Iron spin waves in YFe_2 and UFe_2

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We report the results of neutron inelastic scattering measurements on single crystals of YFe_2 and UFe_2 with special emphasis on the energy region above ~7 meV, which was not reported in our earlier work on UFe_2. Iron spin waves have been observed up to 55 meV in YFe_2, but in UFe_2 such modes become broad at energies above ~35 meV. The most surprising aspects in UFe_2 are the absence of any observable mode involving the U $5f$ electrons, and the strong Fe-Fe interactions as judged by the dispersion of the Fe only mode. Since the spectrum in YFe_2 can be considered as representing ''diluted'' Fe in the cubic Laves phase structure, it is particularly useful to compare the results for UFe_2 and YFe_2. In addition to the increased Fe-Fe interaction (resulting in a larger spin-wave stiffness than found in pure Fe), there is evidence that the Fe spin wave in UFe_2 interacts with a Λ_1 optic phonon mode in the $\langle 111 \rangle$ direction. This phonon represents a breathing between the U and Fe sublattices, and we suggest that it is mediated by the hybridization between the U $5f$ and Fe $3d$ electrons. [S0163-1829(99)00510-X]

I. INTRODUCTION

Following extensive investigation of the heavy rare-earth (RE)Fe_2 compounds (fcc Laves phase crystal structure) some 15–20 years ago,[1] there has been a renewal of interest in the isostructural ferromagnets with cerium and uranium stimulated by theoretical advances[2,3] predicting how the spin and orbital moments will change as the itinerant f electrons hybridize with the $3d$ electrons of Fe. Neutron form factor, x-ray magnetic circular dichroism, and magnetic x-ray Compton scattering experiments have demonstrated the importance of these hybridization effects in the *static* ground-state properties of both $CeFe_2$ (Ref. 4) and UFe_2.[5] Our interest in this work is to investigate the effects of hybridization on the *dynamical* response function of these two unusual materials. This subject has not yet been addressed by theory, but we hope that our measurements will provide motivation for such efforts.

In our first publication using inelastic neutron scattering we showed that a number of unusual effects are present in the spin wave response of UFe_2.[6] The two most important are that *no* response is seen from the uranium spins, and that the Fe spin-wave response is substantially ''hardened'' at low temperature when compared to that of pure iron (i.e., at low temperature the spin-wave stiffness parameter is actually greater than that of pure iron, although T_c is considerably lower in UFe_2). We have recently completed similar neutron inelastic measurements on single crystals of $CeFe_2$.[7] Of the two points mentioned above, the first is common to the results from $CeFe_2$, i.e., no response is seen from the Ce spins, but, in contrast to the situation in UFe_2, the spin-wave stiffness of $CeFe_2$ is greatly reduced compared to that found in pure Fe.

Our inability to observe experimentally a response associated directly with the f element moment in these systems may arise from one (or both) of the following reasons. (1) The response is too weak because the spin moments on both the uranium and cerium ions are of an order of magnitude of $0.2\mu_B$, and/or (2) the response from the f spins is spread over a large energy range. In this latter case, the signal is difficult to separate from the background as the neutron instruments used in these studies, triple-axis spectrometers, have a relatively small resolution ellipsoid in reciprocal space.

The major difference between the compounds $CeFe_2$ and UFe_2 as far as the response of the Fe spins is concerned is certainly surprising, and can be answered only when theoretical estimates of $\chi''(\mathbf{Q},\omega)$ are available with which to com

TABLE I. Information on the three Laves phases ferromagnets (all with the C15 fcc crystal structure, symmetry $Fd3m$) studied by inelastic neutron scattering. In this structure each Fe has six nearest Fe neighbors. Pure Fe, which is bcc with eight nearest Fe neighbors, is added for comparison. T_c is the Curie temperature, μ_{Fe} is the magnetic moment on the Fe site, and ΔE is the ferromagnetic spin-wave gap at the zone center, $q=0$. The spin-wave stiffness constant D (see text) is shown for 100 K. The final row gives the deduced exchange between the Fe moments—see text. Standard deviations in parentheses refer to the least-significant digit.

	Fe	YFe_2	$CeFe_2$	UFe_2
a_0 (Å) (RT)	2.8665	7.370	7.304	7.057
d (Fe-Fe) Å	2.48	2.61	2.58	2.50
T_c (K)	1043(2)	545(5)	235(5)	165(5)
$\mu_{\mathrm{Fe}}(\mu_B)$	2.20(4)	1.5(1)	1.2(1)	0.6(1)
ΔE (meV)	<0.1	0.21(5)	0.25(3)	0.40(5)
D (meV Å^2)				
$T=100$ K	325(10)	250(12)	155(5)	440(30)
J (meV) ($\sim\pm15\%$)	24.0	24.4	19.4	117

pare experiments. Furthermore, measurements of the phonons (at room temperature) for $CeFe_2$, YFe_2, and UFe_2 have shown[8] that the parameters describing the phonons of the former two materials are roughly comparable, whereas in UFe_2 this is not the case. Instead, the phonon parameters in UFe_2 show strong deviations from those expected in a normal Laves-phase material, suggesting that the phonon spectrum is strongly influenced by the electronic susceptibility. Thus, one motivation for the present measurements was to establish whether any direct evidence could be obtained for a magnon-phonon interaction.

Because the Fe moment is only $0.6\mu_B$ in UFe_2 and thus small, the neutron inelastic experiments are difficult, despite the large ($\sim$7 g) single crystal available. Our earlier work extended only to $\sim$7 meV energy transfer. Recently, we have reported briefly[9] on measurements up to $\sim$20 meV. In this paper we summarize all our work extending the spin-wave measurements up to $\sim$35 meV, and we contrast the behavior of the Fe spin waves in the uranium compound with those in YFe_2. Comparative data, which will be useful in discussing the various compounds, are collected in Table I.

YFe_2 is particularly important in these comparisons. Because trivalent Y has no open $\underline{d}$ or f shell, it may be considered as ''diluted'' Fe, with the same Laves phase crystal structure as the other interesting materials discussed in this paper.

II. EXPERIMENTAL DETAILS AND RESULTS

The UFe_2 crystal is the same one described in Ref. 6. Single crystals of YFe_2 were grown in Ames from a Y-rich binary melt. High-purity elements were placed in a sealed Ta crucible and slowly cooled from 1200 °C to 950 °C over a period of a week. After this the excess liquid was decanted from large faceted crystals.

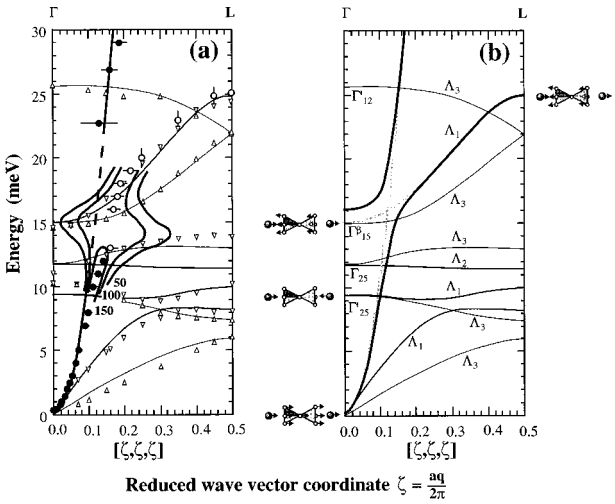

FIG. 1. (a) Spin-wave spectrum at 100 K (filled circles) and phonon dispersion relationships (phonons: $\triangle$, transverse; $\triangledown$, longitudinal) at 300 K in the $\langle 111\rangle$ direction. For the spin wave the quadratic relation is shown as a solid line, and clearly holds up to $\sim$10 meV. The strong interaction of the spin wave with another mode is shown above $\sim$13 meV by contours of equal intensity. The lines through the experimental phonon points represent a fit with a Born–von Kármán model and is discussed in more detail in Ref. 8. The open circles represent what we believe to be a mixed magnon-phonon mode. (b) Simulation of the magnon-phonon interaction proposed in UFe_2. The lines are guides to the eye. The symmetry of certain phonon modes and their relative displacements at the zone center and boundary are represented by showing [111] projections of the Fe tetrahedra along with two of the nearest neighbor U ions. For more details on the displacements see Ref. 8.

The neutron inelastic scattering experiments have been performed on the IN8 (thermal) and IN1 (hot) triple-axis spectrometers at the Institut Laue Langevin, Grenoble, and also with the 1T1 (thermal) instrument at the Laboratoire Leon Brillouin, Saclay. In all cases focusing graphite or copper monochromators have been used with graphite filters to suppress higher-order contamination. At the IN8 and IN1 spectrometers at the ILL a horizontal magnet with a field of 0.4 T aligned along the $\langle 111\rangle$ axis in the scattering plane assured that a single ferromagnetic domain existed during the experiments. Such an arrangement increases the observed spin-wave intensity by a factor of 50% over that in a multidomain sample.

To provide an overview before showing individual scans we show in Fig. 1 the most interesting region of the UFe_2 spectra in the $\langle 111\rangle$ direction. It should be emphasized, as we did earlier,[6] that *all* this response is associated with the Fe spins. An important question is what happens in the energy range above $\sim$12 meV, and it is on this region that we focus in this paper. The dashed line shows the spin-wave response extrapolated from that determined earlier,[6] and it clearly continues to $\sim$10 meV before becoming difficult to follow. Some of the scans used to construct Fig. 1 are shown in Fig. 2. The $\chi''(\mathbf{q},\omega)$ function is shown on the right-hand side and the shading illustrates how the shape of the peak changes over this energy range. Scans with the IN1 triple-axis spectrometer, which is placed on the hot source at the ILL, and thus able to measure excitations at much higher energy trans-

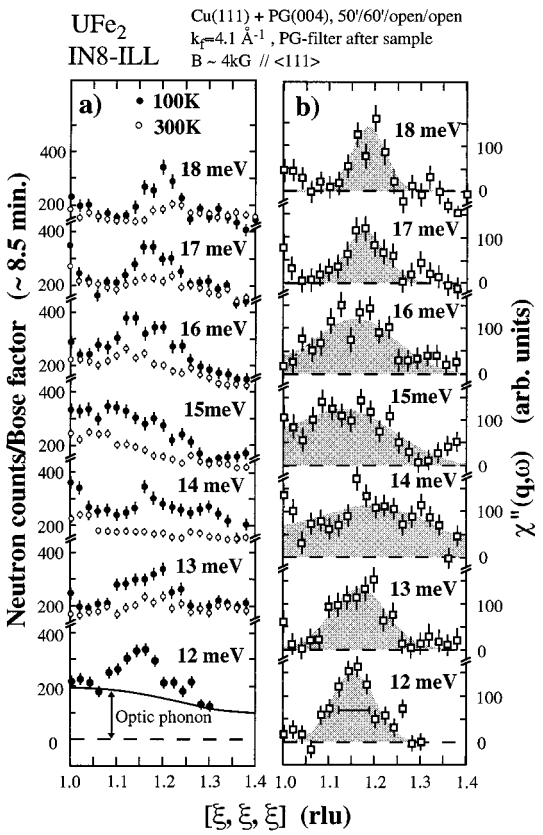

FIG. 2. Data used in the construction of Fig. 1. Data are shown on the left-hand side (a) for both 100 and 300 K. The magnetic signal is much smaller at 300 K, and most of the background is from optic phonons. By performing constant energy scans these phonons do not appear as peaks, as they have little dispersion, see Fig. 1, over this energy range. On the right-hand side (b) is shown the function $\chi''(\mathbf{q},\omega)$, which is obtained by subtracting a background and correcting by the Bose factor. The shading is a guide to the eye. [Data taken with a 0.4 T field along the scattering vector, and using $k_f = 4.1\ \text{Å}^{-1}$ and Cu(111)→PG(004) with IN8 at ILL.]

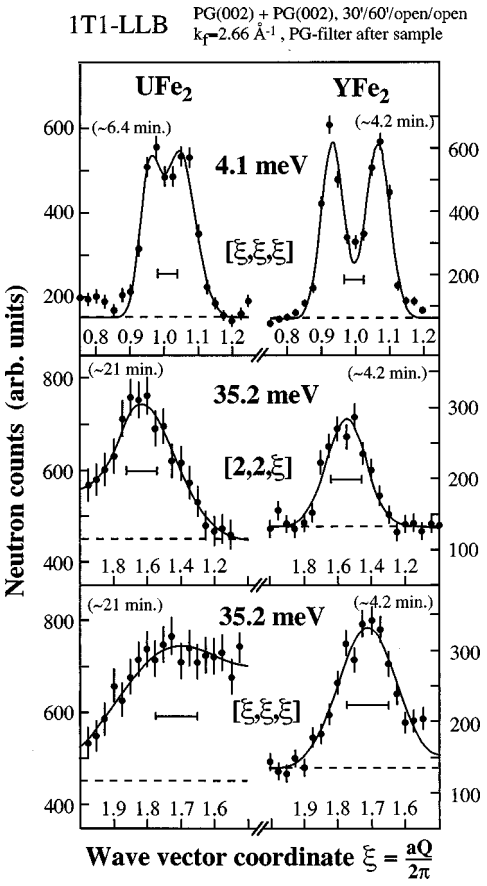

FIG. 3. Scans from UFe_2 ($T = 50$ K, left-hand side) and YFe_2 ($T = 300$ K, right-hand side) for different energy transfers. The upper and lower panels are in the ⟨111⟩ direction, whereas the center panel is for the momentum transfer in the ⟨001⟩ direction. The Γ point for the upper panels is $\xi = 1.0$; whereas for the four lower panels it is $\xi = 2.0$, corresponding to data taken around the reciprocal lattice point [222]. The horizontal bars represent the resolution function of the instrument. (Data from 1T1 at LLB.)

fer, showed no additional *magnetic* excitations in UFe_2 above 40 meV. However, some evidence of magnetic scattering is found up to at least 35 meV, even though the excitations are broad. In principle, of course, there will be many additional optic magnetic modes, but they are presumably too weak to be observed with the size of the present crystal and neutron fluxes.

Comparisons between YFe_2 and UFe_2 are shown in Fig. 3. The Curie temperature T_C of UFe_2 is about a third of that of YFe_2 (Table I), so that these diagrams represent the situation at low temperature in both compounds, and comparisons are valid despite the different temperatures. The top panel demonstrates the greater stiffness of the UFe_2 compared to that of YFe_2, since in the constant-E mode (see Fig. 1), the excitation comes at a smaller distance from the zone center in UFe_2. Considering higher energy the center panel shows the situation along the ⟨001⟩ direction. In both materials a propagating spin wave exists, but there is considerably more broadening in the case of UFe_2. At 35 meV the dispersion for UFe_2 is still greater than that for YFe_2 because the observed peak is closer to the (222) reciprocal lattice point. The lower panel shows the situation at high energy in the ⟨111⟩ direction. Here there is clearly only a ''ridge'' of scattering in the case of UFe_2, whereas a well defined excitation still exists for YFe_2.

III. DISCUSSION

Figure 4(a) gives the spin-wave dispersion curve and the calculated phonon spectra[6] for YFe_2. The thick solid lines give the fit to the spin-wave dispersion with the relationship

$$E = \Delta E + D(1 - \beta q^2)q^2,$$

where we include only the first higher-order term and assume that $\beta \sim 1$ following the work on pure iron.[10,11] ΔE is the energy gap and D is the spin-wave stiffness. It is to be noted that the gap is similar to that found in pure Fe; see Table I. D in pure Fe is 325 meV Å^2. We can use the relationship given in Ref. 10 to deduce the exchange interaction, J, between the Fe moments. In the approximation of small wave vector q, the spin-wave stiffness D is given by $D = 2JSa^2$, where S is the effective spin and a is the distance between atoms.

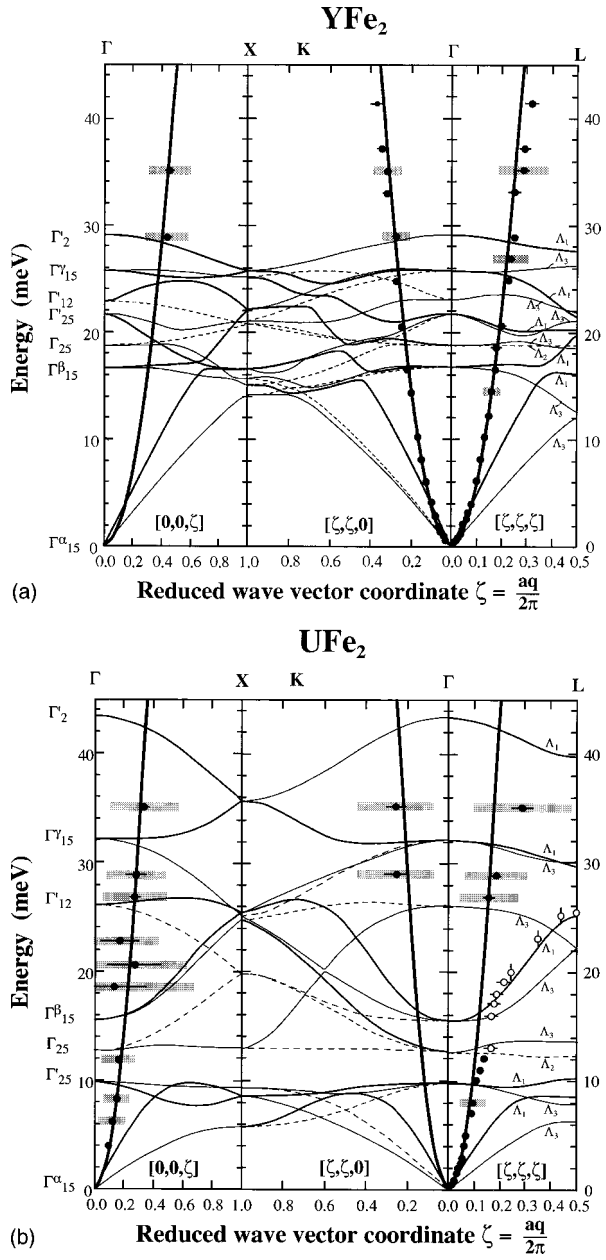

FIG. 4. (a) Spin-wave dispersion curve and calculated phonon spectra (Ref. 8) for YFe_2 at 300 K. The filled points represent the measured spin-wave energies and the thick solid line is a fit. Shading is an attempt to represent the width of the measured spin wave, and has been shown only for a series of scans taken with similar instrumental parameters. Below 14 meV the spin waves are resolution limited. Fitting to the spin-wave relationship gives $D = 250\ (12)$ meV Å^2. Thin solid lines represent calculated phonon spectra, with dashed lines showing phonons forbidden in the geometry of the experiment. (b) Spin-wave dispersion curve and calculated phonon spectra for UFe_2 at 100 K. Symbols are the same as in (a). The open points represent the ''mixed'' magnon and the Λ_1 phonon. The fitting to the low-energy part of the spin wave is with $D = 440\ (25)$ meV Å^2. Note the much greater broadening of the spin waves in UFe_2 than in YFe_2.

Having deduced D from the experimental dispersion curves, the other parameters in Table I then allow a deduction of the exchange interaction J, and this is given in the final row of Table I. Clearly the exchange interaction between Fe moments in pure Fe and YFe_2 is exactly the same, and the reduction in D simply comes from the smaller moment and the larger spacing between Fe atoms. This is further confirmation of our statement that YFe_2 is simply ''diluted'' Fe in the Laves-phase crystal structure. On the other hand, there is a fall in J for $CeFe_2$, consistent with hybridization effects found in that material and the tendency for antiferromagnetic fluctuations.[7] The J value for UFe_2 is large—as pointed out previously.[6]

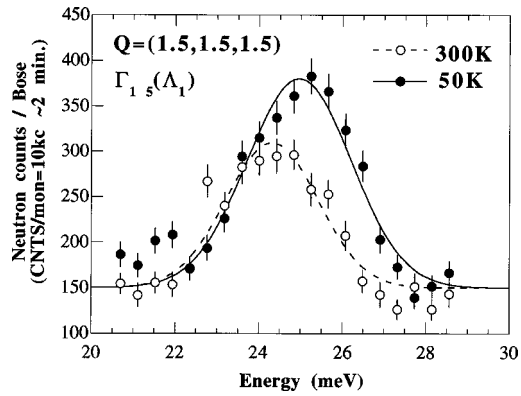

FIG. 5. Data taken at the zone boundary L point, $\mathbf{Q} = (1.5,1.5,1.5)$ at 50 (closed) and 300 K (open points). To compare the data, the signals have been divided by the respective Bose factors at each energy and temperature. (Data taken at 1T1 at LLB.)

Figure 4(b) shows the spin-wave relationship and the calculated phonon spectra in UFe_2. The Fe spin wave in UFe_2 has a much greater width, especially at higher energy, than those found in YFe_2, but magnetic scattering can be observed up to ~35 meV.

Focusing now on the interaction with the Λ_1 optic mode we show experimental data taken at two temperatures at the zone boundary in Fig. 5. Two unusual aspects may be observed once we have normalized the data by the Bose factors for the different temperatures. The first is that there is *more* intensity at 50 K. We have found, as expected, that all other phonons examined, whether in UFe_2 or in YFe_2 obey well the Bose population factors. The second is that the frequency is significantly shifted at 50 K and is higher. Although a small hardening of the phonon spectra might be expected at lower temperature, this shift of ~5%, although small, is not observed in the lower energy phonons in UFe_2 on cooling from 300 to 50 K, and this despite a greater precision in the measurement of the lower-energy phonon frequencies.

The Λ_1 optic phonon—at least at the zone center—may be visualized as a breathing mode in which the Fe tetrahedra and U ions move alternatively towards and away from each other. This is represented by the small figures on each side of Fig. 1(b), which gives a schematic idea of how the interaction may occur between the spin wave and the Λ_1 phonon in the $\langle 111 \rangle$ direction. Table II shows the vibrational amplitudes of the longitudinal $\Gamma_{15}^{\beta}(\Lambda_1)$ phonon branch propagating in the $\langle 111 \rangle$ direction evaluated for different reduced wave vectors $\mathbf{k} = (\zeta,\zeta,\zeta)$ ($\zeta = 0.0,0.1,...,0.5$) and expressed in term of absolute values of the Cartesian components. The atom displacements can be expressed in term of eigenvectors $\{e(\kappa|\mathbf{k})\}$ corresponding to the eigenvalues $\{\omega^2(\mathbf{k})\}$ of the dynamical matrix $D(\mathbf{k})$.[12] The displacement pattern can be described by the Cartesian component $u_\alpha(l\kappa)$ ($\alpha = x,y,z$) of

TABLE II. Vibrational amplitudes $u_\alpha(l\kappa)$ of the longitudinal $\Gamma_{15}^{\beta}(\Lambda_1)$ phonon branch, calculated at different propagation vectors **k**, with coordinates $(\zeta\zeta\zeta)$ along the [111] direction. The atoms positions U(κ) and Fe(κ) label the cluster shown in Fig. 6. The displacements of the Fe atoms 5, 6, and 7, are the same as those of atoms 2, 3, 4, respectively.

Atom positions			Vibrational amplitudes $u_\alpha(l\kappa)$ of $\Gamma_{15}^{\beta}(\Lambda_1)$ phonon (arb. units)					
			$\zeta=0$	$\zeta=0.1$	$\zeta=0.2$	$\zeta=0.3$	$\zeta=0.4$	$\zeta=0.5$
	x	1/8	−0.143	0.129	0.093	−0.054	−0.035	0.033
U(1)	y	1/8	−0.143	0.129	0.093	−0.054	−0.035	0.033
	z	1/8	−0.143	0.129	0.093	−0.054	−0.035	0.033
	x	7/8	−0.143	0.129	0.093	−0.054	−0.035	0.033
U(2)	y	7/8	−0.143	0.129	0.093	−0.054	−0.035	0.033
	z	7/8	−0.143	0.129	0.093	−0.054	−0.035	0.033
	x	1/2	0.465	−0.475	−0.485	0.460	0.353	0.000
Fe(1)	y	1/2	0.465	−0.475	−0.485	0.460	0.353	0.000
	z	1/2	0.465	−0.475	−0.485	0.460	0.353	0.000
	x	1/2	0.144	−0.166	−0.228	0.311	0.420	0.520
Fe(2)	y	−3/4	0.305	−0.317	−0.344	0.363	0.378	0.398
	z	3/4	0.305	−0.317	−0.334	0.363	0.378	0.398
	x	3/4	0.305	−0.317	−0.334	0.363	0.378	0.398
Fe(3)	y	1/2	0.144	−0.166	−0.228	0.311	0.420	0.520
	z	3/4	0.305	−0.317	−0.334	0.363	0.378	0.398
	x	3/4	0.305	−0.317	−0.334	0.363	0.378	0.398
Fe(4)	y	3/4	0.305	−0.317	−0.334	0.363	0.378	0.398
	z	1/2	0.144	−0.166	−0.228	0.311	0.420	0.520

the κ atom inside the l-unit cell and associated with the normal mode j with propagation vector **k** and frequency $\omega(\mathbf{k})$

$$u_\alpha(l\kappa)=[u(\mathbf{k})e_\alpha(\kappa|\mathbf{k})_j/M_\kappa^{1/2}]\exp\{i[\mathbf{k}\cdot\mathbf{x}(l)-\omega_j(\mathbf{k})t]\},$$

where $u(\mathbf{k})$ is an arbitrary amplitude factor. We consider the cluster shown in Fig. 6 in which two adjacent Fe tetrahedrons are shown along the [111] direction, identified as Fe(κ) ($\kappa=1,...,7$) and two uranium atoms $U(\kappa)$ ($\kappa=1,2$) also along the [111]. As shown in Table II, the polarization of Fe(1) is opposite to that of the two U atoms and becomes zero at the zone boundary L; see also Fig. 1(b). The other Fe atoms have a quasilongitudinal polarization, but it is not precisely along ⟨111⟩. The Fe atoms on either side of Fe(1) move in phase, and at the zone boundary it is the center Fe atom that is stationary, resulting in a higher energy for this mode. This optic mode results in a strong interaction between the Fe and U wave functions.

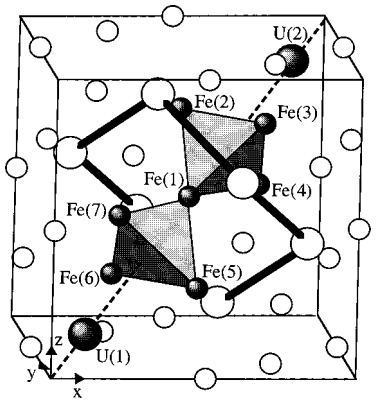

FIG. 6. Crystal structure of cubic Laves phase C15. This is the alternative setting than normally shown, with the U atoms not at the origin, but it places the Fe(1) atom at the body-centered position. Small (large) symbols represent the Fe (U) atoms and those shaded represent the cluster used for the calculation of vibrational amplitudes. The thick lines show the U-U bonding which forms a distorted ring around the central Fe(1) atom.

The nearest d(U-Fe)=2.93 Å. If we take the Hill criterion for U-U hybridization as 3.4 Å, then the effective radius is 1.7 Å, and that for Fe (as taken from the nearest neighbor in pure Fe) is 1.24 Å, so that the sum of these radii are 2.94 Å, exactly that for the nearest U-Fe distance in UFe_2. According to the phonon spectrum[8] the energy necessary to excite this breathing motion is between 10 and 14 meV at the zone center. This is *exactly* the energy at which it becomes difficult to observe the spin wave in the ⟨111⟩ direction, which also represents the closest approach of the two sublattices.

Of course, the ideal measurement would be to perform polarization analysis of the neutron spectra over this energy range, but in view of the weakness of the spin wave scattering in UFe_2 and the great loss associated with polarization analysis at these energy transfers, this is unfortunately beyond our present capabilities.

As discussed in our earlier work,[6] Eremenko *et al.*[13] introduced the idea of magnon-phonon coupling to explain the large change in the Lamb-Mössbauer factor observed at T_C

in UFe_2. They assumed a coupling to the *acoustic* transverse phonons at low energy. There is no evidence for such a coupling, and the anomaly in the Lamb-Mössbauer factor is probably related to the large exchange interactions present in UFe_2. However, interestingly, our present measurements do show that there is a possible magnon-phonon interaction, but with the *optic* longitudinal phonons. Schematically, following Ref. 13, we may represent the two modes as shown in Fig. 1(b). Further details of such an interaction, and consequences to the elastic constants, for example, are beyond the scope of the present experimental study.

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