PHYSICAL REVIEW B VOLUME 57, NUMBER 1 1 JANUARY 1998-I

Magnetism of randomly distributed *f* atoms in $(U_{1-x}Nd_x)Co_2Ge_2$ solid solutions

El'ad Caspi
Nuclear Research Centre-Negev, P.O. Box 9001, 84190 Beer-Sheva, Israel
and Ben-Gurion University of the Negev, P.O. Box 653, 84105 Beer-Sheva, Israel

Moshe Kuznietz, Hanania Ettedgui, Haim Pinto, and Mordechai Melamud
Nuclear Research Centre-Negev, P.O. Box 9001, 84190 Beer-Sheva, Israel

Hagai Shaked
Ben-Gurion University of the Negev, P.O. Box 653, 84105 Beer-Sheva, Israel

(Received 3 April 1997)

Solid solutions of the $(U_{1-x}Nd_x)Co_2Ge_2$ system ($x=0.25$, 0.50, 0.75) are found by neutron diffraction to have tetragonal $ThCr_2Si_2$-type crystal structure (space group $I4/mmm$), same as their end compounds ($x=0$, 1). In the solid solutions, U and Nd atoms are randomly distributed in the basal planes. The end compounds and the solid solutions are paramagnetic at room temperature. At 12 K the (U,Nd) sublattice has the antiferromagnetic AF-I magnetic structure [$+-+-$ stacking of ferromagnetic (U,Nd) planes along c], with moments parallel to the c axis. It is found that within a ferromagnetic basal plane the U magnetic moments are parallel to the Nd magnetic moments, unlike (U,Tb), where they are antiparallel. This shows that the ''spin charge'' rule for lanthanides extends to U-lanthanide systems. Neutron-diffraction and ac-susceptibility results are compared in order to investigate the character of the magnetic transition in the solid solutions. [S0163-1829(97)03646-1]

I. INTRODUCTION

The ternary compounds AM_2X_2 {$A=$lanthanide (Ln) or U [light actinide (An)]; $M=$Co, Ni, Cu; and $X=$Si or Ge}, crystallize in the body-centered tetragonal $ThCr_2Si_2$-type structure (space group $I4/mmm$) (Fig. 1). Most of these materials order magnetically, below room temperature (RT), with a variety of magnetic structures.[1–3] Generally, only the A sublattice orders magnetically. Although the M atoms do not order magnetically, they have a large effect on the magnetic structures.[1,3] In the past we have studied the magnetic phase diagrams of the $U(M,M')_2X_2$ solid solutions.[4] Recently, while studying the validity of the ''spin charge'' concept[5] in the AM_2X_2 systems, we have been interested in the mixing of two atoms in the A site of the $(A,A')Co_2Ge_2$ systems.

The magnetic interactions in these systems can be discussed using the following Hamiltonian:[5]

$$\mathcal{H}=-\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j, \tag{1}$$

where J_{ij} is the exchange coefficient and $\mathbf{S}_i$ is the total spin of the ith atom. Usually, the angular momentum of the $4f$ shell in the lanthanides is not quenched and $\mathbf{S}_i$ is replaced by the total angular momentum $\mathbf{J}_i$ in Eq. (1). $\mathbf{J}_i$ and $\mathbf{S}_i$ are related,[5]

$$\mathbf{J}_i+\mathbf{S}_i=g_i\mathbf{J}_i, \tag{2}$$

where g_i is the Landé factor of the ith atom. Equation (2) leads to

$$\mathbf{S}_i=(g_i-1)\mathbf{J}_i. \tag{3}$$

Also, the ordered magnetic moment of the ith atom is given by

$$\mathbf{m}_i=-\mu_B g_i\mathbf{J}_i, \tag{4}$$

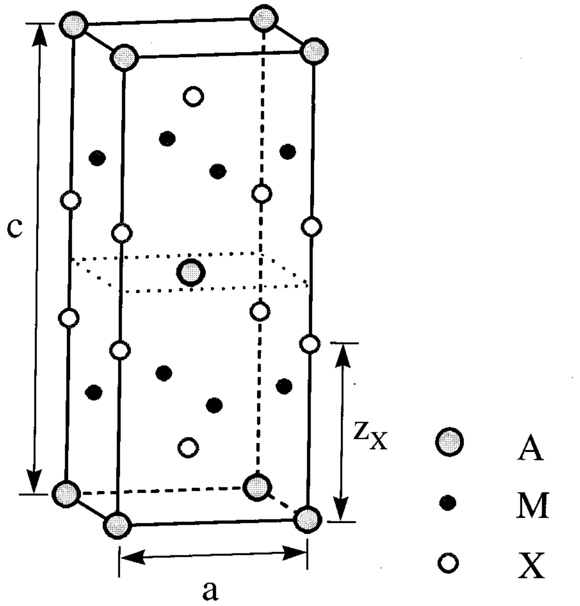

FIG. 1. Crystallographic unit cell of the ternary AM_2X_2 compounds with the atoms A, M, and X occupying the $2a$, $4d$, and $4e$ sites ($z_x\sim\frac{3}{8}$), respectively, in the space group $I4/mmm$ (body-centered tetragonal $ThCr_2Si_2$-type structure). The A atoms [representing the (U,Nd) atoms] delineate the basal planes ($\perp c$).

0163-1829/98/57(1)/449(7)/$15.00

where μ_B is the Bohr magneton.

Substitution of Eq. (3) and Eq. (4) into Eq. (1) yields

$$\mathcal{H} = -\frac{1}{\mu_B^2} \sum_{i,j} J_{ij} \left(\frac{g_i - 1}{g_i} \frac{g_j - 1}{g_j} \mathbf{m}_i \cdot \mathbf{m}_j \right). \tag{5}$$

Since $(g_i - 1)$ can be either positive or negative, a ''spin charge'' [5] can be introduced for each atom. This ''spin charge'' is positive for heavy lanthanides and negative for light ones.[6] For $J_{ij} > 0$, the Hamiltonian in Eq. (5) is minimized for two atoms which have opposite (same) ''spin charge'' signs for antiparallel (parallel) alignment of the magnetic moments.

Previous studies of the ternary compounds, $A\mathrm{Co_2Ge_2}$, show magnetic structures consisting of ''ferromagnetic'' basal planes,[1,3] corresponding to $J_{ij} > 0$ for the intraplane interactions. Thus, it is expected that in the (Ln,Ln′)$\mathrm{Co_2Ge_2}$ systems the ordering of the magnetic moments of (Ln,Ln′) site will obey the following rule: parallel in the case of (light,light), or (heavy,heavy), and antiparallel in the case of (light,heavy). This rule has already been demonstrated in previous studies of (Ln,Ln′) systems.[7,8] Furthermore, it was found that $(\mathrm{U}_{1-x}\mathrm{Tb}_x)\mathrm{Co_2Ge_2}$ a (light-An,heavy-Ln) system orders with the magnetic moments of U antiparallel to Tb,[9] in agreement with this rule. The extension of this rule to (An,Ln) systems is not obvious (see discussion). To consider this extension, one has to show that the (light,light) in an (An,Ln) system also satisfies this rule. This is the motivation of the present study of the magnetic structures in $(\mathrm{U}_{1-x}\mathrm{Nd}_x)\mathrm{Co_2Ge_2}$.

A short summary of the relevant properties of the end compounds of this system $(x = 0,1)$ is given below.

The ternary compounds $\mathrm{UCo_2Ge_2}$ (Ref. 3) and $\mathrm{NdCo_2Ge_2}$ (Ref. 10) crystallize in the body-centered tetragonal $\mathrm{ThCr_2Si_2}$-type, and have similar lattice parameters. Thus, the $(\mathrm{U}_{1-x}\mathrm{Nd}_x)\mathrm{Co_2Ge_2}$ solid solutions are expected to exist throughout the full range $0 < x < 1$.

$\mathrm{UCo_2Ge_2}$ is paramagnetic at RT and was found to undergo an antiferromagnetic transition at $T_N = 175(5)$ K to the AF-I structure.[3] This structure consists of ferromagnetic uranium planes stacked antiferromagnetically along the tetragonal c axis, i.e., with a wave vector $\mathbf{k} = (0,0,1)$. The U magnetic moment was found to be parallel to the c axis with magnitude of $1.9(2)\mu_B$ at 13 K (recalculated using Rietveld profile analysis[11] of the data in Ref. 3).

$\mathrm{NdCo_2Ge_2}$ is paramagnetic at RT and was found to undergo a magnetic transition at 26 K to an incommensurate magnetic structure of the Nd atoms with a wave vector $\mathbf{k} = (0,0,k_z)$ $(k_z = 0.74$ at 12 K).[10] The Nd magnetic moment was found to be parallel to the c axis. This structure was found[10] to coexist below 12 K with the AF-I structure. The amplitude of the incommensurate spin-density wave was found to be $2.1(5)\mu_B$ at $T = 1.5$ K and $4.07(5)\mu_B$ at 12 K (Ref. 10). The magnetic moment corresponding to the AF-I structure was found to be $3.27(5)\mu_B$ at $T = 1.5$ K (Ref. 10).

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $(\mathrm{U}_{1-x}\mathrm{Nd}_x)\mathrm{Co_2Ge_2}$ (with $x = 0.25, 0.50, 0.75, 1$) were prepared by arc melting of the constituents in an argon atmosphere. The obtained buttons were annealed at 1023 K in vacuum for 120 h. Specimens from the samples were examined by x-ray diffraction at 295 K (RT) and were found to be single phase.

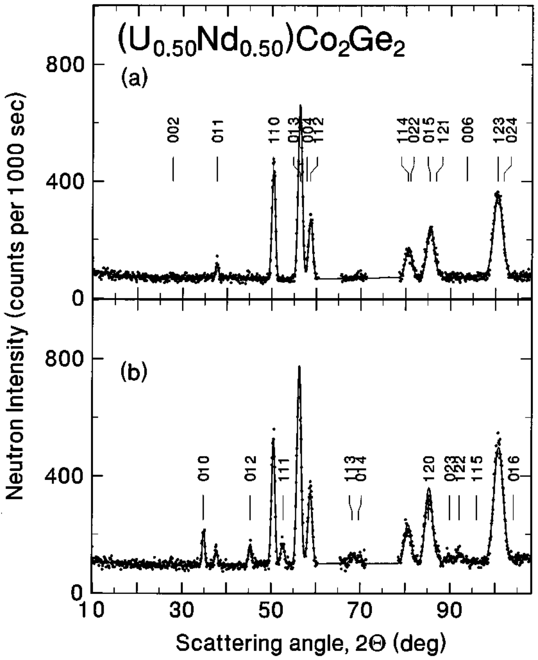

FIG. 2. Neutron $(\lambda = 245$ pm) powder-diffraction data (dots) and Rietveld refinement profile (Ref. 11) (solid line) of the $(\mathrm{U_{0.50}Nd_{0.50}})\mathrm{Co_2Ge_2}$ sample at (a) RT and (b) LT. Crystallographic reflections satisfy the condition $h+k+l=$ even. Magnetic reflections satisfy the condition $h+k+l=$ odd [{010}, {012}, {111}, etc.], consistent with the AF-I structure. Absence of $\{00l\}$ reflections with odd l is consistent with alignment of all the magnetic moments along the tetragonal axis. Observed peaks due to Al sample holder contribution (at ~62° and 74°) were omitted from the analysis.

Neutron- $(\lambda = 245$ pm) diffraction measurements on the four ~20 g $(\mathrm{U}_{1-x}\mathrm{Nd}_x)\mathrm{Co_2Ge_2}$ samples were performed on the KANDI-II diffractometer in the IRR-2 reactor at the Nuclear Research Centre-Negev (NRCN). The sample to be studied was loaded into a cylindrical aluminum container, put into a closed-cycle refrigeration system (DISPLEX, Air Products Inc.). Data were collected at RT and at 12 K (LT). Typical diffractograms at RT and LT are shown (for $x = 0.50$) in Fig. 2. The LT diffractograms show additional reflections, beside those that appear in the RT diffractograms for all samples. The temperature dependence of the integrated intensity of the strongest of these additional reflections was measured by neutron diffraction for each sample (Fig. 3).

Measurements of the ac-magnetic susceptibility were carried out from near RT down to 8 K using liquid He cooling on five small polycrystalline specimens (300–600 mg) (Fig. 3). Four specimens of $(\mathrm{U}_{1-x}\mathrm{Nd}_x)\mathrm{Co_2Ge_2}$ were taken from the present samples, and the fifth specimen (~400 mg) was taken from a sample of $\mathrm{UCo_2Ge_2}$ used in Ref. 3. The amplitude of the applied ac magnetic field $(f \sim 1.5$ kHz) was less than 10 Oe. Calibration of the ac susceptometer was done using a 87 mg powder sample of $\mathrm{Ho_2O_3}$, for which the RT molar susceptibility (χ_M) is 89×10^{-3} emu/mol.

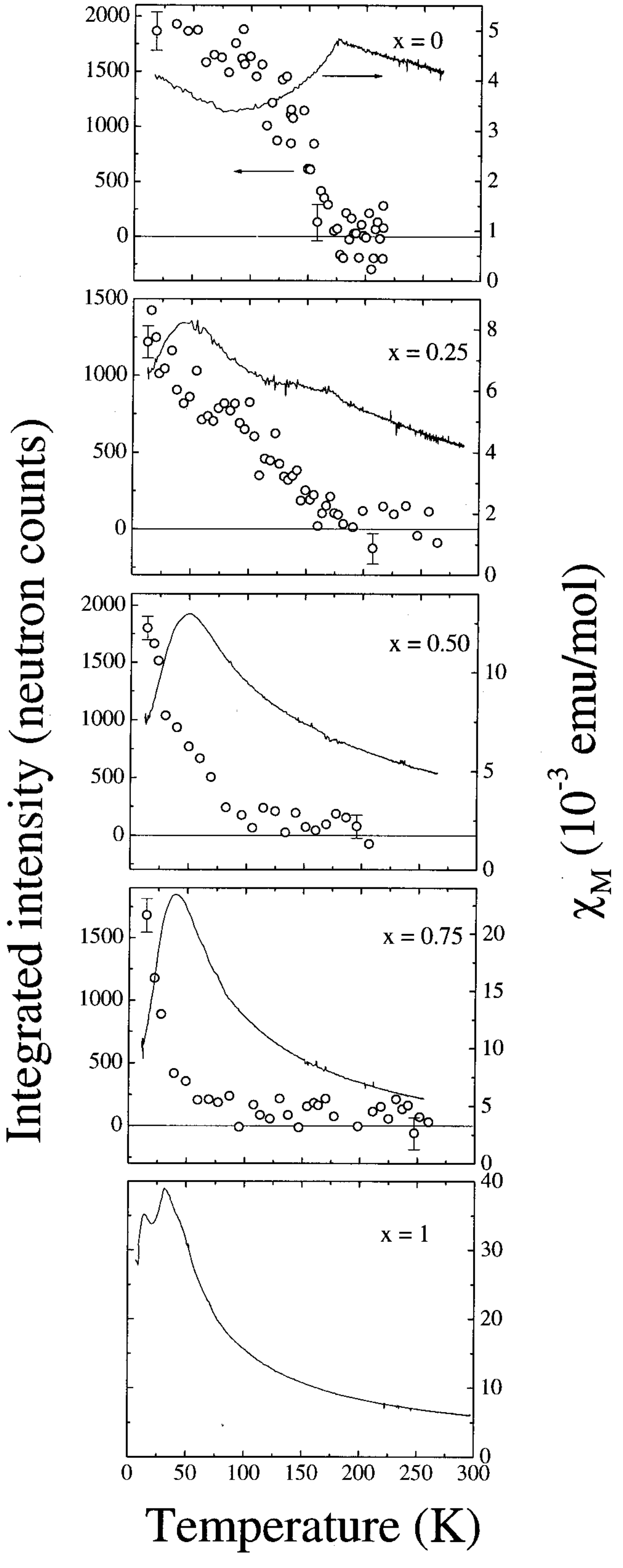

FIG. 3. Observed integrated intensity (after background subtraction) of the magnetic {010} reflections as a function of temperature for the $(U_{1-x}Nd_x)Co_2Ge_2$ compounds (open circles), together with the observed ac-magnetic susceptibility (solid line). Each circle corresponds to a counting time of ~8.5 h for $x=0$, 0.25, and 0.50, and ~3.5 h for $x=0.75$. The neutron data for $x=0$ are taken from Ref. 3. The error bars are representative.

III. RESULTS AND ANALYSIS

The observed reflections in the RT neutron diffractograms of the four $(U_{1-x}Nd_x)Co_2Ge_2$ samples, satisfy the rule $h+k+l=\text{even}$, consistent with the *I*4/*mmm* space group. U and Nd have close neutron-scattering lengths, 8.42 fm and 7.69 fm, respectively. Also, the (U,Nd) site contribution to the stronger diffraction lines, with respect to the contributions of Ge and Co, is small (the latter contributes only to lines with $h+k=\text{even}$). Therefore, the determination of the Nd fraction (x) from the RT neutron diffractograms is not accurate in the present statistics, and the nominal fraction (i.e., the fraction used to prepare the samples) is used in the analysis.

Crystallographic ordering of the U and Nd atoms in the basal planes would require a symmetry reduction, which would result in additional reflections. X-ray diffractograms for several ordered arrangements of U and Nd atoms in the case of $x=0.50$ were calculated using the modified Rietveld profile analysis (the FULLPROF program).[11] The calculated additional reflections due to U and Nd ordering were found to be of the same order of magnitude as the first ({002}) observed reflections (about twice the background). None of these reflections were observed by the x-ray-diffraction measurements. The neutron diffractograms were therefore analyzed, using FULLPROF,[11] with random distribution of U and Nd atoms in the basal planes (2*a* site).

RT structural parameters of $(U_{1-x}Nd_x)Co_2Ge_2$ are found from the RT diffractograms using the analysis described above. As x increases from 0 to 1, the RT lattice parameters a and c, increase by 0.8% and 3%, respectively (Table I; Fig. 4).

In the measurements of the molar ac susceptibility χ_M, vs temperature, the five specimens obey the Curie-Weiss law for $T>200$ K (Fig. 3). The effective magnetic moments μ_{eff}, and the paramagnetic Curie temperatures θ, are obtained (Table II) by fitting the Curie-Weiss relation

$$(\chi_M)^{-1}=(2.83/\mu_{\text{eff}})^2(T-\theta) \tag{6}$$

to the observed data in this temperature range (μ_{eff} given in μ_B, and $T-\theta$ in degrees K). Below this temperature the ac susceptibility goes through a maximum characteristic of magnetic transitions, at temperatures listed in Table II. For the end compounds ($x=0$, 1) these transition temperatures are in agreement with earlier results.[3,10] For $x=0.25$, 0.50, and 0.75, transitions at $T_0^{\text{susc}}\sim 40-50$ K are observed. These peaks are rather broad but their width decreases with increasing x. For $x=0.25$ a second transition is observed at $T_1^{\text{susc}}=160(25)$ K (Fig. 3; Table II).

The LT diffractogram of $NdCo_2Ge_2$ shows additional reflections to those observed at RT. We find that these additional lines are compatible with a commensurate antiferromagnetic structure with a $(+--+-++-)$ stacking of Nd ferromagnetic planes $[\mathbf{k}=(0,0,0.75)]$. The refined Nd magnetic moment in this structure is 3.1(1) μ_B (Table III). Our interpretation is different from the one given in a previous study.[10] A sine modulated magnetic structure with a wave vector $\mathbf{k}=[0,0,0.75(1)]$ and an amplitude $A_{\text{Nd}}=4.0(1)\mu_B$, is also consistent with the neutron diffractogram of $NdCo_2Ge_2$ at 12 K. The lowest magnetic moment, consistent with the sine modulated structure, is obtained with a phase

TABLE I. RT structural parameters of $(U_{1-x}Nd_x)Co_2Ge_2$ obtained by the refinements of the RT neutron diffractograms using the tetragonal space group $I4/mmm$, with the following atoms positions: (U,Nd) at $2a$ (0,0,0), Co at $4d$ $(0,\frac{1}{2},\frac{1}{4})$, and Ge at $4e$ $(0,0,z)$. The neutron-scattering lengths are (in fm) $b_U=8.42$, $b_{Nd}=7.69$, $b_{Co}=2.5$, and $b_{Ge}=8.19$. B is the isotropic thermal displacement parameter. μR is the absorption correction coefficient. Numbers in parentheses represent standard deviation of the last significant digits. The weighted profile (R_{wp}) and expected (R_{exp}) agreement factors are also given.

x	a (pm)	c (pm)	V $(10^{-30}$ m$^3)$	z(Ge)	B $[10^4$ (pm)$^2]$	μR[b]	R_{wp} (%)	R_{exp} (%)
0[a]	400.9(9)	988(2)	158.7(8)	0.375(1)	1.5(8)	0.2(2)	10.10	7.12
0.25	401.6(8)	994(2)	160.4(7)	0.373(1)	0.6(4)	0.7(2)	8.79	6.55
0.50	403.5(8)	1003(2)	163.4(7)	0.371(1)	1.0(3)	1.2(1)	9.56	8.22
0.75	403.2(8)	1009(2)	164.1(7)	0.371(1)	0.1(4)	0.7(2)	9.86	6.79
1	403.9(8)	1017(2)	166.0(7)	0.371(1)	0.8(3)	1.2(2)	9.43	7.29

[a] The results for this sample were obtained by Rietveld profile analysis of data from Ref. 3.

[b] Obtained by LT refinements, assuming close-to-zero value for B in the LT data. The irregularity in μR for $x=0.25$ and 0.50 is due to a higher sample density.

(of the modulation) of $\sim\pi/8$, and is equal to $3.7\mu_B$ (Table III). This value is considerably higher than the ‘‘free-ion’’ value of Nd^{3+} $(3.27\mu_B)$,[6] for which we have no explanation. Hence, we propose that the magnetic structure in $NdCo_2Ge_2$ corresponds to the first solution [i.e., with the $(+--+-++-)$ stacking].

The LT diffractograms of the three $(U_{1-x}Nd_x)Co_2Ge_2$ samples with $x=0.25$, 0.50, and 0.75 (illustrated in Fig. 1 for $x=0.50$) show additional reflections for which $h+k+l=\text{odd}$ (such as {010}, {012}, {111}). These reflections are consistent with ordering of the (U,Nd) sublattice in the AF-I structure. The absence of $\{00l\}$ magnetic reflections with odd l suggests alignment of the ordered magnetic moments parallel to the tetragonal axis. The Rietveld profile analysis of the LT diffractograms (Table III) yields an average moment per (U,Nd) site consistent with a random distribution of the U and Nd atoms in this site.

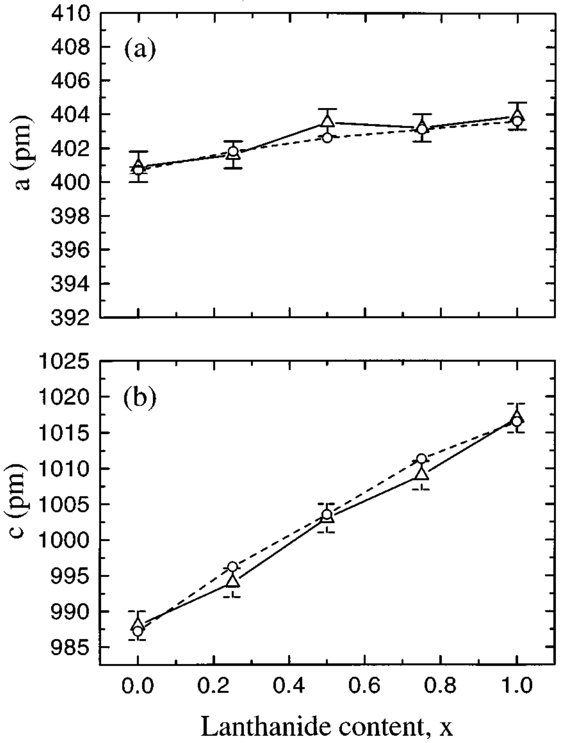

FIG. 4. RT lattice parameters of the $(U_{1-x}Nd_x)Co_2Ge_2$ solid solutions, as a function of x, (a) a, and (b) c. The triangles and circles represent the results of neutron- and x-ray-diffraction measurements, respectively.

The magnetic form factor of the average magnetic moment on the (U,Nd) site, $F(x,\theta_i)$, as used in the Rietveld analysis, is equal to the weighted average of the exponential approximations of the form factors of U and Nd as follows:

$$m_{(U,Nd)}F(x,\theta_i)=|m_U(1-x)\exp\{-C_U[(\sin\theta_i)/\lambda]^2\} \pm m_{Nd}(x)\exp\{-C_{Nd}[(\sin\theta_i)/\lambda]^2\}|, \tag{7}$$

where + (−) designate parallel (antiparallel) alignment of the magnetic moments of U and Nd. The magnetic form factor coefficients of U and Nd in the end compounds are $C_U=4.8\times10^{-20}$ m^2 (Ref. 3), and $C_{Nd}=1.94\times10^{-20}$ m^2 (Ref. 12), respectively. The magnetic moments in the end com-

TABLE II. Magnetic parameters of $(U_{1-x}Nd_x)Co_2Ge_2$ obtained by analysis of the observed ac-magnetic-susceptibility, χ_M. The effective magnetic moment μ_{eff}, and the paramagnetic Curie temperature θ, were obtained by best fitting the observed χ_M vs T to the Curie-Weiss law [Eq. (6)]. T_1^{susc} and T_0^{susc} are the temperatures at which the first and second antiferromagneticlike transitions are observed (Fig. 2). See text for discussion on T_1^{susc} for $x=0.50$ and 0.75.

x	χ_M (RT) $(10^{-3}$ emu/mol)	T_1^{susc} (K)	T_0^{susc} (K)	θ (K)	μ_{eff} (μ_B)
0[a]	4.0(1)	175(5)	-	−331(23)	4.5(2)
0.25	4.1(1)	160(25)	50(10)	−116(3)	3.7(1)
0.50	4.4(1)	[b]	50(10)	−61(4)	3.6(1)
0.75	4.9(1)	[b]	40(10)	−25(2)	3.6(1)
1	5.9(1)	31(3)	12(2)	−49(1)	4.1(1)

[a] The results for this sample are based on data reported in Ref. 3.

[b] Not observed.

TABLE III. Structural and magnetic parameters of $(U_{1-x}Nd_x)Co_2Ge_2$ obtained from the refinements of the LT neutron-diffraction data. Regarding nuclear parameters see Table I. $m_{(U,Nd)}$ is the average ordered magnetic moment on the (U,Nd) site. T_N^{neut} is the para- to antiferromagnetic transition temperature. T_0^{neut} is the lower transition temperature. C is the calculated magnetic form factor coefficient (see text).

x	a (pm)	c (pm)	V (10^{-30} m^3)	z(Ge)	B [10^4(pm)^2]	μR	Magnetic structure	C (10^{-20} m^2)	$m_{(U,Nd)}$ (μ_B)	R_{wp} (%)	R_{exp} (%)	T_N^{neut} (K)	T_0^{neut} (K)
0[a]	400.1(8)	990(2)	158.5(7)	0.375(1)	0.4(4)	0.2(2)	AF-I	4.8	1.9(1)	9.54	7.07	175(5)	-
0.25	400.4(8)	997(2)	159.8(7)	0.373(1)	0.0(3)	0.7(2)	AF-I	3.5	2.3(1)	8.06	6.14	150(10)	40(10)
0.50	401.9(8)	1005(2)	162.3(7)	0.373(1)	0.0(3)	1.2(1)	AF-I	2.9	2.5(1)	9.67	8.27	85(10)	30(10)
0.75	401.8(8)	1010(2)	163.0(7)	0.371(1)	0.1(3)	0.7(2)	AF-I	2.4	2.8(1)	9.08	7.04	70(10)	35(10)
1	402.4(8)	1017(2)	164.8(7)	0.371(1)	0.1(3)	1.2(2)	$(+--+-++-)$[b]	1.94[c]	3.1(1)	10.80	8.01	26[d]	12[d]

[a] The results for this sample are based on data reported in Ref. 3.
[b] Stacking of ferromagnetic Nd planes (see text).
[c] From Ref. 13.
[d] From Ref. 11.

pounds are $m_U = 1.9(2)\mu_B$ (Rietveld analysis of data from Ref. 2), and $m_{Nd} = 3.27\mu_B$ (the ''free-ion'' value of the Nd^{3+} ordered magnetic moment).[6]

A summary of the results of the Rietveld analysis of the LT neutron diffractograms is given in Table III. The average ordered magnetic moment of the (U,Nd) site is found to increase monotonously as a function of x.

The observed ordering temperature T_N^{neut} (i.e., the temperature at which the intensity of the magnetic reflections {010} vanishes, Fig. 3) decreases with the increase of the Nd content (x) in the solid solution (Fig. 3; Table III). A change in slope of the intensity at lower temperatures is interpreted as a second transition for the three solid solutions at $T_0^{neut} \sim 40$ K (Fig. 3). These transition temperatures are similar to those measured by ac susceptibility, T_0^{susc} (Tables II and III).

IV. DISCUSSION

The RT x-ray and neutron-diffraction results described above show, within the accuracy of our data, that the U and Nd atoms are randomly distributed in the $2a$ site of the $ThCr_2Si_2$-type structure, showing a complete miscibility of the end compounds UCo_2Ge_2 and $NdCo_2Ge_2$ in the $(U_{1-x}Nd_x)Co_2Ge_2$ system. The observed lattice parameters are found to depend linearly on x (Fig. 4), in agreement with Vegard's law

$$t_x = (1-x)t_0 + xt_1, \tag{8}$$

[where t_x represents the lattice parameters (a or c) of $(U_{1-x}Nd_x)Co_2Ge_2$] as expected in the case of complete miscibility. This is a special application of Vegard's law, which is usually applied to binary systems, in a pseudoternary system. Moreover, the linear dependence found here is in agreement with the overall linear dependence of the cell parameters on the Ln atomic diameter previously observed for the LnM_2X_2 materials.[13] This linear dependence and the conformity with Vegard's law are in agreement with Pearson's[13] notion of a rigid M-X skeleton in the LnM_2X_2 systems, into which any lanthanide or actinide can fit with some adjustment of the cell parameters. Hence, this notion is valid for one component in LnM_2X_2 (Ref. 13) and two components in $(U,Nd)Co_2Ge_2$ (present work).

The μ_{eff} values, calculated from the ac susceptibilities for $NdCo_2Ge_2$, and for UCo_2Ge_2 (Table II), are found to be higher than the ''free-ion'' values $g\sqrt{J(J+1)})$ for the $4f^3$ electronic configuration of Nd^{3+} (3.62 μ_B),[6] and the $5f^1$ to $5f^4$ electronic configurations of U^{5+} to U^{2+} (2.54–3.62μ_B).[6] Thus, we consider a contribution of Co to the effective magnetic moments according to the equation

$$\mu_{eff} = \sqrt{(1-x)[\mu_{eff}(U)]^2 + x[\mu_{eff}(Ln)]^2 + 2[\mu_{eff}(Co)]^2}. \tag{9}$$

In UCu_2Ge_2, where it can be presumed that there are no copper paramagnetic moments, the effective magnetic moment of U is $2.1(2)\mu_B$.[14] Assuming the above effective magnetic moment for U in the $(U_{1-x}Nd_x)Co_2Ge_2$ system, and the ''free-ion'' value for the Nd moment, we obtain from Eq. (9) the contribution of the Co effective magnetic moments to the observed moments (Table IV). We see that by increasing the Nd content the effective magnetic moment of Co decreases monotonically, but increases again in

TABLE IV. The effective magnetic moment, μ_{eff}, of Co in $(U_{1-x}Ln_x)Co_2Ge_2$ as a function of x, deduced from the observed susceptibility using Eqs. (6) and (9). The value used in this analysis for μ_{eff} of U is $2.1(2)\mu_B$, which is the value of the effective magnetic moment of U in UCu_2Ge_2 (Ref. 14; see text). The values used for Nd and Tb are their trivalent ''free-ion'' values 3.62 and 9.72 μ_B, respectively.

Ln	x	Effective magnetic moment, μ_{eff} (μ_B) Observed	Deduced for Co
Nd	1	4.1(1)	1.3(3)
	0.75	3.6(1)	0.9(4)
	0.50	3.6(1)	1.4(3)
	0.25	3.7(1)	1.9(2)
	0	4.5(2)	2.8(2)
Tb	0.25	5.9(5)	2.0(7)
	0.50	7.3(5)	1.4(14)
	0.75	8.5(5)	0.4(56)
	1	12.0(5)	5.0(6)

$NdCo_2Ge_2$. Making the same assumptions for U and Tb moments in $(U_{1-x}Tb_x)Co_2Ge_2$ (Ref. 15) we obtain similar results (Table IV): the effective magnetic moment of Co decreases monotonically as we add more Tb, but increases again in $TbCo_2Ge_2$.

A comparison between the temperature dependence of the (U,Nd) sublattice magnetization obtained by neutron diffraction and the ac susceptibility (Fig. 3) poses difficulties in the evaluation of transition temperatures. The major maxima in the observed susceptibilities are at lower temperatures (T_0^{susc}) than T_N^{neut} for the solid solutions ($x=0.25$, 0.50, and 0.75) (Fig. 3; Tables II and III). These peaks are broad, in contradiction to the end compounds peaks, which are sharp. For $x=0.25$ a second peak is also observed in the ac susceptibility at approximately the same temperature as T_N^{neut}. Such a peak is not observed for $x=0.50$ and 0.75 (Fig. 3). Also, a change in slope in the neutron intensity vs temperature results can be seen for all three solid solutions at a temperature T_0^{neut}, which approximately corresponds to the low-temperature peak in the ac susceptibility at T_0^{susc} (Tables II and III). A possible explanation to the above observations is that two transitions occur for all three solid solutions, but being smeared out they are hardly separable in the ac susceptibility, and are not clearly seen in the neutron diffractograms because of the low statistics. The character of these transitions is yet to be studied.

Two possible models are suggested here, attempting to explain the data presented above. Both models rely on the idea that the random distribution of two different atoms on the same site may render some clustering and consequently induces a nonuniform magnetic interaction strength. The difference in the full width half maximum (FWHM) between peaks with large l (e.g., {015}), and neighboring peaks (e.g., {114}), is greater in the solid solutions [ΔFWHM $=0.31(6)$] than in the end compounds [ΔFWHM $=0.09(7)$]. This suggests the existence of such clustering in the solid solutions. Both models assume a two-step magnetic ordering process in the solid solutions: (a) Due to the stronger magnetic interaction in U, its moments order at a higher temperature than the Nd moments (e.g., T_N of the end compounds). Thus, the U-rich clusters will order magnetically at higher temperature than the Nd-rich clusters. A similar process has been observed in cases where the two atoms are on different sublattices, both in $ThCr_2Si_2$-type $LnMn_2Ge_2$ systems[16] and in other structures.[17] (b) The in-plane magnetic interactions (mostly direct exchange) are stronger than the intraplane exchange Ruderman-Kittel-Kasuya-Yosida (RKKY-like). As a result, three-dimensional magnetic ordering is established only in the U-rich clusters at a high transition temperature. Full three-dimensional ordering sets in only at a lower temperature, where the intraplane interactions become strong enough.

The Nd ordered magnetic moment is evaluated by best fitting Eq. (10) below to the observed ordered magnetic moments, $m(x)$, on the (U,Nd) site for $x=0$, 0.25, 0.50, and 0.75 [Fig. 5(a)] (see below). Extrapolation of this best fit to $x=1$ results in a Nd magnetic moment of $3.1(2)\mu_B$. This value is in excellent agreement with the Nd moment of the antiferromagnetic structure proposed for $NdCo_2Ge_2$ (Sec. III).

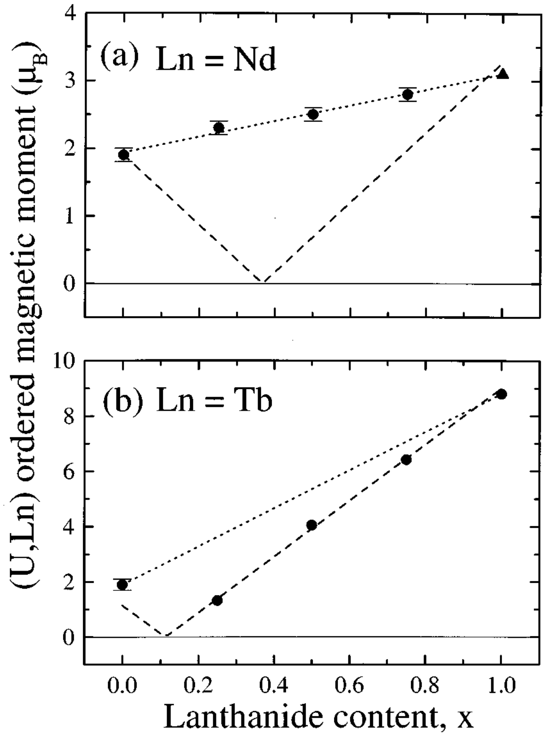

FIG. 5. The observed mean ordered magnetic moment in $(U, Ln)Co_2Ge_2$ (circles) on the (U,Ln) sites for (a) Ln=Nd, and (b) Ln=Tb. The triangle represents the free-ion moment for Nd^{3+}. The experimental error is of the order of the circle size, except where shown. The dashed and dotted lines represent best fits of Eq. (10) to the observed $m_{(\mathrm{U,Ln})}$ for parallel and antiparallel alignments of U and Ln moments, respectively.

The main result of the present work is the confirmation of the "spin charge" model presented in previous works.[7–9] The observed ordered magnetic moments $m(x)$ on the (U, Nd) site (present work) and on the (U,Tb) site,[9] as deduced by Rietveld refinement of the LT neutron diffractograms as a function of x, are found to be distinctly different (Fig. 5). This different x dependence can be modeled as a parallel alignment of m_{U} relative to m_{Nd} and antiparallel alignment of m_{U} relative to m_{Tb} by using the following relation:

$$m(x)^{\pm}=|(1-x)m_{\mathrm{U}}\pm x m_{\mathrm{Ln}}|. \tag{10}$$

A best fit of Eq. (10) to the observed $m(x)$ is achieved with $m(x)^{+}$ and $m(x)^{-}$ for Ln=Nd and Tb, respectively (Fig. 5). This result is in excellent agreement with the "spin charge" concept (see Sec. I) and confirms the validity of the extension of this principle to the (An,Ln) systems. This result implies that U behaves like a light lanthanide in the $ThCr_2Si_2$-type systems, which is unusual for U in other systems. In the actinide series the $5f$ shell is spatially much more extended with respect to the $4f$ shell of the lanthanides series. In the light actinides (e.g., U) the total angular momentum J is usually not a good quantum number due to partial quenching of the orbital angular momentum L. Thus, it is not expected that U will behave like a lanthanide. However, due to the large distances between neighboring atoms in the $ThCr_2Si_2$-type systems, the $5f$ shell is effectively similar to the $4f$ shell (inner and closed). This is in agreement with Pearson's[13] view of the LnM_2X_2 systems, mentioned above. The large ''holes'' in the rigid M-X skeleton accommodate the U atoms, with almost no overlapping of the $5f$ shells. In this case U, a light actinide, will behave like a light lanthanide.

---

[1] H. Pinto, M. Melamud, M. Kuznietz, and H. Shaked, Phys. Rev. B **31**, 508 (1985).

[2] A. Szytuła, in *Handbook of Magnetic Materials*, edited by K. H. J. Buschow (Elsevier, Amsterdam, 1991), Vol. 6, p. 85.

[3] M. Kuznietz, H. Pinto, H. Ettedgui, and M. Melamud, Phys. Rev. B **40**, 7328 (1989).

[4] M. Kuznietz, H. Pinto, and M. Melamud, Physica B **223&224**, 234 (1996).

[5] D. C. Mattis, *The Theory of Magnetism I: Statics and Dynamics* (Springer-Verlag, Berlin, 1981), pp. 232–239.

[6] J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966).

[7] J. R. Cullen, J. P. Teter, and A. E. Clark, IEEE Trans. Magn. **32**, 4779 (1996).

[8] E. Caspi, M. Kuznietz, M. Melamud, H. Pinto, and H. Shaked, Physica B **234-236**, 659 (1997).

[9] M. Kuznietz, H. Pinto, M. Melamud, and K. Tomala, Phys. Rev. B **52**, 15 975 (1995).

[10] G. André, F. Bourée-Vigneron, A. Oleś, and A. Szytuła, J. Magn. Magn. Mater. **86**, 387 (1990).

[11] J. Rodriguez-Carvajal, Physica B **192**, 55 (1993).

[12] H. Pinto and H. Shaked, Phys. Rev. B **7**, 3261 (1973).

[13] W. B. Pearson and P. Villars, J. Less-Common Met. **97**, 119 (1984).

[14] M. Kuznietz, H. Pinto, H. Ettedgui, and M. Melamud, Phys. Rev. B **48**, 3183 (1993).

[15] M. Kuznietz, H. Pinto, and H. Ettedgui, J. Magn. Magn. Mater. **140-144**, 1405 (1995).

[16] G. Venturini, B. Malaman, and E. Ressouche, J. Alloys Compd. **240**, 139 (1996).

[17] H. Shaked, H. Pinto, and I. Felner, in *Modulated Structures*, edited by J. M. Cowley, J. B. Cohen, M. B. Salamon, and B. J. Wuensch, AIP Conf. Proc. No. **53** (AIP, New York, 1979), p. 295.