## Experimental values of the gap exponents for the paramagnetic region

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Results are presented for measurements of the initial paramagnetic susceptibility (i.e., for zero magnetic field) and also of the first three nonlinear terms of the susceptibility expansion in a series relative to a magnetic field, for the compound  $HgCr_2Se_4$  in the temperature range  $T_c + 1$  K  $\leq T \leq T_c + 20$  K (i.e., for  $10^{-2} \le \epsilon \le 10^{-1}$ ). From the temperature functions obtained for the four coefficients of expansion of the paramagnetic susceptibility, three values of the gap exponents were found:  $\Delta_4 = 1.68 \pm 0.04$ ,  $\Delta_6 = 1.64 \pm 0.1$ , and  $\Delta_8 = 1.47 \pm 0.33$ .

The gap exponents  $\Delta_{2n}$  have been determined using the experimental method reported in Ref. I of examining the nonlinear (quadratic) paramagnetic effect. In this method measurements are made of changes in susceptibility  $\Delta X = X_B - X_0$  induced by the application of an external magnetic field, where  $X_B$  is the susceptibility at  $B\neq 0$ , and  $X_0$  is the susceptibility at  $B=0$ . Making use of the measurement method described in Ref. 1, based on binding  $\Delta x$  as a function of  $B$  at various temperatures, the coefficients of the zero, second, fourth, and sixth terms were determined in the expansion of the paramagnetic susceptibility in a series relative to a magnetic field, i.e.,

$$
\chi_B = \chi_0 + \chi_2 B^2 + \chi_4 B^4 + \chi_6 B^6 + \cdots \tag{1}
$$

The main purpose of our work was to determine the experimental values of the gap exponents and, moreover, to compare these values with the theoretical relations  $\Delta_4 = \Delta_6 = \Delta_8$ arising from the scaling laws.

It is known that above the Curie temperature the free energy in a nonzero magnetic field may be expressed as fol $lows$ : $^{2,3}$ 

$$
F(H) = F(0) - \beta^{-1} \sum_{n=1}^{\infty} \frac{(\beta H)^{2n} \Gamma_{2n}}{(2n)!},
$$
 (2)

where  $\beta = 1/kT$ 

$$
\Gamma_{2n} = \sum_{\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_{2n}} G^{\phi \cdots \phi}(\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_{2n}) ,
$$

and  $G^{\phi \cdots \phi}$  are correlation functions described in Refs. 2 and 3. The temperature behavior of the values  $\Gamma_{2n}$  in the vicinity of  $T_c$  is given by the relation

$$
\Gamma_{2n} \sim \epsilon^{-2\Delta_{2n}} \Gamma_{2n-2} \quad , \tag{3}
$$

where  $\epsilon = (T - T_c)/T_c$  and  $\Delta_{2n}$  is the gap exponent. From Eq. (2) and the definition of susceptibility it may be concluded that if we express the paramagnetic susceptibility in the form of a series relative to the magnetic field induction [Eq. (1)], then the following relations are satisfied:

$$
\beta^{-1}\chi_0 \sim \Gamma_2, \quad \beta^{-3}\chi_2 \sim \Gamma_4 \quad ,
$$
  

$$
\beta^{-5}\chi_4 \sim \Gamma_6, \quad \beta^{-7}\chi_6 \sim \Gamma_8 \quad .
$$
 (4)

Knowledge of the temperature relations, such as given by Eq. (3), between the coefficients in the expansion [Eq. (1)] in the vicinity of  $T_c$  permits the explicit determination of the gap exponents  $\Delta_{2n}$ .

Measurements of  $x_0$ ,  $x_2$ ,  $x_4$ , and  $x_6$  were carried out for the compound HgCr<sub>2</sub>Se<sub>4</sub> (polycrystalline sample,  $T_c \approx 106.5$ K) in the temperature range  $T_c + 1$  K  $\leq T \leq T_c + 20$  K, and for a frequency of the measuring field of 1 MHz. Demagnetization effects were taken into account. Values of coefficients in the nonlinear terms of expansion [Eq. (1)] were obtained by measurement of the susceptibility variation  $\Delta x = x_B - x_0$  induced by the magnetic field<sup>1</sup> followed by numerical separation of the particular terms of this expansion. Figure 1 shows the temperature functions  $(T/T_c)\chi_0$ ,  $(T/T_c)^3 \chi_2$ ,  $(T/T_c)^5 \chi_4$ , and  $(T/T_c)^7 \chi_6$  in a log-log scale. Assuming the following exponential temperature depen-



FIG. 1. Functions  $\bar{x}_0 = (T/T_c) x_0$ ,  $-\bar{x}_2 = -(T/T_c) x_2$ ,  $\bar{x}_4 = (T/T_c) x_3$  $T_c$ )<sup>5</sup> $\chi$ <sub>4</sub>, and  $-\bar{\chi}_6 = -(T/T_c)$ <sup>7</sup> $\chi$ <sub>6</sub> vs  $(T - T_c)/T_c$  in a log-log scale. The magnitudes are given in the international system of units (SI). The susceptibility in Eq. (1) is defined by the relation  $X_B = \partial M/\partial H$ , where  $M$  is the magnetic moment per unit volume, and  $H$  is the magnetic field strength.

 $\pmb{\gamma}$  $\Delta_4$  $\Delta_6$  $\Delta_8$  $\gamma_2$  $\gamma_4$  $\pmb{\gamma}_6$ 1.30 4,66 7.94 10.87 1.68 1.47 1.64  $± 0.02$  $± 0.05$  $± 0.15$  $± 0.5$  $\pm$  0.04  $\pm 0.1$  $± 0.33$ 

TABLE I. Experimental values of exponents  $\gamma$ ,  $\gamma_2$ ,  $\gamma_4$ , and  $\gamma_6$  and  $\Delta_4$ ,  $\Delta_6$ , and  $\Delta_8$ .

dences of the magnitudes (cf. Refs. 3 and 4):

$$
\frac{T}{T_c} \chi_0 \sim \epsilon^{-\gamma}, \quad \left(\frac{T}{T_c}\right)^3 \chi_2 \sim \epsilon^{-\gamma_2},
$$
\n
$$
\left(\frac{T}{T_c}\right)^5 \chi_4 \sim \epsilon^{-\gamma_4}, \quad \left(\frac{T}{T_c}\right)^7 \chi_6 \sim \epsilon^{-\gamma_6}, \quad (5)
$$

one can determine (cf. Fig. 1) the values of  $\gamma$ ,  $\gamma_2$ ,  $\gamma_4$ , and  $\gamma_6$  as shown on Table I. This table also gives the values of the gap exponents;  $\Delta_4 = \frac{1}{2}(\gamma_2 - \gamma)$ ,  $\Delta_6 = \frac{1}{2}(\gamma_4 - \gamma_2)$ , and  $\Delta_8 = \frac{1}{2}(\gamma_6 - \gamma_4)$  satisfying the relations

$$
\frac{\Gamma_4}{\Gamma_2} \sim \epsilon^{-2\Delta_4}, \quad \frac{\Gamma_6}{\Gamma_4} \sim \epsilon^{-2\Delta_6}, \quad \frac{\Gamma_8}{\Gamma_6} \sim \epsilon^{-2\Delta_8} \quad . \tag{6}
$$

As is known from scaling laws,<sup>4</sup> the gap exponents appearing in these relations take equal values. Agreement between theoretical and experimental values was satisfactorily confirmed for experimentally determined values of  $\Delta_4$ and  $\Delta_6$  ( $\Delta_4 \approx \Delta_6$ ). The lower value of exponent  $\Delta_8$  may be associated with the considerable error in the determination of exponent  $y_6$ , as may be seen in Fig. 1. The relations

[Eq. (5)] were obtained in a relatively wide range of temperatures, stretching from about  $1^\circ - 20^\circ$  above the Curie temperature. It has not yet been possible, however, to use this method to separate the coefficients of expansion [Eq. (1)] with satisfactory accuracy for the range of temperatures corresponding to values  $\epsilon \leq 10^{-2}$ . For  $\epsilon \leq 10^{-2}$ , very strong temperature dependences of the measured magnitudes are observed, and for this reason, very exact temperature stabilization is required (better than  $10^{-3} - 10^{-4}$  K). Moreover, in this temperature region very weak external magnetic fields should be applied. For T very close to  $T_c$ , the values of the external field for which only the first four terms in Eq. (1) are predominant may be comparable with the value of the magnetic field of the Earth.

The results of our measurements confirm the theoretical predictions (equality of the gap exponents proposed in Refs. 4 and 5). The values of the exponents of the functions  $x_0$ and  $x_2$  vs  $\epsilon$  presented in this paper are also in good agreement [without considering the factors  $T/T_c$  and  $(T/T_c)^3$ , respectively] with those reported in Refs. <sup>1</sup> and 6. However, it is particularly noteworthy that relations [Eq. (5)] hold over a surprisingly wide range of temperatures.

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