# Magnetic correlations and crystal-field levels in the superconductor $(Ce_{0.73}Ho_{0.27})Ru_2$

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Neutron scattering studies have been carried out to investigate the atomic magnetic properties of the "magnetic" superconductor $(Ce_{0.73}Ho_{0.27})Ru_2$. At low temperatures we observe the appearance of elastic or quasielastic magnetic scattering at small momentum transfers, indicating the development of ferromagnetic correlations. The temperature and wave-vector dependence of this scattering can be described to a good approximation by an Ornstein-Zernike correlation function over the entire range of wave vectors (0.035–0.20 $Å^{-1}$) and temperatures (0.05–4.2 K) explored. The range of the spatial correlations $\xi$ $(=1/\kappa)$ increases smoothly with decreasing temperature and suggests the onset of ferromagnetism at $\sim$ 0.5 K. However, below 0.5 K, $\xi$ ceases to increase, saturating at a value of 80 Å with no detectable change in the scattering below that temperature. Thus there is no transition to conventional long-range ferromagnetic order. There is also no indication in the magnetic scattering of the onset of superconductivity at 1.6 K. Measurements of the inelastic magnetic scattering reveal a number of crystal-field transitions, demonstrating that the crystalline electric field removes the 17-fold degeneracy of the $J=8$ $Ho^{3+}$ free ion. The nature of the splittings can be understood on the basis of a crystal field with cubic symmetry, and the ground state is found to be the triply degenerate $\Gamma_5$ state, which possesses a magnetic moment. At low temperatures additional magnetic inelastic scattering is observed at low energies; this suggests that there are substantial exchange effects even though the characteristic magnetic temperature (0.5 K) is very small.

## I. INTRODUCTION

Magnetic impurities drastically alter the superconducting state of most superconducting materials since they provide spin-flip mechanisms which can scatter (singlet) Cooper pairs. Usually the superconducting transition temperature $T_s$ is observed to decrease rapidly as the impurity concentration is increased,[1] consistent with the Abrikosov-Gorkov theory[2] and its extensions.[3] A notable exception to this behavior is the $CeRu_2$ alloy system, in which concentrations of heavy-rare-earth metals can be substituted for Ce without seriously affecting its superconducting properties.[4] For example, small amounts of Ho actually enhance $T_s$, and approximately 35% of the Ce ions can be replaced by Ho before the superconducting state is suppressed. At the other extreme, $HoRu_2$ is a ferromagnet with a Curie temperature $T_C$ which decreases as Ho is replaced by Ce. There is a range of concentrations in which the magnetic-phase boundary, identified by the appearance of a peak in the susceptibility, seems to cross the superconducting boundary thus suggesting coexistence of superconductivity with long-range ferromagnetic order.[5] This behavior is evident in the phase diagram of $(Ce_{1-x}Ho_x)Ru_2$ as given by Wilhelm and Hillenbrand[4] (see Fig. 1). Further indications of coexistence appear in Mössbauer measurements which show hyperfine splitting developing in material which is in the superconducting state.[6]

Qualitatively all the heavy rare earths exhibit similar behavior when substituted into the $CeRu_2$; small concentrations have little effect on the superconducting properties and at larger concentrations there appears to be a region where magnetic order and superconductivity seem to coexist. To investigate the microscopic magnetic properties of this unusual system we have carried out a series of neutron scattering studies on a representative "coexistence" alloy $(Ce_{0.73}Ho_{0.27})Ru_2$. We chose to study the Ho-substituted material for several reasons; first, the free-ion magnetic moment of Ho is very large $(10\mu_B)$

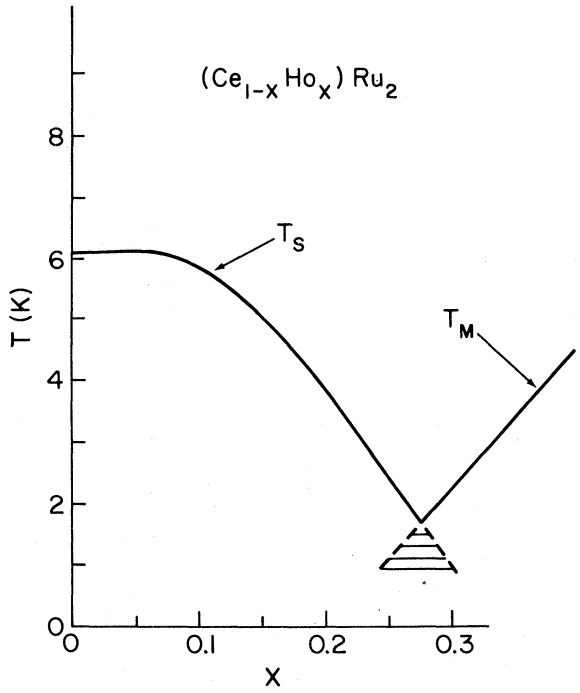


FIG. 1. Schematic diagram showing the concentration dependence of the superconducting transition temperature ($T_S$) and the "magnetic" transition temperature $T_M$ (defined as the observed peak in the measured susceptibility), taken after Wilhelm and Hillenbrand (Ref. 4). The intersection of the two phase boundaries is the so-called "coexistence" region.

and it has a relatively low nuclear absorption cross section. Second, the "coexistence" region has the highest rare-earth concentration for Ho. Finally, these particular samples have been studied by a variety of other techniques including Mössbauer methods as described in the following paper.

Neutron scattering is ideal for studying such a system since the internal magnetic state can be probed without interference from superconducting screening currents. Our neutron measurements show that ferromagnetic correlations develop between the Ho moments in the superconducting state, but the range of correlation does not diverge. There is consequently no transition to true long-range ferromagnetic order. The system seems rather to behave like a ferromagnetic spin glass or "cryptoferromagnet",[7] in contrast to the recently discovered ternary superconductors[8–10] in which the superconducting state is quenched by the onset of long-range ferromagnetic order.[11–14] $(Ce_{1-x}Ho_x)Ru_2$ thus appears to represent an intermediate case in which the system develops *short-range* ferromagnetic order and the superconducting state persists. Preliminary results of our experiments have been briefly reported earlier.[15]

## II. SAMPLE PREPARATION AND CHARACTERIZATION

Two samples were prepared[16] by arc melting the proper proportions of the pure elements together. One pure $CeRu_2$ specimen was prepared as a control and one of composition $(Ce_{0.73}Ho_{0.27})Ru_2$. The samples were annealed at 1300 °C for 24 h to homogenize them. The center of each arc melted "button" was then removed with a spark cutter and doped with $^{57}Co$ for Mössbauer studies; the remainder of each sample ($\sim 0.2$ cm$^3$) was used for the neutron measurements. Chemical analysis of the Ho alloy confirmed a concentration of 27%. The samples were also checked by microprobe analysis and x-ray fluorescence. Only trace amounts of impurity phases (mainly pure Ru) and small inhomogeneities were found. Neutron powder diffraction measurements showed a structure consistent with the expected $C$-15 cubic Laves phase (space group $O_h^7$; $Fd3m$). The lattice constant at helium temperatures was measured to be $7.510 \pm 0.005$ Å for the $(Ce_{0.73}Ho_{0.27})Ru_2$ sample.

ac susceptibility measurements[17] indicate a superconducting transition temperature of 1.6 K for the Ho alloy. Additional measurements[18] extending down to 35 mK establish that the sample is not reentrant, i.e., there is no transition to the normal-conducting phase. The susceptibility measurements[18] at higher temperatures indicate a (ferromagnetic) Curie constant of $\sim 2.2$ K.

## III. EXPERIMENTAL RESULTS

The neutron scattering experiments were carried out utilizing the triple-axis technique. Measurements of the magnetic correlations and high-resolution inelastic scattering were performed at the high-flux beam reactor at Brookhaven National Laboratory, where the sample was mounted in a dilution refrigerator with a low-temperature capability of 50 mK. Measurements of the crystal-field excitations were made primarily at the research reactor at the National Bureau of Standards. In all cases pyrolytic graphite crystals were used as monochromator and analyzer. Söller slit collimators between 10′ and 40′ full width at half maximum (FWHM) were employed both before and after the monochromator and analyzer as required to achieve the necessary resolution and intensity. Pyrolytic graphite filters were used to suppress higher-order wavelength contaminations.

### A. Crystal field

For $Ho^{3+}$ the free-ion ground state has a total angular momentum $J=8$, with $gJ=10\mu_B$. In a crystal, the 17-fold degeneracy of this state may be lifted at

least partially by the effect of the crystalline electric field acting on the orbital part of the wave function; for situations where the exchange energy is small compared to the crystal-field splittings, the latter will determine the magnetic properties at low temperatures. To understand the magnetic properties of these materials, and in particular to study the interplay between magnetism and superconductivity, it is essential to determine the crystal-field ground state of the rare-earth ions. Indeed it is possible for a nonmagnetic state to lie lowest in energy, in which case the rare-earth ions would be nonmagnetic at low temperatures. As we shall see below, however, this is not the case in the system of interest here.

In the $CeRu_2$ structure,[19] the Ce sites possess cubic point symmetry ($\bar{4}3m$), with each Ce having 12 Ru nearest neighbors and 4 Ce next-nearest neighbors. In the substitutional alloy, the replacement of Ce with Ho breaks the cubic symmetry locally, but the influence of this on the crystal-field splittings should be a small perturbation since the dominant effect arises from the 12 Ru nearest neighbors; furthermore, for the range of concentrations presently of interest typically only one in four next-nearest neighbors will be changed. Thus to a first approximation we may expect the overall level scheme to be described by a crystal field with cubic symmetry. The crystal-field Hamiltonian in this case is[20]

$$H = W\left|x\frac{O_4}{F(4)} + (1-|x|)\frac{O_6}{F(6)}\right| , \tag{1}$$

where $W$ is a measure of the overall splitting and $x$ governs the ratio of the fourth- to sixth-order terms. The behavior of the eigenvalues as a function of $x$ is shown in Fig. 2. $|x|=1$ corresponds to the case of only the fourth-order term contributing, whereas $x=0$ corresponds to only the sixth order. Typically the fourth-order term dominates and then the magnitude of $x$ is close to 1. Then if $Wx>0$, the triply degenerate $\Gamma_5$ state (Bethe's notation[21]) lies lowest and the ground state will be magnetic; whereas if $Wx<0$, the $\Gamma_1$ singlet (and therefore nonmagnetic state) will be the ground state.

The cross section per ion for the scattering of neutrons by a crystal-field transition is given in the dipole approximation by[22]

$$\frac{d^2\sigma}{d\Omega\, dE'} = \left(\frac{\gamma e^2}{2mc^2}\right)^2 g_f^2 |f(\vec{K})|^2 \sum_{i,f} P_i |\langle f|\vec{J}_\perp|i\rangle|^2 \delta(E_i - E_f - E') \tag{2a}$$

$$= 2(0.27\times 10^{-12}\ \text{cm})^2 g_f^2 |f(\vec{K})|^2 \sum_{i,f} P_i \sum_{\nu,\nu'} |\langle \Gamma_f, \nu| J_\alpha |\Gamma_i, \nu'\rangle|^2 \delta(E_i - E_f - E') , \tag{2b}$$

where $f(\vec{K})$ is the Fourier transform of the magnetization density, $|i\rangle$ and $|f\rangle$ are the wave functions for the initial and final states, $\vec{J}_\perp$ is the component of the angular momentum perpendicular to the momentum transfer $\hbar\vec{K}$, and the delta function assures conservation of energy. Equation (2b) follows from Eq. (2a) due to cubic symmetry. The matrix elements

$$\sum_{\nu,\nu'} |\langle \Gamma_f, \nu|J_\alpha|\Gamma_i, \nu'\rangle|^2 , \tag{3}$$

have been calculated by Birgeneau[23] as a function of $x$. The labels $\nu, \nu'$ here designate the states within a crystal-field manifold which are degenerate, and the subscript $\alpha$ denotes $x$, $y$, or $z$ which are equivalent in cubic symmetry. The matrix elements for $Ho^{3+}$ are shown in Fig. 3.

The transition probability is proportional to the thermal population $P_i$ of the initial state $|i\rangle$. Since the ions are assumed to be noninteracting magnetically, $P_i$ is given by Boltzmann statistics

$$P_i = \frac{e^{-E_i/kT}}{\mathrm{Tr}\, e^{-H/kT}} , \tag{4}$$

where the denominator is the partition function. We remark that $P_i$ is the thermal population for a particular state $|i,\nu\rangle$ since the degeneracy of the crystal-field states is already taken into account in Eq. (3).

The inelastic scattering at a temperature of 4.6 K is shown in Fig. 4. Since the thermal energy ($kT=0.4$ meV) is small in comparison with the two observed excitations at 3.7 and 15.5 meV, these correspond to transitions out of the crystal-field ground state. The excitations are identified as crystal field in origin since their energies are independent of the momentum transfer (i.e., dispersionless) and because their intensities decrease gradually with increasing $\vec{K}$ in accordance with the magnetic form factor $f(\vec{K})$. The strength of the scattering at 15.5 meV is $4.4 \pm 0.4$ times less than at 3.7 meV.

At elevated temperatures higher-energy crystal-field levels become thermally populated and this may allow additional transitions to be observed. Figure 5 shows the scattering at temperatures of 40 and 80 K, where two transitions become observable at 8.5 and 11.5 meV. We also note that the intensity of the scattering at 15.5 meV (along with that at 3.7 meV) is much reduced at high temperatures due to the de-

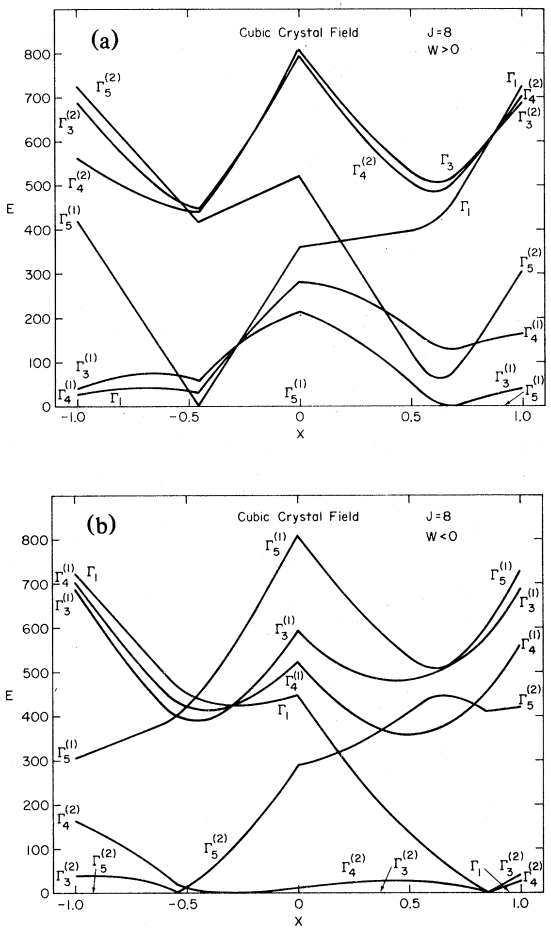


FIG. 2. Crystal-field levels appropriate for $Ho^{3+}(J=8)$ in a cubic field as a function of the parameter $x$, which is related to the ratio of the fourth- to sixth-order terms (see text). $|x|=1$ corresponds to fourth-order terms only, $x=0$ to sixth order only. The level schemes are identical to those given by Lea, Leask, and Wolf (Ref. 20) except that the ground state has been chosen to be at zero energy. (a) $W>0$; (b) $W<0$.

creased thermal occupancy of the crystal-field ground state. This behavior contrasts with that of Bose excitations such as magnons and phonons, whose intensities increase with temperature. We also note that there is a small decrease in the observed energies with increasing temperature which is presumably due to the temperature dependence of the lattice parameter.

To interpret these crystal-field data it is first necessary to determine if any of the scattering originates from the Ce. Inelastic measurements were therefore carried out on the $CeRu_2$ sample, and no excitations of any kind were observed in the small wave-vector region over the energy range 0–20 meV. This behavior is to be expected since susceptibility measurements[4,18,24] on pure $CeRu_2$ show that it is non-

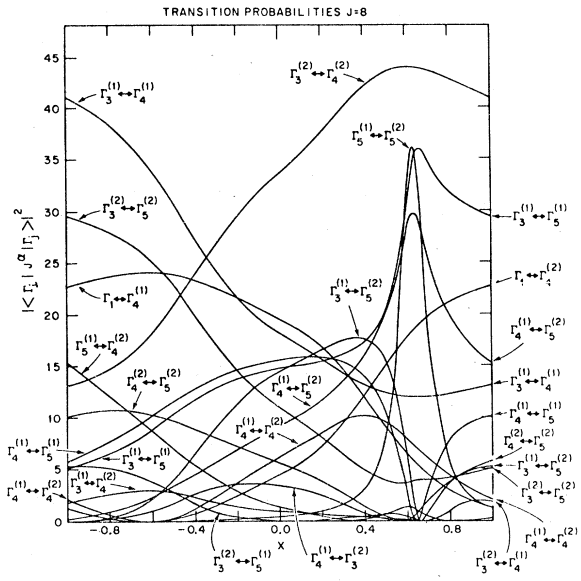


FIG. 3. Matrix elements $|\langle\Gamma_i|J_z|\Gamma_j\rangle|^2$ governing the crystal-field transition probabilities for neutron scattering as a function of the crystal-field parameter $x$ (from Birgeneau, Ref. 23).

magnetic; the Ce being in the +4 valence state with no $4f$ electron. This does not rule out the possibility, however, that a change in valence occurs with increasing Ho content. We therefore had x-ray photoemission[25] measurements carried out on both samples. These data establish that the Ce is in the same valence state in both materials. Finally we note that if a valence change occurred below room temperature, it would inevitably be accompanied by an abrupt change in lattice constant, and no such change has been found in the present investigation or reported elsewhere. We therefore conclude that the observed neutron inelastic scattering originates solely from the Ho.

There are three possible types of crystal-field

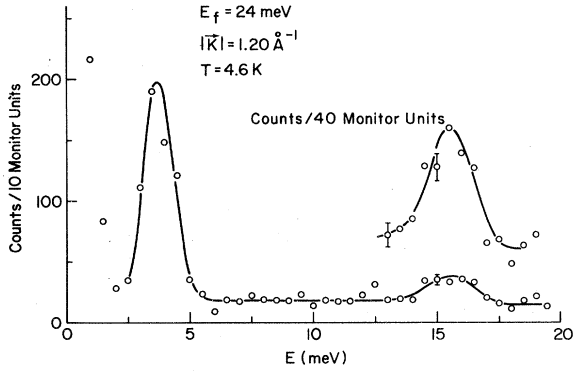


FIG. 4. Inelastic neutron scattering at low temperatures, showing two crystal-field transitions out of the ground state.

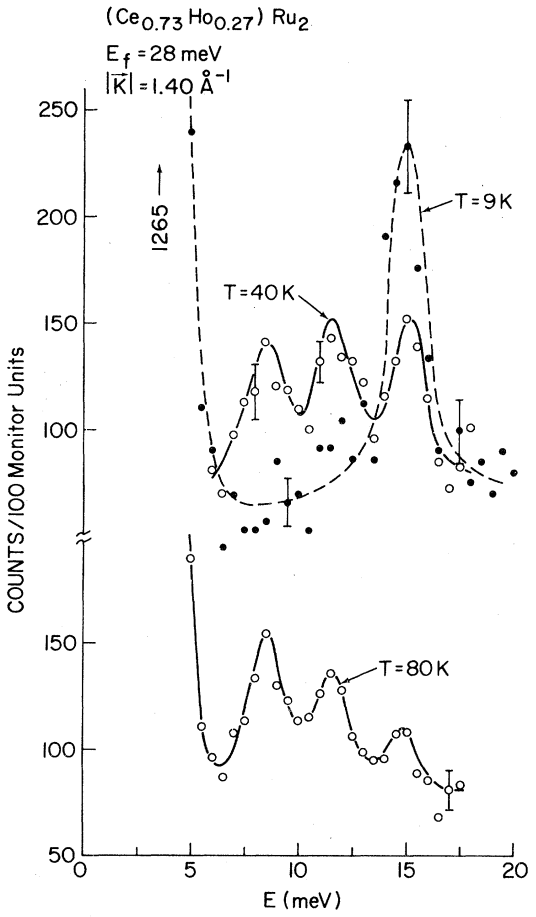


FIG. 5. Inelastic scattering at (a) 40 K and (b) 80 K, showing that the intensity at 15 meV decreases with increasing temperature due to the decreased thermal occupancy of the ground state, and that two excited state crystal-field transitions appear at 8.5 and 11.5 meV.

ground states for $4f^{10}(^5I_8)$ Ho ion in a cubic crystal field[26]; a $\Gamma_1$ singlet, a $\Gamma_3^{(2)}$ nonmagnetic doublet, and the triply degenerate $\Gamma_5^{(1)}$ or $\Gamma_5^{(2)}$. For a level scheme as complicated as $J=8$ it is usually not possible to identify the appropriate crystal-field parameters uniquely without employing intensity information. We can, however, eliminate the possibility of a $\Gamma_1$ ground state immediately. For $W<0$ there is no way to obtain the correct energies to explain the transitions out of the ground state. For $W>0$ it is possible to explain the transitions at 3.7 and 15.5 meV, but not the 8.5-meV transition. Likewise for the $\Gamma_3^{(2)}$ doublet ($W<0$) the only possibility is with $x=+0.57$. This predicts eigenvalues close to the experimental ones, but the predicted intensities are qualitatively incorrect. In particular the level at 3.7 meV would be the $\Gamma_1$, whose transition probabilities with respect to the ground state and the 15.5-meV state are identically zero. We therefore discard this possibility.

The magnetic-triplet $\Gamma_5$ state remains as the only possible ground state. The transitions at low temperatures are superficially consistent with $|x|\sim 1$, $Wx>0$, with $W\sim\pm 0.1$ meV. If it is assumed that $|x|\sim 1$, the sequence of levels would be $\Gamma_5$ (triplet) ground state; $\Gamma_3$ (doublet), $\Gamma_4$ (triplet), $\Gamma_5$ (triplet). The predicted intensities at low temperatures would be in the ratio 4:1 as observed experimentally. But the experimental results as higher temperatures rule out $x\sim\pm 1$ as possibilities. For $x\sim -1$ no 8.5-meV excitation would be expected, while if $x\sim +1$ the 8.5-meV transition would be observable by virture of the thermal population of the 15.5-meV state and would be unobservable at 40 K. In addition, although calculated energies are close to those observed, they are still outside the limits of experimental error. Alternatively, one might try to reduce $W$ so that the $\Gamma_4$ state occurs at 3.7 meV rather than 15.5 meV, which would yield a viable energy-level scheme but qualitatively incorrect intensities.

Only one further value of $x$ remains a possibility; $x\cong -0.3$. The sequence of levels would be: $\Gamma_5^{(1)}$ ground state; $\Gamma_1$, $\Gamma_4^{(1)}$, and $\Gamma_3^{(1)}$ at $\sim$ 3.7 meV; $\Gamma_5^{(2)}$ at 12.5 meV; and the $\Gamma_4^{(2)}$ and $\Gamma_3^{(2)}$ at 15.5 meV, corresponding to a value of $W$ of +0.028 meV. This scheme is the only one which gives the correct energies, intensities, and temperature variations. For this value of $x$, the $\Gamma_5^{(1)}$ ground state has $\langle J_z\rangle = -4.5, 0, 4.5$, with magnetic moments of $(-5.6, 0, +5.6)\mu_B$. We note that a value of -0.3 for $x$ means that the sixth-order contribution to the crystal-field Hamiltonian is large. Although the value of $x$ is difficult to predict in metallic systems as complicated as the present case, this value is nevertheless rather unusual and should be regarded with some caution. This is particularly evident since we observe, as discussed below, additional low-energy scattering at low temperatures whose origin is uncertain. We therefore regard the present values of $W$ and $x$ as tentative. It would be helpful to repeat the measurements for other Ho concentrations, particularly lower concentrations where the Ho-Ho interactions would be reduced. Such measurements are in progress; preliminary results[27] are consistent with the present values of $W$ and $x$.

### B. Magnetic order and correlations

If long-range ferromagnetic ordering of the Ho ions were to coexist on a microscopic scale with superconductivity, then one might expect that any macroscopic magnetization due to the Ho ordering would be compensated by the magnetic field generated by supercurrents. The supercurrents have a large spatial

extent so the associated magnetic form factor would have an appreciable value only at small wave vectors. The magnetic form factor of the Ho ion, on the other hand, is proportional to the Fourier transform of the *atomic* magnetization density of its highly localized $4f$ electrons, and decreases only very slowly with increasing wave vector. Thus at wave vectors corresponding to possible magnetic Bragg peaks, only the Ho contribution to the magnetic intensity would be significant. Conventional neutron-diffraction measurements would therefore be sensitive to any long-range magnetic ordering of the Ho, if it occurs, irrespective of the response of the superconducting electrons.

To determine if long-range ferromagnetic ordering of the Ho occurs at low temperatures, the temperature dependence of the {111} Bragg peak was measured from high temperatures (78 K) down to $\sim 50$ mK. No change in the intensity of the peak outside experimental error was detected, putting an upper limit of $0.3\mu_B$ on any possible net ferromagnetic component to the moment. Complete diffraction patterns were also taken at several temperatures to check for other possible types of order, such as antiferromagnetic, spiral, or spin-density wave order. *No additional Bragg peaks of any kind were observed. Thus there is no conventional long-range magnetic order of any kind in this sample.* We do observe, however, the appearance of elastic or quasielastic small-momentum transfer scattering at low temperatures. Figure 6 shows the temperature variation of the scattering at $|\vec{K}| = 0.035$ Å$^{-1}$. The scattering increases in intensity down to $\sim 0.5$ K, and then no further change is observed. The data shown in the figure are typical of the results throughout the small wave-vector region.

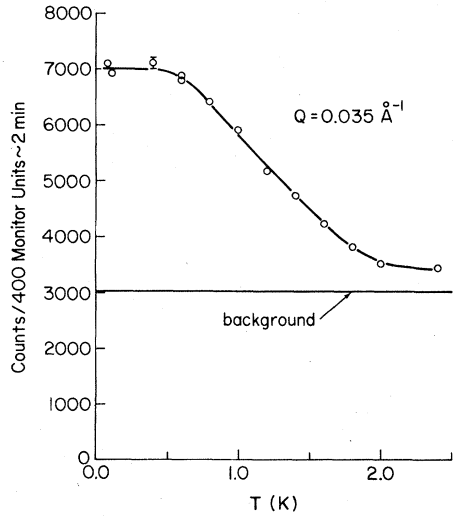


FIG. 6. Temperature evolution of the quasielastic scattering at a wave vector of 0.035 Å$^{-1}$. The enhancement of the scattering at small wave vectors demonstrates that the correlations which develop at low temperatures are ferromagnetic in nature.

The wave-vector dependence of the quasielastic scattering is shown in Fig. 7 at several temperatures. An analyzer crystal was employed in the elastic position to discriminate against higher-energy inelastic scattering, in particular against the crystal-field scattering. In this configuration the energy resolution of the spectrometer was 0.4-meV FWHM. Background was determined by measuring the scattering at high temperatures (40 K). The rapid increase in the background at small momentum transfers is due to overlap with the incident beam. The net scattering decreases with increasing $\vec{K}$ at each temperature as expected from ferromagnetic critical fluctuations. Such small-angle scattering is indicative of ferromagnetic correlations developing between Ho ions which are in the $\Gamma_5$ ground state, since at these low temperatures only the crystal-field ground state is significantly populated. We note that the $\Gamma_5$ triplet state would split isotropically in a magnetic field, so that this could be treated as a system with an effective spin $S = 1$.

Since the magnetic excitation energies at small wave vectors are expected to be much smaller than the instrumental energy resolution, the data collection procedure effectively integrates over energy and the scattering is proportional to the static wave-vector-dependent susceptibility. The cross section in this case should be given to a good approximation by the Ornstein-Zernike form

$$\frac{d\sigma}{d\Omega} \propto \frac{A}{|\vec{K}|^2 + \kappa^2} , \tag{5}$$

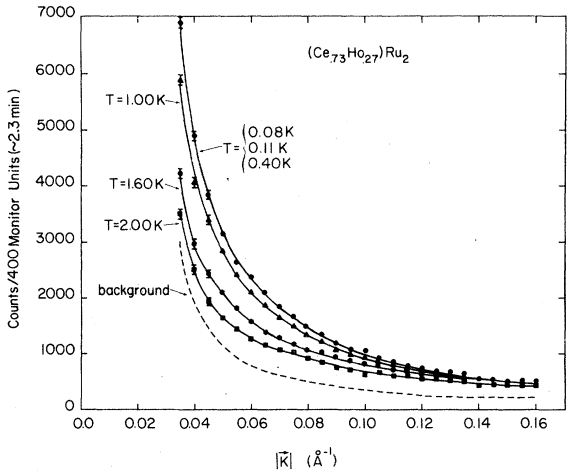


FIG. 7. Wave-vector dependence of the scattering at several temperatures. The solid curves are the result of a least-squares fit to the data assuming an Ornstein-Zernike correlation function.

where $\kappa$ is the inverse of the correlation range $\xi(\xi=1/\kappa)$ in the crystal.[28] In a conventional ferromagnetic phase transition $\xi$ would increase steadily as the transition is approached and diverge at $T_C$.

To determine $\xi$ at each temperature, Eq. (5) was convoluted with the instrumental resolution and least-squares fitted to the data, with $A$ and $\kappa$ as free parameters. This procedure gave good fits to the data, with a quality of fit $\chi^2$ close to unity in each case, showing that Eq. (5) is a good representation of the correlation function over this wave vector and temperature region. The temperature dependence of $\xi$ (and $\kappa$) is shown in Fig. 8. It is evident that the region of correlated spins increases monotonically with decreasing temperature, and in analogy with other ferromagnets these data suggest a divergence at $\sim 0.5$ K. However, for temperatures of 0.5 K and lower we detected no change in the angular variation or overall intensity of the scattering, and we see from Fig. 8 that $\xi$ saturates at a value of 80 Å. There is thus no evidence of a critical divergence. We also found no indication in the small wave-vector magnetic scattering of the onset of superconductivity at 1.6 K.

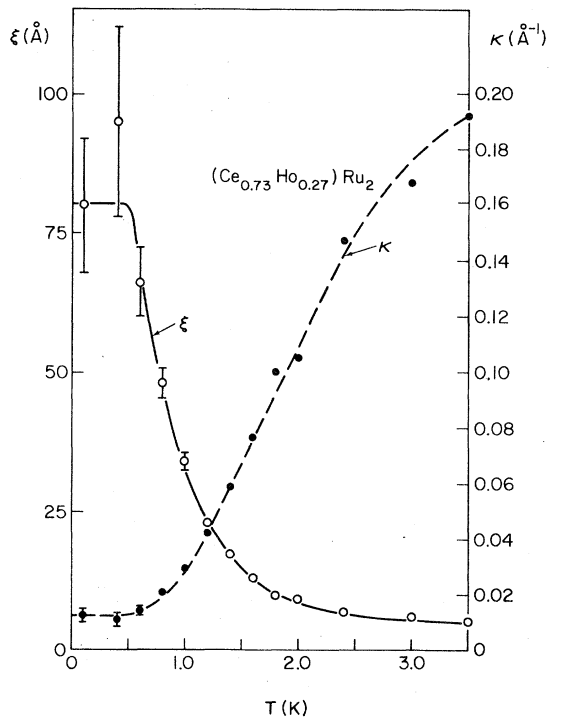


FIG. 8. Temperature dependence of the correlation range $\xi$ (and inverse correlation range $\kappa=1/\xi$). The range of the correlations increases with decreasing temperature, but does not diverge to produce a state with long-range ferromagnetic order. The limiting value of the correlation range is $\sim 80$ Å.

### C. Inelastic scattering at low temperatures

In addition to the crystal-field scattering already discussed, there appears at low temperatures some low-energy scattering which we suspect may be associated with the development of ferromagnetic correlations. Figure 9 shows this scattering at several different temperatures and wave vectors. We note that the width of the scattering is considerably broader than the instrumental resolution (0.2-meV FWHM). Higher-resolution data taken with an incident energy of 5.0 meV confirmed that there is little additional structure to this scattering. Its characteristic energy is $\sim 0.8$ meV. With increasing temperature its width increases; by 3.5 K there are no resolvable peaks at any wave vector, and by 10 K the scattering is no longer identifiable.

## IV. DISCUSSION

The small-angle neutron scattering results demonstrate that ferromagnetic correlations develop in the superconducting phase, but the correlation range $\xi$ saturates at a value of 80 Å rather than diverging to

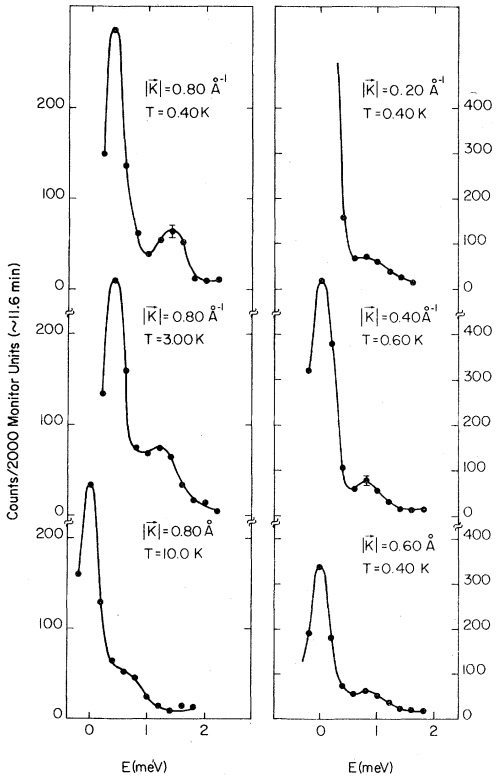


FIG. 9. Inelastic magnetic scattering observed at low energies for several temperatures and wave vectors. The incident energy is 13.5 meV, with an energy resolution of 0.2-meV FWHM.

produce a ferromagnetic state. Similar behavior has been observed[29] in the related system $(Ce_{1-x}Tb_x)Ru_2$, where $\xi$ saturated at a value of 15 Å. This constrasts with the situation found in $HoMo_6S_8$ and $ErRh_4B_4$, in which true ferromagnetic transitions occur with a concomitant reentrance to the normal conducting phase.

In all of these systems the conventional spin-depairing mechanism must either be small or some other compensating mechanisms must be operating in order for these materials to be superconductors. The question arises as to why magnetic transitions are suppressed in the $(Ce\text{-}R)Ru_2$ system. One possibility is that the ferromagnetic transition is inhibited due to the competition with the superconducting state, which can occur in two basic ways. One is via the response of the supercurrents to the internally generated field. As the range of the magnetic correlations increases the superconducting currents will respond to cancel any macroscopic magnetic field. The result is that the long-wavelength magnetic fluctuations will be suppressed because they are energetically costly leaving the short-wavelength fluctuations to dominate the spectrum. Such a competition between the electromagnetic field and the superconducting state has recently been explored theoretically by Blount and Varma.[30] Another possible effect is that the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange interaction also is modified by the superconducting state. Rather than maximizing at zero wave vector, the RKKY susceptibility vanishes at zero wave vector and has a broad peak at finite wave vectors, leading to the "cryptoferromagnetic state" proposed by Anderson and Suhl,[31] or to a "ferromagnetic spin-glass" state. We note that both of these mechanisms lead to the same qualitative effect on the magnetic state; namely that the fluctuations at long wavelengths are suppressed. The theory of Ferrell, Bhattacharjee, and Bagchi[32] incorporates both of these effects in calculating the wave-vector-dependent susceptibility $\chi(\vec{q})$. For the present case of $(Ce_{0.73}Ho_{0.27})Ru_2$ these deviations from Ornstein-Zernike behavior, if present, occur at smaller wave vectors, probably of order $1/\xi_{max} \sim 0.01$ Å$^{-1}$, which is not within the range of our experimental observations. Such effects, however, have been recently observed[33] in $ErRh_4B_4$.

Another possibility is that the concentration of magnetic ions is below the percolation threshold and therefore too small to support a magnetic state. In this case the magnetic correlations would increase with decreasing temperature, but $\xi$ would be bounded by the finite size of the magnetic clusters. In the $CeRu_2$ lattice the number of nearest-neighbor rare-earth sites is four, so that for nearest-neighbor exchange only the percolation concentration[34] is 0.425. The actual exchange interaction is expected to be considerably longer ranged than nearest neighbor, but it is possible that the range may not be sufficient to achieve long-range magnetic order.

The inelastic scattering results demonstrate that the crystal field is of paramount importance in determining the magnetic properties of these materials at low temperatures since the spacing of the crystal-field levels is large in comparison with the magnetic energy. The development of magnetic correlations at low temperatures therefore occurs between ions in triplet $\Gamma_5$ ground states, which split isotropically in a magnetic field and thus behave as effective $S=1$ states.

One unresolved problem concerns the origin of the inelastic scattering observed at low energies and low temperatures. There are two likely possibilities. This scattering could be due to exchange broadening within the $\Gamma_5$ states as the ferromagnetic correlations develop. Relatively well defined collective excitations would be expected to propagate in the system if their wavelength were short in comparison with the correlation length, and the time variation is short compared to the spin relaxation time. Indeed the correlation range becomes quite long (80 Å) at low temperatures and Mössbauer studies show the development of hyperfine splitting. An alternate possibility is that the ground state is split due to the lowering of the symmetry of the local crystal field due to the substitutional disorder on the rare-earth sites. At low temperatures, however, this should affect the intensities of all the crystal-field transitions unless there is exchange mixing of the states within the $\Gamma_5$ manifold of the same order of magnitude as the splitting itself. Either possibility implies that the exchange energy is considerably larger than would be inferred from the "transition temperature" of 0.5 K.

There are at least two other questions that still remain to be answered. One is to establish the values of the crystal-field parameters, and to see if these are sensitive to the Ho concentration. This might also shed light on the question of whether the low-energy scattering is related in any way to crystal-field effects. Another is to determine how varying the Ho concentration alters the development of the magnetic correlations at low temperatures, and in particular to see if the system establishes long-range ferromagnetic order at higher Ho concentrations where the superconducting state is suppressed. Work in this direction is progressing.[27]

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