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^{69,71}Ga-NMR study of the low-energy excitations in $NdGa_2$

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Abstract. We report the results of ^{69}Ga- and ^{71}Ga-NMR measurements on $NdGa_2$ at temperatures between 0.1 and 3 K and in applied magnetic fields between zero and 74 kOe. $NdGa_2$ orders antiferromagnetically below 9.5 K and undergoes several metamagnetic transitions in external magnetic fields. In zero applied magnetic field and below 1 K the temperature dependence of the spin-lattice relaxation rate $T_1^{-1}(T)$ shows a large linear-in-T term, about two orders of magnitude higher than for the reference compound $LaGa_2$. This strong enhancement confirms the presence of low-energy excitations in the antiferromagnetic phase of $NdGa_2$ as was previously indicated by specific heat data. Above 1 K, $T_1^{-1}(T)$ is dominated by an exponential term, which we associate with excitations between the lowest energy levels of the f-electron system. The separation of these energy levels is determined by exchange, crystal-field and Zeeman interactions.

PACS. 75.20.Hr Local moment in compounds and alloys; Kondo effect, valence fluctuations, heavy fermions – 75.30.-m Intrinsic properties of magnetically ordered materials – 76.60.-k Nuclear magnetic resonance and relaxation

1 Introduction

$NdGa_2$ is a hexagonal intermetallic compound whose magnetic properties have extensively been studied in the last decades [1–4]. The magnetic properties of $NdGa_2$ are strongly anisotropic [3], the easy axis of magnetization lying in the basal plane along the [120] direction. In zero applied magnetic field $NdGa_2$displays two subsequent magnetic phase transitions, to an amplitude modulated moment ordering [2,3] at $T_N = 9.5$ K and to an antiphase magnetic structure [3] below $T_t = 7.5$ K. The application of external magnetic fields induces several metamagnetic transitions in moderate fields at low temperatures [3]. The interest in this compound has recently been renewed due to new results of specific heat measurements [5] where an unexpectedly high value of the linear-in-T contribution to the specific heat, of about 150 mJ/molK^2 below 1 K, has been observed.

$NdGa_2$ crystallizes with an AlB_2-type structure [6], belonging to the $hP3$ space group. The lattice parameters are $a = 4.281$ Å and $c = 4.306$ Å. The unit cell has a single Nd site (1a) with $6/mmm$ point-group symmetry and therefore, the Nd^{3+} $J = 9/2$ ground state is split into five doublets by the crystal electric field. The Nd–Nd nearest neighbor distance is 4.281 Å. All the Ga sites are equivalent (2d) and they have the $\bar{6}m2$ point-group symmetry and six Nd nearest neighbors at a distance of 3.278 .

The magnetic structure below T_t has been inferred from the results of neutron diffraction experiments [3,4]. It is an antiphase magnetic structure (so named in Ref. [3]), where the value of the ordered magnetic moment is $M = (3.3 \pm 0.2)\mu_B$ parallel to the [010] direction, very nearly the value expected for the Hund's rule ground-state of the free Nd^{3+} ion. The propagation vector of the antiferromagnetic alignment is $Q = (0.136, 0, 0.014)$ which implies an incommensurate or very long period magnetic structure.

In view of the high value of the linear-in-T contribution to the specific heat, we have performed ^{69,71}Ga-NMR relaxation rate measurements to learn more about the low-energy excitations in this system. These measurements were done at temperatures below 3 K at zero applied magnetic field, *i.e.*, in the low temperature ordered magnetic phase, and in magnetic fields between 17 and 74 kOe where the system is not in an antiferromagnetic state, but rather adopts an induced ferromagnetic state.

For obtaining reference information on the non magnetic background, we have also made NMR experiments on the reference compound $LaGa_2$. This compound has the same crystal structure as $NdGa_2$ and, apart from the Nd $4f$-electrons, similar electronic configurations are expected for both these materials.

2 Experimental aspects

Our $NdGa_2$ and $LaGa_2$ samples were synthesized by arc melting the constituent elements in proper proportions on

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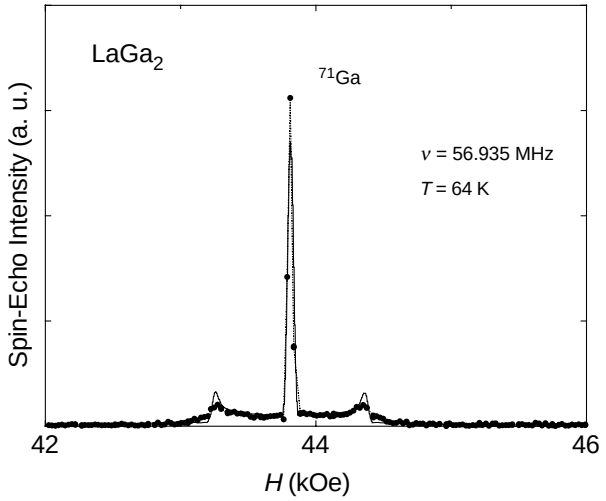

Fig. 1. NMR-spectrum of $LaGa_2$ at a temperature of 64 K and a frequency of 56.935 MHz for a randomly oriented powder. The filled circles, connected by a dotted line, represent the measured data and the solid line represents the result of a computer simulation (see text).

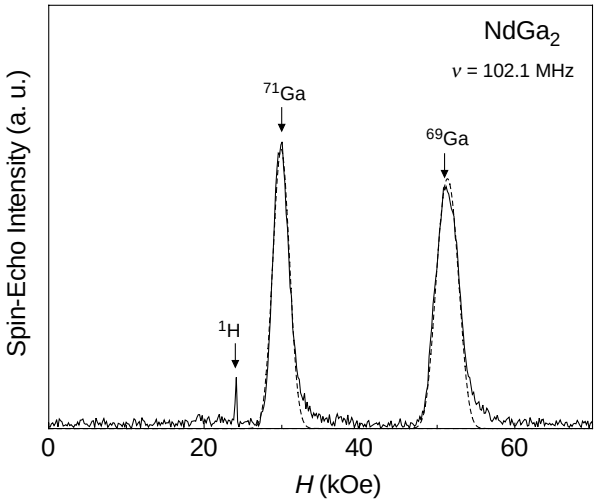

Fig. 2. NMR-spectrum of $NdGa_2$ at a temperature of 0.1 K and a frequency of 102.1 MHz. The solid line represents the measured data, and the dashed line represents the result of a computer simulation for both isotopes (see text). The hydrogen line is due to the sample container.

a copper hearth, placed in a tetra arc furnace and exposed to an argon atmosphere. The samples were subsequently annealed for 50 hours at 950 °C. The results of susceptibility and magnetization measurements performed on our powdered $NdGa_2$ sample show the same antiferromagnetic and metamagnetic transitions that have previously been reported for $NdGa_2$ [3].

All our Ga-NMR measurements were performed using standard spin-echo techniques. We powdered our samples under He atmosphere into grains of linear dimensions smaller than 100 μm, placed them into a teflon container, and immersed them directly into the $^3He-^4He$ mixture of a top-loading dilution refrigerator equipped with an 8 tesla magnet. The NMR spectra at fixed frequencies were obtained by integrating the spin-echo intensities at a given external magnetic field and changing stepwise the magnetic field. For measuring the spin-lattice relaxation rate T_1^{-1} we rotated the nuclear magnetization out of equilibrium by inducing nuclear Zeeman transitions $m \leftrightarrow m-1$ with a single rf-pulse, and monitored the integrated spin-echo intensity $m(t)$ after a variable recovery time t.

3 Experimental results

3.1 NMR spectra

In Figure 1 we show an example of a ^{71}Ga spectrum of $LaGa_2$ at a temperature of 64 K and a frequency of 56.935 MHz. The shape of this spectrum is typical for a randomly-oriented powder containing spin 3/2 nuclei. We fit the NMR spectra assuming a suitable Hamiltonian that contains Zeeman and quadrupole terms and is given, *e.g.*, in reference [7]. We assume also an isotropic Knight-shift, $K_{\rm iso}$, the same for each Ga isotope, due to the polarisation of the conduction electrons. From these fits we obtain for the quadrupolar coupling constants $e^2qQ/h = 3.0$ and 4.6 MHz for ^{71}Ga and ^{69}Ga respectively, reflecting the ratio of the corresponding quadrupolar moments [8]. For the Knight-shift we obtain $K_{\rm iso} = 0.1\%$.

The measurements of the spectra of $NdGa_2$ at several fixed frequencies, with applied magnetic fields above 15 kOe, yield very similar internal magnetic fields $H_{\rm i} \approx$ 48 kOe, indicating that for these experiments the moments are fully aligned in the direction of the applied magnetic fields [9], and that the crystallites are oriented with their c-axis perpendicular to the direction of the applied magnetic field (see Ref. [10]). In Figure 2 we show an example of a spectrum of $NdGa_2$ at a temperature of 0.1 K and at a frequency of 102.1 MHz in the induced ferromagnetic phase. The two prominent peaks in this figure correspond to the isotopes ^{71}Ga and ^{69}Ga. The $NdGa_2$ NMR spectra were fitted again using a Hamiltonian with Zeeman and quadrupole terms and an additional Gaussian inhomogeneous broadening, due to the anisotropy of the magnetization along the [100] and [120] directions and to the distribution of demagnetization factors [11]. The quadrupolar coupling constants were assumed to be the same as for $LaGa_2$. Contrary to the case of $LaGa_2$ all the crystallites were oriented by the applied magnetic field and therefore the fit functions consist of three Gaussians, corresponding to the central line $1/2 \leftrightarrow -1/2$ and the two satellite lines $\pm 3/2 \leftrightarrow \pm 1/2$. The value of the standard deviation σ of the Gaussians is the same for both Ga isotopes, $\sigma = 1$ kG. The difference of the widths of the NMR lines is attributed to the different quadrupolar coupling constants of the two isotopes.

From the measured NMR spectra we obtain the line shift $\Delta H = H_{\rm ref} - H_{\rm m}$, where $H_{\rm ref}$ is the resonant magnetic field for ^{69}Ga or ^{71}Ga in diamagnetic reference compounds and $H_{\rm m}$ is obtained from the center of the Gaussian corresponding to the central line $1/2 \leftrightarrow -1/2$. From our NMR results and the magnetization M data [3],

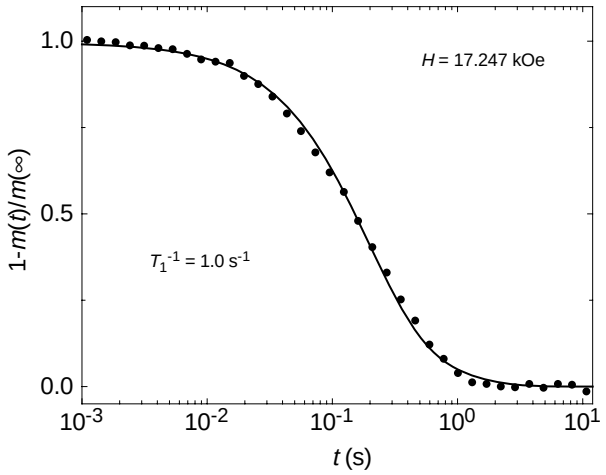

Fig. 3. Nuclear magnetization recovery $m(t)$ of $NdGa_2$ at $T = 0.11$ K and $\nu = 66.81$ MHz. The solid line represents a fit to the data (see text).

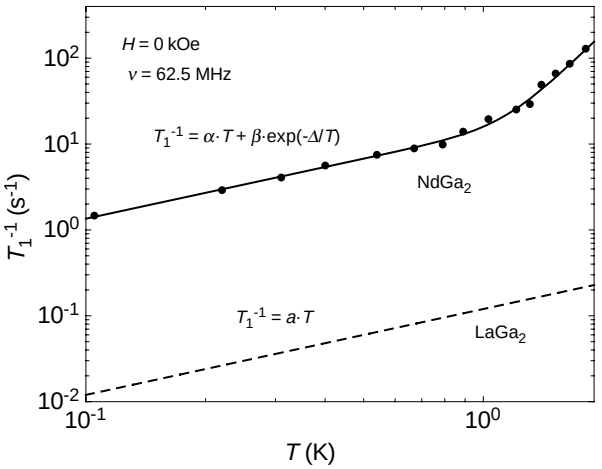

Fig. 4. Spin-lattice relaxation rate of $NdGa_2$ in zero applied magnetic field. The solid line is a fit to the data (see text) with $\alpha = 13.6$ (sK)$^{-1}$, $\beta = 1.4 \times 10^4$ s^{-1} and $\Delta = 8.9$ K. The dashed line show the spin-lattice relaxation rate of $LaGa_2$ extrapolated from data at high temperatures with $a = 0.12$.

we calculate a transferred hyperfine coupling, $H_{\rm hf} = \Delta H/M$, of 15 kOe/$\mu_{\rm B}$ at the Ga site, a much larger value than the 0.1 kOe/$\mu_{\rm B}$ obtained from a simple estimate assuming dipolar fields only. Therefore, we associate the hyperfine coupling with the transfer mechanism provided by conduction electrons.

The adopted magnetic structure below $T_{\rm t}$, in zero applied magnetic field, implies for most of the Ga nuclei a ferromagnetic arrangement of the nearest neighbors (see Ref. [4]). Hence, the internal magnetic fields at most of the Ga nuclei are expected to be of similar magnitude as in the induced ferromagnetic phase, *i.e.*, 48 kOe. This value was confirmed by a measurement of an NMR-spectrum at the fixed frequency of 62.5 MHz, corresponding to the Larmor frequency of ^{71}Ga in a magnetic field equal to the inferred internal field of 48 kOe. Indeed this spectrum showed a broad maximum at zero applied magnetic field.

3.2 Spin-lattice relaxation rates

3.2.1 Spin-lattice relaxation rate of $LaGa_2$

We have measured the ^{69,71}Ga spin-lattice relaxation rate of $LaGa_2$, at a frequency of 56.93 MHz, for temperatures between 22 and 240 K. The corresponding resonant magnetic fields for both isotopes ^{69}Ga and ^{71}Ga are 55.7 kOe and 43.8 kOe respectively. Since for $LaGa_2$ the $+1/2 \leftrightarrow -1/2$ transition is very well defined (see Fig. 1), we performed our T_1^{-1} measurements by irradiating only this transition. In this case the expected [12] and observed functional form of the nuclear magnetization recovery, $m(t)$, is the sum of two exponentials (see Eq. (1) below, with $a = 1$). The spin-lattice relaxation rate may approximately be represented by a Korringa-type law, indicating metallic behavior. For ^{71}Ga we obtain $(T_1T)^{-1} \approx 0.12$ (K s)$^{-1}$ and for ^{69}Ga, $(T_1T)^{-1} \approx 0.08$ (K s)$^{-1}$. The ratio of the relaxation rates measured for the two different isotopes is as expected for a relaxation of magnetic origin where the relaxation rate is proportional to the square of the gyromagnetic ratio.

The Korringa constants for both Ga isotopes are only about a factor two larger than the expected values assuming that the itinerant electrons are represented by a Fermi gas of noninteracting electrons. This situation is similar to that of a typical metal like Cu [13].

3.2.2 Spin-lattice relaxation rate of $LaGa_2$

Due to the inhomogeneous broadening the individual nuclear Zeeman transitions ($m \leftrightarrow m-1$) of the NMR-spectra of $NdGa_2$ could not be resolved. The relaxation rate measurements were performed by irradiating only a narrow range at the center of the NMR spectra, for which we estimated [14] the contribution a of the central transition $1/2 \leftrightarrow -1/2$ and $(1-a)$ of the satellite lines $\pm 3/2 \leftrightarrow \pm 1/2$ from fits to the measured NMR spectra. The function describing the recovery of the nuclear magnetization is then [12]

$$1 - \frac{m(t)}{m(\infty)} = C\{a(0.1\mathrm{e}^{-t/T_1} + 0.9\mathrm{e}^{-6t/T_1}) + (1-a)(0.1\mathrm{e}^{-t/T_1} + 0.5\mathrm{e}^{-3t/T_1} + 0.4\mathrm{e}^{-6t/T_1})\}. \quad (1)$$

This function contains two fitting parameters, *i.e.*, an overall scaling constant C and the spin-lattice relaxation rate T_1^{-1} . In Figure 3 we show an example for the recovery of the magnetization at 0.11 K and for an applied magnetic field of 17.247 kOe, *i.e.*, where the system is in the induced ferromagnetic state. The solid line represents a fit to the data using equation (1), which agrees very well with the measured data over four decades of the recovery time t.

In Figure 4 we present the results of our T_1^{-1} measurements for $NdGa_2$ in zero applied field (in the antiferromagnetically ordered state) at a frequency $\nu = 62.5$ MHz

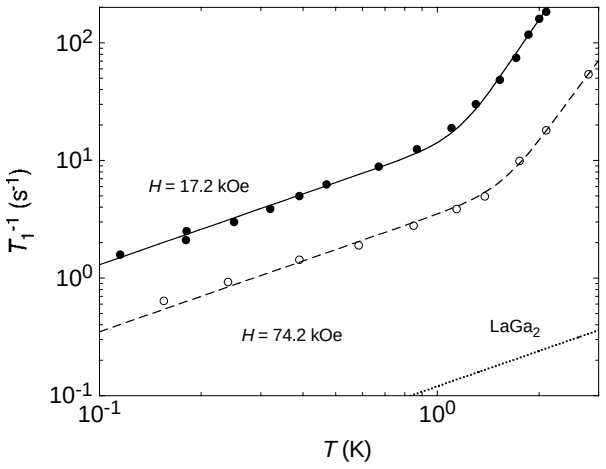

Fig. 5. Spin-lattice relaxation rate of $NdGa_2$ in applied magnetic fields of 17.2 kOe (filled circles) and 74.2 kOe (empty circles). The solid and the dashed lines are fits to the data (see text). The dotted line shows the spin-lattice relaxation rate of $LaGa_2$ extrapolated from data at high temperatures.

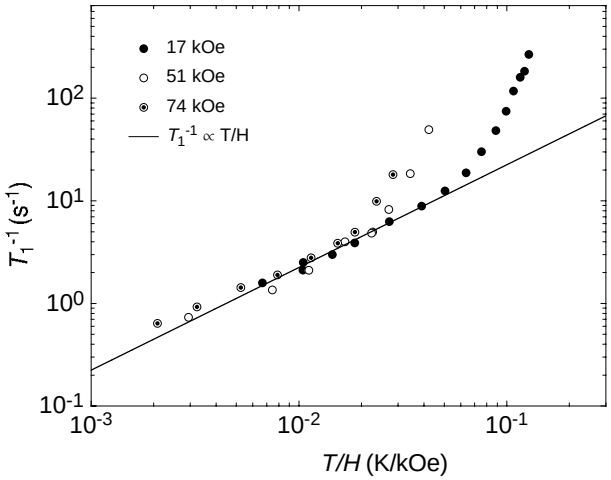

Fig. 6. Spin-lattice relaxation rate of $NdGa_2$ in the induced ferromagnetic phase as a function of T/H for three different applied magnetic fields. The solid line is a linear fit to the data below 1 K.

as function of the temperature. As a reference we have also plotted in the same figure the spin-lattice relaxation rate of $LaGa_2$ extrapolating the Korringa behavior from T_1^{-1} measurements performed between 22 and 240 K as described above. Since the relaxation rate in $NdGa_2$ is much higher than in the reference compound $LaGa_2$, the enhancement of $T_1^{-1}(T)$ of $NdGa_2$ is naturally associated with excitations within the f-electron system. For $NdGa_2$ in zero applied magnetic field we find that $T_1^{-1}(T)$ can be described by a linear and an exponential contribution to the relaxation rate as

$$T_1^{-1}(T) = \alpha T + \beta \mathrm{e}^{-\Delta/T} \qquad (2)$$

with $\alpha = 13.6\ (\mathrm{sK})^{-1}$, $\beta = 1.4 \times 10^4\ \mathrm{s}^{-1}$ and $\Delta = 8.9$ K. The relatively large linear-in-T term, α, in $T_1^{-1}(T)$ confirms the presence of low-energy excitations in the f-electron system. One observes that the ratio $r_\alpha \equiv \alpha_{NdGa_2}/\alpha_{LaGa_2} \approx 110$ and the corresponding ratio for the linear-in-T contribution to the specific heat [5] $r_\gamma \equiv \gamma_{NdGa_2}/\gamma_{LaGa_2} \approx 40$, implying that α scales like $\gamma^{1.3}$ as opposed to a simple Fermi liquid where α is proportional to γ^2. However this type of behavior is often encountered in intermetallic compounds containing f-electron moments [15]. The exponential term in (2) may be associated with a thermally activated process, across an energy gap Δ of the order of 9 K. As will be shown later, we associate this relaxation with excitations between the lowest crystal-field split energy levels of the f-electron system.

In Figure 5 we show the results of the spin-lattice relaxation rate measurements $T_1^{-1}(T)$ for two different magnetic fields, both large enough to flip the moments and suppress completely the antiferromagnetic state. $T_1^{-1}(T)$ can again be well described by equation (2), represented by the solid and dashed lines. Although the relaxation rate is significantly reduced by the application of external fields, it is still strongly enhanced above the values obtained for $LaGa_2$, at least up to $H = 75$ kOe.

The prefactors of the linear-in-T terms of the relaxation rate α, and of the specific heat [5], γ, seem to have different magnetic field dependences. In the case of the specific heat [5], γ decreases rapidly as function of the applied field and at 17 kOe it is roughly 15% of the value in zero field, corresponding to an enhancement of only a factor of 5 relative to the reference compound $LaGa_2$. On the other hand $T_1^{-1}(T, H)$ for 17 kOe is still over two orders of magnitude larger than for the corresponding values of T_1^{-1} for $LaGa_2$. Thus the origin of the high value of α cannot simply be the result of a high density of electronic excitations in the antiferromagnetically ordered state. For higher magnetic fields $T_1^{-1}(T, H)$ scales linearly with (T/H) below 1 K, as indicated by the solid line in Figure 6. This T/H dependence for T_1^{-1} has been observed in other rare-earth compounds, such as $CeAuAl_3$ and $CePd_2In$, in the induced ferromagnetic regime [16,17].

4 Discussion

4.1 Low-energy excitations

The presence of a high density of low-energy excitations in the antiferromagnetic phase below 1 K is clearly shown by the large linear-in-T term of the zero-field NMR relaxation rate which is two orders of magnitude larger for $NdGa_2$ than for the reference compound $LaGa_2$. In $NdGa_2$, in contrast to $LaGa_2$, high internal magnetic fields are present at the studied nuclei, whose fluctuations can produce a high relaxation rate. In this particular case these fluctuations are described as magnon excitations. However, we note that a conventional antiferromagnetic phase, with a gap in the magnon spectrum, would lead to an exponential T-term in the relaxation rate [18] at very low temperatures, and an antiferromagnetic phase with a gapless magnon

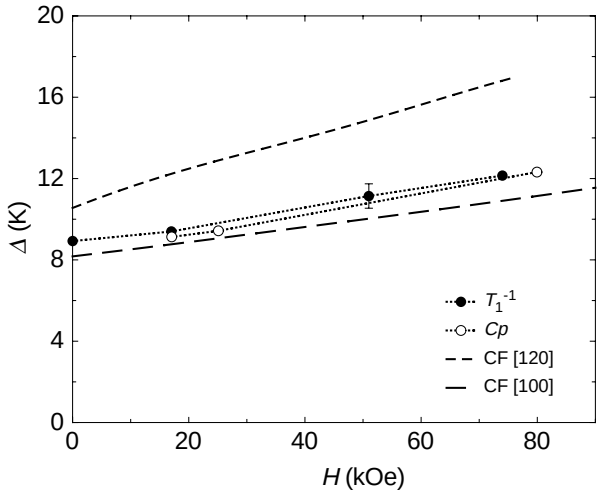

Fig. 7. Comparison of the energy gap inferred from the relaxation rate (filled circles) and from the specific heat (empty circles) data with estimates of the energy separation between the two lowest crystal-field energy levels for applied magnetic fields, H, along the [100] and the [120] direction (dashed lines). A typical error bar is given for the energy gap inferred by the relaxation rate. The corresponding error bars for the specific heat results are estimated to be of the same order.

spectrum would lead to a T^3 term in the relaxation rate [18]. Neither case applies to our observations.

It has previously been suggested [5] that the low energy excitations might be associated with the incommensurability of the magnetic phase of $NdGa_2$ in a similar way as it has been proposed for the case of $CeAl_2$ by Hudák *et al.* [19]. It is interesting to notice that, although $CeAl_2$ and $NdGa_2$ are very different systems, the ratio of the linear-in-T contribution to the relaxation rates of $NdGa_2$ and the reference compound $LaGa_2$, $r_\alpha = 110$, is very similar to the enhancement obtained for $CeAl_2$ if compared to $LaAl_2$ [20], where a substantial part of the linear part in the relaxation rate quite likely originates from features of the incommensurate antiferromagnetic structure [19]. For $NdGa_2$ this picture seems to gain special support from the observed field dependence of the specific heat [5], where the linear-in-T contribution to the specific heat is strongly suppressed by the application of fields of the order of 17 kOe, which is expected to completely suppress the antiferromagnetic ordering. From our NMR results, however, we conclude that a substantial enhancement of the relaxation rate persists in the induced ferromagnetic state. This observation implies an important contribution from a common mechanism for $(T_1T)^{-1}$ in the antiferromagnetic and in the induced ferromagnetic phase. The question of the origin of these low energy excitations thus needs to be reconsidered.

4.2 Energy-gap

In Figure 7 we have plotted the magnetic field dependence of the energy gap in the $4f$ electronic excitation spectrum as it results from fits to the measured data for $T_1^{-1}(T)$ (filled circles) using equation (2) and to the $C_{\rm p}(T)$ data (empty circles) from reference [5]. For the analysis of the specific heat we used a fit function appropriate for a two level system separated by a gap Δ. From Figure 7 it may be seen that the energy gaps obtained from $T_1^{-1}(T)$ and $C_{\rm p}(T)$ are very similar in magnitude and field dependence. Aiming at interpreting these data, we have estimated the energy separation between the two lowest f-electron energy levels as a function of the applied magnetic field assuming an appropriate crystal-field scheme and a molecular field. The crystal-field parameters were obtained from inelastic neutron scattering, susceptibility and specific-heat measurements [4], and the molecular field $H_{\rm Mol}$ of 62 kOe, was inferred from the paramagnetic Curie temperature [4]. We show in Figure 7 (dashed lines) the result of this estimate, for applied magnetic fields along the [120] or the [100] direction. We observe in this figure a qualitatively good agreement of the calculations explained above with the energy gaps deduced from $T_1^{-1}(T)$ and $C_{\rm p}(T)$. Therefore, by extrapolating to $H = 0$, and $H_{\rm Mol} = 0$, we conclude that our NMR data and the new analysis of the $C_{\rm p}(T)$ data support a crystal-field split level scheme in the paramagnetic state with the Γ_7 doublet as the ground state and the $\Gamma_8^{(1)}$ first excited doublet at 9 K.

5 Summary

The presence of low-energy excitations in $NdGa_2$, associated with the f-electron system, is confirmed by a large linear-in-T term in the relaxation rate below 1 K. In zero applied magnetic field the linear-in-T term is about two orders of magnitude higher than for $LaGa_2$, and is only moderately reduced by the application of external fields. It is still considerably enhanced above the values obtained for $LaGa_2$ even in the induced ferromagnetic state, contrary to what may be expected from considering the behavior of $C_{\rm p}(T, H)$. The relaxation rate $T_1^{-1}(T, H)$ in the induced ferromagnetic phase below 1 K shows a gradual reduction proportional to T/H. Additional exponential terms in $T_1^{-1}(T)$ and $C_{\rm p}(T)$ may be associated with electronic excitations between the two lowest crystal-field split energy levels.

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10. The results of measurements of the bulk magnetization of $NdGa_2$ single crystal (Ref. [3]) indicate that when the crystal c-axis is oriented perpendicularly to the applied magnetic field, H, the magnetization saturates for $H \approx 15$ kOe, and for the case where the crystal c-axis is parallel to the applied magnetic field, the magnetization saturates at much larger applied magnetic fields. Our measurements of the NMR spectra of oriented powder are consistent with the former case.
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