

Influence of spin fluctuations on the electrical resistance of UAl_2 and UPt_3 at high pressures and low temperatures

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The compounds UAl_2 and UPt_3 are thought to exhibit ferromagnetic spin fluctuations. We have measured the electrical resistance of both materials to over 1000 K at zero pressure and from room temperature to 1 K at pressures up to 18 kbar. The primary effect of pressure is to reduce the low-temperature slope $\partial R/\partial T$ and thus shift the inflection point in the resistance to higher temperatures. We interpret these observations as arising from an increase in the spin-fluctuation temperature T_s with increasing pressure. Using a Fermi-liquid description of these materials, we calculate the pressure dependence of the $T=0$ susceptibility from the pressure dependence of T_s . We also find that the temperature-dependent resistance at various fixed pressures scales as $T/T_s(P)$ over a remarkably large temperature interval.

I. INTRODUCTION

Recently, increased activity has focused on experimental and theoretical descriptions of materials showing nearly magnetic behavior. Within this class of materials are those that display signatures believed to be characteristic of ferromagnetic spin fluctuations,¹ namely a Curie-Weiss susceptibility at high temperatures that evolves with decreasing temperature into enhanced paramagnetism, a low-temperature resistivity proportional to T^2 , and a $T^3 \ln T$ contribution to the low-temperature heat capacity. Both UAl_2 (Refs. 2 and 3) and UPt_3 (Refs. 4 and 5) are known to comply satisfactorily with all of these properties. Of these two, UPt_3 is particularly interesting because of the recent demonstration⁴ that bulk superconductivity and possibly spin fluctuations coexist in this material.

Preliminary band-structure calculations indicate that in both UAl_2 (Ref. 6) and UPt_3 (Ref. 7) the $5f$ electrons form a narrow band at the Fermi energy, a condition favoring spin fluctuations. Decreasing the U-U separation by the application of hydrostatic pressure should increase the $5f$ -wave-function hybridization and broaden the band causing a decrease in the density of states at the Fermi level. Provided pressure increases the f - f Coulomb repulsion less rapidly than it broadens the $5f$ -band width, decreasing the U-U separation should lower the magnetic susceptibility (χ). Such behavior should be reflected as a systematic change with volume in physical properties, e.g., the magnetic susceptibility and resistivity, that depend sensitively on $5f$ -conduction-band interactions. Evidence substantiating this view has been found in susceptibility measurements⁸ on UAl_2 and through resistivity measurements on UPt_3 (Ref. 9) at pressures up to, respectively, 6.65 and 4.2 kbar.

To study the effect of U-U separation on spin fluctuations in UAl_2 and UPt_3 , we have measured the temperature-dependent resistance of polycrystalline UAl_2 and single crystals of UPt_3 (along both the c axis and in the a - b plane) subjected to hydrostatic pressure. We show that the suppression of spin fluctuations with pressure, as

deduced from susceptibility measurements,⁸ is reflected directly in the temperature-dependent resistance and that, over an appropriate temperature interval, the resistance scales with a pressure-dependent parameter which may be identified with the spin-fluctuation temperature.

II. EXPERIMENTAL PROCEDURE

Our polycrystalline UAl_2 sample was prepared by arc melting the pure elements together on a water-cooled copper hearth in a zirconium-gettered argon atmosphere. Needle-shaped single crystals of UPt_3 were grown from a bismuth flux. Both the UAl_2 and the UPt_3 samples were shown to be single phase by x-ray diffraction and metallographic techniques. Because of geometrical considerations, two separate UPt_3 samples were studied, one on which the resistance was measured along the hexagonal c axis and one for measurements in the a - b plane. As a measure of the quality of our samples, the residual resistance ratios for UAl_2 , UPt_3 c axis, and UPt_3 a - b plane were, respectively, 14, 130, and 47. The superconducting transition temperatures for UPt_3 c axis and a - b plane were 0.51 and 0.38 K, respectively. This difference and that of the residual resistance ratios for the two UPt_3 samples result from the annealing process given the c -axis sample.

Resistance measurements were made by a standard four-lead ac technique at 23 Hz. Platinum leads were attached to the samples by spot welding (UAl_2) or by silver-filled epoxy (UPt_3). The samples were subjected to hydrostatic pressures exceeding 17 kbar in a self-clamping pressure cell, described elsewhere,¹⁰ which used a 1:1 mixture of n -pentane and isoamyl alcohol as the pressure medium. The pressure was determined from a lead manometer situated adjacent to the sample. Resistance measurements in the cell were performed in the temperature range $1 \leq T \leq 300$ K, measured by a calibrated carbon-glass thermometer embedded in the side of the cell.

Resistance measurements made above room tempera-

ture were a by-product of heat-capacity measurements performed at zero pressure. The method, described in detail elsewhere,¹¹ relied on a dc transient-pulse-heating technique.

III. RESULTS AND DISCUSSION

We show in Fig. 1 the temperature-dependent resistance of UAl_2 at four different pressures. From measurements on samples of well-defined geometry cut from the same arc-melted button, we estimate the zero-pressure room-temperature resistivity to be approximately $110 \mu\Omega \text{ cm}$. With increasing pressure, the most notable change is a shift of the inflection point to higher temperatures resulting from a decrease in the low-temperature slope $\partial R/\partial T$. Qualitatively similar behavior is found for UPt_3 when measured along the c axis and in the a - b plane (Fig. 2); however, there are notable differences. In particular, the resistance near room temperature is still increasing noticeably with temperature and is not saturated. This difference is seen most clearly in Fig. 3 where we plot the zero-pressure temperature-dependent resistivity of polycrystalline UPt_3 and UAl_2 for temperatures to over 1000 K.¹¹ We see that the resistivity of polycrystalline UPt_3 continues to increase above room temperature and eventually passes through a maximum near 800 K, while the resistivity of UAl_2 is nearly saturated at temperatures as low as 200 K. What is not apparent in Figs. 2 and 3 is that the resistivity of UPt_3 is markedly anisotropic. Measurements performed by others⁹ on single crystals of UPt_3 have shown that the room-temperature resistivity along the c axis is about $130 \mu\Omega \text{ cm}$, while the resistivity in the basal plane is estimated to be $240 \mu\Omega \text{ cm}$. A comparably huge anisotropy also has been observed⁵ in the paramagnetic susceptibility of UPt_3 single crystals in which $\chi(T=0)$ in the a - b plane is approximately twice the value measured along the c axis, suggesting that the resistivity and susceptibility may be dominated by the same physical effect.

One feature common to nearly all theoretical models of spin fluctuations is that, for temperature much less than the characteristic spin-fluctuation temperature T_s , the resistance is predicted¹² to increase as T^2 , with a slope $\partial R/\partial T^2 \propto 1/T_s^2$. However, if the material is highly disor-

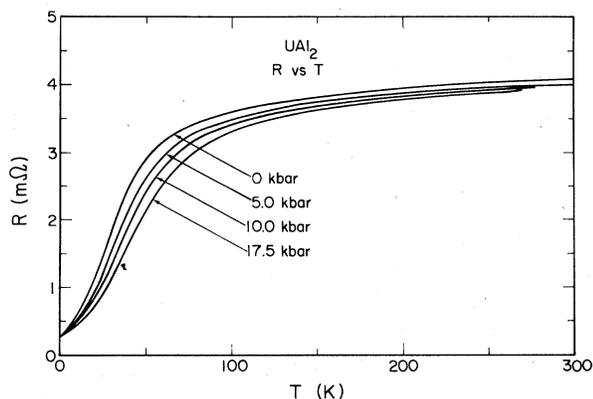


FIG. 1. Resistance as a function of temperature for polycrystalline UAl_2 at four different clamp pressures. For clarity, data at 14 kbar are not shown.

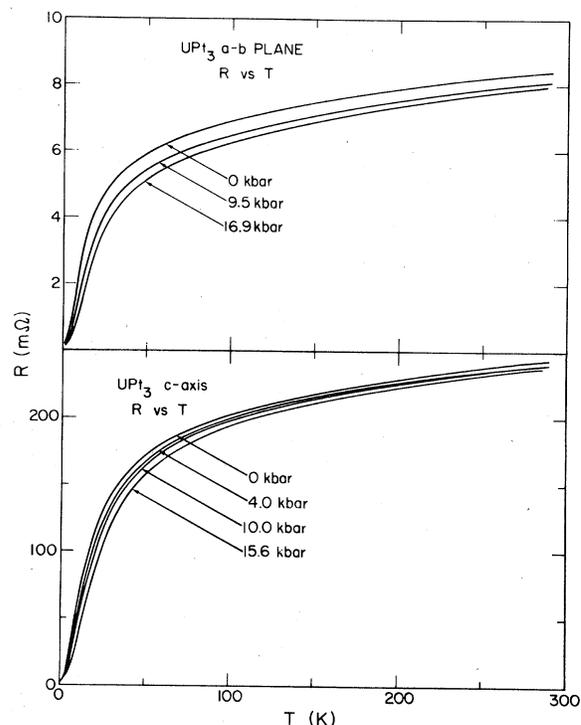


FIG. 2. Resistance vs temperature of single crystal UPt_3 at various fixed clamp pressures. The top set of curves are those measured in the a - b plane and the bottom set measured along the c axis.

dered (large residual resistivity), Riseborough¹³ has shown that one obtains a $T^{3/2}$ dependence of the scattering rate due to spin fluctuations. We have looked closely for a power-law dependence of the low-temperature ($T < 20 \text{ K}$) resistance of UAl_2 and UPt_3 and have found $R \propto T^{3/2}$ in all cases. However, in light of the high residual resistance ratios of our samples, it is not clear that this represents a confirmation of Riseborough's prediction. Indeed, as we shall discuss momentarily, the measured low-temperature dependence may not be a true representation of the ideal (theoretical) resistance when, at higher temperatures, the resistance approaches a saturation limit.

The temperature-dependent resistance of UAl_2 and UPt_3 is reminiscent of that observed in $A15$ compounds¹⁴

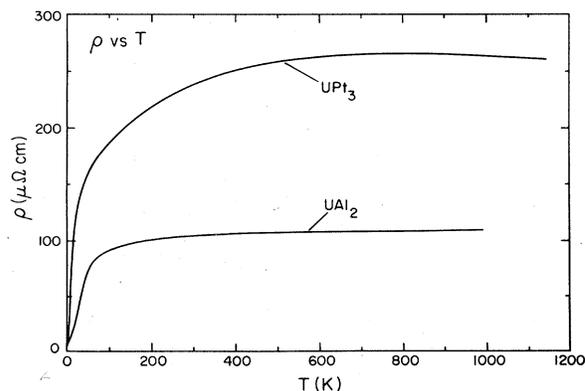


FIG. 3. Resistivity vs temperature for polycrystalline UAl_2 and UPt_3 at zero pressure.

and in some valence-fluctuation materials.¹⁵ In these systems, it is believed that a resistivity saturation limit at high temperatures is reached when the electronic mean free path becomes comparable to an interatomic spacing.¹⁶ Empirically, it has been found^{17,18} that the measured resistance can be described reasonably well as the parallel combination of an ideal resistance R_i and a shunt resistance R_s , whose magnitude is near the saturation value, i.e.,

$$\frac{1}{R} = \frac{1}{R_i} + \frac{1}{R_s}. \quad (1)$$

Although not firmly established, there is some theoretical justification¹⁹ for Eq. (1).

We have applied the "parallel resistor model" [Eq. (1)] to the data of Figs. 1 and 2. Using values of $R_s > R$ (300 K, 0.1 kbar), we find that for each of the samples the "ideal" resistance at low temperatures is proportional to T^2 over an appreciable temperature interval. These results are summarized in Figs. 4–6 where we plot R_i versus T^2 for UAl_2 , UPt_3 c axis, and UPt_3 a - b plane, respectively. The small deviation from T^2 behavior near 4 K in Figs. 4–6 is an experimental artifact due to immersion of the pressure clamp into the liquid-helium bath. Therefore, when allowance is made for saturation effects, the ideal resistance exhibits a temperature dependence in agreement with that expected for a "clean" spin fluctuator. We also note in these figures a clearly systematic decrease in the slope of R_i versus T^2 with increasing pressure that is similar for each sample.

If we assume that R_i in Figs. 4–6 accurately reflects the spin-fluctuation contribution to the resistance, then we may extract the pressure dependence of T_s from the slope $\partial R_i / \partial T^2 = A / T_s^2$. Because we do not have reliably quantitative resistivities for our samples and because theories are at best semiquantitative, we have chosen the proportionality constant A such that at $P=0$ our value of T_s agrees with that determined from other measurements. Once the appropriate normalization is made at $P=0$, we assume that a reasonable description of $T_s(P)$ is provided

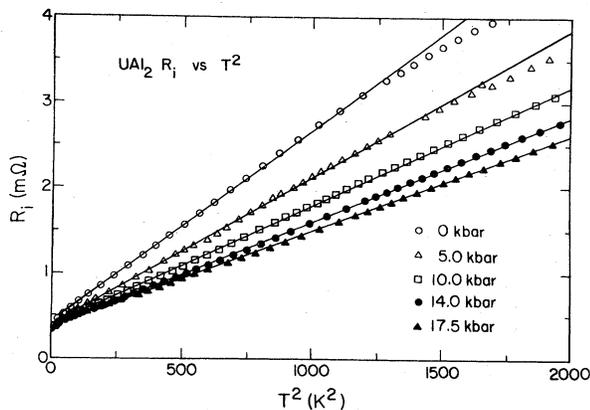


FIG. 4. Ideal resistance as a function of temperature squared for UAl_2 at fixed pressures up to 17.5 kbar. For clarity, only a fraction of the data are shown. To determine the ideal resistance, the value used for R_s in Eq. (1) was 7 mΩ. Lines drawn through the points were used to determine $\partial R_i / \partial T^2$.

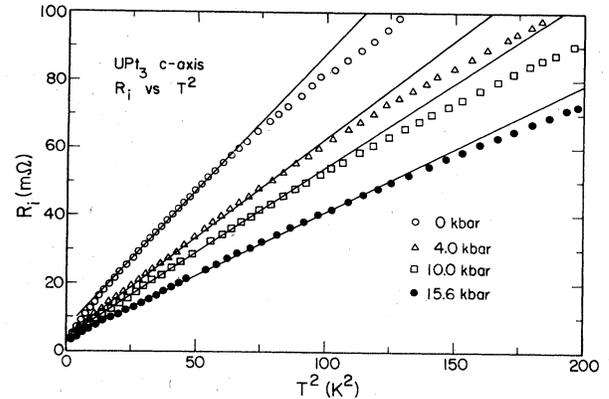


FIG. 5. Ideal resistance vs temperature squared for UPt_3 measured along the c axis at fixed clamped pressures. For clarity, only a fraction of the data points are shown. The shunt resistance used in Eq. (1) was 300 mΩ. Lines drawn through the data were used to determine $\partial R_i / \partial T^2$.

through the pressure dependence of $\partial R_i / \partial T^2$.

Several recent measurements, e.g., specific heat,²⁰ susceptibility,²¹ and magnetoresistance,²¹ place $T_s(P=0)$ at 25–30 K for UAl_2 . We assume the value 26 K. The spin-fluctuation temperature of UPt_3 is less clearly defined. Heat-capacity measurements²² indicate a T_s of 40 ± 25 K for single crystal UPt_3 , while the magnetic susceptibility⁵ shows a departure from Curie-Weiss behavior below ~ 25 K in both the basal plane and along the c axis. Within the framework of a Fermi-liquid description of UPt_3 (which appears to be quite reasonable), one expects the relationship $\chi(T=0) = C / T_s$ to be valid,²³ where C is a constant. The large anisotropy in the $T=0$ susceptibility of UPt_3 (Ref. 5) would then imply that T_s for the c axis is about twice T_s in the a - b plane. If we arbitrarily assume $T_s(P=0) = 42$ K along the c axis, then $T_s(P=0) \approx 20$ K in the basal plane. Because, to our knowledge, there is no theory that treats anisotropic spin-fluctuation scattering, we have no basis justifying this ap-

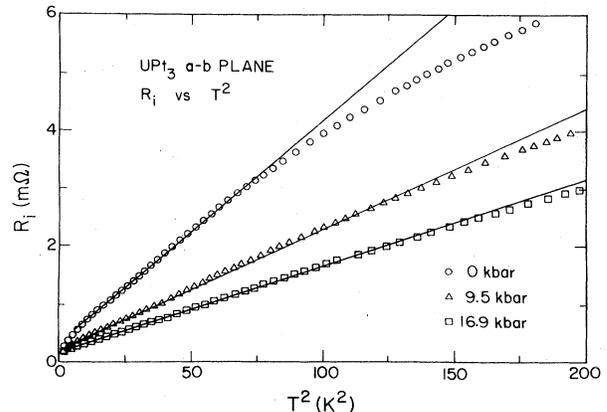


FIG. 6. Ideal resistance vs temperature squared for UPt_3 measured in the a - b plane at pressures up to 16.9 kbar. For clarity, only a fraction of the data points are shown. The shunt resistance used in Eq. (1) was 8.5 mΩ. Lines drawn through the points were used to determine $\partial R_i / \partial T^2$.

proach, although this procedure agrees qualitatively with experimental observations. We emphasize, however, that we attach no significance to the absolute values of T_s but are only using T_s to parametrize the spin-fluctuation characteristics of these materials.

The pressure dependence of the spin-fluctuation temperature, obtained as outlined above, is shown in Fig. 7. We see that T_s increases approximately linearly with pressure for all three samples, suggesting that decreasing the U-U separation drives these materials to a less magnetic state. Again using $\chi(0) = C/T_s$, we can infer the pressure dependence of $\chi(T=0)$ from the slope $\partial T_s / \partial P$, i.e.,

$$\partial \ln \chi(0) / \partial P = -\partial \ln T_s / \partial P. \quad (2)$$

Application of Eq. (2) to the results on UAl_2 shown in Fig. 7 gives

$$\partial \ln \chi(0) / \partial P |_{T \approx 0} = -24 \text{ Mbar}^{-1}.$$

This value agrees remarkably well with the average pressure dependence $\partial \ln \chi / \partial P = -25 \text{ Mbar}^{-1}$ measured directly by Fournier and Beille⁸ and lends credibility to our use of the simple scaling relationship between $\chi(0)$ and T_s . We also point out that our result is *independent* of the value chosen for $T_s(P=0)$. Similar calculations for UPt_3 yield $\partial \ln \chi(0) / \partial P = -30$ and -36 Mbar^{-1} , respectively, for the *c* axis and *a-b* plane. To the extent that Eq. (2) may be applied to UPt_3 , these results imply that pressure suppresses the $T=0$ susceptibility of UPt_3 more rapidly than it does in UAl_2 and that the pressure dependence is anisotropic but not to the extent of $\chi(T=0, P=0)$ or the room-temperature resistivity. [As an aside, we note that a similar analysis of preliminary resistance measurements²⁴ on TiBe_2 under pressure are also consistent with direct measurements²⁵ of $\chi(P)$.]

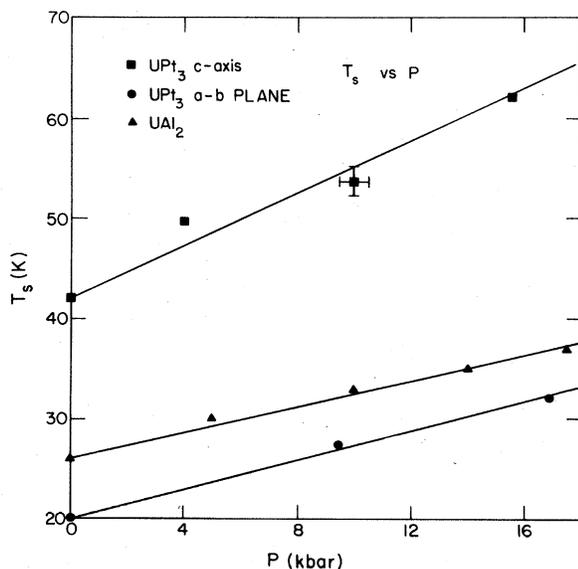


FIG. 7. Spin-fluctuation temperature as a function of pressure for UAl_2 , UPt_3 *c* axis, and UPt_3 *a-b* plane. Values for T_s were deduced from the data presented in Figs. 4–6 as outlined in the text.

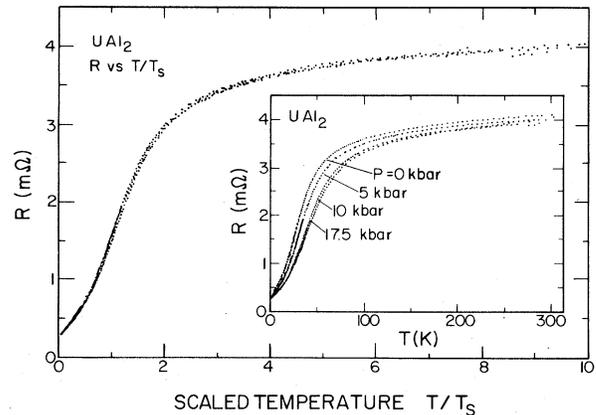


FIG. 8. Resistance vs temperature divided by the pressure-dependent spin-fluctuation temperature for UAl_2 . Values for $T_s(P)$ are given in Fig. 7. Note that this scaling maps all the data at different pressures, shown in the inset and in Fig. 1, onto a single curve.

Because of the way we have derived $T_s(P)$, we expect $R(T, P)$ to be a function of scaled temperature $T/T_s(P)$ over the interval in which $R_i \propto T^2$.¹² However, for UAl_2 this scaling, shown in Fig. 8, holds to at least room temperature. That is, a plot of R versus $T/T_s(P)$ maps all the different pressure curves (Fig. 1) onto a single, unique curve covering the entire temperature range. Such behavior suggests that the resistance is dominated by spin-fluctuation scattering and is consistent with the observation of Fournier and Beille⁸ that the shape of χ versus T is unchanged by pressures up to 6.65 kbar. For UPt_3 , scaling is observed to 50 K for the *c* axis and to 70 K for the *a-b*—plane resistance. We note that quantitatively similar scaling is achieved by using the coefficient of $T^{3/2}$ temperature dependence of the *measured* resistance. Departure from scaling at relatively lower temperatures in UPt_3 may arise from (1) the presence of additional scattering mechanisms, e.g., electron-phonon scattering and/or whatever is responsible for the high-temperature peak in the resistance, (2) a temperature-dependent T_s , (3) thermally induced mixing of the anisotropic spin-fluctuation scattering, or (4) possible combinations of the above.

IV. CONCLUSIONS

We have shown that the pressure dependence of the resistance in UAl_2 and UPt_3 is consistent with an increase in the spin-fluctuation temperature and a concomitant decrease in magnetic scattering as the U-U separation becomes smaller. Using a Fermi-liquid description of these materials, we have presented a new method allowing a determination of $\partial \ln \chi(0) / \partial P$ from resistance measurements on spin-fluctuation materials that is independent of a precise knowledge of $T_s(P=0)$. Results so obtained are

in very good agreement with direct determinations. Finally, we have discovered rather remarkable scaling of the resistance over an extended temperature interval. However, why the scaling regimes in UAl_2 and UPt_3 differ by a factor of 4–5 in apparently similar materials remains an open question. We hope that these results will stimulate additional experimental and theoretical investigations leading to a better understanding of spin-fluctuation behavior in these interesting materials.

Note added in proof. In further support of our observations, we note that others^{9,26} have found the coefficient of the T^2 term in the resistivity to decrease with increasing pressure. In addition, a recent measurement²⁶ of the pres-

sure dependence of χ in polycrystalline UPt_3 has given a value of $\partial \ln \chi(0) / \partial P = -24 \text{ Mbar}^{-1}$, in satisfactory agreement with our results.

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