Magnetic Excitations in an Itinerant Ferromagnet near Quantum Criticality

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We determine the initial temperature dependence of the exchange splitting $\Delta(T)$ in the weak itinerant ferromagnet $ZrZn_2$ ($T_C = 28$ K) using the de Haas–van Alphen effect. There is a large decrease in Δ with temperature in the range $0.5 \leq T \leq 4$ K. A comparison of $\Delta(T)$ with the magnetization $M(T)$ shows that the dominant process responsible for the reduction of M is not the thermal excitation of spin waves, but a repopulation of the spin-$\uparrow$ and spin-$\downarrow$ Fermi surfaces. This contrasts with the behavior in Fe where there is no observable change in Δ and the thermal excitation of spin waves is the only observable spin-flipping process at low temperatures.

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It is well known that at low temperatures the ordered ferromagnetic moment of itinerant ferromagnets such as Fe is reduced initially by the thermal excitation of spin waves. The magnetic moment varies initially as $T^{3/2}$ and the magnitude of the reduction can be understood from independent measurements of the spin-wave stiffness [1]. Fe and Ni are materials with large ordered moments at low temperatures and can thus be regarded as being far from quantum criticality. In this Letter, we measure the temperature dependence of the exchange splitting Δ or magnetic molecular field [$\Delta = \varepsilon(\mathbf{k}_{F,\uparrow}) - \varepsilon(\mathbf{k}_{F,\downarrow})$, where $\varepsilon(\mathbf{k})$ is the paramagnetic band energy]. We show that in an itinerant quantum ferromagnet, i.e., an itinerant ferromagnet close to a quantum critical point (QCP), the mechanism for the destruction of the exchange splitting Δ and magnetization M with increasing temperature is different from that in a large moment itinerant ferromagnet (e.g., Fe) or a localized ferromagnet (e.g., EuO). In the itinerant quantum ferromagnet studied here ($ZrZn_2$), Δ decreases rapidly with temperature and drives the concomitant reduction in M.

The magnetization of a ferromagnet generally decreases with increasing temperature T. Fundamentally, this reduction can only arise by flipping electron spins. At low T, the spin-flipping processes which reduce M in ferromagnetic metals have been split into two types: the thermal excitation of transverse ($\Delta S = 1$) excitations [e.g., spin waves (SW)] and the repopulation of the spin-$\uparrow$ and spin-$\downarrow$ Fermi surfaces by single-particle (SP) excitations (i.e., "Stoner excitations") [2]. One method to obtain information about the nature of the low-energy magnetic excitations in ferromagnets is to compare the T dependence of $\delta\Delta(T) = \Delta(T) - \Delta(0)$ with the corresponding change in the magnetization $\delta M(T) = M(T) - M(0)$ [3,4]. At low T the thermal excitation of spin waves does not affect $\Delta(T)$ [3]. As we demonstrate in this work, $\Delta(T)$ can be measured using high precision by de Haas–van Alphen (dHvA) measurements and $M(T)$ can be determined by SQUID or other magnetometry measurements.

The material we have chosen to study is the small moment itinerant ferromagnet $ZrZn_2$ which crystallizes in the C15 cubic Laves structure with $a = 7.393$ Å. The low T_C and small ordered moment ($0.18\mu_B$ per formula unit) of $ZrZn_2$ indicate that it is close to a ferromagnetic QCP [5] and have led to predictions that longitudinal excitations may cause p-wave superconducting pairing [6]. However, bulk superconductivity has not yet been observed in $ZrZn_2$ [7]. The sample investigated here had a residual resistivity $\rho_0 = 0.6\ \mu\Omega$ cm and a $T_C = 27.5$ K [8] and did not show bulk superconductivity.

The dHvA effect is an oscillatory contribution M_{osc} to the field-dependent magnetization $M(B)$ due to the quantization of the cyclotron orbits of charge carriers. It is well described by the Lifshitz-Kosevich (LK) theory in which each extremal cross section of the FS, of area A, makes an oscillatory contribution to M [9] of the form

$$M_{\mathrm{osc}} \propto B^{1/2} R_D R_T \sin\left[\frac{2\pi F(B)}{B} + \phi\right], \qquad (1)$$

where R_D and R_T are damping factors due to scattering and temperature, respectively, and $F(B) = (\hbar/2\pi e)A(B)$. Our dHvA measurements were made using the field-modulation method in a superconducting 19 T magnet with ^{3}He cryostat insert. Modulation-field amplitude and frequency were $b = 8$ mT and $f = 8.8$ Hz, respectively, and signals were detected phase sensitively at f and $2f$. A single-crystal $ZrZn_2$ sample was mounted in the pickup coil with $[001] \parallel \mathbf{B}$. The field was swept up and down between 5.5 T and 10.5 T with the sample temperature held constant at a value in the range $0.5 \leq T \leq 4.0$ K. Because our main result depends on detecting small relative field shifts, we ensured that the field history of the magnet was identical for all sweeps, thus minimizing changes in field offsets in the superconducting magnet.

The Fermi surface of $ZrZn_2$ has previously been studied in detail by dHvA measurements [10,11]. There are four exchange-split Fermi surface sheets. At low magnetic fields the dHvA signal is dominated by a small exchange-split pocket centered on the Γ point [see Fig. 1(a)]. Figure 2(a) shows raw dHvA data taken at 0.5 K. The oscillations correspond to the exchange-split spin-$\uparrow$ and spin-$\downarrow$ Fermi surfaces. The signal is dominated by components due to the α and β orbits on the spin-$\downarrow$ and spin-$\uparrow$ sheets. These components beat together to give the pattern in Fig. 2(a). We denote the extremal areas (frequencies) of these orbits as $A_\downarrow$ and $A_\uparrow$ ($F_\downarrow$ and $F_\uparrow$).

In this work, we study $\Delta(T)$ in the band giving rise to the Γ pocket. Δ may be defined in terms of the (paramagnetic) band energy $\varepsilon(\mathbf{k})$. Provided Δ is small, we have

$$\Delta = \varepsilon(\mathbf{k}_{F,\uparrow}) - \varepsilon(\mathbf{k}_{F,\downarrow}) \simeq (A_\uparrow - A_\downarrow)\left(\frac{\partial \varepsilon}{\partial A}\right) = \frac{\hbar e}{m_b}(F_\uparrow - F_\downarrow), \tag{2}$$

where m_b is the average band mass. Thus a change in Δ causes a change in $F_\downarrow$ and $F_\uparrow$ which appears as an apparent phase shift of the dHvA oscillations for a given field. If we fit Eq. (1) to experimental data at a low temperature T_1 and then repeat this procedure at a higher temperature T_2, fixing the dHvA frequency F at its low temperature value, then the apparent phase shift for $F_\uparrow$ will be

$$\delta\phi_\uparrow = \phi_\uparrow(T_2, B) - \phi_\uparrow(T_1, B) = \frac{2\pi}{B}[F_\uparrow(T_2) - F_\uparrow(T_1)]. \tag{3}$$

From Eqs. (2) and (3), we can deduce the fractional change in Δ between T_1 and T_2,

$$\frac{\delta\Delta}{\Delta} = \frac{B}{2\pi}\frac{\delta\phi_\uparrow - \delta\phi_\downarrow}{[F_\uparrow(T_1, B) - F_\downarrow(T_1, B)]}. \tag{4}$$

The positions of the beat minima in Fig. 2 are determined by the relative phase of the two frequency components in the beat pattern. Figure 2 shows that the beat minima shift to lower B as T increases, indicating that our measurements are sensitive to the T-induced decrease of Δ. In order to check for possible systematic errors which might give rise to spurious phase shifts, we repeated the experiment on the $\langle 111\rangle$ neck of Au. In this case the zeros of the oscillations were found to move by ≤ 1 mT in the range $1 \leq T \leq 5$ K. To determine $\Delta(T)$ accurately, we performed fits to the data in B ranges of one beat period centered on beat minima. The fitted model includes two terms of the standard LK form and their second harmonics. The fits at the lowest T were made with both $F_{\uparrow,\downarrow}$ and $\phi_{\uparrow,\downarrow}$ free to vary. $F_{\uparrow,\downarrow}$ were then fixed for the fits at higher T, while $\phi_{\uparrow,\downarrow}$ remained free to vary. To calculate the fractional change in Δ [Eq. (4)], we need to correct $F_\uparrow(T_1, B) - F_\downarrow(T_1, B)$ for the field dependence of $F_{\uparrow,\downarrow}$ [12].

In Fig. 3(a) we plot $\delta\Delta(T)/\Delta(0)$ calculated from the best-fit values of $\delta\phi_{\uparrow,\downarrow}$ using $T_1 = 0.5$ K as the reference temperature. The measured change in magnetization $\delta M(T)/M(0)$ [14] is also shown as a solid line. The strik-

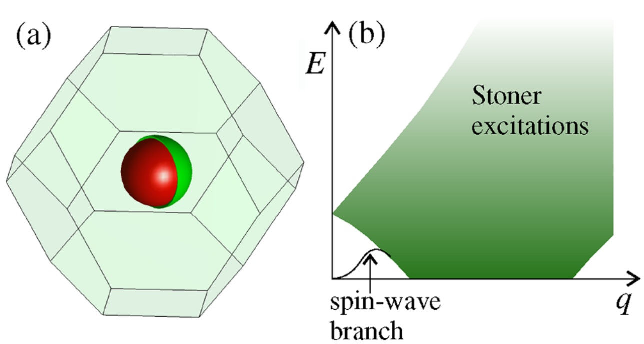

FIG. 1 (color online). (a) Spin-split Fermi surfaces of the Γ pocket in $ZrZn_2$ which give rise to the α (spin-$\downarrow$) and β (spin-$\uparrow$) dHvA components. (b) Sketch of the magnetic excitation spectrum in a conventional itinerant ferromagnet. The shaded area is the Stoner continuum of spin-flipping single-particle excitations. Interactions between Stoner excitations create low-lying transverse modes (spin waves), shown as a single branch. Our results suggest that interactions also create over-damped longitudinal modes at low energy (not shown).

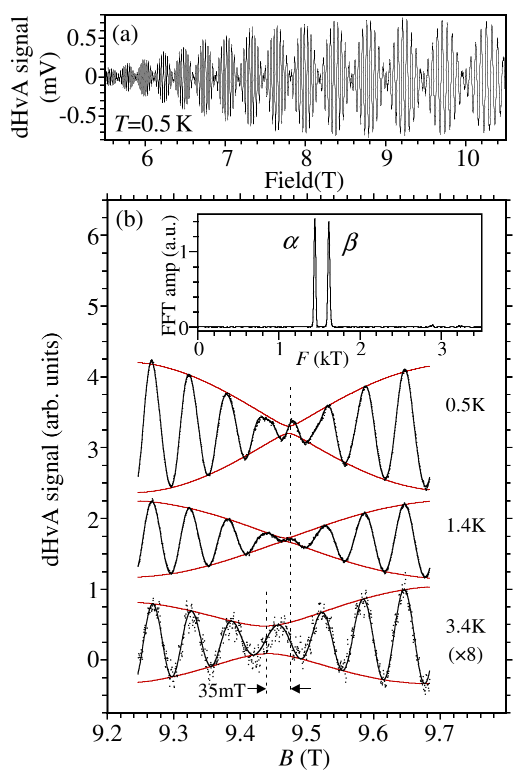

FIG. 2 (color online). (a) Raw dHvA data from a field sweep 5.5 → 10.5 T at 0.5 K. Beating oscillations from the α and β orbits dominate the signal. (b) T dependence of the dHvA data near a beat minimum. The points show raw dHvA signal at various T. Solid lines are fits of a two-component dHvA model to the data (see text). Red lines indicate the beat envelope (fundamental oscillations only) from the fit. The beat minimum shifts down in field as T increases, showing that $\Delta(T)$ is falling with T. The T dependence of this shift, determined by the fits, allows calculation of $[\Delta(T) - \Delta(0)]/\Delta(0)$ which is plotted in Fig. 3. Inset: Fourier transform of the data in panel (a).

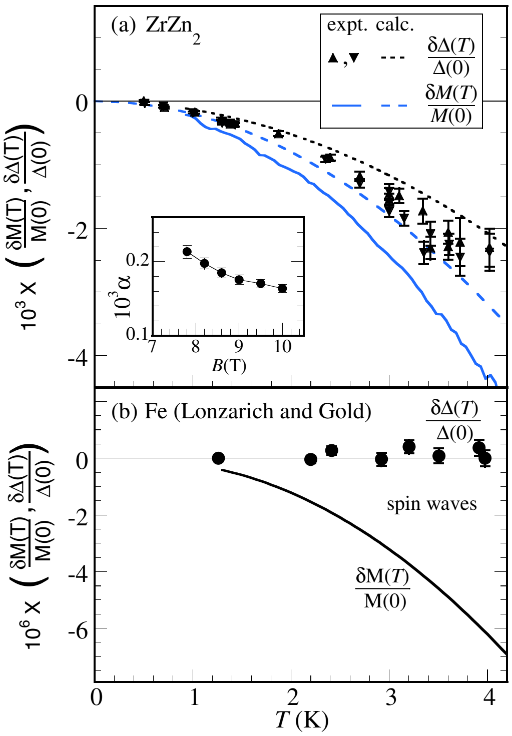

FIG. 3 (color online). Low temperature reduction of $\Delta(T)$ and $M(T)$ in $ZrZn_2$ and Fe. Panel (a) shows $\delta\Delta(T)/\Delta(0)$ at $B = 9.5$ T for $ZrZn_2$ calculated from dHvA measurements (this work). Results from up (▲) and down (▼) sweeps are shown. The solid line is the measured $\delta M(T)/M(0)$ from Ref. [14]. The dashed and dotted lines are, respectively, $\delta M(T,B)/M(0,B)$ and $\delta\Delta(T,B)/\Delta(0,B)$ calculated at $B = 9.5$ T using a dynamical Ginzburg-Landau model (see text). The inset shows the T^2 coefficient from fits to $\delta\Delta(T)/\Delta(0)$ at other beat minima. For $B < 7.8$ T signal to noise was too low to extract the phase reliably. Panel (b) shows results for Fe [4,15] where no decrease of Δ was resolved for $T < 4.2$ K.

ing result is that there is a pronounced decrease in $\Delta(T)$, and it is close to the change in $M(T)$ over the same temperature range. This means that in $ZrZn_2$, most of the reduction in the magnetization on raising the temperature is due to the concomitant reduction of Δ. This behavior contrasts with that expected from the Bloch $T^{3/2}$ law and the results of similar experiments on Fe [4,15] [see Fig. 3(b)]. In Fe it is found that $\Delta(T)$ is constant within experimental error for $1 \leq T \leq 4.2$ K, and $\delta\Delta(T)$ is at least an order of magnitude smaller than $\delta M(T)$. We can understand the significance of our result by remembering that the thermal excitation of spin waves does not affect Δ in first order [3]. Thus there must be other spin-flipping processes present in $ZrZn_2$ in addition to spin waves. Stoner excitations which cause a repopulation of spin-$\uparrow$ and spin-$\downarrow$ Fermi surfaces are a likely candidate [2].

We now discuss the dramatic collapse of $\Delta(T)$ in terms of the dynamical Ginzburg-Landau model of Lonzarich and Taillefer [16] used to describe weak ferromagnets such as $ZrZn_2$. The model uses a classical vector order parameter $\mathbf{M}$ which is allowed to fluctuate about its mean value $M\hat{\mathbf{z}}$, such that $\mathbf{M}(\mathbf{r},t) = M\hat{\mathbf{z}} + \mathbf{m}(\mathbf{r},t)$. $\mathbf{m}$ is a zero-meaned stochastic function characterized by the variance of its components, $\langle m_\parallel^2\rangle$ and $\langle m_\perp^2\rangle$, parallel and perpendicular to $\hat{\mathbf{z}}$. The angle brackets $\langle\ldots\rangle$ denote an average in time and space over the thermally induced fluctuations [16]. The free energy $\mathcal{F}$ is expanded in ensemble averages of even powers of $\mathbf{M}(\mathbf{r},t)$ [17]. Setting $\partial\mathcal{F}/\partial M = 0$ gives the equation of state

$$\frac{B}{bM} = \frac{a}{b} + 3\langle m_\parallel^2\rangle + 2\langle m_\perp^2\rangle + M^2, \qquad (5)$$

where a and b are the coefficients of the Ginzburg-Landau free energy which are renormalized with respect to the Landau coefficients of pure Stoner theory.

In conventional Stoner theory [2] we assume $\delta\Delta \propto \delta M$. This approximation slightly overestimates $\delta\Delta(T)$ in $ZrZn_2$ [Fig. 3(a)] and gives completely the wrong answer in Fe [Fig. 3(b)]. A better approximation [18] in the limit when small wave vector and low-energy excitations are thermally excited is

$$\delta\Delta \propto \delta\langle|\mathbf{M}(\mathbf{r})|\rangle. \qquad (6)$$

In this approximation Δ is sensitive only to the local degree of spin polarization and not to the direction of the quantization axis. The effect of thermally excited fluctuations on $\langle|\mathbf{M}(\mathbf{r})|\rangle$ can be seen from the expansion,

$$\begin{aligned}\langle|\mathbf{M}(\mathbf{r})|\rangle^2 &= \langle\sqrt{[M\hat{\mathbf{z}} + \mathbf{m}(\mathbf{r})]\cdot[M\hat{\mathbf{z}} + \mathbf{m}(\mathbf{r})]}\rangle^2 \\ &= \left\langle M\sqrt{1 + \frac{2m_\parallel(\mathbf{r})}{M} + \frac{m_\parallel^2(\mathbf{r})}{M^2} + \frac{2m_\perp^2(\mathbf{r})}{M^2}}\right\rangle^2 \\ &= M^2 + 2\langle m_\perp^2\rangle + \ldots, \qquad (7)\end{aligned}$$

where all terms in odd powers of $\mathbf{m}(\mathbf{r})$ vanish when the average is taken. Equation (7) expresses the geometrical fact that adding a random transverse fluctuation to a fixed vector $M\hat{\mathbf{z}}$ gives on average a larger resultant moment, but adding a fluctuation that is longitudinal to $M\hat{\mathbf{z}}$ does not affect the mean value.

Equation (5) determines the equilibrium value of M as a function of B, $\langle m_\parallel^2\rangle$, and $\langle m_\perp^2\rangle$. It follows from Eq. (5) with $B = 0$ that $M^2(T,0) = M^2(0,0) - 3\langle m_\parallel^2\rangle - 2\langle m_\perp^2\rangle$. Substituting this into Eq. (7) gives $\Delta^2(T,0) \propto M^2(0,0) - 3\langle m_\parallel^2\rangle$. Thus both longitudinal and transverse fluctuations reduce M but, at zero field, only longitudinal fluctuations affect Δ. To interpret our dHvA results, we need to solve the cubic equation [Eq. (5)] for $B > 0$. As our measurements are made at very low T where $\delta M(T,B)/M(0,B) \ll 1$, we expand about $\langle m_\parallel^2\rangle = \langle m_\perp^2\rangle = 0$ in $\langle m_{\parallel,\perp}^2\rangle/M^2$ to order $\langle m_{\parallel,\perp}^2\rangle/M^2$, and find

$$\frac{\delta M(T,B)}{M(0,B)} = -\frac{3\langle m_\parallel^2\rangle + 2\langle m_\perp^2\rangle}{2zM(0,B)^2}. \qquad (8)$$

Substituting into Eq. (7) gives

$$\frac{\delta\langle|\mathbf{M}(\mathbf{r})|\rangle}{M(0,B)} = -\frac{3\langle m_{\parallel}^2\rangle + 2(1-z)\langle m_{\perp}^2\rangle}{2zM(0,B)^2}, \qquad (9)$$

where z is defined as $z(B) = \frac{3}{2} - \frac{1}{2}[M(0,0)/M(0,B)]^2$. The effects of applying a magnetic field are (i) to decrease δM and $\delta\Delta$ for a given fluctuation amplitude and (ii) to introduce some dependence of $\delta\Delta$ on the transverse fluctuation amplitude. From measurements of $M(B)$ [13] we find $z(9.5\ \mathrm{T}) = 1.30$.

The most robust prediction of the present model is the ratio between $\delta\Delta$ and δM. Provided the fluctuations are approximately isotropic ($\langle m_{\perp}^2\rangle = \langle m_{\parallel}^2\rangle$), Eqs. (8) and (9) show that the ratio of $\delta\Delta(T,0)/\Delta(0,0)$ and $\delta M(T,0)/M(0,0)$ equals 3/5. The corresponding ratio at $B = 9.5$ T is 0.47. The dotted line in Fig. 3 shows $0.47 \times \delta M/M(0,B)$ from the results in Ref. [14]. This simple parameter-free model gives a result that is in fair agreement with $\delta\Delta(T)/\Delta(0)$ calculated from our dHvA measurements. In the case of Fe the saturated nature of the moment means that $\langle m_{\parallel}^2\rangle = 0$ and $M(0,0) = M(0,B)$, yielding $\delta\Delta(T) = 0$ at low temperatures as observed in Fig. 3(b).

A second quantitative test of the model is the absolute magnitude of $\delta M(T,B)/M(0,B)$. For this we need to make an assumption about the T and B dependence of the fluctuation amplitudes. The onset of FM in the Landau model is marked by the divergence of $\chi_{\parallel} = \partial M/\partial B$ at $M = 0$. Putting this condition into Eq. (5) determines the magnitude of the isotropic fluctuations at T_C, $\langle m_c^2\rangle = M^2(0,0)/5$. The detailed calculations of Ref. [16] suggest $\langle m_{\parallel}^2\rangle$ and $\langle m_{\perp}^2\rangle$ are approximately proportional to T^2 throughout the FM state so we set $\langle m_{\parallel}^2\rangle = \langle m_{\perp}^2\rangle = (T^2/T_C^2)M^2(0,0)/5$ in Eq. (8). The result is $\delta M(T,0)/M(0,0) = 6.6 \times 10^{-4}T^2$ and $\delta M(T, 9.5\mathrm{T})/M(0, 9.5\mathrm{T}) = 1.9 \times 10^{-4}T^2$. The latter underestimates the drop of the exchange splitting by about 33% [see dashed line in Fig. 3(a)]. In making this comparison it should be remembered that this is a parameter-free model and a number of approximations have been made. These include neglecting the field dependence of $\langle m_{\parallel}^2\rangle$ and $\langle m_{\perp}^2\rangle$ and any underlying temperature dependence of the Ginzburg-Landau parameters a and b.

In summary, we have shown that there is a rapid initial collapse of the exchange splitting Δ with increasing temperature in $ZrZn_2$ at low temperatures. To our knowledge, this is the first observation of such a collapse in an itinerant ferromagnet. The only other materials studied by the dHvA technique used here are Fe [4], Ni [19], and Au [20]. In each case, temperature induced changes in the Fermi surface volume were several orders of magnitude smaller than in $ZrZn_2$ or not observable. In $ZrZn_2$, the observed collapse of Δ and the concomitant reduction of the magnetization M with increasing temperature suggests that the dominant spin-flipping process is the repopulation of the spin-$\uparrow$ and the spin-$\downarrow$ Fermi surfaces. Our observations support the notion that longitudinal fluctuations (excitations) of the local magnetization are important in $ZrZn_2$ [6]. The special feature of $ZrZn_2$ is that it is one of the few examples of a "quantum itinerant ferromagnet", i.e., an itinerant ferromagnet close to quantum criticality. Thus it would be interesting to see whether similar behavior is observed in related materials such as MnSi and Ni_3Al.

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