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Voigtian Kubo–Toyabe muon spin relaxation

M R Crook and R Cywinski†
School of Physics and Astronomy, University of St Andrews, St Andrews KY16 9SS, UK

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Abstract. The superposition of independent Gaussian and Lorentzian field distributions at an implanted muon site is shown, using Monte Carlo simulations, to result in a Voigtian generalization of the well known dynamic Kubo–Toyabe muon spin-relaxation function from which information on the internal field distribution within a sample can be recovered. The physical origins of the associated pseudo-Voigtian internal field distribution are discussed in the context of recent experimental zero-field μSR results from spin-fluctuating and heavy-fermion systems.

1. Introduction

The longitudinal muon spin-relaxation (μSR) function appropriate for spin-polarized muons implanted in a sample consisting of an ensemble of randomly oriented static (nuclear or atomic) magnetic dipoles is well known in the extreme limits of both concentrated and dilute dipolar systems. In the concentrated limit each orthogonal component of the magnetic field at the muon site is represented by a Gaussian probability distribution [1] about zero and the muon spin relaxation for a stationary muon is described by the semi-classical Gaussian Kubo–Toyabe function [2, 3]. In the dilute limit each of the orthogonal field components is assumed to be distributed about zero with a Lorentzian probability [4] and in this case the resulting muon spin relaxation is given by the static Lorentzian Kubo–Toyabe function [5]. However, there are numerous reports [6–9] in the literature of the observation of muon spin-relaxation (μSR) spectra with lineshapes which are neither purely Gaussian nor purely Lorentzian, but which have a character lying somewhere between the two limits. In such circumstances a 'power Kubo–Toyabe' function, namely

$$g_z^V(t) = a_0\left[\frac{1}{3} + \frac{2}{3}(1-(\lambda t)^\beta)\exp\left(-\frac{(\lambda t)^\beta}{\beta}\right)\right] \tag{1}$$

is frequently invoked as a convenient analytical form through which the μSR spectra can be parametrized. In equation (1) a_0 is the initial asymmetry (ideally $a_0 = 0.33$) and λ is the muon spin-relaxation rate. Although the parameter β provides a continuous interpolation between purely Gaussian ($\beta = 2$) and purely Lorentzian ($\beta = 1$) static Kubo–Toyabe lineshapes, there has, as yet, been no interpretation of the physical significance of β itself. However, it is clear that β must reflect, to some extent, the nature of the internal field distribution at the muon site. It is therefore worthwhile to consider the origins and implications of the power Kubo–Toyabe function in more detail.

In addressing this problem we have employed conventional Monte Carlo techniques to simulate muon spin-relaxation spectra for well defined internal field distributions for both

† Author to whom any correspondence should be addressed.

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stationary and mobile muons. We show that the relaxation function of equation (1) is simply a static Voigtian form of the generalized dynamic Kubo–Toyabe function, representing muon spin relaxation in the presence of a pseudo-Voigtian field distribution resulting from a superposition of independent Gaussian and Lorentzian components. Moreover, we show that analysis of the resulting μSR spectra can provide the relative Gaussian and Lorentzian contributions to the field distribution.

2. The simulation procedure

We have approached the simulations from the perspective of a model dipolar spin system for which the magnetic field probability distribution is specified. In this model the fate of each muon is determined by a Monte Carlo process, intended to closely represent a realistic longitudinal geometry μSR experiment, until a suitable number of events have been recorded. The experimental geometry that we have employed in the simulation corresponds to that of the MuSR spectrometer at the ISIS pulsed muon source at the Rutherford Appleton Laboratory [10]: spin-polarized muons are implanted within a sample at time $t = 0$, and the decay positrons, emitted preferentially along the final muon spin direction with an asymmetry of approximately 0.33, are collected and time stamped in detectors in the forward ($+z$-) and backward ($-z$-) directions. Each implanted muon is randomly assigned a lifetime t_l defined by the exponential decay probability distribution, $\exp(-t/\tau_\mu)$, where τ_μ (=2.19 μs) is the muon lifetime. During the lifetime of the muon its spin, initially polarized along the $+z$-direction, undergoes Larmor precession about its local magnetic field. If the muon is stationary, then the muon experiences a unique field for its entire lifetime and it is therefore only necessary to monitor the z-component of the muon spin to enable the time evolution of the asymmetry to be calculated. If the muon is diffusing, however, in accordance with the strong-collision model, the muon's residence time, τ_r, at a particular site is randomly selected with the exponential probability , $e^{-\nu t}$, where ν is the muon diffusion rate. After allowing the muon spin components to evolve for the time τ_r in this field, a further residence time and magnetic field, representative of a new site, are randomly generated. This process is repeated until the muon decays.

At the instant of decay the probability of detecting the emerging positron in both the forward (F) and the backward (B) detectors is calculated according to the muon decay anisotropy function, incorporating the solid angle subtended by the detectors at the sample position. Each muon decay thus contributes to both the forward and backward detectors as a weighted event and in this manner the numbers of events occurring at time t in each detector are recorded. The muon spin-asymmetry plot may then be constructed in the standard way by taking the ratio

$$G_z(t) = \frac{F(t) - B(t)}{F(t) + B(t)} \tag{2}$$

where $F(t)$ and $B(t)$ are the positron count rates in the forward and backward detectors respectively, and $G_z(t)$ is the longitudinal muon spin-relaxation function, which, at $t = 0$, extrapolates to $a_0 = 0.33$.

3. Stationary muons

Muon spin-relaxation spectra have been simulated for a superposition of Gaussian and Lorentzian field distributions at the muon site. Thus, each orthogonal component, H_i, where $i = x, y, z$, of the magnetic field at the muon site is obtained by a simple addition of

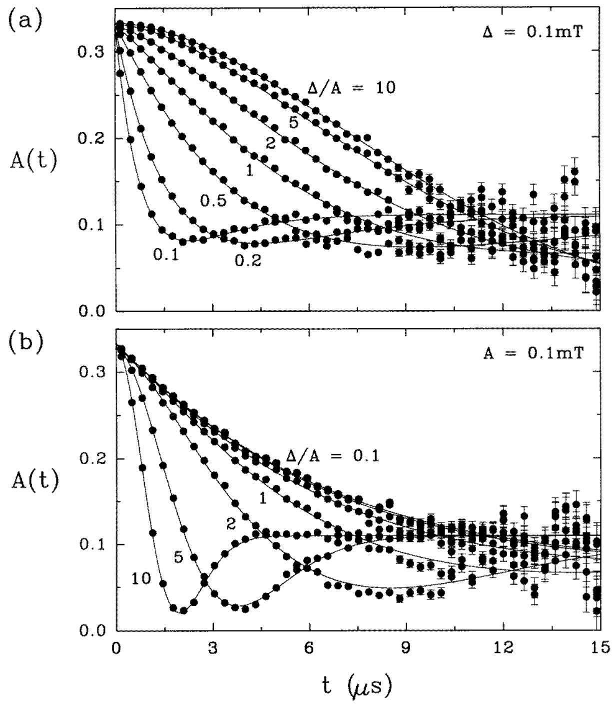

Figure 1. Muon spin-relaxation spectra resulting from a superposition of Gaussian and Lorentzian static field distributions. Δ is the width of the Gaussian component and A is the width of the Lorentzian component.

two independent contributions, one randomly selected according to a Gaussian probability distribution, $P^G(H_i)$,

$$P^G(H_i) = \frac{1}{\sqrt{2\pi}\,\Delta} \exp\left(-\frac{H_i^2}{2\Delta^2}\right) \tag{3}$$

and the other randomly selected according to a Lorentzian probability distribution, $P^L(H_i)$,

$$P^L(H_i) = \frac{\gamma_\mu}{\pi} \frac{\alpha}{(\alpha^2 + \gamma_\mu^2 H_i^2)}. \tag{4}$$

The vector sum of these components then determines the magnitude and direction of the resultant magnetic field at that particular muon site. By adjusting the ratio of the Gaussian distribution width, Δ, to the Lorentzian distribution width, α, the resultant distribution varies between the Gaussian and Lorentzian limits.

When each orthogonal field component is distributed about zero as a Gaussian of width Δ, the magnitude of the resultant field is simply a Maxwellian distribution of width Δ and it is this width which appears in the Gaussian Kubo–Toyabe function in the standard form as $\sigma = \gamma_\mu \Delta$, where σ is the relaxation rate and γ_μ is the gyromagnetic ratio of the muon. However, when each orthogonal field component is distributed about zero as a Lorentzian of width α, the magnitude of the resultant field is given by the Lorentzian equivalent of the Maxwellian distribution (see equation (4)) but in this case with a width $A = \alpha/\langle K\rangle$. For a system of dilute dipoles the factor $\langle K\rangle$ results from the angular average over the anisotropic factor in the dipolar field expression. We have determined numerically, from the simulations, that $\langle K \rangle$ takes the value 0.6919(11). This value can be compared with a value of 0.6909 obtained from a previous numerical treatment [11]. We emphasize here that it is the width A, not as commonly believed α, which appears in the Lorentzian Kubo–Toyabe function

$$g_z^L(t) = a_0 \left[\frac{1}{3} + \frac{2}{3}(1 - at)\exp(-at) \right] \tag{5}$$

in the standard form $a = \gamma_\mu A$.

Table 1. A summary of the least-squares fits of $g_z^V(t)$ (equation (1)) to the simulated relaxation spectra shown in figure 1.

Δ (mT)	A (mT)	$a_{0\,fit}$	λ_{fit} (MHz)	β_{fit}
0.1	0.01	0.3331(3)	0.0881(3)	1.780(9)
0.1	0.02	0.3317(4)	0.0917(3)	1.647(8)
0.1	0.05	0.3294(4)	0.1063(3)	1.411(7)
0.1	0.1	0.3292(6)	0.1354(4)	1.219(6)
0.1	0.2	0.3290(6)	0.2080(6)	1.103(5)
0.1	0.5	0.3292(6)	0.451(1)	1.028(4)
0.1	1.0	0.3327(6)	0.886(3)	0.992(4)
0.01	0.1	0.3338(6)	0.0878(3)	0.996(5)
0.02	0.1	0.3335(6)	0.0903(3)	1.009(5)
0.05	0.1	0.3322(6)	0.1013(3)	1.063(5)
0.1	0.1	0.3293(5)	0.1354(4)	1.217(6)
0.2	0.1	0.3264(5)	0.2163(5)	1.477(6)
0.5	0.1	0.3274(5)	0.4700(8)	1.769(6)
1.0	0.1	0.3292(5)	0.897(2)	1.881(6)

Figure 1(a) shows a selection of simulations of μSR spectra resulting from a typical Gaussian field distribution width of $\Delta = 0.1$ mT summed with a range of Lorentzian field distribution widths varying from $A = 0.1\Delta$ to $A = 10\Delta$. Simulations for the corresponding range of Gaussian distribution widths summed with a Lorentzian distribution width of $A = 0.1$ mT are shown in figure 1(b). The simulated spectra have been analysed by least-squares fitting the power Kubo–Toyabe function of equation (1). As can be seen, all of the spectra in figures 1(a) and 1(b) are found to be accurately described by $g_z^V(t)$ of equation (1). The parameters obtained from the least-squares fits are summarized in table 1.

We find that the distribution of the magnitude of the magnetic field at the muon site, in all of the above simulations, is accurately described by a three-dimensional equivalent of the pseudo-Voigtian lineshape, $P^V(H)$,

$$P^V(H) = A_0 \left[(1-\eta)\left(\frac{2}{\pi}\right)^{1/2} \Gamma^{3/2} H^2 \exp\left(-\frac{H^2}{2\Gamma^2}\right) + \eta\left(\frac{4}{\pi}\right)\frac{\Gamma H^2}{(\Gamma^2 + H^2)^2} \right]. \tag{6}$$

Figure 2 shows several least-squares fits of $P^V(H)$ of equation (6) to the field distributions obtained from the simulations of figure 1(a). The parameters obtained from these fits, together with those obtained from the field distributions extracted from the simulations shown in figure 1(b), are summarized in table 2.

As each orthogonal magnetic field component is derived from a simple superposition of fields selected from pure Lorentzian and pure Gaussian distribution functions, the general form of the three-dimensional pseudo-Voigtian function given by equation (6) for the magnitude of the field distribution is entirely justified: equation (6) is essentially a

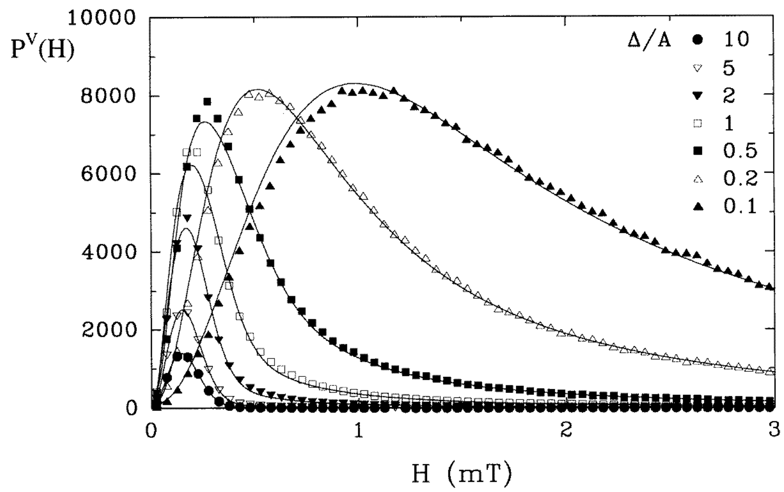

Figure 2. Probability distributions, $P^V(H)$, for the magnitude of the magnetic field at the muon site obtained from the simulations shown in figure 1(a). The data points are obtained from the numerical simulations, while the solid lines represent least-squares fits of equation (6) to the data.

Table 2. The results of a least-squares fit of the pseudo-Voigtian field distribution, $P^V(H)$, to the field profiles at the muon site obtained from the simulations shown in figure 1.

Δ (mT)	A (mT)	Γ (mT)	$\gamma_\mu\Gamma$ (MHz)	λ (MHz)	η	β	$\beta+\eta$
0.1	0.01	0.104 39(8)	0.0889(1)	0.0881(3)	0.115(1)	1.780(9)	1.895(9)
0.1	0.02	0.109 69(8)	0.0934(1)	0.0917(3)	0.217(1)	1.647(8)	1.864(8)
0.1	0.05	0.1274(1)	0.1085(1)	0.1063(3)	0.458(2)	1.411(7)	1.869(7)
0.1	0.1	0.1611(2)	0.1372(2)	0.1354(4)	0.691(2)	1.219(6)	1.910(6)
0.1	0.2	0.2410(4)	0.2052(3)	0.2080(6)	0.885(2)	1.103(5)	1.988(5)
0.1	0.5	0.5153(9)	0.439(1)	0.451(1)	0.993(2)	1.028(4)	2.021(4)
0.1	1.0	1.001(2)	0.852(2)	0.886(3)	1.015(2)	0.992(4)	2.007(4)
0.01	0.1	0.0989(2)	0.0842(2)	0.0878(3)	0.997(2)	0.996(5)	1.993(5)
0.02	0.1	0.1031(2)	0.0878(2)	0.0903(3)	0.994(2)	1.009(5)	2.003(5)
0.05	0.1	0.1202(2)	0.1023(2)	0.1013(3)	0.882(2)	1.063(5)	1.945(5)
0.1	0.1	0.160 47(6)	0.1366(1)	0.1354(4)	0.691(1)	1.217(6)	1.908(6)
0.2	0.1	0.2551(3)	0.2172(3)	0.2163(5)	0.468(2)	1.477(6)	1.945(6)
0.5	0.1	0.547 90(4)	0.466(1)	0.4700(8)	0.233(2)	1.769(6)	2.002(6)
1.0	0.1	1.0455(8)	0.890(1)	0.897(2)	0.083(2)	1.881(6)	1.964(6)

superposition of the Maxwellian and the equivalent Lorentzian field distributions. However, the use of only a single parameter, Γ, to define the overall width of this distribution, rather than using a combination of the independent Gaussian and Lorentzian distribution widths, is perhaps less justified. Nevertheless, with the introduction of a mixing parameter, η, the form of the pseudo-Voigtian field distribution function is clearly consistent with the observed field distributions.

The muon spin-relaxation spectrum is determined by the projection of the muon spin Larmor precessional motion along the longitudinal ($+z$-) direction averaged over the magnetic field distribution. It is the width of this distribution that determines the

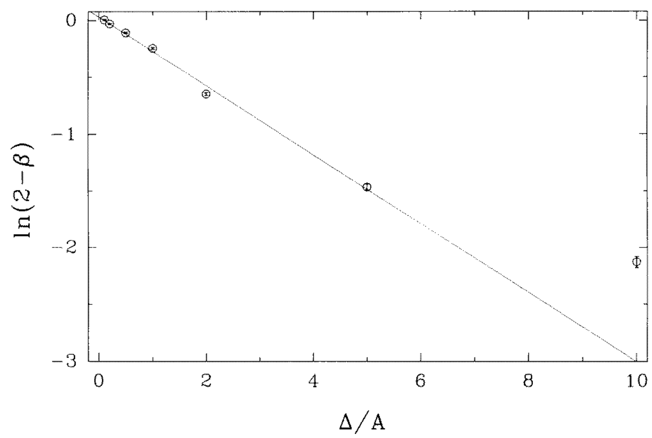

Figure 3. The relationship between β and the ratio of the Gaussian, Δ, to Lorentzian, A, field distribution widths obtained from the numerical simulations shown in figure 1.

relaxation rate. The advantage, therefore, of characterizing the width of the pseudo-Voigtian distribution by a single parameter is that this parameter may then be used to determine the muon spin-relaxation rate in the same way that the parameters which define the Gaussian and Lorentzian field widths determine the relaxation rate in the pure Gaussian and Lorentzian static Kubo–Toyabe functions. That this is the case, for Δ/A spanning two orders of magnitude, is immediately apparent from an inspection of the values of $\gamma_\mu\Gamma$ in table 2. These values are in excellent agreement with the values of λ listed in table 2 which have been extracted through least-squares fits of the power Kubo–Toyabe function (1) to the simulated spectra of figure 1. We also show, in figure 3, that the value of β obtained from these least-squares fits is adequately represented for most of the range of Δ/A by the relationship

$$\beta = 2 - \exp\left(-k\frac{\Delta}{A}\right) \tag{7}$$

where k is found to be 0.3035 and, as shown in table 2, $\eta = 2 - \beta$. Clearly the power Kubo–Toyabe function given in equation (1) can aptly be named a Voigtian Kubo–Toyabe function.

4. Diffusing muons

If the muon is rapidly diffusing, then we expect the motional narrowing process to affect the Gaussian component of the magnetic field but not the Lorentzian component. When the muon diffusion rate, ν, is sufficiently fast, such that $\nu \gg a$, then the relaxation arising from a Lorentzian field distribution becomes independent of the diffusion rate and is given simply by the quasistatic function $e^{-4at/3}$. The modulation of the Gaussian field component may then be considered as a distinct event and the total relaxation is thus derived from two concurrently occurring independent statistical processes. Under these circumstances the polarization at any time, t, is the product of the two individual polarizations at that time.

If the muon is slowly diffusing, however, such that $\nu \approx a$, then the relaxation arising from a Lorentzian field distribution is not independent of the diffusion rate. To obtain an

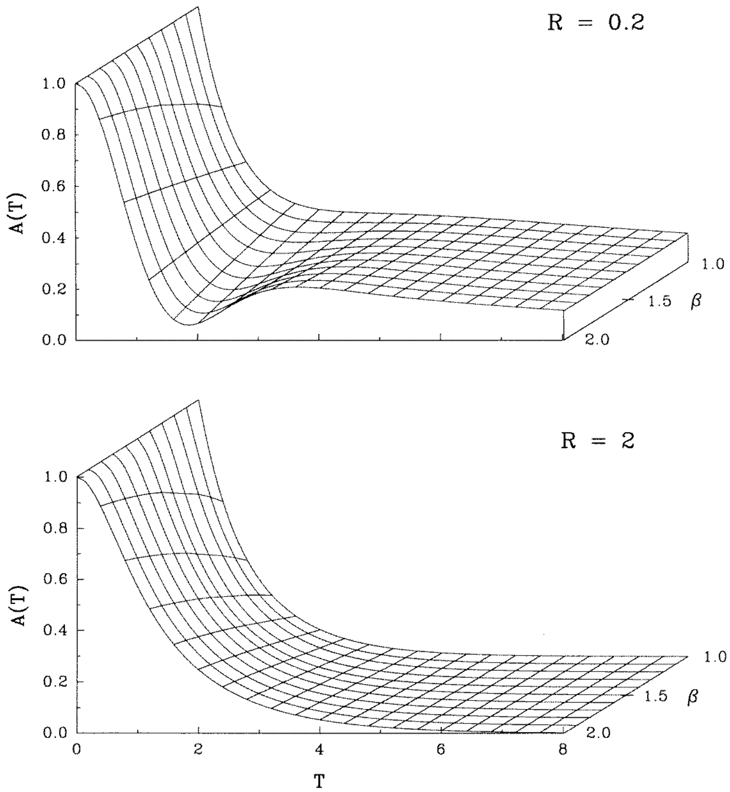

Figure 4. The dynamic Voigtian Kubo–Toyabe muon spin-relaxation function resulting from a superposition of Gaussian and Lorentzian static field distributions modulated according to the strong-collision model (see the text). Top panel: $R = 0.2$ and $1 \leqslant \beta \leqslant 2$; bottom panel: $R = 2$ and $1 \leqslant \beta \leqslant 2$.

accurate description of the muon relaxation function in this situation the strong-collision model should be applied to the Voigtian Kubo–Toyabe function. The depolarization, $G_z^V(t)$, is then given by

$$G_z^V(t) = g_z^V(t) + \nu \int_0^t G_z^V(t - t')g_z^V(t')\ \mathrm{d}'t \tag{8}$$

where $g_z^V(t)$ is the static Voigtian Kubo–Toyabe function of equation (1). $G_z^V(t)$ is shown in figure 4 as a function of the dimensionless parameters R and T, where $R = \nu/\gamma_\mu\Gamma$ and $T = \gamma_\mu\Gamma t$, for $R = 0.2$ and $R = 2$ with $1 \leqslant \beta \leqslant 2$.

Figure 5 shows a set of simulations which cover a range of slow muon diffusion rates for three particular ratios of Δ/A. The first set, (a), has $\Delta = 0.1$ mT and $A = 0.2$ mT and is close to the Lorentzian limit. The second set, (b), has $\Delta = 0.2$ mT and $A = 0.2$ mT and is intermediate in character, while the third set, (c), has $\Delta = 0.5$ mT (5 G) and $A = 0.1$ mT and is close to the Gaussian limit. The solid lines in figure 5 are least-squares fits of the numerically evaluated dynamic Voigtian Kubo–Toyabe function of equation (8) to the simulated data. The parameters obtained from these fits are summarized in table 3, together

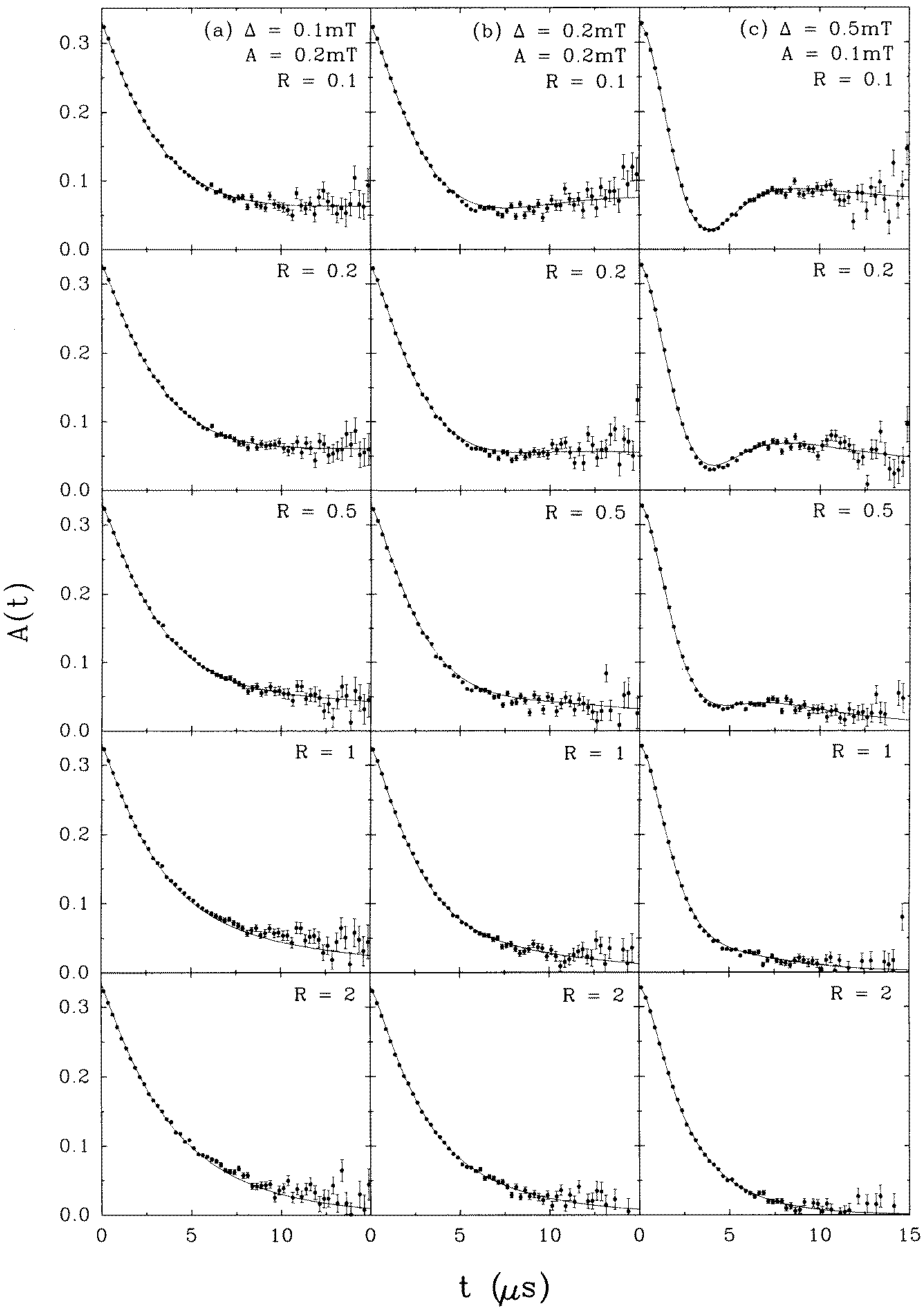

Figure 5. Muon spin-relaxation spectra for a slowly diffusing muon in the presence of both Gaussian, Δ, and Lorentzian, A, static field distributions. The spectra are characterized by $R = \nu/\gamma_\mu \Gamma$ and β, where Γ is the width of the resultant Voigtian field distribution and β defines the extent to which this is either Gaussian or Lorentzian (see the text). The data points are the results of the numerical simulations, while the solid lines are least-squares fits of the dynamic Voigtian Kubo–Toyabe function, given by equation (8).

Table 3. A summary of the least-squares fits of the dynamic Voigtian Kubo–Toyabe function, $G_z^V(t)$, to the simulated spectra shown in figure 5. (a) $\Delta = 0.1$ mT, $A = 0.2$ mT, $\beta_{exp} = 1.10$, $\gamma_\mu\Gamma_{exp} = 0.208$; (b) $\Delta = 0.2$ mT, $A = 0.2$ mT, $\beta_{exp} = 1.22$, $\gamma_\mu\Gamma_{exp} = 0.271$; (c) $\Delta = 0.5$ mT, $A = 0.1$ mT, $\beta_{exp} = 1.77$, $\gamma_\mu\Gamma_{exp} = 0.470$.

	ν (MHz)	R_{exp}	$a_{0\,fit}$	$\gamma_\mu\Gamma_{fit}$ (MHz)	ν_{fit} (MHz)	β
(a)	0.020 80	0.1	0.3300(6)	0.2076(7)	0.0449(2)	1.100(3)
	0.041 60	0.2	0.3295(6)	0.2103(6)	0.0492(2)	1.101(4)
	0.1040	0.5	0.3301(5)	0.2100(7)	0.0998(1)	1.100(3)
	0.2080	1	0.3295(6)	0.2118(6)	0.209(1)	1.100(5)
	0.4160	2	0.3314(6)	0.2114(6)	0.3263(3)	1.099(4)
(b)	0.027 08	0.1	0.3249(6)	0.2730(7)	0.0322(2)	1.285(6)
	0.054 16	0.2	0.3254(6)	0.2750(6)	0.0706(5)	1.300(6)
	0.1354	0.5	0.3279(6)	0.2737(6)	0.1380(1)	1.250(6)
	0.2708	1	0.3273(5)	0.2816(7)	0.310(3)	1.250(7)
	0.5416	2	0.3289(6)	0.2790(7)	0.4255(3)	1.250(6)
(c)	0.0470	0.1	0.3261(6)	0.4698(9)	0.0398(2)	1.807(8)
	0.0940	0.2	0.3291(5)	0.4702(8)	0.0859(3)	1.736(7)
	0.2350	0.5	0.3302(5)	0.4715(8)	0.2259(2)	1.784(9)
	0.4700	1	0.3289(5)	0.4702(8)	0.4871(2)	1.798(8)
	0.9400	2	0.3295(5)	0.4770(8)	1.004(1)	1.704(8)

with the expected values of both β and $\gamma_\mu\Gamma$ obtained from the fits of the corresponding static functions given in table 1. As can be seen from this table, despite the similarities of the spectra, the extracted parameters agree remarkably well with the expected values.

5. Conclusions

With the aid of Monte Carlo simulation techniques we have examined the longitudinal μSR spectra resulting from a pseudo-Voigtian distribution of the magnetic fields at the muon site for both stationary and a slowly diffusing muons. It has been found that, for a stationary muon, the static Voigtian Kubo–Toyabe function $g_z^V(t)$, given by equation (1), accurately describes these spectra for field distributions spanning the entire range between the Gaussian and Lorentzian limits. In addition, we have demonstrated that the relaxation rate of the μSR spectrum is determined solely by the width of the Voigtian field distribution. We have also established a relationship (equation (7)) between the relative widths of the Gaussian and Lorentzian components of the pseudo-Voigtian distribution and the mixing parameter, η, and the variable power, β, appearing in $g_z^V(t)$.

In the case of a diffusing muon the application of the strong-collision model to the Voigtian Kubo–Toyabe model is appropriate. Within the framework of this model we can successfully recover the expected parameters (diffusion rate, relaxation rate etc) from simulations of slow to moderate muon diffusion rates and for a range of field distributions.

It is interesting to note that Voigtian Kubo–Toyabe muon spin relaxation has recently been observed in the Zr-substituted spin-density-wave-like weak itinerant electron magnet $NbFe_2$ [7], and in the Y-substituted heavy-fermion compounds UPd_3 [8] and URu_2Si_2 [9]. A framework for interpreting the zero-field μSR spectra from these systems is provided by the current analysis. For example pure URu_2Si_2 develops antiferromagnetic order of a spin-density-wave character, with a very small ordered moment at $T_N = 17.5$ K. Small additions of Y leave T_N essentially unchanged, but produce locally somewhat better defined moments which can behave as Kondo impurities. As temperature decreases, we therefore expect the muons implanted in $U_{1-x}Y_xRu_2Si_2$ to sense an increasingly pronounced Lorentzian field distribution, related to the condensation of the dilute magnetic impurities, against an otherwise Gaussian field distribution arising from the local nuclear dipole fields. Indeed, on cooling, the μSR spectra obtained from $U_{1-x}Y_xRu_2Si_2$ show the Voigtian Kubo–Toyabe form, with the exponent β decreasing from $\beta = 2$ at high temperatures to $\beta = 1$ at 5 K [9]. The behaviour of $NbFe_2$ and UPd_4 can similarly be discussed in terms of an essentially inhomogeneous moment condensation from a spin-fluctuating matrix.

In general the Voigtian Kubo–Toyabe function might be expected to provide a reasonable description of the muon spin relaxation in any matrix within which dilute atomic or magnetic cluster dipoles are embedded in a concentrated matrix of nuclear dipoles, or perhaps in any situation wherein a pseudo-Voigtian internal field distribution arises naturally (for example a dipolar system which is intermediate between the dilute and concentrated limits).

Acknowledgments

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