THERMOELECTRIC POWER (S, *dS/dT*) IN ANTIFERROMAGNETIC Cr-Al ALLOYS (*) (**)

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ABSTRACT — Accurate measurements of the thermoelectric power (S) of antiferromagnetic Cr-Al dilute alloys are presented over a wide range of temperature (80-350 K).

Particular attention is given to the critical behaviour of the temperature derivative dS/dT near the Néel temperature (T_N). The anomalous concentration dependence of T_N in Cr-Al is also considered.

1 — INTRODUCTION

The antiferromagnetic b.c.c. Cr-Al dilute alloys constitute a very complex system, as revealed by the pronounced changes which occur in the electronic density of states [1], the Debye temperature [1] and the peculiarities in the magetic behaviour for Al compositions in the range $0<c<6$ at. %. Further difficulties arise in this range of compositions due to the exceptionally small magnetic anomalies found in most physical properties, even in those sensitive properties as the magnetic susceptibility [2] or the electrical resistivity [3, 4]. This occurs in spite of the steady increase in the magnetic moment

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(*) Results presented at the Conference of the Portuguese Physics Society (Lisbon, February, 1978).

(**) Work fiancially supported by INIC (Portugal) and NATO Research Grant no. 1481.

per atom caused by the addition of Al, as shown by neutron diffraction studies [5, 6]. These studies revealed an abrupt transition from an incommensurate antiferromagnetic structure to a commensurate one when Al content exceeds about 1 at. %. This has been corroborated by recent latent heat measurements [7]. The information available on the Néel temperature also suggests a very complex and anomalous behaviour of T_N in the concentration range $0<c<6$ at. % Al [8].

The anomalous behaviour in T_N and the excepcionally small magnetic anomalies which occur in Cr-Al, were good reasons to start a detailed and systematic study of transport properties in this system. To our knowledge, the available information on transport properties in Cr-Al is rather scarce, and essentially restricted to the *general* features of the electrical resistivity (ρ). The first experimental account has been given by Chakrabarti [3] (including the Hall effect), but a more precise study of ρ for low Al content has been done subsequently by Arajs et al [4] ($0.3<c<6.2$ at % Al). For one particular case (1.2 at % Al) general information was also given for the thermoelectric power over a restricted range ot temperature [4].

In this preliminary report we present very accurate data on the thermoelectric power (S) of Cr-Al dilute alloys over a wide range of temperatures (~80-320 K) and, for the first time, detailed and precise information is published on the behaviour of the temperature derivative of the thermoelectric power (dS/dT) for this system.

The importance of the thermal treatments on the behaviour of the thermoelectric power is also investigated.

A detailed analysis of the critical behaviour of dS/dT, its intimate connection with the critical behaviour of $d\rho/dT$, and the peculiar concentration dependence of the Néel temperature will be object of a forthcoming publication, when the present investigations (ρ, $d\rho/dT$, S, dS/dT) have been extended to the *whole* set of available Cr-Al samples in the range $0<c<6$ at % Al.

2 — EXPERIMENTAL TECHNIQUE

The thermoelectric power was measured by a static method [9]. The temperature difference across the sample ($\Delta T \simeq 0.5$ K), produced by Joule heating (H_1), was measured with a differential copper/constantan thermocouple, in good contact with, but electrically insulated from the sample. Another similar thermocouple (T) allowed the determination of the sample mean temperature. The thermoelectric voltage (ΔV) was measured using Cu leads spotwelded to the sample. Both thermocouple and thermoelectric voltages were measured directly on sensitive digital voltmeters, with 10^{-2} and 10^{-3} μV resolution respectively.

The sample, attached at one end to a copper «basis» (thermocouple T), was surrounded by a radiation shield with a heater (H_3), and the assembly suspended inside a metallic vaccuum chamber ($\sim 10^{-6}$ torr) by a thin tube of stainless steel.

The electrical leads were carefully twisted, thermal anchored, and with reasonable lengths between sample and basis to minimize heat losses. The heating power delivered at the free end of the sample (H_1) could be transferred to an equal heater (H_2) on the basis.

The measuring technique was as follows: (*i*) with the power switched to H_2 in the basis (current stability $\pm 1/10^4$) a set of readings was taken (T_0, ΔV_0, ΔT_0) to define a zero reference for all circuits. (*ii*) Then the power was transferred abruptally to H_1 and a difference of temperature gradually established across the sample. A period of 15 min was adopted before taking the new readings (T_1, ΔV_1, ΔT_1). In general $T_1 \simeq T_0$, since the transfer of the same power from H_2 to H_1 practically does not affect the temperature at the basis when heat losses are small. (*iii*) Power is transferred again to H_2, and the whole process repeated as before. Obviously, the mean temperature of the sample can be adjusted with the heater H_3 in the radiation shield. In general we adopted very low rates of change of temperature, of the order of 15 mK/minute.

Since the differences of temperature between consecutive points were reasonably small ($\delta T/T_c \sim 10^{-3}$), we used the «sliding rule» process to obtain our final (smooth) dS/dT values. Therefore, groups of five consecutive points were slid along the whole set of initial dS/dT values, but without mixing dS/dT values on both sides of T_c.

The typical scatter in our S curves is generally fairly small ($\sim 1/10^3$) and of the order of a few per cent for dS/dT curves. Reproducibility of S values in different runs is of the order of 10^{-3}. This indicates that under the careful experimental conditions adopted, and by taking readings always at the same time intervals, the parasitic thermal emfs do not interfere in any significant way in our results.

3 — SAMPLES

The samples used in this work contain 0.06, 0.8, 1.93 and 4.8 at % Al. Except for 0.06 at % Al, all the other samples were prepared at the Clarendon Laboratory, Oxford, by melting the constituent elements (Johnsson Mathey spectrographically pure Cr and Al) in alumina crucibles using radiofrequency heating under argon atmosphere. Several melting operations were performed in order to improve the homogeneity of the samples. The final compositions were determined by the atomic absorption method. Metallographic analysis and hardness tests, made at the Faculty of Engineering in Porto, revealed a fairly homogeneous and single-phase system for these low Al compositions. The samples were cut from the melted ingots using a spark machine and a slab geometry ($\mathrm{cm} \times \mathrm{mm} \times 10^{-1}\,\mathrm{mm}$) obtained at the end.

The 0.06 at % Al sample was prepared at Imperial College of London, by the arc discharge method under argon atmosphere. Several melting operations were made in order to improve the homogeneity of the alloy. A cylindric-shaped sample was finally obtained by the suction method, through a cold copper mould. For this sample, the exact Al content is difficult to measure by the usual methods of analysis, including electron scanning microscopy. Therefore, its actual composition had to be inferred from residual resistivity measurements, assuming $\Delta\rho = 12\,\mu\Omega$. cm per 1 at % Al, which is an average figure obtained from a linear plot of ρ_0 vs Al composition, for a series of dilute alloys (< 4 at % Al) prepared under the *same* conditions. Annealing at about 1050 °C for 5 days was done for the 0.06, 1.93 and 4.8 at % Al samples, under argon atmosphere. In order to study the importance of heat treatments, we did not anneal the 0.8 at % sample, neither a second sample with the same nominal composition as 0.06 at % Al.

4 — EXPERIMENTAL RESULTS

Let us consider first the case of the annealed samples. All these measurements have been performed at the University of Porto.

Fig. 1 shows the temperature dependence of the thermoelectric power for the sample with 0.06 at % Al over a wide range of temperatures (120–340 K), and a direct comparison is made with the corresponding behaviour of the electrical resistivity measured in the *same* sample.

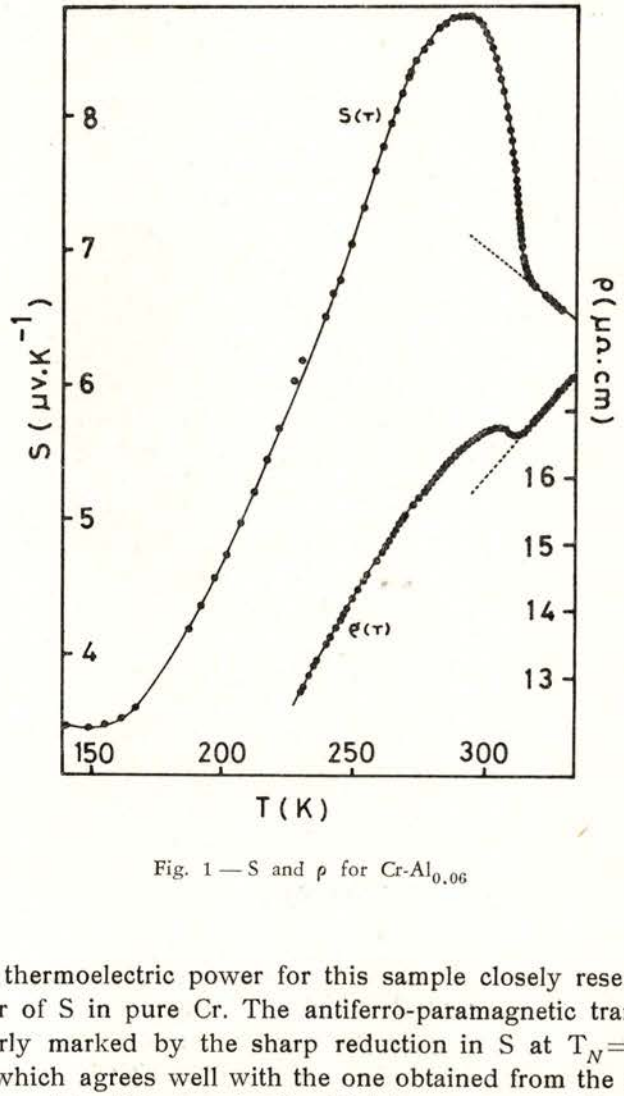

Fig. 1 — S and ρ for Cr-Al$_{0.06}$

The thermoelectric power for this sample closely resembles the behaviour of S in pure Cr. The antiferro-paramagnetic transition is also clearly marked by the sharp reduction in S at $T_N = 309.8$ K, a value which agrees well with the one obtained from the minimum in $d\rho/dT$, and is still close to the Néel temperature of Cr ($\simeq$ 311K). This reduction in S is essentially due to the sharp increase in the effective number of conduction electrons as the antiferromagnetic gap in the electron spectrum goes to zero at T_N. Similar effects have been observed in the thermoelectric power of Cr-Si [10], Cr-V, Cr-Mn [11] and Cr-Ni [12] alloys.

The temperature derivatives dS/dT and $d\rho/dT$ for $Cr_{99.94}Al_{0.06}$ are shown in Fig. 2, as a function of temperature. In both cases the antiferro-paramagnetic transition is clearly marked by a pronounced and very narrow dip of both derivatives at T_N. The general trend of both curves is remarkably similar, which suggests the possible existence of a close relationship between dS/dT and $d\rho/dT$ in the vicinity of the Néel temperature. The analysis of this important question and its possible theoretical implications for critical phenomena studies will be object of a forthcoming paper (with M. Ausloos).

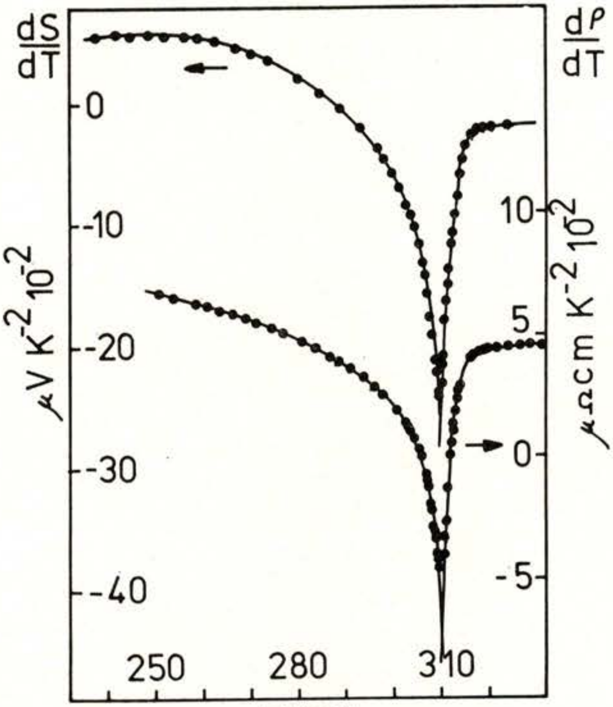

Fig. 2 — dS/dT and dρ/T for $Cr\text{-}Al_{0.06}$

Fig. 3 shows the temperature dependence of S for the sample with 1.93 at% Al. This curve is similar to that of $Cr_{99.94}Al_{0.06}$. However, the transition is not as sharp, probably due to the attenuation of the first-order character of the antiferromagnetic transition, as impurities are added to Cr [13]. Again, the Néel temperature as given by $d\rho/dT$ studies ($T_N = 290$) agrees well with the inflexion

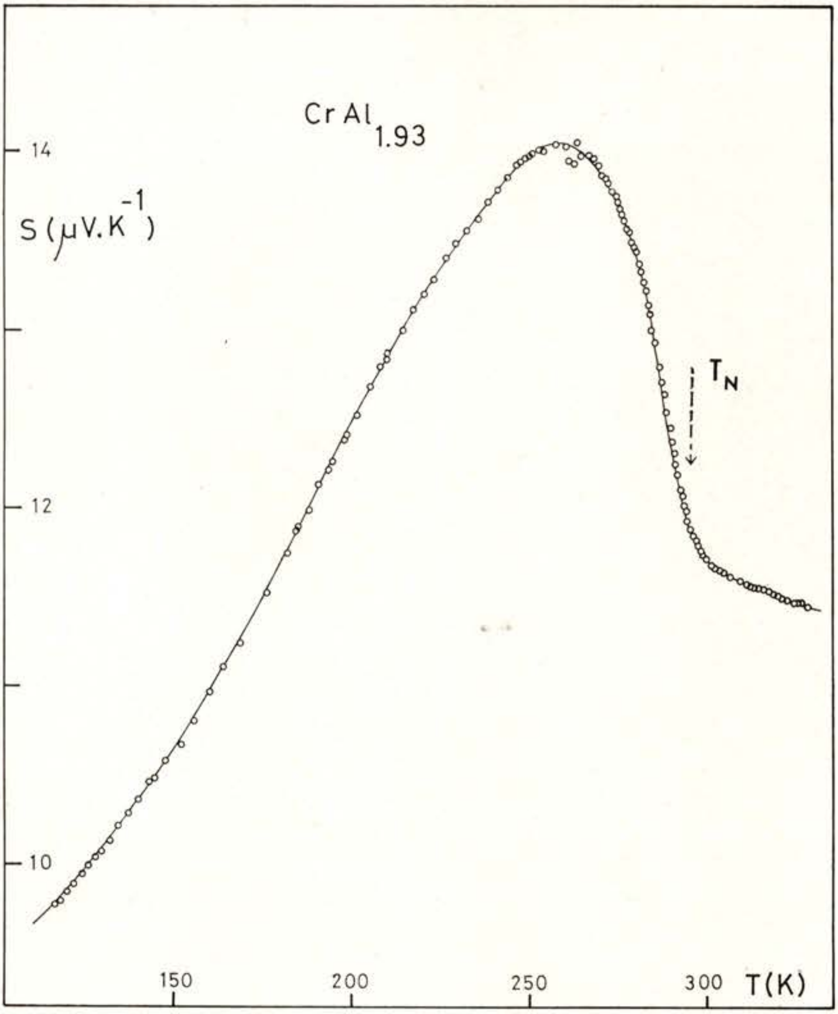

Fig. 3 — S for $Cr\text{-}Al_{1.93}$

point in the S(T) curve, as shown in the figure by the dotted arrow.

The temperature derivative of the thermoelectric power for $Cr_{98.07}Al_{1.93}$ (Fig. 4) still exhibits a marked «critical» behaviour, with a sharp dip at T_N, in the same manner as for $d\rho/dT$.

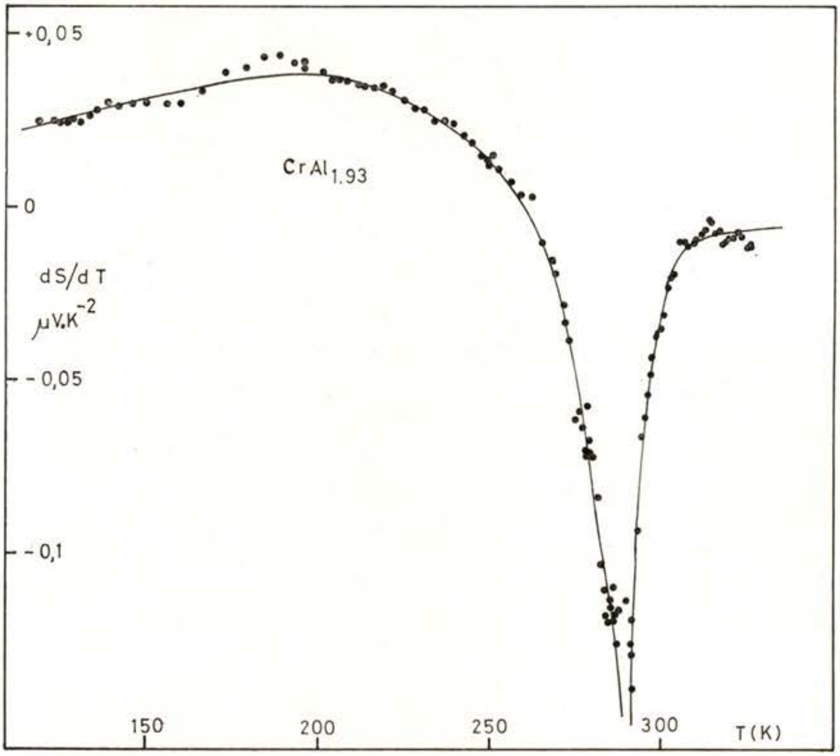

Fig. 4 — dS/dT for $Cr\text{-}Al_{1.93}$

The behaviour of S(T) for $Cr_{98.2}Al_{4.8}$ is shown in Fig. 5. The anomaly in the transition region is now much attenuated, and dS/dT (Fig. 6) shows a considerable broadening in the dip around T_N ($\simeq$ 288K). It is possible that this behaviour might be associated with a complex mixing of magnetic phases in the transition regime between incommensurate and commensurate antiferromagnetism [5], [14].

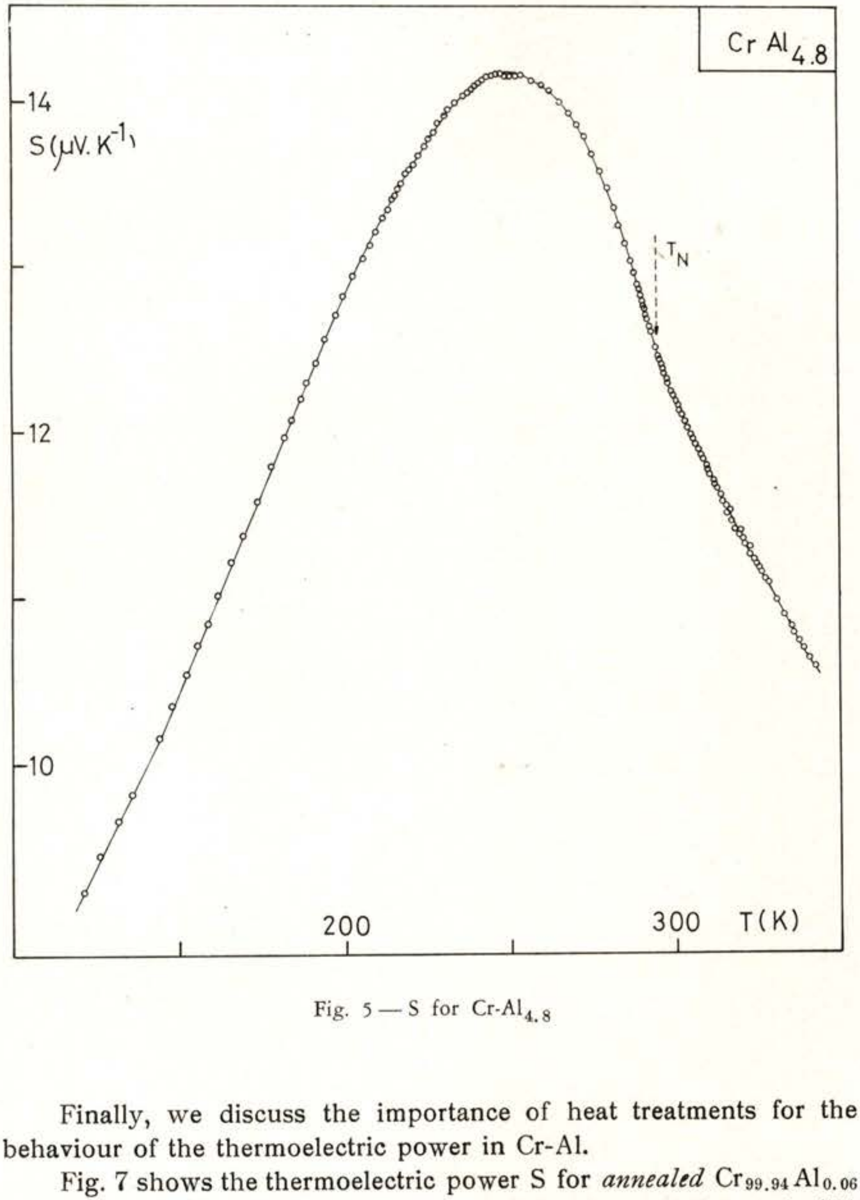

Fig. 5 — S for Cr-$Al_{4.8}$

Finally, we discuss the importance of heat treatments for the behaviour of the thermoelectric power in Cr-Al.

Fig. 7 shows the thermoelectric power S for *annealed* $Cr_{99.94}Al_{0.06}$ sample (I), compared with S measured in non-annealed sample (II) with the *same* nominal composition. Several points deserve mention here. First, the considerable reduction in the critical temperature (309.8 → 280K), the general broadening of the transition, and the greater dispersion of the experimental points for sample II. Second,

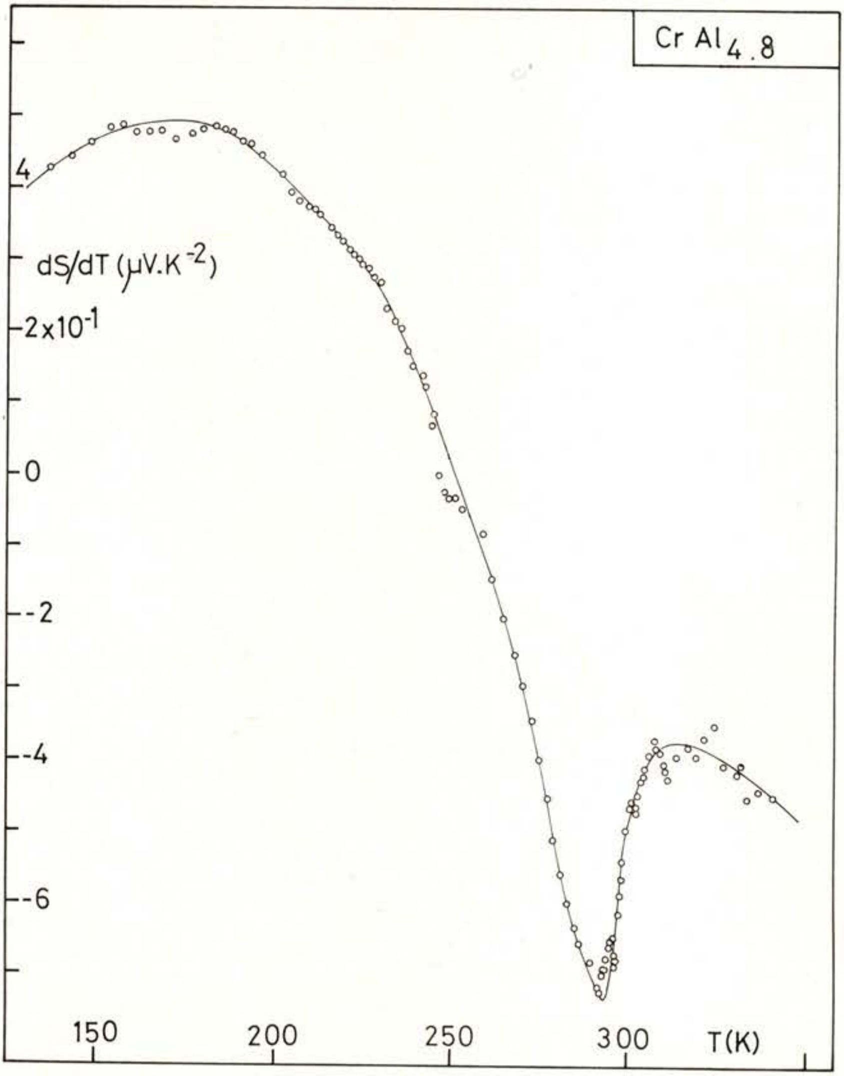

Fig. 6 — dS/dT for Cr-$Al_{4.8}$

two opposite effects appear in S: whereas for sample II (not annealed) S has much higher values in the cooperative phase, the reverse happens in the paramagnetic phase. However, the general shape and the pronounced reduction in S(T) near T_N are still present in the non--annealed sample.

We have also measured S in a sample with higher Al content (0.8 at %, nominal) in the non-annealed condition. As can be seen in Fig. 8, there is a dramatic enhancement in the magnitude of S in the antiferromagnetic state. On the other hand, in the paramagnetic phase there is again a marked reduction with respect to sample I.

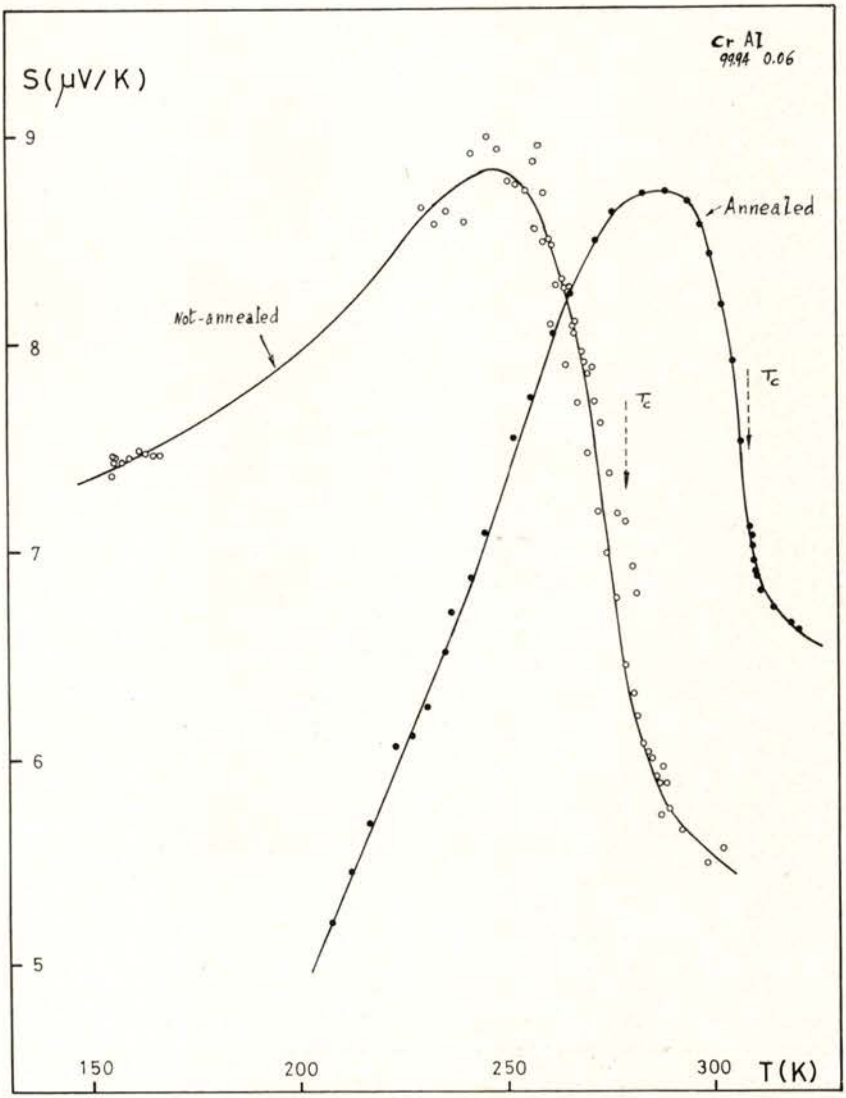

Fig. 7 — S for annealed (and non-annealed) Cr-Al$_{0.06}$

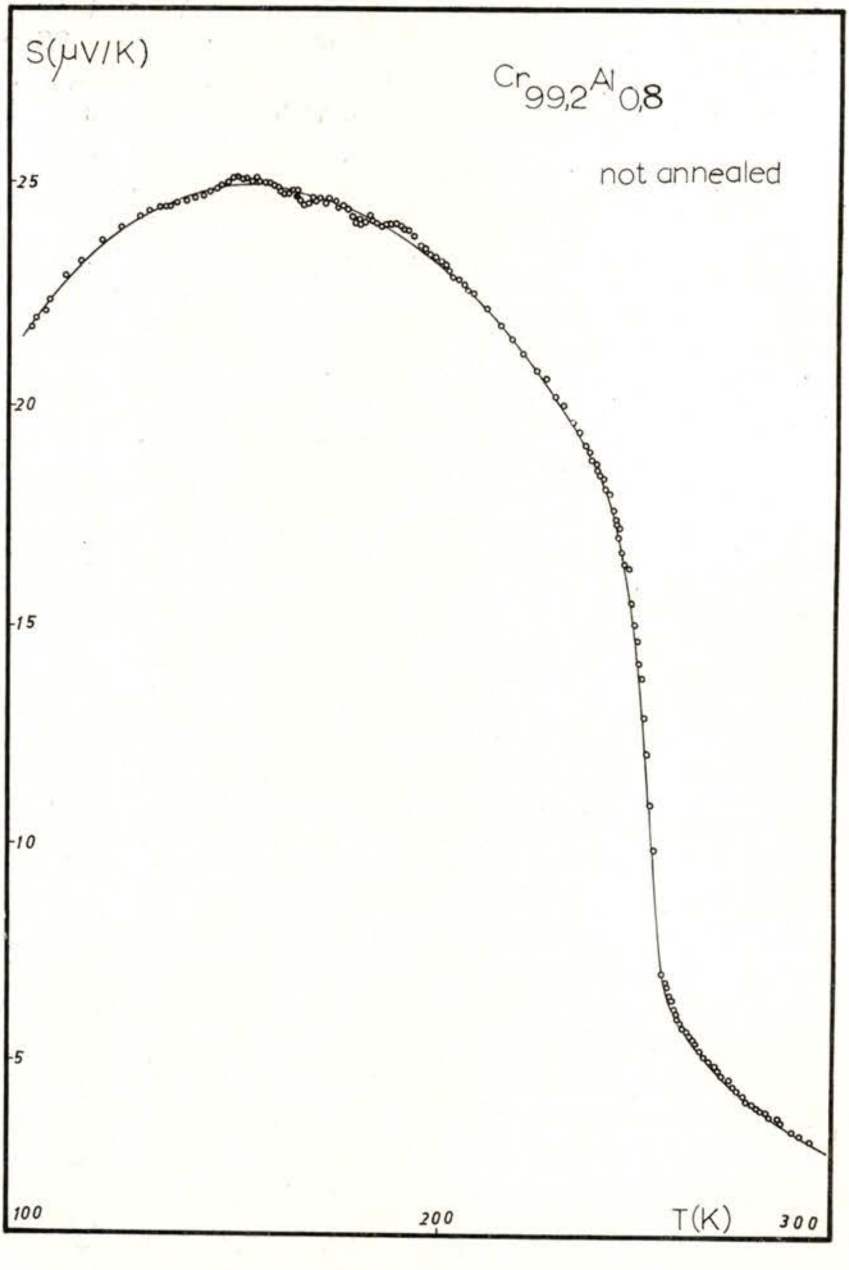

Fig. 8 — S for Cr-$Al_{0,8}$ (non-annealed)

5 — DISCUSSION

Taking the S curve for the sample with lowest Al content (Fig. 1), let us sumarize the main features in the thermoelectric power:

(i) S has fairly high values in the paramagnetic phase. In this region S is approximately linear in T, with a negative slope ($-dS/dT \sim 10^{-2}$ μV/K^2), *but* not extrapolating through the origin.
(ii) S is always positive (hotter junction negative).
(iii) The transition to the antiferromagnetic state enhances S (*positive* contribution).
(iv) A slight minimum is observed at low temperatures.

In order to discuss these features let us review briefly some relevant theory of the thermoelectric power, starting from Mott formulae [15]:

$$S = + \frac{\pi^2 k^2}{3e} . T . \left(\frac{\partial \, ln \rho}{\partial \varepsilon}\right)_{\varepsilon_F} \qquad \rho = \left(\frac{m^*}{e^2}\right) \cdot \frac{1}{n} \cdot \frac{1}{\tau} \tag{1}$$

where e, m^*, n have the usual meanings and τ is the electron relaxation time due to the scattering mechanisms. Combining these two expressions we obtain:

$$S = - \frac{\pi^2 k^2}{3e} . T . \left[\left(\frac{\partial \, ln \, n}{\partial \varepsilon}\right) + \left(\frac{\partial \, ln \, \tau}{\partial \varepsilon}\right)\right]_{\varepsilon = \varepsilon_F} \tag{2}$$

Assuming a one-band normal metal, several scattering mechanisms for the s-electrons and isotropic relaxation times $\tau_i \propto \varepsilon^{r_i}$ (r_i = small number), one can show that:

$$S = - \frac{\pi^2 k^2}{3 e \varepsilon_F} . T . \left[\left(\frac{\partial \, ln \, n}{\partial \, ln \, \varepsilon}\right)_{\varepsilon_F} + \sum_i r_i \frac{\rho_i}{\rho}\right] \tag{3}$$

where $\rho = \sum_i \rho_i$.

The first contribution in the second member depends directly on the band structure of the *carriers*. Assuming these to be s-electrons (as usually happens in normal and transition metals), and a free electron model with $n \propto \varepsilon^{3/2}$, we get a negative contribution to S, linearly dependent on T:

$$S = -\frac{\pi^2}{2} \cdot \frac{k}{e} \cdot \left(\frac{kT}{\varepsilon_F}\right) \tag{4}$$

Its magnitude is fairly small at ordinary temperatures, since $k/e \sim 86$ μV/Kelvin, $kT \sim 10^{-2}$ *e*V, $\varepsilon_F \sim 5$ *e*V, and $S \approx$ μV/Kelvin.

The remaining contributions in (3) do not generally give, for the usual scattering mechanisms (impurities, phonons, spin-disorder in the paramagnetic phase) much different results. For example, assuming the predominance of *one* single scattering mechanism we get simply:

$$S = -\frac{\pi^2}{3} \cdot \frac{k}{e} \cdot \left(\frac{kT}{\varepsilon_F}\right) \cdot r_1 \quad \approx \mu\text{V/Kelvin} \tag{5}$$

Therefore, in order to obtain much higher values for S, as is experimentally observed in most transition metals (~ 10 – 20 μV/Kelvin), one must seek alternative explanations.

There is no doubt that contributions from n in (3) are essentially due to *s*-electrons, since the *d*-electrons in transition metals have very low mobility (narrow *d*-bands, high effective mass). The successful explanation was given by Mott [15], who noticed hat in metals with an incomplete *d*-band the conduction *s*-like electrons can be effectively scattered (e.g. by phonons) *into* vacant states in the *d*-band. Since the transition probability is proportional to the density of states in the *d*-band, $N_d(\varepsilon)$, and this is very high for a narrow *d*-band, the *s*-*d* interaction dominates the scattering process so that approximately

$$\frac{1}{\tau} \propto N_d(\varepsilon) \tag{6}$$

Therefore, for transition metals we have from (2):

$$S \simeq -\frac{\pi^2}{3} \cdot \frac{k}{e} . kT . \left[\left(\frac{\partial \ln n}{\partial \varepsilon}\right)_{\varepsilon_F} - \left(\frac{\partial \ln N_d}{\partial \varepsilon}\right)_{\varepsilon_F}\right] \tag{7}$$

In transition metals, $N_d(\varepsilon)$ is a very irregular function of ε with very sharp peaks, where $|\partial N_d / \partial \varepsilon|$ is very high. If ε_F, for a particular element, occurs near such a peak, $|S|$ will be very large. The sign of S can be $(+)$ or $(-)$, depending on whether ε_F is to the right or left of such a peak, respectively. Similar reasonings can be made near a sharp minimum in N_d.

A simple physical explanation of this effect has been given by Kolomoets et al [16]. Electrons in the interval kT around ε_F are subjected to selective scattering due to pronounced variation of N_d over this interval. In the case of an almost filled *d*-band, the electrons which are diffused from the cold junction of the specimen to the hot one are subjected to the more intense scattering. This facilitates a still greater accumulation of electrons at the cold end of the specimen, which means $S < 0$ in *this* case $(dN_d / d\varepsilon < 0)$.

We may now understand the high values of S for Cr in the paramagnetic region and from $S > 0$ we conclude that ε_F sits in a point where $(\partial N_d / \partial \varepsilon) > 0$. Due to the irregular shape of $N_d(\varepsilon)$, a slight shift in ε_F within the *d*-band, produced e. g. by an increase in temperature or slight changes in the lattice parameters [17], may produce dramatic changes in $\partial N_d / \partial \varepsilon$. This may be the cause for the reversal in the sign of S which occurs in Cr at fairly high temperatures (~ 1500 K) [17].

Since $(\partial N_d / \partial \varepsilon)_{\varepsilon F}$ will thus generally depend on T, we may understand why the paramagnetic thermoelectric power does not extrapolate through the origin. However, the small linear contribution to S in the paramagnetic state, with a negative slope of $\sim 10^{-2}$ μV/K is probably due to the *n*-term in (7). The negative sign indicates $(\partial n / \partial \varepsilon)_{\varepsilon F} > 0$, i.e., the Fermi level occurs in the first part of the *s*-band.

Let us examine now the pronounced enhancement of S in the antiferromagnetic state, certainly associated with the severe reduction in the electronic concentration *n*, as confirmed from electrical resistivity measurements. Such reduction is caused by the new gaps which appear in the system, due to the extra periodicity introduced by the antiferromagnetic structure [18, 19]. These gaps appear in the conduction *s*-band, affecting therefore the first term in equation (2). Obviously, one such gap (Δ) must occur in the vicinity of the Fermi level in order to affect drastically the transport properties.

This gap increases with the enhancement of the magnetic periodic structure, i. e., with the sub-lattice magnetization; a BCS — like theory describes well the temperature dependence of Δ [20]. By coincidence, this function is fairly close to the Brillouin function $B_{1/2}(T/T_N)$. At absolute zero we have

$$\Delta(0) \simeq 1.76\ kT_N$$

so that larger gaps are expected for the antiferromagnets with higher Néel temperatures.

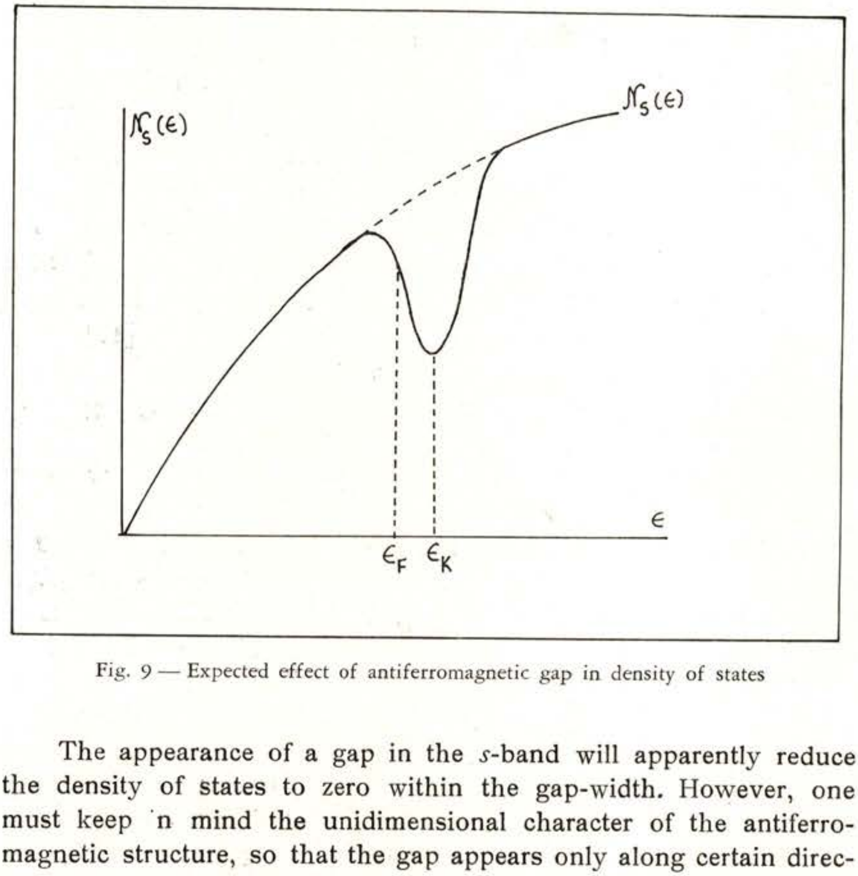

Fig. 9 — Expected effect of antiferromagnetic gap in density of states

The appearance of a gap in the *s*-band will apparently reduce the density of states to zero within the gap-width. However, one must keep in mind the unidimensional character of the antiferromagnetic structure, so that the gap appears only along certain directions in k-space. Therefore, the integrated density of states over all directions, $N_s(\varepsilon)$, will simply present a dip in the gap region around a particular energy ε_K [21], as shown in Fig. 9.

Two different cases may appear, whether ε_F occurs below or above ε_K [22]. In the first case, as shown in Fig. 9, the appearance of the magnetic gap produces negative $(\partial n/\partial \varepsilon)_{\varepsilon_F}$ values, which means positive S-values, as occurs in our Cr-Al alloys. The sharp increase in S is certainly due to the dramatic rise in Δ as T is slightly lowered below T_N. However, this increase soon gets fairly slow, and for $T/T_N \approx 0.7$ almost 85% of the gap full value $\Delta(0)$ has been attained. This means that $(\partial n/\partial \varepsilon)_{\varepsilon_F}$ has reached now an almost constant negative value, and the kT factor in eq. (2) will dominate the physics, making S decrease towards fairly low values. We see therefore the origin of the well marked maximum in S which occurs at intermediate temperatures in our alloys.

Some difficulty arises with the ($Cr_{99.94}Al_{0.06}$) sample at the lowest temperatures reached, where a minimum occurs in S, certainly preceeding a maximum at still lower temperatures, since S must ultimately vanish as T → 0K (3rd. principle). This maximum has in fact been observed in pure Cr between 50 and 100K, and has been atributted to a phonon-dragg efect [23]. It is known that the addition of impurities always decreases the magnitude of these effects. This may explain why we do not observe such a minima in our curves for the other (more impure) Cr-Al samples in the range of temperatures investigated.

Finally, let us examine the efects of alloying [23], assuming the validity of Mathiessen's rule

$$\rho = \rho_i + \rho_j \tag{8}$$

where ρ is the total resistivity (Cr-Al), ρ_i is the ideal resistivity of the pure solvent metal (Cr), and ρ_j is the resistivity attributable to scattering of conduction electrons by impurities of type *j*. Substitution of (8) into (1) gives

$$S = + \frac{\pi^2 k^2 T}{3e} \left[\frac{\partial \ln(\rho_i + \rho_j)}{\partial \varepsilon} \right]_{\varepsilon_F} \tag{9}$$

and a bit of algebra leads to the Gorter-Nordheim relation

$$S = S^{Al} + \frac{\rho^{Cr}}{\rho} (S^{Cr} - S^{Al}) \tag{10}$$

Here S^{Cr} is the diffusion thermopower of pure Cr, and

$$S^{Al} = \frac{\pi^2 k^2 T}{3e} \left[\frac{\partial \ln \rho^{Al}}{\partial \varepsilon} \right]_{\varepsilon_F} \quad , \tag{11}$$

where ρ^{Al} is the part of the resistivity attributable to scattering of conduction electrons by Al impurities.

In the paramagnetic region, and at a definite temperature, a plot of S vs $1/\rho$ should be a straight line

$$S = a + b(1/\rho) \tag{12}$$

Since ρ increases monotonically with the addition of Al (and most likely $S^{Cr} >> S^{Al}$), we should expect a corresponding monotonic decrease of S in the paramagnetic phase. Although a pronounced decrease occurs in all our samples with respect to Cr, it is by no means monotonic.

This discrepancy is probably due to the violation of the basic assumptions of the theory, *i.e.*, the validity of Matthiessen's rule and the presumption that alloying does not alter the Fermi surface and thereby change S^{Cr}. We think that this last assumption is not justified in Cr-Al, in view of the pronounced alterations in such basic parameters as the electronic specific heat constant (γ) and the Debye temperature (Θ), which are known to occur in this system for low Al concentrations. Thus, thermopower measurements give another indirect confirmation of the great complexity of the Cr-Al system.

Further work is now in progress, in order to obtain a better understanding of the very peculiar physical properties of Cr-Al alloys with low Al content.

The authors wish to thank Dr. M. Ausloos from Univ. Liège, for the very iluminating discussions and his permanent interest and encouragement. Thanks are due also to: Dr. Margarida Costa, from Univ. Coimbra, for providing the inital samples which made possible to start this work; Dr. H. Stone, from Imp. College of London, for preparing the $Cr_{99.94}Al_{0.06}$ sample; Prof. J. M. Araújo, for useful comments and help during the correction of proofs; Eng. Jaime Bessa for the valuable technical assistance; and to Prof. Maia e Costa for the metalographic studies. Finally, the financial support given by the Instituto Nacional de Investigação Científica (INIC, Portugal) and by the NATO Organization (Research Grant no. 1481) is gratefully acknowledged.

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