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2008 J. Phys.: Condens. Matter 20 475208

(http://iopscience.iop.org/0953-8984/20/47/475208)

IOP Publishing — Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter **20** (2008) 475208 (5pp) doi:10.1088/0953-8984/20/47/475208

Field-dependent AC susceptibility of itinerant ferromagnets

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Received 12 September 2008, in final form 12 October 2008
Published 6 November 2008
Online at stacks.iop.org/JPhysCM/20/475208

Abstract
Whereas dc measurements of magnetic susceptibility, χ, fail to distinguish between local and weak itinerant ferromagnets, radio-frequency (rf) measurements of χ in the ferromagnetic state show dramatic differences between the two. We present sensitive tunnel-diode resonator measurements of χ in the weak itinerant ferromagnet $ZrZn_2$ at a frequency of 23 MHz. Below the Curie temperature, $T_C \approx 26$ K, the susceptibility is seen to increase and pass through a broad maximum at approximately 15 K in zero applied dc magnetic field. Application of a magnetic field reduces the amplitude of the maximum and shifts it to lower temperatures. The existence and evolution of this maximum with applied field is not predicted by either the Stoner or self-consistent renormalized (SCR) spin-fluctuation theories. For temperatures below T_C both theories derive a zero-field limit expression for χ. We propose a semi-phenomenological model that considers the effect of the internal field from the polarized fraction of the conduction band on the remaining, unpolarized conduction band electrons. The developed model accurately describes the experimental data.

(Some figures in this article are in colour only in the electronic version)

DC measurements of magnetic susceptibility fail to distinguish between local and itinerant ferromagnets. A common method of determining χ_{dc} is measuring the magnetic moment and then dividing by the applied field, H. However, this is only applicable provided the magnetization is linear in H from $H = 0$ up to the measurement field. For magnetically soft or small moment ferromagnets, this criterion may not be satisfied. Perhaps a more careful method is to measure M in two slightly different magnetic fields and then calculate $\Delta M/\Delta H$ as shown in figure 1. Further, due to limited sensitivity, DC measurements are usually conducted in significant magnetic fields, on the order of 1–10 Oe. In exceptionally soft materials these fields may be sufficient to smear certain zero-field features.

In itinerant systems the situation is even more complicated. In order to deduce the size of a magnetic moment per ion, one has to be in a saturation regime by applying a large field. However, this tells us nothing about the magnitude of this moment in zero field that, in itinerant systems, is field dependent. Yet, even with a conventional definition, observation of a magnetic moment that is a fraction of a Bohr magneton per ion is not bulletproof evidence for the itinerant nature of magnetism. For example, the local moment metallic rare-earth compound $CeAgSb_2$ [1] and the insulating titanate $YTiO_3$ [2] both possess a fractional magnetic moment per ion. Possible explanations of such fractional local moments could be a canted antiferromagnetic structure as may occur in $YTiO_3$ [3], or crystalline electric field effects as has been proposed for $CeAgSb_2$ [4].

Compared with dc measurements, ac susceptibility can be measured in much lower magnetic fields. It is a valuable technique in studying magnetic materials. However, interpretation of the results, especially in itinerant systems, is complicated. For example, low frequency ac susceptibility measurements on the insulating two dimensional ferromagnet K_2CuF_4 [5] are strikingly similar to those measured in palladium slightly doped with manganese [6] as well as in an Fe–Ni–B–Si alloy [7]. While it is clear that the insulating compound is a local moment, the nature of the latter two is open to debate. It is suggested in these works that the similarities in the data are due to a combination of demagnetization effects and domain-wall motion.

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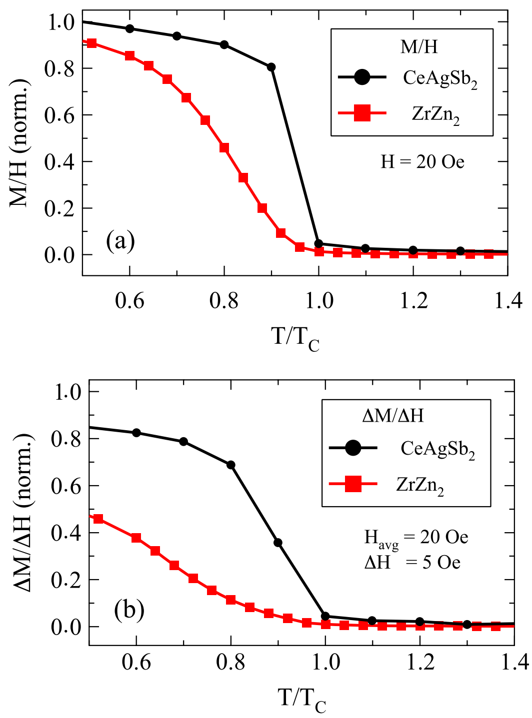

Figure 1. Comparison of normalized measurements of dc susceptibility of the 4f local moment ferromagnetic $CeAgSb_2$ (black circles, $T_C \approx 9.8$ K) and the weak itinerant ferromagnet $ZrZn_2$ (red squares, $T_C \approx 27$ K) on a reduced temperature scale. (a) The conventional dc susceptibility, M/H. (b) The result of dc delta measurements as explained in the text. Both sets of measurements were carried out with $H_{avg} = 20$ Oe. Normalization is relative to the 5 K value of M/H and performed for each sample separately.

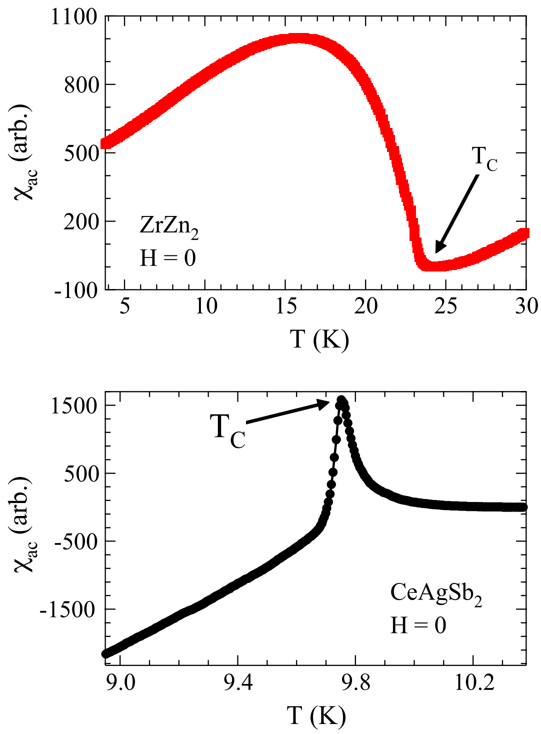

Figure 2. Radio-frequency susceptibility of the weak itinerant ferromagnet $ZrZn_2$ (top) and the 4f local moment ferromagnet $CeAgSb_2$ (bottom) in zero applied field. T_C marks the feature at the Curie temperature for each material. The general decrease in the measured χ (above T_C for $ZrZn_2$, and below for $CeAgSb_2$) is caused by a decrease in resistivity. Note the different temperature scales in each plot.

In this contribution we report the radio-frequency temperature-dependent ac susceptibility of the well studied commonly accepted itinerant ferromagnet $ZrZn_2$, and examine its evolution with an applied magnetic field. We then present a semi-phenomenological model that describes our data. Low frequency ac susceptibility measurements on $ZrZn_2$ have been reported [8]; however, as the focus of that work was not the ferromagnetism of the compound, the only presented data are for $T < 2$ K.

Recently, it has been shown [9, 10] that rf measurements of ac magnetic susceptibility, χ_{ac}, seem to distinguish between local moment and itinerant ferromagnetism. Further, in [9] an explanation ruling out the demagnetization or magnetic domain effects was presented. It is clear from figure 2 that the rf susceptibility of the weak itinerant ferromagnet $ZrZn_2$ [11] is distinctly different from that of the 4f local moment $CeAgSb_2$ [1]. The purpose of this work is to present an effective Weiss-type model to describe the data derived from itinerant ferromagnets.

The design and operation of a tunnel-diode resonator (TDR) are described in detail elsewhere [12–14]. The device is built around a tunnel diode, a semiconducting device with a voltage bias region of negative differential resistance. Biasing to this voltage region allows the tunnel diode to drive an LC tank circuit at its natural resonant frequency. In magnetic measurements, a sample is placed in the coil of the tank circuit, thereby changing the total inductance, and, hence, the tank circuit's resonant frequency. It can be shown [15] that the frequency shift of the tank circuit is directly proportional to the real part of the magnetic susceptibility, χ, of the sample in the coil as

$$\frac{\Delta f}{f_0} \approx -\frac{1}{2}\frac{V_s}{V_c}4\pi\chi_m. \quad (1)$$

Here V_s and V_c are the volumes of the sample and coil, respectively, and χ_m is the measured susceptibility of the sample. Careful design and construction allows one to resolve changes in resonant frequency induced by the sample on the order of 1–10 mHz. The resulting tuned circuit, operating at 10–20 MHz, gives frequency sensitivity on the order of a few parts per billion. This translates to a typical sensitivity of 10^{-7}–10^{-8} change in χ induced by temperature or magnetic field. In order to measure the absolute value of χ it is necessary to know the difference between the empty coil resonance and the resonance with the sample in place at the measurement temperature. However, in order to obtain the sensitivity quoted, all components must be held at constant low temperature to within approximately 5 mK. This makes performing such a measurement quite difficult, though not impossible. Due to the operating frequency the measured susceptibility is composed of two parts. The first is due to the magnetic moments in the sample, and may be either para or diamagnetic depending on the material studied. The second is due to the screening of an rf field via the normal skin effect in metals. This screening is a diamagnetic contribution and is a measure of changes in resistivity [16].

Radio-frequency susceptibility data presented herein were collected in a TDR operating at 23 MHz mounted in a ^{4}He cryostat. The design is similar to that presented in [9]. The temperature of the sample can be varied from 3 to 100 K and a dc magnetic field up to 2.5 T coaxial with the rf excitation field (∼20 mOe) can be applied with a superconducting magnet. The magnet is mounted inside the vacuum can of the cryostat resulting in no trapped magnetic field at the beginning of each run. As the effects studied herein are completely suppressed by fields on the order of 500 Oe, any such trapped magnetic field could affect the data. Single crystal samples were used in this study. In all data presented herein, the magnetic easy axis was aligned with the rf excitation field and the dc bias field. The $CeAgSb_2$ sample was prepared as described in [1], while the $ZrZn_2$ sample was prepared as described in [17].

Figure 1 compares temperature-dependent dc susceptibility for the local moment $CeAgSb_2$ with that for the itinerant $ZrZn_2$ as measured in a *Quantum Design* MPMS-5. Two different techniques were used to determine these susceptibilities. Panel (a) shows the usual χ_{dc} where the magnetic moment is measured in a small applied field ($H = 20$ Oe) and M/H is calculated. While this method is appropriate for temperatures well above T_C, where magnetization is linearly dependent on field over a fairly large field range, it should be expected to fail in the ferromagnetic state because M is not necessarily linear in H all the way down to $H = 0$. Bearing this in mind, a delta measurement of χ_{dc} was performed (results in panel (b)) as follows. Magnetic moment versus temperature was measured first in a 17 Oe field and then in a 22 Oe field. The difference in the resulting moment was divided by the 5 Oe difference in applied fields to determine $\Delta M/\Delta H$. The advantage of this method is that it only requires approximate linearity in $M(H)$ over the 5 Oe window defined by the upper and lower fields. Thus, it can be expected to approximate $\chi = \frac{dM}{dH}$ more closely. Obviously, a smaller H window is more likely to conform to the linear $M(H)$ approximation.

While the delta measurement results in a lower susceptibility in both samples, both dc techniques produce quite similar $\chi(T)$ curves. This is to be contrasted with the results of zero-field radio-frequency susceptibility versus temperature as shown in figure 2. Whereas the local moment system shows a sharp, well defined peak in χ_{ac} at the Curie temperature, the itinerant system exhibits a broad maximum well below T_C.

Conventional theories of itinerant ferromagnetism fail to predict the behavior seen in the TDR data of $ZrZn_2$. The development of the Stoner theory [18] was driven by a desire to understand how a fractional Bohr magneton magnetic moment could be created in nickel. The failures of Stoner theory, i.e. Curie temperatures that are too high and the lack of a Curie–Weiss susceptibility above T_C, were impetus for the development of the spin-fluctuation theory of Moriya and Kawabata [19]. While spin-fluctuation theory does indeed predict a Curie–Weiss-type paramagnetic state and largely correct the Curie temperatures, neither it nor the Stoner theory adequately describe the broad maximum seen in the rf data. Indeed, both theories derive a strictly zero-field limit of χ just below T_C of the same form,

$$\chi(T < T_C) = \chi_0 \left(1 - \left(\frac{T}{T_C}\right)^n\right)^{-1}. \quad (2)$$

The difference between the two theories is the value of n. For Stoner theory $n = 2$ while for spin fluctuations $n = 1$ [20, 21] (and there is an intermediate regime, where $n = 4/3$ [22]). The Stoner theory does predict a nonzero χ at $T = 0$ and $H = 0$ [23].

A zero-field limit calculation, however, is not representative of a ferromagnetic system below T_C. As the system begins to order there is a nonzero field in the sample from the ordered moments themselves. In itinerant systems this mean field should continue to increase as T decreases and the fraction of spin polarized conduction electrons increases. The increase in the itinerant mean field may be expected to be more dramatic than in a local moment mean field because in the former there are physically more magnetic carriers as temperature decreases, while in the latter there is merely less thermal randomization of the moment directions. To account for the self-field and for the effect of an applied external magnetic field we propose a modified Brillouin function for a spin-1/2 system. We choose a spin-1/2 system because in itinerant systems it is the single electron spin that is of interest.

$$m^*(t, h) = m_0^* \tanh \frac{h}{1 - t^n}. \quad (3)$$

Here $t = T/T^*$, where T^* is a characteristic temperature not necessarily equal to T_C, and h is a dimensionless field term that is assumed to include both the self-field from the magnetization and the applied field. It should be noted that this form does not represent the magnetization of the sample. Typically the conduction band in itinerant magnets is split into spin majority and spin minority subbands. Here a slightly different view is taken. The conduction band is split into a polarized subband, consisting of only one type of spin, and a compensated subband, consisting of equal numbers of spin up and spin down electrons. The former subband gives rise to the magnetic moment. m^* may be taken as the population of the compensated subband. In the absence of a theory that gives clear $\chi(T)$ curves in different fields it is necessary to have some starting point. The denominator of the tanh argument in equation (4) is chosen, so that susceptibility is compatible with zero-field limits predicted for magnetic susceptibility by Stoner and spin-fluctuation models. Moreover, if we plot $(1 - m^*)$, it behaves almost exactly as the observed magnetization as a function of temperature. However, *dynamic* magnetic susceptibility is given by unpolarized electrons represented by m^*. Differentiating equation (3) with respect to h gives

$$\chi(t, h) = \frac{\chi_0}{1 - t^n} \cosh^{-2} \frac{h}{1 - t^n}. \quad (4)$$

In the limit $h \to 0$, this reduces to equation (2) if $T^* \to T_C$, providing the proper zero-field limit for χ. The advantage is that there is a parameter in the expression for susceptibility that can be tuned to consider the effect of a magnetic field.

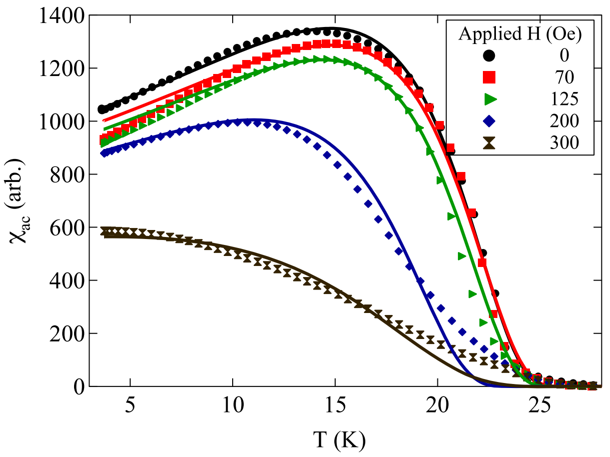

Figure 3. Comparison of data (points) and fits (solid lines) from the model presented in equation (4) with $n = 1$. For clarity only every seventh data point is shown. In all fits, R^2 values were greater than 0.98.

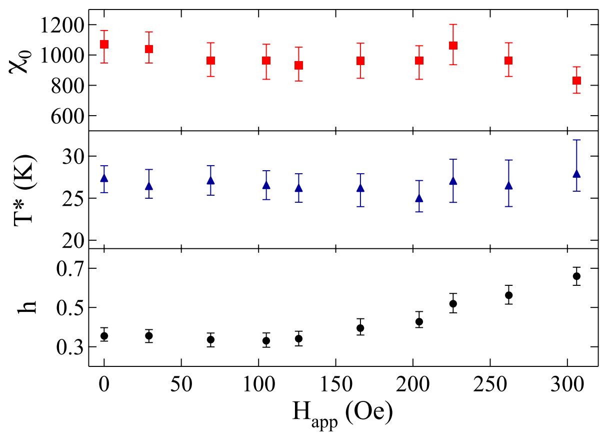

Figure 4. Values of fitting parameters χ_0, T^*, and h (top to bottom) for $ZrZn_2$ single crystal TDR data as functions of applied field (H_{app}) derived from equation (4). Errors were determined by individually varying the fit parameters until the R^2 value dropped below 0.95.

To account for the resistivity contribution to the measured susceptibility, data collected in a dc field of 1 kOe were subtracted from lower field runs. This field was sufficiently large to completely suppress the maximum in χ_{ac} while being small enough that it is not expected to result in a significant magnetoresistance.

Data fits to the model were attempted for various values of n. It was found that $n = 1$, corresponding to the spin-fluctuation theory, gave the best agreement. Figure 3 shows best fits of equation (4) to the TDR data for $ZrZn_2$. The resulting values of the fitting parameters are shown in figure 4. χ_0 decreases with applied H. T^* is constant within the errors and it is approximately equal to T_C. The value of h is approximately constant up to applied fields of about 125 Oe and thereafter grows monotonically as H is increased.

In weak itinerant ferromagnets, like $ZrZn_2$, the susceptibility in the ferromagnetic state is dominated by contributions from the polarized fraction of conduction electrons and the unpolarized fraction. This second contribution comes from a band with a large Stoner enhancement, so it should be expected to have a correspondingly large susceptibility. We suggest that our model accounts for the behavior of the unpolarized portion of the conduction band.

The operating frequency in this study is commensurate with domain-wall resonance techniques [24]. However, in $ZrZn_2$, dc magnetization measurements suggest that single crystals are forced into a single domain in fields as small as 30 Oe [25], effectively ruling out any domain-wall motion.

It would be advantageous to have frequency resolved measurements. However, the technique used here does not lend itself to the decade changes of frequency needed to draw any reasonable conclusions. The actual frequency used here corresponds to an excitation energy on the order of 100 neV. Typical Stoner excitations are on the order of meV. The tacit assumption of the model presented here is that the unpolarized, fluctuating component of the conduction band is what is actually probed. In principle, such fluctuations can have very low energy. Still, to resolve the differences between the two processes requires an increase of frequency by four orders of magnitude.

In conclusion, we have presented rf *dynamic* magnetic susceptibility measurements on the weak itinerant ferromagnet $ZrZn_2$ in various applied dc fields. While the results themselves are interesting, indicating significant dynamic polarizability of the electronic subsystem, they also show that zero-field limit expressions for χ predicted, for example, by Stoner and spin-fluctuation theories are insufficient to explain the data. A Weiss-like model based on the assumption that the rf response of the itinerant ferromagnet is dominated by the fluctuating, unpolarized fraction of the conduction band was shown to fit the data quite well. It is hoped that these data will spur theoretical effort in understanding the dynamic properties of the ferromagnetic state of such systems.

Acknowledgments

We thank P C Canfield and G Lapertot for supplying the samples, and J Schmalian, P C Canfield, S L Bud'ko, G Samolyuk, and V Antropov for fruitful discussions. Work at the Ames Laboratory was supported by the Department of Energy, Office of Basic Energy Sciences, under contract No DE-AC02-07CH11358. RP acknowledges support from the Alfred P Sloan foundation.

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