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# Spontaneous magnetic order in $Ce_{0.73}Ho_{0.27}Ru_2$ below the superconducting transition temperature

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Mössbauer-effect studies on the Laves-phase alloy $Ce_{0.73}Ho_{0.27}Ru_2$ have been carried out as a function of temperature to investigate the possible coexistence of spontaneous magnetic order and superconductivity. The alloy was dilutely doped with $^{57}Co$, which substitutes for the ruthenium atoms. At a temperature of about 2 K, the $^{57}Fe$ Mössbauer spectra showed the onset of line broadening that increased with decreasing temperature. The broadening is attributed to magnetic ordering of the holmium atoms, which produces magnetic interactions at some of the $^{57}Fe$ impurity nuclei, and is interpreted in terms of a model which yields temperature-dependent hyperfine fields $H_i$. Saturation of $H_i$ is suggested below 0.5 K. A molecular-field model was fit to $H_i$, giving a saturation hyperfine field of 3.47 ± 0.13 T and a magnetic ordering temperature of 1.72 ± 0.03 K. Low-field-susceptibility measurements determined the superconducting transition temperature to be 1.57 K with a half-width of 0.18 K. The paramagnetic data were extrapolated to a Curie temperature of 1.3 ± 0.2 K. There is no evidence for reentrant behavior down to 35 mK. Magnetization measurements gave a magnetic moment of $5.08 \pm 0.2\mu_B$ per formula unit, and the derived high-field-susceptibility data gave a Curie temperature of 1.5 ± 0.7 K. The reported results are consistent with the recent neutron scattering measurements on $Ce_{0.73}Ho_{0.27}Ru_2$. The coexistence of superconductivity with some type of short-range ferromagnetism is supported.

## I. INTRODUCTION

The coexistence of magnetic order and superconductivity is suggested in the phase diagrams determined by a number of magnetic-susceptibility studies[1–4] on a class of rare-earth substituted, Laves-phase compounds. These compounds are of the form $Ce_{1-x}R_xRu_2$, where $R$ denotes a magnetic rare-earth atom. Starting from pure $CeRu_2$, a slow depression of the superconducting transition temperature $T_c$ is observed with increasing $x$. The ferromagnetic-ordering temperature $\theta$ is also depressed with decreasing $x$, starting from a region rich in the magnetic rare-earth element. The two curves, $T_c(x)$ and $\theta(x)$, then intersect at some finite $T$ and $x$. Extrapolation through the intersection of the two-phase boundaries defines the region of possible coexistence. Intersection temperatures and concentrations taken from the phase diagrams of Wilhelm and Hillenbrand[2] range from 3.8 K at $x = 0.135$ for gadolinium to 1.6 K at $x = 0.27$ for holmium.

Because of the diamagnetism associated with superconductivity, susceptibility measurements cannot be used to determine magnetic ordering at temperatures below $T_c$. Other types of macroscopic and microscopic techniques are not affected by the superconducting screening currents. In fact, such techniques have been used to detect magnetic order in the superconducting state of these materials. Specific-heat measurements below $T_c$ by Peter *et al.*[3] and Rupp[5] on $Ce_{1-x}Gd_xRu_2$ and by Rupp[5] on the terbium-substituted alloy show contributions attributed to magnetic ordering of the rare-earth ions. A study by Roth *et al.*[6] on $Ce_{0.8}Tb_{0.2}Ru_2$ ($T_C \sim 2.6$ K), using small-angle inelastic neutron scattering, identified the presence of short-range ferromagnetic ordering at 1.5 K. Other nuclear methods have also sampled directly the presence of magnetic order. Kumagai *et al.*[7] used a spin-echo NMR technique on gadolinium in $Ce_{1-x}Gd_xRu_2$ for $x \geq 0.105$. Their results support coexistence. Ruebenbauer *et al.*[8] have used the Mössbauer effect (ME) of $^{155}Gd$ to study this same material in the concentration range $0.05 \leq x \leq 0.5$. Temperature-dependent line broadening was observed in the ME spectra below $T_c$ for concentrations in the coexistence region. The broadening was attributed to unresolved magnetic hyperfine splitting resulting from at least partially ordered gadolinium ions. Because of difficulties associated with many of the existing rare-earth ME transitions and because not every rare-earth nucleus has a transition, we developed a technique where dilute $^{57}Co$ impurities can be introduced into these materials.[9] The $^{57}Fe$ Mössbauer resonance may then be used to detect the magnetic ordering of the rare-earth ions. Using this technique, spontaneous spin order was observed in the gadolinium-substituted alloys in the superconducting state.[9] The spin order was manifested by a partially resolved, temperature-dependent hyperfine component in the ME spectra. In all of the observations of magnetic order just described, no significant differences were detected between ordering in the normal and in the superconducting phases.

The largest rare-earth concentration associated with the coexistence regions of the $(Ce_{1-x},R_x)Ru_2$ alloys is for the holmium-substituted material. Two parallel studies have been conducted on samples from the same melt of $Ce_{0.73}Ho_{0.27}Ru_2$. This composition approximates that at the intersection of the phase boundaries. The first study is reported by Lynn *et al.*[10] in the preceding paper, where small-angle inelastic neutron scattering measurements show short-range ferromagnetic correlations between the holmium moments in the superconducting state of the alloy. The correlation length is seen to saturate at low temperature, precluding long-range ferromagnetic order. The second study, reported here, employs the Mössbauer spectroscopy of $^{57}Fe$ impurities to examine spontaneous magnetic spin order of the holmium ions above and below $T_c$. Low-field-susceptibility and bulk-magnetization measurements have also been made to determine $T_c$ and $\theta$ for this composition.

Because of the large concentration of magnetic ions, the substituted Laves-phase alloys remain extremely important in resolving the question of coexistence. These systems are indeed different from the recently discovered reentrant ternaries. For example, $ErRh_4B_4$, which is better behaved metallurgically than $Ce_{1-x}R_xRu_2$, undergoes a complete quenching of the superconducting state below $T_c$ by the onset of long-range ferromagnetic order.[11] Including the present data, the accumlated phenomenological evidence obtained from a variety of techniques confirms the simultaneous existence of some type of limited-range, ferromagnetic order in the superconducting state of certain $Ce_{1-x}R_xRu_2$ compounds. Although a definitive theoretical interpretation is yet to be made, explanations are evolving that incorporate short-range magnetic order. Examples include the spin glass and the mictomagnetic states, recently reviewed by Fischer[12] and Roth.[13]

## II. SAMPLE PREPARATION AND CHARACTERIZATION

The sample was prepared from the elemental materials by arc melting. Pure ruthenium powder was arc melted separately to degas and purify the starting material. Appropriate amounts of high-purity holmium and cerium were added to the ruthenium button to give the correct final compostion, $Ce_{0.73}Ho_{0.27}Ru_2$, hereafter referred to as CHR. The ternary sample was then arc melted twenty times, inverting between each melt, to produce a more homogeneous sample. The button was finally subjected to a homogenization heat treatment of 24 h at 1600 K.

A cylindrically shaped piece was spark cut from the center of the button. This cylinder was sliced into five disks approximately 1 mm thick by 8.6 mm in diameter. The disks were used in the low-field ac-susceptibility and ME experiments. The outer ring of the button with the central core removed was used in other susceptibility and magnetization measurements and in the neutron scattering work of Lynn *et al.*[10]

The several samples were subjected to various tests to confirm the composition and to assess the homogeneity. In particular, several electron-microprobe scans of a central disk indicated that only small amounts ( < 5%) of impurity phases were present (mostly pure ruthenium), and that the bulk of the sample was quite homogeneous. Chemical analysis confirmed the composition, and neutron-powder-diffraction measurements were consistent with the cubic Laves-phase *C*-15 structure.[10]

## III. BULK-MAGNETIC CHARACTERIZATION

### A. Low-temperature ac susceptibility

Because the ME is not sensitive to superconductivity, ac magnetic susceptibility measurements were made to measure the paramagnetic response and to determine the superconducting transition temperature $T_c$ for CHR. The susceptibility was measured over the temperature range 1.2 to 12 K. The temperature was determined from a calibrated germanium resistance thermometer. The 273-Hz measuring field strength was $\leq 0.1$ mT. A temperature-dependent background, determined from a scan with no sample present, was subtracted from the data. Several disks sliced from the central cylinder, one doped with $^{57}Co$, were measured. The data presented in Fig. 1 are for the doped sample used in the ME experiments discussed below. The abrupt diamagnetic response

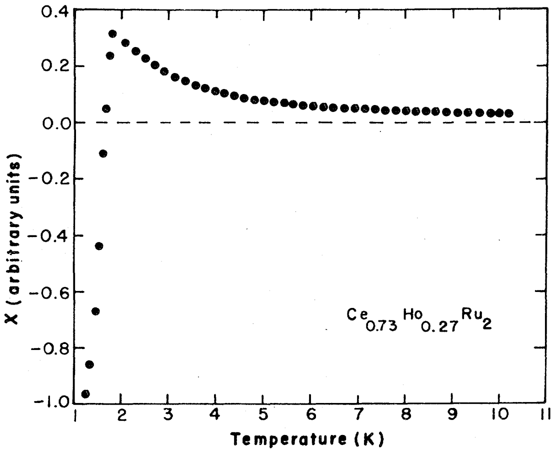

FIG. 1. Low-field ac susceptibility as a function of temperature for $^{57}Co$-doped CHR. The superconducting transition occurs at 1.57 K and has a half-width of 0.18 K.

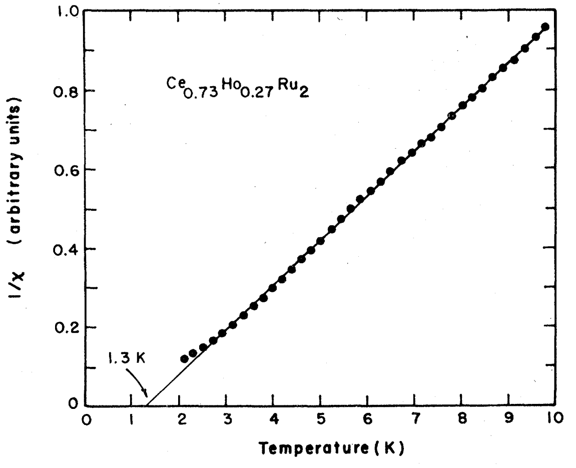

FIG. 2. Reciprocal low-field susceptibility as a function of temperature for CHR. The straight-line fit (Curie-Weiss behavior) extrapolates to $\theta = 1.3 \pm 0.2$ K.

below 1.8 K signals the onset of superconductivity. The midpoint of the transition $T_c$ is 1.57 K, in good agreement with the results of Wilhelm and Hillenbrand.[2] Although we were not able to obtain a low enough temperature in this particular apparatus to see a constant diamagnetic response of the sample, the characteristic $S$ shape of the transition and subsequent ultralow-temperature measurements show that essentially the full transition had been observed. The half-width of the transition is 0.18 K. Separate measurements performed in a $^3$He-$^4$He dilution refrigerator showed that the ac susceptibility was constant from 1.2 K down to 35 mK, indicating no reentrant behavior in this specimen. Other disks and sections of the original button gave $T_c$ values ranging from 1.3 to 1.87 K with the lowest values coming from sample material near the surface of the button. We feel that this range in $T_c$ values is due to local variations in the chemical composition of the sample material.

The paramagnetic response of the sample above $T_c$ was used to obtain a value of the magnetic-ordering temperature $\theta$. A Curie plot of $1/\chi$ is shown in Fig. 2. A straight-line fit to the data between 3 and 10 K extrapolates to a $\theta$ of 1.3 $\pm$ 0.2 K. Deviations from a straight line below 3 K may be caused by a high-temperature tail of the superconducting transition.

### B. Magnetization measurements

Magnetization measurements were performed on part of the outer region of the CHR button which had been crushed to a powder. The measurements were made in the temperature range 1.2 – 220 K and in fields up to 1.17 T in a vibrating sample magnetometer. The dependence of magnetization $M$ on magnetic field $H$ was nonlinear below 20 K. The magnetization measurements yielded a moment of $5.08 \pm 0.2\mu_B$ per formula unit. The dc susceptibility was determined from $M/H$ at 1.17 T for $T > 20$ K. A least-squares straight-line fit to the data gives a $\theta$ value of 1.5 $\pm$0.7 K, in good agreement with the low-field ac-susceptibility result.

## IV. MÖSSBAUER-EFFECT MEASUREMENTS

### A. Source prepartion

The $^{57}$Fe Mössbauer effect was employed as a microscopic probe of spontaneous magnetic spin order. Basic to our undertanding of the results is the assumption that radioactive $^{57}$Co impurity atoms will enter the CHR lattice substitutionally at ruthenium sites. This is expected from the structural isomorphism between $CeRu_2$, $CeCo_2$, and $CeFe_2$.

The source of 14.4-keV $^{57}$Fe $\gamma$ rays used in these measurements was prepared using a central disk taken from the CHR button. To maintain or increase source intensity, this particular disk was doped several times. The disk was prepared by coarse and fine polishing of one of the surfaces. Up to 1 mCi of ammoniacal $^{57}CoCl_2$ was electroplated onto the central 6-mm-diameter region of the prepared surface. After a second polished CHR disk was placed facing the plated side of the first, the sandwiched pair was placed in a covered molybdenum crucible that was inserted into a high-vacuum induction furnace and pumped to $10^{-7}$ Torr. The sample was subjected to a diffusion anneal for, typically, 10 h at 1350 K. Some activity transferred to the facing piece. Both sources in all of the dopings gave essentially the same ME spectra. However, all of the data discussed here were obtained with the source after its final doping in which the effective impurity concentration of cobalt and iron is estimated at $< 500$ ppm. The presence of this much $^{57}$Co is not expected to affect the superconducting properties of CHR significantly; $CeCo_2$ is superconducting with $T_c = 1.5$ K.

### B. Measured spectra

Mössbauer-effect spectra using the above CHR source were obtained in a standard transmission geometry using a room-temperature, single-line $K_4Fe(CN)_6 \cdot 3H_2O$ velocity-modulated absorber. Primary velocity calibration was determined by laser interferometry. The source was mounted in one of two variable-temperature cryostats. The first attained temperatures down to 1.1 K by pumping on liquid $^4$He. The second was a $^3$He cryostat which allowed source temperatures as low as 0.37 K. The temperature of the source was determined by $^4$He or $^3$He vapor-pressure thermometry.

Representative spectra measured at temperatures of 3.95 K and below are shown in Fig. 3. At temperatures above 3 K (including 75 and 298 K), the features of the measured spectra are identical to the 3.95-K spectrum. These spectra are characterized by a resolved symmetric doublet with an effective splitting of 0.76 mm/s and a component linewidth [full width at half maximum (FWHM)] of 0.32 mm/s. These same characteristics were found[9] in $^{57}$Fe spectra for gadolinium-substituted CeRu$_2$ systems above the ordering temperatures and for CeRu$_2$ at all temperatures. Second-phase pure ruthenium, if present, did not contribute to the broadened ME spectra. A $^{57}$Co source of pure ruthenium shows a single broadened line at all temperatures with a different isomer shift relative to $^{57}$Fe in CeRu$_2$.[9] Such a line was not observed in any of the spectra in the present work. As the source temperature is lowered below 3 K, the sharp doublet behavior begins to broaden systematically, as shown in the 2.16-, 1.61-, and 1.08-K spectra of Fig. 3. At the lowest temperatures the broadening suggests additional emerging structure, exemplified by the 0.37-K spectrum of Fig. 3. A qualitative measure of the observed broadening was obtained by fitting each of the spectra to a symmetric Lorentzian doublet with a constrained splitting of 0.76 mm/s. Figure 4 shows the component linewidth from this simple analysis. The analysis ignores any possibilities for additional structure and, indeed, results in progressively poorer fits at low temperatures. The width begins to broaden rapidly as the

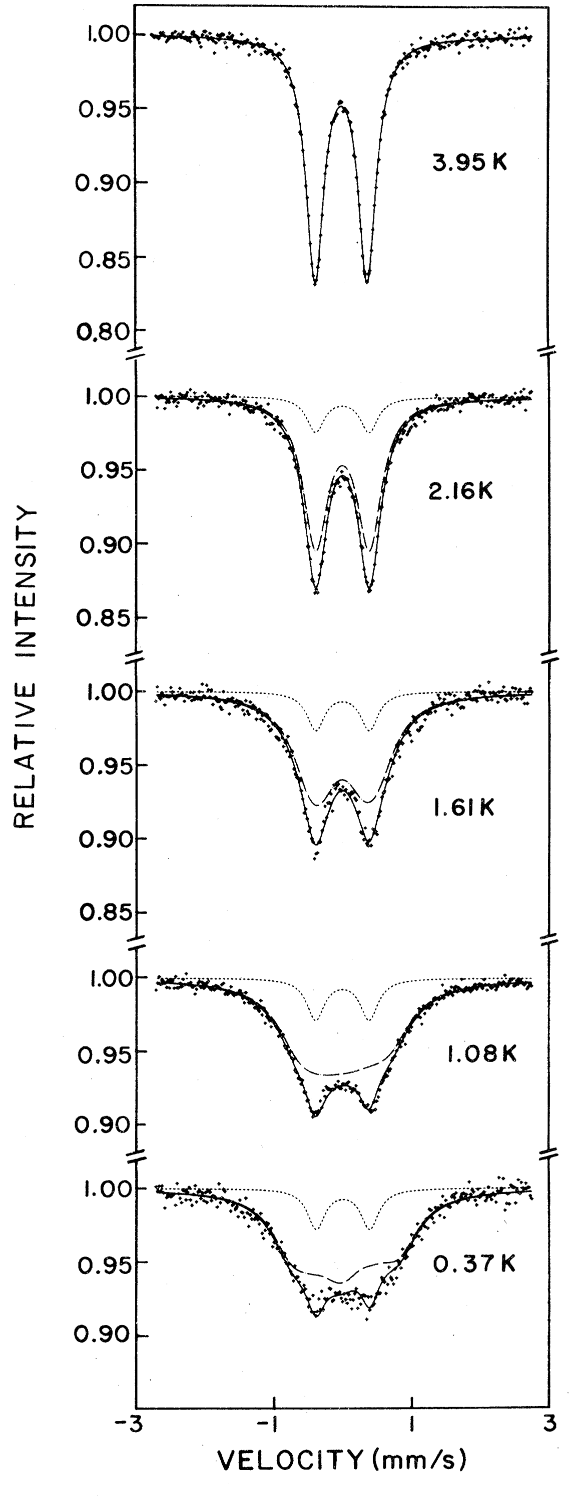

FIG. 3. $^{57}$Fe ME spectra of Ce$_{0.73}$Ho$_{0.27}$Ru$_2$ at 3.95, 2.16, 1.61, 1.08, and 0.37 K showing the development in the two-site model of magnetic order. The dotted curve denotes the pure quadrupole site, and the dashed curve represents the magnetic plus quadrupole site.

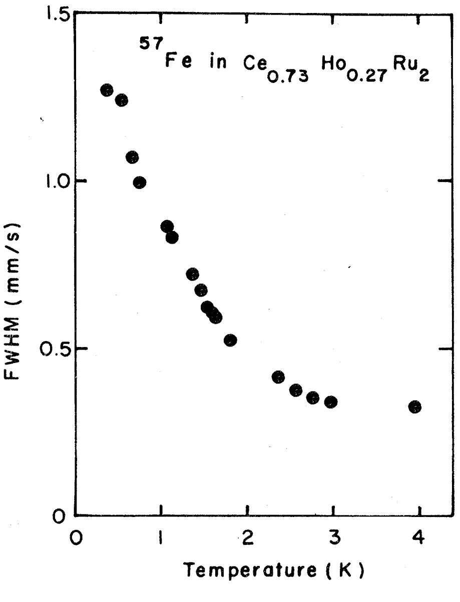

FIG. 4. Component linewidth for a pure quadrupole fit to the $^{57}$Fe ME spectra of CHR as a function of temperature.

temperature is decreased below 2 K with some type of saturation suggested at the lowest temperatures.

Relaxation effects on the low-temperature ME spectra are unlikely with $^{57}$Fe in such a metallic system. Supporting this view is the temperature dependence of the line broadening shown in Fig. 4. The onset of the broadening agrees qualitatively with the transition temperature expected from susceptibility measurements. Furthermore, the FWHM is not inversely dependent on temperature but tends to a low-temperature saturation value.

The behavior of the line broadening with temperature is qualitatively similar to the behavior of the quasielastic neutron scattering intensity (indicative of ferromagnetic correlations) observed by Lynn *et al.*[10] and shown in their Fig. 6. We believe that this low-temperature line broadening reflects the presence of some type of magnetic order at the holmium sites transferred to the $^{57}$Fe impurity sites and is a measure of spontaneous magnetic ordering in CHR, even at temperatures for which CHR is known to be a superconductor.

### C. Interpretation

As in our earlier study of $Ce_{1-x}Gd_xRu_2$,[9] we interpret the behavior of the spectra discussed here in terms of a model that assumes (i) all iron impurities have substituted at one of the crystallographically equivalent ruthenium sites, and (ii) only those iron impurities which have one or more near-neighbor holmium atoms respond to any spontaneous magnetic order present. Within these assumptions, the model permits two nonequivalent sites for the $^{57}$Fe impurities, a pure quadrupole site and a magnetic-plus-quadrupole site.

Qualitatively, the first site in our model represents iron in a $CeRu_2$-like lattice. To describe the ME spectrum resulting from the nuclei at these sites, we consider the quadrupole interaction resulting from an axially symmetric electric field gradient (EFG) directed along the [111] direction of the unit cell. The strength of this interaction and its direction result from the tetrahedral coordination of the ruthenium atoms along the body diagonal. Iron nuclei at this type of site would yield a ME spectrum consisting of a quadrupole-split symmetric doublet. Implicit in our model is the assumption that only those iron impurities residing near magnetically ordered holmium atoms develop a time-averaged magnetic hyperfine splitting that is generally taken to be proportional to the average magnetization. The second site in our model, therefore, represents iron nuclei with at least one holmium atom on the near-neighbor cerium coordination postions. To describe the ME spectrum associated with these second sites, it is necessary to include both a quadrupole interaction and a magnetic-dipole interaction. The EFG tensor is allowed to be asymmetric for this second site.

The Hamiltonians used for the two-site description are expressed in the notation of Kündig[14] as

$$\mathcal{H}_Q = \frac{eQV_{zz}}{4I(2I-1)}[3I_z^2 - I(I+1) + \eta(I_+^2 + I_-^2)] \ , \quad (1)$$

$$\mathcal{H}_M = -g\mu_N H_i[I_z\cos\beta + (1/\sqrt{2})(I_+e^{-i\alpha} + I_-e^{+i\alpha})\sin\beta] \ , \quad (2)$$

$$\text{Site 1: } \mathcal{H}_1 = \mathcal{H}_Q \ , \quad (3)$$

$$\text{Site 2: } \mathcal{H}_2 = \mathcal{H}_Q + \mathcal{H}_M \ . \quad (4)$$

Equation (1) defines an electric quadrupole interaction, where $I$ is the nuclear spin; $I_\pm = I_x \pm iI_y$ with $I_x$, $I_y$, and $I_z$ being angular-momentum operators; $V_{zz}$ is the principal component of the EFG tensor; and $\eta = |V_{xx} - V_{yy}|/V_{zz}$ is the asymmetry parameter for the EFG tensor. Equation (2) specifies the magnetic hyperfine interaction, where $H_i$ is the magnitude of the internal magnetic field at the iron nucleus, $\alpha$ is the polar angle relating the direction of $H_i$ to the $xy$ plane of the EFG frame, and $\beta$ is the azimuthal angle between the direction of magnetization and the $z$ direction of the EFG frame. The other parameters in Eqs. (1) and (2) have their usual meanings. The Hamiltonians appropriate to the two sites of the model are given by Eqs. (3) and (4). A computer program has been developed[15,16] which incorporates these Hamiltonians specified for the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition in $^{57}$Fe. It was used to perform a powder-average least-squares fit of Eqs. (3) and (4) to the measured spectra.

The results of such fits are shown as the curves in Fig. 3. The solid curves represent composite sums of site-1 and -2 results. The component denoted by dots corresponds to a site-1 designation. The component denoted by dashes results from a site-2 description. The fits were obtained using several constraints. The FWHM for site 1 and the effective quadrupole splitting relevant to Eq. (1) were constrained to those values obtained from the 3.95-K spectrum. The values for the asymmetry parameter ($\eta = 0.6$) and the angles ($\alpha = 0$ and $\beta = 54.73°$) fixing the direction of magnetization, [100], were assumed from previous results for $Ce_{0.75}Gd_{0.25}Ru_2$, which has a similar content of a different substituted rare earth. These symmetry values were used because the lack of structure in the present measurements precluded their independent determination. The relative weighting of the spectral areas of site-2 to site-1 components was fixed at 5.6:1. This is the ratio obtained using the binomial theorem to calculate the probability, assuming the proper concentration and nearest-cerium-neighbor coordination, of a ruthenium ($^{57}$Co) site having one or more near-neighbor holmium atoms relative to the probability of

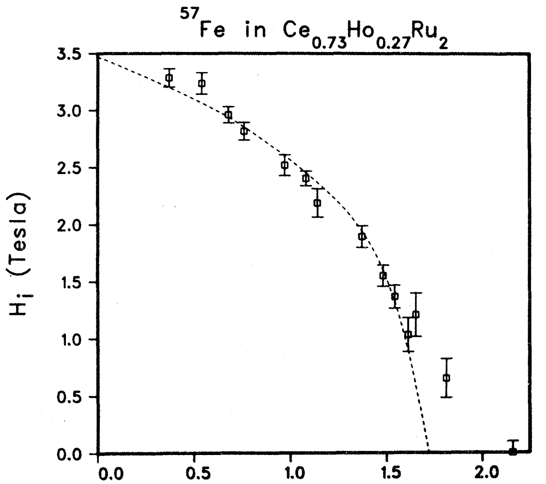

FIG. 5. Two-site-model internal field of the $^{57}$Fe ME in CHR as a function of temperature. The dashed line is a molecular-field-model fit to the data and is described in the text.

having no near-neighbor holmium atoms. The free parameters in these fits are basically associated with site 2 and include the FWHM and the internal field $H_i$ at the $^{57}$Fe nucleus. It was found necessary to allow the linewidth parameter for site 2 to vary, presumably as compensation for a more complicated distribution of local-neighbor configurations. Plotted in Fig. 5 is $H_i$ vs $T$ as obtained from the fitting of the two-site model. The sign of $H_i$ was not determined in this experiment. The dashed curve is a least-squares fit of the data to a molecular-field model. The parameters determined by the fit were $3.47 \pm 0.13$ T for $H_i$ at $T=0$, infinity for the spin value, and $1.72 \pm 0.03$ K for the magnetic-ordering temperature.

## V. DISCUSSION

We believe that the measurements reported here point to the coexistence of superconductivity and spontaneous magnetic order in CHR. The low-field ac-susceptibility measurements clearly indicate the onset of superconductivity at temperatures below 1.8 K, with a midpoint of 1.6 K. The susceptibility measurements in the paramagnetic region extrapolate to a magnetic-ordering temperature of $1.3 \pm 0.2$ K for low-field measurements and of $1.5 \pm 0.7$ K for the high-field, high-temperature data. These data are consistent with a ferromagnetic type of order ($\theta > 0$) and with each other.

The fits of the ME data to the two-site model show a well-defined onset of $H_i$ near 1.8 K and a saturation below 0.5 K (Fig. 5). (The molecular-field model fit gives an ordering temperature of about 1.7 K.) The sharp onset is partly an artifact of the fitting procedure affecting those spectra taken above 2 K. The lack of resolved structure in these spectra caused the site-2 parameters (FWHM and $H_i$) to be essentially equivalent. As a result, all the fits to the spectra for $T > 2$ K gave $H_i = 0$, absorbing any magnetic hyperfine-splitting contribution into a broadened linewidth. Magnetic hyperfine splitting probably begins slightly above 2 K and is reflected in the line broadening seen in Fig. 4.

The $^{57}$Fe ME measurements[9] on $Ce_{1-x}Gd_xRu_2$ exhibit similar behavior in $H_i$ vs $T$, although the saturation internal field is three times greater than in CHR due to the iron atoms being in a different electronic state. Consequently, the spectra for CHR are much less resolved compared to those for the gadolinium system. Nevertheless, the highly constrained fits used on the CHR data could be applied to the data on the gadolinium system with no change in the previous results.

The present ME results on $^{57}$Fe in CHR are in good agreement with the neutron scattering work of Lynn *et al.*[10] on the same system. At the wave vector $Q = 0.035$ Å$^{-1}$, the increase in the magnetic-scattering intensity, which saturates below 0.5 K, closely follows the broadening observed in Fig. 3. It also qualitatively matches the internal field behavior shown in Fig. 4. Lynn *et al.* interpret their neutron scattering data in terms of the Ornstein-Zernike model to show ferromagnetic correlations near 2 K that grow in spatial extent down to 0.5 K. Below this temperature, the correlation length saturates at 80 Å instead of diverging as expected in a normal transition to long-range ferromagnetism. No Bragg peaks indicative of long-range order nor any evidence of behavior attributable to the superconducting transition were observed. Their work supports the contention of the coexistence of superconductivity and ferromagnetic order of limited spatial extent. Much the same conclusion was reached by Roth *et al.*[6] for $Ce_{0.80}Tb_{0.20}Ru_2$; the maximum observed correlation length in this system was 16 Å.

If the order is of a spin-glass type with no long-range order, then the bulk magnetization is zero in zero applied field. Any hyperfine splitting would be proportional to $q^{1/2}$, where $q$ is the spin-glass order parameter of the Edwards-Anderson model.[17] The susceptibility measurements reported by Davidov *et al.*[4] and the $^{155}$Gd ME work by Ruebenbauer *et al.*,[8] both on $Ce_{1-x}Gd_xRu_2$, support the presence of a spin-glass type of magnetic order coexisting with superconductivity. A very slow onset in the appearance of the hyperfine field, interpreted as nearly critical behavior of the system over an extended temperature range, is consistent with the Edwards-Anderson model of spin-glass behavior. The present CHR work shows this same qualitative behavior of the hyperfine field $H_i$ vs $T$.

The present results on CHR indicate the coexistance of superconductivity with some type of spontaneous magnetic order. The magnetic order, exemplified by the presence of a hyperfine field at the $^{57}$Fe nucleus, begins near 2 K and appears to saturate below 0.5 K. This is in qualitative agreement with the temperature dependence of the quasielastic neutron scattering intensity (signaling ferromagnetic correlations) reported by Lynn *et al.* The type of magnetic order thus suggested is short-range ferromagnetism.

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