

CRITICAL BEHAVIOUR OF THE CdCr_2Se_4 , HgCr_2Se_4 AND CuCr_2Se_4 SPINELS NEAR THEIR CURIE POINTS*

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For the chalcogenide spinels CdCr_2Se_4 , HgCr_2Se_4 and CuCr_2Se_4 the specific magnetization σ vs magnetic field H and temperature T were determined for temperatures close to their Curie points T_c . Sharp kinks in the $\sigma(T)$ plots were observed in magnetic fields up to 20 Oe. These kinks were used for the determination of the Curie points, the temperature dependence of the spontaneous magnetization σ_0 near T_c and the critical exponent β ($\sigma_0 \sim (T_c - T)^\beta$). The results of the magnetization measurements as a function of the magnetic field intensity for $5 \cdot 10^{-4} < \varepsilon = T - T_c / T_c < 5 \cdot 10^{-3}$ were used for the determination of critical exponent γ , which characterizes temperature changes of the initial magnetic susceptibility χ_0 near T_c ($\chi_0^{-1} \sim (T - T_c)^\gamma$), and exponent δ for the critical isotherm ($\sigma \sim H_{\text{eff}}^{1/\delta}$). The obtained values of the critical exponents $\beta \simeq 1/3$, $\gamma \simeq 4/3$ and $\delta \simeq 4.2$ seem to suggest that the spinels investigated are the Heisenberg ferromagnet.

1. Introduction

The critical behaviour of ferromagnet near the Curie point T_c has been the subject of many experimental [1-10] and theoretical works [11-21]. It was found that changes of the spontaneous magnetization σ_0 and initial magnetic susceptibility χ_0 with temperature T near T_c follow the dependences:

$$\sigma_0(T) = A(T_c - T)^\beta \quad T \lesssim T_c, \quad (1)$$

$$\chi_0^{-1}(T) = B(T - T_c)^\gamma \quad T \gtrsim T_c, \quad (2)$$

whereas changes of magnetization with the effective magnetic field intensity H_{eff} satisfy the dependence

$$\sigma(H_{\text{eff}}) = CH_{\text{eff}}^{1/\delta} \quad T \simeq T_c. \quad (3)$$

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Here β , γ and δ are the critical exponents characteristic of a given ferromagnetic. Their experimental values lie in the ranges:

$$0.33 < \beta_{\text{exp}} < 1.96$$

$$0.91 < \gamma_{\text{exp}} < 1.37$$

$$2.0 < \delta_{\text{exp}} < 7.0$$

Theoretical considerations of the critical behaviour of ferromagnet near T_c have revealed that in the case of the cubic lattice, the exponents β , γ and δ assume the following values: in the molecular field theory $\beta = 1/2$, $\gamma = 1$, $\delta = 3$; in the three-dimensional Ising's model $0.3 < \beta < 5/16$, $\gamma = 5/4$, $\delta = 5.2$; and in the Heisenberg's model $\beta = 1/3$, $\gamma = 4/3$, $\delta = 5$. It means that the experimental determination of the values of the critical exponents β and γ for a given ferrimagnet enables us to attribute it a suitable model elucidating its magnetic properties.

In the present work I would like to show the results of study of critical behaviour near the Curie point for selenospinel CdCr_2Se_4 , HgCr_2Se_4 , and CuCr_2Se_4 .

2. Experimental

Measurements in magnetic fields $H_z > 100$ Oe were performed using an electromagnet of the Weiss type. The magnetic properties of ferromagnet in weak magnetic fields (up to 100 Oe) were measured using a specially designed measuring assembly (Fig. 1) consisting

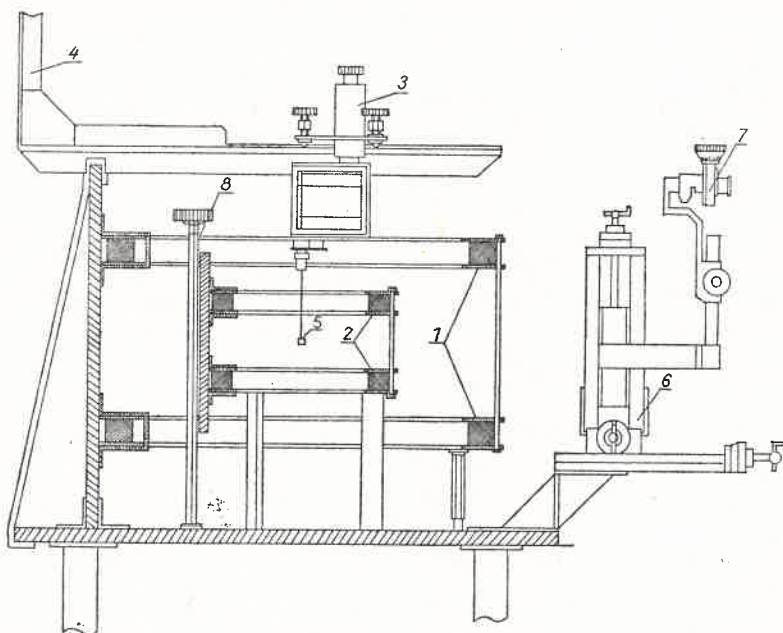


Fig. 1. The schema of the set-up for measuring magnetic properties in weak magnetic fields. 1 — Helmholtz coils creating weak magnetic fields. 2 — Helmholtz coils creating the magnetic field gradient. 3 — A magnetic balance. 4 — Suspension of the magnetic balance. 5 — A sample. 6 — A cruciform support. 7 — A micro-ocular. 8 — A support for changing the position of the coils 1 relative to the coils 2

of two pairs of Helmholtz coils coaxially fixed on a common stand. The coils (1) were connected in series to create a uniform weak magnetic field $H_z = 24 i_1$ directed along the vertical z axis. This field is proportional to the current intensity i_1 flowing through the coils. The coils (2) are connected in the push-pull system and create a gradient of the magnetic field intensity $dH_z/dz = 2.86 i_2$, which is directed along the z axis and proportional to the current intensity i_2 in the gradient coils.

The specific magnetization σ and magnetic susceptibility χ were measured using the Faraday method [23].

The specific magnetization and magnetic susceptibility of polycrystalline selenospinel CdCr_2S_4 , HgCr_2S_4 and CuCr_2Se_4 were measured in outer magnetic fields from 2 to 8000 Oe at 90–150 K for CdCr_2S_4 and HgCr_2S_4 and 400–430 K for CuCr_2Se_4 .

The specific magnetization and magnetic susceptibility were measured to within 1% and the temperature of a sample was stabilized and measured to within 0.1 K. The study was carried out for spherically shaped samples. The sample demagnetizing field was taken into account in the treatment of the results.

3. Results and discussion

Figs 2, 3 and 4 present the temperature changes of specific magnetization near Curie points, as obtained in weak magnetic fields for CdCr_2S_4 , HgCr_2S_4 and CuCr_2Se_4 , respectively. It can be seen that near T_c , in effective magnetic fields up to 20 Oe, there occurs a sharp kink on the $\sigma(T)$ curves. This kink shifts towards lower temperatures with an increase in magnetic field intensity. The kink in low magnetic fields was used to determine the Curie point of the selenospinel investigated and the temperature changes of their spontaneous magnetization near T_c [22].

The critical coefficient β (Eq. (1)) was determined from the slope of the dependence $\ln(\sigma_0^2) = f[\ln(T_c - T)]$ (Fig. 5) plotted for such the temperature range, for which $5 \cdot 10^{-4} < \varepsilon = T_c - T/T_c < 5 \cdot 10^{-3}$.

Extrapolation of the isotherms $H_{\text{eff}}/\sigma = f(\sigma^2)$ to $\sigma^2 = 0$ gave values of the reciprocals of initial magnetic susceptibility χ_0^{-1} near T_c . From the slope of the line $\ln(\chi_0^{-1}) = f[\ln(T - T_c)]$ for $5 \cdot 10^{-4} < \varepsilon < 5 \cdot 10^{-3}$ (Fig. 6) the critical coefficient γ was determined (Eq. (2)).

The critical coefficient δ (Eq. (3)) was determined from the slope of the line $\ln \sigma = f[\ln H_{\text{eff}}]$, plotted for $\varepsilon = 2 \cdot 10^{-4}$ (Fig. 7).

The obtained values of the critical exponents β , γ , δ and Curie temperatures for the selenospinel investigated are given in Table I.

It can be seen that the values of the critical exponents β and γ are, within the experimental error limits, 1/3 and 4/3 which were theoretically predicted for the Heisenberg ferromagnets [12, 19]. It was also found that the critical exponents β , γ and δ follow, within the experimental error limits, the scaling law

$$\gamma \geq \beta(\delta - 1).$$

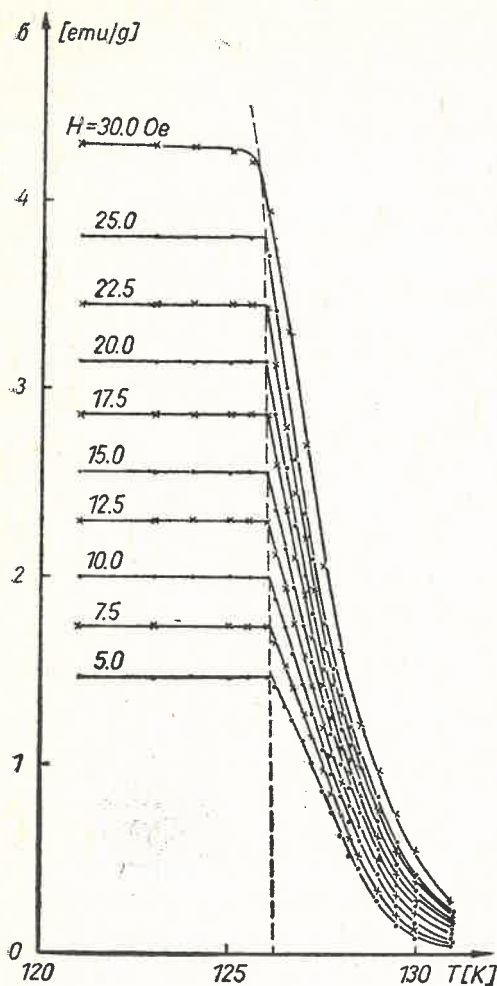


Fig. 2

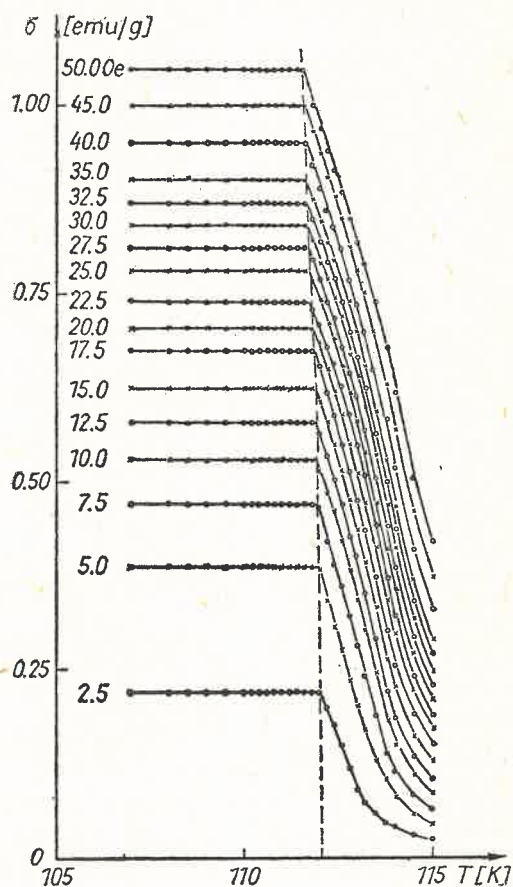


Fig. 3

Fig. 2. Temperature changes of the specific magnetization of CdCr_2Se_4 near the Curie point, measured in effective magnetic fields up to 30 Oe

Fig. 3. Temperature changes of the specific magnetization of HgCr_2Se_4 near the Curie point, measured in effective magnetic fields up to 50 Oe

To check the magnetic state equation all the experimental data were plotted as a function $H_{\text{eff}} \varepsilon^{\beta\delta} = f[\sigma \varepsilon^{\beta}]$.

Fig. 8 shows the above mentioned dependence for CdCr_2Se_4 . The same dependences $H_{\text{eff}} \varepsilon^{\beta\delta} = f[\sigma \varepsilon^{\beta}]$ were obtained for HgCr_2Se_4 and CuCr_2Se_4 . The experimental points lie on two curves for $T < T_c$ and $T > T_c$, with that the latter curve passes through the origin of the coordinates.

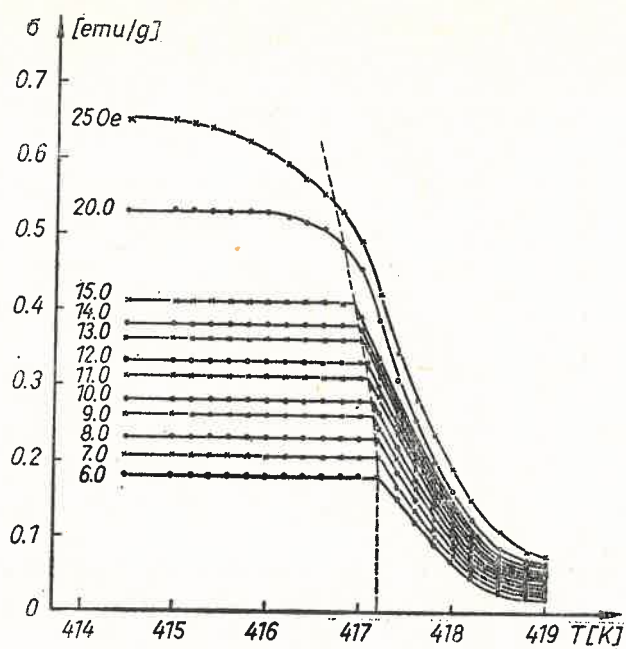


Fig. 4. Temperature changes of the specific magnetization of CuCr_2Se_4 near the Curie point, measured in effective magnetic fields up to 25 Oe

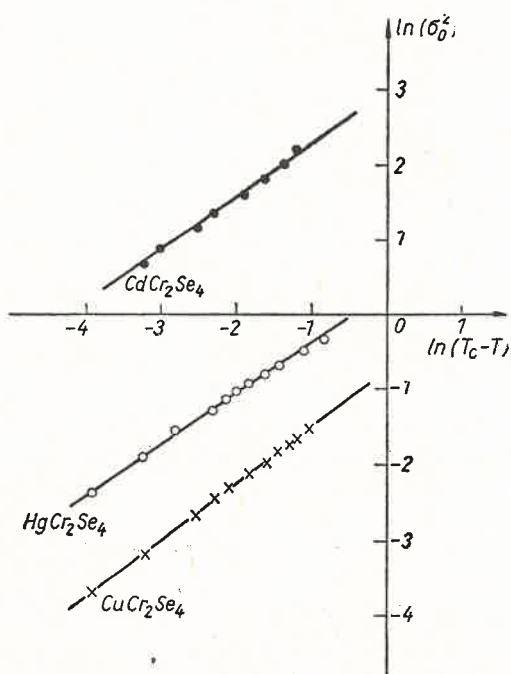


Fig. 5. The dependence $\ln(\sigma_0^2) = f[\ln(T_c - T)]$ near the Curie point for the selenospinel investigated

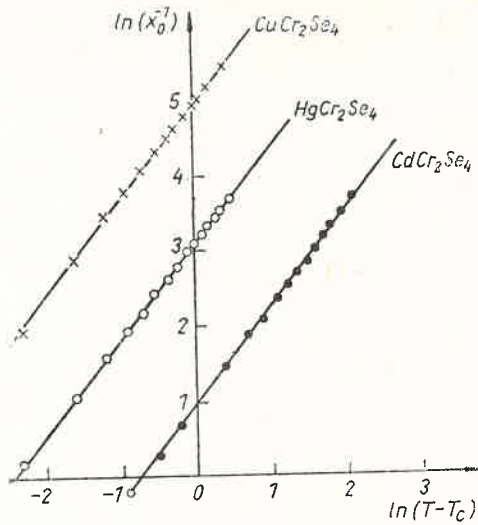


Fig. 6. The dependence $\ln(\chi_0^{-1}) = f[\ln(T - T_c)]$ near the Curie point for the selenospinel investigated

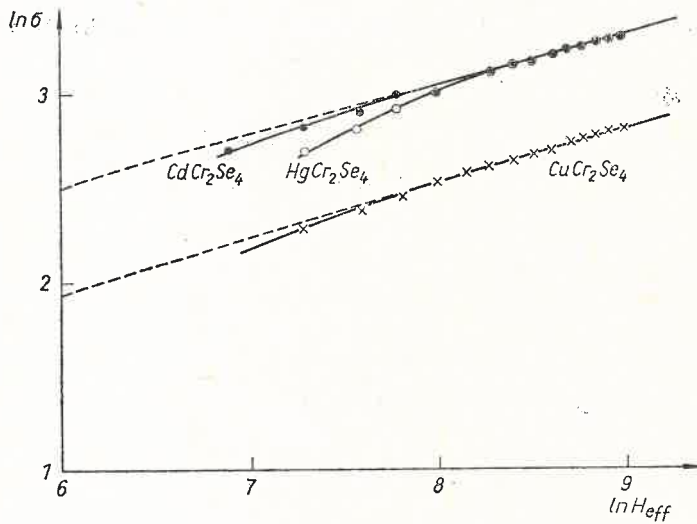


Fig. 7. The dependence $\ln \sigma = f[\ln H_{eff}]$ obtained for $T \simeq T_c$ ($\epsilon = 2 \times 10^{-4}$)

TABLE I

Spinel	T_c [K]	β	γ	δ
$CdCr_2Se_4$	126.20 ± 0.05	0.34 ± 0.02	1.29 ± 0.02	4.2 ± 0.3
$HgCr_2Se_4$	112.04 ± 0.05	0.34 ± 0.02	1.30 ± 0.02	4.1 ± 0.3
$CuCr_2Se_4$	117.20 ± 0.05	0.37 ± 0.02	1.32 ± 0.02	4.2 ± 0.3

The results obtained are in a good agreement with those obtained for CdCr_2Se_4 [9, 10] and show that the selenospinel investigated fulfill the equation of magnetic states.

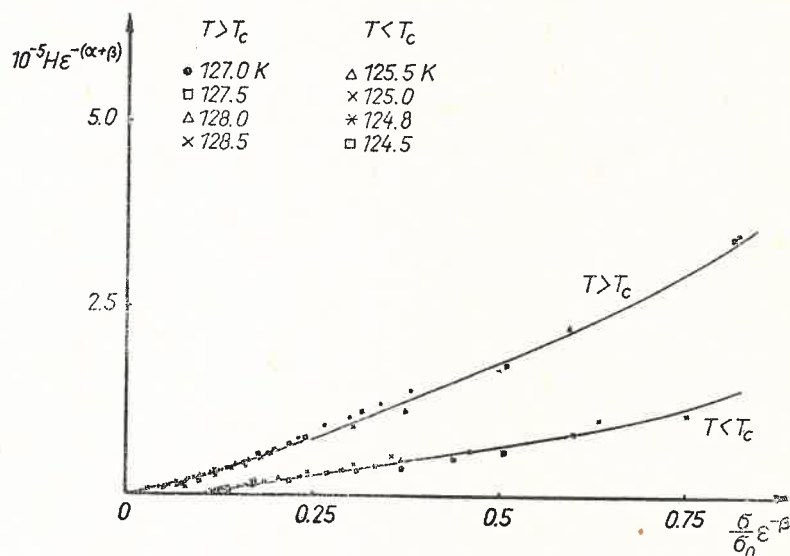


Fig. 8. The dependence $H_{\text{eff}}\epsilon^{\beta\delta} = f(\sigma\epsilon^{\beta})$ for the selenospinel CdCr_2Se_4 .

REFERENCES

- [1] J. E. Noakes, A. Arrott, *J. Appl. Phys.* **35**, 931 (1964).
- [2] S. Araj, R. V. Colvin, *J. Appl. Phys.* **35**, 2424 (1964).
- [3] J. S. Kouvel, M. E. Fisher, *Phys. Rev.* **136A**, 1626 (1964).
- [4] S. Araj, *J. Appl. Phys.* **36**, 1136 (1965).
- [5] J. S. Kouvel, D. S. Rodbell, *J. Appl. Phys.* **38**, 979 (1967).
- [6] G. Devey, *J. Phys.* **29**, 74 (1968).
- [7] J. T. Ho, J. D. Lister, *J. Appl. Phys.* **40**, 1270 (1969).
- [8] J. T. Ho, J. D. Lister, *Phys. Rev. Lett.* **22**, 606 (1969).
- [9] K. Miyatani, *J. Phys. Soc. Jap.* **28**, 259 (1970).
- [10] K. Miyatani, *J. Appl. Phys.* **41**, 1272 (1970).
- [11] G. A. Baker, *Phys. Rev.* **124**, 768 (1961).
- [12] C. Domb, M. F. Sykes, *Phys. Rev.* **128**, 168 (1962).
- [13] J. Gammel, W. Marshall, *Proc. Roy. Soc. A* **275**, 257 (1963).
- [14] G. A. Baker, *Phys. Rev.* **129**, 99 (1963).
- [15] D. S. Gaunt et al., *Phys. Rev. Lett.* **13**, 713 (1964).
- [16] R. B. Griffiths, *Phys. Rev.* **158**, 176 (1967).
- [17] M. E. Fisher, *J. Appl. Phys.* **38**, 981 (1967).
- [18] H. E. Stanley, *J. Appl. Phys.* **38**, 977 (1967).
- [19] S. Freeman, P. J. Woytowicz, *Phys. Lett.* **26A**, 231 (1967).
- [20] N. Menyuk, *Phys. Rev.* **166**, 510 (1968).
- [21] P. Schofield, *Phys. Rev. Lett.* **22**, 606 (1969).
- [22] P. J. Woytowicz, M. Rayl, *Phys. Rev. Lett.* **20**, 1489 (1968).
- [23] Z. Obuszko, *Acta Phys. Pol.* **24**, 135 (1963).