

## Individual Co site contributions to the magnetic anisotropy and NMR investigation of $Y_2(Co_{1-x}M_x)_{17}$ ( $M = Cu, Al$ )

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Co-site crystalline fields in  $YCo_5$  and  $Y_2Co_{17}$  are calculated using a point-charge model. It is shown that the  $2c$  site in  $YCo_5$  and the  $18j$  site in  $Y_2Co_{17}$  make positive contributions to the magnetic anisotropy, while the  $3g$  site in  $YCo_5$  and the  $18h$ ,  $9d$ , and  $6c$  sites in  $Y_2Co_{17}$  make negative contributions, in agreement with Streever's NMR result. Through an NMR investigation,  $M$  substitution for Co in  $Y_2(Co_{1-x}M_x)_{17}$  ( $M = Cu, Al$ ) is demonstrated to occur at random in  $M = Cu$  and with a site preference in  $M = Al$ . Al is found mainly to prefer a  $6c$  dumbbell site. Consequently, it is shown that the different magnetic anisotropy behaviors between  $Y_2(Co_{1-x}Cu_x)_{17}$  and  $Y_2(Co_{1-x}Al_x)_{17}$  arise from the differences in site preference between Cu and Al for Co.

### I. INTRODUCTION

Rare-earth cobalt intermetallic compounds, such as  $RCo_5$  and  $R_2Co_{17}$ , have been attractive as high-energy-product permanent magnets, which possess high magnetic anisotropy. The high magnetic anisotropy of the compounds is mainly contributed from the crystalline field acting on rare-earth atoms coupled with spin-orbit interaction which governs the magnetic easy-axis directions of the compounds.<sup>1</sup> However, the rare-earth cobalt compounds with nonmagnetic rare-earth elements, such as Ce, La, and Y, also have a large magnetic anisotropy, which suggests the importance of the cobalt atom's contribution to the magnetic anisotropy of the compounds.

$Y_2Co_{17}$  possesses an easy direction of magnetization along the basal plane, while  $YCo_5$  possesses an easy  $c$  axis. Recently, Hamano *et al.*<sup>2</sup> measured magnetic anisotropy of  $Y_2(Co_{1-x}M_x)_{17}$  single crystal ( $M = Al$  or  $Cu$ ) and revealed that magnetic anisotropy hardly changes with Cu substitution, while it drastically changes with Al substitution and goes from negative (easy  $c$  plane) to positive (easy  $c$  axis) at  $x$  exceeding 0.12.

The easy axis change with substituted atoms for Co has also been observed in  $Y_2(Co, Fe)_{17}$  compounds.<sup>3</sup> This easy axis change has been interpreted in terms of a site preference of Fe atoms for Co atoms.<sup>4</sup> This suggests that the anisotropy behavior in  $Y_2(Co_{1-x}M_x)_{17}$  might be explained by the difference in the site preference between Cu and Al for Co.

The idea of site-preference contribution to the easy-axis change implies the existence of the significant difference in the crystallographically different in-

dividual Co site anisotropy, which has been suggested by the author.<sup>4</sup> Recently, Streever<sup>5</sup> has revealed the anisotropy difference of an individual Co site in  $R$ -Co intermetallic compounds by NMR study.

This paper first calculates the Co anisotropy in  $YCo_5$  and  $Y_2Co_{17}$ , based on the crystalline field using a point-charge model, to reveal the difference in the site preference between Al and Cu for Co in  $Y_2(Co_{1-x}M_x)_{17}$  through NMR study and then shows how site preference can be used to determine anisotropy behavior in  $Y_2(Co_{1-x}M_x)_{17}$  compounds.

### II. Co ANISOTROPY IN $YCo_5$ AND $Y_2Co_{17}$ COMPOUNDS

Recently, an analysis was carried out on the crystalline field in  $3d$  metals using the localized-moment model.<sup>6</sup> In the analysis, expressions have been obtained for the contributions from lattice ions and conduction electrons to the crystalline field. Numerical values of the potential for hexagonal Co metal were found to be well coincided with the values calculated using point-charge models in sign and also in magnitude. Therefore, the contributions from Co atoms to the crystalline field were calculated, using a point-charge model to estimate Co anisotropy in the  $YCo_5$  and  $Y_2Co_{17}$  compounds.

Crystal structures for  $RCo_5$  and rhombohedral  $R_2Co_{17}$  are shown in Fig. 1.  $R_2Co_{17}$  structure, which crystallizes in a rhombohedral or a hexagonal form, depending on the rare-earth element, is derived from the more basic  $RCo_5$  structure by ordered substitutions of  $R$  atoms by pairs of Co atoms.  $RCo_5$  has

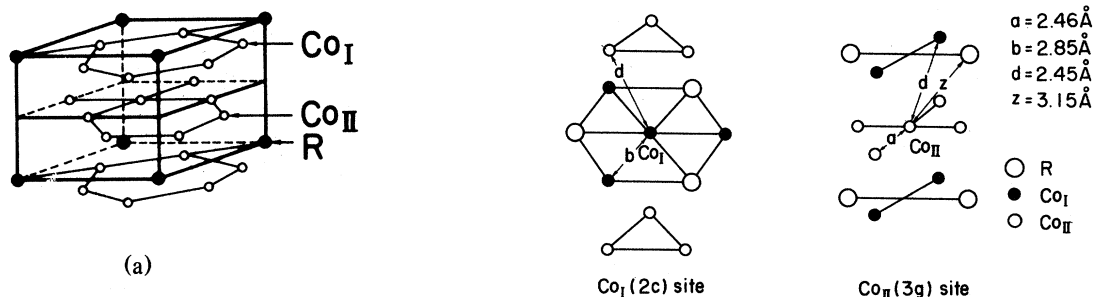


FIG. 1. Crystal structures (a)  $R\text{Co}_5$  and (b) rhombohedral  $R_2\text{Co}_{17}$ .

two crystallographically different sites applicable to Co atoms.  $R_2\text{Co}_{17}$  structures of rhombohedral and hexagonal types are closely related. Both structures have four crystallographically different Co sites, whose corresponding sites in the two structures have nearly the same environment. The local environments for each Co site, including the first and second neighbors, are drawn in Figs. 2 and 3 for  $R\text{Co}_5$  and rhombohedral  $R_2\text{Co}_{17}$  structure, respectively.

#### A. $R\text{Co}_5$

The  $\text{Co}_I$  site in Fig. 2 has  $C_3$  and  $\sigma_h$  symmetries, so that crystalline-field parameters, other than  $B_2^0$  and  $B_4^0$ , are zero. The  $\text{Co}_{II}$  site has a  $c$ -plane symmetry plane, which makes  $B_2^1$ ,  $B_4^1$ , and  $B_4^3$  zero. There are two additional  $\text{Co}_{II}$  positions in the  $R\text{Co}_5$  unit cell having local environments equivalent to the  $\text{Co}_{II}$  site described in Fig. 2, whose neighbor  $\text{Co}_I$  atoms in Fig. 2 are rotated by  $\pm 2\pi/3$  around the  $c$  axis. Parameters  $B_2^2$ ,  $B_4^2$ , and  $B_4^4$  are, therefore,

FIG. 2. Local environment around Co in  $R\text{Co}_5$ . Atomic distances are shown for  $\text{YCo}_5$ .

eradicated by averaging over the three  $\text{Co}_{II}$  positions. Thus, the  $\text{Co}_{II}$  site has also only  $B_2^0$  and  $B_4^0$  in the crystalline-field potential.

#### B. $R_2\text{Co}_{17}$

The  $\text{Co}_I$  site in Fig. 3 has a  $c$ -plane symmetry plane, so that  $B_2^1$ ,  $B_4^1$ , and  $B_4^3$  vanish. There are two additional  $\text{Co}_I$  positions in the  $R_2\text{Co}_{17}$  unit cell, other than the  $\text{Co}_I$  site described in Fig. 3, in which the surrounding atoms around  $\text{Co}_I$  in Fig. 3 are rotated by  $\pm 2\pi/3$  around the  $c$  axis. Parameters  $B_2^2$ ,  $B_4^2$ , and  $B_4^4$  are eradicated by averaging over the three  $\text{Co}_I$  positions.

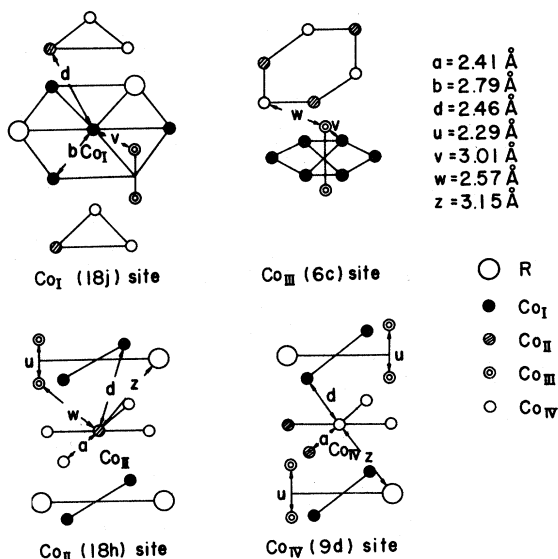


FIG. 3. Local environment around Co in rhombohedral  $R_2\text{Co}_{17}$ . Atomic distances are shown for  $\text{Y}_2\text{Co}_{17}$ .

Likewise, there are three  $\text{Co}_{\text{II}}$  positions in the  $\text{R}_2\text{Co}_{17}$  unit cell equivalent to the  $\text{Co}_{\text{II}}$  site described in Fig. 3. Parameters  $B_2^1$ ,  $B_4^1$ , and  $B_4^3$  are eradicated by averaging over the four  $\text{Co}_{\text{II}}$  positions.  $B_2^2$ ,  $B_4^2$ , and  $B_4^4$  values are calculated to be an order of magnitude smaller than those of  $B_2^0$  and  $B_4^0$ , respectively.

The  $\text{Co}_{\text{III}}$  site in Fig. 3 has  $C_3$  and  $\sigma_h$  symmetries, so that the crystalline-field parameters, other than  $B_2^0$  and  $B_4^0$ , are zero. There is one more  $\text{Co}_{\text{IV}}$  position other than  $\text{Co}_{\text{IV}}$  described in Fig. 3, in which each atom around  $\text{Co}_{\text{IV}}$  in Fig. 3 is rotated by  $\pi$  around the  $c$  axis. Then,  $B_2^1$ ,  $B_4^1$ , and  $B_4^3$  are eliminated by averaging over the two  $\text{Co}_{\text{IV}}$  positions.  $B_2^2$ ,  $B_4^2$ , and  $B_4^4$  are calculated to be an order of magnitude smaller than those of  $B_2^0$  and  $B_4^0$ , respectively.

Thus, neglecting the smaller  $B_2^2$ ,  $B_4^2$ , and  $B_4^4$  terms, the crystalline-field potential, acting on each Co site in  $\text{RCo}_5$  and  $\text{R}_2\text{Co}_{17}$ , can be described as

$$V = B_2^0 [3L_z^2 - L(L+1)] + B_4^0 \times [35L_z^4 - 30L(L+1)L_z^2 + 25L_z^2 - 6L(L+1) + 3L^2(L+1)^2], \quad (1)$$

where

$$B_2^0 = -\frac{e}{4} \frac{5}{\pi} \gamma_{20} \alpha \langle r^2 \rangle, \quad B_4^0 = -\frac{3e}{16} \frac{1}{\pi} \gamma_{40} \beta \langle r^4 \rangle, \quad (2)$$

$$\gamma_{no} = 4\pi \sum_j \frac{q_j}{(2n+1)R_j^{(n+1)} Y_n^0(\theta_j, \phi_j)},$$

and  $-e$  is electronic charge.  $q_j$  is atomic charge, assumed to be  $+3e$  for Y atoms and  $+Z^*e$  for Co atoms. The summation of  $\gamma_{no}$  is carried out to the environment atoms with distance  $R_j$  from the reference cobalt atom drawn in Figs. 2 and 3.  $\alpha$  and  $\beta$  are the Stevens multiplicative factors. The magnetic moment of the cobalt atoms in  $\text{YCo}_5$  and  $\text{Y}_2\text{Co}_{17}$  is 1.46 (Ref. 7) and  $1.64\mu_B$ ,<sup>8</sup> respectively, with the assumption that the Y atom be nonmagnetic, which leads to 0.74 in  $\text{YCo}_5$  and 0.82 in  $\text{Y}_2\text{Co}_{17}$  for the cobalt spin moment. If the cobalt spin moment is roughly assumed to be  $S=1$  in  $\text{YCo}_5$  and  $\text{Y}_2\text{Co}_{17}$  from the localized model, cobalt atoms have a  $3d^8$  configuration, which leads to  $\alpha = \frac{2}{105}$  and  $\beta = \frac{2}{315}$ .  $\langle r^2 \rangle$  and  $\langle r^4 \rangle$  are taken from the Slater function.  $Y_n^m(\theta, \phi)$  is spherical harmonic function with spherical coordinates  $(R, \theta, \phi)$ , and  $L$  is the angular momentum.

At first, consider the neighboring Y-atom contribution to the Co magnetic anisotropy in  $\text{YCo}_5$ . As seen in Fig. 2, three Y atom neighbors around  $\text{Co}_I$  are in the  $c$  plane, which leads to positive  $B_2^0$  in Eq. (1).  $B_4^0$  is shown to be one order smaller than  $B_2^0$  in magnitude. Therefore, the  $\text{Co}_I$  spin orientation arising from the Y-atom neighbors favors a  $c$  plane. This is in conflict with Streever's result of an easy  $c$  axis for the  $\text{Co}_I$  spin. Four Y-atom neighbors around a  $\text{Co}_{\text{II}}$  atom are in a plane perpendicular to the  $c$  plane,

which leads to negative value for  $B_2^0$ .  $B_4^0$  is also shown to be one order smaller than  $B_2^0$  in magnitude. Consequently,  $\text{Co}_{\text{II}}$  spin orientation arising from Y atom neighbors favors a  $c$  axis, which is again in conflict with Streever's result that the  $\text{Co}_{\text{II}}$  spin is favorable in the  $c$  plane. Accordingly, the contribution of Y neighbors to each Co anisotropy in  $\text{YCo}_5$  will be considered not to be dominant. Likewise, it can also be shown that Y-neighbors contribution to the crystalline field in  $\text{Y}_2\text{Co}_{17}$  is not dominant.

The Co neighbors contribution to each Co-site crystalline field can be calculated using Eqs. (1) and (2). For the  $^3F$  state  $(3d)^8$  of the Co atom, the energy levels for each Co site in  $\text{YCo}_5$  and  $\text{Y}_2\text{Co}_{17}$  are shown in Fig. 4. As seen in the figure, the ground states for  $\text{Co}_I$  and  $\text{Co}_{\text{II}}$  in  $\text{YCo}_5$  are  $L_z = \pm 3$  and 0, respectively, supporting the polarized neutron studies<sup>9</sup> that  $\text{Co}_I$  atoms have large  $3d$  orbital moment, and magnetization densities of  $\text{Co}_{\text{II}}$  atoms are more isotropic. This implies that  $\text{Co}_I$  site spin favors  $c$  axis and  $\text{Co}_{\text{II}}$  spin favors  $c$  plane through spin-orbit coupling. This result agrees with Streever's result. For  $\text{Y}_2\text{Co}_{17}$ , the  $\text{Co}_I$  ground state is  $L_z = \pm 3$ , while states for  $\text{Co}_{\text{II}}$ ,  $\text{Co}_{\text{III}}$ , and  $\text{Co}_{\text{IV}}$  are all  $L_z = 0$ . This result also signifies that, in  $\text{Y}_2\text{Co}_{17}$ ,  $\text{Co}_I$  spin favors  $c$  axis and other Co site spins favor  $c$  plane, which agrees again with Streever's result.

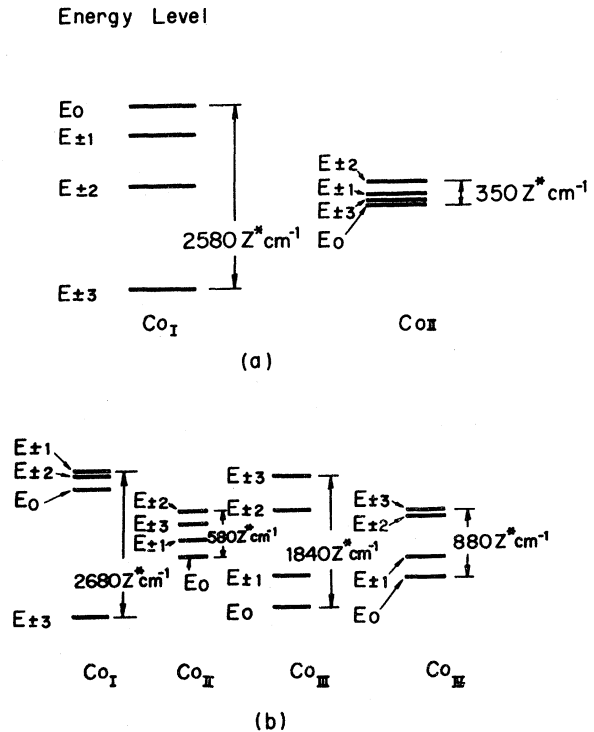


FIG. 4. Energy levels for each Co site calculated by Eqs. (1) and (2) in (a)  $\text{YCo}_5$  and in (b)  $\text{Y}_2\text{Co}_{17}$ .

It is noted that, in  $YCo_5$ , first excited state ( $L_z = \pm 3$ ) energy difference from ground-state energy  $\Delta E$  of  $Co_{II}$  is very small, compared with that of  $Co_I$ . The small  $\Delta E$  in  $Co_{II}$  will make a smaller  $Co_{II}$  contribution to the magnetic anisotropy. Namely, magnetic anisotropy of  $YCo_5$  will mainly arise from  $Co_I$  contribution. In  $Y_2Co_{17}$ , on the other hand, magnetic anisotropy will mainly arise from  $Co_{II}$ ,  $Co_{III}$ , and  $Co_{IV}$  contributions, which all favor  $c$  plane of Co spin in agreement with experimental magnetic easy axis of  $Y_2Co_{17}$ . In particular,  $Co_{III}$  contribution to the  $c$  plane of Co spin seems to be the largest. It is also noted that in  $YCo_5$  ground orbital states for crystal-line fields are  $L_z = \pm 3$ , that is, degenerate and in  $Y_2Co_{17}$   $L_z = 0$ , that is, nondegenerate. Consequently, magnetic anisotropy will be larger in  $YCo_5$  than in  $Y_2Co_{17}$  because degenerate ground orbital states make rather large magnetic anisotropy through the first-order spin-orbit coupling. This may explain the difference in the magnitude of magnetic anisotropy between  $YCo_5$  and  $Y_2Co_{17}$ .

### III. NMR IN $Y_2(Co_{1-x}M_x)_{17}$ ( $M = Cu$ OR $Al$ ) COMPOUNDS

Alloys for NMR study were prepared by arc melting using 99.9 wt. % Y, 99.5 wt. % Co, 99.99 wt. % Cu, and 99.9 wt. % Al constituent elements. Thus obtained ingots were annealed for 10 days at 1100°C and then rapidly cooled to room temperature. Single phase of each sample was confirmed by x-ray analysis. The annealed ingots were ground into powders whose particles were 10  $\mu m$  in diameter and which were used for NMR measurement.

NMR observations were accomplished using the spin-echo method at liquid- $N_2$  temperature within a cold-finger-insert Dewar, which fits into a resonance coil. The NMR equipment used is conventional. The NMR intensity was measured by obtaining a maximum echo signal for each frequency observed on the oscilloscope. Echo intensities obtained at each frequency were divided by that frequency.

The  $^{59}Co$  NMR spectra in  $Y_2(Co_{1-x}Cu_x)_{17}$  and  $Y_2(Co_{1-x}Al_x)_{17}$  at liquid- $N_2$  temperature are shown in Fig. 5. The  $^{59}Co$  NMR spectrum in  $Y_2Co_{17}$  has four peaks, corresponding to four crystallographically different Co atom sites in  $R_2Co_{17}$  structure. The four peaks at 166, 174, 203, and 219 MHz in Fig. 5 correspond to  $Co_{II}$ ,  $Co_{IV}$ ,  $Co_I$ , and  $Co_{III}$  in Fig. 3, respectively.<sup>10</sup> Namely, the greater the number of local cobalt atoms, the higher the  $^{59}Co$  resonance frequency. As shown in Fig. 5, the NMR spectra behave differently between Cu and Al substituted compounds. In the Cu substituted compounds, the peak resonance frequency at each Co site scarcely changes up to  $x = 0.10$ . At  $x = 0.15$  a new resonance peak appears at 152 MHz and the resonance peak

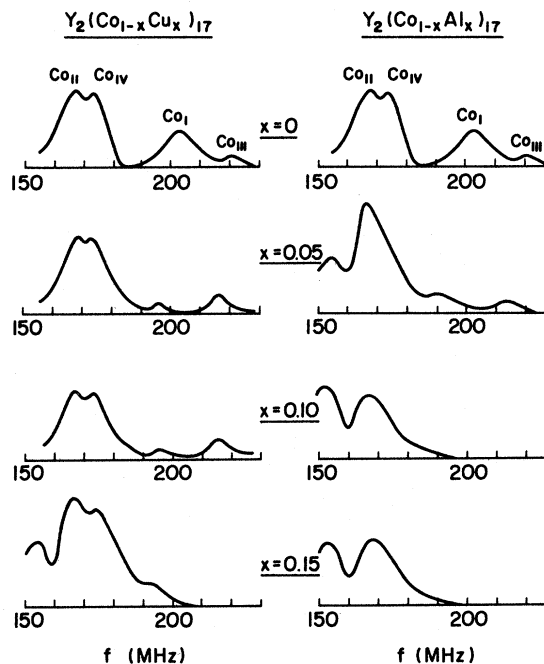


FIG. 5. NMR spectra for  $^{59}Co$  in  $Y_2(Co_{1-x}Cu_x)_{17}$  and  $Y_2(Co_{1-x}Al_x)_{17}$  at liquid- $N_2$  temperature.

corresponding to  $Co_{III}$  disappears. In the Al-substituted compounds, on the other hand, a new resonance peak at 152 MHz appears at  $x = 0.05$  and the resonance peaks disappear at  $x \geq 0.05$  for  $Co_{IV}$  and  $x \geq 0.10$  for  $Co_I$  and  $Co_{III}$ , respectively.

The NMR intensities at  $Co_I$ ,  $Co_{IV}$ , and  $Co_{III}$  sites normalized by the intensity at  $Co_{II}$  site are shown in Fig. 6 as a function of  $x$  in  $Y_2(Co_{1-x}Cu_x)_{17}$  and  $Y_2(Co_{1-x}Al_x)_{17}$ . In the Al-substituted compounds the  $Co_{III}$  intensity rapidly decreases, which is suggestive of site preference of Al for  $Co_{III}$ . On the other hand, Cu substitution for Co atoms in  $Y_2(Co_{1-x}Cu_x)_{17}$  seems to be at random, because the intensity at each Co site gradually changes with  $x$ .

Hyperfine fields of  $^{59}Co$  in  $Y_2Co_{17}$  can be estimated<sup>4</sup> by

$$H_{Co_i} = (-30.4 - 7.8N_{Co_i})\mu_{Co} \quad (3)$$

where,  $i$  implies I, II, III, and IV,  $\mu_{Co}$  is the magnetic moment, and  $N_{Co_i}$  is the number of Co neighbors around  $Co_i$  atom site which are 11, 9, 13, and 10 for  $Co_I$ ,  $Co_{II}$ ,  $Co_{III}$ , and  $Co_{IV}$ , respectively, as shown in Fig. 3. If some  $Co_{III}$  atom pairs are substituted for by Al,  $Co_{III}$  resonance intensity will diminish. The hyperfine field for  $Co_{II}$  whose neighbor one  $Co_{III}$  pair is substituted for by Al, can be estimated as 152 kOe, using Eq. (3) with  $\mu_{Co} = 1.64\mu_B$  and  $N_{Co_{II}} = 8$ , which leads to 154 MHz resonance frequency. This value

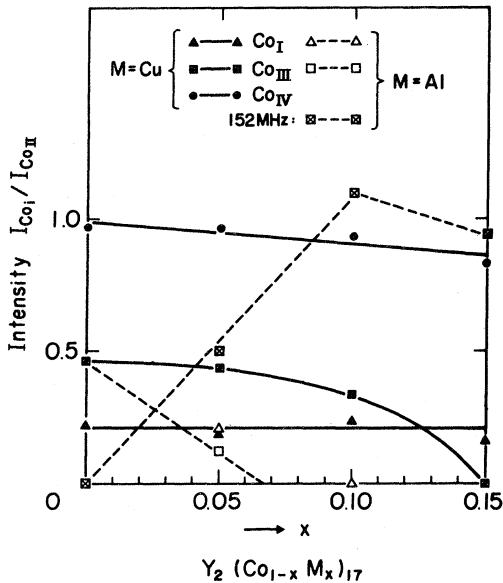


FIG. 6. NMR intensity of each cobalt site, normalized by that of  $\text{Co}_{\text{II}}$  atom in  $\text{Y}_2(\text{Co}_{1-x}\text{Cu}_x)_{17}$  and  $\text{Y}_2(\text{Co}_{1-x}\text{Al}_x)_{17}$ .

coincides well with the observed 152 MHz resonance peak frequency. The resonance frequency corresponding for  $\text{Co}_{\text{IV}}$ , whose neighbor two  $\text{Co}_{\text{III}}$  pairs are substituted for by Al, is also estimated as 154 MHz. Because the numbers of  $\text{Co}_{\text{II}}$  and  $\text{Co}_{\text{IV}}$  atoms whose neighbor one or two  $\text{Co}_{\text{III}}$  pairs are substituted for by Al atoms increase with increasing Al concentration, 154 MHz resonance intensity will increase with Al substitution, which is observed in Fig. 6.

#### IV. INTERPRETATION OF ANISOTROPY IN $\text{Y}_2(\text{Co}_{1-x}\text{M}_x)_{17}$ ( $M = \text{Cu OR Al}$ )

It was shown in Sec. II, that  $\text{Co}_{\text{I}}$  contribution to the magnetic anisotropy was positive, while  $\text{Co}_{\text{II}}$ ,  $\text{Co}_{\text{III}}$ , and  $\text{Co}_{\text{IV}}$  contributions were negative. In particular,  $\text{Co}_{\text{III}}$  was the largest in the negative contribution. The Al substituted compounds are first considered. According to the NMR results, Al atoms prefer  $\text{Co}_{\text{III}}$  atom sites. In  $\text{Y}_2(\text{Co}_{1-x}\text{Al}_x)_{17}$ , the resonance of  $\text{Co}_{\text{II}}$  and  $\text{Co}_{\text{III}}$  with negative contributions to magnetic anisotropy are still observed at  $x = 0.05$ . At  $x \geq 0.10$ , on the other hand,  $\text{Co}_{\text{III}}$  resonance disappears and only two peaks at 152 and 165 MHz are observed. As indicated in Sec. III the resonance at 152 MHz corresponds to the contributions from  $\text{Co}_{\text{II}}$  and  $\text{Co}_{\text{IV}}$

with  $\text{Co}_{\text{III}}$  atom neighbors substituted by Al. The resonance at 165 MHz, on the other hand, corresponds to the contributions from  $\text{Co}_{\text{I}}$ ,  $\text{Co}_{\text{II}}$  with  $\text{Co}_{\text{III}}$  atom neighbors substituted by Al and  $\text{Co}_{\text{IV}}$  with one  $\text{Co}_{\text{III}}$  atom pair of the two substituted by Al, which have  $N_{\text{Co}} = 9$ , respectively. The crystalline field at each Co atom site with  $N_{\text{Co}} = 8$  or 9 at  $x \geq 0.10$  in  $\text{Y}_2(\text{Co}_{1-x}\text{Al}_x)_{17}$  is similar to that of the respective Co atom site with  $N_{\text{Co}} = 8$  or 9 in  $\text{YCo}_5$ , except that one or two Y atom neighbors around each Co atom are replaced with two nonmagnetic Al atoms. Thus, the magnetic anisotropy in  $\text{Y}_2(\text{Co}_{1-x}\text{Al}_x)_{17}$  will become positive from negative at  $x$  exceeding 0.1 because  $\text{YCo}_5$  has positive anisotropy. This explains the magnetic anisotropy behavior of  $\text{Y}_2(\text{Co}_{1-x}\text{Al}_x)_{17}$  observed by Hamano *et al.*

In  $\text{Y}_2(\text{Co}_{1-x}\text{Cu}_x)_{17}$ , on the other hand, Cu replaces Co atoms at random, thus,  $\text{Co}_{\text{III}}$  resonance peak is still observed up to  $x = 0.10$  and  $\text{Co}_{\text{II}}$  and  $\text{Co}_{\text{IV}}$  resonance peaks up to  $x = 0.15$ , whose contributions to magnetic anisotropy are all negative, as calculated in Sec. II. Consequently, the sign of magnetic anisotropy in  $\text{Y}_2(\text{Co}_{1-x}\text{Cu}_x)_{17}$  will not change with  $x$  up to 0.15, at least. This also supports the experiments reported by Hamano *et al.*

#### V. SUMMARY

Co site crystalline field in  $\text{YCo}_5$  and  $\text{Y}_2\text{Co}_{17}$  was calculated using the point-charge model. It was shown that Co neighbor contribution to the each Co site magnetic anisotropy was larger than the Y neighbor contribution. Consequently,  $2c$  site in  $\text{YCo}_5$  and  $18j$  site in  $\text{Y}_2\text{Co}_{17}$  make positive contributions to the anisotropy, while  $3g$  site in  $\text{YCo}_5$  and  $18h$ ,  $9d$ , and  $6c$  sites in  $\text{Y}_2\text{Co}_{17}$  make negative contributions, in agreement with Streever's NMR result.

NMR study in  $\text{Y}_2(\text{Co}_{1-x}\text{M}_x)_{17}$  ( $M = \text{Cu, Al}$ ) revealed that Cu substitution for Co was at random, while Al substitution for Co succeeded with a site preference. Al mainly prefers  $6c$  dumbbell site. Thus, in  $\text{Y}_2(\text{Co}_{1-x}\text{Al}_x)_{17}$  the change of the magnetic anisotropy in sign appears to depend on diminution of the negative  $6c$  site contribution to the magnetic anisotropy through Al preference for  $6c$  site, while in  $\text{Y}_2(\text{Co}_{1-x}\text{Cu}_x)_{17}$  it appears not to occur with Cu substitution because Cu diminishes each cobalt site contribution, equally.

#### ACKNOWLEDGMENT

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