

# Magnetic structure and anisotropy of Ga- and Al-substituted $\text{LaCo}_5$ and $\text{YCo}_5$ intermetallics

O. Moze

*Istituto Nazionale per la Fisica della Materia, Dipartimento di Fisica, Università di Parma, Viale delle Scienze, Parma, 43100, Italy*

L. Pareti and A. Paoluzi

*Istituto Maspec del Consiglio Nazionale delle Ricerche, via Chiavari 18/a, Parma, 43100, Italy*

K. H. J. Buschow

*Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstrasse 65 1018 XE Amsterdam, The Netherlands*

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The crystal and magnetic structures of hexagonal compounds  $\text{LaCo}_5$ ,  $\text{LaCo}_4\text{Ga}$ , and  $\text{YCo}_4\text{Ga}$  (space group  $P_6/mmm$ ) have been investigated by time-of-flight neutron diffraction at 293 K. The Ga atoms are found to preferentially occupy the  $3g$  site. For  $\text{LaCo}_5$ , Co moments at crystallographic sites  $2c$  and  $3g$  of  $1.60\mu_B$  and  $1.76\mu_B$  have been refined and these moments decrease substantially with substitution of one Ga atom for Co. The magnetic anisotropy field has been measured for  $\text{LaCo}_5$ ,  $\text{LaCo}_4\text{Ga}$ ,  $\text{YCo}_4\text{Ga}$ , and  $\text{LaCo}_4\text{Al}$  on oriented powders using the singular point detection technique. The Co sublattice displays an axial anisotropy for all compounds at temperatures from 77 to 293 K. The anisotropy field is decreased by up to 50% with substitution of one Co atom by Ga (Al). The Co moments for  $\text{LaCo}_5$  are in agreement with results of band structure computations. The single-site approximation for the Co sublattice anisotropy is found to be not generally applicable.

## I. INTRODUCTION

Permanent magnet materials with the  $\text{CaCu}_5$  structure have been a source of intense study for many years now for both technological and fundamental reasons.<sup>1,2</sup> This is highlighted in the material  $\text{YCo}_5$ ,<sup>3-5</sup> which is an ideal permanent magnetic material because of an extremely high uniaxial magnetocrystalline anisotropy and a high Curie point. Materials with this structure are still being investigated for possible synthesis of compounds which can present even better permanent magnet performance parameters. The crystal structure is extremely simple (Fig. 1) and comprises Cu atoms at  $2c$  sites in the same plane as Ca ( $1a$  site) atoms and layers of Cu ( $3g$  site) atoms in between the layers containing Ca and Cu atoms. It has been demonstrated by <sup>155</sup>Gd Möss-

bauer spectroscopy and band structure computations<sup>6</sup> that the electric field gradient at the rare-earth site in such materials can reach high values when  $d$  and  $sp$  atoms show a strong preferential site occupation in the  $\text{CaCu}_5$  structure. A precise knowledge of such site occupancies in ternary compounds is, in general, useful for preparation of intermetallics with high field gradients and hence a high magnetocrystalline anisotropy. A resultant uniaxial anisotropy which is rather substantial also stems from the Co sublattice, as demonstrated in  $\text{YCo}_5$  and  $\text{LaCo}_5$  compounds<sup>7,8</sup> and sophisticated spin-polarized band structure calculations show that this most probably arises from a significant orbital magnetization at Co sites.<sup>9,10</sup>

In order to investigate the effects of preferential site occupancies on the magnetic properties of  $R\text{Co}_5$  compounds, and in particular those with La and Y, we have chosen to investigate a series of Ga- and Al-substituted compounds. An understanding of the mechanism that favors the preferential site occupation in  $R(\text{Co,Ga})_5$  compounds has been obtained from the results of time-of-flight neutron diffraction, which have been compared with model predictions based on size and electronegativity arguments. Furthermore, a meaningful interpretation of the electrical transport, specific heat, and magnetic data can only be given once data for the site occupation of the  $d$  and  $sp$  atoms are available. A well-documented amount of magnetization data for Ga compounds is readily available,<sup>11</sup> but information on dependence of the magnetic anisotropy on Ga substitution is lacking. The site preference of Ga in  $\text{CeCo}_4\text{Ga}$  has been previously investigated by neutron diffraction.<sup>12</sup> Current data obtained on the magnetic moments at  $2c$  and  $3g$  sites for  $\text{LaCo}_5$ ,  $\text{LaCo}_4\text{Ga}$ , and  $\text{YCo}_4\text{Ga}$  are compared with previous polarized neutron measurements, where they exist, as well as with results of band structure calculations.

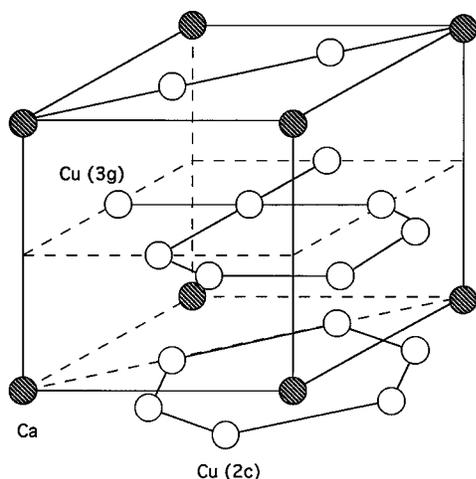


FIG. 1. The hexagonal  $\text{CaCu}_5$  structure.

## II. EXPERIMENTAL DETAILS

The samples used in the present investigation were prepared by arc melting and subsequent annealing at 800 °C for at least three weeks. Sample quality and homogeneity were verified by x-ray powder diffraction and thermomagnetic analysis. The samples were predominantly single-phase material as the diffraction lines showed the characteristics of the hexagonal CaCu<sub>5</sub> structure. Thermomagnetic analysis, however, very clearly revealed the presence of impurity phases. These were found to consist of cubic La(Co,Ga)<sub>13</sub> (NaZn<sub>13</sub> structure) with a corresponding Curie point of 672 °C for the La compounds while the Y compound had impurity phases which consisted of hexagonal Y<sub>5</sub>(Co,Ga)<sub>19</sub> (Curie point = 572 °C) and rhombohedral Y<sub>2</sub>(Co,Ga)<sub>17</sub> (Curie point = 684 °C). The total contribution of the impurity phases was estimated at less than 2% on the basis of the relative peak intensities from the major and impurity phases.

Neutron powder diffraction measurements were performed on the POLARIS high-intensity powder diffractometer at the ISIS neutron spallation source, Didcot, U.K. Data were collected in two different configurations, one with a detector bank in backscattering  $2\theta = 145^\circ$  and the other in forward scattering,  $2\theta = 35^\circ$ . The instrumental resolution  $\Delta d/d = 0.004$  ( $d$  is the lattice  $d$  spacing) is constant for the backscattering bank as it also is for the forward-scattering bank, but the resolution for the forward-scattering bank is a factor of 2 smaller. The conversion from neutron time of flight (TOF), expressed in microseconds, to  $d$  spacing (in Å) is  $\text{TOF} = 505.55568dL\sin\theta$ , where  $L$  is the total neutron flight path. For the POLARIS diffractometer, the values of  $L$  are 12.7981 and 14.2042 m for the backscattering and forward-scattering detector banks, respectively. Diffraction data for both compounds were collected at 293 K. The data were normalized and corrected for absorption prior to use for refinement. Data collected in the backscattering mode allow for a particularly reliable decoupling of preferential occupation and thermal vibration parameters, since small lattice  $d$  spacings are accessed (down to approximately 0.2 Å).

The magnetic anisotropy fields for all compounds were measured from 77 to 293 K on powders aligned in an 8 kOe magnetic field using the singular point detection (SPD) technique.<sup>13</sup> Thermomagnetic analysis was also performed from room temperature to 1100 K.

## III. RESULTS AND ANALYSIS

The diffraction data collected on the diffractometer POLARIS were analyzed by the Rietveld technique,<sup>14</sup> using the program TF104M based on the Cambridge Crystallography Subroutine Library CCLSL,<sup>15</sup> and developed for refinement of both crystal and magnetic structures. The appropriate neutron nuclear scattering lengths employed in the refinement were, respectively,  $b_Y = 0.775 \times 10^{-12}$  cm,  $b_{Co} = 0.249 \times 10^{-12}$  cm,  $b_{Ga} = 0.7288 \times 10^{-12}$  cm, and  $b_{La} = 0.824 \times 10^{-12}$  cm. All refinements were carried out in the space group  $P_6/mmm$ . The sections of the diffraction patterns arising from the impurity phases were neglected, since it is highly unrealistic to attempt to refine one or more impurity phases which form a very small fraction (less than 2%) of the total sample volume. The origin of the unit cell was chosen with

Y and La at (0,0,0). Consequently, the Co and Ga atoms occupy sites at the special positions  $2c$  ( $\frac{1}{3}, \frac{2}{3}, 0$ ) and  $3g$  ( $\frac{1}{2}, 0, \frac{1}{2}$ ). Hence the following parameters were refined: a scale factor, peak profile and background function parameters, isotropic thermal vibration parameters for all sites, Co site populations for sites  $2c$  and  $3g$ , and finally Co magnetic moments for these sites. The refinement strategy consisted of first fixing the Co moments on all sites to those obtained from bulk magnetization measurements and refining only the structural parameters for data from the backscattering bank. Data for this bank allowed for refinement of approximately 200 independent and overlapping reflections. The highly anisotropic neutron spin-magnetic moment interaction allows the overall moment direction and magnitude with respect to the unique axis to be determined even for powder materials with a configurational symmetry lower than cubic.<sup>17</sup> For a simple ferromagnet, with a single spin axis, the scattered intensity, for unpolarized neutrons, is proportional to the sum of the squares of the nuclear and magnetic structure factors:

$$I = F_{\text{nuc}}^2 + \langle q_{hkl}^2 \rangle F_{\text{mag}}^2, \quad (1)$$

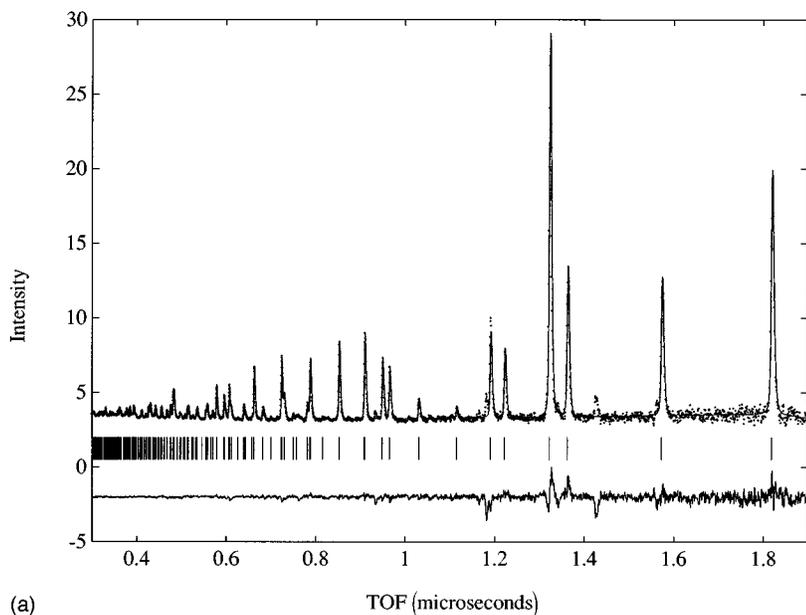
where  $\langle q_{hkl}^2 \rangle$  is the average value of  $\sin^2 \gamma$ , with  $\gamma$  the angle between the moment direction and the scattering vector. This further simplifies to the form

$$\langle q_{hkl}^2 \rangle = 1 - \langle \cos^2 \gamma \rangle \quad (2)$$

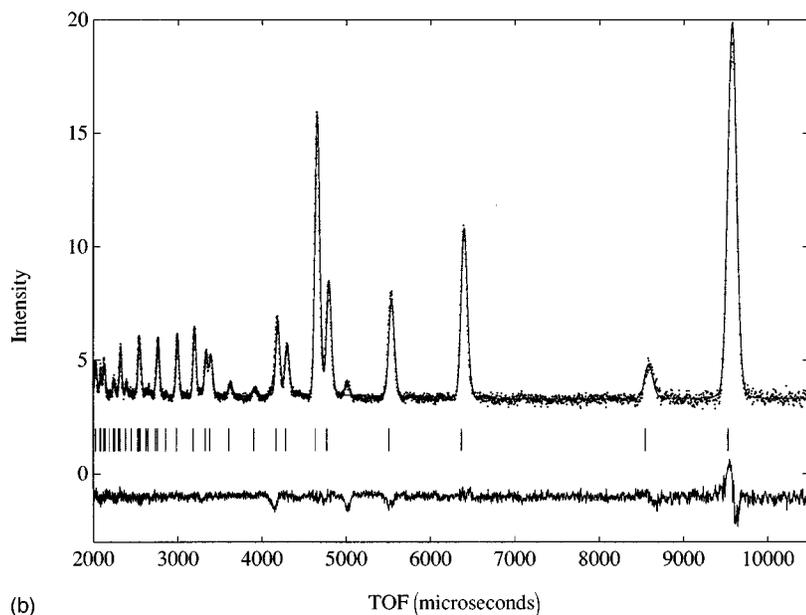
with

$$\langle \cos^2 \gamma \rangle = \left[ \frac{1}{2} (h^2 + hk + k^2) a^{*2} \sin^2 \phi + l^2 c^{*2} \cos^2 \phi \right] d^2, \quad (3)$$

where  $\phi$  is the angle between the moment direction and the  $c$  axis,  $a^*$  and  $c^*$  are reciprocal space lattice parameters, and  $d$  is the lattice spacing for reflection  $(hkl)$ . The Co site moments were refined by fixing all the structural parameters obtained beforehand and subsequently refining only the Co moments at  $2c$  and  $3g$  sites for the forward-scattering bank data. For this data set there are 41 independent and overlapping reflections available, of which about ten have a substantial magnetic contribution, with the first peak, the (100), in particular, being almost totally magnetic in origin. The appropriate magnetic form factor for metallic Co was employed for the refinement. The best agreement was obtained with moments aligned along the  $c$  axis, in proper agreement with bulk magnetization measurements. The Ga atoms were found to show a distinct preference for occupying the  $3g$  site. There is some discrepancy with previously reported site occupancies in Ce compounds. For instance, these data report an almost exclusive occupancy by Co of the  $2c$  site in CeCo<sub>4</sub>Ga.<sup>12</sup> The measurements were carried out at room temperature, well above the magnetic ordering temperature for this compound, and hence it would appear that the crystal structure refinement is highly reliable. The present investigation reports a reduced Co occupancy for  $2c$  sites for both Y and La compounds. This could be due to different annealing treatments for these compounds, but this is considered unlikely, as all compounds were annealed at the same temperature for the same period of time. Another reason for the different behavior for the Ce compound might be that the unit cell has a larger  $c$  axis than in corresponding La and Y



(a)



(b)

FIG. 2. (a) Observed, calculated, and difference neutron time-of-flight diffraction patterns for  $\text{LaCo}_5$  at 293 K. Vertical bars indicate calculated peak positions. Data displayed are for the backscattering bank at  $2\theta = 145^\circ$ . Neutron intensity is in arbitrary units and neutron time of flight is in units of  $10^4$  ms. (b) Observed, calculated, and difference neutron time-of-flight diffraction patterns for  $\text{LaCo}_5$  at 293 K. Vertical bars indicate calculated peak positions. Data displayed are for the forward-scattering bank at  $2\theta = 35^\circ$ . Neutron intensity is in arbitrary units.

compounds, although its  $a$  axis is shorter.<sup>1</sup> In order to check if this discrepancy has any effect on the refined magnetic moments, the Co moments were also refined by also fixing the Co occupancies to those observed for the Ce compound. Only slight differences were noted, all within the experimental errors of the refinement. Refinement quality factors in all three cases were better than 5%. The observed and calculated diffraction patterns for the compounds  $\text{LaCo}_5$ ,  $\text{LaCo}_4\text{Ga}$ , and  $\text{YCo}_4\text{Ga}$  are displayed in Figs. 2, 3, and 4 for both forward- and backscattering banks, while final refined structural and magnetic moment parameters are displayed in Table I.

Magnetization measurements yielded Curie points of 470 and 380 K for  $\text{YCo}_4\text{Ga}$  and  $\text{LaCo}_4\text{Ga}$ , respectively. The temperature dependence of the magnetic anisotropy field is displayed in Fig. 5.

#### IV. DISCUSSION

The gallium site occupation is in agreement with a previous neutron diffraction investigation of the site preference of

$s, p$  elements in several  $\text{CaCu}_5$  compounds of cerium.<sup>12</sup> This site preference was found to be governed mostly by size effects. The lattice parameters of  $\text{LaCo}_5$  and  $\text{YCo}_5$  are seen to increase with Ga substitution, in accordance with the larger atomic radius of Ga.

Data on the Co site moments for  $\text{LaCo}_5$  are extremely scarce or nonexistent, in contrast with accurate polarized neutron data available for  $\text{YCo}_5$ .<sup>18</sup> Heidemann, Richter, and Buschow<sup>19</sup> report on the hyperfine fields at  $2c$  and  $3g$  sites for  $\text{LaCo}_5$  extracted from inelastic neutron scattering measurements and obtain a different site dependence via a scaling constant. The magnetic moments for  $\text{LaCo}_5$  reported here appear to be a more direct measurement of the moments. In the present investigation, the refined Co site moments appear reasonable and are in good agreement with bulk magnetization measurements, in particular with those reported by Bartashevich *et al.*,<sup>10</sup> who report a saturation magnetization of  $8.46\mu_B$  per formula unit at 4.2 K for  $\text{LaCo}_5$ . The present neutron data give a saturation moment of  $8.48\mu_B$  at 293 K.

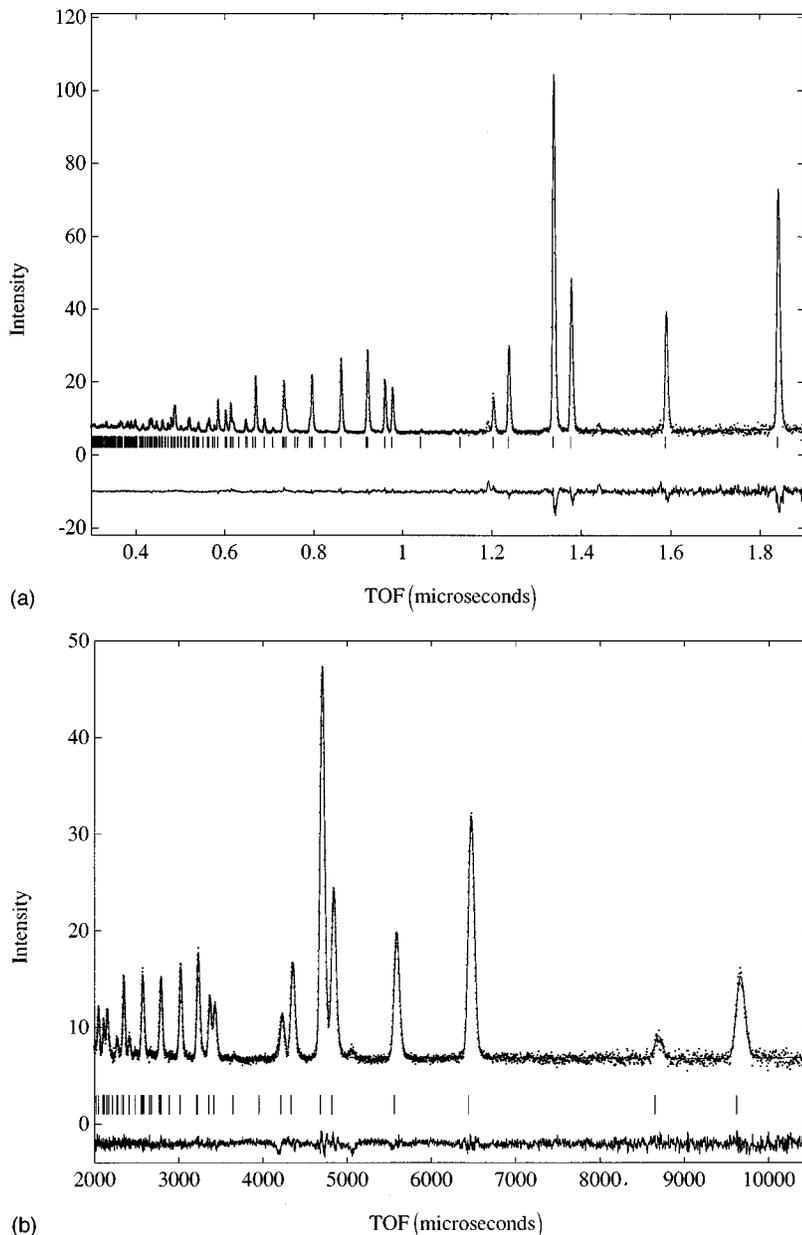


FIG. 3. (a) Observed, calculated, and difference neutron time-of-flight diffraction patterns for  $\text{LaCo}_4\text{Ga}$  at 293 K. Vertical bars indicate calculated peak positions. Data displayed are for the backscattering bank at  $2\theta = 145^\circ$ . Neutron intensity is in arbitrary units and neutron time of flight is in units of  $10^4$  ms. (b) Observed, calculated, and difference neutron time-of-flight diffraction patterns for  $\text{LaCo}_4\text{Ga}$  at 293 K. Vertical bars indicate calculated peak positions. Data displayed are for the forward-scattering bank at  $2\theta = 35^\circ$ . Neutron intensity is in arbitrary units.

Nothing at all, however, can be inferred about the orbital and spin contributions. This is due to the inherent nature of the neutron powder technique. It can, however, be assumed that there is also a consistent orbital contribution which is even perhaps larger than that observed for  $\text{YCo}_5$ , as predicted by band structure calculations. The effects of substitution of even one Ga atom for Co on the magnetic properties are drastic. This is reflected in the observed site moments for both  $\text{YCo}_4\text{Ga}$  and  $\text{LaCo}_4\text{Ga}$  as well as in the behavior of the magnetocrystalline anisotropy. The various band structure calculations<sup>8,9,20–22</sup> do not take into consideration the different site symmetries for the  $2c(6\bar{m}2)$  and  $3g(mmm)$  positions, while calculation of the magnetic anisotropy for these systems is still a formidable task.

The behavior of the magnetocrystalline anisotropy, as reflected by the anisotropy fields plotted for the Ga-substituted compounds in Fig. 5, is in marked contrast to that of B-substituted compounds. Boron is isoelectronic with Ga and Al and it might be expected that approximately the same

behavior would be observed. In fact, the anisotropy field of  $\text{YCo}_4\text{B}$  decreases with decreasing temperature and reaches at room temperature a value of 1.6 T.<sup>25</sup> This is substantially lower than the value of 6.4 T observed here for the compound  $\text{YCo}_4\text{Ga}$  at room temperature. One could argue that the reason for this difference in behavior can be easily explained on the basis of simple considerations of the preferential site entrance. It is well established that magnetic anisotropy for  $\text{YCo}_5$  and  $\text{LaCo}_5$  can be totally ascribed to the Co atoms at  $2c$  and  $3g$  sites. The  $2c$  site contributes positively to the magnetocrystalline anisotropy, favoring an easy  $c$  axis, while the  $3g$  site contributes only modestly or negatively.<sup>25–28</sup> Even if magnetism in  $\text{YCo}_5$  and  $\text{YCo}_5$  has a mostly itinerant character (the main features can be accounted for by existing sophisticated electronic band structure calculations), the  $3d$  anisotropy can be considered as originating from the residual part of the moment localized at the  $3d$  atom, or even from the presence of band states having a well-defined orbital character, the specific mechanism be-

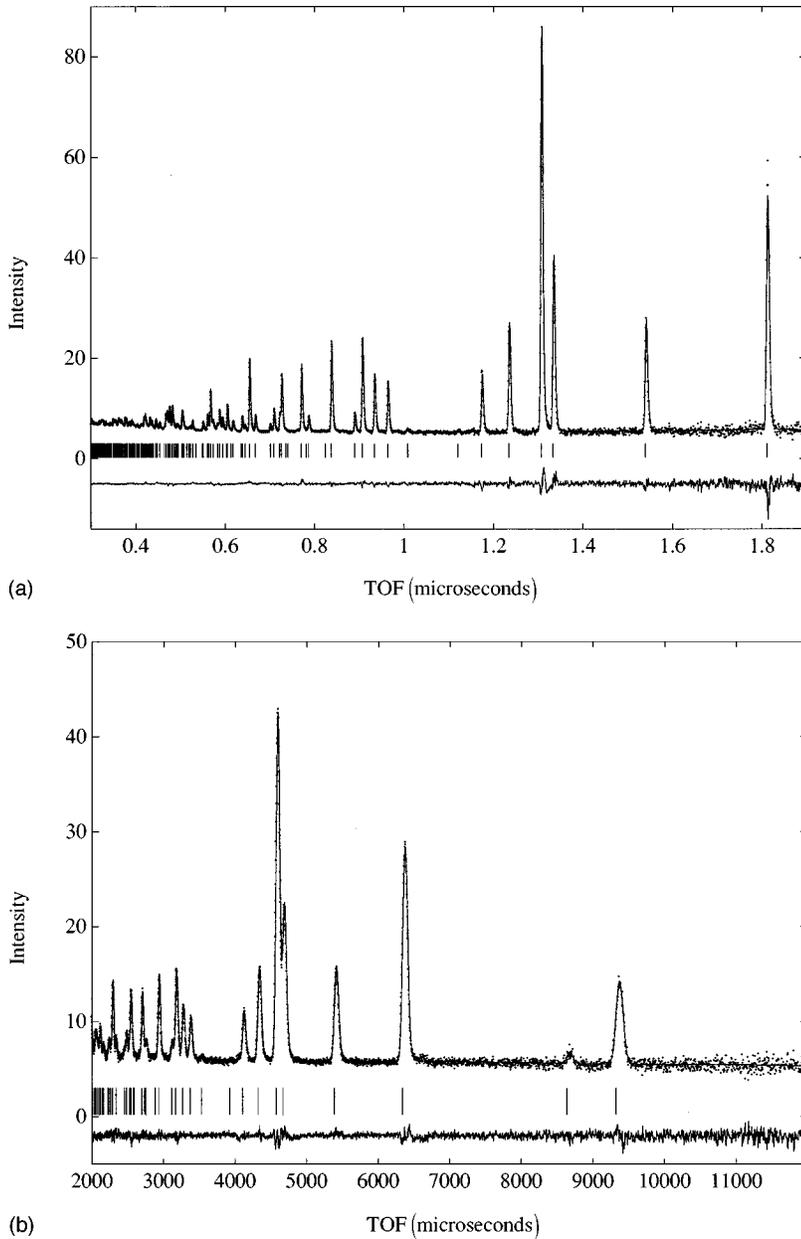


FIG. 4. (a) Observed, calculated, and difference neutron time-of-flight diffraction patterns for  $\text{YCo}_4\text{Ga}$  at 293 K. Vertical bars indicate calculated peak positions. Data displayed are for the backscattering bank at  $2\theta = 145^\circ$ . Neutron intensity is in arbitrary units and neutron time of flight is in units of  $10^4$  ms. (b) Observed, calculated, and difference neutron time-of-flight diffraction patterns for  $\text{YCo}_4\text{Ga}$  at 293 K. Vertical bars indicate calculated peak positions. Data displayed are for the forward-scattering bank at  $2\theta = 35^\circ$ . Neutron intensity is in arbitrary units.

ing the spin-orbit coupling. Thus a localized picture for the Co anisotropy appears to be realistic for a phenomenological description of the composition dependence of the magnetocrystalline anisotropy present in substituted compounds. This is provided that substitution does not significantly modify the “localized”  $3d$  band states.

The behavior of the anisotropy in  $\text{YCo}_{5\pm x}$  has indeed been satisfactorily described in the framework of a localized model.<sup>29</sup> Dumbbell Co sites are present in the case of Co excess. A competition between opposite contributions of Co in the  $2c$  sites (axial anisotropy, positive second-order anisotropy constant  $K_1$ ) and  $3g$  sites (planar anisotropy, negative  $K_1$ ), and a planar anisotropy attributed to the dumbbell sites, with a relative intensity of the contributions in the ratios  $1:-0.4:-2.5$  accounts for the observed variation of the anisotropy with Co excess and defect. The resultant second-order anisotropy constant  $K_1$ /formula unit, applying the model to Ga and Al compounds, can be expressed as

TABLE I. Refined site occupancies and magnetic moments at 293 K.  $N$  refers to the Co site occupation. Lattice parameters and magnetic moments for  $\text{YCo}_5$  have been taken from Refs. 18, 23, and 24.

	Site	$a$ (Å)	$c$ (Å)	$N$	$\mu_z$ ( $\mu_B$ )
LaCo <sub>5</sub>	Co <sub>2c</sub>	5.1085(3)	3.9667(3)	1.00	1.60(2)
	Co <sub>3g</sub>			1.00	1.76(2)
YCo <sub>5</sub>	Co <sub>2c</sub>	4.935	3.964	1.00	1.72
	Co <sub>3g</sub>			1.00	1.77
LaCo <sub>4</sub> Ga	Co <sub>2c</sub>	5.1600(2)	4.0180(3)	0.90(2)	0.6(2)
	Co <sub>3g</sub>			0.78(2)	1.2(2)
YCo <sub>4</sub> Ga	Co <sub>2c</sub>	5.0016(2)	4.0086(3)	0.88(2)	0.9(2)
	Co <sub>3g</sub>			0.75(2)	1.0(2)

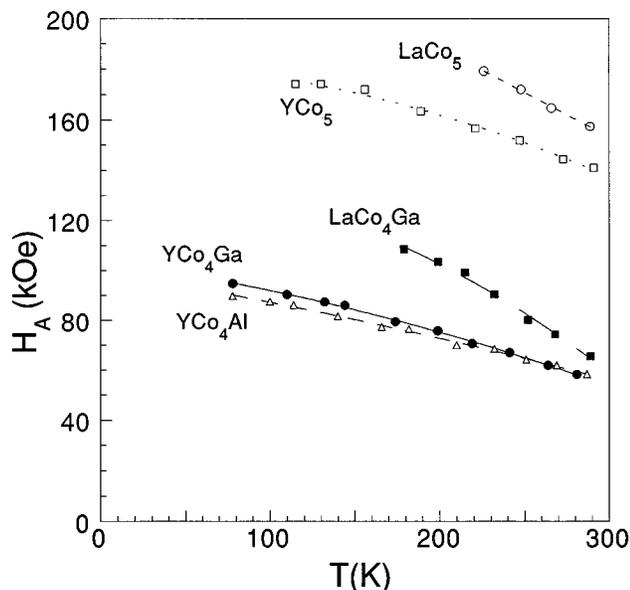


FIG. 5. Temperature dependence of the magnetic anisotropy field for  $\text{LaCo}_5$ ,  $\text{YCo}_5$ ,  $\text{YCo}_4\text{Ga}$ ,  $\text{LaCo}_4\text{Ga}$ , and  $\text{YCo}_4\text{Al}$ .

$$K_1 = [2 - p]K_1^{2c} + [3 - (1 - p)]K_1^{3g}, \quad (4)$$

where  $p$  is the fraction of Ga or Al atoms per formula unit which enter the  $2c$  sites, and  $K_1^{2c}$  and  $K_1^{3g}$  are the individual  $2c$  and  $3g$  site contributions to the Co anisotropy. One would thus expect, on this basis, that Ga or Al substitution would cause the observed reduction in anisotropy only if a marked preference of Ga or Al for the axial  $2c$  sites of approximately 60% were present. The neutron data clearly indicate a strong preference of Ga and Al for the planar  $3g$

sites. Returning to the case of the compound  $\text{YCo}_4\text{B}$  cited above, the same general considerations must be taken with care, since, even though B atoms are known to enter the  $2c$  sites exclusively, and would cause a strong decrease in the anisotropy, the crystal structure is different (ordered  $\text{CeCo}_4\text{B}$  structure) and the known occurrence of a first-order field-induced transition<sup>30</sup> first-order magnetization process implies that higher-order anisotropy terms arise from the substitution.<sup>31</sup> The single-site model, however, does account for the anisotropy when Co is substituted by a  $3d$  atom such as Cu in  $\text{LaCo}_{5-x}\text{Cu}_x$  and  $\text{YCo}_{5-x}\text{Cu}_x$ ,<sup>32</sup> considering the known random nature of the Cu substitution on the  $2c$  and  $3g$  sites<sup>12,33,34</sup> (the random substitution reported in these references has been observed by three separate accurate neutron diffraction investigations but it is in contrast with a less detailed neutron study which suggests Cu substitution in  $2c$  sites<sup>35</sup>).

These combined results indicate that the single-site approximation is not generally applicable. The failure of the localized model in the case of Ga, Al, and B substitutions appears to be due to the fact that the contributions of the remaining Co atoms are drastically modified by entrance of non- $3d$  metals. It can be expected to lead to wrong results in cases where the valence states of the substituted atoms hybridize strongly with the  $3d$  electrons of the Co atoms. It seems, however, to be applicable in the case of  $3d$ - $3d$  metal substitution.

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