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Magnetoresistivity of the spin-fluctuation materials TiBe$_2$ and UAl$_2$

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High-field magnetoresistivity (MR) measurements are presented for high-purity samples of TiBe$_2$ and UAl$_2$. The MR of TiBe$_2$ gives strong evidence for scattering due to spin fluctuations. The peculiar features in the MR around 5 T are apparently connected to the maxima in the susceptibility and may provide a clue to a theoretical description of the contributions of spin fluctuations to the resistivity. A value for the spin-fluctuation temperature is obtained and it is in agreement with earlier estimates. The MR of UAl$_2$ is consistent with the behavior of a compensated metal. From the high-field behavior of the MR we expect to observe de Haas—van Alphen oscillations above approximately 10 T, but up to 40 T no signals are found. A small negative MR at higher temperatures may be due to spin-fluctuation scattering, but the evidence is not as clear as for TiBe$_2$.

I. INTRODUCTION

The $C\,15$ materials TiBe$_2$ and UAl$_2$ are well-known examples of exchange-enhanced paramagnetic compounds, in which spin fluctuations (SF's) are believed to play a dominant role. UAl$_2$ is the first homogeneous metallic compound that showed a $T^3\ln(T)$ term in the low-temperature specific heat, attributed to the effect of SF's.[1] The value for the electronic specific heat coefficient is $\gamma=143$ mJ/mol K^2. UAl$_2$ shows a high susceptibility at low fields, $\chi_0=5.7\times10^{-6}$ m^3/mol.[2] Experimental results for UAl$_2$, in contrast to those for TiBe$_2$, have only a small dependence on sample quality. The values reported for χ_0 agree all to within 10%.[1-3] The field dependence of both the magnetization and magnetoresistivity (MR) at 4.2 K are reported (Ref. 2) to show a bending over to a smaller variation with increasing field at approximately 15 T. As of yet, the theoretical and experimental information on the Fermi surface of UAl$_2$ is very limited.[4-6]

The properties of TiBe$_2$ have been extensively studied by a wide range of techniques; for a summary see Ref. 7. Most of the investigations concentrate on the maximum in the susceptibility $\chi(T)$ (Ref. 8) and $\chi(H)$ (Ref. 9) and on the low-temperature anomaly in the specific heat (Ref. 10). The maximum in $\chi(H)$ has been explained by a simple model involving fine structure in the density of states (DOS) close to the Fermi energy.[7,11,12] The anomaly in the low-temperature specific heat was, like that for UAl$_2$, fitted to a $T^3\ln(T)$ term and ascribed to SF's.[10] The Fermi surface of TiBe$_2$ and its field dependence have been studied using the de Haas—van Alphen effect.[7,13]

Here we present MR results on high-purity samples of TiBe$_2$ and UAl$_2$, concentrating on the longitudinal MR in fields up to 20 T and at temperatures down to 1.5 K.[14] The resistance of a metallic material at a temperature T in a magnetic field H is determined by several mechanisms. For the SF materials considered here we write

$$\rho(H,T)=\rho(0,T)+\Delta\rho_0(H,T)+\Delta\rho_{\text{SF}}(H,T)\ . \tag{1}$$

The resistivity at $H=0$, $\rho(0,T)$, is determined by scattering on impurities, phonons, and SF's. The MR is defined by $\Delta\rho(H,T)=\rho(H,T)-\rho(0,T)$. In Eq. (1) two contributions to the MR are distinguished; $\Delta\rho_0(H,T)$ represents the ordinary MR due to the orbital motion of the electrons in a magnetic field. The ordinary MR is always positive for all orientations of the magnetic field. It is larger for fields perpendicular to the current (transverse MR, $\Delta\rho_t$) than for fields parallel to the current (longitudinal MR, $\Delta\rho_l$).

The contribution $\Delta\rho_{\text{SF}}(H,T)$ to the MR represents the change in the scattering of the conduction electrons by the SF's when a magnetic field is applied. Theoretical results on the effects of SF's on MR for enhanced paramagnetic materials are given by Ueda[15] and Hertel *et al.*[16] For low temperatures Hertel *et al.* give an expression

$$\Delta\rho_{\text{SF}}(H,T)=g(H)T^2\ , \tag{2}$$

where $g(H)$ is a complicated function of the field. According to Eq. (2) the total MR, $\Delta\rho(H,T)$, at $T=0$ K is equal to the ordinary MR, $\Delta\rho_0(H,0)$. Both Ueda and Hertel *et al.* find that $\Delta\rho_{\text{SF}}$ is negative. This is important

since $\Delta\rho_0$ is always positive. In contrast, Beal-Monod[17] conjectures that for low fields and low temperatures $g(H)$ is proportional to $[\mathcal{X}^n(H)-\mathcal{X}^n(0)]$, where the power n is not specified. This would make $g(H)$ positive when $\mathcal{X}(H)$ increases with a field applied and negative in case of a decreasing susceptibility.

The separation of the MR into two distinct contributions as given in Eq. (1) is rather crude. When the magnetic field changes the electron-SF scattering rate, this will affect the ordinary MR, $\Delta\rho_0(H,T)$, as well. Moreover, in an experiment it is often difficult to separate the two contributions. However, Eq. (1) is useful in the limiting situation where one of the two terms dominates $\Delta\rho(H,T)$. For the moment we ignore effects of a field-dependent DOS at the Fermi level.

The major part of the experiments are performed using a four-probe ac technique; some have been repeated using a dc method. The fields are generated by a Bitter magnet of the Nijmegen High Magnetic Field Laboratory. The samples are mounted on a spiral gear rotator, with the long axis of the sample and the current direction either parallel or perpendicular to the axis of rotation.

The UAl_2 sample is a single crystal of dimensions $0.6\times0.6\times2.5$ mm^3, with the long axis along [110]. The resistivity ratio $\rho(294$ K$)/\rho(1.5$ K$)\approx160$. The actual sample quality may be better than indicated by this number, since at 1.5 K the resistance still decreases about linearly with decreasing T. The $TiBe_2$ sample is a polycrystalline piece of $0.7\times0.7\times5.0$ mm^3 with a resistivity ratio $\rho(294$ K$)/\rho(1.5$ K$)\approx80$.

II. THE MAGNETORESISTANCE OF $TiBe_2$

Figure 1 shows the transverse and longitudinal MR of $TiBe_2$ at 1.5 and 4.2 K. We find that $\Delta\rho_t$ is larger than $\Delta\rho_l$, as one generally expects. We wish to concentrate on the longitudinal MR. Figure 2 shows $\Delta\rho_l(H)$ for temperatures between 1.5 and 42.0 K. Part of the data is
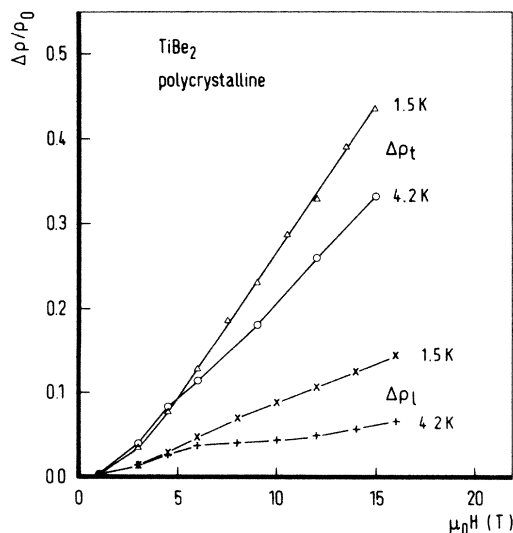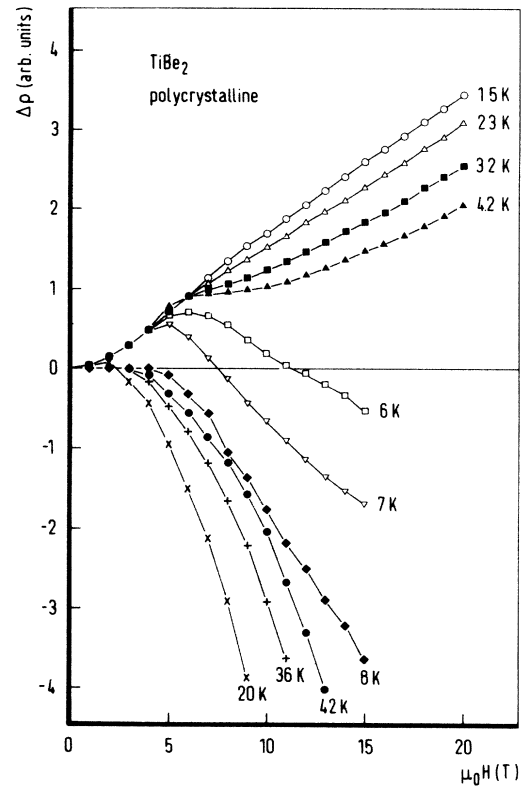

FIG. 2. Longitudinal magnetoresistivity of polycrystalline $TiBe_2$ as a function of applied field for temperatures between 1.5 and 42.0 K. The data are recorded while changing the field linearly in time from 0 to 20 T (or 15 T) in 5 min.

plotted as $\Delta\rho_l(T)$ at several values of the field in Fig. 3 and as a percentage of the zero-field resistivity at the various temperatures in Fig. 4.

For temperatures below 7 K and for initially increasing

FIG. 1. Transverse and longitudinal magnetoresistivity of polycrystalline $TiBe_2$ at 1.5 K and 4.2 K.
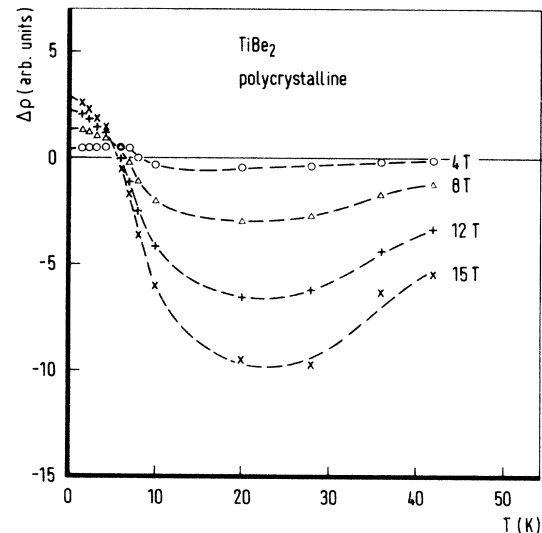

FIG. 3. The longitudinal magnetoresistivity results from Fig. 2, plotted as a function of temperature at several values of the field. The dashed curves serve as a guide to the eye.
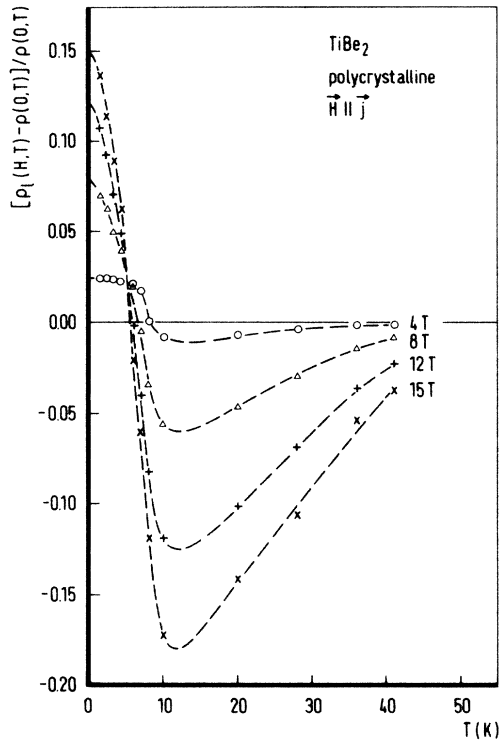
FIG. 4. The same data as in Fig. 3, plotted relative to the zero-field resistivity. The dashed curves serve as a guide to the eye.

H the curves in Fig. 2 all show the same behavior. A negative deviation from the lowest-temperature curve sets in above approximately 5 T; this deviation grows with increasing T. For temperatures above approximately 8 K the MR is dominated by a strong negative contribution at all fields. No saturation of the negative MR was observed for fields up to 15 T. Figure 3 shows that the positive $\Delta\rho_l$ at low temperatures is small compared to the negative value at high T; relative to $\rho(0,T)$ they are of comparable magnitude (Fig. 4). The temperature dependence of $\Delta\rho_l$ in Fig. 3 attains a minimum value at about 25 K and increases for higher temperatures. The minimum of Fig. 3 is moved to lower temperature when plotted as a ratio to the zero-field resistivity (Fig. 4). This is caused by the strong increase of $\rho(0,T)$ with temperature.[8]

III. THE MAGNETORESISTANCE OF UAl₂

In Fig. 5 the results for the low-temperature transverse and longitudinal MR, $\Delta\rho_t$ and $\Delta\rho_l$, for UAl₂ are plotted as a function of the square of the magnetic field. It is found that $\Delta\rho_l(H)$ saturates and that $\Delta\rho_t(H)$ approaches a H^2 behavior for fields above approximately 10 T. Again, the longitudinal MR is much smaller than the transverse effect. The bending over in the field dependence at about 15 T, observed by Franse et al.,[2] was reproduced for $\Delta\rho_t(H)$ with the field along [110]. For other orientations the bending point is less clear or not found at all. The subtle structure is better resolved for the lower temperatures.

In Fig. 6 the results for $\Delta\rho_l(H)$ are presented for several temperatures. Part of the data is plotted again in Fig. 7 as
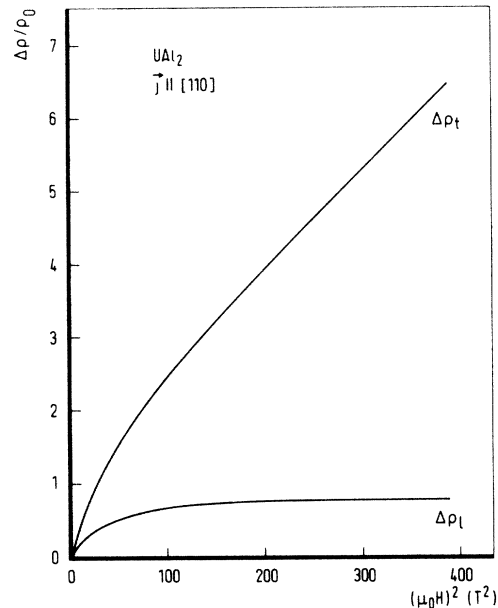

FIG. 5. Transverse and longitudinal magnetoresistivity for a single crystal of UAl₂ at 1.5 K. The field is parallel to [001] for $\Delta\rho_t$. The data are recorded in a 5-min sweep from 0 to 19.74 T.

$[\rho_l(H,T)-\rho(0,T)]/\rho(0,T)$ as a function of T at three different values of the field. We see that the MR decreases when T is raised and that $\Delta\rho$ becomes negative for temperatures above approximately 20 K. The negative $\Delta\rho(H,T)$ at higher temperatures is of the same magnitude as the positive MR at low T. The change relative to the zero-field value at the respective temperatures, however, is much smaller for high temperatures, due to the strong increase in $\rho(0,T)$ with increasing temperature.[18]

IV. DISCUSSION

We will first discuss the results on TiBe₂. The higher temperature negative $\Delta\rho_l$ in Figs. 2, 3, and 4 can only be
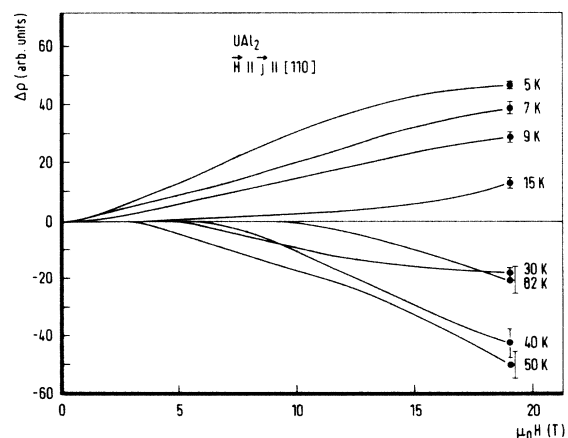

FIG. 6. Longitudinal magnetoresistivity of a UAl₂ single crystal as a function of applied field for temperatures between 1.5 and 82.0 K.
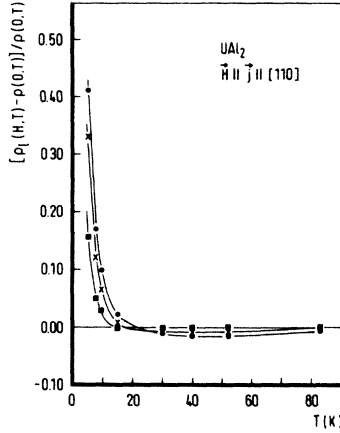
FIG. 7. The same data as in Fig. 6, plotted as the relative change in resistivity as a function of temperature for a magnetic field of 6.3 T (■), 12.7 T (×), and 19.0 T (●), respectively.

explained as originating from the term $\Delta\rho_{SF}$ in Eq. (1). In contrast, at the lowest temperatures $\Delta\rho_l(H,T)$ is positive and increases monotonically. This is the ordinary behavior, due to the deflection of the current by the field. The observation that at low temperatures $\Delta\rho_0$ dominates, and that at higher temperatures $\Delta\rho_{SF}$ becomes more important, may be explained by a cooperation of two effects. First and probably the most important is the temperature dependence of the SF scattering; towards lower temperatures $\Delta\rho_{SF}$ becomes smaller, giving way to the ordinary MR $\Delta\rho_0$. Second, $\Delta\rho(0,T)$ itself increases, as for all metals, when T decreases.

The most striking feature in Fig. 2 is that in the temperature range from 1.5 to 7 K the negative contribution to $\Delta\rho(H)$ seems to set in only at fields above 5 T. This behavior bears resemblance to that observed for the field dependence of the specific heat.[10] However, the resistivity in zero-field $\rho(0,T)$ changes by 50% between 1.5 and 7 K for the material used. Thus one may expect that $\Delta\rho_0(H,T)$ decreases with increasing T. A small temperature dependence of $\Delta\rho_0(H,T)$ would imply that $\Delta\rho_{SF}(H,T)$ is positive for fields smaller than 5 T. This would lend support to the conjecture in Ref. 17 that

$$\Delta\rho_{SF}(H,T) \propto [\chi^n(H)-\chi^n(0)] \ ,$$

since the susceptibility also rises to a maximum at 5 T. As a heuristic argument, one may observe that with a field increasing to 5 T the Stoner factor $S=\chi_0/\chi_{Pauli}$ is growing and the material is approaching a magnetic phase transition. Thus the SF's become stronger and the resistivity increases. Above 5 T the Stoner factor decreases again, the SF's become weaker, and the resistivity decreases. For lower temperatures the maximum in $\Delta\rho$ disappears, in agreement with Eq. (2). For higher temperatures the maximum smears out, as is observed for the maximum in $\chi(H)$.[12] In Refs. 15 and 16 only a negative behavior for $\Delta\rho_{SF}$ is found, which is probably due to simplifying assumptions concerning the band structure of the material.

In Fig. 3 a minimum in $\Delta\rho(H,T)$ is found at 25 ± 10 K. The position of this minimum is not very sensitive to the temperature dependence of $\Delta\rho_0$, since this term is small compared to $\Delta\rho_{SF}$, and $\Delta\rho_0$ is a smooth function of the temperature. The temperature at the minimum in $\Delta\rho$ may be interpreted as the characteristic temperature for the SF's, T_{SF}; the value observed here is in agreement with earlier estimates based on specific-heat measurements.[10] However, we must bear in mind that our value is determined in high fields. A more detailed comparison of the present results with theory requires a calculation of the SF effects using a realistic band structure.

The MR of UAl_2 is consistent with the behavior of a compensated metal (Fig. 5). For a compensated metal, $\Delta\rho_t(H)$ increases proportional to H^2 for any direction of H perpendicular to the current, provided that

$$\omega_c\tau \gg 1, \quad \omega_c = e\mu_0H/m^* \ . \tag{3}$$

Here ω_c is the cyclotron frequency and τ is the lifetime of the particles averaged over the orbit. The longitudinal MR is expected to saturate in all cases where $\omega_c\tau \gg 1$. The observed saturation indicates that this condition is satisfied for $\mu_0H > 10$ T. When $\omega_c\tau > 1$ one might expect to observe the de Haas—van Alphen effect above 10 T. However, several attempts to observe oscillations in this very pure material in pulsed fields up to 40 T and at temperatures down to 1.2 K were unsuccessful. This is in contrast to earlier reports[4] of the observation of the de Haas—van Alphen effect in UAl_2.

The ordinary positive longitudinal MR in UAl_2 is much larger than the one for the $TiBe_2$ sample. Above 20 K we see again a negative $\Delta\rho=\rho(H,T)-\rho(0,T)$, in absolute value of the same magnitude as the low-temperature positive $\Delta\rho$. Due to the strong increase of $\rho(0,T)$ going from 1.5 to 20 K, the relative change in resistivity $\Delta\rho/\rho(0,T)$ is only 1% or 2% here. Therefore, the negative MR cannot unambiguously be attributed to SF's. The decrease in ρ in a field of 19 T is at most 2%; this could easily be explained as a field dependence of the total DOS as well. A smaller SF effect than in $TiBe_2$ is expected, when one compares the Stoner factors, $S=15$ for UAl_2, $S=65$ for $TiBe_2$.[19]

The MR for UAl_2 clearly does not offer the strong evidence for SF scattering that is found in the case of $TiBe_2$. Recently de Groot *et al.*[5] performed a finely meshed band-structure calculation of UAl_2. They find the Fermi level at the top of a Van Hove singularity in the DOS. From this they suggest that the temperature dependence of the resistivity, the anomalous susceptibility, and low-temperature specific heat may be explained by band-structure effects alone. In their calculation there is another peak in the DOS at 8 meV distance from the Fermi level. The field required to make $S\mu_B\mu_0H$ equal to 8 meV is 9 T, using $S=15$. This is close to the field at which the anomalies mentioned above are observed.[2] It is not clear whether a band-structure calculation is reliable to such fine detail. Note that the many-body enhancement factor $1+\lambda\approx7$, which follows from the calculated DOS at the Fermi level and the experimental specific heat.[5] However,

in a material with strongly localized f electrons at the Fermi level a finely peaked DOS may be anticipated. In the case of $TiBe_2$, band-structure effects are invoked to explain the susceptibility and, indirectly, the MR behavior

in fields around 5 T. Also for UAl_2 they should be considered as contributing to the anomalies in the field and temperature dependence on the magnetization and resistivity.

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[19]Dividing the measured susceptibility $X_0 = 5.6 \times 10^{-3}$ m^3/mol for UAl_2 (Ref. 2) by the Pauli susceptibility with a DOS at the Fermi level, $N_F = 245$ states/eV atom (Ref. 5). The value $S = 2$ quoted in Ref. 5 is in error.