Suppression of Spin Fluctuations in UAl$_2$ in High Magentic Fields

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Magnetization and magnetoresistivity measurements near 4.2 K on UAl$_2$ in fields up to 35 and 25 T, respectively, show a suppression of spin fluctuations in this material between 15 and 20 T. The experiments strongly suggest that the field and temperature effects on the paramagnon contributions are closely related. The validity of various theoretical models on the suppression of paramagnon effects are also discussed in light of these measurements.

PACS numbers: 72.15.Gd

The compound UAl$_2$ has been known for some time to be a spin-fluctuation (SF) system whose temperature dependence of specific heat,[1] resistivity,[2] and magnetic susceptibility[3] fit neatly into various theories explaining these phenomena.[4-6] However, UAl$_2$ is an exceptional SF system in the sense that the maximum value of its susceptibility is observed at $T = 0$ K. For other SF materials, the temperature dependence at which this maximum is found range from 80 K for Pd (Ref. 7) to 360 K for LuCo$_2$.[8] The characteristic spin-fluctuation temperature (T_{SF}) of UAl$_2$ is readily determined from the quadratic temperature dependence of its electrical resistivity and believed to be about 25 or 30 K, although other estimates from Armbrüster *et al.*[9] have ranged to as low as 4 to 7 K. Interestingly enough, the magnetic field dependence of the transport properties of UAl$_2$ has never been studied at low temperatures where paramagnon effects are expected to be largest. We have undertaken a study of the transport properties since it has been argued that the application of high magnetic fields will reduce paramagnon effects. The Zeeman splitting of the spin-up and spin-down electron states will prevent certain excitations with reversal of spin direction to occur. Since the characteristic paramagnon excitations in UAl$_2$ are of the order of $k_B T_{SF}$, we would expect a strong suppression of these effects at high magnetic fields and low temperatures.

Magnetization and magnetoresistivity measurements near 4.2 K on UAl$_2$ in fields of 35 and 25 T, respectively, show a suppression of the SF. These measurements performed at 4.2 K indicate a freezing out of SF between 15 and 20 T. Magnetization measurements at 77 K show that the SF effects are absent at this temperature.

With $g = 2$ and the relationship between H_{SF} and the characteristic temperature T_{SF} (25 K) through $g\mu_B H_{SF} = k_B T_{SF}$, H_{SF} is calculated to be near 19 T. In consequence, our experiments suggest that the field and temperature effects on the paramagnon contributions to the magnetization and magnetoresistivity are closely related.

Freezing out of SF in magnetic fields has been reported previously for YCo$_2$,[10] LuCo$_2$,[10,11] and Pd.[12] In contrast with these compounds, we have achieved complete suppression of SF in UAl$_2$ (e.g., H_{SF} for Pd is believed to be 190 T!). A similar conclusion may be inferred from the magnetization measurements on TiBe$_2$.[13]

The validity of various theoretical models on the suppression of paramagnon effects is discussed in light of these measurements.

Magnetization measurements up to 35 T were performed on a polycrystalline sample at the high magnetic field installation[14] of the Natuurkundig Laboratorium at the University of Amsterdam. The magnetoresistivity has been measured on a single-crystal rod using a four-point ac technique in a 25-T hybrid magnet at the High-Field Magnet Laboratory of the University of Nijmegen.[15] In addition to the magnetization and magnetoresistivity, the temperature dependence of the suscep-
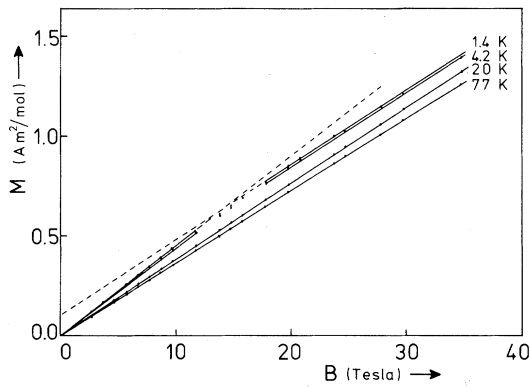
FIG. 1. Magnetization of UAl_2 at 1.4, 4.2, 20, and 77 K.

TABLE I. Molar susceptibilities of UAl_2. The units of $\partial M/\partial H$ are 10^{-9} m^3/mole and the experimental error is of the order of 1%.

$T(K)$	$\partial M/\partial H$ [a]	$\partial M/\partial H$ (2 – 5 T)	$\partial M/\partial H$ (20 – 35 T)
1.4	$\cdots$	56.5	47.3
4.2	55.0	55.4	47.3
20	$\cdots$	47.7	47.7
25	47.3	$\cdots$	$\cdots$
50	46.6	$\cdots$	$\cdots$
77	45.5	45.9	45.9

[a]Reference 3, field value unknown.

tibility was measured in fields of 1.2 and 8.0 T in a sensitive magnetometer.[16]

The samples for the magnetization measurements were prepared by arc melting the appropriate amounts of constituents in a titanium-gettered argon atmosphere. The uranium had a nominal purity of 99.5% (Al, 5 ppm; Fe, 5 ppm; Mn, 11 ppm; Cu, 55 ppm; Co,Ni, < 10 ppm) and the Al was supplied by Johnson and Matthey–England. The ingots were melted, turned over, and remelted five times to increase sample homogeneity. The ingots were annealed at 800 °C for 10 days in an evacuated (2×10^{-6} Torr) sealed tube and subsequently furnace cooled.

The single crystals used for the magnetoresistance measurements were grown from the constituents and were initially melted in an arc furface. The button was then crushed and melted into a rod in a silver boat with induction heating. The single crystals were grown from these rods by an induction-melting floating-zone technique. An oriented single crystal was spark cut from the rod and annealed at 1000 °C for 24 h—yielding a residual resistivity ratio of 120 (at 1.5 K).

Magnetization curves of UAl_2 at 1.4, 4.2, 20, and 77 K are shown in Fig. 1. There is an increase in the initial susceptibility at low fields when the temperature is lowered from 77 to 1.4 K.

The susceptibility, derived from the magnetization at 1.4 and 4.2 K, decreases with increasing field reaching a field-independent value above 20 T. At 20 and 77 K the susceptibility is found to be independent of magnetic field over the entire field interval. Values for the differential susceptibility, presented in Table I, for various field intervals as a function of temperature indicate that the high-field values at 4.2 and 1.4 K

correspond to those low-field values of Brodsky and Trainor[3] at 25 K.

We wish to emphasize the perfect linearity of the magnetic isotherms up to 10 T. This implies that within the accuracy of the measurements magnetic impurities, which at liquid-helium temperatures must be saturated above 1 or 2 T, do not contribute to the observed magnetization.

The temperature and field dependence of the magnetic susceptibility suggest that an equivalent contribution can be suppressed either by applying a magnetic field of about 20 T or by increasing the temperature to T_{SF}. Ascribing the contribution to paramagnon effects, we must conclude that the characteristic field and temperature are related by $g\mu_B H_{SF} = k_B T_{SF}$, with $g = 2$.

The temperature-dependent susceptibility has also been measured from 4.2 to 270 K at 1.2 and 8.0 T. At low temperatures the data corresponds to an expression $\chi = \chi_0(1 - T^2/T_{SF}^2)$ with $T_{SF} \sim 28$ K. The temperature dependence of the differential susceptibility at 1.2 or 8.0 T could not be fitted satisfactorily with a $(T/T_{SF})^2 \ln(T/T^*)$ term, where T^* is an adjustable parameter, as has been previously suggested.[17]

The change of the specific heat in a magnetic field can be predicted from the susceptibility through the Maxwell relation,

$$(\partial S/\partial \mu_0 H)_{T,p} = (\partial M/\partial T)_{H,p}. \tag{1}$$

On taking the temperature derivative we write

$$(\partial c_p/\partial \mu_0 H) = T(\partial^2 M/\partial T^2)_{H,p}. \tag{2}$$

Using $M = \chi H$ we find that

$$(\partial c_p/\partial \mu_0 H) = TH(\partial^2 \chi/\partial T^2)_{H,p}. \tag{3}$$

For the field effect on the coefficient γ of the linear temperature term in the specific heat we obtain

$$(\partial \gamma/\partial \mu_0 H) = H(\partial^2 \chi/\partial T^2)_{H,p}. \tag{4}$$
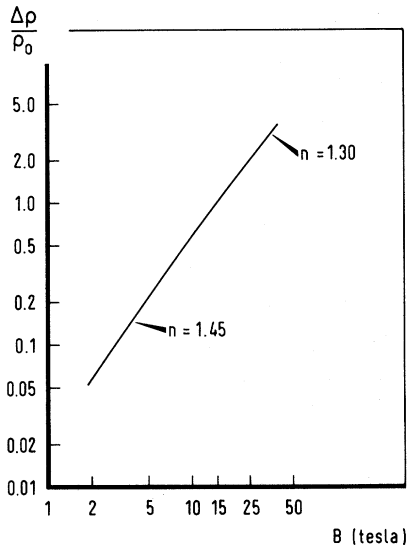
FIG. 2. Magnetoresistivity of UAl_2 at 4.2 K.

Since the T^2 fit to the susceptibility is known at low temperature, Eq. (4) can be written as

$$(\partial\gamma/\partial\mu_0 H) = -2H(\chi_0/T_{SF}^2)_{H,p}. \qquad (5)$$

According to the data of Fig. 1 χ_0 is found to decrease with increasing field. By consequence, the diminution of the specific-heat coefficient γ is less than H^2. At 4.3 T, $\Delta\gamma/\gamma$ is calculated to be -1.25% which is in very good agreement with the experiments by Trainor, Brodsky, and Culbert.[1]

The paramagnon contribution to the effective band mass is related to the S factor by the formula of Brinkman and Engelsberg,[4] $m^*/m = 1 + \frac{9}{2}\ln(S/3)$. For the S's previously derived[1] (~ 3.5), $(m^*/m)_{\text{paramagnon}} = 1.7$. As a result of the suppression of SF in high magnetic field (above H_{SF}) it is quite plausible that the de Haas–van Alphen effect can only be seen when all paramagnon contributions to the effective mass are frozen out. In preliminary experiments, de Haas–van Alphen oscillations have been observed in UAl_2 only above 20 T.[18]

The magnetoresistance also shows a bending over at fields of about 15 T, as shown in Fig. 2. The power law is $\Delta\rho/\rho_0 = H^n$; for fields between 2 and 15 T, $n = 1.45$, while above 15 T, $n = 1.3$. This has been seen for various crystallographic sample orientations with respect to the field and can probably not be attributed to $\vec{k}$-dependent band-structure effects.

Recently, Hertel, Appel, and Fay[19] calculated the magnetic field dependence of the electron-paramagnon interaction on the resistivity of UAl_2; here S was taken as 4, where a 14% suppression of the SF contribution is predicted at 10 T. If we assume as a simple model that the magnetoresistivity due to paramagnons and other effects (band structure, electron-electron, electron-phonon, etc.) are in series then the total resistivity may be written as $\Delta\rho = \Delta\rho_{SF} + \Delta\rho_{\text{other}}$. If we additionally assume that ρ_{SF} is maximum at zero field (contributing 30% at $H = 0$, from an extrapolation above H_{SF} in Fig. 2) then the paramagnon contribution is suppressed by 75% at 10 T.

To conclude, we find that the paramagnon effects at low temperature in UAl_2 seem to be large and can be suppressed in high magnetic fields above H_{SF}. Our experiments suggest that the field and temperature effects on the paramagnon contributions to the transport properties are closely related.

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Evidence for Interaction Effects in the Low-Temperature Resistance Rise in Ultrathin Metallic Wires

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New measurements are reported of the low-temperature resistance rise in ultrathin wires of Cu, Ni, and AuPd, which confirm the proportionality to $T^{-1/2}$ predicted by the interaction model. Moreover, these results and those in the literature show an absolute magnitude consistent within a factor of ~ 2 with the predictions of this model, using independently determined parameters of similar accuracy. It is inferred that interaction effects are at least as important as localization effects in these systems.

Recently there has been much theoretical[1-5] and experimental[6-9] work concerning the resistance rise at low temperatures observed in metallic samples of reduced dimensionality, attributed to either "localization" or "interaction" effects. In two-dimensional (2D) samples, the relative importance of the two mechanisms can be sorted out by application of a magnetic field. In the one-dimensional (1D) case of interest here, magnetic effects are smaller and less helpful; thus, one must put greater weight on the absolute magnitude of the effect and its dependence on material parameters and on temperature and sample size. On the other hand, the 1D regime has the advantage that the effect scales linearly with the characteristic length rather than only logarithmically as in the 2D case.

In this Letter, we report new experimental results on ultrathin wires of copper, nickel, and AuPd alloy, and also the results of a careful reanalysis of the data in the published literature. In all cases, the quantitative prediction of the interaction model of Altshuler *et al.*,[4] using independently determined parameter values, consistently accounts for much of the observed resistance rise, and in the case of Cu (our most reliable results), it accounts for essentially all of it. Accordingly, we infer that interaction effects are at least of comparable importance with, and may dominate over, localization effects in the metallic samples reported to date.

In either theory, so long as the resistance increase is small, it can be written as

$$\delta R/R \approx \Lambda/L_q. \tag{1}$$

The length Λ has a different meaning in the two models, while the length $L_q = (A/\rho)(4\hbar/e^2)$ is the length of conductor having the characteristic quantum resistance $4\hbar/e^2 \approx 16\,400\ \Omega$. In the free-electron model, L_q can be expressed in more microscopic terms as $L_q \approx k_F^2 A l$, so that it would be of the order of the mean free path l in the case of a "wire" made up of a single chain of metal atoms, but it is proportional larger for wires of realistic cross sections.

In the localization model,[1-3] the length Λ is essentially the inelastic diffusion length[10] $\Lambda_E = (D\tau_E)^{1/2}$, where D is the electronic diffusion coefficient and τ_E is the inelastic scattering time. τ_E is usually determined by the electron-phonon coupling strength, which can vary markedly between metals, and τ_E is normally expected to vary as T^{-p}, where $p \approx 3$. Alternatively, Abrahams *et al.*[5] have recently argued that if electron-electron effects dominate the inelastic relaxation time, p should be $\frac{1}{2}$ in 1D wires.