# Magnetic response function of the itinerant ferromagnet CeFe$_2$

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Neutron inelastic scattering experiments on single crystals of the itinerant ferromagnet CeFe$_2$ show that there is a strong competition between the ferromagnetic ground state and an antiferromagnetic (AF) ground state with the wave vector $\mathbf{q}=[\frac{1}{2}\frac{1}{2}\frac{1}{2}]$. The ferromagnetic spin wave has a small temperature-independent gap of 0.25 meV and a reduced (compared to other rare-earth Fe$_2$ Laves phases) stiffness constant of $D = 155(5)$ meV Å$^2$. The strong fluctuations around the AF wave vector give rise to an AF spin-wave dispersion relationship that can be followed across the reduced AF Brillouin zone. The gap in the AF excitation spectrum is $\sim$1 meV at 15 K and rises to $\sim$5 meV above 100 K. At low temperature with a window of $\pm20$ GHz, we observe an apparent static AF component of $\sim0.05\mu_B$ superimposed on the ferromagnetic component of $1.2\mu_B$ per Fe atom. The spatial correlations of these AF fluctuations extend over many unit cells at low temperature. Our measurements have not detected any response directly from the Ce moments; so we assume that their response is spread over a wide energy range. Mössbauer spectra show an anomalous behavior of the Lamb-Mössbauer factor as a function of temperature and also show that the magnetic system is not saturated even at large (5 T) fields, suggesting that short-range AF order may persist to higher temperatures than the medium-range order observed in the neutron experiments. [S0163-1829(98)09241-8]

## I. INTRODUCTION

The numerous studies of the magnetic excitations in the heavy $R$Fe$_2$ ($R$=rare earth) compounds with cubic Laves phase structure have provided important information on interactions between the localized rare-earth $4f$ and itinerant $3d$ electrons of Fe.[1,2] By far the strongest interaction is the ferromagnetic one between the nearest-neighbor Fe atoms, which have a separation almost identical to that in elemental Fe. Thus YFe$_2$ has as large a Curie temperature (545 K) as the heavy $R$Fe$_2$ compounds, and the main influence of the rare earth is to introduce anisotropy, as would be expected.[3,4] Since many of these compounds have practical applications,[3] the early interest was in the easy directions of magnetization, which can frequently change as a function of temperature due to complex nature of the RE single-ion anisotropy.[5] This early work[5] already noted the unusual situation in CeFe$_2$ ($a_0=7.304$ Å, ferromagnetic $T_C\sim230$ K), but it was not until Ericksson *et al.*[6] suggested that the Ce $4f$ electrons would be *itinerant* in this compound, and hence quite different from the *localized* $4f$ electrons associated with other $R$Fe$_2$ compounds, that it became of more general interest. A major question has become the values of the moments (both $4f$ and $5d$) associated with Ce, and experiments to probe

this quantity involving dichroism[7–10] (on both the Ce $L$ and $M$ edges) as well as polarized-neutron[11] and Compton[12] scattering have been reported. A consensus emerges that the net moment on the Ce site of $-0.15\mu_B$ is mostly spin and that it is polarized opposite to the Fe moment of $1.2\mu_B$. Although the magnitude is smaller than that of $-0.50\mu_B$ proposed by theory,[6,13] the general physics of the itinerant nature of the $4f$ states in CeFe$_2$ is accepted. An implicit assumption is that there may be considerable hybridization between the itinerant Fe $3d$ and Ce $4f$ states. The consequences of this hybridization remain to be elucidated.

Additional work was stimulated by the experiments of Roy and Coles in the late 1980s, that a small amount of doping of another metal into the Fe site would induce a stable antiferromagnetic ground state (rather than the ferromagnetism of CeFe$_2$) at low temperature.[14,15] In neutron-diffraction experiments on polycrystalline samples of CeFe$_2$ doped with Al, Co, and Ru, Kennedy *et al.*[16,17] showed that the antiferromagnetic (AF) structure (stable at the lowest temperatures) had a wave vector of $\mathbf{q}=[\frac{1}{2}\frac{1}{2}\frac{1}{2}]$. These authors also reported, albeit briefly, that AF reflections of the same type were present in their neutron powder pattern[17] below $\sim$60 K in the parent compound CeFe$_2$, but the magnitude of the moment was too small for an accurate determination,

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although they estimated $\sim 0.15 \mu_B$ per Fe atom. They did not address the question of how AF reflections can appear in the context of an apparently stable ferromagnetic ground state. This was followed by a report[18] on ac susceptibility measurements on $CeFe_2$. Despite the observation of anomalies below $\sim 80$ K, the authors concluded that there was no AF phase at low temperature. However, work on the doped systems [of Al (Ref. 19), Ru (Refs. 20 and 21), and Re and Ir (Ref. 22)] shows the prevalence of a stable AF ground state at low temperature, and there is also a report that in the Ru system the AF phase may be suppressed with a magnetic field; this leads to an appreciable magnetoresistance.[21]

An obvious question to ask about pure $CeFe_2$ is whether the hybridization between the itinerant $4f$ and $3d$ electron states is responsible for the AF interactions between the Fe-Fe nearest neighbors. This might explain the apparent instability of $CeFe_2$ at low temperature and the ease with which stable AF is found with a small change in electron concentration. Unfortunately, we know of no theoretical treatments of the generalized susceptibility of $CeFe_2$, and one of the objects of our present study is to motivate such studies.

Our own interest in $CeFe_2$ actually came from yet another direction. We have recently reported on the low-energy spin-wave spectra of the isostructural compound $UFe_2$, in which the $5f$ electrons are also *itinerant*.[23] Briefly, in $UFe_2$, we were unable to find any sign of the U $5f$ moment (which is known to be very small) in the inelastic response, but we did observe a considerable enhancement of the Fe spin-wave stiffness constant $D$. This latter is defined in the conventional way where the Fe spin-wave energy at small wave vector $\mathbf{q}$ is given by $E = \Delta E_F + Dq^2 + \cdots$, where $\Delta E_F$ is the ferromagnetic gap and we neglect higher-order terms. Surprisingly, in view of the fact that $UFe_2$ orders ferromagnetically at only 165 K, $D$ is even higher than found in pure iron, which of course orders near 1000 K. We ascribe this enhancement of the Fe-Fe nearest-neighbor ferromagnetic interactions to the hybridization between the U $5f$ and Fe $3d$ states. Thus we had considerable interest in extending our neutron inelastic studies to $CeFe_2$, as it promises to be a second isostructural system in which the $f$ electrons are itinerant.

## II. SAMPLE PREPARATION AND EXPERIMENTAL DETAILS

The compound $CeFe_2$ melts incongruently at 925 °C, and a stoichiometric melt is only fully liquid for temperatures above $\sim 1200$ °C. This paratectic formation makes synthesis of sizable single crystals via the Bridgman growth, Czochralski growth, or zone refining extremely difficult, if not impossible. On the other hand, sizable crystals can be grown from a binary melt using excess Ce as the solvent.[24] Using this technique, single crystals of up to 5.2 g were grown at Ames Laboratory out of a Ce-rich binary melt. The crystals are highly faceted and consist of a number of excellent crystals, but with an overall mosaic of $\sim 1°$. As usual with Ce compounds, the samples are sensitive to oxygen and the samples were kept at all times in either a glove box or in a sealed (with He exchange gas) can. However, a more important sensitivity of the large crystal was found to thermal shock.
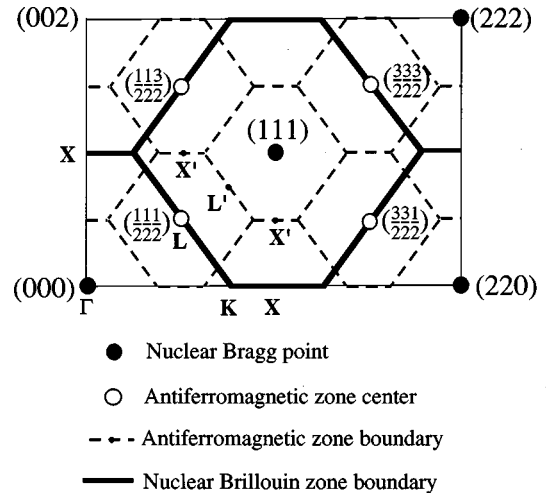


FIG. 1. $(1\bar{1}0)$ plane projection as used for the experiment. The solid points and thick solid lines give the allowed nuclear reflections and ferromagnetic Brillouin zones (BZ), respectively. [Note that the (002) is forbidden in the Laves phase symmetry.] Also shown as open circles and dashed lines are the antiferromagnetic zone centers and their BZ, respectively.

Thus, after our first experiments at low temperature, the displex was warmed fairly rapidly ($\sim 1$ h) to room temperature and the largest crystal was found to have disintegrated into many small grains. A neutron powder pattern showed that the sample was pure $CeFe_2$ with the correct lattice parameter and there was no sign of lines associated with $CeFe_2$. Fortunately, more crystals could be fabricated and these have not (yet) been destroyed because all subsequent experiments have employed slow cooling and heating. This disintegration does not occur for small pieces and is probably more a function of the growth morphology than the compound itself.

The neutron inelastic scattering experiments have been performed on the IN8 (thermal) and IN12 (cold) triple-axis spectrometers at the Institut Laue Langevin, Grenoble, and also with the 1T1 (thermal) and 4F2 (cold) machines at the Laboratoire Leon Brillouin at Saclay. In all cases focusing graphite monochromators have been used with graphite filters to suppress higher-order contamination. An initial study of the increase of the intensity at the (111) reflection verified that $T_C$ is 235 K, which is the accepted value for stoichiometric $CeFe_2$.

The $^{57}Fe$ Mössbauer measurements have been carried out using a $^{57}Co/Rh$ source and a powdered single-crystal absorber containing about 5 mg $Fe/cm^2$. The spectra have been recorded with the $CeFe_2$ absorber maintained at different temperatures between 4.2 and 300 K in either zero or external fields up to 8 T applied parallel to the $\gamma$-ray direction.

## III. RESULTS

### A. Overview of inelastic response at low temperature

We show in Fig. 1 the Brillouin zones (BZ) in the $(1\bar{1}0)$ plane. The thick solid lines define the ferromagnetic BZ. For the acoustic spin-wave excitations near the BZ centers, corresponding to $\mathbf{q}$ (the reduced wave vector from the BZ origin) small, the structure factors will correspond to those for elastic scattering at the zone center. The rele-
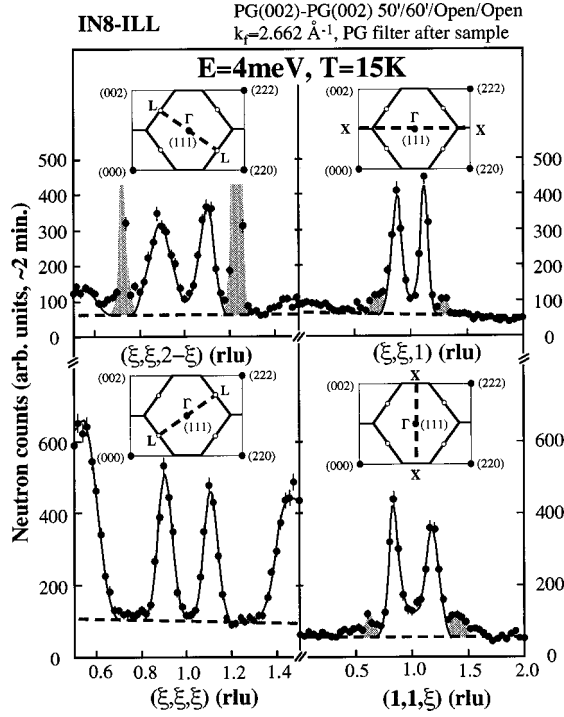
FIG. 2. Constant-$E$ scans at an energy transfer of 4 meV at $T$ = 15 K in different directions around the (111) ferromagnetic zone center. The directions of the scans are marked in the insets. The center of each scan $\xi = 1$ is the (111) ($\Gamma$) point, and the two peaks close to that in $\xi$ are the Stokes and anti-Stokes components of the ferromagnetic Fe spin wave. The slightly different intensities and shapes arise from the resolution of the instrument. The shaded peaks correspond to phonons. Note the strong antiferromagnetic fluctuations present around the $L$ points (left-hand panels) and its absence around $X$ in the right-hand panels. The lines are guides to the eye. Data taken on IN8 (ILL) with PG(002)→PG(002), $k_f = 2.662$ Å$^{-1}$, and collimation 50′/60′/open/open.

vant structure factors are $F(000) = \mu_{Ce} + 2\mu_{Fe}$, $F(002)$ = 0, $F(111) = \mu_{Fe} - \mu_{Ce}/(2^{1/2})$, $F(220) = \mu_{Ce}$, and $F(222)$ = $2\mu_{Fe}$. From these relationships, which have been well exploited in past studies,[1,2,23] we can determine whether both or only one of the moments is contributing to the dynamic response. Our studies of CeFe$_2$ show that the *entire* inelastic signal reported in this paper arises from the iron moments. For example, no acoustic spin waves are observed around the (220) zone center. This is exactly the same situation as was found in the study of UFe$_2$.[23] It does not mean that no moment is associated with Ce, but the most probable explanation is that the Ce inelastic response is spread over a wide energy range, as is found,[25] for example, in CeNi$_2$, and such a response is hard to establish with a triple-axis spectrometer as used in the present experiments.

We start by showing in Fig. 2 constant-$E$ scans ($E = 4$ meV) in the (111) BZ in different directions at the lowest temperature $T = 15$ K. The central signal around $\xi = 1$ in each case represents the ferromagnetic spin wave, which propagates from the ferromagnetic zone center ($\xi = 1$ or $q = 0$), and this cone of the ferromagnetic response function is thus cut in both the Stokes and anti-Stokes directions for a constant energy transfer of 4 meV. The slightly different intensities and peak shapes registered are a consequence of the spectrometer resolution. What is also clear is that a second
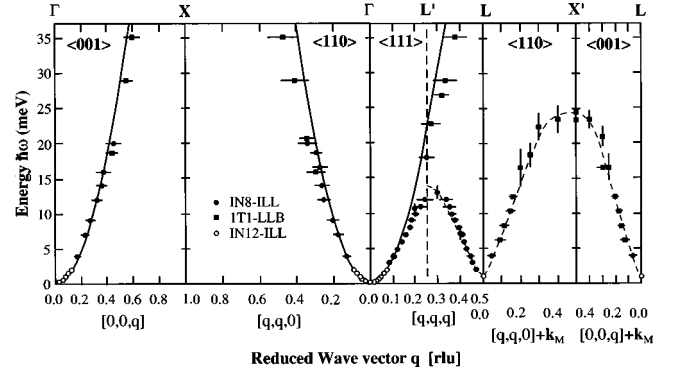


FIG. 3. Dispersion curves measured at low temperature (15 K) in the three principal directions. For the ferromagnetic spin wave (originating from $\Gamma$), the quadratic relation fitted at low $\mathbf{q}$ is indicated by solid lines. The dashed lines indicate the antiferromagnetic fluctuations, which originate at the point $L = \mathbf{k}_M = [\frac{1}{2}\frac{1}{2}\frac{1}{2}]$ in the BZ.

mode is centered at the $L$ points of the BZ and appears much stronger in the ($\xi,\xi,\xi$) (bottom left panel) than in the ($\xi,\xi,2-\xi$) (top left panel) direction. As we shall see, this is the *antiferromagnetic* fluctuation spectrum and structure factor arguments show that it arises only from the Fe moments.

The total dispersion curves for the magnetic response are given in Fig. 3. The ferromagnetic spin waves are similar to those observed in other $R$Fe$_2$, except that the spin-wave stiffness is substantially reduced, but we observe also *antiferromagnetic* spin waves propagating from the $L$ points in the BZ. One of the surprising features of these dispersion curves is that there appears very little, if any, interference between the two. Unfortunately, at the one point in the diagram, close to the point $L'$ where the two spin waves may be observed together, the AF spin waves have little intensity, and so this question of interference is difficult to answer experimentally.

More details of the AF spin waves at low temperature are shown in Fig. 4 in which constant-$E$ scans are shown around the $L$ points, but in two different directions. These show that the spin waves are indeed well defined. In fact, at the lowest temperature both magnetic response functions appear to be resolution limited in energy space, which signifies that there is little damping of the response.

## B. Temperature dependence

An overview of the total magnetic response across the BZ in the [111] direction as a function of temperature is shown in Fig. 5. As the temperature is raised to 200 K ($T_C - 35$ K), the Bose factor (at an energy transfer of 4 meV) leads to a substantial increase of ~4.7 in observed intensity of the ferromagnetic spin wave, but there is little change in its position. On the other hand, the AF fluctuations (around $L$) clearly do not follow the Bose factor, which implies that the $\chi(\mathbf{Q},\omega,T)$ reduces as a function of $T$ at this wave vector and energy, and they also shift their position. On further warming to 300 K ($T_C + 65$ K), the ferromagnetic response renormalizes and spreads over a wider energy range. The ferromagnetic spin waves at higher energy are shown as a function of temperature in Fig. 6. There is only a small shift of the energy of the ferromagnetic spin wave over the whole temperature range; however, it clearly becomes considerably broader
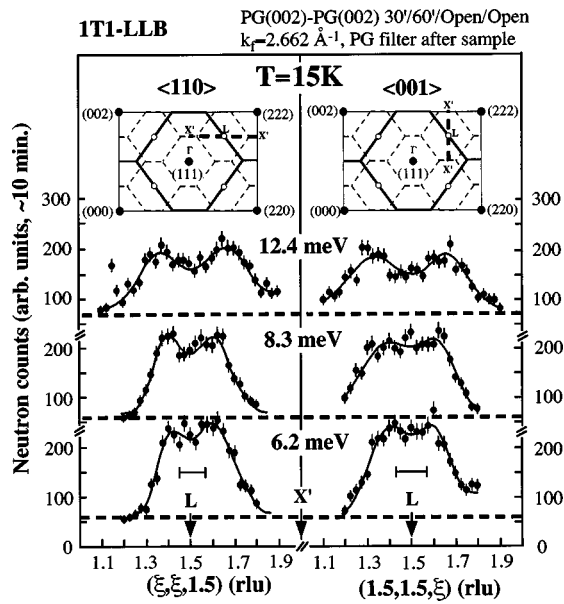
FIG. 4. Details of the AF spin waves around the $L$ point $(\frac{3}{2}\frac{3}{2}\frac{3}{2})$ taken with constant-$E$ scans. The horizontal bar shows the experimental resolution. The data were taken on 1T1 at LLB with the configuration PG(002)→PG(002) with collimation 30'/60'/open/open and $k_f = 2.662$ Å$^{-1}$.

above $T_C$. It is then difficult to separate the ferromagnetic and AF responses in the zone.

## IV. DISCUSSION OF NEUTRON RESULTS

We start by returning to the point made earlier that the *entire* spectral response found in these experiments arises from the Fe moments. This may be seen most clearly by the absence of any signal around the (220) nuclear zone center, for which the elastic structure factor arises from the Ce mo-
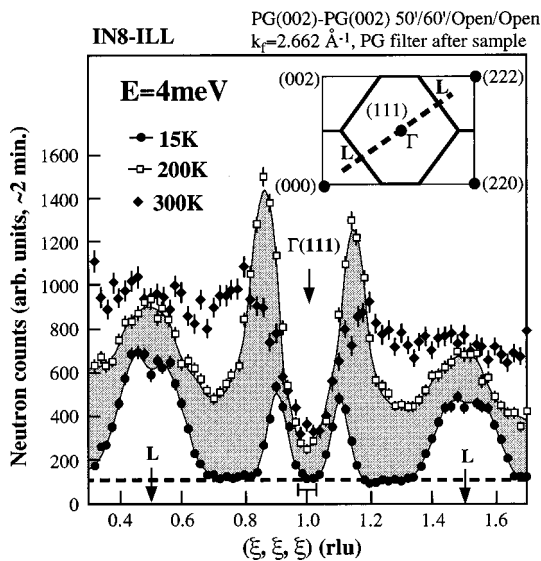


FIG. 5. Constant-$E$ scans ($E = 4$ meV) as a function of temperature across the zone in the longitudinal direction as shown in the inset. All the scattering is magnetic; i.e., no phonons are seen in this configuration. The shading is to help to distinguish the different contributions. Spectrometer configuration as in Fig. 2.
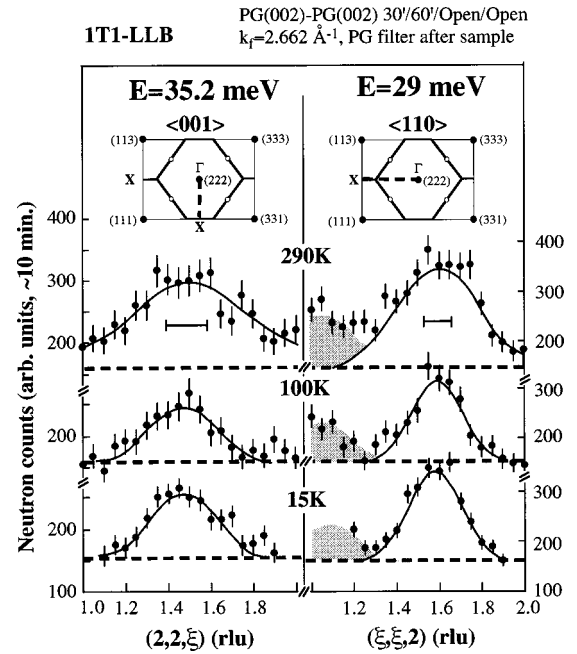


FIG. 6. Details of the ferromagnetic spin wave at high energy as a function of temperature. The horizontal bar shows the experimental resolution, and the shaded part is an estimated contribution from phonons. The spectrometer configuration was the same as described in Fig. 4. The lines are Gaussian fits to the spectra.

ments only. Moreover, the crystal-field-like mode that is found in studies of $R$Fe$_2$ with heavy rare earths[1] is most easily identified around the (002) zone center, for which the acoustic modes are forbidden as the elastic structure factor is zero, and here also we have found no measurable inelastic intensity. For the AF spin waves we find that the intensities follow closely those expected from an iron-only contribution. A good example is shown in the two left-hand panels of Fig. 2. In the lower panel the scans trace a line in the BZ that signifies a maximum contribution of the Fe moments, whereas the line traced between $L$ points in the upper panel connects BZ with zero Fe contribution to the acoustic modes, so that the intensity is much reduced compared to that observed in the lower panel. We have similar considerations throughout all Brillouin zones. Of course, at higher energies we would expect optic modes involving Fe, and perhaps also Ce, moments, but since these have not been mapped out even for the heavy $R$Fe$_2$ compounds, in which the rare-earth moments are much larger, they are beyond our present capability. The nature of the Ce inelastic response remains unanswered by our experiments. This is the same situation as found for UFe$_2$ and probably signifies that the Ce (and U) response is spread over a wide range of both wave vector and energy, much like is found[25] in CeNi$_2$, and this is especially difficult to detect with a triple-axis spectrometer. Even with a time-of-flight experiment on a polycrystalline sample, as performed in the CeNi$_2$ studies, the difficulty would be in detecting a presumably weak signal in the presence of the strong dispersive scattering from the Fe moment sublattice.

At low temperature the analysis of the low-energy and wave vector part of the spectral response is shown in Fig. 7. Figure 7(a) indicates that the ferromagnetic spin-wave stiffness constant of 155(5) meV Å$^2$ is substantially reduced
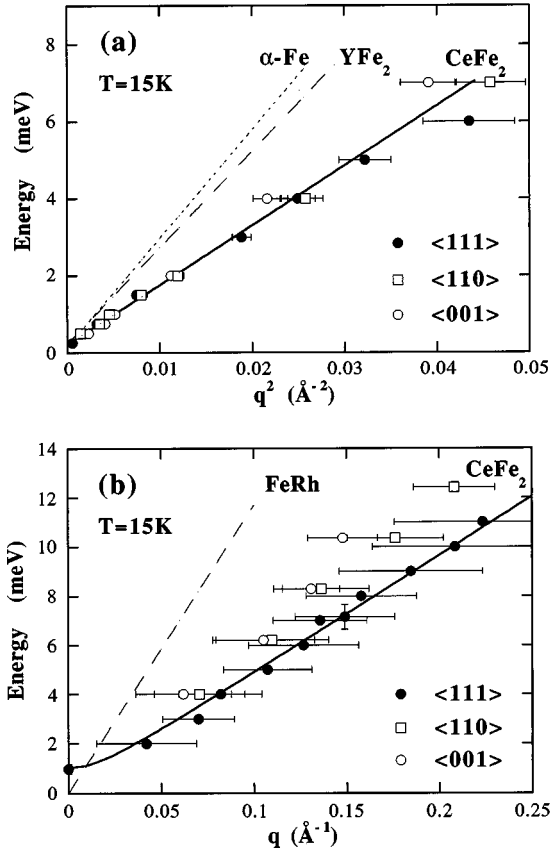
FIG. 7. (a) Experimental data for the ferromagnetic spin waves from different directions fitted (solid line) to a quadratic dispersion relationship $E = \Delta E_F + Dq^2$, where $\Delta E_F$ is the spin-wave gap and $D$ is the spin-wave stiffness. The value for $D$ is 155(5) meV Å$^2$. Also shown are the slopes for elemental $\alpha$-Fe and YFe$_2$. (b) Experimental data for the antiferromagnetic spin wave from different directions fitted (solid line) to the relationship $E^2 = (\Delta E_{AF})^2 + (Gq)^2$. The value of $G = 49(5)$ meV Å. Also shown as a dashed line is the relationship (with $G = 115$ meV Å) found in FeRh (Ref. 26).

from that of 250 meV Å$^2$ found in isostructural YFe$_2$. Such a reduction is consistent with the reduced $T_C$ of CeFe$_2$ compared to the other $R$Fe$_2$ materials. As a function of temperature, at least up to $\sim 180$ K, there is little change in the ferromagnetic response (see Figs. 5 and 6) so that $D$ is essentially independent of $T$. We recall here that this is a completely different situation from that found[23] in UFe$_2$ in which $D$ was found *increased* at low temperature (to as high as $\sim 450$ meV Å$^2$), but was also found to be $T$ dependent. If the changes in the ferromagnetic spin-wave constant may be ascribed to the hybridization between the itinerant $f$ (of the rare-earth or uranium atoms) and the Fe $3d$ states, then the effects are quite different depending on whether we are considering $4f$ (in the case of Ce) or $5f$ (in the case of uranium) electrons hybridizing with the $3d$ band. Of course, whereas these are both $f$ electrons being hybridized with the $3d$ band, their spatial extent is different. Moreover, there is normally one $4f$ electron associated with cerium, whereas three $5f$ electrons are associated with uranium. Although the $D$ values for YFe$_2$, CeFe$_2$, and UFe$_2$ are all different, their ferromagnetic gaps ($\Delta E_F$) in the spin-wave spectrum are all very

much the same [0.21(5), 0.25(3), and 0.40(5) meV, respectively] and this is consistent with little anisotropy introduced from the RE or uranium sublattice. For comparison, recall that the gap in pure Fe is $\sim 0.1$ meV. As expected, this is quite different when the RE atom carries a large moment (as in the case of Tb or Ho), in which case $\Delta E_F$ can be as large as 10 meV.[2]

The dispersion of the AF spin waves at low temperature and small $q$ is shown in Fig. 7(b). The gap ($\Delta E_{AF}$) at low temperature is 1.0(1) meV, and in the relationship $E^2 = (\Delta E_{AF})^2 + (Gq)^2$ the value of $G = 49(5)$ meV Å. We have found one material, FeRh, in which neutron experiments[26] report a phase transition from AF at low temperature to ferromagnetism at $\sim 350$ K and then to paramagnetism at $\sim 700$ K. This situation is not the same, of course, as in CeFe$_2$, in which both AF and ferromagnetic fluctuations exist *simultaneously*, but it does give us some guidance as the excitations involve principally iron. The AF spin-wave stiffness $G = 116(3)$ meV Å and is shown in Fig. 7 as a dashed line.

We now turn to the temperature dependence of the effects, especially the AF response. Recall that the ferromagnetic spin wave shows little change (Figs. 5 and 6) up to 200 K (0.85$T_C$), and even above $T_C$ (Fig. 6) this spin-wave response is still present at high energies, albeit damped. The situation is different for the AF response. Details of this are shown in Fig. 8. In Fig. 8(a) we show the gaps $\Delta E_F$ and $\Delta E_{AF}$ as a function of temperature. $\Delta E_F$ is independent of temperature, whereas $\Delta E_{AF}$ is strongly $T$ dependent until about 150 K and then becomes difficult to measure, but is certainly larger than $\sim 4$ meV. Another way to represent the AF response is to construct the function $\chi''(\mathbf{Q}, \omega, T)$ as a function of temperature at the AF BZ center, here chosen as the $L$ point $\mathbf{Q} = (\frac{3}{2}\frac{3}{2}\frac{3}{2})$. These points are determined by taking the neutron counts, subtracting a background, and dividing by the Bose factor $\{1 - \exp(-E/kT)\}^{-1}$. Although at higher energy, $E \sim 10$ meV and above, the phonon response becomes important and is difficult to subtract, the essential physics of how the response function sharpens and lowers in frequency as the temperature is decreased can be seen from this figure.

The presentation of $\chi''(\mathbf{Q}, \omega, T)$ for the AF fluctuations allows one a relatively clear picture of the temporal aspect of the AF fluctuations, but not of their spatial extent. To examine these more closely we have used the 4F cold-source triple-axis spectrometer at the LLB and examined the *elastic* scattering as a function of temperature. To define this in more detail, we note that the window of acceptance in energy space of the spectrometer (used with $k_f = 1.55$ Å$^{-1}$) is $\sim 150$ $\mu$eV full width at half maximum (FWHM) or, equivalently, a frequency window of $\pm 20$ GHz. The results of this experiment are shown in Fig. 9. This figure shows two important features of the AF fluctuations. The first is that within the resolution of the instrument we do see what appears to be a stable AF component. The finite resolution of the instrument must always be borne in mind. Certainly, with better energy resolution we would see a different temperature dependence. With no energy resolution, as, for example, in the powder experiments reported by Kennedy and Coles,[17] a very small AF component may be visible at a relatively high temperature. In brief, since we believe the AF fluctuations are tem-
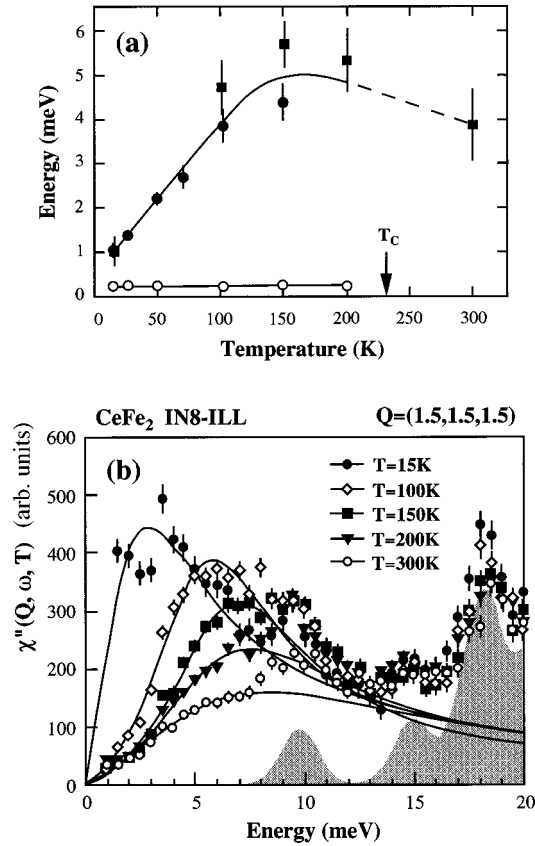
FIG. 8. (a) Ferromagnetic ($\Delta E_F$, open circles) and antiferromagnetic ($\Delta E_{AF}$, solid circles and squares) gaps as a function of temperature. The results for $\Delta E_{AF}$ are shown for two AF BZ centers, circles $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$ and squares $\mathbf{Q} = (\frac{3}{2}\frac{3}{2}\frac{3}{2})$. (b) $\chi''(\mathbf{Q}, \omega, T)$ as a function of energy at $\mathbf{Q} = (\frac{3}{2}\frac{3}{2}\frac{3}{2})$ for different temperatures. The solid hatched areas represent the dynamical response from the phonons at this $\mathbf{Q}$ and must be subtracted from the experimental points to get the true magnetic response function.

poral rather than static in nature, their effects will be seen differently by different techniques, depending on their temporal resolution.

We can make an estimate of the magnitude of the magnetic moment responsible for the peak at low temperature as shown in Fig. 9. Although there is considerable uncertainty associated with knowing how to calibrate this intensity (as the intensities of the Bragg peaks from the crystal certainty suffer from extinction), we estimate a moment of $\sim 0.05 \mu_B$ associated with the AF fluctuations of the Fe atoms.

The spatial correlations are also interesting and may be derived from the data shown in the inset. Here we show the peak intensity and the experimental width of the peak. The latter must be deconvoluted with the resolution of the instrument (also shown) and is then directly proportional to $1/\zeta$, where $\zeta$ is the real-space correlation length. The correlations of the AF fluctuations vary from $\sim 400$ Å at $T < 25$ K to about half this value at 60 K, at which temperature the temporal aspect of the fluctuations is such that they are outside the energy window of the spectrometer. These scans have been performed perpendicular to $\mathbf{Q}$, where the resolution is normally the best. In the longitudinal direction the instrumental resolution is not sufficient to determine any experimental broadening; however, anisotropy in the correlation
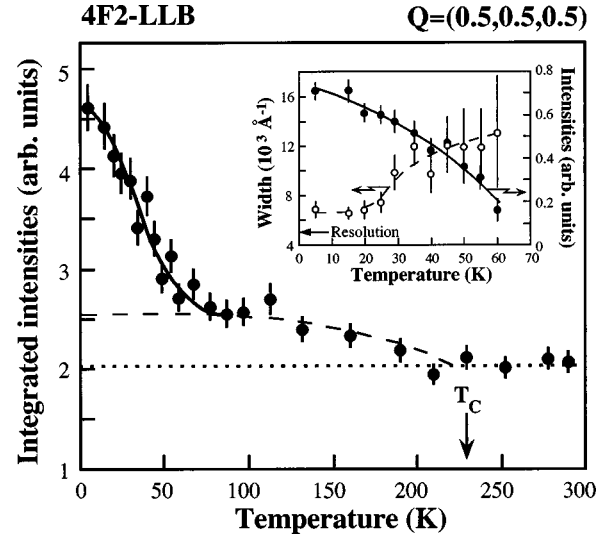


FIG. 9. Integrated intensities of the signal at $\mathbf{Q} = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$ observed with the 4F2 cold-source triple-axis spectrometer at LLB ($k_f = 1.55$ Å$^{-1}$). The integration was performed by rocking the crystal through the Bragg position (transverse scans). The dotted line is the contribution from $\lambda/2$ from the nuclear (111) reflection, and the additional contribution to this reflection from the ferromagnetic ordering at the (111) position is shown as a dashed line. The additional magnetic scattering is shown as the experimental points, with the solid line as a guide to the eye. The inset shows the *peak* intensity (solid circles and line) together with the FWHM of the transverse scans as a function of temperature.

lengths would not be expected on the basis of the cubic symmetry of CeFe$_2$ and the isotropic dispersion curves (Fig. 3). Such a range for the AF correlations, over many lattice sites, demonstrates the unusual nature of the magnetic ground state in CeFe$_2$.

## V. MÖSSBAUER EFFECT EXPERIMENTS

From the temperature dependence of the spectra (quadrupole doublet in the paramagnetic state and magnetically split sextet in the ordered state), we confirm that CeFe$_2$ orders at $T_C = 230(2)$ K (Fig. 10). More interesting is the observation of an anomalous behavior of the recoil-free fraction (Lamb-Mössbauer $f$ factor), proportional to the area of the spectra. As observed previously for UFe$_2$,[27] the $f$ factor abruptly increases below $T_C$, whereas it exhibits a linear dependence above (Fig. 11). From the slope of the linear part, one deduces a Debye temperature $\theta_D$ of 323 K very close to the one (325 K) reported for UFe$_2$.[27] The unusual trend of the $f$ factor below $T_C$, which implies that the mean-square displacement of the Fe atoms decreases, can possibly be ascribed to the different Fe-Fe exchange interactions in UFe$_2$ and CeFe$_2$ than found in standard $R$Fe$_2$ compounds.[23]

The shape of the spectra, related to the number and population of the magnetically inequivalent Fe sites, is usually considered as a fingerprint of the direction of the iron easy axis of magnetization.[28] This led to the claim that the easy direction of the iron moment in CeFe$_2$ varied from parallel to [100] below 150 K to a direction [$uuw$], which makes an angle of about 20° with respect to [100], above 150 K. However, the analysis of our high quality data (Fig. 10) indicates
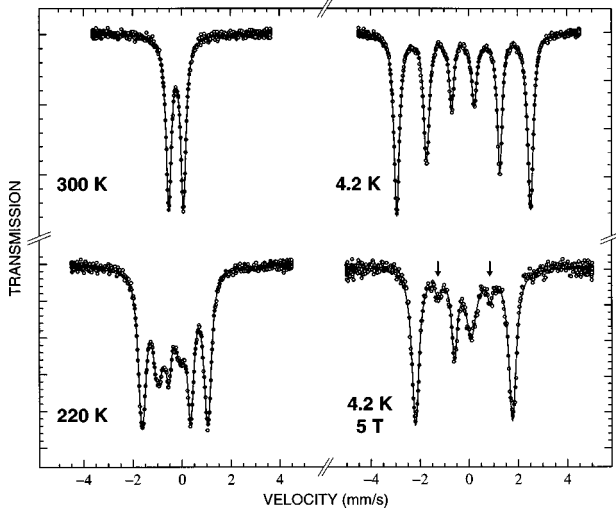
FIG. 10. $^{57}$Fe Mössbauer spectra of CeFe$_2$ at different temperatures and in an external field of 5 T at 4.2 K. The quadrupole splitting was negative ($e^2qQ \approx -1.19$ mm/s), and the average saturation hyperfine field $H_{hf}$ was 16 T. Note that the intermediate lines (arrows) do not vanish at high fields.

that the magnetic configuration of CeFe$_2$ is more complex. Indeed, the observation of asymmetric spectra with inhomogeneous line broadening down to 4.2 K rules out a [100] easy direction. Moreover, fits implying four magnetically inequivalent iron site (this is the situation for a ferromagnet with Fe moments pointing along an arbitrary [$uvw$] direction) could not reproduce the experimental data.

To shed more light on the actual spin configuration, we performed additional experiments by applying an external magnetic field. Due to the weak magnetic anisotropy of CeFe$_2$, as revealed by the small gap in the ferromagnetic spectrum and single-crystal magnetization data,[29] we anticipate that a moderate field (>0.2 T) will align the Fe moments along the field direction and lead to an extinction of the two intermediate lines of the magnetically split sextet.[30,31] This was not the case as Fig. 10 shows. The intermediate lines persist to at least 150 K in an applied field as large as 5 T. This clearly shows that the hyperfine field ($H_{hf}$) does not align with the applied external field. Although an-
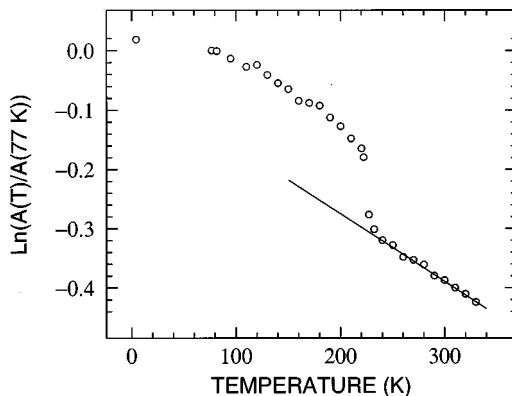


FIG. 11. Temperature dependence of the normalized (at 77 K) spectra are in CeFe$_2$. Notice the abrupt change of the Lamb-Mössbauer factor at $T_C$. The Debye temperature of 323 K was deduced from a fit of the linear part above $T_C$.

isotropic contributions to $H_{hf}$ (dipolar fields of less than 1 T) could lead to a small misalignment of $H_{hf}$ with respect to the iron moment direction,[32] they can hardly be invoked to explain the observed intensities of the intermediate Mössbauer lines. A more natural explanation would be that CeFe$_2$ is a canted ferromagnet; i.e., a small antiferromagnetic component is superimposed on a dominant ferromagnetic contribution. Note that one observes a monotonous temperature dependence of the average hyperfine field. This suggests that the AF component is present up to $T_C$.

## VI. CONCLUSIONS

Following our experiments on UFe$_2$, we anticipated some surprises in the dynamical response function of CeFe$_2$. We did not, however, expect it to be so radically different from that of UFe$_2$: that *no* signal associated specifically with either the uranium or cerium moments in these materials can be understood on the basis of a wide (in energy) response function that is difficult to detect with a triple-axis spectrometer. In CeFe$_2$ we have found a strong reduction in the ferromagnetic exchange interactions and associated antiferromagnetic fluctuations in what is nominally regarded as a ferromagnet.

The dynamics presented in Figs. 8 and 9 bears a superficial resemblance to the classic ''soft-mode phase transition,'' typified by the studies[33] of SrTiO$_3$. We imagine a ferromagnetic ground state (analogous to a stable structure A) and the development of AF fluctuations that prefer an AF ground state (analogous to structure B). The frequency of the fluctuating AF phase, if we follow the classic Landau theory, would then be expected to decrease quadratically with $T$; i.e., $E^2$ would be proportional to $T-T_0$, where $T_0$ is the temperature of the phase transition. At the same time, intensity (the central peak) appears in the elastic channel ($E=0$), its magnitude depending on a coupling constant to some unspecified relaxing degree of freedom. This is an attractive analogy, giving a qualitative (phenomenological) frame within which to relate the observations reported in Figs. 8 and 9. Unfortunately, on closer inspection, any quantitative analogy is flawed; the frequency of the soft mode (the AF fluctuations) decreases *linearly*, not quadratically (see Fig. 8), and there is evidence for a distinct change in the dynamics around ~60 K.

Our neutron measurements suggest that these AF fluctuations are *dynamic* in origin, and their observation therefore depends on the time resolution of the measuring probe. They are unusually long range in their spatial extent—at least 400 Å (i.e., >50 unit cells) at low temperature. As the temperature is raised, their spatial extent reduces and their characteristic frequency increases. At the lowest temperature a rough estimate of the amplitude of these fluctuations at the Fe site is ~0.05$\mu_B$ compared to the ferromagnetic component of 1.2$\mu_B$. Averaging over a long time, such as in the Mössbauer measurements (which have a frequency scale much lower than the neutron probe), should therefore give no signal, at least for $T>60$ K. This is not the case—effects of the AF fluctuations can be seen also in the Mössbauer spectra and even up to relatively high temperature. One way to reconcile this apparent conflict is by realizing that Mössbauer measurement is sensitive to correlations at short distances (a

few Å), whereas neutron measurements are sensitive only to correlations that give reasonable ''peaks'' in reciprocal space. In the neutron case the correlation length must be of the order of 50 Å to observe such a peak. At a given temperature this implies that for longer real-space correlations there is a correspondingly higher characteristic frequency. As the temperature is raised, a given real-space correlation length naturally develops a higher characteristic frequency, but for the shortest Fe-Fe distance there is still enough correlation for a signal to fall in the relatively small frequency window of the Mössbauer measurements. It would be interesting to explore this relationship between real-space correlation length and frequency with high-resolution neutron spectroscopy.

The wave vector of these AF fluctuations is $\mathbf{q}=[\frac{1}{2}\frac{1}{2}\frac{1}{2}]$, and this is exactly the AF wave vector found in many compounds in which Fe is replaced by another metal.[19–22] It corresponds to a doubling of the unit cell along the $\langle 111 \rangle$ direction. This suggests that there is an electronic instability in the $\chi''(\mathbf{q},\omega)$ of CeFe$_2$ and that a small change in the electron concentration leads to a stable AF ground state. Since these effects are driven by a small amount of Ru doping, and Fe and Ru are nominally isoelectronic, it is not simply a question of elec-

tron concentration, but must include the effects of doping with electrons of a different spatial extent. No doubt this aspect is sensitive to the mixing between the $3d$ and Ce $4f$-$5d$ band, but the full details need to be worked out in more detail before our observations can be put into a proper context. It would naturally be interesting to perform similar experiments as described in this paper on doped CeFe$_2$ to determine whether ferromagnetic fluctuations remained in the stable AF state. The presence of an electronic instability is further suggested by the observation of giant magnetoresistance effects in the Ru-doped materials.[21]

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