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CRITICAL BEHAVIOUR OF THERMAL RESISTIVITY OF Ni

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Accurate data are presented on the behaviour of the thermal conductivity K as a function of temperature for a pure Ni sample near its Curie point. Previous results on the electrical resistivity (ϱ, $d\varrho/dT$) are used to explain the temperature-dependence of $K(T)$. The results are analysed in terms of electron-phonon and $s-d$ exchange interactions. The critical behaviour of the thermal resistivity $W(=K^{-1})$ has also been investigated.

Study of the transport properties of magnetic phase transitions provides a sensitive and often rather simple means of investigation details of the microscopic interactions. The electrical resistivity ϱ in particular has received a good deal of attention [1–7]. The scanty experimental information available on the thermal conductivity K of magnetic materials and on the complex behaviour which occurs in the transition region earlier precluded any discussion on such matters as the values of the critical exponents. Accordingly, only general features were considered [8–12]. The critical exponents are of interest because many different kinds of physical systems behave in a similar way near the critical point T_c. This work reports for the first time the critical exponents of the thermal resistivity of pure Ni both below and above T_c. The universality concept [4, 5, 13] is also tested.

In ferromagnetic metals and alloys, the most characteristic interaction is the $s-d$ interaction, i.e. the spin exchange interaction between the conduction (s) and unfilled shell (d) electrons. According to Kasuya [1], this exchange interaction depends on the relative orientation of the spins of both electrons. Therefore, at $T=0$ all the spins of d-electrons being in order, there is no electrical resistance, while at a finite temperature this order is disturbed and thus a resistance appears and increases with temperature. Above T_c, the directions of the d-electron spins become perfectly random, and the electrical resistance remains constant. The resistivity caused by such a process is called the spin-fluctuation [3] or spin-disorder contribution [10].

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In the present work, we present accurate data on the thermal conductivity of pure Ni near its Curie point. The results show a well-defined anomaly in $K(T)$, precisely located at T_c and in good agreement with the electrical resistivity data (ϱ, $d\varrho/dT$) obtained for the same sample [7]. The results are discussed in terms of electronic, phonon and $s-d$ exchange interactions. The critical exponents of the thermal resistivity are calculated and compared with the unique rough estimate reported for Ni by Graig et al. [14], who analysed the thermal diffusivity data measured by Kirichenko [15].

Experimental

A nickel rod 2.34 mm in diameter and 5.5 cm in length was supplied by the National Physics Laboratory, Budapest, Hungary. It was stated to be of high spectrographic purity. The electrical resistivity ϱ of the same sample was previously measured by using a standard four-probe technique [7].

The thermal conductivity K was measured by using an apparatus described in detail earlier [11, 12]. It is mainly based on the electrical and thermal potential distributions along a thin rod that is heated by passing a direct electric current through it. The quantity $K\varrho$ was calculated by using the measured voltage *vs.* temperature (V *vs.* θ) relation. The estimated uncertainty in K is about 3%.

Results and discussion

1 Electrical resistivity

It was shown previously that the electrical resistivity ϱ of ferromagnetic metals and alloys displays anomalous behaviour during transition from the ferromagnetic to the paramagnetic state. This anomaly results from the exchange interaction between the s and d-electrons [1, 3]. In a previous paper [7] on the electrical resistivity of Ni, particular emphasis was put on analysis of the critical behaviour related to the nature and the type of the singularities in the temperature coefficient of resistivity (TCER) in the immediate vicinity of T_c.

In this work, we are interested only in decomposing the electrical resistivity into its different contributions according to the corresponding scattering mechanisms, and in using the results in the analysis of the thermal resistivity. If Mattheissen's rule is assumed provide an adequate approximation, the total electrical resistivity $\varrho(T)$ is the sum of a spin-fluctuation component $\varrho_s(T)$, a component due to electron-phonon scattering $\varrho_{ph}(T)$, and a contribution due to the scattering of electrons by static imperfections and lattice defects ϱ_D, i.e.:

$$\varrho(T) = \varrho_D + \varrho_{Ph}(T) + \varrho_s(T) \tag{1}$$

where the component ϱ_D is temperature-independent and was found to be negligibly small, especially in the high-temperature range [16, 17]. According to Wilson's theory of metals [18] and the analysis given by Parrott et al. [19], the electron-phonon scattering gives rise to an electrical resistivity term $\varrho_{ph}(T)$:

$$\varrho_{ph}(T) = \varrho_\theta T/\theta_D \tag{2}$$

where ϱ_θ is the electrical resistivity at the Debye temperature θ_D.

Kasuya [1] used a spin-wave method to calculate the magnetic resistivity term $\varrho_s(T)$ in terms of a deviation of the spin s from the average orientation, i.e. $S-\sigma$. At absolute zero temperature, $\sigma = S$, and thus $\varrho_s = 0$. When the temperature is raised, σ decreases and ϱ_s increases. For $T > T_c$, $\sigma = 0$ (complete disorder) and ϱ_s remains constant. Figure 1 gives the electrical resistivity as a function of temperature for the

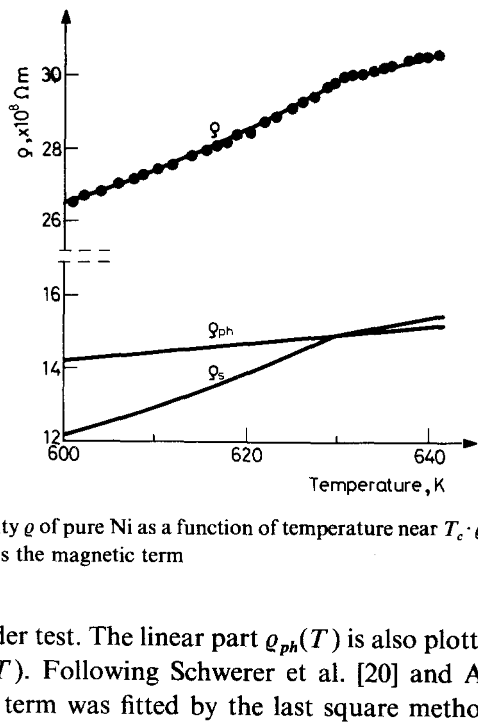

Fig. 1 Electrical tesistivity ϱ of pure Ni as a function of temperature near $T_c \cdot \varrho_{ph}$ is the electron-phonon contribution; ϱ_s is the magnetic term

pure Ni sample under test. The linear part $\varrho_{ph}(T)$ is also plotted, together with the magnetic term $\varrho_s(T)$. Following Schwerer et al. [20] and Awad [10], this spin-disorder resistivity term was fitted by the last square method to an exponential function of the form:

$$\varrho_s(T) = a \exp(bT) \tag{3}$$

where a and b are constants. Our data give the following results: $a = 2.17 \times 10^{-9}$ $\Omega \cdot m$ and $b = 6.7 \times 10^{-3}$ K^{-1} with a regression coefficient of 0.997.

2 Thermal conductivity

The measured thermal conductivity K of the pure Ni specimen is presented in Fig. 2, as a function of absolute temperature T. The absolute value of K for our sample at T_c (50.0 Wm^{-1} K^{-1}) compares favourably well with the TPRC reported data [21]

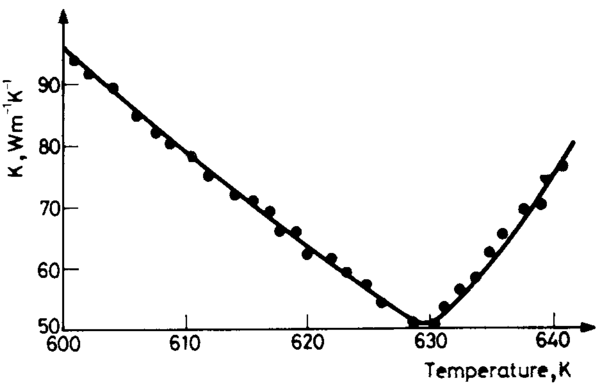

Fig. 2 Thermal conductivity K as a function of temperature

for a recommended 99.5% purity Ni and with that of Powell et al. [8] for a highly pure Ni sample. It can be seen from Fig. 2 that K falls sharply and steadily with increase in temperature up to T_c, where a nearly sharp minimum is observed. Above T_c, K has a positive temperature coefficient. This behaviour has also been observed for pure Ni by Powell et al. [8], for Ni–Cu alloys by Jackson et al. [9], and for Ni–Mn dilute alloys by Ammar et al. [11].

In an attempt to understand the reason for the observed minimum in K, let us first assume that the lattice contribution to K, for $T > \theta_D$, is either constant or very weakly temperature-dependent, and then consider what would be expected from the electronic contribution K_e. If we use a simple kinetic theory model, K_e can be expressed as:

$$K_e = C_e V_f l_e / 3 \tag{4}$$

where C_e is the electronic specific heat, which is proportional to T, V_F is the Fermi velocity, and l_e is the electron mean free path. Then, we can write:

$$\frac{1}{K_e}\frac{dK_e}{dT} = \frac{1}{T} + \frac{1}{l_e}\frac{dl_e}{dT} \tag{5}$$

Below T_c, l_e decreases rapidly with increasing T, due to the decreasing order in the system, i.e. dl_e/dT is negative [22]. However, there is no quantitative information on the way in which l_e varies with T. Thus, we may suggest that the absolute value of the second term in Eq. (5) is larger than $1/T$, with the result of a negative slope for K_e. This peculiar behaviour of dK_e/dT can also be understood by using the Wiedemann–Franz law:

$$K_e = L_0/\varrho T \tag{6}$$

where L_0 is the Sommerfield–Lorenz number, a constant equals to 2.45×10^{-8} V^2 K^{-2}. On differentiating Eq. (6), we get:

$$\frac{1}{K_e}\frac{dK_e}{dT} = \frac{1}{T} - \frac{1}{\varrho}\frac{d\varrho}{dT} \tag{7}$$

According to our previous results: $\frac{1}{\varrho}\frac{d\varrho}{dT} > \frac{1}{T}$, which is quantitatively in support of the negative slope of $K_e(T)$ for $T < T_c$. Referring again to Eq. (5), spin-disorder scattering dominates in the paramagnetic phase ($T > T_c$) and this imposes a small and practically constant mean free path l_e. A positive slope for $T > T_c$ follows, in qualitative agreement with the experimental results in Fig. 2. In addition, since l_e is practically constant for $T > T_c$ (spin-disorder dominance) and $C_e \propto T$, from Eq. (4) one would expect a linear increase of K with T, i.e. K = constant $\times T$, which seems consistent with the results.

3 *Critical behaviour of thermal resistivity*

If Matthiessen's rule is assumed to be applicable to the case of thermal resistivity W, by analogy to Eq. (1) we can write:

$$W = W_D + W_{ph} + W_s \tag{8}$$

$$= \frac{\varrho_D}{L_0 T} + \frac{\varrho_{ph}}{L_0 T} + W_s \tag{9}$$

where the first term W_D is due to defects and is neglected in the temperature range under investigation [17]. The phonons contribute the second term W_{ph}, and since $\varrho_{Fh}T$ was assumed to be linear with T (Eq. (2)), this term is a constant:

$$W_{ph} = \frac{\theta_D}{L_0 \varrho_\theta} = 9.67 \times 10^{-3} \text{ W}^{-1} \text{ mK} \tag{10}$$

By substracting this calculated phonon contribution from the total resistivity, we obtained the temperature-dependence of the spin-scattering term $W_s(T)$. The

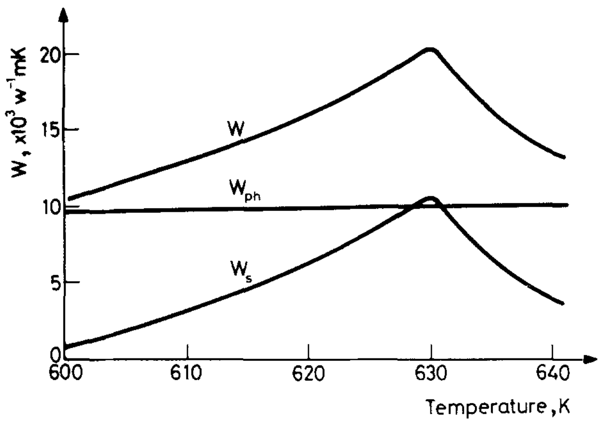

Fig. 3 Thermal resistivity W together with the phonon contribution term W_{ph} and the magnetic contribution term W_s as a function of temperature

results are shown in Fig. 3. It can be seen that the anomaly in $W(T)$ comes mainly from $W_s(T)$.

The natural step now is to calculate the critical exponents of the magnetic thermal resistivity $W_s(T)$. It was previously shown [7, 14, 23], that the regular form of a singular quantity is to be used:

$$W = A(\varepsilon^{-\lambda} - 1)/\lambda + B \tag{11}$$

where A and B are constants, λ is the critical exponent, and ε is the reduced temperature $[(T-T_c)/T_c]$. Thus on differentiating the non-linear part in Eq. (11):

$$W_s(T) = W(T) - B$$

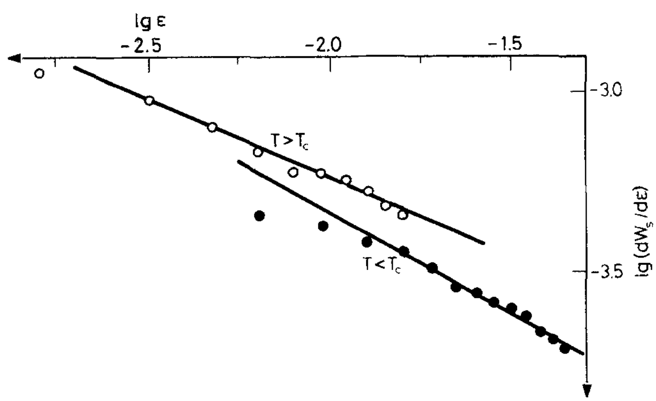

Fig. 4 $\log dW_s/d\varepsilon$ *vs.* $\log \varepsilon$, where ε is the reduced temperature $(T-T_c)/T_c$

and plotting log dW_s/dε against log ε, we obtain a straight line with slope $-(\lambda+1)$. Figure 4 shows such plots for $T > T_c$ and $T < T_c$, and gives the following exponents:

$$\lambda^+ = 0.44 \quad \text{for } T > T_c;$$

$$\lambda^- = -0.47 \quad \text{for } T < T_c.$$

These may be compared with the values reprted by Craig et al. [14]:

$$\lambda^+ = 1.50 \quad \text{for } T > T_c;$$

$$\lambda^- = 0.67 \quad \text{for } T < T_c.$$

Craig et al. found these exponents indirectly by first analysing reported thermal diffusivity data (Kiricheko et al. [15]) and then using the available critical exponents of the specific heat of Ni. However, their values have never been tested either theoretically or experimentally.

Ausloos [4, 5] and Helman et al. [13] have predicted theoretically that all transport properties possess universal behaviour, irrespective of the scattering process. It has also been found experimentally by Sousa et al. [6] that this is true for the antiferromagnet $Cr_{1-x}Al_x$ ($x = 0.06$). The universality hypothesis means:

$$C_M \approx \frac{\mathrm{d}\varrho}{\mathrm{d}T} \approx \frac{\mathrm{d}s}{\mathrm{d}T} \tag{12}$$

where C_M is the magnetic specific heat and s is the thermoelectric power. We suggest that, besides relations (12), the magnetic thermal resistivity, as a singular quantity, is also proportional to dϱ/dT and to C_M, i.e.

$$W_s \approx \frac{\mathrm{d}P}{\mathrm{d}T}, \qquad W_s \approx C_M \tag{13}$$

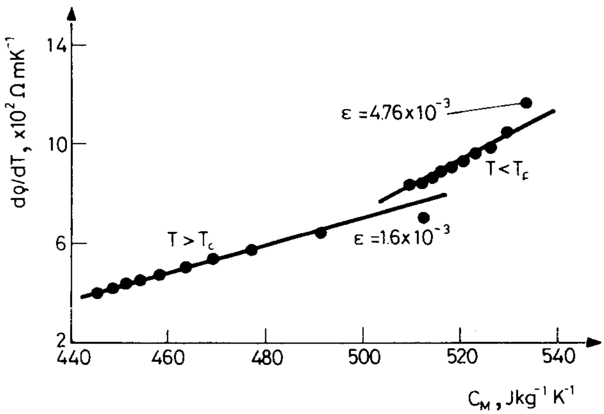

Fig. 5 dϱ/dT vs. magnetic specific heat C_M

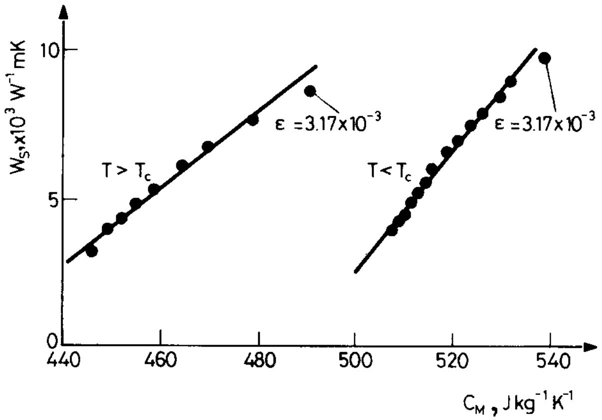

Fig. 6 Magnetic thermal resistivity W_s *vs.* magnetic specific heat C_M

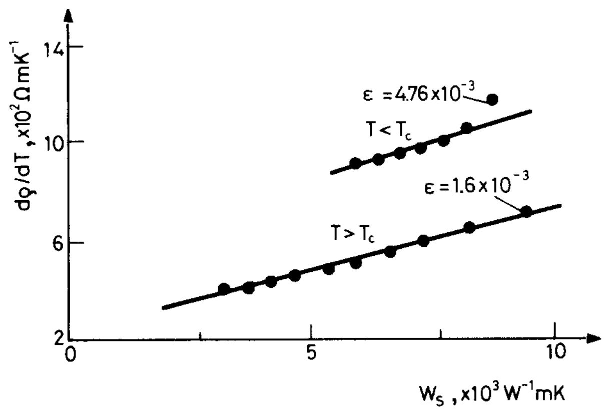

Fig. 7 dϱ/dT *vs.* magnetic thermal resistivity W_s

Figures 5–7 show this proportionality, where we have used our previous measurements on the same sample for $d\varrho/dT$ [7] and for C_M [23]. It is clear that the universality prediction is valid up to $\varepsilon \simeq 3 \times 10^{-3}$, where deviation from the straight lines appears. This means that short-range interaction become dominant when T approaches T_c from either side of the transition. The complexity of the Fermi surface and band structure of Ni near the transition and electron–electron scattering may contribute to both electrical and thermal resistivities [24]. The effects of inelastic scattering and umklapp processes are also important is systems with a complex band structure [19].

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Zusammenfassung — Die Wärmeleitfähigkeit K einer reinen Nickelprobe wurde in der Nähe des Curiepunktes in Abhängigkeit von der Temperatur beschrieben. Vorangestellte Untersuchungen des elektrischen Widerstandes $\left(\varrho, \frac{d\varrho}{dT}\right)$ wurden benutzt, um die Temperaturabhängigkeit von K zu erklären. Die Ergebnisse wurden unter Inbetrachtnahme von Elektron-Photon und $s-d$-Austausch Wechselwirkungen interpretiert. Das kritische Verhalten des Wärmewiderstandes W $(=K^{-1})$ wurde ebenfalls untersucht.

Резюме — Представлены точные данные о поведении термической проводимости K в зависимости от температуры для чистого никеля в окрестностях его точки Кюри. Для объяснения температурной зависимости $K(T)$ были использованы ранее полученные результаты по электрическому удельному сопротивлению (ϱ, $d\varrho/dT$). Результаты обсуждены на основе электрон-фононовых и $s-d$ обменных взаимодействий. Исследовано также критическое поведение термического удельного сопротивления $W(=K^{-1})$.