + \left[1 - \left(\frac{B_{\rm loc}^Z}{B_{\rm loc}} \right)^2 \right] \cos(\omega_\mu t) \right\}, \\
S_\mu^Z(t) &= S_\mu \left[\cos^2\theta + \sin^2\theta \cos(\omega_\mu t) \right],
\end{aligned}
$$



with $B_{\rm loc}^2 = \left(B_{\rm loc}^X\right)^2 + \left(B_{\rm loc}^Y\right)^2 + \left(B_{\rm loc}^Z\right)^2$ and $\omega_\mu = \gamma_\mu B_{\rm loc}$.

Let $D_v(\mathbf{B}_{\rm loc})$ be the distribution of static fields probed by the muons,

$$P_Z^{\rm stat}(t) = \left\langle \frac{S_\mu^Z(t)}{S_\mu} \right\rangle = \int [\cos^2\theta + \sin^2\theta \cos(\omega_\mu t)] D_v(\mathbf{B}_{\rm loc})\, \mathrm{d}^3\mathbf{B}_{\rm loc}.$$

Zero-field polarisation function

Case of isotropic distribution

Recall

$$P_Z^{\mathrm{stat}}(t) = \int [\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)] D_v(\mathbf{B}_{\mathrm{loc}})\, \mathrm{d}^3\mathbf{B}_{\mathrm{loc}}.$$

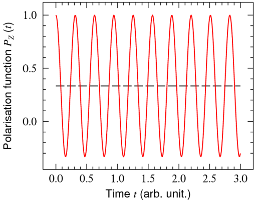
Assume $D_v(\mathbf{B}_{\mathrm{loc}})\, \mathrm{d}^3\mathbf{B}_{\mathrm{loc}} = D_v(B_{\mathrm{loc}}) B_{\mathrm{loc}}^2 \mathrm{d}B_{\mathrm{loc}} \sin\theta\, \mathrm{d}\theta\, \mathrm{d}\varphi$,

$$P_Z^{\mathrm{stat}}(t) = \frac{1}{3} + \frac{2}{3}\int 4\pi D_v(B_{\mathrm{loc}}) B_{\mathrm{loc}}^2 \cos(\omega_\mu t)\, \mathrm{d}B_{\mathrm{loc}},$$

with $\omega_\mu = \gamma_\mu B_{\mathrm{loc}}$.

Example: $4\pi D_v(B_{\mathrm{loc}}) B_{\mathrm{loc}}^2 = \delta(B_{\mathrm{loc}} - B_0)$
ideal magnetic polycrystal

$$P_Z^{\mathrm{stat}}(t) = \frac{1}{3} + \frac{2}{3}\cos(\gamma_\mu B_0 t)$$



Zero-field polarisation function

Example: Maxwell-Boltzmann distribution for B_{loc}

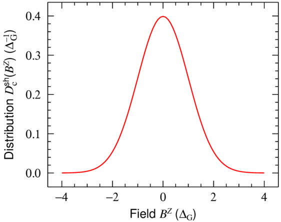
For isotropic Gaussian distributed B^{α} with rms Δ_{G},

$$D_v(\mathbf{B})\,\mathrm{d}^3\mathbf{B} = \left(\frac{1}{\sqrt{2\pi}\Delta_{\mathrm{G}}}\right)^3 \exp\left(\frac{-B^2}{2\Delta_{\mathrm{G}}^2}\right) B^2\,\mathrm{d}B \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi,$$

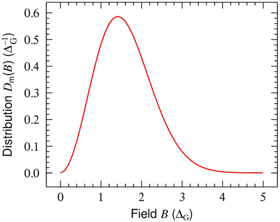
$$D_m(B) = 4\pi D_v(B) B^2,$$

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

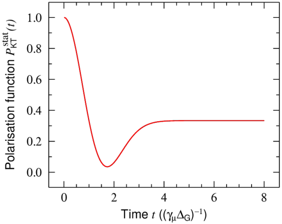
which is the so-called Kubo-Toyabe function.



Component distribution



Modulus distribution



Kubo-Toyabe function. Minimum at $t = \sqrt{3}/\gamma_\mu\Delta_{\mathrm{G}}$

Outline

Effect of external field

Case of transverse $\mathbf{B}_{\rm ext}$

If $\mathbf{B}_{\rm ext}$ is strong enough, recall

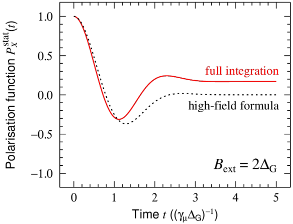
$$P_X^{\rm stat}(t) \quad = \quad \underbrace{\left[\int D_c^{\rm sh}(x)\cos(\gamma_\mu t x)\,\mathrm{d}x\right]}_{\text{damping factor}}\ \underbrace{\cos(\gamma_\mu B_{\rm ext}\, t)}_{\text{oscillating factor}}\ .$$

Trivial effect of $\mathbf{B}_{\rm ext}$ on oscillation frequency.

If width of distribution is non-negligible compared to $\mathbf{B}_{\rm ext}$, resort to general formula

$$P_X^{\rm stat}(t) = \int [\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)] D_v(\mathbf{B}_{\rm loc})\,\mathrm{d}^3\mathbf{B}_{\rm loc}.$$

Example: Gaussian field distribution



Towards the Kubo-Toyabe function

Effect of external field

Case of longitudinal $\mathbf{B}_{\mathrm{ext}}$

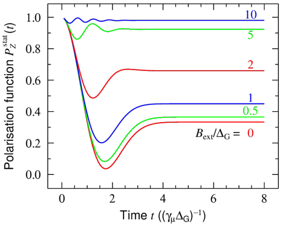
Recall,

$$P_Z^{\mathrm{stat}}(t) = \int [\cos^2\theta + \sin^2\theta\cos(\omega_\mu t)] D_v(\mathbf{B})\,\mathrm{d}^3\mathbf{B},$$

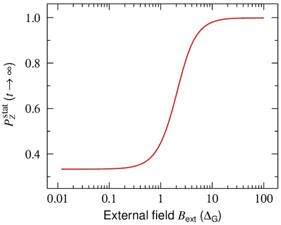
and the former isotropic Gaussian distribution.

Now the Z component of $D_v(\mathbf{B})$ is shifted:

$$D_v(\mathbf{B})\,\mathrm{d}^3\mathbf{B} = \left(\frac{1}{\sqrt{2\pi}\Delta_{\mathrm{G}}}\right)^3 \exp\left(\frac{-B_X^2 - B_Y^2}{2\Delta_{\mathrm{G}}^2}\right) \exp\left(\frac{-(B_Z - B_{\mathrm{ext}})^2}{2\Delta_{\mathrm{G}}^2}\right) \mathrm{d}B_X \mathrm{d}B_Y \mathrm{d}B_Z.$$



- at large field: muon spin decoupling
- oscillations at $\gamma_\mu B_{\mathrm{ext}}$
- field dependence serves to ascertain the model
- sensitivity in the range $\Delta_{\mathrm{G}}/5 \lesssim B_{\mathrm{ext}} \lesssim 5\Delta_{\mathrm{G}}$



Outline

Outline

Origin of field at the muon site

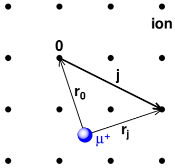
- nuclei
  - high concentration of magnetic moments
  - quasi-static on τ_μ scale
  - disordered and no correlation
- electrons
  - high concentration of magnetic moments/structural order
    $\longrightarrow$ magnetically ordered phase
    $\longrightarrow$ paramagnetic phase (dynamical on τ_μ scale)
  - low concentration of magnetic moments/structural disorder (spin-glass)
    $\longrightarrow$ frozen state
    $\longrightarrow$ paramagnetic state (dynamical on τ_μ scale)

muon life time $\tau_\mu = 2.2\ \mu$s

The magnetic field at the muon site

Dipolar and Fermi contact fields

The dipolar field arising from localized spins $\mathbf{J}_j$ with Landé factors g is



$$\mathbf{B}_{\mathrm{dip}} = -\frac{\mu_0}{4\pi} g \mu_{\mathrm{B}} \sum_j \left[-\frac{\mathbf{J}_j}{r_j^3} + 3\frac{(\mathbf{J}_j \cdot \mathbf{r}_j)\mathbf{r}_j}{r_j^5} \right].$$

$\mathbf{r}_j$ is the vector distance from the spin to the muon.

When a polarised electron density is present at the muon, an additional contribution is present, the Fermi contact field:

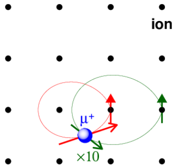
$$\mathbf{B}_{\mathrm{con}} = -\frac{\mu_0}{4\pi} g \mu_{\mathrm{B}} \sum_{j \in \mathrm{NN}} H_j \mathbf{J}_j.$$

Only the muon nearest neighbors (NN) usually contribute to $\mathbf{B}_{\mathrm{con}}$.

When both $\mathbf{B}_{\mathrm{dip}}$ and $\mathbf{B}_{\mathrm{con}}$ contribute to $\mathbf{B}_{\mathrm{loc}}$ (*i.e.* in metals) they generally have the same order of magnitude.

The magnetic field at the muon site

Dipolar and Fermi contact fields



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The magnetic field at the muon site

Dipolar and Fermi contact fields

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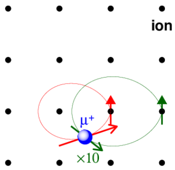

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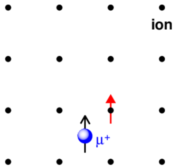


Only the muon nearest neighbors (NN) usually contribute to $\mathbf{B}_{\mathrm{con}}$.

When both $\mathbf{B}_{\mathrm{dip}}$ and $\mathbf{B}_{\mathrm{con}}$ contribute to $\mathbf{B}_{\mathrm{loc}}$ (*i.e.* in metals) they generally have the same order of magnitude.

The magnetic field at the muon site

Reciprocal space

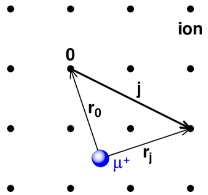
$\mathbf{B}_{\rm dip}$ and $\mathbf{B}_{\rm con}$ linearly depending on $\mathbf{J}_j$,

$$\mathbf{B}_{\rm loc} = \mathbf{B}_{\rm dip} + \mathbf{B}_{\rm con} = -\frac{\mu_0}{4\pi}\frac{g\mu_{\rm B}}{v_{\rm c}}\sum_j \boldsymbol{G}_j \mathbf{J}_j.$$

$\boldsymbol{G}_j$ is the muon-spin j coupling tensor.

It is often a good idea to introduce the Fourier space quantities:

$$\begin{aligned}\boldsymbol{G}_{\mathbf{q}} &= \sum_j \boldsymbol{G}_j \exp(i\mathbf{q}\cdot\mathbf{r}_j),\\ \mathbf{J}_{\mathbf{q}} &= \frac{1}{\sqrt{n_c}}\sum_j \mathbf{J}_j \exp(-i\mathbf{q}\cdot\mathbf{j}).\end{aligned}$$



Then,

$$\mathbf{B}_{\rm loc} = -\frac{\mu_0}{4\pi}\frac{g\mu_{\rm B}}{\sqrt{n_c}\,v_{\rm c}}\sum_{\mathbf{q}} \exp(-i\mathbf{q}\cdot\mathbf{r}_0)\boldsymbol{G}_{\mathbf{q}}\mathbf{J}_{\mathbf{q}}.$$

Origin of field at the muon site

- nuclei
  - high concentration of magnetic moments
  - quasi-static on τ_μ scale
  - disordered and no correlation
- electrons
  - high concentration of magnetic moments/structural order
    $\longrightarrow$ magnetically ordered phase
    $\longrightarrow$ paramagnetic phase (dynamical on τ_μ scale)
  - low concentration of magnetic moments/structural disorder (spin-glass)
    $\longrightarrow$ frozen state
    $\longrightarrow$ paramagnetic state (dynamical on τ_μ scale)

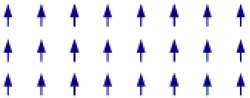
muon life time $\tau_\mu = 2.2\ \mu$s

Outline

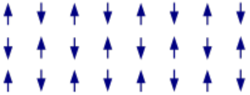
Zero-field polarisation function in magnets

Reminder:

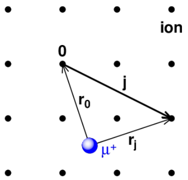
- $\mathbf{J_q} = \frac{1}{\sqrt{n_C}} \sum_\mathbf{j} \exp(-i\mathbf{q}\cdot\mathbf{j})\mathbf{J_j}$, $\mathbf{J_j} = \frac{1}{\sqrt{n_C}} \sum_\mathbf{q} \exp(i\mathbf{q}\cdot\mathbf{j})\mathbf{J_q}$
- Ferromagnet: $\mathbf{J_{q=0}}$ ($\mathbf{J_{q\neq 0}} = 0$)



- Antiferromagnet: $\mathbf{J_q}$ is finite only for $\mathbf{q} = \pm\mathbf{k}$, where $\mathbf{k}$ is the propagation wavevector of the magnetic structure.



In a μSR experiment several millions muons are implanted: they randomly localise in different unit cells of the crystal structure.



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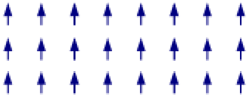

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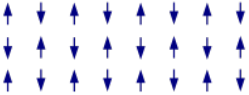

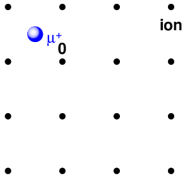

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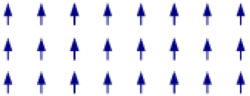

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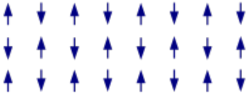

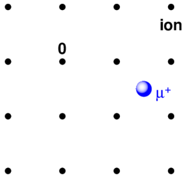

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Zero-field polarisation function in magnets

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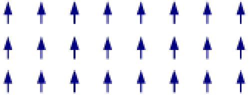

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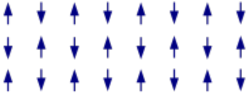

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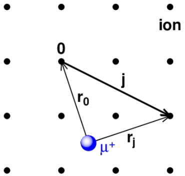

Zero-field polarisation function in magnets

Commensurate magnets

Recall,

$$\mathbf{B}_{\mathrm{loc}} = -\frac{\mu_0}{4\pi}\frac{g\mu_{\mathrm{B}}}{\sqrt{n_c}v_{\mathrm{c}}}\sum_{\substack{\mathbf{q}=0\\ \mathrm{or}\\ \mathbf{q}=\pm\mathbf{k}}}\exp(-i\mathbf{q}\cdot\mathbf{r}_0)\boldsymbol{G}_{\mathbf{q}}\mathbf{J}_{\mathbf{q}}.$$

An antiferromagnetic structure is commensurate if $\mathbf{k} = r\mathbf{Q}$ where $\mathbf{Q}$ is a reciprocal lattice vector and r is a rational number.

$\longrightarrow$ $\exp(-i\mathbf{q}\cdot\mathbf{r}_0)$ takes a finite number of values, so $\mathbf{B}_{\mathrm{loc}}$ does.

Obviously, this is also true for a ferromagnet in which $\mathbf{q} = \mathbf{k} = 0$.

$\longrightarrow$ One (or more) muon spin precession frequency(ies).

μSR cannot directly tell whether a system is a ferro- or an antiferromagnet.

Zero-field polarisation function in magnets

Incommensurate magnets — spin density wave

Recall,

$$\mathbf{B}_{\rm loc} = -\frac{\mu_0}{4\pi}\frac{g\mu_{\rm B}}{\sqrt{n_c}v_{\rm c}}\sum_{\mathbf{q}=\pm\mathbf{k}}\exp(-i\mathbf{q}\cdot\mathbf{r}_0)\boldsymbol{G}_{\mathbf{q}}\mathbf{J}_{\mathbf{q}}.$$

For an incommensurate magnetic structure, $\mathbf{k} = s\mathbf{Q}$ where s is an irrational number.
$\longrightarrow$ $\exp(-i\mathbf{q}\cdot\mathbf{r}_0)$ takes an infinite number of values,
$\longrightarrow$ a continuous distribution of $\mathbf{B}_{\rm loc}$ is expected.

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Zero-field polarisation function in magnets

Spin density wave, simple case (1)

Recall,

$$\mathbf{B}_{\rm loc} \quad = \quad -\frac{\mu_0}{4\pi}\frac{g\mu_{\rm B}}{\sqrt{n_c}v_{\rm c}}\sum_{\mathbf{q}=\pm\mathbf{k}} \exp(-i\mathbf{q}\cdot\mathbf{r}_0)\boldsymbol{G}_{\mathbf{q}}\mathbf{J}_{\mathbf{q}}.$$

Assume that the vectors $\mathbf{B}_{\rm loc}$ remain collinear when $\mathbf{q}\cdot\mathbf{r}_0$ spans the interval $[0,2\pi[$, then

$$\mathbf{B}_{\rm loc} \quad = \quad \cos\alpha\,\mathbf{B}_{\rm max}, \quad \text{with } \alpha\in[0,2\pi[.$$

Zero-field polarisation function in magnets

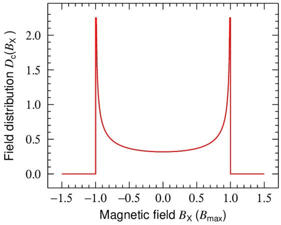
Spin density wave, simple case (2)

Assume for simplicity $\mathbf{B}_{\max} \parallel \mathbf{X}$,

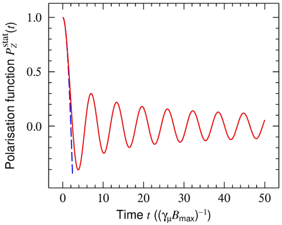
$$D_{\rm c}(B_X) = \int \delta(B_X - B_{{\rm loc},X})\,{\rm d}B_{{\rm loc},X} = \frac{\int_0^{2\pi} \delta(B_X - B_{\max}\cos\alpha)\,{\rm d}\alpha}{\int_0^{2\pi} {\rm d}\alpha} = \frac{1}{\pi}\frac{1}{\sqrt{B_{\max}^2 - B_X^2}},$$

$$P_Z^{\rm stat}(t) = \int_{-B_{\max}}^{B_{\max}} D_{\rm c}(B_X)\cos(\gamma_\mu B_X t)\,{\rm d}B_X = J_0(\gamma_\mu B_{\max} t)$$

$J_0(x)$: zeroth-order Bessel function of the first kind.



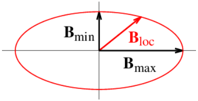
- For $x \ll 1$, $J_0(x) \to 1 - x^2/4$
- For $x \to \infty$, $J_0(x) \to \sqrt{2/\pi x}\cos(x - \pi/4)$: $\pi/4$ dephasing of oscillations



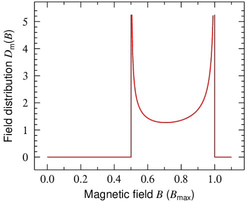
Zero-field polarisation function in magnets

Spin density wave, general case

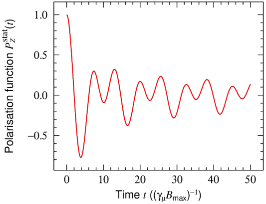
$\mathbf{B}_{\rm loc} = \cos\alpha\,\mathbf{B}_{\rm max} + \sin\alpha\,\mathbf{B}_{\rm min}$, with $\mathbf{B}_{\rm max} \perp \mathbf{B}_{\rm min}$.
The ellipse follows from the anisotropy of the dipolar interaction.

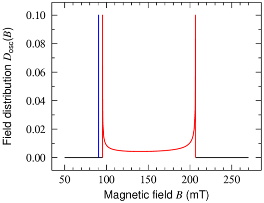


$$D_{\rm m}(B) = \frac{2}{\pi}\frac{B}{\sqrt{B_{\rm max}^2 - B^2}\sqrt{B^2 - B_{\rm min}^2}}.$$



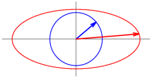
$D_{\rm m}(B)$ and $P_Z(t)$ in the case
$B_{\rm max} = 2B_{\rm min}$
and
$\mathbf{B}_{\rm max}, \mathbf{B}_{\rm min} \perp \mathbf{Z}$

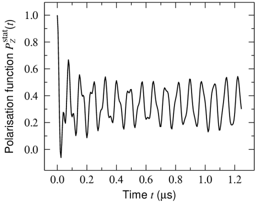




Real life case of MnSi
(helical spin density wave):

- $\mathbf{B}_{\rm max}$ and $\mathbf{B}_{\rm min}$ not $\perp$ $\mathbf{Z}$,
- four magnetic muon sites,
- four magnetic domains.





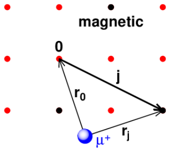
Outline

Computation of field distributions

Uncorrelated moments, high-transverse field case

Consider magnetic impurities randomly distributed in a matrix of non-magnetic sites. With the notations

- j for a site among a total of N,
- $c_{\rm imp}$ for the occupation probability of an impurity (possibly $c_{\rm imp} = 1$),
- $B_{Z,j}$ for the Z component of the field at the muon arising from atom at site j,
- $w_j(B_{Z,j})$ for the distribution of field $B_{Z,j}$ produced at the muon by impurity at site j,



$$D_{\rm c}^{\rm sh}(B_Z) = \int \cdots \int \delta \left(B_Z - \sum_{j=1}^{N} B_{Z,j} \right) \prod_{j=1}^{N} [(1 - c_{\rm imp})\delta(B_{Z,j}) + c_{\rm imp} w_j(B_{Z,j})] \, {\rm d}B_{Z,1} \ldots {\rm d}B_{Z,N}.$$

The distributions due to the impurities are assumed to be independent, hence $\prod_{j=1}^{N}$.

We will take $B_{Z,j} = -\frac{\mu_0}{4\pi} J_{Z,j} \frac{g_i \mu_{\rm B}}{r_j^3} (3\cos^2\theta_j - 1)$, i.e. the impurity dipole field.

Computation of field distributions

Uncorrelated moments, high-transverse field case, extreme dilution limit ($c_{\mathrm{imp}} \ll 1$)

Computation of the characteristic function

$$G_{\mathrm{TF}}(t) = \int \exp(i\gamma_\mu B_Z t) D_{\mathrm{c}}^{\mathrm{sh}}(B_Z)\,\mathrm{d}B_Z,$$

for $c_{\mathrm{imp}} \ll 1$, i.e. the large dilution limit:

$$G_{\mathrm{TF}}(t) = \exp(-\gamma_\mu \Delta_{\mathrm{L}} |t|),$$

with $\Delta_{\mathrm{L}} = K_{\mathrm{L}} \frac{\mu_0}{4\pi} \rho_{\mathrm{vol}} c_{\mathrm{imp}} g \mu_{\mathrm{B}} \langle |m| \rangle$, where ρ_{vol} is number of sites per unit volume, the m's are the eigenvalues of J_Z and $K_{\mathrm{L}} \approx 2.5325$ (case where each impurity has its own quantisation axis).
From an inverse Fourier transform of $G_{\mathrm{TF}}(t)$,

$$D_{\mathrm{c}}^{\mathrm{sh}}(B_Z) = \frac{1}{\pi} \frac{\Delta_{\mathrm{L}}}{\Delta_{\mathrm{L}}^2 + B_Z^2}$$

i.e. a Lorentzian or Cauchy distribution.

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Computation of field distributions

Uncorrelated moments, high-transverse field case, $c_{\mathrm{imp}} = 1$

The characteristic function is

$$G_{\mathrm{TF}}(t) \approx \exp\left(-\frac{\gamma_\mu^2 \Delta_{\mathrm{G}}^2 t^2}{2}\right),$$

in the short-time limit, with $\Delta_{\mathrm{G}}^2 = \frac{1}{3}\left(\frac{\mu_0}{4\pi}\right)^2 \sum_{j=1}^{N} \frac{g^2 \mu_{\mathrm{B}}^2}{r_j^6} \left\langle J_Z^2 \right\rangle (1 - 3\cos^2\theta_j)^2$.

Extremely fast convergence of the sum, due to the r_j^{-6} factor.

Case of nuclear dipoles: the $2J+1$ Zeeman levels of J_Z are equipopulated, hence $\left\langle J_Z^2 \right\rangle = J(J+1)/3$. The initial $1/3$ factor drops when all the nuclei have the same quantisation axis.

From an inverse Fourier transform of $G_{\mathrm{TF}}(t)$,

$$D_{\mathrm{c}}^{\mathrm{sh}}(B_Z) = \frac{1}{\sqrt{2\pi}\Delta_{\mathrm{G}}} \exp\left(-\frac{B_Z^2}{2\Delta_G^2}\right),$$

i.e. a Gaussian distribution.

Computation of field distributions

Uncorrelated moments, zero-field case, $c_{\mathrm{imp}} \ll 1$

Procedure similar to the high transverse field case:

$$D_{\mathrm{v}}(\mathbf{B}) = \int \cdots \int \delta\left(\mathbf{B} - \sum_{i=1}^{N} \mathbf{B}_i\right) \prod_{i=1}^{N} \left[(1 - c_{\mathrm{imp}})\delta(\mathbf{B}_i) + c_{\mathrm{imp}} w_i(\mathbf{B}_i)\right] \mathrm{d}\mathbf{B}_1 \ldots \mathrm{d}\mathbf{B}_N.$$

For $c_{\mathrm{imp}} \ll 1$,

$$G_{\mathrm{ZF}}(\mathbf{t}) = \exp(-\gamma_\mu \Delta_{\mathrm{L}} t),$$

with $\Delta_{\mathrm{L}} = K_{\mathrm{L}} \frac{\mu_0}{4\pi} \rho_{\mathrm{vol}} c_{\mathrm{imp}} g \mu_{\mathrm{B}} \langle |m| \rangle$, where $K_{\mathrm{L}} \approx 4.5406$.
Since $G_{\mathrm{ZF}}(\mathbf{t})$ only depends on t, $D_{\mathrm{v}}(\mathbf{B})$ is isotropic with

$$D_{\mathrm{v}}(\mathbf{B}) = D_{\mathrm{v}}(B) = \frac{1}{\pi^2} \frac{\Delta_{\mathrm{L}}}{\left(\Delta_{\mathrm{L}}^2 + B^2\right)^2}.$$

Recap on the static polarisation functions

- Computation of $P_{X,Z}^{\mathrm{stat}}(t)$ assuming a field distribution
- Nature of field at the muon site (dipole and Fermi contact)
- Derivation of $D_{\mathrm{c}}(B_Z)$ and $D_{\mathrm{v}}(\mathbf{B})$ for usual physical situations

Outline

Outline

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Introduction to the dynamical polarisation functions

The Larmor equation

$$\frac{\mathrm{d}\mathbf{S}_\mu(t)}{\mathrm{d}t} = \gamma_\mu \, \mathbf{S}_\mu(t) \times \mathbf{B}_{\mathrm{loc}}(t),$$

is still valid.

However it is difficult to solve it when $\mathbf{B}_{\mathrm{loc}}(t)$ is a stochastic variable.

Stochastic account of dynamics

We compute $P_\alpha(t)$ for two different models.

Hypothesis for both models:
$\mathbf{B}_{\rm loc}(t)$ follows a stationary Gaussian-Markovian process, i.e.

- independent of origin of time
- $B^\alpha_{\rm loc}(t)$ belongs to a Gaussian distribution
- $\mathbf{B}_{\rm loc}(t)$ evolves in jumps, with a hopping probability which does not depend on the system state before the jump.

Doob's theorem (1942):

$$\langle B^\alpha_{\rm loc}(t_0) B^\alpha_{\rm loc}(t_0 + t)\rangle = \langle (B^\alpha_{\rm loc})^2\rangle \exp\left(-\nu_{\rm c}|t|\right)$$

where $\nu_{\rm c}^{-1} = \tau_{\rm c}$ is the field correlation time.

Outline

The weak collision model (1)

Computation of $P_X(t)$

Recall, for a single static field $B^Z_{\rm loc} = B_0$,

$$P_X^{\rm stat}(t) = \cos(\omega_0 t)$$

with $\omega_0 = \gamma_\mu B_0$.

For $B^Z_{\rm loc}(t)$, the phase at time t is

$$\gamma_\mu B^Z_{\rm loc}(t_0)(t_1 - t_0) + ... + \gamma_\mu B^Z_{\rm loc}(t_{n-1})(t_n - t_{n-1}) = \int_0^t \gamma_\mu B^Z_{\rm loc}(t')\mathrm{d}t'.$$

After averaging over the muon ensemble

$$P_X(t) = \mathcal{R}e\left\{\left\langle \exp\left[i\int_0^t \gamma_\mu B^Z_{\rm loc}(t')\mathrm{d}t'\right]\right\rangle\right\}.$$

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The weak collision model (2)

Computation of $P_X(t)$

Now, for a stationary Gaussian process,

$$\left\langle \exp\left[i\int_0^t \gamma_\mu \delta B^Z_{\rm loc}(t')\mathrm{d}t'\right]\right\rangle = \exp\left[-\int_0^t \mathrm{d}t' \int_0^t \gamma_\mu^2 \left\langle \delta B^Z_{\rm loc}\delta B^Z_{\rm loc}\left(t'-t''\right)\right\rangle \mathrm{d}t''\right],$$

where $\delta B^Z_{\rm loc}(t') = B^Z_{\rm loc}(t') - \langle B^Z_{\rm loc}\rangle$. Using Doob's theorem and the relation

$$\int_0^t \mathrm{d}t' \int_0^t f(t'-t'')\mathrm{d}t'' = 2\int_0^t (t-\tau)f(\tau)\mathrm{d}\tau$$

where $f(t)$ is an even function, we get

$$P_X(t) = \exp\left\{-\frac{\gamma_\mu^2\Delta_{\rm G}^2}{\nu_{\rm c}^2}\left[\exp(-\nu_{\rm c}t) - 1 + \nu_{\rm c}t\right]\right\}\cos\left(\gamma_\mu\langle B^Z_{\rm loc}\rangle t\right),$$

with $\Delta_{\rm G}^2 = \langle\left(\delta B^Z_{\rm loc}\right)^2\rangle$.

This is the so-called Abragam formula (Anderson, 1954).

The weak collision model (2)

Computation of $P_X(t)$

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with $\Delta_{\rm G}^2 = \langle \left(\delta B_{\rm loc}^Z\right)^2 \rangle$.

This is the so-called Abragam formula (Anderson, 1954).

The Abragam function

$$P_X(t) = \exp\left\{-\frac{\gamma_\mu^2 \Delta_\mathrm{G}^2}{\nu_\mathrm{c}^2}\left[\exp(-\nu_\mathrm{c} t) - 1 + \nu_\mathrm{c} t\right]\right\} \cos\left(\gamma_\mu \langle B_\mathrm{loc}^Z \rangle t\right)$$

- For $\nu_\mathrm{c} \ll \gamma_\mu \Delta_\mathrm{G}$,

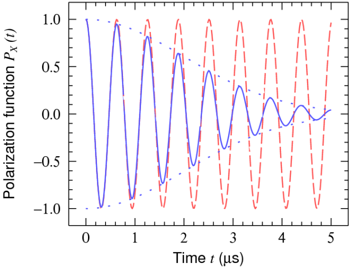
  $$P_X(t) = \exp\left(-\gamma_\mu^2 \Delta_\mathrm{G}^2 t^2/2\right) \cos\left(\gamma_\mu \langle B_\mathrm{loc}^Z \rangle t\right) .$$

- For $\nu_\mathrm{c} \gg \gamma_\mu \Delta_\mathrm{G}$,

  $$P_X(t) = \exp\left(-\lambda_X t\right) \cos\left(\gamma_\mu \langle B_\mathrm{loc}^Z \rangle t\right) ,$$

  with $\lambda_X = \gamma_\mu^2 \Delta_\mathrm{G}^2/\nu_\mathrm{c} = \gamma_\mu^2 \Delta_\mathrm{G}^2 \tau_\mathrm{c}$.

  This is the so-called extreme motional narrowing limit (NMR language).



Examples of Abragam function

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The strong collision model (1)

Computation of $P_Z(t)$

- Let ℓ be the number of changes for $\mathbf{B}_{\rm loc}(t)$ during the muon life time,

$$P_Z(t) = \sum_{\ell=0}^{+\infty} R_\ell(t),$$

where $R_\ell(t)$ is the contribution to $P_Z(t)$ of muons which have experienced ℓ field changes between 0 and t.

- Now,

$$R_0(t) = P_Z^{\rm stat}(t)\exp(-\nu_{\rm c} t),$$

since the probability for $\mathbf{B}_{\rm loc}(t)$ to be unchanged between 0 and t is $\exp(-\nu_{\rm c} t)$.

The strong collision model (2)

Computation of $P_Z(t)$

- For $\ell = 1$ field change and since the process is Gaussian-Markovian,

$$R_1(t) = \left\langle \int_0^t \frac{S^Z_{\mu,j}(t-t')}{S_\mu} \exp[-\nu_\mathrm{c}(t-t')]\nu_\mathrm{c} \frac{S^Z_{\mu,i}(t')}{S_\mu} \exp(-\nu_\mathrm{c} t')\mathrm{d}t' \right\rangle_{ij}$$

$$= \nu_\mathrm{c} \int_0^t R_0(t-t')R_0(t')\mathrm{d}t'.$$

- Recursion relation:

$$R_{\ell+1}(t) = \nu_\mathrm{c} \int_0^t R_\ell(t-t')R_0(t')\mathrm{d}t'.$$

- From the previous relation and the definition $P_Z(t) = \sum_{\ell=0}^{+\infty} R_\ell(t)$,

$$\sum_{\ell=0}^{+\infty} R_{\ell+1}(t) = \nu_\mathrm{c} \int_0^t P_Z(t-t')R_0(t')\mathrm{d}t' = P_Z(t) - R_0(t),$$

...

The strong collision model (3)

Computation of $P_Z(t)$

which can be rewritten as the integral equation

$$P_Z(t) = P_Z^{\mathrm{stat}}(t)\exp(-\nu_{\mathrm{c}} t) + \nu_{\mathrm{c}} \int_0^t P_Z(t - t')P_Z^{\mathrm{stat}}(t')\exp(-\nu_{\mathrm{c}} t')\mathrm{d}t',$$

or in terms of Laplace transforms ($f(s) = \int_0^{+\infty} f(t)\exp(-st)\mathrm{d}t$),

$$P_Z(s) = \frac{P_Z^{\mathrm{stat}}(s + \nu_{\mathrm{c}})}{1 - \nu_{\mathrm{c}} P_Z^{\mathrm{stat}}(s + \nu_{\mathrm{c}})}.$$

- Laplace transforms useful for studying analytical behaviour of $P_Z(t)$
- For numerical purposes, solving numerically the integral equation is efficient

Dynamical polarisation functions

$P_Z(t)$ in zero external field for an isotropic Gaussian distribution

Recall

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

- For $\nu_{\mathrm{c}} \ll \gamma_\mu \Delta_{\mathrm{G}}$,

$$P_Z(t) \simeq \frac{1}{3}\exp\left(-\frac{2}{3}\nu_{\mathrm{c}} t\right) + \frac{2}{3}(1 - \gamma_\mu^2 \Delta_{\mathrm{G}}^2 t^2)\exp\left(-\frac{\gamma_\mu^2 \Delta_{\mathrm{G}}^2 t^2}{2}\right).$$

  High sensitivity to slow dynamics.

- For $\nu_{\mathrm{c}} \gtrsim \gamma_\mu \Delta_{\mathrm{G}}$,

$$P_Z(t) = \exp\left\{-2\frac{\gamma_\mu^2 \Delta_{\mathrm{G}}^2}{\nu_{\mathrm{c}}^2}\left[\exp(-\nu_{\mathrm{c}} t) - 1 + \nu_{\mathrm{c}} t\right]\right\}.$$

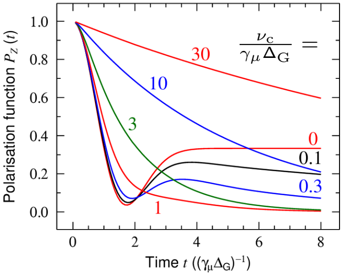
- For $\nu_{\mathrm{c}} \gg \gamma_\mu \Delta_{\mathrm{G}}$,

$$P_Z(t) = \exp(-\lambda_Z t),$$

  with

$$\lambda_Z = 2\gamma_\mu^2 \Delta_{\mathrm{G}}^2 / \nu_{\mathrm{c}}.$$

  (extreme motional narrowing limit).



Dynamical polarisation functions

$P_Z(t)$ in a longitudinal field for an isotropic Gaussian distribution

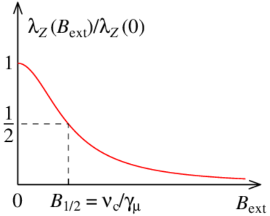
- For $\nu_c \gg \gamma_\mu \Delta_G$,

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

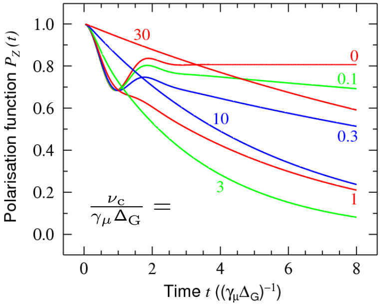
with

$$\lambda_Z = \frac{2\gamma_\mu^2 \Delta_G^2 \nu_c}{\nu_c^2 + \omega_\mu^2}$$

(Redfield formula) and $\omega_\mu = \gamma_\mu B_{ext}$.



Determination of ν_c from $\lambda_Z(B_{ext})$



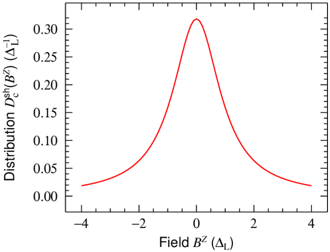
$P_Z(t)$ for $B_{ext} = 3\Delta_G$.

Dynamical polarisation functions

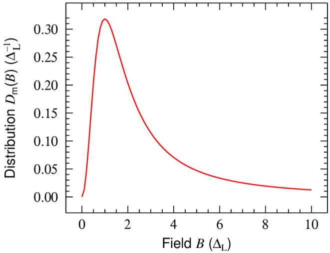
The case of dilute spin glasses (1)

Recall

$$D_{\mathrm{c}}^{\mathrm{sh}}(B_Z) = \frac{1}{\pi}\frac{\Delta_{\mathrm{L}}}{\Delta_{\mathrm{L}}^2 + B_Z^2}, \qquad D_{\mathrm{v}}(\mathbf{B}) = D_{\mathrm{v}}(B) = \frac{1}{\pi^2}\frac{\Delta_{\mathrm{L}}}{(\Delta_{\mathrm{L}}^2 + B^2)^2},$$



Transverse-field



Zero-field

Muons far from any magnetic site have no chance to experience a large field
$\longrightarrow$ Gaussian-Markovian hypothesis breaks.

Dynamical polarisation functions

The case of dilute spin glasses (2)

To cope with the breakdown, we compute the dynamical polarisation function for muons at a given position and perform the spatial average in a second step.
We write

$$P_Z^{\mathrm{stat}}(t) = \int P_{\mathrm{KT}}(t)\rho_{\Delta_{\mathrm{L}}}(\Delta_{\mathrm{G}})\mathrm{d}\Delta_{\mathrm{G}},$$

such that

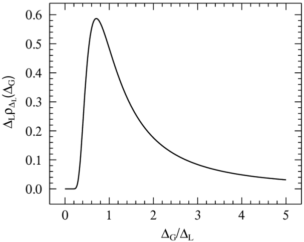
$$P_Z^{\mathrm{stat}}(t) = \frac{1}{3} + \frac{2}{3}(1 - \gamma_\mu \Delta_{\mathrm{L}} t) \exp\left(-\gamma_\mu \Delta_{\mathrm{L}} t\right),$$

is the static function for muons in a dilute magnetic system.
The function

$$\rho_{\Delta_{\mathrm{L}}}(\Delta_{\mathrm{G}}) = \sqrt{\frac{2}{\pi}} \frac{\Delta_{\mathrm{L}}}{\Delta_{\mathrm{G}}^2} \exp\left(-\frac{\Delta_{\mathrm{L}}^2}{2\Delta_{\mathrm{G}}^2}\right),$$

fulfils the requirement. Then

$$P_Z(t) = \int P_{\mathrm{DKT}}(t)\rho_{\Delta_{\mathrm{L}}}(\Delta_{\mathrm{G}})\mathrm{d}\Delta_{\mathrm{G}}.$$



Dynamical polarisation functions

The case of dilute spin glasses (3)

- For $\nu_\mathrm{c} \ll \gamma_\mu \Delta_\mathrm{L}$,

$$P_Z(t) \simeq \frac{1}{3}\exp\left(-\frac{2}{3}\nu_\mathrm{c} t\right) + \frac{2}{3}(1-\gamma_\mu\Delta_\mathrm{L} t)\exp\left(-\gamma_\mu\Delta_\mathrm{L} t\right).$$

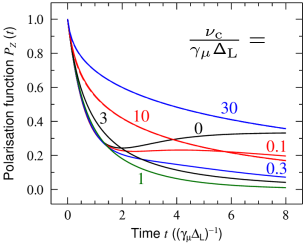
  High sensitivity to slow dynamics.

- For $\nu_\mathrm{c} \gtrsim \gamma_\mu \Delta_\mathrm{L}$,

$$P_Z(t) = \exp\left\{-\sqrt{\frac{4\gamma_\mu^2\Delta_\mathrm{L}^2}{\nu_\mathrm{c}^2}\left[\exp(-\nu_\mathrm{c} t) - 1 + \nu_\mathrm{c} t\right]}\right\}.$$

- For $\nu_\mathrm{c} \gg \gamma_\mu \Delta_\mathrm{L}$,

$$P_Z(t) = \exp\left(-\sqrt{\frac{4\gamma_\mu^2\Delta_\mathrm{L}^2 t}{\nu_\mathrm{c}}}\right).$$

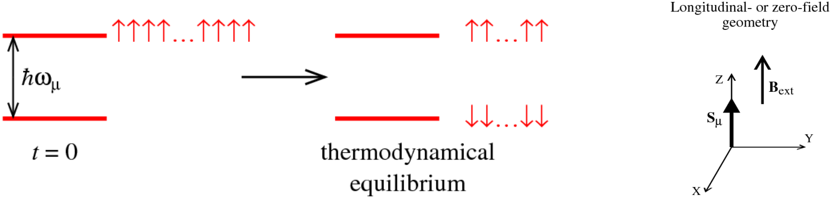


Outline

The polarisation functions from a quantum approach

A flavour for zero- and longitudinal-field experiments

Consider the Zeeman states of the muon spin (spin 1/2),



At thermodynamical equilibrium, the populations of the two states are equal since $\hbar\omega_\mu \ll k_B T$.

Indeed, for $B_{loc} = 1$ T, $\hbar\omega_\mu = 0.56$ μeV ($= k_B T$ for $T = 6.5$ mK).

The polarisation functions from a quantum approach

Derivation of $P_Z(t)$ (1)

Recall Stephen Blundell's lecture,

$$P_Z(t) = 2\,\mathrm{Tr}\left[\rho_{\mathrm{s}} S_\mu^Z S_\mu^Z(t)\right]$$

with

$$S_\mu^Z(t) = \exp\left(i\frac{\mathcal{H}t}{\hbar}\right) S_\mu^Z \exp\left(-i\frac{\mathcal{H}t}{\hbar}\right)$$

where ρ_{s} is the density operator and $\mathcal{H}$ is the Hamiltonian for the muon-system ensemble.

The polarisation functions from a quantum approach

Derivation of $P_Z(t)$ (2)

After some computation,

$$P_Z(t) \simeq \exp[-\psi_Z(t)]$$

with

$$\psi_Z(t) = 2\pi\gamma_\mu^2 \int_0^t (t-\tau)\cos(\omega_\mu \tau)\left[\Phi^{XX}(\tau) + \Phi^{YY}(\tau)\right] \mathrm{d}\tau.$$

where $\Phi^{\alpha\beta}(\tau) = \frac{1}{4\pi}\left[\left\langle \delta B_{\mathrm{loc}}^{\alpha}(\tau)\delta B_{\mathrm{loc}}^{\beta}\right\rangle + \left\langle \delta B_{\mathrm{loc}}^{\beta}\delta B_{\mathrm{loc}}^{\alpha}(\tau)\right\rangle\right]$ is the field correlation function and $\omega_\mu = \gamma_\mu B_{\mathrm{ext}}$.

The polarisation functions from a quantum approach

Derivation of $P_Z(t)$ (3)

Assuming $\Phi^{\alpha\beta}(\tau)$ to decay rapidly on the μSR time t scale, we get $\psi_Z(t) = \lambda_Z t$ with

$$\lambda_Z = \pi\gamma_\mu^2 \left[\Phi^{XX}(\omega_\mu) + \Phi^{YY}(\omega_\mu)\right].$$

$\Phi^{\alpha\beta}(\omega_\mu)$ is the *time* Fourier transform of $\Phi^{\alpha\beta}(\tau)$.

If $\Phi^{\alpha\alpha}(\tau) = \frac{1}{2\pi}\langle(\delta B_{\rm loc}^{\alpha})^2\rangle \exp\left(-\nu_{\rm c}|\tau|\right)$

- $B_{\rm ext} = 0$,

  $$\lambda_Z = \gamma_\mu^2 \left(\langle(\delta B_{\rm loc}^{X})^2\rangle + \langle(\delta B_{\rm loc}^{Y})^2\rangle\right)/\nu_{\rm c},$$

  which can be identified to

  $$\lambda_Z = 2\gamma_\mu^2 \Delta_{\rm G}^2/\nu_{\rm c}.$$

- for any $B_{\rm ext}$, assuming $\Phi^{\alpha\alpha}(\tau)$ independent of $B_{\rm ext}$,

  $$\lambda_Z = \frac{2\gamma_\mu^2\Delta_{\rm G}^2\nu_{\rm c}}{\nu_{\rm c}^2 + \omega_\mu^2}.$$

  This is again Redfield's formula.

Outline

The magnetic field at the muon site

The dipolar field arising from localized spins $\mathbf{J}_j$ with Landé factors g is

$$\mathbf{B}_{\mathrm{dip}} = -\frac{\mu_0}{4\pi} g \mu_{\mathrm{B}} \sum_j \left[-\frac{\mathbf{J}_j}{r_j^3} + 3\frac{(\mathbf{J}_j \cdot \mathbf{r}_j)\mathbf{r}_j}{r_j^5} \right].$$

$\mathbf{r}_j$ is the vector distance from the spin to the muon.

When a polarised electron density is present at the muon, an additional contribution is present, the Fermi contact field:

$$\mathbf{B}_{\mathrm{con}} = -\frac{\mu_0}{4\pi} g \mu_{\mathrm{B}} \sum_{j \in \mathrm{NN}} H_j \mathbf{J}_j.$$

Only the muon nearest neighbors (NN) usually contribute to $\mathbf{B}_{\mathrm{con}}$.
When both $\mathbf{B}_{\mathrm{dip}}$ and $\mathbf{B}_{\mathrm{con}}$ contribute to $\mathbf{B}_{\mathrm{loc}}$ (*i.e.* in metals) they generally have the same order of magnitude.
Altogether

$$\mathbf{B}_{\mathrm{loc}} = \mathbf{B}_{\mathrm{con}} + \mathbf{B}_{\mathrm{con}} = -\frac{\mu_0}{4\pi} \frac{g \mu_{\mathrm{B}}}{v_{\mathrm{c}}} \sum_j \boldsymbol{G}_j \mathbf{J}_j.$$

$\boldsymbol{G}$ is the muon-spin j coupling tensor.

Spin-lattice relaxation rate λ_Z and spin-correlation function

From

$$\lambda_Z = \pi\gamma_\mu^2 \left[\Phi^{XX}(\omega_\mu) + \Phi^{YY}(\omega_\mu)\right],$$

introducing the *space* Fourier transform,

$$\mathbf{J}(\mathbf{q}) = \frac{1}{\sqrt{n_c}} \sum_j \mathbf{J}_j \exp(-i\mathbf{q}\cdot\mathbf{j}),$$

we get

$$\lambda_Z = \frac{\mathcal{D}}{2} \int \sum_{\alpha\beta} \mathcal{A}^{\alpha\beta}(\mathbf{q}) \Lambda^{\alpha\beta}(\mathbf{q},\omega_\mu) \frac{\mathrm{d}^3\mathbf{q}}{(2\pi)^3}.$$

$$\Lambda^{\alpha\beta}(\mathbf{q},\omega) = \frac{1}{2}\left[\left\langle \delta J^\alpha(\mathbf{q},\omega)\delta J^\beta(-\mathbf{q})\right\rangle + \left\langle \delta J^\beta(-\mathbf{q})\delta J^\alpha(\mathbf{q},\omega)\right\rangle\right]$$

is the spin correlation tensor,

$$\mathcal{A}^{\alpha\beta}(\mathbf{q}) = G^{X\alpha}(\mathbf{q})G^{X\beta}(\mathbf{q}) + G^{Y\alpha}(\mathbf{q})G^{Y\beta}(\mathbf{q})$$

is the muon-system coupling factor, and $\mathcal{D} = \left(\frac{\mu_0}{4\pi}\right)^2 \gamma_\mu^2 (g\mu_\mathrm{B})^2/v_\mathrm{c}$.

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Spin-lattice relaxation rate λ_Z and spin-correlation function

Recall

$$\lambda_Z = \frac{\mathcal{D}}{2} \int \sum_{\alpha\beta} \mathcal{A}^{\alpha\beta}(\mathbf{q}) \Lambda^{\alpha\beta}(\mathbf{q}, \omega_\mu) \frac{\mathrm{d}^3\mathbf{q}}{(2\pi)^3}. \tag{1}$$

λ_Z is an integral of the spin-correlation function taken near 0 energy (neV to μeV range) over the Brillouin zone with a weighting factor depending on the muon site.

Outline

Superposition of uncorrelated field distributions

The distribution resulting from independent distributions is the convolution product of each of the distributions.

- High transverse field case
  - The evaluation of $P_X^{\mathrm{stat}}(t)$ is trivial since its envelope is the inverse Fourier transform of $D_{\mathrm{c}}^{\mathrm{sh}}(B_Z)$
  - Example: a dilute spin glass in a matrix of atoms with nuclear moments

$$P_X^{\mathrm{stat}}(t) = \exp\left(\frac{-\gamma_\mu^2 \Delta_{\mathrm{G}}^2 t^2}{2}\right) \exp\left(-\gamma_\mu \Delta_{\mathrm{L}} t\right) \cos(\gamma_\mu B_{\mathrm{ext}} t)$$

- Zero-field case
  - Trivial case of Gaussian distributions, since the convolution of Gaussians is a Gaussian
  - Much trickier situation in the other cases, since $P_Z^{\mathrm{stat}}(t)$ is not expressed as an inverse Fourier transform
  - Beware that the so-called Kubo golden formula is not of general validity

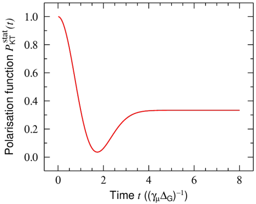
Presence of short-range correlations in the field distribution

Zero-field case (1)

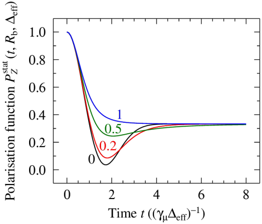
- Occasionally, ZF spectra in quasi-static magnetic systems are found similar to the Kubo-Toyabe function but with a minimum less pronounced than predicted.
- Taking the average of Kubo-Toyabe polarisation functions with Gaussian-distributed field widths,

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

provides the required spectral shape. This is the so-called Gaussian-broadened-Gaussian function (Noakes and Kalvius, 1997).



$P_{\mathrm{KT}}(t)$



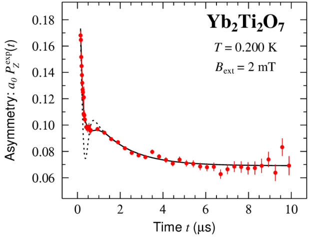
$P_Z^{\mathrm{GbG}}(t)$ as a function of $R \equiv \Delta_{\mathrm{GbG}}/\Delta_0$, with $\Delta_{\mathrm{eff}}^2 \equiv \Delta_0^2 + \Delta_{\mathrm{GbG}}^2$.

Presence of short-range correlations in the field distribution

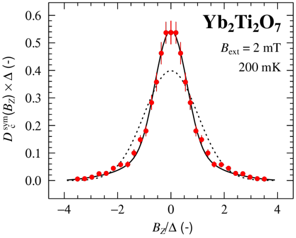
Zero-field case (2)

- Monte Carlo simulations suggest the presence of short-range correlations to be responsible for the weak dip (Noakes, 1999)
- The spectral shape close to the Kubo-Toyabe lineshape suggests the field distribution to be close to a Gaussian
- Therefore, $D_c(B_Z) \propto \exp\left(\frac{-B_Z^2}{2\Delta^2}\right) \longrightarrow D_c(B_Z) \propto \exp\left[-g\left(\frac{B_Z}{\delta}\right)\right]$ with $g(x) = \frac{1}{2}x^2 + \frac{1}{3}(\eta_3 x)^3 + \frac{1}{4}(\eta_4 x)^4$.

Example of $Yb_2Ti_2O_7$, a geometrically frustrated magnet with $T_c \approx 0.25$ K.



Fits with the new distribution (full line) and the Kubo-Toyabe function (dotted line)



New distribution compared to Gaussian distribution

$\longrightarrow$ Presence of short-range correlations in the magnetically ordered state (Yaouanc *et al*, 2013)

Outline

The stretched exponential function

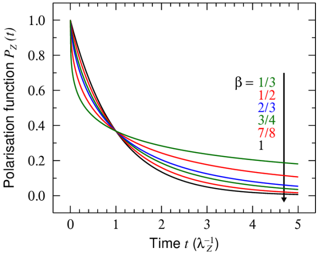
The function

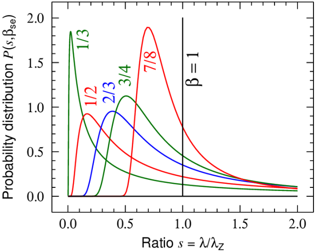
$$P_Z(t) = \exp\left[-(\lambda_Z t)^\beta\right],$$

with $0 < \beta \leq 1$ is often used for the interpretation of μSR data. Sometimes, $\beta > 1$ is even allowed (compressed exponential function).
It was introduced by Kohlrausch (1854), and can be understood as resulting from a collection of exponential functions $\exp(-\lambda t)$ with a distribution $P(s, \beta)$ of relaxation rates,

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

where $s \equiv \lambda/\lambda_Z$ is a dimensionless relaxation rate.





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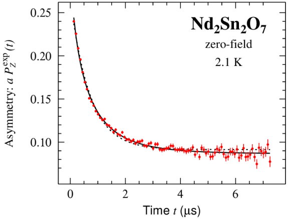
The stretched exponential function

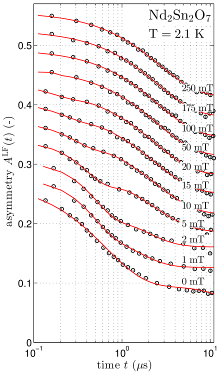
- It is rarely physically justified except in the case of dilute spin glasses, where $\beta = 1/2$ in the extreme motional narrowing limit. Recall

$$P_Z(t) = \exp\left(-\sqrt{\frac{4\gamma_\mu^2 \Delta_{\rm L}^2 t}{\nu_{\rm c}}}\right).$$

- Sometimes a physically sound model approaches very well the stretched exponential function. Example of $Nd_2Sn_2O_7$, a geometrically frustrated magnet with $T_{\rm N} = 0.91$ K.



Full line: stretched exponential with $\beta = 0.70\,(3)$. *Dotted line: exponential.*



A set of LF spectra fitted to the dynamical Kubo-Toyabe model.

⟶ Presence of quasi-static correlations in the paramagnetic phase (Dalmas de Réotier *et al*, 2017)

Outline

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Summary

- Computation of $P_{X,Z}(t)$ in a static $\mathbf{B}_{\rm loc}$, for different field distributions
- Origin and nature of the field at the muon site
- Derivation of the form of the field distribution in selected cases
- Computation of $P_{X,Z}(t)$ when $\mathbf{B}_{\rm loc}$ is dynamical
- Effect of spatial correlations

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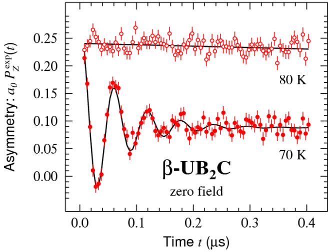
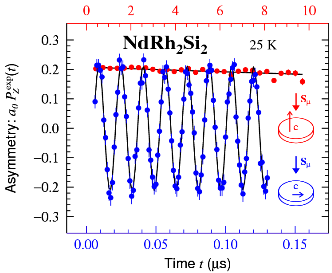
Zero-field polarisation function in magnets

Commensurate magnets: examples



Ferromagnetic transition at T_C = 74.5 K. Powder sample.



Antiferromagnetic transition at T_N = 57 K. Axial magnet, single crystal

μSR cannot directly tell whether a system is a ferro- or an antiferromagnet.

Computation of the field distribution width

Alternative approach, case of nuclear moments (1)

Start from

$$P_X(t) = \frac{1}{2}\mathrm{Tr}\{\rho_{\mathrm{sys}}\sigma^X\sigma^X(t)\}$$

with

$$\sigma^X(t) = \exp\left(i\frac{\mathcal{H}_{\mathrm{tot}}}{\hbar}t\right)\sigma^X\exp\left(-i\frac{\mathcal{H}_{\mathrm{tot}}}{\hbar}t\right),$$

and $\mathcal{H}_{\mathrm{tot}} = \mathcal{H}_{\mathrm{Z},\mu} + \mathcal{H}_{\mathrm{Z,sys}} + \mathcal{H}_{\mathrm{dip}}$.
The field distribution arises from $\mathcal{H}_{\mathrm{dip}}$, truncated to (high field and secular approximations)

$$\tilde{\mathcal{H}}_{\mathrm{dip},\parallel} = \sum_j \frac{\mu_0}{4\pi}\frac{\gamma_\mu\gamma_j\hbar^2}{2r_j^3}(1-3\cos^2\theta_j)\sigma^Z I_j^Z.$$

$\mathbf{I}_j$: nuclear spin at site j (distance r_j and polar angle θ_j to the muon).

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Computation of the field distribution width

Alternative approach, case of nuclear moments (2)

Expanding $P_X(t)$ up to second order in t, we recover the formula

$$\Delta_{\rm G}^2 = \left(\frac{\mu_0}{4\pi}\right)^2 \sum_j \frac{\gamma_j^2 \hbar^2}{r_j^6} \frac{J_j(J_j+1)}{3} (1 - 3\cos^2\theta_j)^2,$$

already given.

Outlook:

- The method allows the electric field gradient acting on the nuclei to be accounted for in the computation of $\Delta_{\rm G}^2$.
- The above method is equivalent to the Van Vleck formula (1948) $\Delta_{\rm G}^2 \propto -\frac{1}{2\gamma_\mu^2\hbar^2} {\rm Tr}\{[\tilde{\mathcal{H}}_{{\rm dip},\parallel}, \sigma^X]^2\}$,
- Similar method for computation of the ZF field width $\Delta_{\rm G}^2 \propto -\frac{1}{2\gamma_\mu^2\hbar^2} {\rm Tr}\{[\mathcal{H}_{{\rm dip},\perp}, \sigma^Z]^2\}$.