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Nonlinear Pauli Susceptibilities in $Sr_3Ru_2O_7$ and Universal Features of Itinerant Metamagnetism

B. S. Shivaram, Jing Luo, and Gia-Wei Chern
Department of Physics, University of Virginia, Charlottesville, VA. 22901, USA.

Daniel Phelan
Materials Science Division, Argonne National Labs, Lemont, IL. 60637.

R. Fittipaldi and A. Vecchione
CNR-SPIN, Fisciano, Salerno I-84084, Italy
and
Dipartimento di Fisica "E.R. Caianiello",
Universit di Salerno, Fisciano, Salerno I-84084, Italy
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We report, for the first time, measurements of the third order, χ_3 and fifth order, χ_5, susceptibilities in an itinerant oxide metamagnet, $Sr_3Ru_2O_7$ for magnetic fields both parallel and perpendicular to the c-axis. These susceptibilities exhibit maxima in their temperature dependence such that $T_1 \approx 2T_3 \approx 4T_5$ where the T_i are the position in temperature where a peak in the i-th order susceptibility occurs. These features taken together with the scaling of the critical field with the temperature T_1 observed in a diverse variety of itinerant metamagnets find a natural explanation in a single band model with one Van Hove singularity (VHS) and onsite repulsion U. The separation of the VHS from the Fermi energy Δ, sets a single energy scale, which is the primary driver for the observed features of itinerant metamagnetism at low temperatures.

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Metamagnetism (MM), the sudden rise of the magnetization at a critical field, is a phenomenon observed in a diverse range of metals, and extensively studied in both d and f-electron based itinerant systems [1]. In many itinerant metamagnets as is the case with heavy fermion materials there is a clear presence of local moments, often antiferromagnetically coupled (as ascertained, for instance, from a high temperature Curie Weiss plot), which however develop a strong net moment at the critical field, B_c. The predominant antiferromagnetic correlations are strongly suppressed in high fields [2] with possibly new ferromagnetic correlations arising [3] in the vicinity of B_c. When the critical field is below the Kondo energy scale MM in heavy fermions can be understood based on an itinerant picture with strongly renormalized quasiparticle bands. In contrast, itinerant electron MM can also be found in systems where there is no clear evidence for local moments [4] as in the case of the metallic oxide $Sr_3Ru_2O_7$ (SRO). Given these distinctions between the two cases it would be natural to ask what universal features exist in various metamagnets and what are the significant departures from such common behavior.

The bilayer ruthenate SRO shows a complex phase diagram where multiple MM transitions may be tuned by varying the angle of the applied field with respect to the crystal axes. Upon increasing temperature, these first-order MM transitions end at critical end points, which are themselves connected by a line of second order phase transitions [5, 6]. Enclosed between these transition lines is an anomalous phase with unusual transport properties. The resistivity in this regime is unusually high and shows anisotropy. Earlier attempts to understand these anomalous behaviors have been focused on the emergence of a nematic phase associated with the MM transitions [8–10]. It has also been proposed that the anomalous phase can be viewed as a magnetic analogue of the spatially inhomogeneous superconducting Fulde-Ferrell-Larkin-Ovchinnikov state [11, 12]. Interestingly, recent magnetic neutron scattering showed that a spin-density-wave phase is induced in this regime of the phase diagram [5], which provides a natural explanation for the strong resistivity anisotropy or the electronic nematic behavior.

Although the nature of the anomalous phase remains to be solved, most theoretical models [8–12] for the MM transitions assume the presence of a Van Hove singularity (VHS) that is proximate to the Fermi surface in SRO. Experimentally, the existence of VHS within a few meV distance from the Fermi level has indeed been observed in recent high resolution ARPES measurements [13]. Indeed, Binz and Sigrist have demonstrated that MM transitions can be produced in a minimum single-band model with a logarithmically divergent VHS close to the Fermi surface [14]. Incorporating weak local Coulomb repulsion between the electrons, they showed that when the magnetic field tunes the Fermi surface of one spin species close enough to the VHS, there is a jump in magnetization. The VHS also plays an important role in models of other metamagnetic materials [15–17], such as the Kondo-lattice model for heavy fermions. For example, divergent density of states (DOS) at the edge of the so-called hybridization gap is a generic feature of quasi-

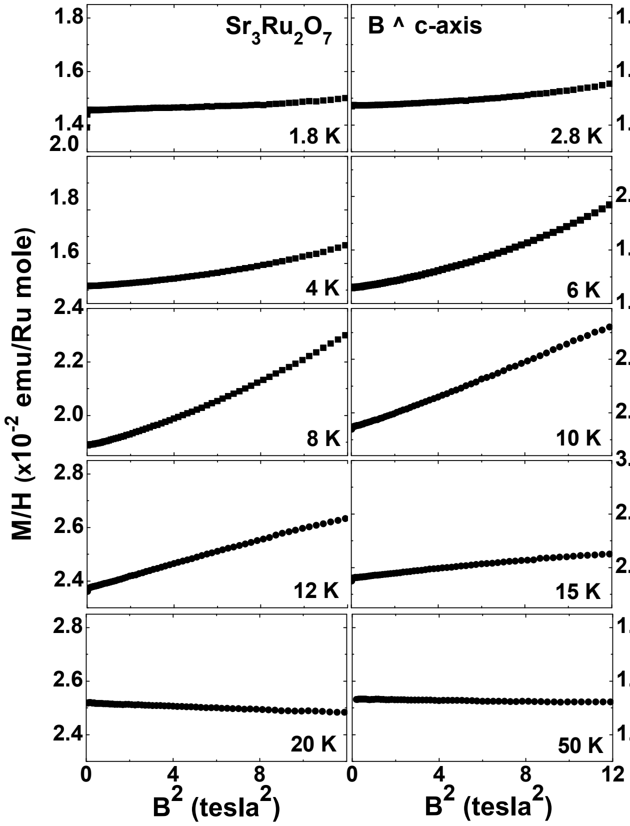

FIG. 1. The magnetization isotherms for B perpendicular to the c-axis plotted as per Eq. (1) for various temperatures as indicated. Since the slope yields χ_3 in such a plot it is immediately seen that χ_3 is weakly negative at high temperatures, $T > 20$ K, turns positive as T is lowered reaching a maximum around 10 K and decreasing thereafter. Also noticeable between 3 K and 8 K is the positive curvature which implies a non-zero (+ve) value for the next higher susceptibility, χ_5.

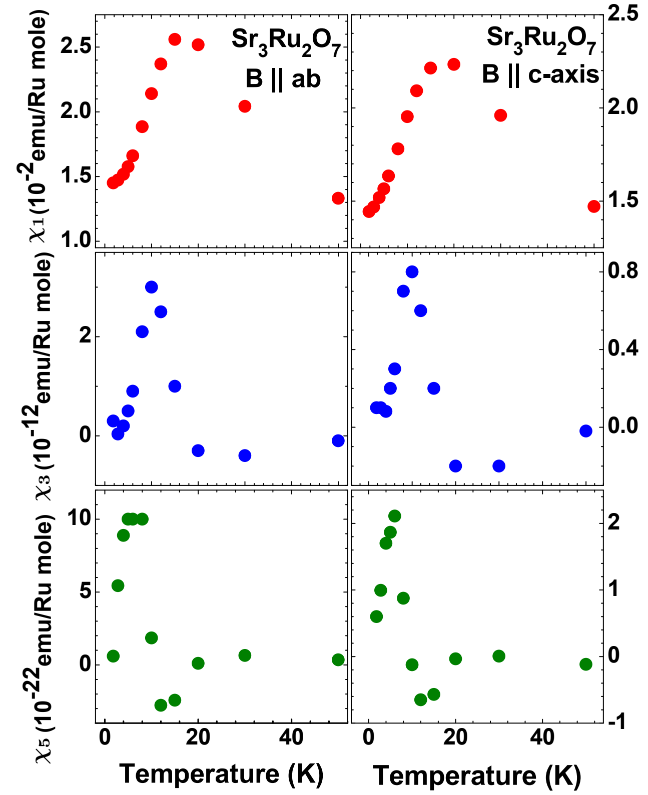

FIG. 2. The values of χ_1, χ_3 and χ_5 obtained from quadratic fits to the lines such as those shown in Fig. 1. Note the distinct peaks in all three of these susceptibilities with each higher order susceptibility exhibiting a positive peak at temperatures successively lower by approximately a factor of two.

particle band structures in heavy fermions [18].

In this paper, we present new measurements of nonlinear susceptibilities in high quality single-crystals of SRO for magnetic fields both parallel and perpendicular to the c-axis of the crystal. The experimental results bear a surprising resemblance to the recent work reported by us on heavy fermions and the universal behavior noted there [19–21]. We show that the universal behavior of the nonlinear susceptibilities is a generic feature of Pauli paramagnetism for electronic systems whose Fermi level lies close to a VHS.

For our study we used single crystals synthesized with a flux growth technique at the University of Salerno [22]. Their quality was checked by X-ray rocking curves and only the best samples were selected for the present work. Measurements of the DC magnetization were carried out at Argonne in a commercial SQUID magnetometer (Quantum Design MPMS-3). The magnetization isotherms obtained at the lowest temperature for B perpendicular to c-axis exhibit from the large jump in the magnetization at 5 T a smaller feature at 5.8 T thus confirming the high quality of our samples in accordance with previous work [23, 24].

In Fig.1 we show the experimental magnetization isotherms plotted in a particular manner so that the extraction of the nonlinear susceptibilities is facilitated. The equilibrium magnetization may be written as an expansion in odd powers of the applied field B as:

$$M = \chi_1 B + \chi_3 B^3 + \chi_5 B^5 \quad (1)$$

Dividing both sides of Eq. (1) by B indicates that a plot of M/B vs B^2 yields a straight line with the intercept giving the linear susceptibility and the slope yielding the leading nonlinear susceptibility χ_3 (provided that χ_5 is negligible). A significant non-zero value of χ_5 would show up as a curvature in the lines in such a plot. It is indeed observed in Fig. 1 that the lines have a negative slope at the high temperature end, hence negative χ_3 as might be expected in any paramagnet. However, the slope turns positive as the temperature is lowered and goes through a maximum at a temperature $T_3 \approx 10K$. It is well known through several previous measurements [7] that the linear susceptibility in SRO has a maximum at a temperature $T_1 = 18$ K. We thus observe that $T_3 \approx 0.5T_1$. Fig. 1 also demonstrates that the next higher order susceptibility

χ_5 which is negligible at $T > 10$ K is non-zero for $T < 10$ K where it has a significant positive value. At the lowest temperature measured $T = 1.8$ K the value of χ_5 is nearly zero again. Thus χ_5 also goes through a maximum albeit at an even lower temperature labelled $T_5 \approx 0.25T_1$. Fig. 2 presents all the three susceptibilities extracted from plots such as those in Fig. 1 for both orientations of the magnetic field. The raw data plots for the parallel orientation are shown in the supplementary section.

Apart from the discussion above several additional points about the behavior of the three susceptibilities shown in Fig. 2 are noteworthy. The linear susceptibility has a large non-zero value as $T \to 0$ as it should be in a Pauli enhanced paramagnet [25, 26] with $\chi_1(0)$ being nearly the same for for both B $\parallel$ c and B $\perp$ c. In contrast the peak values of the nonlinear susceptibilities are significantly different between the two orientations. A magnetic field in the basal plane appears to generate a higher nonlinearity as measured by χ_3 and χ_5.

Experimental results analogous to the above in a completely different family of materials, the f-electron based heavy fermion systems, were considered recently using a simple phenomenological model. In this approach an effective spin-1 Hamiltonian is employed with a large anisotropy term at the single site level [19, 27]. This model reproduced with a remarkable degree of success the experimental correlations such as $T_5 \approx 1/2T_3 \approx 1/4T_1$, the high field magnetic response as well as the ultrasound velocity measurements [21]. While the same model could also be applied to our present work, we note however that this model produced all susceptibilities tending to zero as $T \to 0$, contrary to the observations here as well as with the heavy fermions.

In an attempt to further understand results on the nonlinear susceptibilities, here we investigate the minimum theoretical model that includes a VHS proximate to the Fermi edge and a local Hubbard repulsion U [14, 28]. In the Hartree-Fock mean-field approximation, the Gibbs free energy of the system is given by

$$\mathcal{F} = -T \sum_{\sigma=\uparrow,\downarrow} \int d\varepsilon\, \rho(\varepsilon) \ln\left(1 + e^{-\beta(\varepsilon-\mu_\sigma)}\right) + (\mu_\uparrow n_\uparrow + \mu_\downarrow n_\downarrow) + U n_\uparrow n_\downarrow - Bm, \tag{2}$$

where m is the normalized magnetization along the field direction, $n_{\uparrow,\downarrow} = n/2 \pm m$ are the densities of up- and down-spin electrons, respectively, $\mu_{\uparrow,\downarrow}$ are the corresponding chemical potentials, and $\rho(\varepsilon)$ is DOS. Specifically, we consider a DOS with a logarithmically divergent VHS: $\rho(\varepsilon) = (1/W)\ln|W/(\varepsilon - \varepsilon_{\rm VHS})|$, where the parameter W is of the order of the bandwidth. An important term in the model is $\Delta \equiv \varepsilon_{\rm VHS} - \mu$ which controls the "distance" of the Fermi level to the singularity. For a given external field B, the magnetization is determined from the minimization $\partial\mathcal{F}/\partial m = 0$ subject to the condition that $n_\uparrow + n_\downarrow = n$ [14, 28, 29]. More details can be found in the supplemental information [30].

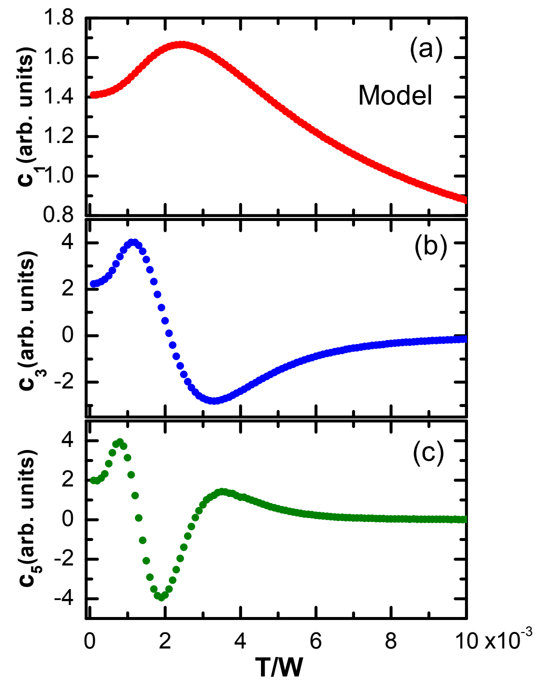

FIG. 3. Shows the calculated susceptibilities χ_1, χ_3, χ_5 in the model in arbitrary units. For the results shown the difference between the chemical potential at $T = 0$ and the van Hove singularity $\Delta/W = (\varepsilon_{\rm VHS} - \mu)/W = 0.006$, and $U/W = 0.16$.

Our numerical calculations find a number of remarkable correlations relevant to the interpretation of our experiments. Fig. 3 shows a comparison of the the calculated susceptibilities from this model with the experimental results of Fig. 2. Qualitatively, the maxima in all the three susceptibilities are reproduced fairly well. Any quantitative discrepancies can be attributed to finer details of the DOS and bandstructures not employed in our calculations. We emphasize that our calculation is based on a minimum yet very general theoretical model which assumes a local Hubbard repulsion and a VHS in the DOS; the model is characterized only by two dimensionless parameters Δ/W and U/W. Consequently, the results summarized in Fig. 4 represent generic behavior of Pauli linear and nonlinear susceptibilities of electron systems whose Fermi level lies close to a VHS. Moreover, we have checked that similar curves are obtained with other types of singularities, e.g. a power-law divergent DOS: $\rho(\varepsilon) \sim 1/|\varepsilon - \varepsilon_c|^\alpha$. Further quantitative agreement may be obtainable if additional structure/energy scale in the DOS and the FS are incorporated into the model. While the choice of a single energy scale gives rise to a sole metamagnetic transition, as noted earlier SRO exhibits multiple closely spaced MM transitions.

It is worth noting that nonlinear susceptibility measurements can offer unique information about the magnetic properties of materials. For example, a negatively divergent $\chi_3(T)$ provides a direct signature of spin-glass transitions [32, 33]. It has also been widely used as a probe of quadrupolar spin fluctuations in rare-earth and heavy fermion compounds [34–36]. However, nonlinear susceptibilities have not been systematically investigated in the context of Pauli paramagnetism within a mean-field treatment of Hubbard interaction. Our calculation here thus provides a tell-tale signature of nonlinear Pauli susceptibilities when the Fermi surface is close to a VHS. In particular, the temperatures T_i at which the maximum χ_i occur display a universal relation $T_5 \approx 1/2T_3 \approx 1/4T_1$, which indeed is observed in many itinerant metamagnets.

We note that the decreasing order of characteristic temperatures $T_1 > T_3 > T_5$ is not uncommon and has indeed been noted in at least two other models [19, 38]. However, we note this relation (or the more precise relation $T_1 \approx 2T_3 \approx 4T_5$) seems to be a characteristic feature of itinerant metamagnetim. For such a relation to be valid the existence of a VHS close to the Fermi level appears to be crucial. For example, the linear Pauli susceptibility computed from a constant DOS (e.g. free electrons in 2D) exhibits no maximum with respect to temperature, while the nonlinear Pauli susceptibilities are negative at low temperatures [37]. At the same time, in at least one system, a frustrated Kagome magnet with localized spins, peaks in the linear and nonlinear susceptibilities are observed at nearly the same temperature [31] .

In parts (a) and (b) of Fig. 4, we show the ratio T_1/T_3 computed for a range of values of the parameters Δ and U. Remarkably we find that the values for this ratio cluster in the range 2.0 to 2.4 over a wide range of Δ (i.e. for $\Delta > 0.01W$) irrespective of the value of U and over a large range of $U < 0.15W$ irrespective of the value of Δ. As shown in Fig. 4(c), very significantly we find that the peak temperature T_1, which depends linearly on Δ, does *not* depend on the value of U for small values of Δ. Furthermore for a noticeable peak to appear in the temperature dependence of the linear susceptibility, the ratio $\chi_1(T_1)/\chi_1(0)$ must be larger than unity. And as shown in Fig. 4(d), this occurs for a value of $U > 0.1W$. Given the experimental value of $\chi_1(T_1)/\chi_1(0) \approx 1.7$ this indicates a Hubbard repulsion $U \approx 0.18W$ for SRO. This value is consistent with those inferred in previous studies, indicating that SRO is a moderately correlated system. In contrast the heavy fermion systems are generally considered to have a large U.

These theoretical results prompt us to consider to what extent the current model extends to all itinerant MMs. We have already noted the ratio T_3/T_1 approximates $1/2$ in heavy fermion systems. The empirical correlation of T_1 and B_c is a phenomenon that has been even more widely established (Fig. 5). As stated earlier these correlations were captured in a simple "local" $S = 1$ model [19]. It is remarkable that in the present model where we calculate only the Pauli part, albeit with a specific band structure feature, we find very similar correlations as in the "local" model employed earlier. Such correlations are also reproduced in an infinite range model for clusters of spins worked out recently by Kumar and Wagner [38]. These

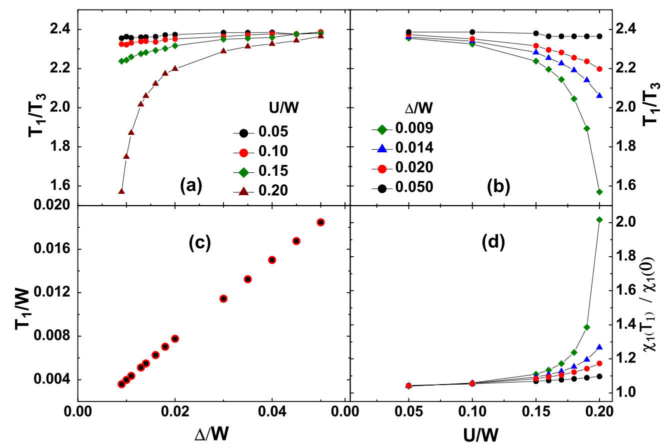

FIG. 4. Parts (a) and (b) show the ratio T_1/T_3 computed in the model for a range of values Δ for specific values of U (left panel) and for a range of values of U for specific values of Δ (right panel). Except for very small $\Delta/W \lesssim 0.01$ and very large $U/W \gtrsim 0.15$ the values of this ratio cluster in the range between 2.4 and 2.0. Part (c) demonstrates that T_1 is independent of U but depends only on Δ. Part (d) shows the ratio $\chi_1(T_1)/\chi_1(0)$ as a function of U for four different values of Δ as indicated.

are unexpected and remarkable coincidences and could very well explain why the diverse set of materials, with varying crystal structures and belonging to different d and f-electron systems (Fig. 5) exhibit the same universal features. From the success of the present theoretical work it appears that a common factor in all these materials is the occurrence of a Van Hove type singularity and its proximity to the Fermi edge. Nevertheless, the significant difference in the Hubbard U i.e. the weak/moderate correlation seen in SRO versus the strong correlation in the HFS with both instances revealing similar evolution of nonlinear susceptibilities needs to be accounted for. In addition other physical effects such as the presence of critical spin fluctuations known to be present in LiV_2O_4 (represented in fig. 5) [39] are not considered in the present model. The linear relation could suggest that the critical metamagnetic field scales with the energy scale of spin fluctuations and and in the case of VHS, this energy scale is determined by the separation between Fermi level and the VHS. In addition multiple energy scales as seem appropriate to Yb based HFS metamagnets [42] will alter the scaling relations noted here [42, 43].

It is also natural to ask whether experimentally there is evidence for the existence of such singularities in DOS uniformly across all systems. As discussed above, recent high resolution ARPES measurements [13] in SRO have shown that such a singularity in close proximity (within a few meV) to the Fermi edge indeed exists. Its observation in heavy fermion systems has also been noted in several systems. In particular in $CeCoGe_2$ the Kondo temperature is very large, ∼250 K, and this facilitates the successful identification/separation of a VHS/Kondo

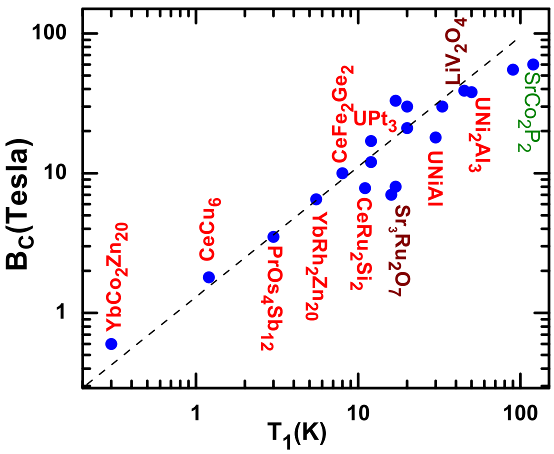

FIG. 5. Shows the linear correlation of T_1 vs B_c in a wide variety of heavy fermions, oxides, and pnictides.

type resonance [40, 41]. Identifying similar features in the vast majority of the compounds referred to in Fig. 5 is an experimental task worth undertaking.

It is clear from the above discussion and the new experimental results on SRO that a simple microscopic model with a single energy scale albeit with a specific band structure feature is successful to a large extent in providing a full explanation of the linear and nonlinear magnetic response of this strongly correlated itinerant MM. It is also remarkable that the observed behavior in a system with no evidence for local moments bears a strong resemblance to our earlier work on heavy fermions. Other correlations related in general to the thermodynamics of MMs originating from the current model will be presented in a forthcoming longer paper.

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