$BaVS_3$: from spin gap insulator to non-Fermi-liquid

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Because of their conveniently low critical temperatures and pressures, f-electron systems have long served as model systems for studying quantum phase transitions and non-Fermi-liquid (NFL) behavior [1]. Recently, the interest has turned to analogous phenomena in d-electron systems, such as ruthenates and vanadates. The ruthenates are near a magnetic instability [2], while for vanadates and titanates, the interplay of spin and orbital fluctuations is important. We present $BaVS_3$ as a system governed by spin–orbital fluctuations.

Fig. 1 shows the measured, and some extrapolated, features of the phase diagram of $BaVS_3$ in the p–T–B space (T: temperature, p: pressure, B: magnetic field), summarizing results from Refs. [3–5]. Let us first consider the $B = 0$ plane. At atmospheric pressure, there is a metal–insulator transition (MIT) from a bad metal to a non-magnetic Mott insulator at $T_{MI} = 69$ K, and a further transition to long-period antiferromagnetic (AFM) order at $T_X = 30$ K [3,6]. We determined the pressure dependence of T_{MI} up to the $T = 0$ insulator-to-NFL-metal transition at $p_{cr} = 20$ kbar [4].

The $T_{MI}(p)$ phase boundary of the canonical Mott system V_2O_3 [7] is similar to that of $BaVS_3$, but the character of the transition, and of the adjoining phases, is quite different. The MIT of V_2O_3 is of first order, leading to either an AFM insulator or a paramagnetic insulator. The latter shows that on-site correlations are the driving force of the MIT, and intersite correlations are secondary. In contrast, the MIT of $BaVS_3$ is continuous, at least up to $p = 15$ kbar [5]. It is not associated with any known kind of symmetry breaking, though it is certainly accompanied by the change of short-range spin *and* orbital correlations, as shown by the specific heat anomaly [8]. The $T_X < T < T_{MI}$ intermediate insulating phase is a spin–orbital liquid with a spin gap Δ_s.

Comparing the various estimates [5,6,9,10] for the $p = 1$ bar spin gap which range from $\Delta_s \approx 100$ to 200 K, to the standard $\Delta_c \approx 600$ K for the charge gap (a value reconfirmed by our recent TEP measurements), we conclude that $BaVS_3$ is not in the large-U limit of some effective Hubbard model but rather of intermediate U. We might infer that $BaVS_3$ can become an insulator only because it develops simultaneously a spin gap which is, as yet, the best candidate for an "order parameter" of the intermediate phase.[1] We determined the pressure dependence of the spin gap along the entire MI phase boundary, relating it to the characteristic (critical) field obtained from a scaling analysis of the magnetoresistivity data [5]. The 3D phase boundary

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[1] Not literally, since it does not distinguish between degenerate ground states; however, it may be related to the true order parameter like a spin-Peierls gap.

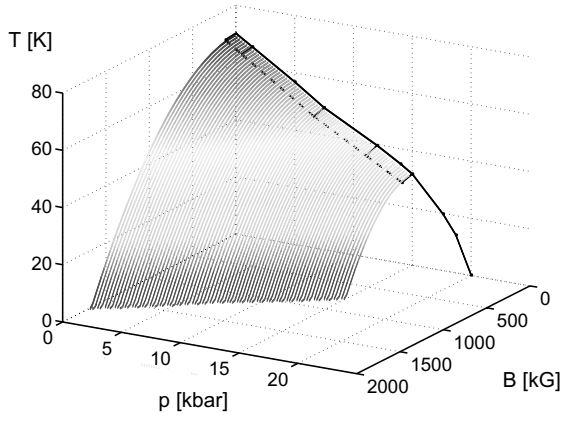

Fig. 1. The phase boundary between the spin–orbital liquid insulator and bad metallic phases in the T–p–B space (based on [4,5]). The bold lines are measured, the thin lines are obtained from a quadratic extrapolation to high fields, or by interpolation. The $T < T_X$ AFM phase is notshown.

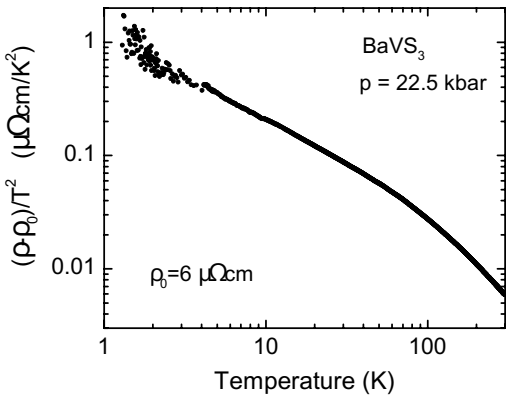

Fig. 2. The divergence of the effective electron–electron scattering amplitude at $p = 22.5$ kbar indicates a non-Fermi-liquid.

shown in Fig. 1 encloses the spin-gapped spin–orbital liquid insulator.

For $p > p_{cr}$, we find a NFL metal [4]. The NFL nature is seen from the fact that the effective electron–electron scattering amplitude $A = (\rho - \rho_0)/T^2$, instead of saturating at low T, increases by atleasttwo orders of magnitude, apparently diverging as T is lowered (Fig. 2). Similar behavior is often found for f-electron systems [11], but it is quite rare with 3d-electrons.[2] Our $p = 22.5$ kbar data show that $\rho \approx \rho_0 + A'T^{1.25}$ holds atleastfor 1 K $< T <$ 40 K. The peak value of $A \approx 1$ μΩ cm/K^2 is 1–2 orders of magnitude lower than for Ce-based alloys [11], indicating that the V d-electrons are not so heavy, but we should keep in mind that $p = 22.5$ kbar is not particularly near to p_{cr}.

The vicinity of a quantum critical point does not necessarily explain why $BaVS_3$ has a NFL state similar to that of nearly AFM f-systems. The customary picture of heavy fermion systems relies on overlapping wide and narrow bands, and there have been attempts to invoke a similar feature for d-electrons, as for the recently discovered NFL ruthenate $La_4Ru_6O_{19}$ [12]. We do not yet have any indication that a similar reasoning should apply to $BaVS_3$.

To conclude, a variety of anomalous conducting and insulating states makes $BaVS_3$ unique among the $3d^1$ vanadium compounds. Assuming that the picture of Mott localization is valid, we infer that there is an exponentially large number of nearly degenerate spin–orbital configurations, and this gives rise to the spin–orbital liquid phase [3]. On the other hand, it is not clear that $BaVS_3$ should be considered as strongly correlated as, say, V_2O_3 is. The resistivity is ruled by an energy scale which is surprisingly small in view of the size of the spin gap, but the system never becomes a good metal. A hitherto unrecognized feature is needed to arrive at a consistent description of $BaVS_3$.

[2] For 4*d* systems, see Refs. [2,12].

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