HIGH-FIELD MAGNETORESISTANCE OF POLYCRYSTALLINE ZrZn₂

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(Received 29 May 1980 by J. Tauc)

We report magnetoresistance measurements of polycrystalline ZrZn₂ as a function of temperature (4.2—48 K) and magnetic field up to 19 T. The results indicate the presence of both positive and negative contributions to the magnetoresistance. The latter is due to spin fluctuations. Below \( T_e \) the resistivity varies with temperature like \( T^2 \) over the entire field range (0—19 T). The coefficient of the \( T^2 \)-term decreases with increasing field and fits a \( H^{1/3} \) dependence above ~10 T, in accordance with theoretical predictions. Complex behaviour of the magnetoresistance is found in the paramagnetic regime above ~5 T.

ZrZn₂ [1] is perhaps the prime example of itinerant electron ferromagnetism in the limit of very low exchange splitting energies [2]. There is now renewed interest in this material following the discovery of itinerant antiferromagnetic TiBe₂ [3] and the hindered “p-state pairing” that was recently proposed to explain the ferromagnetism of ZrZn₂ [4]. These recent studies stress the importance of the electron-phonon interaction in itinerant magnets. Evidence for this in ZrZn₂ may be seen in the anomalous temperature dependence of the elastic moduli [5, 6], among others.

Ogawa has measured the electrical resistivity [7] and magnetoresistance [8] of polycrystalline ZrZn₂ up to 5 T. The temperature dependence of the electrical resistivity was found [7, 8] to follow closely the predictions of self-consistent renormalized (SCR) theory of spin fluctuations [9, 10]. The magnetoresistance was in qualitative agreement with the SCR theory for low fields [8]. In the present communication we report the magnetoresistance of polycrystalline ZrZn₂ at fields up to 19 T. Our results below \( T_e \) are in agreement with the predictions of the SCR theory for strong magnetic fields [10]. Complex behavior of the magnetoresistance is observed in paramagnetic ZrZn₂ at high fields above ~10 T.

The preparation and the X-ray and electron microprobe analyses of the ZrZn₂ samples were described elsewhere [5]. By repeated sintering we were able to obtain samples with 91% of the X-ray density, as compared with 84% in Ogawa’s work [7]. The higher density of our samples is expected to reduce a possible contribution of voids to the magnetoresistance [11]. The samples are slightly off stoichiometry with an actual composition of ZrZn₁.₉₅. This, however, was shown to have only a slight effect on the ferromagnetism of the sample [12].

A standard four-probe technique was used for the magnetoresistance measurements with pressure contacts for the voltage leads. The measurements were carried out at the Francis Bitter National Magnet Laboratory on a 19 T solenoid by continuously sweeping the field at constant temperature, between 4.2 and 48 K and at 78 K. The temperature was measured using a calibrated carbon-glass resistor and stabilized to within 0.1 K during a field sweep, using a temperature controller with a capacitance cryogenic sensor. Magnetization measurements were also performed as a function of temperature (4.2—300 K) and magnetic field up to 0.95 T.

From an Arrott plot we find \( T_e = 21.0 \pm 0.5 \) K. The 4.2 K isotherm is given by \( M^2 = 7.5 + 0.8 H/M \), where \( M \) and \( H \) are in emu g⁻¹ and kOe, respectively. The spontaneous magnetization at 0 K is therefore \( M_0 = 0.110 \pm 0.003 \mu_B/\text{ZrZn}_2 \), slightly lower than in earlier studies [7, 10]. The slope of our Arrott isotherms, however, is only about half that of Ogawa’s samples [7, 10]. Such inconsistencies are attributed to differences in stoichiometry and the details of preparation and have been noted before in ZrZn₂ [6, 12, 13].

The zero-field resistivity of our sample is generally in agreement with Ogawa’s observations [7] and the predictions of the SCR theory [9]. Our residual resistivity ratio (RRR) of ~3.5 is lower than Ogawa’s [7] RRR ~5. The room temperature resistivity (~330 \( \mu\Omega \text{cm} \)) is close to the average between values reported for stoichiometric ZrZn₂ [7] and for ZrZn₁.₉ [14]. The temperature dependence of the zero-field resistivity below \( T_e \) follows the expression [7, 9],

\[
\rho(H = 0, T) = R_0(0) + R(0)T^2
\]

with \( R_0(0) = 95 \pm 10 \mu\Omega \text{cm} \) and \( R(0) = (3.5 \pm 0.2) \times 10^{-8} \Omega \text{cm K}^{-2} \). Ogawa [7] finds \( R_0(0) = 121 \mu\Omega \text{cm} \) and \( R(0) = 4.7 \times 10^{-8} \Omega \text{cm K}^{-2} \). Due to uncertainty

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in the dimensions of the sample (\sim 10\%) we present our magnetoresistance data in terms of \( \Delta \rho/\rho_0 = [\rho(H) - \rho(H = 0)]/\rho(H = 0) \), as a function of field at several temperatures. Bars indicate amount of noise around measured values. Dashed line shows Ogawa's result at 4.2 K [8].

The measured field dependence of the magnetoresistance of ZrZn\(_2\), \( \Delta \rho/\rho_0 \), is shown in Fig. 1 at several temperatures. Figure 2 shows the temperature dependence of \( \Delta \rho/\rho_0 \) at several fields. The bars in Figs. 1 and 2 indicate the level of noise around the measured value. The results shown were obtained with the magnetic field perpendicular to the current direction. Identical results were obtained, within experimental accuracy, with the field parallel to the current direction.

There is qualitative agreement between the results of Figs. 1 and 2 up to 5 T and Ogawa's magnetoresistance work [8]. At low temperature (e.g. 4.5 K), \( \Delta \rho/\rho_0 \) is positive and increases with \( H \) over the entire 19 T range (Fig. 1). As the temperature exceeds \sim 10 K, \( \Delta \rho/\rho_0 \) becomes increasingly negative at low and intermediate fields. Well below \( T_c \) (10 \sim \sim 15 K) \( \Delta \rho/\rho_0 \) passes through a shallow minimum at \sim 10 T and then becomes positive at higher fields (Fig. 1). Close to \( T_c = 21 \) K, up to 26 K, \( \Delta \rho/\rho_0 \) is almost field independent above \sim 10 T and negative up to 19 T. At higher temperatures a broad minimum reappears in \( \Delta \rho/\rho_0 \) vs \( H \) (Fig. 1). It becomes more pronounced with increasing temperature up to \sim 38 K and then decreases appreciably. This behavior cannot be expected on the basis of previously reported data on ZrZn\(_2\) [8] or Sc \(_3\)In [15] at lower fields. The 47.8 K \( \Delta \rho/\rho_0 \) vs \( H \) curve is very similar to those obtained around \( T_c \), in particular above \sim 10 T (Fig. 1). At 78 K (not shown in Fig. 1) \( \Delta \rho/\rho_0 = 0 \) within experimental error up to 19 T. The variation of \( \Delta \rho/\rho_0 \) with temperature (Fig. 2) shows a minimum at all fields. The minimum shifts from about \( T_c \) to higher temperature and becomes narrower and more pronounced with increasing field up to \sim 13 T, at which it is found at \sim 38 K. The minimum shifts slightly to lower temperature, broadens and decreases again at higher fields. It should be stressed that for \( H \sim 5 \) T the maximum negative magnetoresistance occurs in paramagnetic ZrZn\(_2\), between 30 and 40 K (Fig. 2).

The change of sign of \( \Delta \rho/\rho_0 \) at \sim 10 to \sim 18 K, depending on the field strength (Fig. 2), indicates, as already noted by Ogawa [8], the presence of an ordinary Lorentz effect along with a negative contribution due to spin fluctuations [7, 8]. An additional positive contribution due to voids [11] in the sintered material may be indicated, recalling the higher density of our samples, by the much larger positive effect observed by Ogawa at low temperature [8] (dashed line in Fig. 1). The orbital and void contributions are expected to diminish with increasing temperature. A third source of positive magnetoresistance would be associated with the possible presence of a charge density wave (CDW) in ZrZn\(_2\) [6]. The onset of a CDW at \sim 45 K was postulated [6] on the basis of sound velocity [5] and other measurements. Recent Hall measurements also show a change of sign of
From the slope of the low-temperature Arrott isotherms of our sample and \( AE_F = 8/3 \) eV, as obtained by \(^{91}\)Zr NMR spin-lattice relaxation rate measurements \([19]\), we estimate \( E = 1.2 \), much larger than Ueda's \( E = 5/8 \) for \( \text{ZrZn}_2 \) \([10]\). This difference reflects the different Arrott slopes of the particular \( \text{ZrZn}_2 \) sample under study.

Acknowledgements — Helpful discussions with Professor M. Weger and correspondence with Professor S. Ogawa are gratefully acknowledged. We are grateful to the Francis Bitter National Magnet Laboratory for the use of the high-field magnet and cryogenic equipment. This work was supported in part by a grant from the Materials Research Laboratory, DMR76-01111.

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