Eur. Phys. J. B **16**, 233–239 (2000)

THE EUROPEAN PHYSICAL JOURNAL B

EDP Sciences

Thermal expansion and magnetostriction in an anomalous Gd intermetallic compound

J.W. Taylor[1], H. Capellmann[2], K.-U. Neumann[1], and K.R.A. Ziebeck[1,a]

[1] Department of Physics, Loughborough University, Loughborough LE11 3TU, UK
[2] Institut für Theoretische Physik, RWTH Aachen, Germany

Received 28 July 1999

Abstract. Thermal expansion and forced magnetostriction measurements are reported on two Gd intermetallic compounds which order magnetically below ∼10 K. The relative influence of the electronic, lattice and magnetic degrees of freedom was determined using results obtained on a non-magnetic isostructural compound. A Grüneisen analysis revealed that whilst the magnetic contribution to the specific heat is similar for both Pd_2GdIn and Cu_2GdIn the spontaneous magnetostriction was significantly smaller in the Pd compound. Forced magnetostriction measurements suggest that the thermal expansion in Pd_2GdIn is primarily associated with spin fluctuations in the Pd 4*d* band. It is suggested that these additional degrees of freedom give rise to the enhanced specific heat observed in Pd_2GdIn.

PACS. 75.30.-m Intrinsic properties of magnetically ordered materials – 65.70.+y Thermal expansion and density changes; thermomechanical effects – 75.80.+q Magnetomechanical and magnetoelectric effects, magnetostriction

1 Introduction

Inter-metallic compounds containing rare earth elements exhibit a wide variety of properties, which arise from the relative importance of the lattice and electronic degrees of freedom and their possible interplay. For systems in which coherent lattice and magnetic degrees of freedom are involved polarised neutron scattering can sometimes be employed to determine not only the temporal and spatial dependence of the modes but also the absolute magnitude. In many cases it is not possible to experimentally separate the different contributions and unambiguously identify the mechanism or mechanisms involved. Analysis of experimental data therefore usually proceeds with resort to models. In order to quantify the influence of certain mechanisms, studies have been made on systems in which the number of degrees of freedom have been restricted. This has led to considerable interest in compounds in which the sole magnetic element is gadolinium. Gadolinium has a particularly stable $4f$ shell, it is an S state ion, and is therefore unaffected by the crystal field. Furthermore the coupling of the lattice and electronic degrees of freedom arising from spin-orbit interaction is expected to be negligible. Despite this potential simplification, some Gd compounds are found to have unusual low temperature properties. In particular for $Gd_{1-x}Y_xNi_2Si_2$ [1–3] alloys, $GdCu_2Si_2$, $GdGa_2$, $GdCu_5$ [3] and Pd_2GdIn [4] an enhanced specific heat has been reported above the Néel temperature giving rise to an anomalously large linear contribution. For the former alloys this behaviour was thought to arise from extensive short-range ferromagnetic correlations extending well above the Néel temperature. However for Pd_2GdIn extensive analysis suggests that lattice degrees of freedom were involved. The present paper reports new results on one of these compounds, namely Pd_2GdIn, concerning the thermal expansion and forced magnetostriction.

Resistivity, specific heat [4] and magnetisation [5] measurements indicate that Pd_2GdIn orders magnetically below ∼9 K. However the order appears to be complex being antiferromagnetic in zero field but transforming to ferromagnetism under the influence of an applied field. The saturation moment extrapolated to 0 K is approximately 8 μ_B, which is 1 μ_B more than the 7 μ_B expected for a Gd^{3+} (gJ) ion. In the paramagnetic phase the observed effective moment is still slightly larger than the free ion value of Gd^{3+}. The additional moment observed in the ground state was tentatively attributed to the polarisation of the Pd ($4d$) band by the Gd ($4f$) local moments. However the specific heat is enhanced and remains so beyond 20 K. Measurements made on isostructural compounds in which the Gd was replaced by Lu [4] or the Pd by Cu [6] did not reveal anomalous behaviour which suggested a possible $4f$-$4d$ involvement in Pd_2GdIn.

[a] e-mail: K.R.Ziebeck@lboro.ac.uk

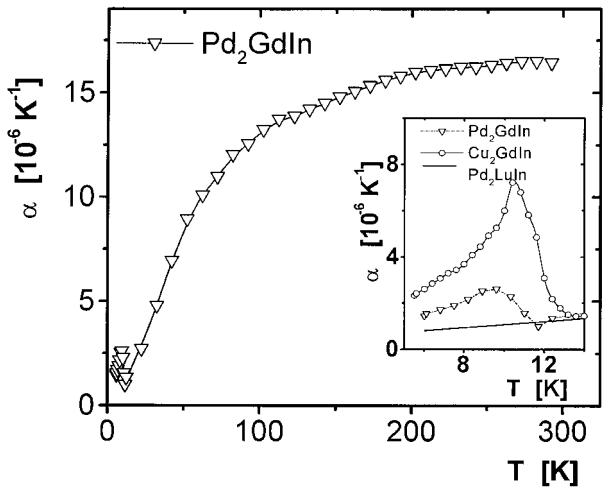

Fig. 1. The thermal expansion coefficient of Pd_2GdIn between 2 and 300 K. The inset shows the low temperature variation of Cu_2GdIn (o), Pd_2GdIn ($\triangledown$) and Pd_2LuIn (—). The lines connecting the data points for the two Gd compounds are only a guide for the eye.

2 Experimental

20 g polycrystalline samples of Pd_2GdIn, Cu_2GdIn and Pd_2LuIn were prepared by melting the appropriate quantities of spectrographically pure elements, in an argon arc furnace. X-ray powder diffraction confirmed the materials to be single phase and to have the Heusler structure and cell parameters as reported in the literature [4]. Pillar shaped specimens of dimension $4 \times 4 \times 20$ mm were spark eroded from each ingot.

Measurements of the thermal expansion and forced magnetostriction were made using a sensitive three terminal capacitance method which enabled length changes of $\frac{\Delta L}{L} \sim 10^{-10}$ to be detected. The specimens were mounted in a parallel plate capacitance cell made from oxygen free high conductivity copper. By placing the cell in a superconducting magnet, measurements could be made over the temperature range 2 to 300 K and in magnetic fields up to 7 T. Temperatures rendered stable to $\pm$ 0.1 K were measured using a Cernox sensor thermally anchored to the cell.

Although magnetisation measurements on these materials have previously been reported [5], further measurements were undertaken as a result of anomalies observed in the forced magnetostriction investigations. The magnetisation measurements were carried out as before using a SQUID magnetometer manufactured by Quantum Design.

3 Thermal expansion

The thermal expansion coefficient of Pd_2GdIn observed between 2 and 300 K is shown in Figure 1 together with the low temperature variation observed for all three compounds (inset). The total volume change between 0 and 300 K as given by

$$\int_0^{300\,\mathrm{K}} 3\alpha(T)\mathrm{d}T \tag{1}$$

is 1.12, 1.15% and 1.01% for Pd_2GdIn, Cu_2GdIn and Pd_2LuIn respectively. Above approximately 20 K the variation of α with temperature is similar for all three compounds. Since the compound containing Lu does not order magnetically the thermal expansion may be considered to arise only from the electronic and lattice contributions. If they are not coupled $\alpha(T)$ will comprise of two terms proportional to T and T^3 namely

$$\alpha(T) = AT + B\left(\frac{T}{\theta_\mathrm{D}}\right)^3 \int_0^{\theta_\mathrm{D}/T} \frac{x^4 \mathrm{e}^x}{(\mathrm{e}^x - 1)^2}\mathrm{d}x \tag{2}$$

where A and B are constants and θ_D is the Debye temperature. Therefore at low temperatures $T \leq \theta_\mathrm{D}/12$ a plot of $\alpha(T)/T \, versus \, T^2$ should within a Grüneisen regime produce a linear variation. A linear relation between $\alpha(T)/T$ and T^2 holds for Pd_2LuIn below $\sim$30 K. A least square fit was then used to determine the coefficients of the linear and cubic terms. The results of this analysis are given in Table 1 together with those obtained for Pd_2GdIn and Cu_2GdIn.

Table 1. The coefficients A and B obtained by least square regression of the low temperature approximation $\alpha(T) = T + BT^3$ to the low temperature thermal expansion data of Pd_2GdIn, Cu_2GdIn and Pd_2LuIn.

	$A \times 10^{-6}$ [K^{-2}]	$B \times 10^{-11}$ [K^{-4}]
Pd_2GdIn	0.101 $\pm$ 0.002	4.4 $\pm$ 0.3
Pd_2LuIn	0.107 $\pm$ 0.001	4.0 $\pm$ 0.2
Cu_2GdIn	0.062 $\pm$ 0.002	5.0 $\pm$ .03

Since the latter two compounds order magnetically only data between 12 K and 30 K was used in the analysis. It may be seen from Table 1, that the values for the coefficients are very similar for all three compounds. The Pd_2LuIn data was then used to extract the "magnetic thermal expansion" in the two compounds containing Gd. The magnetic thermal expansion so obtained is plotted as a function of temperature in Figure 2 for Pd_2GdIn and in Figure 3 for Cu_2GdIn. The spontaneous volume magnetostriction at 0 K which the lattice undergoes as a result of magnetic long range order is obtained using

$$\int_{T>T_\mathrm{N}}^{0} 3\alpha_\mathrm{m}(T)\mathrm{d}T \tag{3}$$

to give 2×10^{-3} % and 10×10^{-3} % for Pd_2GdIn and Cu_2GdIn respectively. The data was extrapolated to zero Kelvin assuming a linear variation.

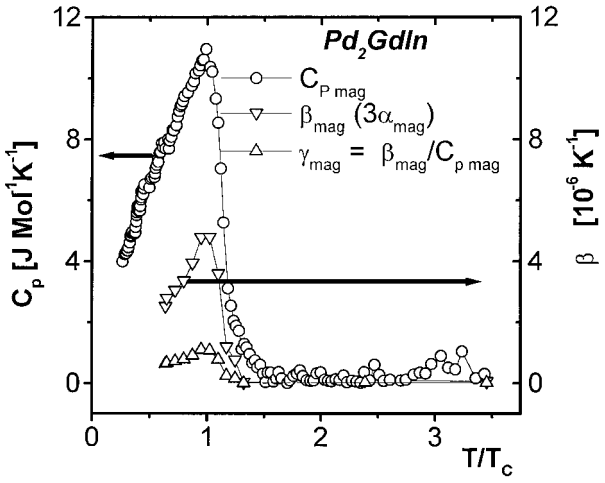

Fig. 2. The magnetic specific heat C_m, magnetic contribution to the coefficient of the thermal expansion β_m and ratio $\beta_\mathrm{m}/C_\mathrm{m}$ as a function of reduced temperature in Pd_2GdIn.

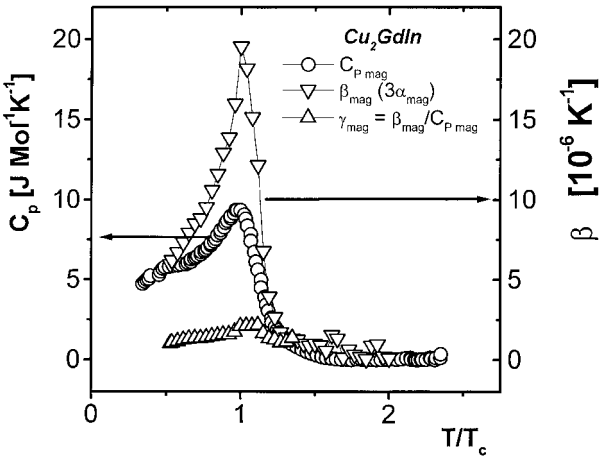

Fig. 3. The magnetic specific heat C_m, magnetic contribution to the coefficient of the thermal expansion β_m and ratio $\beta_\mathrm{m}/C_\mathrm{m}$ as a function of reduced temperature in Cu_2GdIn.

From Figures 2 and 3 spontaneous magnetostriction is clearly visible with a maximum in the magnetic contribution to the thermal expansion coefficient in the vicinity of the Néel temperatures. For both compounds the maximum in the specific heat and magnetic thermal expansion are in good agreement. At T_N the spontaneous volume magnetostriction ($\beta_\mathrm{m} = 3\alpha_\mathrm{m} = \frac{\mathrm{d}}{\mathrm{d}T}\frac{\Delta V}{V} = 3\frac{\mathrm{d}}{\mathrm{d}T}\frac{\Delta L}{L}$) is estimated to be $\sim 5 \times 10^{-6}$ K^{-1} and $\sim 20 \times 10^{-6}$ K^{-1} for Pd_2GdIn and Cu_2GdIn respectively. The change in magnetostriction is clearly larger in the copper compound whereas the relative jump in specific heat at T_N is essentially the same for both compounds [4], the ratio $C_\mathrm{m}/\beta_\mathrm{m}$ (10^6 J/mol) being 11/5 for Pd_2GdIn and 10/20 for Cu_2GdIn. In the case of Pd_2GdIn the magnetic specific heat was obtained after subtraction of an enhanced linear component as explained in reference [4]. Below T_N the specific heat of the palladium compound has an unusual linear dependence on temperature and is extended above the Néel temperature suggesting a significant degree of short range order. On the other hand, the thermal expansion does not exhibit the same degree of short-range order. As a consequence C_m is not proportional to β_m over the entire temperature range where magnetic order occurs. The specific heat of Cu_2GdIn has a thermal variation more generally observed in magnetic phase transitions. The short-range order is less pronounced than in Pd_2GdIn but follows that observed in the thermal expansion. However C_m is still only proportional to β_m over a limited range of temperature. In the low temperature limit, a similar analysis can also be made of the electronic a/γ and lattice b/β contributions to the specific heat and thermal expansion. Using the values of a and b given in Table 1 and the reported electronic γ and lattice β components of the specific heat [4] yields ratios of $\sim 10^{-8}$ and 10^{-11} for the electronic and lattice contributions for both Pd_2GdIn and Cu_2GdIn. The ratio of the magnetic to other contributions (electronic and lattice) at T_N is about 12:1 for specific heat measurements and for thermal expansion 7:1 (Cu_2GdIn) and 2:1 (Pd_2GdIn). The ratio $\beta(T)/C_\mathrm{m}$, which is plotted in Figures 2 and 3 for the two Gd compounds, leads to the determination of a magnetic Grüneisen parameter $\gamma_\mathrm{m} \cong \partial \ln U_\mathrm{m}/\partial \ln V$ which simplifies to $\alpha_\mathrm{m} = -(B\gamma_\mathrm{m}/3V)\, C_\mathrm{m}$ where B is the isothermal bulk modulus. The magnetic Grüneisen parameter provides information on the dependence of the isotropic magnetic interaction energy U_m on the inter-atomic separation.

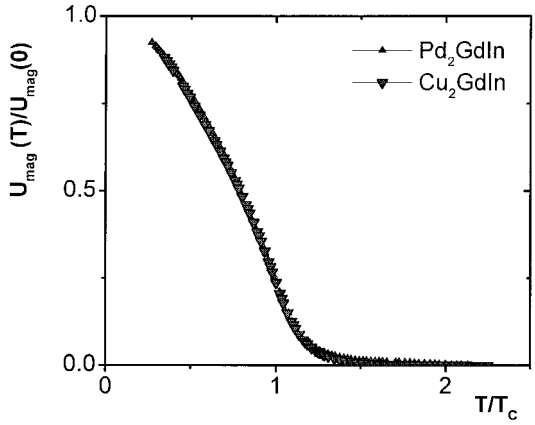

Fig. 4. The relative internal magnetic energy of Pd_2GdIn and Cu_2GdIn as a function of temperature obtained from the magnetic specific heat.

The internal magnetic energy at 0 K can be determined from the area of the specific heat anomaly as a function of temperature. In Figure 4 the relative internal magnetic energy is plotted as a function of reduced temperature.

$$\frac{U_\mathrm{m}(T)}{U_\mathrm{m}(0)} = \frac{\int_T^\infty C_\mathrm{m}\left(\widetilde{T}\right)\mathrm{d}\widetilde{T}}{\int_0^\infty C_\mathrm{m}\left(\widetilde{T}\right)\mathrm{d}\widetilde{T}}\,. \tag{4}$$

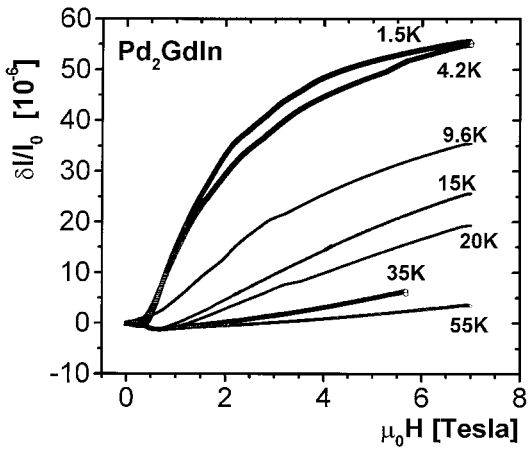

Fig. 5. Longitudinal forced magnetostriction isotherms for Pd_2GdIn.

In the absence of any coupling to the lattice this gives directly the thermal evolution of the spin-spin correlation function. This ratio is plotted for Pd_2GdIn and Cu_2GdIn as a function of reduced temperature in Figure 4, from which it may be seen that the variation is very similar in both compounds. If α_m is proportional to C_m the internal magnetic energy can also be obtained using the magnetic thermal expansion. However the magnetic Grüneisen relation does not hold for these compounds. In particular the spontaneous magnetostriction of Pd_2GdIn appears anomalously low. If a similar analysis is carried out using equation (4) and the magnetic thermal expansion coefficient β_m for both compounds, the results are similar to those obtained using C_m. Whilst the thermal variation is similar to that calculated for a two spin cluster model with nearest neighbour interaction [7], more information is required concerning the magnetic structure. Spin wave measurements on several ferromagnetic Heusler compounds [8,9] including one close to an antiferromagnetic instability [10] indicate that the exchange interactions are of long range and oscillatory as expected for an RKKY mechanism. In these isostructural compounds *e.g.* Pd_2MnIn the magnetic moment is confined to the Mn atoms which occupy the same crystallographic site as the Gd. There is negligible overlap of the $3d$ wavefunctions and the compounds provide good model wavefunctions for studying localised transition metal magnets. For the compounds containing $4f$ elements the magnetic order is associated with the Gd atoms which in the case of Pd_2GdIn induces a small moment on the palladium atoms. The field dependence of the magnetic isotherms in the magnetisation measurements [5] below T_N suggests that the magnetic order is complex with a continuous evolution towards ferromagnetism rather than a spin flop transition. Since Gd is a S state ion the moment is relatively stable and unaffected by crystalline electric fields. Furthermore the cubic environment would suggest anisotropy effects are small. For fields of $\sim$ 2 T the magnetic energy μH associated with the Gd moments is comparable to $k_B T_N$ and therefore only moderate fields are required to establish ferromagnetic alignment of the Gd moments. The Curie-Weiss behaviour of the susceptibility above T_N is consistent with local moment behaviour in which the magnetic phase transition is driven by transverse fluctuations of the Gd moment. Although below T_N in Pd_2GdIn the Gd moments partially polarise the $4d$ Pd band producing an extra $\sim 1\mu_B$ to the observed magnetisation this does not constitute a local moment. Magnetisation measurements show that magnetic short range order above T_N has disappeared by $\sim$ 15 K. Within a local moment model, *e.g.* Heisenberg system, the magnetic contribution to the thermal expansion arises from the thermal variation of the spin-spin correlation function and the dependence of the exchange integral with distance. For an itinerant system amplitude fluctuations are important and lead to an additional degree of freedom which can influence thermal expansion.

4 Magnetostriction

The isothermal longitudinal forced magnetostriction $\frac{\Delta L}{L} = \frac{\ell(H)-\ell(0)}{\ell(0)}$ was measured at several temperatures in fields up to 7 T for each sample. The results for Pd_2GdIn are shown in Figure 5. With the exception of the isotherm corresponding to approximately the Néel temperature, *i.e.* $T = 9.59$ K, all the isotherms are initially negative before having a positive slope. Although the change in magnetostriction is small it is significant with the minimum in the forced expansion occurring for fields $\leq$ 0.5 T. Previous magnetic isotherms [5] were measured in field steps of 0.5 T and so it was not possible to ascertain whether there is an associated change in magnetisation. Therefore the magnetisation was remeasured in the ordered phase using field steps of 0.1 T. Although the isotherm obtained at 2 K and shown in Figure 6 does not appear to have any anomaly, if the data is plotted in the form of an Arrott plot (M^2 *versus* H/M), inset to Figure 6, a change in slope is observed around 0.5 T. Clearly the magnetic transition is not a spin flop but a continuous transformation. The forced magnetostriction observed in Cu_2GdIn as shown in Figure 7 has a different field dependence compared to Pd_2GdIn. Although only the longitudinal magnetostriction has been measured on polycrystalline specimen the differences observed between the two samples are significant. The precise dependence of the volume on applied field depends amongst other criteria on the details of the magnetisation process and may be obtained using the Maxwell equation

$$\left(\frac{\partial V}{\partial H}\right)_{T,P} = -\left(\frac{\partial M}{\partial P}\right)_{T,H} \tag{5}$$

where M is the magnetisation in the field direction. For a simple antiferromagnet $M = \chi V_o H$ and equation (5) becomes

$$\frac{1}{V_o}\left(\frac{\partial V}{\partial H}\right)_{T,P} = -H\left(\frac{\partial \chi}{\partial P}\right)_{H,T}. \tag{6}$$

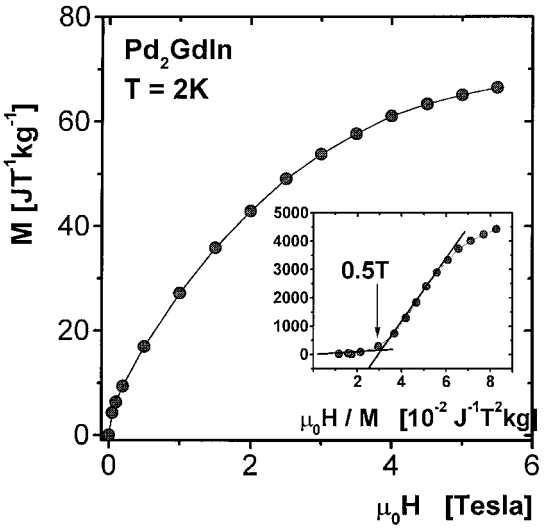

Fig. 6. The magnetisation of Pd_2GdIn as a function of field at 2 K. The inset shows the same data plotted in the form of an Arrott plot M^2 *versus* H/M. For clarity not all the data points have been plotted.

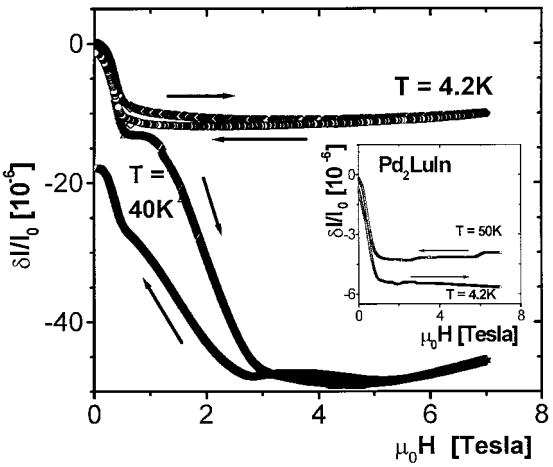

Fig. 7. Longitudinal forced magnetostriction isotherms for Cu_2GdIn. The inset shows the data obtained for the non-magnetic compound Pd_2LuIn.

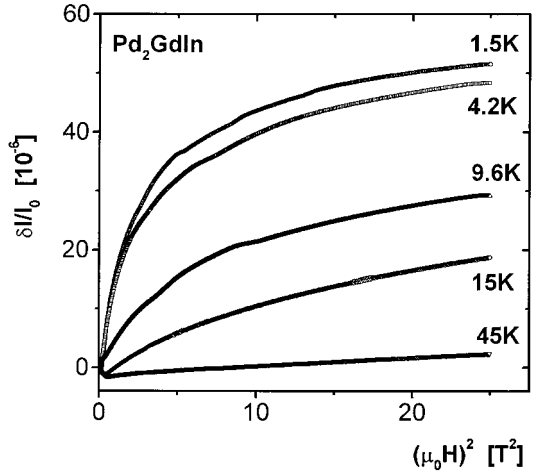

Fig. 8. The magnetostriction of Pd_2GdIn plotted against the square of the applied magnetic field.

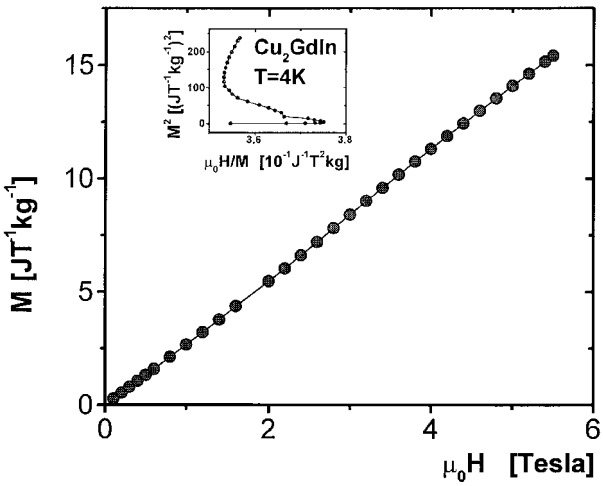

Fig. 9. The magnetisation of Cu_2GdIn as a function of field at 4 K. The inset shows the same data plotted in the form of an Arrott plot M^2 *versus* H/M.

On the application of a field the sublattice magnetisation rotates and for fields greater than H_{sf} becomes canted producing a ferromagnetic component. Eventually for fields $H \geq 2H_E$ the system becomes ferromagnetic. For intermediate fields $H_{sf} < H < 2H_E$ the susceptibility is essentially field independent and equation (6) can be integrated to give

$$\frac{V_H - V_{sf}}{V_o} = -\frac{1}{2}\left(\frac{\partial \chi}{\partial P}\right)\left(H^2 - H_{sf}^2\right). \qquad (7)$$

Thus for fields greater than H_{sf} the volume magnetostriction $\Delta V/V$ varies linearly with H^2. More detailed model calculations [7, 11] also predict that the forced magnetostriction will have a quadratic dependence on the applied field for paramagnetic and antiferromagnetic behaviour. However it may be seen from Figure 8 that this dependence is only approached at high temperatures. Below T_N the forced magnetostriction of Cu_2GdIn (Fig. 7) shows a distinct anomaly at approximately 0.5 T. However the anomaly is not the same as that observed in Pd_2GdIn since it involves a zero change in the magnetostriction as a function of field. The anomaly is more pronounced for increasing field and at 4.2 K a significant hysteresis is observed. Above approximately 3 T there is very little field dependence observed in the magnetostriction. This anomaly coincides with that reported in magnetisation measurements below T_N. More detailed magnetisation measurements undertaken in the magnetically ordered phase are shown in Figure 9. Again the magnetic transitions which are continuous can be seen in the Arrott plot inset to Figure 9. On warming above the Néel temperature the discontinuity in the isotherms which occurs around 3 T disappears with the magnetostriction becoming field independent at approximately the lower anomaly

of 0.5 T. The general variation of the forced magnetostriction with field is similar to that observed for non-magnetic Pd_2LuIn Figure 7.

5 Discussion

The magnitude of the thermal expansion coefficient observed for all three compounds is similar and appears quite "normal". It may be compared with 10^{-3} observed for materials exhibiting giant magnetostriction or with zero for Invar materials. The total volume change between 0 and 300 K again is very close for the three compounds indicating a small magnetic component. This was confirmed by subtracting an electron and phonon contribution from the two Gd compounds, assuming all three components are uncoupled. If the magnetic contributions to the specific heat and thermal expansion are compared for the two magnetic compounds differences are revealed. At T_N the jump ΔC_m in the magnetic specific heat is similar for both compounds, whereas the spontaneous magnetostriction is significantly smaller in Pd_2GdIn. A comparison of the thermal evolution of the magnetic specific heat and thermal expansion reveals that a magnetic Grüneisen relation does not hold. The magnitude of V and B in the Grüneisen expression may be considered similar for both compounds and not to vary significantly over the temperature range considered. Thus the coupling to the lattice is different in both compounds. This difference is highlighted in the results obtained in the forced magnetostriction measurements. Magnetometry measurements reveal that the magnetisation process is different in both magnetic compounds with Pd_2GdIn having a complex structure. Unfortunately the high absorption cross section for thermal neutrons hinders the establishment of the magnetic structure. In the absence of this information it is not possible to comment in detail on the magnetisation process. However if the Gd moments form a triple Q structure similar to that in the isostructural compound Pd_2MnIn this will lead to a negligible magnetostriction [12]. If this is the case in Pd_2GdIn, the forced magnetostriction would arise from the polarisation of the Pd 4*d* band. This would then decrease as a function of temperature as observed. Since the applied field $\mu H \sim k_B T_N$ the forced magnetostriction may be expected to remain finite for large fields above T_N.

The longitudinal magneto resistance $\Delta\rho/\rho$ observed [1] at 5 K in the ordered magnetic phase of Pd_2GdIn is reported to have a variation similar to that expected for a weak itinerant ferromagnet. Since Gd is expected to have a stable moment of fixed amplitude the possible itinerant behaviour could arise from the Pd 4*d* band. The possibility of amplitude fluctuations provides an additional mechanism for thermal expansion. Within the itinerant electron model the thermal expansion arising from the magnetisation process predicts an expansion coefficient below T_c given by

$$\alpha_m = -2/3KCM^2(0,0)/T_c^2 \quad (8)$$

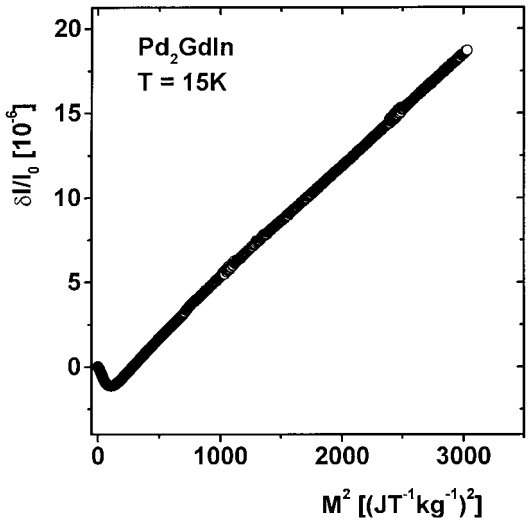

Fig. 10. The forced magnetostriction of Pd_2GdIn at 15 K plotted against the square of the specific magnetisation.

where K is the isothermal compressibility and C is the magneto elastic coupling constant. Above T_c,, α_m is zero since the spontaneous magnetisation in this model becomes zero. However if spin fluctuations are taken into account a positive magneto volume strain is predicted in the paramagnetic state, as a result of the increase in the mean squared amplitude of the spin density fluctuations. There is currently no information with regard to the compressibility of Pd_2GdIn or Cu_2GdIn but the parameters K and C also appear in the expression for the volume magnetostriction

$$\omega_m(H,T) - \omega_m(0,0) = KC\left[M^2(H,T) - M^2(0,0)\right] \quad (9)$$

where ω is the relative volume change and M the magnetisation. Thus combining the magnetostriction and magnetisation data enables KC to be determined, which according to equation (6) is given by the gradient if $\Delta\omega$ is plotted against M^2. For $T \geq T_N$ the isotherms indicating positive magnetostriction are linear with approximately the same gradient; shown in Figure 10 is the isotherm corresponding to 15 K. The isotherms in the magnetically ordered phase are not linear but the "best" straight line fit to these data yields the same gradient. The curvature may arise from the complex nature of the magnetic order and possible differences in the precise field dependence between the magnetisation and expansion measurements. As indicated the gradient yields directly the product KC in equation (9) namely $\sim 7\times10^{-9}$ $(\mathrm{kg\,T/J})^2$ essentially independent of temperature over the measurement range. This value is approximately two orders of magnitude smaller than that observed for weak itinerant ferromagnets [13]. A similar discrepancy is obtained if the derived value of KC is then used in equation (8) to determine α_m. Furthermore within this model the magnetic contribution to the thermal expansion above T_c is predicted to be zero. Clearly the forced magnetostriction remains significant up to at least $6T_N$ and is different in form from that

expected for disordered Gd local moments as exemplified by Cu_2GdIn. The possibility of spin fluctuations associated with 4*d* Pd band requires consideration. Above $\sim 1.5 T_N$ the uniform susceptibility is Curie Weiss with an effective moment slightly larger than that expected for disordered Gd local moments. The effects of spin correlations and in particular amplitude fluctuations on the thermal expansion of weakly and nearly ferromagnetic materials has been considered [14]. In this model the magnetic thermal expansion above the transition temperature is linear in temperature with the coefficient α_m given by

$$\alpha_m = \frac{1}{5} KC \frac{1}{B' C_m} \tag{10}$$

where B' is the coefficient of the fourth order term M^4 in the Landau expansion of the free energy and C_m is the Curie constant. Substituting the appropriate values obtained at 20 K yields $\alpha_m \sim 0.5 \times 10^{-6}$ in reasonable agreement with the results shown in Figure 2. It is therefore tempting to identify the enhanced specific heat with the presence of spin fluctuations associated with the Pd 4*d* band.

6 Conclusions

Thermal expansion and forced magnetostriction measurements have been carried out on two intermetallic compounds containing Gd which order magnetically and one paramagnetic isostructural compound for which Gd has been replaced by Lu. Using a Grüneisen analysis in which the electronic and lattice degrees of freedom are assumed to be uncoupled, the results for the non magnetic Lu compound were used to obtain an estimate of the spontaneous magnetostriction in the two Gd compounds. This analysis revealed that the spontaneous magnetostriction in Pd_2GdIn was significantly less than that obtained for Cu_2GdIn possibly as a result of the Gd moments forming a triple Q structure. On the other hand the magnetic contribution to the specific was found to be similar in both compounds. Significant differences in the magnetoelastic properties between the two compounds were observed in the forced magnetostriction measurements. Analysis of these data suggests that the thermal expansion in Pd_2GdIn is primarily associated with the induced moment on the Pd lattice. Magnetoresistance measurements also suggest the presence of spin fluctuations associated with the 4*d* Pd band. On the basis of these results it is suggested that the enhanced specific heat observed in Pd_2GdIn occurs as a result of these additional degrees of freedom.

The authors would like to thank Bryan Dennis for his technical assistance and the Paul Instrument fund of the Royal Society for its support in the construction of the dilatometer.

References

1. E.V. Sampathkumararan, I. Das, Phys. Rev. B **51**, 8178 (1995).
2. E.V. Sampathkumararan, I. Das, Phys. Rev. B **51**, 8631 (1995).
3. M. Bouvier, P. Lethuillier, D. Schmitt, Phys. Rev. B **43**, 13137 (1991).
4. M.J. Parsons, J. Crangle, K.-U. Neumann, K.R.A. Ziebeck, J. Mag. Magn. Mat. **184**, 184 (1998).
5. K.-U. Neumann, J. Crangle, R.T. Giles, D. Visser, N.K. Zayer, K.R.A. Ziebeck, Solid State Comm. **84**, 577 (1992).
6. K.-U. Neumann, J. Crangle, D. Visser, N.K. Zayer, K.R.A. Ziebeck, Phys. Lett. A **177**, 99 (1993).
7. E. Callen, H.B. Callen, Phys. Rev. **139**, A455 (1965).
8. K. Tajima, Y. Ishikawa, P.J. Webster, M.W. Stringfellow, D. Tocchetti, K.R.A. Ziebeck, J. Phys. Soc. Jpn **43**, 483 (1977).
9. Y. Noda, Y. Ishikawa, J. Phys. Soc. Jpn **40**, 690 (1977).
10. P.J. Webster, K.R.A. Ziebeck, J. Mag. Magn. Mat. **50**, 7 (1985).
11. A. del Morel, M.S.S. Brooks, J. Phys. C **7**, 2540 (1974).
12. J.W. Taylor, K.-U. Neumann, K.R.A. Ziebeck (work in progress).
13. M. Matsunga, Y. Ishikawa, T. Nakajima, J. Phys. Soc. Jpn **51**, 1153 (1982).
14. T. Moriya, K. Usami, Solid State Comm. **34**, 95 (1980).