Features of the thermodynamic properties of dysprosium as a quasi-two-dimensional magnetic system

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The anomaly in the heat capacity of dysprosium in the vicinity of the Néel point has been investigated in specimens with $R_{300}/R_{4.2} = 82$ and 200. The magnetic component of the specific heat was approximated by a power-law function. It is shown that the critical behavior of the thermodynamic properties of dysprosium can not be explained either on the model of second-order phase transitions[1] or by the assumption of a first-order phase transition.[2] The observed features of the critical properties can be explained on the assumption that a large exchange-interaction anisotropy can lead to appreciable anisotropy of the magnetic moment fluctuations. In this case the thermodynamics of the system can be described within the framework of the two-dimensional degenerate XY model. The experimental results near the transition point are adequately described by a function corresponding to the Kosterlitz-Thouless transition.

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1. INTRODUCTION

The picture of a disordering phase transition in a number of magnetic systems with a multicomponent order parameter $n \geqslant 4$ is still not clear in spite of an appreciable amount of theoretical and experimental work on this problem. Dysprosium is one of the most studied systems with $n = 4$ and a "simple spiral" type of magnetic structure. The disordering in dysprosium is usually regarded as a second order phase transition in the Bak and Mukamel (BM) description.[1] It has recently been shown[2,3] that for certain values of the coefficients of the Hamiltonian describing the magnetic structure of dysprosium, a first-order phase transition can be realized. There are insufficient detailed data in the literature on the thermodynamic properties which are suitable for analyzing the form of the singularity and the nature of the transition. The existing results are ambiguous and this may be connected with imperfection of the specimens. In the present work measurements of the heat capacity of high-purity dysprosium have been carried out. The results obtained and also existing results on the electrical resistivity of dysprosium show that there are a number of features of the critical behavior: a different form of the singularity below and above the Néel point T_N; an anomalous increase in the critical exponent near the transition point; a sharp difference in the effect of impurities on the nature of the singularity below and above the transition point. The results obtained are not explained satisfactorily either within the framework of the BM model of a second-order phase transition, mentioned above, or from the point of view of a first-order transition. One of the possible reasons for this disagreement may be the formation of local magnetic-moment configurations of the soliton type, recently discussed theoretically, which can change appreciably the thermodynamics of the phase transition. For large exchange-interaction anisotropy such configurations can have a vortex structure.[4] The features of the critical behavior of dysprosium mentioned above can be explained by the assumption of a non-trivial structure to the magnetic-moment fluctuations near the transition point. This assumption is confirmed by the adequacy of the approximations of the experimental results near the transition point using the function obtained for the Kosterlitz-Thouless model.[5]

2. EXPERIMENTAL RESULTS

The heat capacity of two specimens, 1 and 2, with $R_{300}/R_{4.2} = 82$ and 180 to 200 respectively, was measured in a vacuum adiabatic microcalorimeter of volume 0.3 cm^3 (Ref. 6). The masses were 0.140 and 0.675 g. The amount of nongaseous impurity in the specimens was about 8×10^{-3} at.%. The total oxygen, nitrogen and carbon content was not more than 0.02 at.%. Specimen 1 was the same as that used for studying the electrical resistivity.[7] About 160 experimental values of $C_p(T)$ were obtained in the neighborhood of the phase transition temperature for specimen 1 and about 300 for specimen 2. The mean square deviation of the experimental values of $C_p(T)$ from the smoothed curve in the region of the transition was $\approx 0.6\%$ for specimen 1 and 0.1% for specimen 2. The magnetic components of the specific heat for both specimens were determined on the assumption that the regular part of $C_p(T)$ coincides with the heat capacity of nonmagnetic lutecium, which is very close to dysprosium in its lattice properties.

As was shown before,[7] the temperature dependence of the derivative of the resistivity dR/dT near the Néel point is asymmetrical for $T < T_N$ and $T > T_N$. When studying electrical resistivity, the question of the region where the magnetic component of the specific heat C_M/T is proportional to dR/dT always arises. Comparison of the experimental results for dR/dT and for C_M/T was made for specimen 1. The Néel temperatures determined both from the maxima of $C_p(T)$ and dR/dT agreed to within the limits of experimental error, so that the comparison was made at identical temperatures. The results are shown in Figs. 1 and 2. It can be seen that below T_N, in the region of reduced temperature

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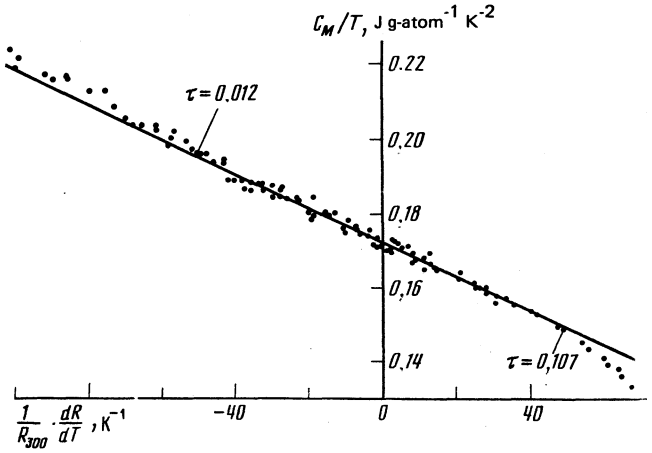

FIG. 1. Results of comparing values of $C_M(T)/T$ and $R_{300}^{-1}dR/dT$ measured at the same temperatures for specimen 1. The region $T<T_N$.

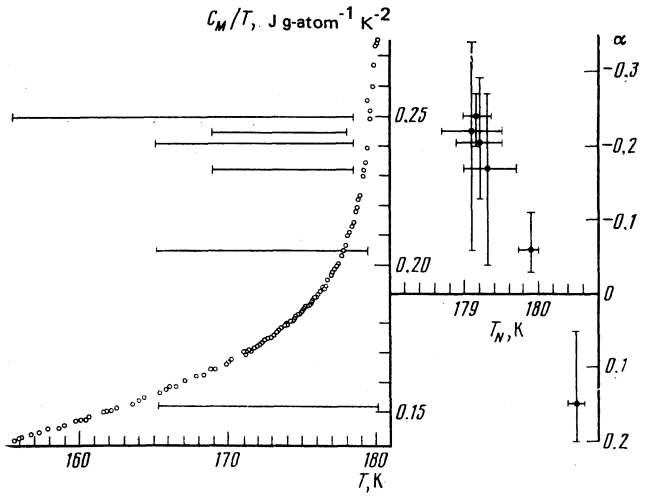

FIG. 3. Intervals of the approximations (on the left) and the critical exponents and transition temperatures corresponding to them (on the right) for specimen 2. The region $T<T_N$.

$0.012<\tau<0.107$, proportionality between C_M/T and dR/dT is observed. Above T_N, C_M/T and dR/dT are reliably proportional only for $\tau>0.044$. Below and above T_N there is appreciable departure from proportionality in the immediate neighborhood of T_N. Where C_M/T and dR/dT are not proportional one must give preference to the thermodynamic results in analyzing the singularity.

The values of $C_M(T)$ were approximated by the least squares method to the function

$$C_M/T=A+B\tau^{-\alpha}. \quad (1)$$

The agreement between the approximation and the experimental results and the reliability of the description were tested as before.[7] The results of looking for the intervals where the experimental results for specimen 2 are reliably described by Eq. (1) are illustrated in Fig. 3 for $T<T_N$ and in Fig. 4 for $T>T_N$. The experimental values of C_M/T are shown in the left-hand part of Fig. 3 and the temperature intervals for various analyses are shown by the lines. On the right are shown values of α and T_N obtained from the approximation and their confidence limits. Similar results are obtained for specimen 1, but the confidence limits for values of the critical exponent are wide because of the larger scatter in the experimental results. The most reliable values of the critical exponent for the maximum temperature range are shown in Table I. All the confidence limits in our treatment, shown in Table I and in the figures, correspond to a 95% confidence level.

The region $T<T_N$. The results of approximating the $C_M(T)$ results below the transition point show that a temperature range can be isolated for both specimens where the critical exponent is close to that determined from dR/dT (Ref. 7) and corresponds to $\alpha\approx-0.2$ (see Table I). We should note that although the values of the critical exponent with allowance for the confidence limits correspond to those predicted theoretically for a system with a four-component order parameter ($\alpha=-0.17$, Ref. 1), their modulus is somewhat greater in all cases. The exponent was calculated[1] taking account of two terms in the ε-expansion; taking the

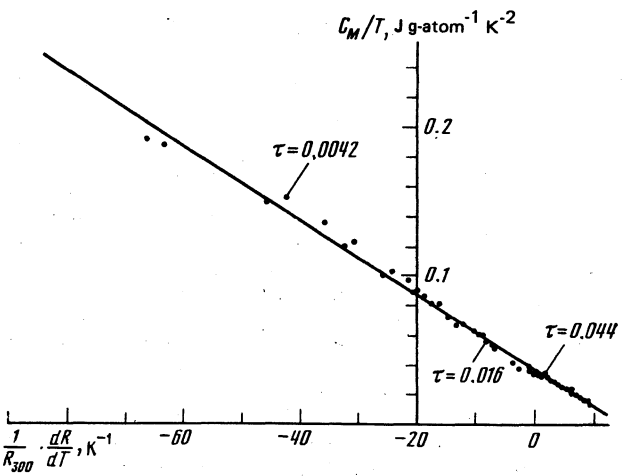

FIG. 2. Results of comparing values of $C_M(T)/T$ and $R_{300}^{-1}dR/dT$ measured at the same temperatures for specimen 1. The region $T>T_N$.

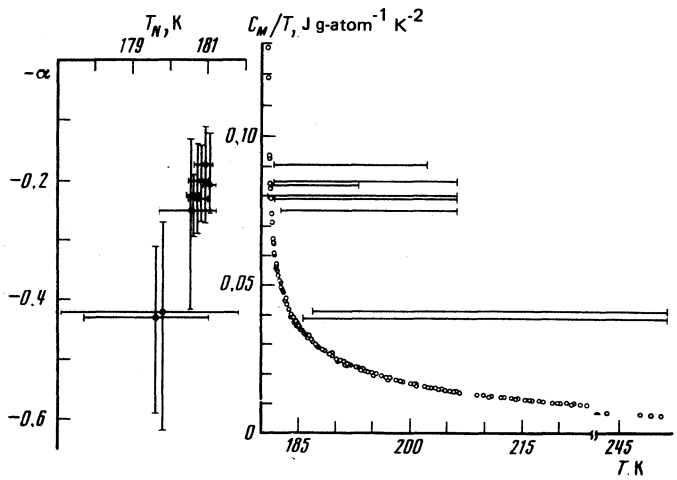

FIG. 4. Intervals of the approximation (on the right) and the critical exponents and transition temperatures corresponding to them (on the left) for specimen 2. The region $T>T_N$.

Table I,

	$\frac{R_{300}}{R_{4.2}}$	Interval	α	T_N, K
		$T<T_N$		
C_p	82	$0.005<\tau<0.092$	-0.25 ± 0.2	$180.0^{+1.2}_{-0.7}$
C_p	200	$0.0094<\tau<0,082$	$-0.205^{+0.07}_{-0.09}$	179.2 ± 0.3
$\frac{dR}{dT}$ [7]	82	$0.0038<\tau<0.055$	$-0.22^{+0.08}_{-0.06}$	$180.8^{+0.4}_{-0.3}$
		$T>T_N$		
C_p	200	$0.0081<\tau<0.12$	$0.17^{+0.1}_{-0.06}$	$180.9^{+0.15}_{-0.3}$
		$0.056<\tau<0.39$	0.49 ± 0.29	178 ± 5
C_p	82	$0.015<\tau<0.30$	$0.12^{+0.22}_{-0.12}$	$182.2^{+0.4}_{-1.0}$
C_p [8]	–	$0.012<\tau<0.11$	-0.15 ± 0.4	176.5 ± 0.7
		$0.083<\tau<0.438$	0.39 ± 0.3	175.0 ± 4.5
$\frac{dR}{dT}$ [7]	82	$0.015<\tau<0.15$	$0.33^{+0.11}_{-0.10}$	$180.5^{+0.5}_{-0.6}$
$\frac{dR}{dT}$ [7]	30	$0.021<\tau<0.053$	$-0.26^{+0.17}_{-0.12}$	$182.9^{+0.4}_{-0.6}$
		$0.034<\tau<0.23$	0.51 ± 0.9	179.1 ± 0.9

third term into account only increases the difference noted. Attempts to take account of non-asymptotic additions, by introducing a term $D\tau^{\Delta-\alpha}$ into Eq. (1), showed that just as in the analysis (Ref. 7) of dR/dT this term does not reduce the total square deviation.

The region $T>T_N$. On treating the $C_M(T)$ results for specimen 2 according to Eq. (1), two regions can be isolated where the approximation is reliable and adequate, as can be seen from Fig. 4. For $0.0081<\tau<0.12$ the parameter $\alpha=0.17^{+0.1}_{-0.06}$; in the range $0.056<\tau<0.39$, $\alpha=0.49\pm0.29$. It can be seen that the parameters α and T_N are determined considerably more crudely for large values of τ than for small τ, owing to the decrease of C_M far from the transition. The boundary of the transition from one type of behavior to the other is very smeared out, so that the intervals given overlap. Comparison with the results of analyzing dR/dT for specimens with $R_{300}/R_{4.2}=82$ and 101 shows that the revealed[7] tendencies of the exponent to change agree with the results of the detailed analysis of $C_M(T)$ of specimens 1 and 2. In any case, it seems to us rather clear that there is no scaling behavior such as occurs for $T<T_N$. Besides our results, the results on the heat capacity[8] of a dysprosium specimen containing 0.8 at.% impurity has been analyzed. The results of a statistical treatment of these data differ appreciably from those obtained on specimens 1 and 2 for $T>T_N$. Although the results of Ref. 8 are not sufficiently detailed for a complete check on the reliability of the approximation (only 40 experimental points in the region studied) and the confidence intervals $\Delta\alpha$ are large, nevertheless there is a temperature interval over which the exponent obtained corresponds to the BM model.

For comparison, the results[9] of analysing dR/dT for a specimen[7] with $R_{300}/R_{4.2}=30$ are shown in Fig. 5, as well as the results described above on heat capacity. It can be seen that for $T>T_N$ the critical behavior of the thermodynamic functions for the purer specimens 1 and 2 and specimens with larger impurity content[8,9] are fundamentally different. It can also be seen from Fig. 5 that if we include in the analysis of $C_M(T)$ according to Eq. (1) those experimental values for which $|\tau|<0.008$, then the value of the critical exponent becomes $\alpha\gtrsim1$. In this case the description becomes unreliable and inadequate, i.e., the experimental results are not described by a function of the type of Eq. (1) in the immediate vicinity of T_N. A possible reason for the distortion in the function could be the influence of the compressibility at temperatures close to the transition point. Compressibility was taken into account,[10] but it appears that the function there does not give a reliable and adequate approximation to the experimental results in the region close to T_N. It seems that

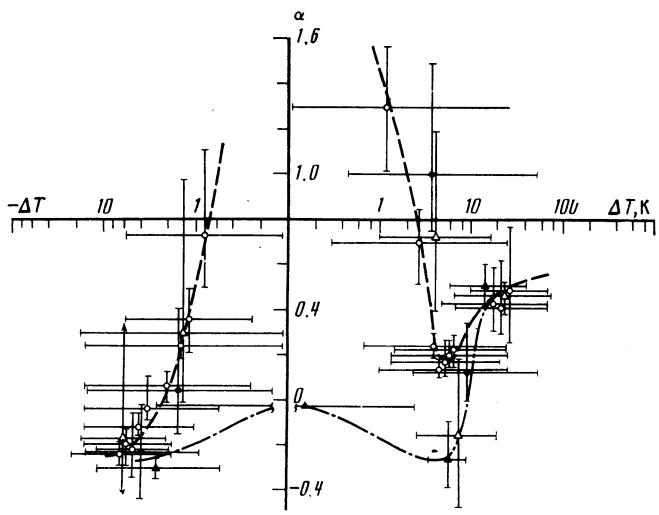

FIG. 5. Comparison of the results of the approximation: ●—specimen 1, $R_{300}/R_{4.2}=82$; ○—specimen 2, $R_{300}/R_{4.2}=200$; △—analysis of the results of Ref. 8; ▲—data[7] for a specimen with $R_{300}/R_{4.2}=30$ analyzed in Ref. 9.

compressibility is not important in the present case and there must be other mechanisms that distort the nature of the singularity near T_N.

3. DISCUSSION OF THE RESULTS

The results obtained for the approximation of the experimental results show that although there are systematic departures from the calculated value of α, the results below T_N do not contradict the BM model of a second-order phase transition. At the same time, above T_N there is no exponent corresponding to the BM model. The anomalous growth in the critical exponent for $|\tau| < 0.008$ also indicates the inapplicability of the BM model in the immediate vicinity of T_N. This increase of the exponent can be regarded as a result of the effect of a smeared-out first-order phase transition δ-function in dysprosium, proposed by Barak and Walker.[2] According to them, the Hamiltonian has the form

$$-H=\frac{1}{2}\sum_i [r(\eta_i^2+\bar{\eta}_i^2)+(\nabla\eta_i)^2+(\nabla\eta_i)^2]$$

$$+U_1\Big[\sum_i(\eta_i^2+\bar{\eta}_i^2)\Big]^2+4U_2(\eta_i^2+\bar{\eta}_i^2)(\eta_2^2+\bar{\eta}_2^2),$$

where η_i and $\bar{\eta}_i$ are the components of the magnetic moment of the spiral structure with right and left rotation.[2] The disordering process at $U_2<0$ proceeds as a second-order phase transition,[1] while the case of $U_2>0$ corresponds to a first-order transition. We see from a parametric diagram of a renormalization-group analysis in U_1, U_2 coordinates[2] that stationary points located on the $U_2=0$ line can influence the critical properties of the system in the case of a first-order transition. A non-Gaussian point for which $\alpha=-0.02$ and a Gaussian point with $\alpha=0.5$ are situated on this line.[3] A certain intermediate exponent $\alpha=0.2$, observed for specimen 2 at $T>T_N$, can be attributed to the close positioning of these two points to the trajectories on the parametric diagram. There is, however, no satisfactory explanation for the exponent $\alpha=-0.2$ observed for the $T<T_N$ branch. The existence of a first-order transition in Dy, Tb and Ho has not so far been demonstrated by direct experimental results. There is practically no experimental observation of a jump in volume and magnetic moment. A special study of magnetic susceptibility carried out on specimen 1 showed that there is no hysteresis of the transition temperature, to an accuracy of 0.1 K. The question of the influence of impurities on the critical behavior is also unclear. The interval over which the properties are distorted by impurities both for a second-order transition and for a first-order δ-function transition should broaden with increasing impurity density, but this is not observed experimentally. It is also unclear how the presence of $\approx 0.1\%$ impurity in a specimen is sufficient to change completely the critical behavior for $T>T_N$, while the impurities have no noticeable effect in the region $T<T_N$. We are thus unable to obtain a satisfactory description of the experimental results either within the framework of a model of a first-order phase transition, or from the point of view of a second-order transition.

We shall now consider what in our view is yet another possible mechanism for a transition to the ordered state in the magnetic structures studied. It is known that the exchange constants in rare-earth metals are related in the following way: $J_0>J_1>J_2$,[11] where J_0 is the constant characterizing exchange in the plane, while J_1 and J_2 characterize exchange between nearest and next nearest planes along the helicoid axis. From this we may suppose that the correlation radius r_0 in the plane can be greater than that along the hexagonal axis, r_1. If the interplanar interaction were appreciable (the correlation radii were close, $r\approx r_1$), the phase transition would be described within the framework of the theory discussed above. But if the interplanar interaction is small and $r_0 \gg r_1$, then near the transition and for $T>T_N$ a situation evidently occurs which one might try to describe within the framework of a quasi-degenerate XY model.[4,12,13] A detailed consideration of the configuration of the magnetic moments by Kosterlitz and Thouless leads to the conclusion of the existence in the plane of vortex "molecules" consisting of two vortices with opposite senses of rotation.[4] We shall now estimate whether the existence of such a vortex structure is energetically possible in the case of dysprosium. The increase in the energy of interaction with neighboring planes when vortices arise can be written as[14]

$$\Delta E_1=I_1\varphi^2R^2,$$

where φ^2 is the square of the spontaneous moment and R is the vortex dimension in interatomic distances. We compare this quantity with the energy of a vortex, which can be written as[14]

$$\Delta E_2=\pi J_0\, ln\,(R/a),$$

where a is the interatomic distance in the plane. Using results on the exchange constants[11] $J_0/J_1=5$ to 11 and the fact that $\varphi\sim\tau^\beta$, where $\beta=0.39$,[1] we can obtain the ratio $\Delta E_1/\Delta E_2\approx 0.06$ at a distance of ~ 2 K from the transition, i.e., for $\tau\approx 0.01$, taking $R=5a$.[14] This approximate calculation shows that a vortex structure is energetically possible for dysprosium. Then according to Kosterlitz[5] the magnetic part of the heat capacity near the temperature where the vortex molecules are destroyed should be represented by the function

$$C_M/T=A+Bt^{-2}\exp(-b/t^{1/2}), \qquad (2)$$

where $t=|T-T_0|/T_0$ and T_0 is the Kosterlitz-Thouless transition temperature. The maximum in the experimental $C_M(T)$ dependence should then correspond to the maximum of the function in Eq. (2). Approximating the experimental results by Eq. (2) showed that there is a reliable and adequate description of the C_M/T dependence in the temperature range from 178.5 to 180.7 K. In this range there are 23 experimental heat-capacity values. From this approximation the parameters obtained are $T=180.8$ K and $b=0.22$. According to calculation,[5] $b=1.5$. The two-dimensional Kosterlitz-Thouless model is evidently a very crude approximation for describing the complicated quasi-two-dimensional structure of dysprosium. It is thus difficult to expect full agreement between the calculated parameters and those obtained by experiment. The maximum in $C_M(T)$ occurs at

180.22 K. We should note that in the above interval the value of $C_M(T)$ decreases by 20% for $T < T_{max}$ and by 50% for $T > T_{max}$ from the value of $C_M(T_{max})$. The adequate description of such a rapidly changing function by a relatively simple expression with a mean square deviation $\approx 0.3\%$ can not, in our view, be accidental.

The experimentally observed features of the critical behavior of dysprosium below and above the phase-transition point can be explained with the help of the quasi-two-dimensional degenerate XY model. The dissociation of vortex pairs is essentially complete at temperatures above T_0 and spin-wave excitations start to contribute appreciably to the heat capacity. As shown by calculation,[15,16] the exponential dependence of Eq. (2) then goes over to a power law. Unfortunately, the value of the corresponding power was not given there. It is possible that the exponent $\alpha \approx 0.2$ observed experimentally above T_N corresponds to this transition region. At higher temperatures the observed exponent, within the confidence limits, corresponds to the molecular-field approximation, which agrees with calculations.[13] The increase in the crystal-field anisotropy at temperatures below the Kosterlitz-Thouless region leads[17] to the destruction of the vortex structure and to the appearance of a spontaneous magnetic moment. The interplanar interaction increases with the growth of the spontaneous moment, the system becomes three-dimensional, and a helicoidal structure is established in it. This shows up in an experiment by the appearance below T_N of an exponent corresponding to the BM three-dimensional model.[1]

The fact that the Néel points obtained from the approximation of Eq. (1) lie about 1 K below, at $T < T_N$, and 1 K above, at $T > T_N$, the temperature of the maximum heat capacity could serve as indirect confirmation of the suggestion made above. For a first-order phase transition the approximated T_N usually lies above the point of maximum $C_M(T)$ for the left-hand branch and below it for the right-hand branch. In addition, on varying the range of approximating the experimental results by the Kosterlitz-Thouless equation,[2] it turned out that the limits of reliability of this approximation agree roughly with the limits of reliability of the power-law function described (Eq. (1)), which are determined independently. The experimental results obtained on the pure specimen 2 are thus satisfactorily explained within the Kosterlitz-Thouless model.

We shall now consider the distinguishing features of results obtained on specimens with larger impurity content.[8,9] The formation of vortices is evidently hampered by the presence of impurities in the specimen. In fact, for impurity densities 10^{-1} to 1 at%, corresponding to the specimens used,[9,8] the mean distance between impurities is comparable with the vortex dimensions. The limitation on the growth of the correlation radius in the plane, due to the presence of impurity, assists the magnetic structure in becoming three-dimensional. It is probable that an exponent close to that obtained on the BM model is thus observed above T_N for these specimens.[8,9] The results agree with results of a neutron diffraction study[18] on polycrystalline dysprosium, probably containing an appreciable amount of impurity, where helicoidal correlation was observed above the Néel point. On the other hand, an anomalous growth of critical exponent in the immediate neighborhood of the heat-capacity maximum indicates a change in the nature of the singularity in this region (see Fig. 5). We think that for pure specimens this change is connected with the formation of a vortex structure. It is possible that a helix and helicoidal formation coexist and the relation between them can be determined by the impurity content. Indications of a vortex magnetic structure in specimens with larger amounts of impurity[8,9] appear therefore only in the immediate vicinity of the transition point. Since a vortex structure is absent below the transition point, the sensitivity of the critical behavior to the amount of impurity is appreciably less in this case.

The influence of impurities in the case of a quasi-two-dimensional system manifests itself in a wider temperature interval than usual. While in the pure specimens 1 and 2 a function connected with the destruction of the vortex structure is observed over an interval of ≈ 10 K, the presence of impurities changes the nature of the $C_M(T)$ relation over the whole of this interval. For a normal three-dimensional system the effect of impurities shows up[7] in the much narrower temperature interval at a distance of ≈ 0 to 1 K from the transition point.

The Kosterlitz-Thouless model thus describes the heat-capacity behavior in the immediate vicinity of the transition point better than other formulations considered above, explains the whole collection of experimental results on the critical behavior of dysprosium, the asymmetry of the observed critical exponents and the difference in sensitivity of the singularity below and above T_N to the amount of impurity in the specimen. We note, however, that the correspondence between the system considered and the two-dimensional Kosterlitz-Thouless model depends on the ratio of the exchange constants for intraplane and interplane interaction. Unfortunately there is no quantitative description in the literature of the behavior of a quasi-two-dimensional system as a function of the magnitude of the parameter which characterizes the degree of degeneracy of the system. Taking account of the quasi-two-dimensionality can lead to the formation of nonlinear configurations of the soliton type, which are more complicated than plane vortices. The possibility of their arising in ferro- and antiferromagnets is discussed by, for example, Ivanov and Kosevich.[19] However, the thermodynamics of such systems is not yet developed. It is possible that it may be very close in the nature of the singularity to that obtained by Kosterlitz and Thouless.

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