Helicity Order: Hidden Order Parameter in URu_2Si_2

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(Received 28 March 2005; published 25 January 2006)

We propose that the ''hidden order parameter'' in URu_2Si_2 is a helicity order that must arise if the Pomeranchuk criteria for the spin-antisymmetric Landau parameters with respect to the stability of a Fermi liquid state are violated. In a simple model, we calculate the specific heat, the linear and nonlinear magnetic susceptibilities, and the change of transition temperature in a magnetic field with such an order parameter, and obtain quantitative agreement with experiments in terms of two parameters extracted from the data. The peculiar temperature dependence of the NMR linewidth and the nature of the loss of excitations in the ordered phase seen by neutron scattering are also explained, and experiments are suggested to directly confirm the proposed order parameter.

DOI: 10.1103/PhysRevLett.96.036405 PACS numbers: 71.27.+a, 71.10.Hf, 75.10.−b, 75.40.Cx

The ''hidden order'' phase in the heavy fermion compound URu_2Si_2 below the second order transition at 17.5 K [1] has remained a puzzle for about 20 years. The magnitude of the specific heat at the transition is equivalent to that of ordering of a moment of about $0.5\mu_B$ per unit cell. No change in spin-rotational symmetry or lattice translational symmetry consistent with this specific heat has been discovered. Detailed neutron diffraction experiments [2] reveal a moment of only about $0.03\mu_B$ per unit cell, which as NMR and μSR experiments [3] reveal, is due to the presence of a second phase. Some very interesting proposals for new types of order have been made [4–6], which have not been supported by experiments designed to look for them.

Some of the other properties measured at the transition to the ''hidden order'' phase and in it are the linear magnetic susceptibility which only changes slope at the transition, the nonlinear magnetic susceptibility which shows a singularity at the transition similar to that of the specific heat [7], the change of the transition temperature with an applied magnetic field [8,9], the loss of low energy excitations observed by neutron scattering for a range of wave vectors [2,10,11] and in transport measurements [12], and the NMR relaxation rate [13] which exhibits the extraordinary result that there is an extra inhomogeneous relaxation rate in the ordered phase which increases below the transition temperature *proportional* to an order parameter.

We suggest here that the transition is to a state proposed [14] as a cure to the spin-antisymmetric Landau-Pomeranchuk instability (LPI) of the Fermi liquid. For reasons that will be clear, we call such states *helicity-ordered states* [15]. We calculate the thermodynamic properties near the transition and account quantitatively for the observed thermodynamic features and qualitatively for the NMR and the excitation spectra with parameters extracted from the experiments. We also suggest experiments that can provide direct evidence for the proposed phase.

In Landau's Fermi liquid theory [16], the change in the free energy due to a small change of the equilibrium distribution function $\delta n(\mathbf{k}\sigma)$ is

$$\delta F = \sum_{\mathbf{k}\sigma} \epsilon^0_{\mathbf{k}} \delta n(\mathbf{k}\sigma) + \frac{1}{2}\sum_{\mathbf{k}\sigma\mathbf{k}'\sigma'} f(\mathbf{k}\sigma, \mathbf{k}'\sigma')\delta n(\mathbf{k}\sigma)\delta n(\mathbf{k}'\sigma'). \quad (1)$$

The interaction functional $f(\mathbf{k}\sigma, \mathbf{k}'\sigma')$ has spin-symmetric (s) and spin-antisymmetric (a) parts:

$$f(\mathbf{k}\sigma, \mathbf{k}'\sigma') = f^s(\mathbf{k}\sigma, \mathbf{k}'\sigma') + f^a(\mathbf{k}\sigma, \mathbf{k}'\sigma'). \quad (2)$$

The coefficients of an expansion of $f^{s,a}$ in terms of the irreducible representations of the Fermi surface are the Landau parameters $F_l^{s,a}$, in terms of which Pomeranchuk [17] obtained a set of conditions for the stability of the Fermi liquid: $1 + (2l+1)^{-1}F_l^{s,a} > 0$. Any violation of these conditions leads to a Landau-Pomeranchuk instability, which must be cured by a broken symmetry in corresponding irreducible representation l and spin symmetry s or a. For example, the ferromagnetic instability occurs for $F_0^a \to -1$. Spin-symmetric instabilities in finite l channels have attracted much recent interest [14,18,19]. A spin-ordered state, which is anisotropic in momentum space (without change in translational symmetry) is the obvious cure to the finite l antisymmetric LPIs [14,20]. We develop this idea here; we find that besides the LPI criteria, additional conditions must be satisfied so that the instability is of second order.

Consider the model Hamiltonian,

$$\mathcal{H} = \sum_{k\sigma} \epsilon^0_k c^\dagger_{k\sigma} c_{k\sigma} + \frac{1}{2}\sum_{\mathbf{k},\mathbf{k}';\mathbf{q}} J_{\mathbf{k},\mathbf{k}'}(\mathbf{q})(c^\dagger_{k+q}\vec{\sigma}c_k)\cdot(c^\dagger_{k'-q}\vec{\sigma}c_{k'}), \quad (3)$$

where ϵ^0_k is the spectrum of a noninteracting Fermi gas, $\vec{\sigma}$ Pauli matrices. $\mathbf{q}$ is the momentum transfer; of interest is the instability in the forward scattering limit ($q \to 0$). $J_{\mathbf{k},\mathbf{k}'}$ is the interaction in spin-antisymmetric channels, which can be expanded as $J_{\mathbf{k},\mathbf{k}'}(0) = \sum_l J_l P_l(\cos\theta_{\mathbf{k},\mathbf{k}'})$, where $\theta_{\mathbf{k},\mathbf{k}'}$ is the angle between $\mathbf{k}$ and $\mathbf{k}'$, and $P_l(x)$ are Legendre polynomials.

In the normal state of a Fermi liquid, helicity is disordered since the spin-quantization axes at each $\mathbf{k}$ can be independently rotated. The proposed order parameter for the model has the general form [14,20]:

$$\langle \delta n(\mathbf{k}, \sigma)\rangle = \sigma \cdot \mathbf{D}(\hat{\mathbf{k}}_f). \quad (4)$$

The spin-quantization axis is thereby fixed in relation to the direction on the Fermi surface [21].

We need consider only one specific l channel and write $J_{\mathbf{k},\mathbf{k}'}$ in a separable form $J_l P_l(\cos\theta_k) P_l(\cos\theta_{k'})$. The simplest order parameter has $D^z(\hat{\mathbf{k}}_f) \neq 0$, so that the associated energy parameter is

$$\Delta_l = \langle J_l \sigma^z D^z(\hat{\mathbf{k}}_f)\rangle = \left\langle \sum_k J_l P_l(\cos\theta_k)(n_{k\uparrow} - n_{k\downarrow})\right\rangle. \quad (5)$$

This Ising order parameter is especially useful to discuss the tetragonal compound URu_2Si_2, which has a large anisotropy in the magnetic susceptibility favoring the c axis. With Eq. (5) we have a noninteracting model with the effective spectrum

$$E_{\uparrow,\downarrow}(\mathbf{k}) = \epsilon_k^0 \mp \mu_0 H \pm \Delta_l P_l(\cos\theta_k), \quad (6)$$

where H is the external magnetic field, and μ_0 the effective single-electron magnetic moment. Imposing the requirement of a constant chemical potential, the Fermi surfaces for the up and down spins are split as schematically illustration in Fig. 1.

We calculate the free energy following standard methods. The following results are for $l = 1$, but can easily be generalized to higher-l channels. The free energy can be in general separated into two parts,

$$\mathcal{F} = \mathcal{F}_0(T, H) + \mathcal{F}_m(T, H, \Delta_1), \quad (7)$$

where $\mathcal{F}_0$ describes a paramagnetic phase ($\Delta_1 = 0$). The specific heat and the magnetic susceptibility [$M = \chi_1 H + (\chi_3/3!)H^3 + \cdots$, χ_1 and χ_3 are the linear and nonlinear spin susceptibilities, respectively] can be easily obtained. Including terms of $O(H^2)$, $O(T^2)$, and the variation of the density of states $\rho(\epsilon)$ near the chemical potential to $O[\rho''(\epsilon_F)]$, we obtain

$$\frac{C_H^0}{T} = \frac{2\pi^2}{3} k_B^2 \rho\left[1 + \frac{1}{2}\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}\right)(\mu_0 H)^2\right], \quad (8a)$$

$$\chi_1^0 = 2\mu_0^2 \rho\left[1 + \left(\frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}\right)\frac{\pi^2}{6}(k_B T)^2\right], \quad (8b)$$

$$\chi_3^0 = 3!\mu_0^4 \rho\left[\left(\frac{\rho''}{3\rho} - \frac{\rho'^2}{\rho^2}\right) + 3\frac{\rho'^2}{\rho^2}\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{3\rho^2}\right)\frac{\pi^2}{6}(k_B T)^2\right]. \quad (8c)$$

For noninteracting electrons, these are standard results (see, e.g., Ref. [22]); we list them here to use them to extract parameters from the normal state experimental

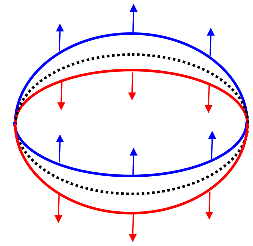

FIG. 1 (color online). Schematic Fermi surface for the two different spin directions. The dotted line denotes the Fermi surface of the paramagnetic phase with vanishing order parameter while the solid lines illustrate the Fermi surface of the helicity-ordered phase.

results. For interacting electrons (in the limit of zero field) they are multiplied by Landau parameters, m^*/m for the specific heat, $(m^*/m)/(1 + F_0^a)$ for the susceptibility and $(m^*/m)/(1 + 4F_0^a)$ for the nonlinear susceptibility.

$\mathcal{F}_m(T, H, \Delta_1)$, is the additional contribution to the free energy for $\Delta_1 \neq 0$. Expressing it in series of the order parameter Δ_1 gives

$$\begin{aligned}
\mathcal{F}_m &= \frac{1}{2}A\Delta_1^2 + \frac{1}{4}B\Delta_1^4,\\
A &= \frac{2\rho}{3}\left(1 + \frac{3}{2\rho J_1}\right) + \frac{2\rho}{3}\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}\right)\\
&\quad \times\left[\frac{\pi^2}{6}(k_B T)^2 + \frac{(\mu_0 H)^2}{2}\right]\\
&\quad + \rho\frac{\rho'^2}{\rho^2}\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{3\rho^2}\right)\frac{\pi^2}{6}(k_B T)^2(\mu_0 H)^2,\\
B &= \rho\left(\frac{\rho''}{5\rho} - \frac{\rho'^2}{3\rho^2}\right).
\end{aligned} \quad (9)$$

When $H = 0$ and $T = 0$, the criterion to have a continuous phase transition is $A < 0$ and $B > 0$, i.e.,

$$1 + \frac{3}{2\rho J_1} < 0; \qquad \frac{\rho''}{5\rho} - \frac{\rho'^2}{3\rho^2} > 0. \quad (10)$$

The first gives $J_1 < 0$ and $\rho|J_1| > (2l + 1)/2$ (here $l = 1$), which is precisely the LPI criterion. The second is an additional criterion, on the form of the density of states at the Fermi surface to have a second order transition. If $B < 0$, one must expand the free energy to terms of order Δ_1^6. In that case, a first-order phase transition is favored.

At $H = 0$, the critical temperature is given by

$$\frac{\pi^2}{6}(k_B T_c)^2 = -\left(1 + \frac{3}{2\rho J_1}\right)\Big/\left(\frac{\rho''}{\rho} - \frac{\rho'^2}{\rho^2}\right). \quad (11)$$

This, together with Eq. (10), requires $\rho''/\rho > \rho'^2/\rho^2$. In the presence of a small magnetic field, T_c varies as

$$\begin{aligned}
T_c^H &\approx T_c[1 - (H/H_0)^2],\\
H_0 &= \frac{(2/3)^{1/2}\pi k_B T_c/\mu_0}{[1 + \frac{\pi^2(k_B T_c)^2}{2}\frac{\rho'^2}{\rho^2}h_\rho]^{1/2}},
\end{aligned} \quad (12)$$

where $h_\rho \equiv [\rho''/\rho - \rho'^2/(3\rho^2)]/(\rho''/\rho - \rho'^2/\rho^2)$. Below T_c we have the nontrivial solution $\Delta_1^2 = -A/B$, i.e., a helicity-ordered state. The changes in some thermodynamical quantities at and below T_c from their values for $T > T_c$ are calculated to be

$$\delta c_H/c_H^0 = \frac{\rho}{81}\pi^4 k_B^4 g_\rho (3T^2 - T_c^2), \tag{13a}$$

$$\delta\chi_1/\chi_1^0 = \frac{2\rho}{9}\frac{\mu_0^2}{\chi_1^0} g_\rho \frac{\pi^2}{6} k_B^2 (T^2 - T_c^2), \tag{13b}$$

$$\delta\chi_3/\chi_3^0 = \frac{2\rho}{3}\frac{\mu_0^4}{\chi_3^0} g_\rho \left[1 + h_\rho \frac{\rho'^2}{\rho^2}\frac{\pi^2}{6}(k_B T)^2\right], \tag{13c}$$

where $g_\rho \equiv (\rho''/\rho - \rho'^2/\rho^2)^2/[\rho''/(5\rho) - \rho'^2/(3\rho^2)]$. The specific heat shows, of course, a characteristic mean-field discontinuity at the transition point; more interesting is the fact that χ_3 also shows a discontinuity while the linear magnetic susceptibility shows merely a change of slope. A singularity in the nonlinear magnetic susceptibility is to be expected for any order parameter O when a term $|O|^2 H^2$ is allowed in the free energy. Usually the coefficient of this term is so small that the singularity in χ_3 is not noticed. What is special about URu_2Si_2 is that the dimensionless mean-field jump in χ_3 is similar to the dimensionless mean-field jump in the specific heat. This and several other properties are quantitatively explained below.

We have ignored the Landau parameters in the dimensionless quantities in Eqs. (8) and (13). The reason is that the change in the Landau parameters near the transition may be shown following Leggett [23], for the case of transition in superfluid ^{3}He, to be proportional to $(\rho\Delta_1)^2$. To this order, assumed $\ll 1$, they vanish in dimensionless quantities.

In the following, we try to fit the experimental data: the specific heat from Ref. [1], and the linear and nonlinear magnetic susceptibilities from Ref. [7]. The data are shown in Fig. 2 for the reader's convenience. From Eqs. (8) and (13), in addition to the prefactors, $\gamma_0 = \frac{2\pi^2}{3}\rho k_B^2$, $\chi_0 = 2\mu_0^2\rho$, $\tilde{\chi}_0 = 3!\,\mu_0^4\rho$, all other quantities can be determined by two additional dimensionless variables, $C_1 = (\rho''/\rho - \rho'^2/\rho^2)(\pi^2/6)(k_B T_c)^2$ and $C_2 = (\rho'^2/\rho^2)/(\rho''/\rho)$, where T_c is taken as 17.5 K.

Consider the linear magnetic susceptibility. It is continuous with a change in the slope at the transition point, which is consistent with the result in Eq. (13b). Also notice that in the normal state it shows a significant linear temperature dependence besides the constant Pauli term. This is, indeed, required by the theory in order that there be a second order transition to the helicity-ordered phase [see Eq. (10)]. Near T_c,

$$\chi_1(T \gtrsim T_c) = \chi_0(1 + C_1) + 2\chi_0 C_1 (T - T_c)/T_c,$$
$$\delta\chi_1(T \lesssim T_c) = \chi_0 \frac{10}{9} C_1 \frac{1 - C_2}{1 - 5C_2/3}(T - T_c)/T_c. \tag{14}$$

By fitting them with the experimental data, we can extract

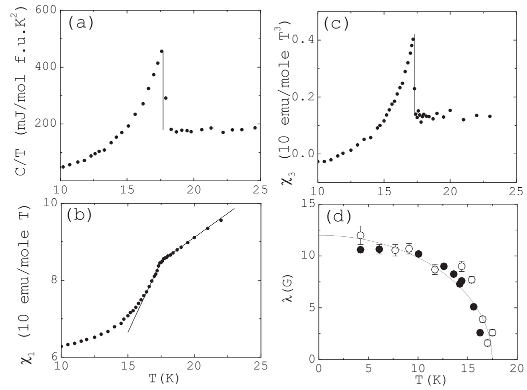

FIG. 2. The experimental data of direct relevance to the calculations in this Letter. The specific heat data (a) are extracted from Fig. 1 in Ref. [1]. In the specific heat data a contribution to C/T proportional to T^2, presumably mostly due to phonons, has been subtracted. Data of the linear (b) and nonlinear (c) magnetic susceptibilities (along the c axis) are extracted from Fig. 2 in Ref. [7]. (d) The inhomogeneous linewidth of Si-NMR for magnetic fields in the c direction and in the plane, which is extracted from Fig. 4 in Ref. [13]; the solid line is the fitting function $\lambda = 12[1 - (T/T_c)^2]^{1/2}(G)$.

$$C_1 \approx 0.35, \qquad C_2 \approx 0.52 \tag{15}$$

(with $\chi_0 \approx 63.0$ emu/mole T). We then can calculate other thermodynamic quantities. At T_c, the discontinuities of the specific heat coefficient ($\gamma = C/T$) and the nonlinear magnetic susceptibility are calculated from Eq. (13) to be

$$\delta\gamma(T_c)/\gamma_0 = 1.4, \qquad \delta\chi_3(T_c)/\chi_3(T_c^+) = 2.1, \tag{16}$$

while the corresponding experimental quantities from Fig. 2 are approximately 1.5 and 2.4, respectively. In finite magnetic field, Eq. (12) predicts that T_c decreases as $(H/H_0)^2$, where $H_0 = 38.2T$ if we take μ_0 to be one Bohr magneton. This again, is in agreement with the experiments, where H_0 is estimated as 48.5(1) T in Ref. [8], while 35.3 T in Ref. [9]. We can also determine the order parameter ($H = 0$)

$$\Delta_1(T) = 77[1 - (T/T_c)^2]^{1/2}\ \mathrm{K}. \tag{17}$$

To summarize, we get the qualitatively correct behavior of the linear susceptibility, and extracting two parameters from it can explain quantitatively the relative jump in the specific heat and that of the nonlinear susceptibility as well as the characteristic field for the suppression of the transition. (The fact that a simple model for the Fermi surface of the paramagnetic phase gives these quantities quite well is doubtless due to the fact that we are comparing dimensionless quantities.) We do not do well on the specific heat just below T_c; our slope is about a factor of 2 smaller than the straight line fit one might make in Fig. 2. Note that the proposed order parameter has an Ising symmetry. So, the actual exponents in the critical regime of the transition are expected to be that of an Ising model in three dimensions;

for example, the specific heat is expected to show a lambda shape. While further experiments are required to test this, the measured specific heat is not inconsistent with such a form (see Fig. 2). In such a case, a mean-field fit to the data always gives a slope smaller than the experiment.

The values of C_1, C_2 required above imply that $\rho''/\rho \approx 2\rho'^2/\rho^2$; i.e., the chemical potential in the normal phase lies near a local minima of the density of states. This is consistent with the density of states calculated by band-structure calculations [24].

Let us next consider the NMR measurements. Si-NMR [13] reveals no change in Knight shift but an increase in the inhomogeneous linewidth below T_c, which within experimental uncertainty can be fitted to be $\propto [1 - (T/T_c)^2]^{1/2}$ [see Fig. 2(d)], i.e., *proportional* to an order parameter. This is quite unusual. We note first that in a perfectly pure sample, we expect no change in Knight shift or linewidth. In the presence of impurities, which *locally* break the reflection symmetry about the basal plane, a local ferromagnetic region forms, as is evident from Fig. 1. The magnitude of the local field then is proportional to the order parameter, but its magnitude as well as direction are random. This gives no Knight shift but a linewidth consistent with observations. The magnitude depends on details of the defect, but a $O(0.5\mu_B)$ defect, expected from the magnitude of the order parameter, need be present only in concentrations of a few parts in a thousand to produce the observed linewidth of order 10 Gauss. The observed linewidth is almost independent of the direction of the applied magnetic field. This can be shown to occur for generic distribution of impurities about the Si sites [25].

We intend to calculate the excitation spectra in the future. One can, however, see that, given the Fermi surfaces shown in Fig. 1, a decrease in inelastic scattering below a certain characteristic energy of the order of Δ_1 is expected for spin-flip particle-hole excitations. This is what is observed in neutron scattering [2,10,11]. A quantitative calculation requires a more realistic model of the normal state Fermi surface than used here. We note that the characteristic magnitude of this energy scale observed in inelastic neutron scattering experiments is $\sim$70 K [2], which compares well with our estimate [see the coefficient in Eq. (17)].

Finally, we turn to how the proposed order parameter may be directly observed. On applying an electric field parallel to the c axis, a spin current would be generated but no such effect should occur on applying electric field parallel to the basal plane [27]. Another direct possibility is through spin-polarized positron annihilation suggested to us [28].

We acknowledge useful discussions with V. Aji, W. Buyers, D. MacLaughlin, A. Mills, A. Migliori, J. Mydosh, and A. Ramirez.

[1] T. T. M. Palstra *et al.*, Phys. Rev. Lett. **55**, 2727 (1985); M. B. Maple *et al.*, Phys. Rev. Lett. **56**, 185 (1986); W. Schlabitz *et al.*, Z. Phys. B **62**, 171 (1986).

[2] C. Broholm *et al.*, Phys. Rev. Lett. **58**, 1467 (1987); C. Broholm *et al.*, Phys. Rev. B **43**, 12 809 (1991).

[3] K. Matsuda *et al.*, Phys. Rev. Lett. **87**, 087203 (2001); H. Amitsuka and M. Yokoyama, Physica (Amsterdam) **329–333B**, 452 (2003).

[4] P. Santini and G. Amoretti, Phys. Rev. Lett. **73**, 1027 (1994).

[5] V. Barzykin and L. P. Gor'kov, Phys. Rev. Lett. **74**, 4301 (1995).

[6] P. Chandra *et al.*, Nature (London) **417**, 831 (2002).

[7] A. P. Ramirez *et al.*, Phys. Rev. Lett. **68**, 2680 (1992).

[8] N. H. van Dijk *et al.*, Phys. Rev. B **56**, 14 493 (1997).

[9] J. S. Kim *et al.*, Phys. Rev. B **67**, 014404 (2003).

[10] M. J. Bull *et al.*, Acta Phys. Pol. B **34**, 1265 (2003).

[11] C. R. Wiebe *et al.*, Phys. Rev. B **69**, 132418 (2004).

[12] T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. B **33**, 6527 (1986).

[13] O. O. Bernal *et al.*, Phys. Rev. Lett. **87**, 196402 (2001).

[14] C. M. Varma, Philos. Mag. **85**, 1657 (2005).

[15] This should be distinguished from *helical* spin states, such as found in MnSi and in many rare-earth metals.

[16] L. D. Landau, Zh. Eksp. Teor. Fiz. **30**, 1058 (1956) [Sov. Phys. JETP **3**, 920 (1957)]; see also D. Pines and P. Nozières, *The Theory of Quantum Liquids* (Benjamin, New York, 1966), Vol. 1, Chap. 1.

[17] I. Ya. Pomeranchuk, Zh. Eksp. Teor. Fiz. **35**, 524 (1958) [Sov. Phys. JETP **8**, 361 (1959)].

[18] C. J. Halboth and W. Metzner, Phys. Rev. Lett. **85**, 5162 (2000).

[19] V. Oganesyan, S. A. Kivelson, and E. Fradkin, Phys. Rev. B **64**, 195109 (2001).

[20] The possibility of such states from quite different arguments has been pointed out by J. E. Hirsch, Phys. Rev. B **41**, 6820 (1990).See also C. Wu and S. C. Zhang, Phys. Rev. Lett. **93**, 036403 (2004).

[21] Spin-orbit interactions also fix the spin relative to **k**. However, contrary to the helicity order, the point group symmetry of the lattice is respected when they are taken into account.

[22] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Saunders College, Philadelphia, 1976), Chaps. 2, 31.

[23] A. J. Leggett, Phys. Rev. Lett. **14**, 536 (1965).

[24] G. J. Rozing, P. E. Mijnarends, and D. D. Koelling, Phys. Rev. B **43**, 9515 (1991).

[25] We thank Professor D. MacLaughlin for an explicit calculation based on Ref. [26] checking that the anisotropy is about 20%.

[26] R. E. Walstedt and L. R. Walker, Phys. Rev. B **9**, 4857 (1974).

[27] A. Migliori (private communication).

[28] A. Mills (private communication).