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Intersublattice exchange coupling in Gd–Mn compounds studied by INS

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Abstract

Inelastic neutron scattering (INS) has been used to study the exchange interaction in polycrystalline samples of $GdMn_2Si_2$, $GdMn_2Ge_2$, $GdMn_6Sn_6$ and Gd_6Mn_{23}. Direct values of the molecular fields experienced by the Gd moments were obtained from the position of the dominant transition at low momentum transfer in the INS spectra. By using a simple spin wave model, also values of the Gd–Mn exchange constants in these compounds were derived. Our results are compared with results from different magnetic measurements.

Keywords: Rare earth transition metal compounds; Gd–Mn compounds; Inelastic neutron scattering; Exchange interaction

1. Introduction

The class of intermetallic R–T compounds (R=rare earth and T=Transition metal) exhibits a wide range of magnetic properties [1]. For a description and explanation of many of these properties the knowledge of the strength of the R–T coupling is important. A well approved method to determine the magnetic R–T coupling strength is inelastic neutron scattering (INS) since it can provide a direct measure of the molecular field experienced by the rare earth spin in the magnetically ordered state [2–5]. In most rare earth compounds the crystal field leads to a complicated level splitting of the rare earth ground multiplet and therefore to a complicated INS spectrum. In the case of Gd compounds, the crystal field effects are negligible due to the non-existence of an orbital moment of the Gd ion and the ground state multiplet is exclusively Zeeman-split [6].

The R–T coupling strength can also be expressed by the R–T exchange constant. This constant can be determined from the INS spectra by using a well established spin wave model [2–4], if the spin of the transition metal is known from other experimental methods.

The molecular fields and the R–T exchange constants in several Gd–Co and Gd–Fe compounds were determined by our group with INS [7,8] and by another group with high-field measurements on free powder particles (HFFP method) [9]. In the present investigation we have concentrated on compounds of Gd with Mn. There is only little information on the intersublattice interaction in these compounds. The reason for this is probably the fact that in most intermetallic R–Mn compounds there is a fairly strong antiferromagnetic Mn intrasublattice interaction. The latter interaction hampers the determination of the intersublattice coupling from measurements of the magnetic ordering temperature or from HFFP measurements [9,10].

The antiferromagnetic Mn–Mn interaction is expected to lead also to a reduced molecular field at the Gd sites and for this reason gives rise to difficulties with INS measurements.

This has limited our choice for INS measurements to the compounds $GdMn_2Si_2$, $GdMn_2Ge_2$, $GdMn_6Sn_6$ and Gd_6Mn_{23} for the following reasons:

$GdMn_2Si_2$, $GdMn_2Ge_2$ and $GdMn_6Sn_6$ belong to the class of the layered RMn_2X_2 and RMn_6X_6 compounds (X=Ge, Si and Sn) which have recently attracted much interest because of the competing influence of intra and interlayer exchange interaction on the magnetic structures [11]. In $GdMn_2Ge_2$ the Mn sublattice orders antiferromagnetically at $T=365$ K and the Gd sublattice orders ferromagnetically at $T=96$ K. The Gd magnetic ordering then forces the Mn sublattice to order also ferromagnetically but in the opposite direction to the Gd magnetization [13]. Although the Mn sublattice order in these layered

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structures depends strongly on the Mn layer distances [10,12], a somewhat similar behavior should be expected for $GdMn_2Si_2$.

Concerning $GdMn_6Sn_6$, it was concluded from magnetic measurements that the Mn–Mn coupling must be very weak and that the Mn moments are not exactly antiparallel but canted [10,12]. Measurements of the magnetic phase diagram of the isotypic $GdMn_6Ge_6$ compound led to a low-temperature phase where ferromagnetically ordered Gd and Mn planes are present [14].

Neutron diffraction on Y_6Mn_{23} revealed an antiferromagnetic order of the Mn sublattice but with strongly different values of the Mn moments [16].

2. Experimental details and results

The samples of the compounds $GdMn_2Si_2$, $GdMn_2Ge_2$, $GdMn_6Sn_6$ and Gd_6Mn_{23} were prepared by arc melting from the starting materials of at least 99.9% purity. After arc melting the samples were wrapped into Ta foil, sealed in evacuated quartz tubes and annealed at 800°C for about 3 weeks. X-ray diffraction was used to confirm that after annealing the samples were single phase, the amount of impurity phases being less than 5%.

The inelastic neutron scattering experiments were performed at temperatures below 20 K on the HET time-of-flight spectrometer at the ISIS spallation source of the Rutherford Appleton laboratories. The powdered samples were mounted in thin-foil Al containers of dimension 4×4 cm^2 at the cold finger of a closed cycle He-refrigerator. The mass of the samples in the neutron experiments was typically 10 g with thicknesses between 1 and 2 mm.

Samples containing natural Gd are less suitable for INS experiments owing to the high absorption coefficient of the Gd nucleus. However, earlier neutron scattering experiments on compounds prepared of naturally occurring Gd had shown that a satisfactory energy resolution combined with sufficient intensity can be reached when using an incident neutron energy of 250 meV [6,7]. This incident energy was also employed in the present experiment.

Results of the INS experiments on the various Gd–Mn compounds are shown in Fig. 1(a)–(d). The spectra presented here are already background corrected. For information on the neutron scattering conditions and on the procedure of the background subtraction, see Refs. [5,6].

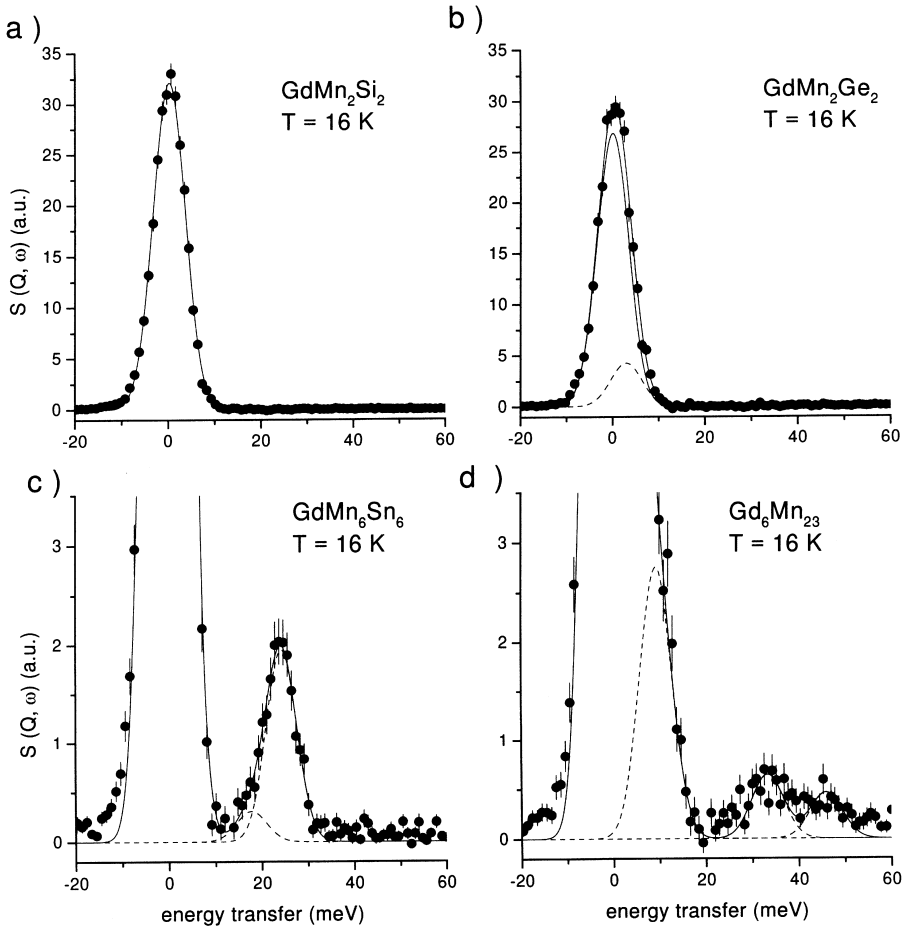

Fig. 1. Inelastic neutron spectra of (a) $GdMn_2Si_2$, (b) $GdMn_2Ge_2$, (c) $GdMn_6Sn_6$ and (d) Gd_6Mn_{23} all measured at $T=16$ K after background subtraction. The incident neutron energy was 250 meV. Solid line: total fit (including elastic line). Individual inelastic contributions are marked by dashed lines.

Table 1
Energy transfer Δ observed in inelastic neutron scattering experiments and the corresponding molecular field B_{mol} at the Gd site derived from the INS data. Z_{GdMn} are the next-nearest Mn neighbors to the Gd ion which, by Eq. (3), leads to the product of the exchange coupling constant J_{GdMn} and the average Mn spin S_{Mn}. Taking S_{Mn} from the references given, we can also determine the exchange constants J_{GdMn}

Compound	Δ (meV)	B_{mol} (T)	Z_{GdMn}	$\lvert J_{GdMn}\, S_{Mn}\rvert$ (meV)	S_{Mn}	J_{GdMn} (meV)
$GdMn_2Si_2$	<3	<26	8	<0.187	-0.75 [15]	<-0.25
$GdMn_2Ge_2$	3 ± 2	26	8	0.187 ± 0.14	-0.85 [16]	-0.22
$GdMn_6Sn_6$	24.4 ± 0.5	211	12	1.02 ± 0.02	-1.1 [10]	-0.93
Gd_6Mn_{23}	9.2 ± 1	79.5	13	0.354 ± 0.04	–	-1.96

The solid lines in the spectra mark the results of a least-squares fit of the background-corrected data points. The dashed lines correspond to inelastic peaks of Gaussian shape with widths approximately equal to the width of the elastic line representing the instrumental resolution (FWHM ~7 meV).

In $GdMn_2Si_2$ and $GdMn_2Ge_2$ (Fig. 1(a) and (b)) no clear inelastic scattering was observed. For $GdMn_2Ge_2$ the width of the elastic peak is slightly broader than the resolution width and we therefore fitted a small inelastic peak as a shoulder to the elastic peak.

In $GdMn_2Si_2$ the width of the elastic peak corresponds nearly exactly to the resolution.

For $GdMn_6Sn_6$ (Fig. 1(c)) one elastic and two inelastic lines were fitted. A weak inelastic peak occurs as a shoulder at the low-energy part of a dominant inelastic peak. In the spectra of Gd_6Mn_{23} (Fig. 1(d)) a dominant inelastic contribution can be clearly observed as a shoulder on the elastic peak. Beside this, some weak inelastic scattering at higher energies is observed and fitted by two more inelastic peaks.

In the following section we will give an interpretation of the INS spectra, concentrating our main interest on the position Δ of the dominant inelastic peak. The experimental results for Δ are listed in Table 1, together with the values deduced for the molecular field and the exchange constants. The additional inelastic scattering in the $GdMn_6Sn_6$ and Gd_6Mn_{23} spectra will be briefly discussed at the end of the next section.

3. Interpretation and discussion

Based on results of our earlier neutron scattering studies made on various types of rare earth intermetallics we have interpreted these dominant inelastic peaks as resulting from the excitation of a dispersionless spin wave mode that corresponds to an out-of-phase precession of the Gd spins in the exchange field of the neighbouring spins [5,7,8].

For the dispersionless mode the following relation holds between the neutron energy transfer and the molecular field B_{mol} experienced by the Gd spins ($g_R=2$ for Gd):

$$\Delta = \mu_B g_R B_{mol} \quad (1)$$

In order to compare the strengths of the intersublattice interactions in compounds of different crystal structure and composition one may use the Gd–Mn exchange constant J_{GdMn} defined by means of an isotropic Heisenberg-type Hamiltonian

$$H_{ex} = -\Sigma 2 J_{GdMn} S_{Gd} S_{Mn} \quad (2)$$

where the summation is over all Mn spins S_{Mn}. Using this Hamiltonian the energies of three spin wave modes (Mn–Mn, Gd–Mn and Gd–Gd) can be calculated within a simple spin wave model [2]. Assuming that only the number Z_{GdMn} of nearest Mn neighbors contribute to the interaction and neglecting the Gd–Gd exchange one obtains for the energy of the flat dispersionless mode

$$\Delta = 2 Z_{GdMn} J_{GdMn} S_{Mn} \quad (3)$$

Without any further assumptions we can deduce from the measured quantity Δ the value of the product $J_{GdMn}\, S_{Mn}$. The results for the molecular field B_{mol} and the product $J_{GdMn}\, S_{Mn}$, both derived directly from the energy transfer Δ, are listed in Table 1. The values of S_{Mn} reported for $GdMn_2Si_2$ [15], $GdMn_2Ge_2$ [16] and $GdMn_6Sn_6$ [10] in Table 1 were derived from results of magnetic measurements. These values have been used in conjunction with Eq. (3) to calculate the corresponding values of J_{GdMn}, which we have also listed in the table.

For Gd_6Mn_{23} the situation is somewhat more complicated. As mentioned above neutron diffraction measurements made on the isotypic compound Y_6Mn_{23} have shown that the Mn moments at the four crystallographic sites 4 b, 24 d, 32 f_1 and 32 f_2 possess the different values $-2.8\ \mu_B$, $-2.1\ \mu_B$, $+1.8\ \mu_B$ and $+1.8\ \mu_B$ [17]. The number of Mn neighbors to a rare earth atom associated with these four sites equals $Z_{RMn}=1$, 4, 4 and 4, respectively. These data have been used in Eq. (3) to calculate J_{GdMn} under the assumption that the situation in the Gd compound is basically the same. In that case one obtains $J_{GdMn}=-1.96$ meV. This value has to be regarded, however, with some reservation because the low value reported for the saturation moment of this compound (38.5 μ_B per formula unit Gd_6Mn_{23}) suggests a canted moment arrangement.

In the following we will compare the intersublattice exchange constants obtained in the course of the present investigation with experimental data obtained by means of other techniques elsewhere and with our former results obtained for Gd–Co and Gd–Fe compounds [7,8].

Results of magnetic measurements made on a single crystal of $GdMn_2Ge_2$ were analysed in terms of the mean field model by Iwata et al. [16]. For the intersublattice exchange constant the authors found a value that corresponds to $J_{GdMn} = -0.32$ meV, which is in good agreement with our INS result listed in Table 1. The susceptibility of $GdMn_2Si_2$ and $GdMn_2Ge_2$ has been investigated by Brabers et al. [18] and explained in terms of a simple model. For $GdMn_2Si_2$ the exchange constant $J_{GdMn} = -0.17$ meV and for $GdMn_2Ge_2$, $J_{GdMn} = -0.34$ meV were found, which is in good agreement with our INS results.

The intersublattice coupling in several compounds of the type RMn_6Sn_6 were studied by Brabers et al. [10] by means of the HFFP method. For the $GdMn_6Sn_6$ compound the applied field was not sufficient to determine the intersublattice coupling. For the $GdMn_3Cr_3Sn_6$ compound diluted with non-magnetic Cr-ions an exchange constant, $J_{GdMn} = -0.76$ was determined. From the results of a series of other $RMn_{6-x}Cr_xSn_6$ compounds (R = Tb and Dy) with $x = 1$, 2 and 3 it was concluded that the exchange constant J_{GdMn} for the full compound $GdMn_6Sn_6$ should be 0.2–0.3 meV higher than for $GdMn_3Cr_3Sn_6$, which would be in good agreement with our INS result.

Up to now the magnetism in the Gd_6Mn_{23} compound is not understood well and we did not find any published results about the exchange coupling in this compound.

We will now compare the exchange constants J_{GdMn} listed in Table 1 with the intersublattice coupling obtained for the Gd–Co and Gd–Fe compounds [7,8]. The exchange constant found for $GdMn_6Sn_6$ is of the same magnitude and has the same sign as in the Gd compounds of Co and Fe of similar 3d concentration, as for instance in Gd_2Fe_{17} and Gd_2Co_{17}.

However, the values of the intersublattice exchange constants found for $GdMn_2Si_2$ and $GdMn_2Ge_2$ are surprisingly low. In fact, such low values have not been observed in Gd compounds of Fe or Co for any 3d concentration. Because of the uncertainty regarding the magnetic structure of Gd_6Mn_{23} we will leave this compound out of consideration.

The tendency of increasing intersublattice coupling with decreasing 3d content, which we observed in the Gd–Co and Gd–Fe compounds can not be confirmed for the Gd–Mn compounds where a decrease of J_{GdMn} with increasing Mn concentration is observed.

Finally, we will discuss the additional weak inelastic scattering which was observed for $GdMn_6Sn_6$ and Gd_6Mn_{23}. As mentioned above the simple spin wave model which was used to calculate the energy of the dispersionless mode leads to three spin modes. But this is a strong simplification since the real number of spin modes is determined by the number of atoms per unit cell. The excitations from all spin modes are measured in the INS spectra. The INS spectra is therefore proportional to the magnon density-of-states. To reproduce in detail the measured spectra one has to know the exact magnon density-of-states for the real, rather complicated crystal structure. This, however, is beyond the scope of the present investigation.

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