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EFFECT OF COMPOSITION ON THE MAGNETIC BEHAVIOUR OF Gd_2X COMPOUND

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Abstract

The eigenvalues of the isotropic magnetic interaction system extraction from Internal Field Approximation (IFA) are computed on the basis of RKKY for the complicated magnetic behaviour of Gd_2X system X= (Au^{+1}, Pt^{+2}, Al^{+3}, In^{+3}) as well as Gd. From the strength and slope of eigenvalue curves in different ranges of interaction the strength and sharpness of the magnetic transition can be suggested. It is shown that the values of the first transition temperature "T_c" as ferromagnetic ordering "F.M", as well as its sharpness in the short-range interaction "R_c", includes at least two atomic layers, this being in agreement with reported experimental results. In the long-range interaction, the second transition is predicted to be of the antiferromagnetic order "AF.M" for Gd_2Al and Gd_2In. It becomes impossible to see the second transition for Gd_2Pt and Gd_2Au as interaction range is increased. For pure metallic "Gd", however, the transition is shown to be of the helicoidal type in the range of "12-17" A°, where all of these results correspond to experimental observations.

Introduction

The magnetic properties of rare earth "RE" metals are mainly due to the localized and partially filled 4f-shell. The magnetic moments are coupled to their neighbours by exchange interaction through the conduction electrons RKKY [1]. The effect of the crystal field on the transition temperature and magnetic moment in gadolinium compounds can be neglected to a first approximation, as "Gd" is in the S-state. So the only factor that determines the magnetic properties is indirect exchange term "J_{ij}^{RR}".

Much effort has been devoted in recent years to trying to understand the cause of competing exchange [2] or random anisotropy [3] where double magnetic transition or unstable phase exists, (especially in Gd_2X intermetallic systems). To our understanding, the difference in magnetic behaviour of the four complicated Gd_2X systems, that have been studied experimentally since 1975 [9-16], are due to:

(i) Effect of conduction-electron concentration "C.E.C" on the range of interaction, both from mean free-path and the high spin polarization at low temperatures.

(ii) Topological composition of magnetic ions as the crystal structure of Gd_2Au, Gd_2Pt, Gd_2Al is the same, as illustrated in Tables 1 and 2 [4-8]. The contradiction in the magnetic behaviour of these compounds from the various experimental reports is also illustrated in Table 3. The isostructure of (X= Au, Al, Pt) compound and the position of magnetic ion suggest that the topological position site should be the main cause, which is due to "C.E.C".

Keywords: Double magnetic transition compound

Table 1. Crystal structure data of Gd_2X compound and Gd

Compound	Lattice	a(Å)	b(Å)	c(Å)	N	Ref
Gd	hep	3.6315		5.777	2	4
Gd_2In	dhep	5.413			4	5
Gd_2Al	orthorhombic(Pnma)	6.606	5.146	9.53	8	6
Gd_2Au	orthorhombic(Pnma)	7.116	4.962	8.998	8	7
Gd_2Pt	orthorhombic(Pnma)	7.186	4.813	8.854	8	8

Table 2. Composition data of orthorhombic systems

Compound	Type of Gd	Position	X	Y	Z	Ref
Gd_2Al	Gd(I)	4(c)	0.83	0.25	0.07	6
	Gd(II)	4(c)	0.96	0.25	0.075	
Gd_2Au	Gd(I)	4(c)	0.845	0.25	0.0671	7
	Gd(II)	4(c)	0.478	0.25	0.817	
Gd_2Pt	Gd(I)	4(c)	0.8538	0.25	0.081	8
	Gd(II)	4(c)	0.9883	0.25	0.6703	

Table 3. The reported experimental magnetic behaviour of Gd_2X system

Compound	Tc or TN	θP	Ref
Gd	Tc= 293.5		9
Gd_2In		212	10
	T(demag)= 103		10
	Tc= 187		5
	T(demag)= 99.5		5
Gd_2Al		150	11
	Tc= 281	284	12
	TN= 44	170	13
Gd_2Au	Tc= 161	125	13
	TN= 61.6		14
	Tc= 266	260	15
	TN= 59		15
Gd_2Pt	T(or)= 165		16
$Gd_{2-x}La_xAl$	T≤284	$100 \leq \theta_p \leq 290$	17

Model and Calculation

In examining the problem of exchange mechanism (between the localized spins), the only explicit calculation can be applied by the following Hamiltonian,

$$H= \sum_{RR'} \sum_{ij}^{F} J_{ij}^{RR'} S_i^R S_j^{R'} \quad (1)$$

where J_{ij}^{RR} is the effective exchange constant between two spins and F is the number of magnetic ions in the unit cell.

In this work, an investigation into the effects of topological composition on the J_{ij}^{RR} in the different ranges of interaction that are based on the following equation is presented (1).

$$J_{ij}^{RR'} = (9\pi Z^2 J^2(0) / 4E_f) F(2K_f |R_i - R_j|) \quad (2)$$

where the function $F(x)= \frac{Cosx - xSinx}{x^4}$ with $x= 2k_f(R_i - R_j)$ being the characteristic which has to be calculated with the following constant assumption.

1) Since it is effectively dependent on the mean free path "MFP", it is assumed to be "1-2" atomic layers at the high temperature corresponding to short-range interaction. As the temperature decreases to the lowest state, the long-range oscillation makes it necessary to take into consideration a few magnetic ions, i.e. "6 atomic layers".

2) $K_f= (3\pi^2 n/v)^{1/3}$ is the electron effective wavenumber at the Fermi level within the approximation of spherical effective Fermi surface, where $z= \frac{n}{N}$ is the ratio of total conduction electron to the total number of magnetic sites in the unit cell. The spherical approximation is a good approximation in RE-intermetallic compound, particularly for Gd.

3) $E_f= h^2 K_f^2/2m^*$ is the Fermi energy, where m^* is the effective mass of free electrons equal to $3m_0$ (m_0 is the rest mass of electrons which is frequently taken for the rare earth elements [17]).

4) J(0) is the exchange integral between the conduction electron and the localized 4f-orbital electrons. It is supposed to be constant and identical for all the above compounds in the whole ranges of temperature in this paper.

Now by applying calculated J_{ij} to the "IFA" method for a case of isotropic interaction, in which the transition symmetry implies that $J_{ij}^{RR'} = J_{ij}^{R-R'', R-R'}$, the ordering temperature, average spin vector σ_j can be obtained by solution of the following eigenvalue and eigenvector system [18].

$$\sum_{j,R'} J_{ij}^{RR'} \sigma_{j,\mu}^{R} = \lambda \sigma_{i,\mu}^{R'} \text{ for all i, R, } \mu = x,y,z$$

$$\text{where} \quad \sigma_{j,\mu}^{R} = \sigma_{j,\mu}^{0} \exp(ik.R) \quad (3)$$

therefore the problem is reduced to finding the eigenvalue and eigenvector of the following f-dimensional system to get the transition temperature:

$$\sum_{j} \xi_{ij} \sigma_{j,\mu}^{R'} = \lambda(k) \sigma_{i,\mu}^{R'} \text{ for all i, } \mu = x,y,z$$

$$\text{where} \quad \xi_{ij} = \sum_{R'} J_{ij}^{RR'} \exp(ik.R') \quad (4)$$

in which for each value of k there are F eigenvalues of $\lambda(k)$. Since we are considering transition from disordering to the first ordering state, the solution should consider the lowest lying eigenvalue of $\lambda(k)$.

With calculation of $\lambda(k)$ for different lattice directions we will be able to obtain the $\lambda(k)$ according to k in different axes which suggests the following point:

(a) There is an F.M ordering when minimum of $\lambda(k)$ is found for k=(000).

(b) When the minimum of λ is in the first Brillouin zone "BZ", then it denotes the existence of AF.M in the lattice.

(c) The spiral spin ordering causes the minimum of λ in the range of centre to the first "BZ" distance.

(d) Then by using $\lambda_{min}(k)$ extracted from Equation 3 which corresponds to $\mu H_{in}= \frac{3}{2} k_B T_0$ with respect to Equation 1 for Gd-base, we can obtain the transition temperature from the disorder phase to the first order phase from the following equation:

$$T_0 = \frac{S(S+1)\lambda_{min}}{3k_B} \quad (5)$$

(e) It should also be mentioned that the strength of the magnetic phase transition is caused by the slope of $\lambda(k)$ curves.

Results and Discussion

As mentioned above, by knowing the crystal structure parameter, occupation (x,y,z) and counting the magnetic ions in the unit cell (Tables 1,2); firstly we calculate the number of magnetic ions in the different ranges of correlation length for each kind of Gd_I and Gd_{II} (Table 4). It is considered that Gd contributes three-electrons to the conduction band [19], the same applies to Al and In. Pt and Au, however, contribute two and one electron respectively. Secondly, on the basis of the above

calculations and Equations 2 to 4, we can concentrate on the $\lambda(k)$ curves and their minimum value in different correlations for the following cases:

A) Pure Gd

The minimum eigenvalue of $\{\xi_{ij}\}$ has been calculated for the propagation vectors k along three principal reciprocal lattice vector directions <100>, <010>, <001>, in the interaction path of 6 to 18 A°. In the correlation length 6-11 A°, the minimum of $\lambda(k)$ was found for k= (000), which corresponds to an F.M order (Figs. 1a, 1b). But in the range 11-17 A°, the minimum of $\lambda(k)$ is reached in the direction of <010> for k = 0.25q (Fig. 1c) which corresponds to a helicoidal spin structure. Above 18 A°, the helicoidal structure disappears (Fig. 1d). This behaviour and the decrease of λ_{min} as a function of distance up to R= 12 A° are in agreement with experimental results [20], demonstrating a sharpness of phase transition and a change of the easy direction of magnetization from c-axis to a-axis in the temperature range 230-300°K.

B) Gd_2 Au-Gd_2 Pt

The result of calculations for both the short and long range correlation length 4-16 A° is shown in Figures 2 and 3. Both cases are in unaltered magnetic order configuration and values of $\lambda_{min}(k)$ remained on k=(0,0,0) in the F.M state. There is no phase change due to the change in correlation length on the form of $\lambda(k)$. Ferromagnetic order seems to be stable for Gd_2 Pt and Gd_2 Au with assumption made on the free-electron concentration and magnetic composition [21,22].

Functional values of $\lambda_{min}(k)$ with correlation length for Gd_2 Pt as well as the $\lambda(k)$ curve suggest that there should be at least one other phase transition which must appear in shorter correlation length.

C) Gd_2In-Gd_2Al

The calculations of $\lambda(k)$ for both Gd_2 In and Gd_2 Al are shown in Figures 4a and 5a in the correlation length of 5 A° and 6 A°, respectively.

The value of $\lambda_{min}(k)$ was found for k=(0,0,0) for Gd_2In which corresponds to F.M state. For Gd_2Al, however, $\lambda_{min}(k)$ is in the AF.M state at k = 1!?. for Gd_2In the value of $\lambda_{min}(k)$ in the principal directions coincides with $\lambda(k=1)$ in the range of 7-16 which signifies AF.M state (Figs. 5b, 5c, 5d).

An existing strength competition between the values of $\lambda_{min}(k=0)$ and $\lambda_{min}(k=1)$ as well as the slow decreasing

Table 4. Calculated data in the range of interaction and their number of neighbour; where indicated the balance and competition number of neighbour shows the symmetric ordering of the crystal structure network

Distance	Gd_2In		Gd_2Al		Gd_2Au		Gd_2Pt		Gd
(A)	GdI	GdII	GdI	GdII	GdI	GdII	GdI	GdII	Gd
4	8	6	8	8	8	8	8	10	12
5	8	12	10	8	12	12	12	12	12
6	20	24	21	17	22	16	24	18	20
7	40	32	34	40	34	36	36	38	50
8	40	38	45	43	54	50	54	52	56
9	70	80	77	73	85	77	85	77	92
10	106	104	107	105	109	111	106	112	134
11	120	122	125	133	133	139	143	145	158
12	180	176	181	185	176	182	179	185	208
13	216	218	229	221	238	232	250	244	268
14	254	256	286	282	289	283	299	303	334
15	350	328	342	352	345	355	361	372	424
16	392	388	421	419	440	442	450	446	526
17	472	484	514	506	517	511	542	540	604
18	580	574	604	602	610	614	622	620	744

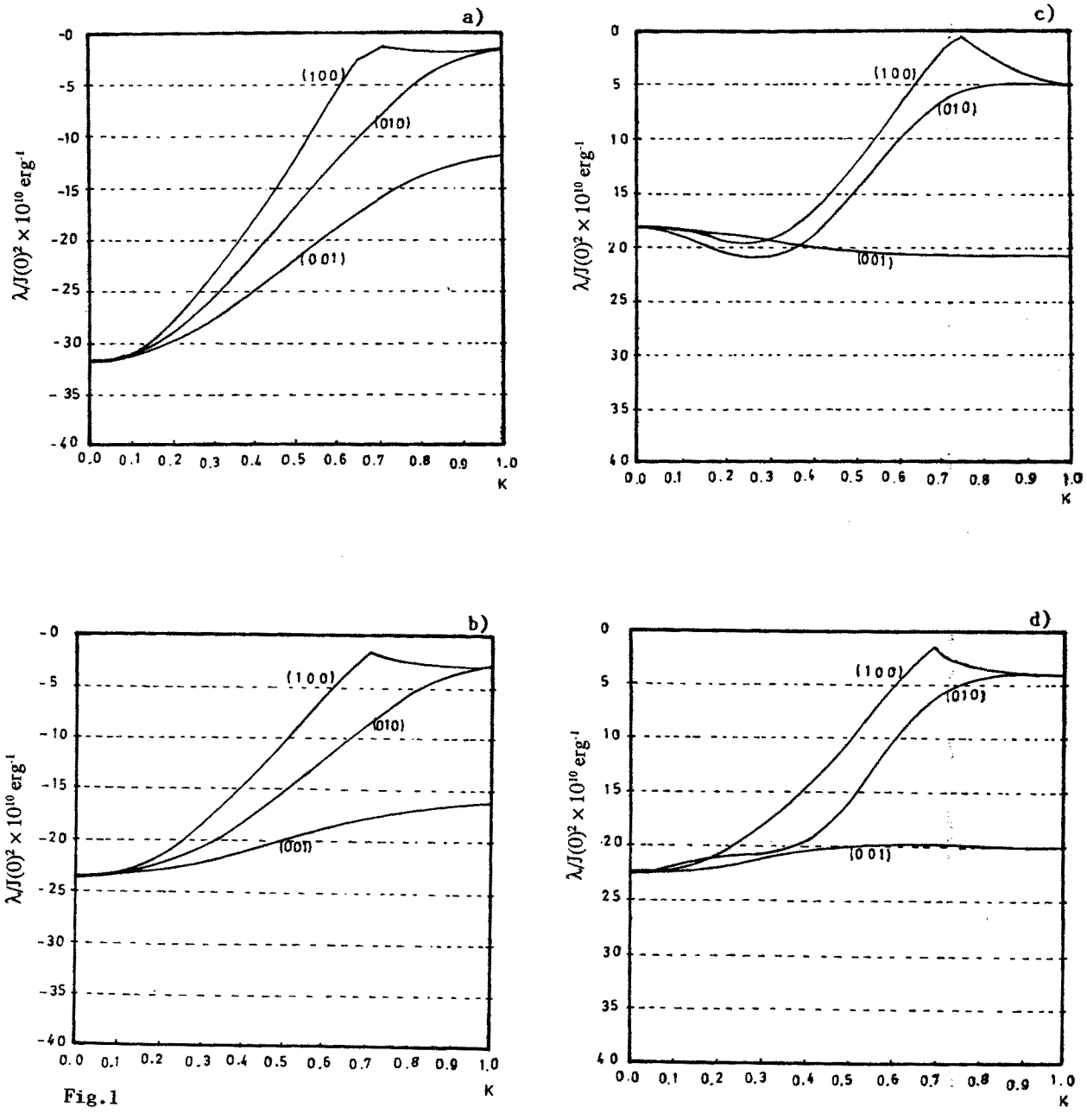

Figure 1. Curves $\lambda_{min}(k)$ for Gd in two principal directions <001> and <010>, interaction range (a) 6 A° (b) 8 A° (c) 12 A° (d) 18 A°

slope of $\lambda(k)$ curves makes Gd_2In a good candidate for percolation problem [23].

For Gd_2Al, as $\lambda_{min}(k=0)$ decreases strongly it remains at a "BZ" only in the principal direction of <100> in the range of 9-16 A° (Figs. 4b,4c,4d). It shows a delicate change of curve from 8 A° to 12 A° at F.M state and from 12 A° to 16 A° a slow-slope and AF.M order in <100> direction [24].

However, from the above-mentioned results of $\lambda(k)$, it can be explained that:

1) The abrupt slope in three principal axes, which is shown in Figures 1a and 5a, suggests the strength of magnetization and sharp F.M phase transition, due to the short range of 1-2 atomic layers. This is similar to

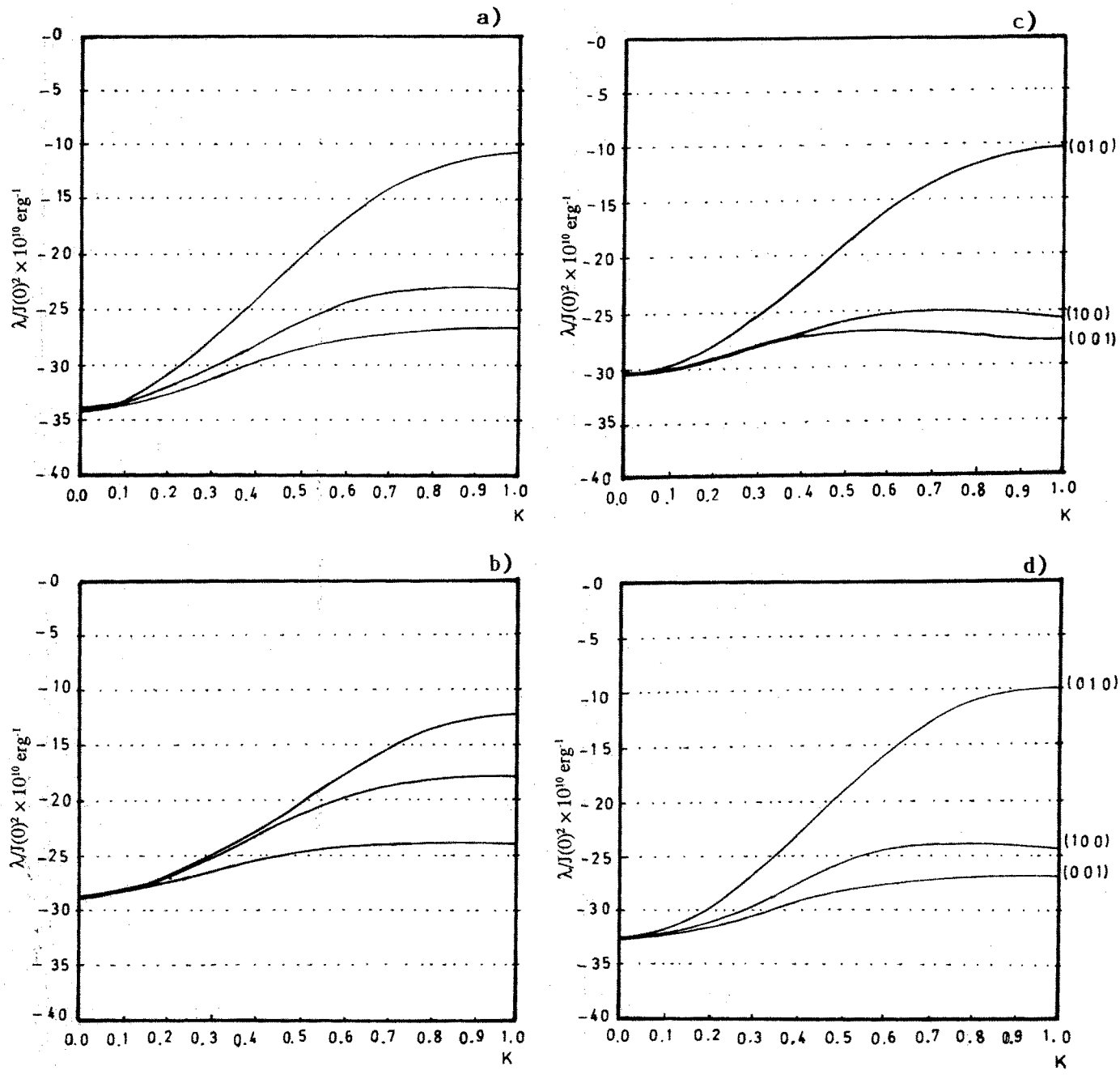

Figure 2. Curves $\lambda_{min}(k)$ for Gd_2Au in three principal directions <100>, <001> and <010>, interaction range (a) 5 A° (b) 8 A° (c) 12 A° (d) 16 A°

experimental results obtained for T_c of Gd and Gd_2In [5,9,10].

2) As the interaction range increases in each case, the slopes of $\lambda(k)$ and its absolute value will also change. In the case of "Gd" the absolute value decreases from 32 to 18 as the strength of interaction varies, on the other hand as the slope of $\lambda(k)$ changes it can be argued that the strength of F.M phase transition decreases.

3) The slope of $\lambda(k)$ for Gd_2 Au shows F.M phase transition only in the <010> direction, and a very slow slope in the <100> and <001> direction as well as an increase in values of λ_{min} with increasing interaction range from 8° to 16 A° which suggests that there is not a sharp F.M transition for this compound, as is shown

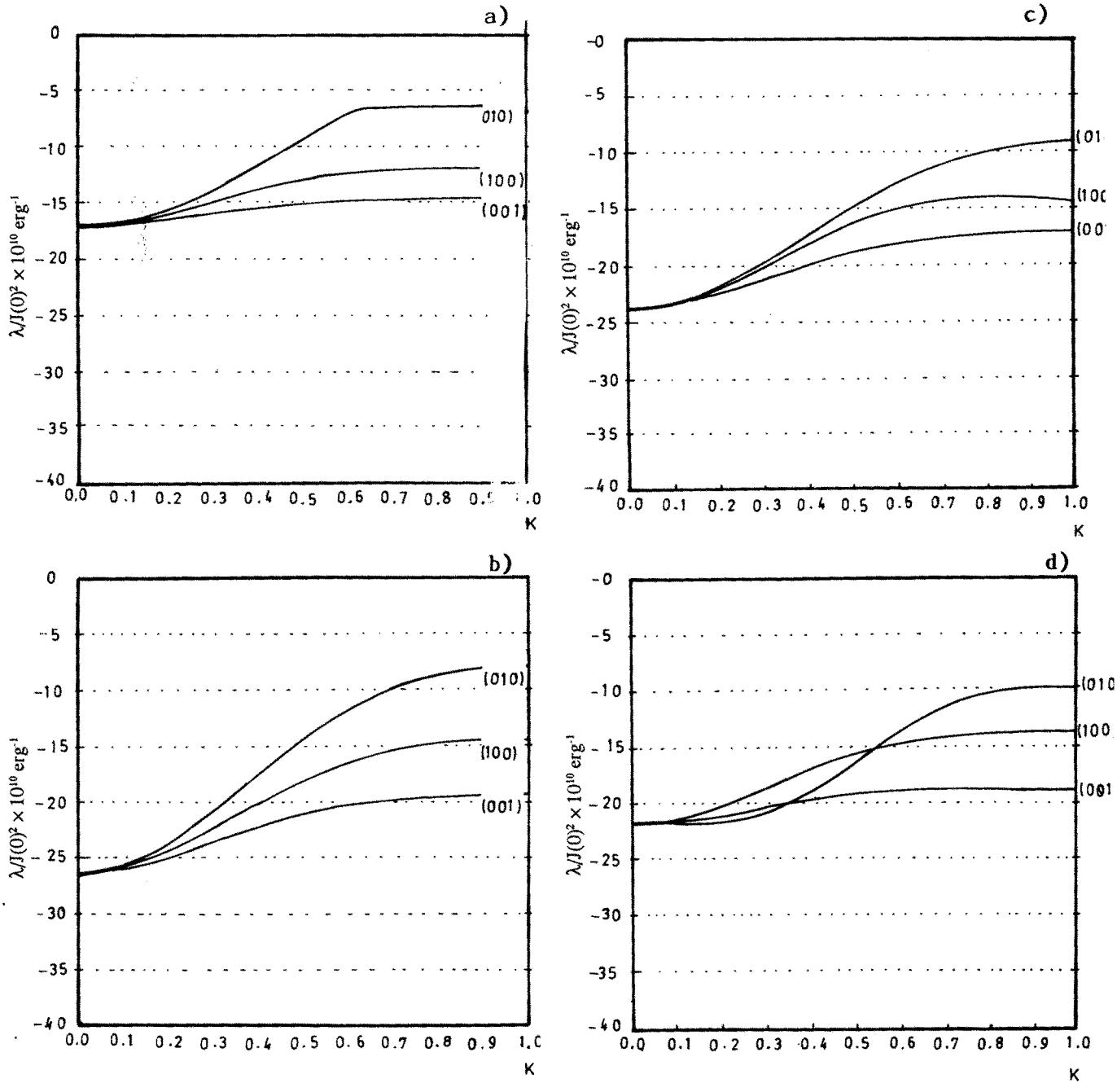

Figure 3. Curves $\lambda_{min}(k)$ for Gd_2Pt in three principal directions <100>, <001> and <010>, interaction range (a) 5 A° (b) 8 A° (c) 12 A° (d) 16 A°

experimentally [13,15].

4) For Gd_2Pt, the $\lambda(k)$ curve shows a delicate behaviour in the short-range interaction (Fig. 3a). As the interaction range is increased (5-8 A°), the absolute value of $\lambda(k)$ increases strongly, and also the slope of $\lambda(k)$ is increased, suggesting a rapid increase in the strength of F.M phase transition.

From the above calculation in the short range interaction "first correlation length" and the absolute constant of $J_{(0)}$= 0.095 eV in the Equations (2 and 5) the calculated values of Curie temperature are given in Table 5. The observed discrepancy between the calculated and experimental results could be attributed to the exponential dependence range to the electron mean free path [25].

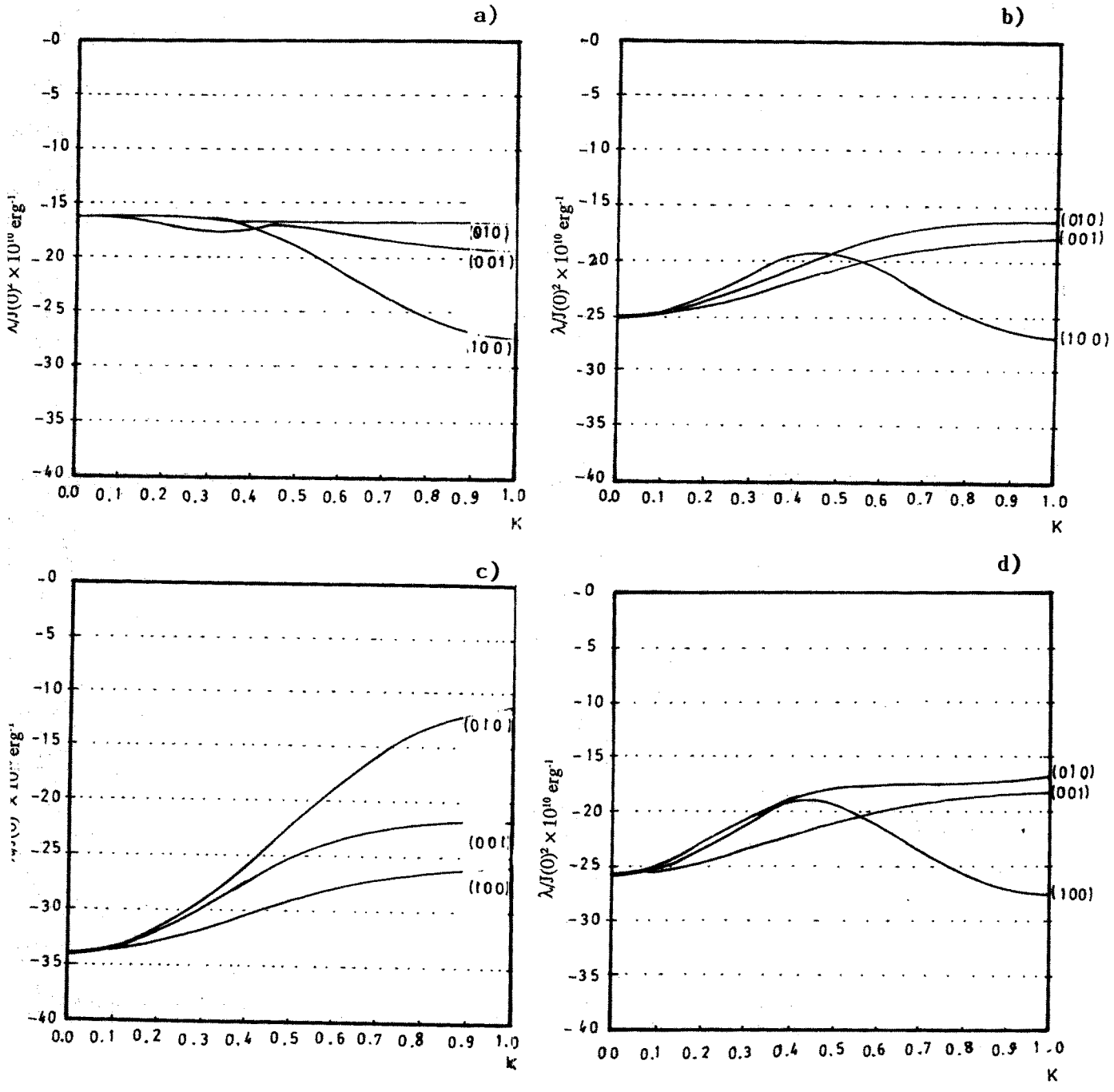

Figure 4. Curves $\lambda_{min}(k)$ for Gd_2Al in three principal directions <100>, <001> and <010>, interaction range (a) 6 A° (b) 9 A° (c) 12 A° (d) 16 A°

It should be mentioned that the selected value of $J_{(0)}$ in short-range interactions for the material under study is reasonable, as it has been found for Gd in various materials [26,27,28]. Also, all the above calculations in three principal networks, in particular in the orthogonal direction, show most of the magnetic structures. For this reason, the rectangular network, which is an orthogonal structure, was selected. For the hexagonal network, the calculations were performed on the a-axis and c-axis which have special importance. As an example the curves of λ(k) in three directions <110>, <011> and <101> are shown in Figures, 6a, 6b, 6c and 6d.

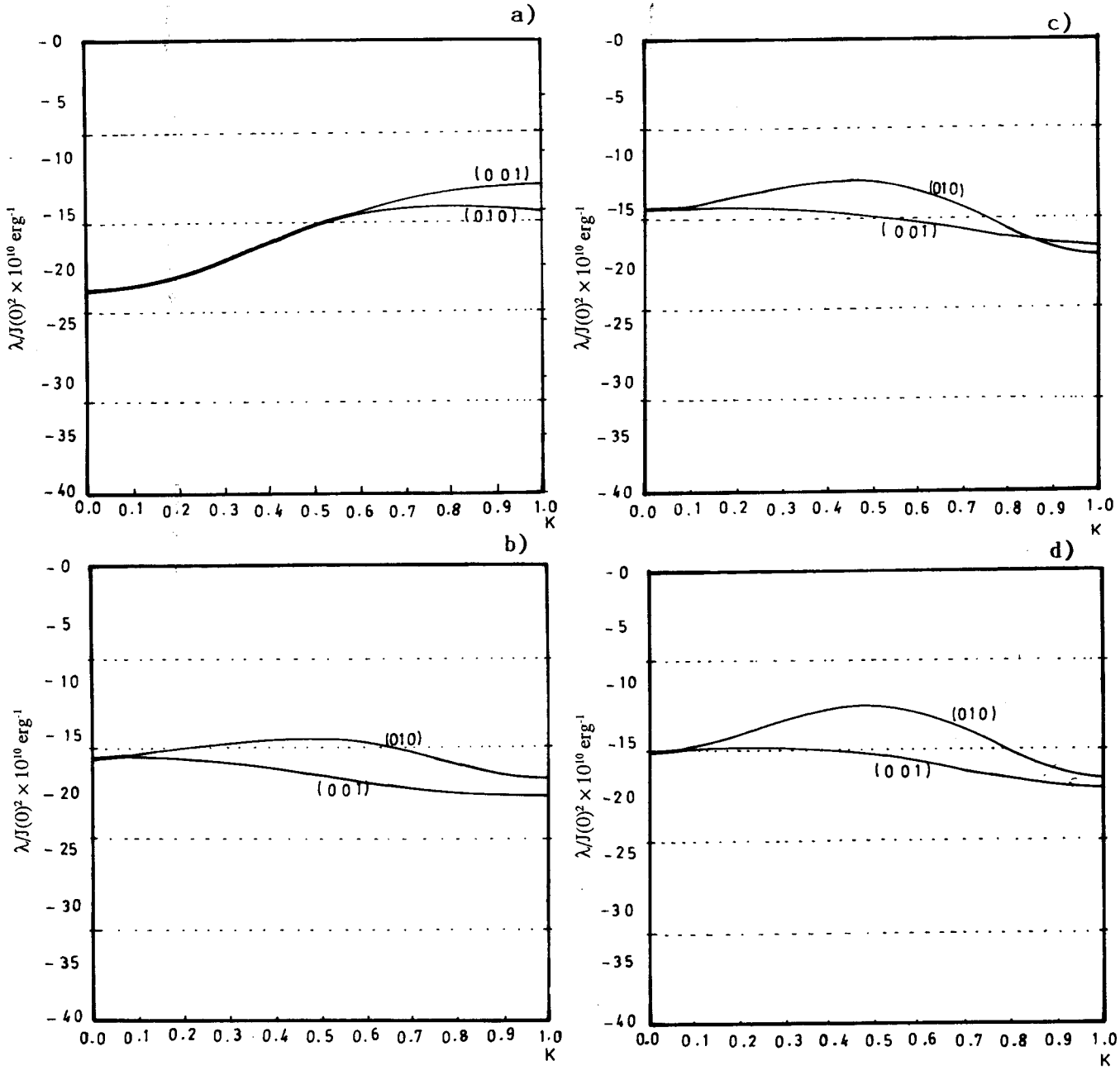

Figure 5. Curves $\lambda_{min}(k)$ for Gd_2In in two principal directions <001> and <010> (or <100>), interaction range (a) 5 A° (b) 8 A° (c) 12 A° (d) 16 A°

Table 5. Calculated data of Fermi surface and eigenvalue of (K) used in obtaining T_0

Compound	Kf(cm-1)	λ_{min}/J(0)2x10e10 erg-1	Tc(ob)(k)	Tc(cal)(k)
Gd	1.39	-32	293.5	282
Gd_2Al	1.49	-34.1	281	300.5
Gd_2In	1.46	-22.3	187	196.5
Gd_2Au	1.37	-24	266	252
Gd_2Pt	1.45	-17.2	165	151.6

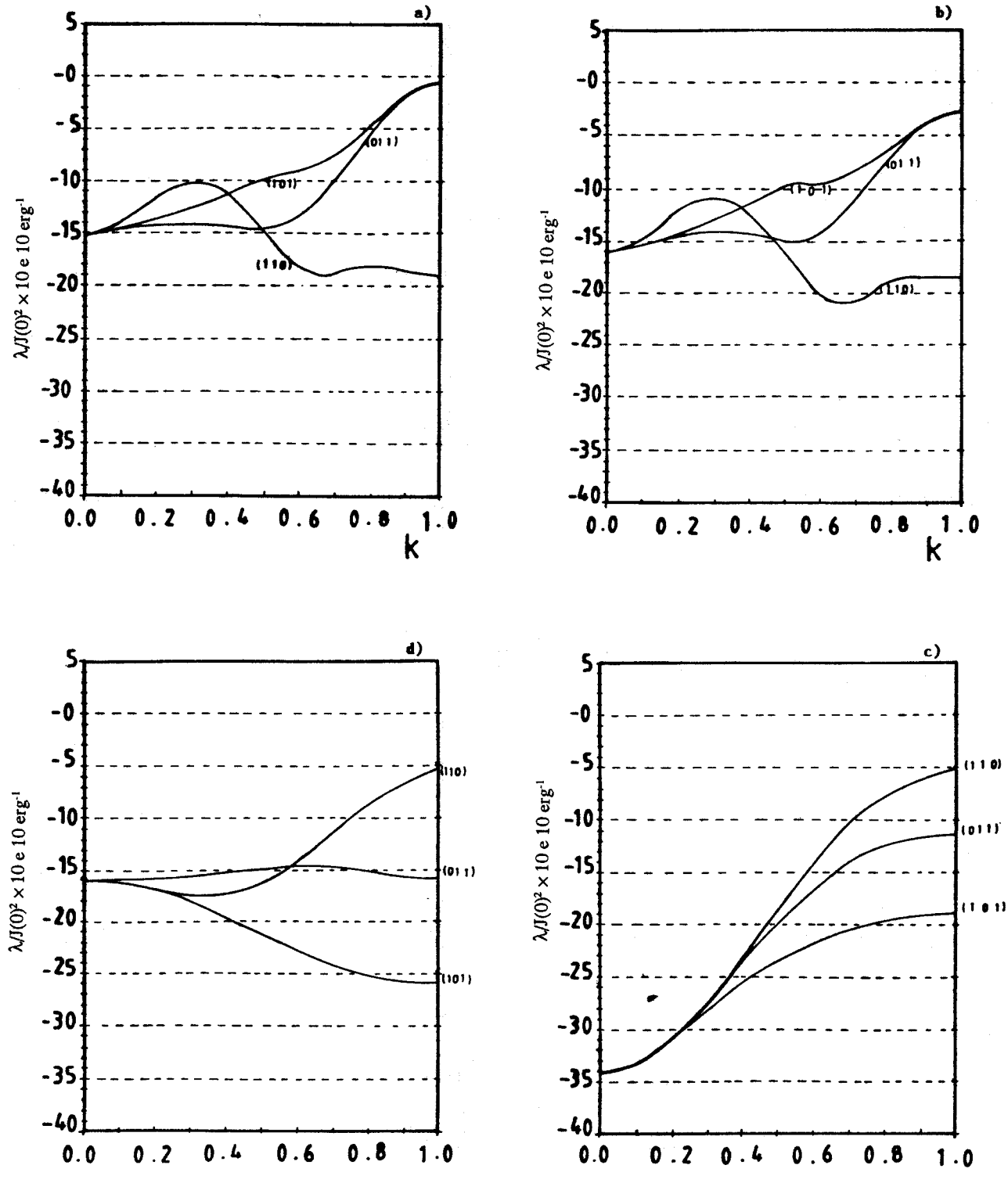

Figure 6. Curves $\lambda_{min}(k)$ in three principal directions <110>, <101> and <011> for (a) Gd_2In, interaction range 12 A° (b) Gd_2In, interaction range 16 A° (c) Gd_2Al, interaction range 6 A° (d) Gd_2Al interaction range 9 A°

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