Quasicritical behaviour of the nuclear magnetic resonance and muon spin-rotation relaxation in cubic ferromagnets and antiferromagnets

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Abstract
A theoretical model describing quasicritical behaviour of the nuclear magnetic resonance (NMR) relaxation rates $1/T_1$, $1/T_2$ and muon relaxation rate λ for cubic ferromagnets and antiferromagnets with spin fluctuations is presented. The model extends and generalizes the model of Moriya and explains why the observed experimental values of the critical exponents are different from the values predicted by the theory of critical phenomena. The formulae for the relaxation rates obtained from this model are used for fitting to the experimental NMR data for YMn_2D_x, $TbMn_2D_2$ and the muon spin-rotation (μSR) data for YMn_2, YMn_2D_x, $GdMn_2$ and $Y(Co_{1-x}Al_x)_2$ (ferromagnetic). It is shown that the dependence of the exchange coupling constant on the wavevector k is important for understanding the observed quasicritical effects.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The behaviour of magnetic materials above their magnetic transition temperatures has been of great interest both theoretically and experimentally over the past few years. Of most interest is the mechanism of the transition from the thermally activated paramagnetic state to the ordered state of strongly interacting magnetic moments. In the temperature range above but close to the ordering temperature (the Curie temperature T_C or the Néel temperature T_N), the interactions evolve from short-range to long-range interactions, but the processes are not yet clear or satisfactorily described; the related phenomena are referred to as critical behaviour. Many physical quantities show critical behaviour when the temperature T approaches the transition point $T_{C,N}$, i.e. their temperature dependence can be described by the function

$$F(\delta) = \text{constant} \times \delta^{-\eta} \tag{1}$$

where $\delta = \frac{T - T_{C,N}}{T_{C,N}}$ and η is a critical exponent. Theoretically calculated critical exponents are constants independent of the chemical composition of the materials investigated, but their specific values depend crucially on the theoretical model assumed. For example, the critical exponent for magnetic susceptibility given by the Ginzburg–Landau model is 1.0, that given by the three-dimensional Ising model is 1.2378 and that given by the Heisenberg model is 1.388 [1]. The problem of critical behaviour for itinerant electron materials is very difficult, because of the spin fluctuations that have to be considered. To explain the hyperfine broadening of the nuclear magnetic resonance (NMR) linewidth as the transition point is approached from above, Moriya [2] proposed a model in the framework of the theory of spin fluctuations. He obtained asymptotic values of the NMR linewidth near the transition point proportional to $\delta^{-3/2}$ for cubic ferromagnets and to $\delta^{-1/2}$ for cubic antiferromagnets. Lovesey *et al* [3, 4] discussed the critical behaviour of the muon relaxation rate λ near the critical point and obtained values proportional to $\delta^{-3\nu/2}$ (where $\nu = 0.70$) for the Heisenberg ferromagnet and $\delta^{-\nu/2}$ for the Heisenberg antiferromagnet. Strictly speaking, equation (1) is valid only in the limit $T \rightarrow T_{C,N}$ and the behaviour can be quite different when T differs from $T_{C,N}$ by a finite amount.

Our interest was focused on the Laves phases RMn_2 (R = rare earth or Y) and their hydrides, where spin fluctuations must be taken into account. In our approach to explaining the NMR relaxation rates $1/T_1$, $1/T_2$ and the muon relaxation rate λ in YMn_2D_x, we have shown [5] that these quantities change according to equation (1) over quite a wide temperature range, which is rather surprising. Moreover, the values of η obtained by us depend on the deuterium concentration x and differ from the $1/2$ value. Although the behaviour described by the empirical formula (1) looks like a critical one, it cannot be explained within the standard theory of critical phenomena, and thus we call it 'quasicritical' [5]. The YMn_2D_x hydrides are cubic ferrimagnets with a very small ferromagnetic component; hence they were treated by us as antiferromagnets. To explain their behaviour in the paramagnetic region, we proposed a model in the framework of the theory of spin fluctuations [5]. In the present paper we extend this model to both ferromagnetic and antiferromagnetic cubic materials. The proposed model represents a generalized, extended version of Moriya's theory [2]. We present, discuss and apply this model not only to our data for YMn_2D_x [5, 6], but also to our NMR data for $TbMn_2D_2$ [7] and muon spin-rotation (μSR) data for YMn_2 [8], $GdMn_2$ [9] and $Y(Co_{1-x}Al_x)_2$ (ferromagnetic) [10].

2. Theoretical considerations

There are three important types of nuclear spin interaction mechanism in magnetically ordered systems [2]:

 (1) hyperfine interactions;
 (2) indirect nuclear spin interaction via the hyperfine interactions;
 (3) direct nuclear dipole–dipole interaction in which the electron spins do not participate.

In the following calculations we concentrate on the first two mechanisms, because at high temperatures they are determined by the electron spin motion. This motion is generally very rapid at high temperatures; thus the nuclear spin motion can be neglected in this range.

According to Moriya's theory of spin fluctuations [2], the spin–lattice relaxation time T_1 for a cubic structure at the temperature T can be written approximately as follows:

$$\frac{1}{T_1} = \frac{2A^2 k_B T}{\hbar^2 g^2 \mu_B^2} \frac{1}{N} \sum_k \frac{\chi(k)}{\Gamma_k} \tag{2}$$

where A is a hyperfine coupling constant, N is the number of magnetic atoms per unit volume, $\chi(\mathbf{k})$ is a staggered susceptibility, Γ_k determines the decay of the time correlation function:

$$f_k(t) = \exp(-\Gamma_k t) \tag{3}$$

and the other symbols have their usual meanings. The hyperfine coupling is in principle a tensor and depends on the wavevector $\mathbf{k}$ if we work in the $\mathbf{k}$-representation. However, because the hyperfine interaction is of short range and isotropic, we can approximate it by a scalar [11, 12]. If we consider muon spin depolarization the dipolar interaction may become important. This will be discussed later. The calculation of $1/T_1$ could be carried out if the analytical forms of $\chi(\mathbf{k})$ and Γ_k were known. Both quantities depend on the local magnetic symmetry of the compound investigated. In the following we shall consider the ferromagnetic and antiferromagnetic cases separately.

2.1. Ferromagnetic materials

For a cubic ferromagnetic crystal the scalar susceptibility $\chi(\mathbf{k})$ has the form

$$\chi(\mathbf{k}) = \frac{g^2\mu_B^2 S(S+1)/3k_B T_C}{\delta + \{1 - (J(\mathbf{k})/J(0))\}} \tag{4}$$

where S is the spin of the magnetic ion and $J(\mathbf{k})$ is the Fourier transform of the exchange coupling constant. For small k the susceptibility can be approximated by

$$\chi(\mathbf{k}) = \frac{C_1}{\delta + \gamma k^2}. \tag{5}$$

The coefficient Γ_k in the time correlation function for a ferromagnet can be expanded into powers of k as follows [2]:

$$\Gamma_k = \Lambda k^2 + \Theta k^4 + \cdots \tag{6}$$

where Λ is proportional to δ and Θ remains finite at T_C. Moriya introduced the term Θk^4 making an extension of the van Hove phenomenological theory [13] in which Γ_k depends on k^2 only. Thus the nuclear relaxation time T_1 can be rewritten approximately as

$$\frac{1}{T_1} = \frac{2A^2 k_B T}{\hbar^2 g^2 \mu_B^2} \frac{1}{N} \sum_k \frac{C_1}{(\delta + \gamma k^2)(\Lambda k^2 + \Theta k^4)}. \tag{7}$$

To obtain the asymptotic behaviour

$$\frac{1}{T_1} \propto \delta^{-3/2} \tag{8}$$

as $T \to T_C$, Moriya assumed

$$\frac{\Lambda}{\Theta} = \frac{\delta}{\gamma}. \tag{9}$$

We abandon this assumption because it has no justification and we introduce a dimensionless parameter α:

$$\frac{\Lambda}{\Theta} = \alpha\frac{\delta}{\gamma}. \tag{10}$$

Changing summation into integration in a standard way, we obtain

$$\frac{1}{T_1} = \frac{2A^2 k_B T}{\hbar^2 g^2 \mu_B^2} \nu \frac{1}{(2\pi)^3} \int \frac{\gamma C_1 \, \mathrm{d}^3 \mathbf{k}}{\Theta(\delta + \gamma k^2)(\alpha\delta + \gamma k^2)k^2} \tag{11}$$

where $\nu = V/N$ is the atomic volume. Performing the integration over a sphere within the first Brillouin zone for $\alpha \neq 1$, we get

$$\frac{1}{T_1} = \frac{A^2 \nu S(S+1)}{3\pi^2 \hbar^2 \gamma \Theta} \frac{T}{T_C} \left(\frac{\delta}{\gamma}\right)^{-3/2} \left[\frac{1}{1-\alpha} \arctan\left(k_m \left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right.$$
$$\left. + \frac{1}{(1-\alpha)\sqrt{\alpha}} \arctan\left(\frac{k_m}{\sqrt{\alpha}} \left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right] \tag{12}$$

where k_m is a cut-off parameter. It is easy to see that we can recover the asymptotic behaviour given by equation (8) when $T \rightarrow T_C$, but our expression (12) is valid over a much larger temperature range and consequently the related phenomena are referred to as quasicritical behaviour.

2.2. Antiferromagnetic materials

For a cubic antiferromagnetic crystal, the scalar susceptibility has a similar form:

$$\chi(k) = \frac{g^2 \mu_B^2 S(S+1)/3k_B T_N}{\delta + \{1 - (J(k)/J(K_0))\}} \tag{13}$$

where T_N is the Néel temperature, $\delta = \frac{T-T_N}{T_N}$ and K_0 is the vector where $J(k)$ has the maximum. We can now expand the denominator and get

$$\chi(k) = \frac{g^2 \mu_B^2 S(S+1)/3k_B T_N}{\delta + \gamma \kappa^2} \tag{14}$$

where $\kappa = k - K_0$.

The coefficient Γ_k for an antiferromagnet can be approximately represented as

$$\Gamma_{k+K_0} = \Lambda' + \Theta' k^2 + \cdots \tag{15}$$

where $\Lambda' \propto \delta$ and Θ' is finite at T_N as before. Again Moriya introduced the parameter Θ' making an extension of the de Gennes and Villain phenomenological theory for antiferromagnets [14] which—as he stated—was inadequate even qualitatively. The term $\Theta' k^2$ is necessary to take into account the k-dependence of the time correlation function. Moving the centre of the coordinate system, we can write

$$\sum_k \frac{\chi(k)}{\Gamma_k} = \sum_k \frac{\chi(K_0+k)}{\Gamma_{K_0+k}}. \tag{16}$$

Then the relaxation time T_1 takes the form

$$\frac{1}{T_1} = \frac{2A^2 k_B T}{\hbar^2 g^2 \mu_B^2} \frac{1}{N} \sum_k \frac{C_2}{(\delta + \gamma k^2)(\Lambda' + \Theta' k^2)}. \tag{17}$$

For the same reasons as for ferromagnets, we introduce the parameter α:

$$\frac{\Lambda'}{\Theta'} = \alpha \frac{\delta}{\gamma} \tag{18}$$

and we get for $\alpha \neq 1$

$$\frac{1}{T_1} = \frac{A^2 \nu S(S+1)}{3\pi^2 \hbar^2 \gamma \Theta'} \frac{T}{T_N} \left(\frac{\delta}{\gamma}\right)^{-1/2} \left[\frac{1}{1-\alpha} \arctan\left(k_m \left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right.$$
$$\left. + \frac{\sqrt{\alpha}}{(\alpha-1)} \arctan\left(\frac{k_m}{\sqrt{\alpha}} \left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right] \tag{19}$$
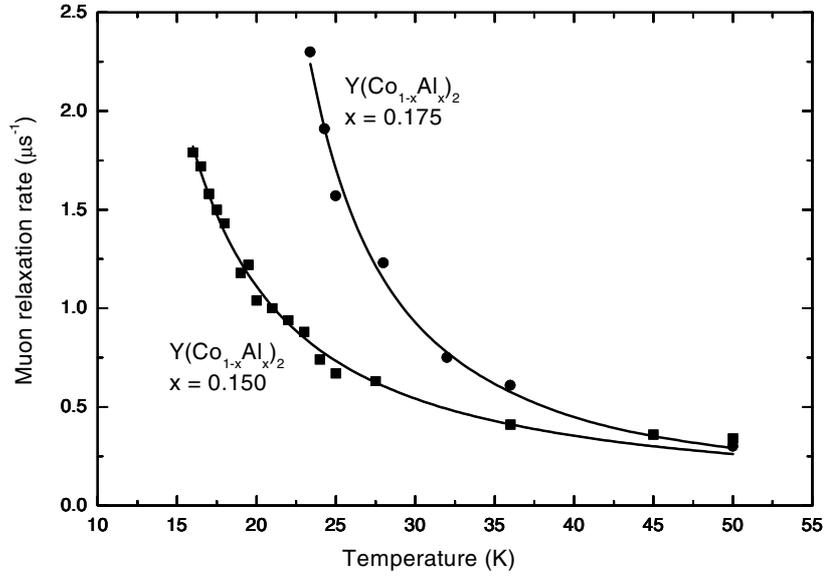
Figure 1. The muon relaxation rate λ as a function of temperature for Y(Co$_{1-x}$Al$_x$)$_2$. The experimental points taken from [10]; the solid curves are the curves fitted with equation (21).

as we have shown in [5]. We can also recover here the asymptotic behaviour

$$\frac{1}{T_1} \propto \delta^{-1/2} \tag{20}$$

as in the original Moriya's work for $T \to T_N$, but the range of applicability of (19) is much wider than (20) and this behaviour is also referred to as quasicritical as in the former case.

3. Numerical results and discussion

3.1. Ferromagnetic materials

In our literature search we had difficulty in finding an appropriate set of T_1-relaxation data; thus to test our formula (12) we analysed the μSR results for the weak ferromagnet Y(Co$_{1-x}$Al$_x$)$_2$ [10]. In this Laves-phase-type material, muons locate in A2B2 interstitial sites, like D atoms in YMn$_2$D$_x$. We compared our formula (12) for T_1^{-1} with experimental data for the muon depolarization rate λ. We found it reasonable, because the behaviour of muons is caused by the same physical interaction as in the case of the NMR T_1-relaxation rate for nuclear magnetic moments of the magnetic or non-magnetic atoms. That is, spin fluctuations cause nuclear spin relaxation as well as muon magnetic moment depolarization in the interstitial positions. According to Yaouanc et al [11], if we identify λ with the inverse muon spin–lattice relaxation time, the μSR expression should be equivalent to the formula given by Moriya for NMR [2]. Some care has to be taken here because the dipolar interaction may in general be important for muons. However, its contribution should be crucial only in strong ferromagnets and for weak ferromagnets it can be neglected. Thus the formula (12), when adapted for λ in the case of weak ferromagnets, should differ only by a numerical prefactor:

$$\lambda = \text{constant} \times \frac{T}{T_C}\left(\frac{\delta}{\gamma}\right)^{-3/2}\left[\frac{1}{1-\alpha}\arctan\left(k_m\left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right.$$
$$\left. + \frac{1}{(1-\alpha)\sqrt{\alpha}}\arctan\left(\frac{k_m}{\sqrt{\alpha}}\left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right]. \tag{21}$$

Table 1. The values describing quasicritical behaviour for the materials analysed. The values of the critical exponent η and the $T_{C,N}$ fitted using equation (1) or taken from literature are given for comparison. In the last two columns we present the values of $T_{C,N}$ and γ obtained from the fits using equations ((12), (19), (21) and (22)).

Material	Data source	η	$T_{C,N}$	$T_{C,N}$ (fit)	γ
$Y(Co_{1-x}Al_x)_2$ $x = 150$	λ [10]	0.68	13	6.8(3)	0.073(4)
$Y(Co_{1-x}Al_x)_2$ $x = 175$	λ [10]	0.64	21.5	14.7(6)	0.067(5)
YMn_2, $T\downarrow$	λ [8]	0.35 [8]	92.5	75(1)	0.288(7)
YMn_2, $T\uparrow$	λ [8]	0.35 [8]	113	87.4(8)	0.295(9)
$GdMn_2$, 0.0 GPa	λ [9]	0.34 [9]	99 [9]	—	0.06(1)
				92.6(3)	0.096(2)
$GdMn_2$, 0.3 GPa	λ [9]	0.43 [9]	81 [9]	—	0.12(1)
				79(1)	0.13(1)
$GdMn_2$, 0.6 GPa	λ [9]	0.43 [9]	71 [9]	—	0.12(1)
				69.0(7)	0.146(9)
$TbMn_2D_2$	T_1 [7]	0.937 [7]	270.5 [7]	—	0.21(1)
				265(1)	0.19(2)
$TbMn_2D_2$	T_2 [7]	0.937 [7]	273.6 [7]	—	0.209(9)
				270.4(4)	0.19(1)

We fitted the function $\lambda(T)$, given by equation (21), to the experimental data for the $Y(Co_{1-x}Al_x)_2$ compound with $x = 0.150$ and 0.175. The results are presented in figure 1. The parameters γ and T_C obtained by the fitting procedure are given in table 1. The parameters α and k_m, which have no physical meaning, are omitted in the table. The agreement between experimental points and the theoretical curve is almost ideal, though the calculated Curie temperature T_C is slightly lower than the one estimated from the experiment. However, we have to keep in mind that the determination of T_C depends somewhat on the experimental set-up and on the definition used as well. The γ-values obtained for $Y(Co_{1-x}Al_x)_2$ are comparable with those for YMn_2D_x [5, 6] for $x > 1$ (see figure 4). The small γ-values can be attributed to the small itinerant Co magnetic moments in this compound and to the weak dependence of the exchange coupling constant J on k.

3.2. Antiferromagnetic materials

For antiferromagnets, above the Néel temperature T_N, the relaxation rates ($1/T_1$, $1/T_2$) should depend on temperature according to equation (19), and the expression for the muon relaxation rate λ should also differ only by a constant numerical prefactor and has the form

$$\lambda = \text{constant} \times \frac{T}{T_N}\left(\frac{\delta}{\gamma}\right)^{-1/2}\left[\frac{1}{1-\alpha}\arctan\left(k_m\left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right.$$
$$\left. + \frac{\sqrt{\alpha}}{(\alpha-1)}\arctan\left(\frac{k_m}{\sqrt{\alpha}}\left(\frac{\delta}{\gamma}\right)^{-1/2}\right)\right]. \tag{22}$$

The μSR data for YMn_2 taken after [8] and $GdMn_2$ taken from [9] were analysed using equation (22). The fitting parameters are given in table 1 and the temperature dependences of the relaxation rates are presented in figures 2 and 3, respectively. For YMn_2 the results for heating and cooling runs give comparable values of γ. For $GdMn_2$ (figure 3) we show fits with T_N as the free parameter (solid line) and with fixed T_N (broken line) to the values calculated in
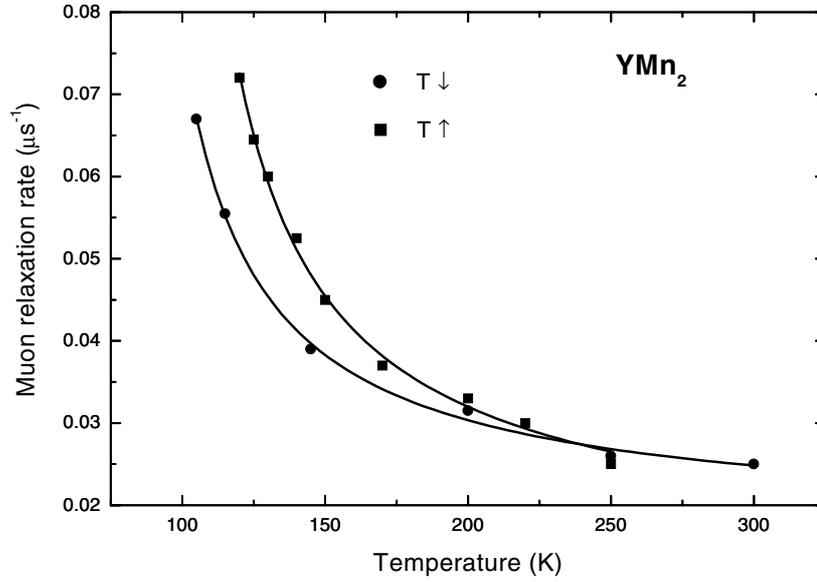
Figure 2. The muon relaxation rate λ as a function of temperature for YMn$_2$. The experimental points are taken from [8] ($\downarrow$: measured on cooling; $\uparrow$: measured on heating); the solid curves are the curves fitted with equation (22).

the paper [8] using equation (1). It is interesting that curves fitted with fixed and free T_N-values do not differ too much, whereas the γ-values are apparently sensitive to T_N (see table 1). For GdMn$_2$, at zero pressure, the curve for fixed T_N is far out from the experimental points. The reason is that the fixed Néel temperature (99 K) is very close to the first experimental point (100 K), so the fit with T_N as a free fitting parameter is much better. For GdMn$_2$, γ is smaller than for YMn$_2$. It is interesting that γ increases with pressure. This means that decreasing interatomic distances cause stronger dependence of the exchange interactions on the k-vector, which seems quite reasonable.

In our previous paper [5] we fitted the μSR relaxation rates λ for YMn$_2$D$_x$ with $x = 0.5$, 1, 2, 3 and in [6] the NMR relaxation rates T_1 and T_2 of ^{2}D for $x = 0.65$, 1, 1.5, 2, 2.5. In figure 4 the relation between the γ-parameter and deuterium concentration x as obtained from the fits to the formulae for λ, T_1 and T_2 [6] together with γ for YMn$_2$ (see table 1) are presented. It is also interesting that the γ-value for YMn$_2$ is higher than that for YMn$_2$D$_{0.5}$ in accordance with the tendency visible for YMn$_2$ deuterides (see figure 4). The curve in figure 4 is just a guide to the eye and visualizes the evident decrease of γ with deuterium content. It is also noteworthy that γ obtained from the fits of λ, T_1 and T_2 agrees reasonably well for each deuterium concentration. The γ-parameter describes the strength of the influence of the wavevector on the susceptibility dispersion relation (14). Making an expansion as in equation (13), we get a dependence of the exchange coupling constant J on γ in the form

$$\frac{J(\boldsymbol{k})}{J(\boldsymbol{K_0})} = 1 - \gamma(\boldsymbol{k} - \boldsymbol{K_0})^2. \tag{23}$$

This led us in [6] to the conclusion that the decrease of the γ-value with increasing deuterium concentration presented in figure 4 can be understood as a change towards more uniform (less k-dependent) exchange interactions for higher deuterium concentrations, for which Mn–Mn distances increase and Mn moments are better localized.
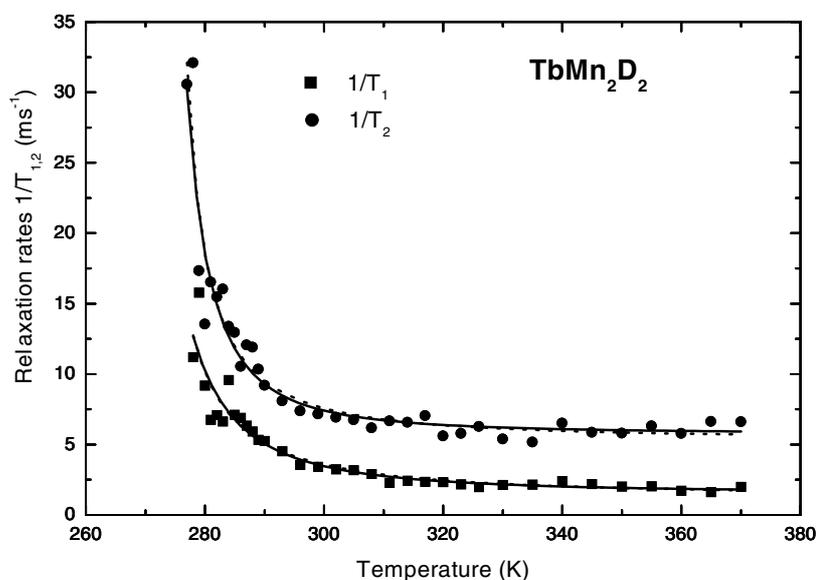
Figure 3. The muon relaxation rate λ as a function of temperature for GdMn$_2$ for three values of the external pressure. The experimental points are taken from [9]; the solid curves are the curves fitted by use of equation (22) with T_N as a free parameter; the dotted curves are the curves fitted with fixed T_N to the values from [9].

Figure 4. The compositional dependence of γ for YMn$_2$D$_x$ taken from [6] and a value of γ obtained by fitting to the μSR data for YMn$_2$ taken from [8]. The dot–dashed curve is a guide to the eye only.

The parameters obtained from fitting the formula (19) to the recent results for ^{2}D relaxation times T_1 and T_2 for TbMn$_2$D$_2$ [7] are collected in table 1. In figure 5 we present the results of

Figure 5. The relaxation rates $1/T_1$ and $1/T_2$ as a function of temperature for TbMn$_2$D$_2$. The experimental points are taken from [7]; the solid curves are the curves fitted using equation (19) with T_N as a free parameter; the dotted curves are curves fitted with T_N fixed to the values from [7].

the fitting of T_1 and T_2 for TbMn$_2$D$_2$. It is clear that the results are not very sensitive to the temperature T_N, because the fits with T_N as a free parameter (solid line) differ only slightly from those with T_N fixed to the values determined from the empirical formula [7]. In our fits we have also assumed that the temperature-independent part of relaxation rates related to the terbium exists [7]. The values of γ for TbMn$_2$D$_2$ are about three times higher than those for YMn$_2$D$_2$. We suggest that this is caused by terbium atoms with large (about 9 μ_B) magnetic moments, that may indeed cause a stronger dependence of the exchange interactions on the $\boldsymbol{k}$-vector.

The comparison of changes of γ as a function of deuterium content in YMn$_2$D$_x$ and changes of γ with pressure in GdMn$_2$ is also very interesting. Deuterium atoms in interstitial sites cause increase of the unit volume, acting as 'negative pressure'. Indeed, we see that positive pressure increases the γ-value (see table 1) whereas 'negative pressure' causes decrease of γ (see figure 4). These findings led us to the conclusion that the increase of the Mn–Mn distances causes flattening of the $J(\boldsymbol{k})$ function, whereas shortening of the Mn–Mn distances causes $J(\boldsymbol{k})$ to be peaked, which seems quite reasonable.

4. Conclusions

The model presented for the quasicritical behaviour of the NMR relaxation rates T_1 and T_2 as well as the muon relaxation rate for ferromagnetic and antiferromagnetic cubic materials was discussed and used to analyse the data for Y(Co$_{1-x}$Al$_x$)$_2$ (weakly ferromagnetic), YMn$_2$, GdMn$_2$, YMn$_2$D$_x$ and TbMn$_2$D$_2$ (antiferromagnets). The proposed formulae (12), (19) are compatible with the simplified Moriya model of critical behaviour, as they give appropriate limiting values of the critical exponent η close to the critical point. In the results of the numerical calculations with use of our formulae, the most meaningful physical parameter of

the model (γ) has been obtained and analysed. The parameter γ describes the dependence of the staggered susceptibility on wavevector $\boldsymbol{k}$ (equations (4) and (13)) and the $\boldsymbol{k}$-dependence of the exchange coupling constant $J(\boldsymbol{k})$ as well. The fits made by means of our formulae (12), (19) to the experimental data taken from different sources (μSR and NMR data) gave consistent results (table 1).

On the other hand the values of $T_{C,N}$ obtained from fits within our model are systematically lower than the values obtained from other static measurements (e.g. magnetic susceptibility) or from fits to the empirical formulae with a critical exponent (1). This discrepancy results from a systematic deviation in the temperature measurements, different definitions used to determine $T_{C,N}$ and the sharpness of the critical transition. The γ-values are in a reasonable range, which attests to the correctness of our model. The increase of γ with increasing pressure, positive in GdMn$_2$ and 'negative' in YMn$_2$D$_x$, can be related to $J(\boldsymbol{k})$ dependence on the Mn–Mn distance, which seems very reasonable. Lovesey *et al* presented sophisticated theoretical models for μSR in ferromagnets [3] and in antiferromagnets [4]. However, even for the authors of [3, 4] it was difficult to verify their theory with experimental data. So we propose a parallel model, which—as we showed—is in reasonably good agreement with the available experimental results. We hope that the proposed extension of Moriya's theory provides better insight into the origin of critical behaviour and a better understanding of the role of spin fluctuations in relaxation phenomena preceding magnetic ordering.

Acknowledgment

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