Critical thermal expansion of yttrium iron garnet

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A capacitance dilatometer (precision 1%, sensitivity 10^{-8}) was used in measurements of the thermal expansion coefficient α near the Curie point T_c of an yttrium iron garnet crystal. The influence of magnetic fields H (up to 3 kOe) on the temperature dependence of α was studied. The experimental data on $\alpha(H, T)$ were used to show that α can be described by static scaling. The relevant critical indices were calculated and a scaling function, which could be used to describe $\alpha(H, T)$ by a single curve, was constructed.

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1. INTRODUCTION

A phenomenological theory of second-order phase transitions can give a number of general relationships between the anomalies of various thermodynamic quantities in the vicinity of the temperature T_c of a transition of a magnetically ordered crystal to a paramagnetic state.[1,2] In particular, the magnetic component of the specific heat C_m and the isobaric thermal coefficient α were shown by Pippard[3] (see also Ref. 4) to be related by

$$C_m = T_c(dP_c/dT)V\alpha. \quad (1)$$

Here, $P = P_c(T)$ is the equation for the phase transition points in the (P, T) plane. It follows from Eq. (1) that the thermal expansion coefficient should become infinite on approach to $T \to T_c$ and it should obey the same law as the specific heat. The linear relationship between C_m and α follows also from microscopic concepts based on the Heisenberg model in which the relationship between the magnetic and elastic degrees of freedom is introduced via the bulk dependence of the exchange integral I. Callen and Callen[5] showed that near the Curie temperature

$$C_m = N[I_1 z_1(\partial\langle SS'\rangle_1/\partial T)_V + I_2 z_2(\partial\langle SS'\rangle_2/\partial T)_V + \ldots] \quad (2)$$

and

$$\alpha_{me} = (kN/3V)[z_1 I_1\gamma_1(\partial\langle SS'\rangle_1/\partial T)_V + z_2 I_2\gamma_2(\partial\langle SS'\rangle_2/\partial T)_V + \ldots]. \quad (3)$$

Here, α_{me} is the magnetoelastic contribution to thermal expansion; k is the isothermal compressibility; z_n is the number of the nearest neighbors; $\gamma_n = \partial(\ln I/\partial(\ln V)$ is the magnetic Grüneisen constant; N is the Avogadro number. In the case of magnetically ordered crystals in which the exchange interaction between the nearest neighbors predominates ($z_1 I_1 \gg z_2 I_2$) we find from Eqs. (2) and (3) that C_m and α_{me} are related linearly. It has been shown experimentally that Eqs. (1)–(3) are well satisfied by magnetic insulators EuO (Ref. 6) and $Y_3Fe_5O_{12}$ (Ref. 7).

We can thus see that the proportionality between C_m and α, which follows from the thermodynamic and model representations, and which has been demonstrated experimentally, allows us to describe the thermal expansion coefficient in the critical region using fluctuation theory of phase transitions and applying the ideas of static scaling to the specific heat when analyzing the experimental data for α (Refs. 8 and 9).

2. EXPERIMENTAL RESULTS AND DISCUSSION

The ability to describe the critical behavior of α within the static scaling framework can be illustrated by considering the example of yttrium ion garnet the substance most thoroughly investigated by us and for which experimental data are available on the specific heat, magnetization, and elastic wave velocity all measured in the same crystal.[10] Moreover, in the case of $Y_3Fe_5O_{12}$ the isotropic $Fe_a^{3+}-O^{2-}-Fe_d^{3+}$ exchange interaction predominates and, therefore, we can ignore the second terms in Eqs. (2) and (3). Then, the relationship between C_m and α is indeed linear:

$$\alpha_{mv} = k\gamma C_m/3V, \quad (4)$$

and the magnetic Grüneisen constant found from direct measurements is $\gamma = 3.13$ and it is indeed independent of temperature.

We measured the thermal expansion coefficient with a capacitance dilatometer with an error not exceeding 1% and a sensitivity of 10^{-8} in relative measurements. The values of α were obtained in a wide temperature range (100–700°K) under static and dynamic thermal conditions. Under static conditions the temperature was stabilized to within 0.005°K and measurements were made at 0.1°K. The rate of change of temperature under dynamic conditions was kept constant at 1°K/h.

The experimental curve shown in Fig. 1 represents repeated measurements of the temperature dependence of α in the vicinity of T_c of $Y_cFe_5O_{12}$. In the critical

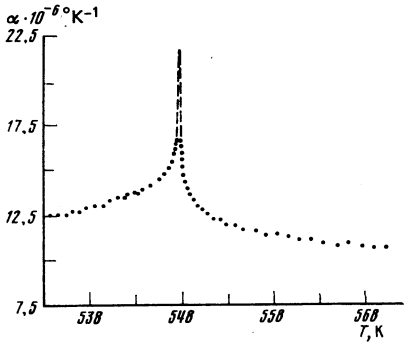

FIG. 1. Temperature dependence of the thermal expansion coefficient α of $Y_3Fe_5O_{12}$ along the [100] axis near a phase transition at $T_c = 548.25$°K. The points are the experimental values and the dashed curve is calculated.

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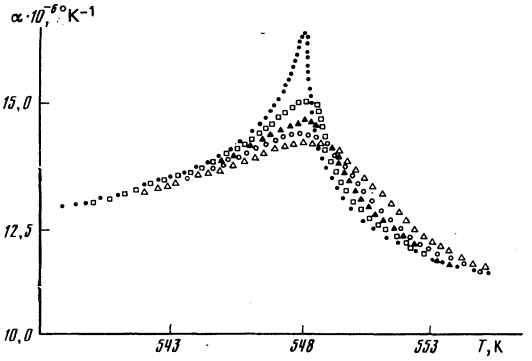

FIG. 2. Temperature dependences of α for $Y_3Fe_5O_{12}$ along the [100] direction in the critical region, recorded in various magnetic fields (Oe): •) 0; □) 300; ▲) 1140; ○) 2500; △) 3000.

region there is a characteristic λ anomaly and a peak value of α occurs at a temperature coinciding with T_c deduced from the specific heat and spontaneous magnetization data. The experimental values of α near T_c can be approximated by a power law

$$\alpha^{\pm}=\frac{A^{\pm}}{a^{\pm}}(\tau^{-a^{\pm}}-1)+E^{\pm}\tau+B^{\pm}. \qquad (5)$$

Here, $\tau=(T-T_c)/T_c$ and the "+" and "−" signs refer to $T>T_c$ and $T<T_c$, respectively. The parameters in Eq. (5) were selected in such a way that the rms deviation of the experimental points from the dependence (5) did not exceed 1%. The critical indices a^+ and a^- depended largely on the selection of T_c and, therefore, we used the value of T_c deduced by minimizing the total rms deviation $\Delta^2=(\Delta^+)^2+(\Delta^-)^2$, i.e., we utilized the equality $T_c^+=T_c^-$. All the calculations were carried out by the least-squares method on a BÉSM-4M computer. The experimental points satisfied the fitting function with the following parameters:

$$a^+=-0.10\pm0.02,\quad A^+=1.84\cdot10^{-6}\ °K^{-1},\quad B^+=7.7\cdot10^{-6}\ °K^{-1},$$
$$E^+=6.0\cdot10^{-6},\quad T_c^+=T_c^-=548.25\ K,$$
$$a^-=-0.12\pm0.02,\quad A^-=1.67\cdot10^{-6}\ °K^{-1},\quad B^-=6.0\cdot10^{-6}\ °K^{-1},$$
$$E^-=5.0\cdot10^{-6},\quad 4\cdot10^{-4}\leqslant\tau\leqslant6\cdot10^{-2}.$$

The curve $\alpha=f(T)$ calculated from these data is shown dashed in Fig. 1. The deviation of the experimental points from the calculated curve occurs in the temperature interval $T-T_c\leqslant\pm0.2°K$ indicating a high quality of the investigated crystal. The experimental data for α showed that $Y_3FE_5O_{12}$ can be described satisfactorily by the three-dimensional Heisenberg model for which the ε expansion predicts $a=-0.1$ (Ref. 12). The calculated values of a satisfy the scaling relationships

$$a^+=a^-,\quad 2-a=\beta(\delta+1) \qquad (6)$$

($\beta=0.365$ and $\delta=4.6$ are the critical indices of the spontaneous magnetization and of the critical isotherm).

The validity of the above relationships shows that it should be possible to plot the magnetic equation of state which describes, in terms of reduced coordinates, the dependences of α on τ and H. Applying the ideas developed for the specific heat[9] and using Eqs. (1)–(5), we find that the magnetic equation of state describing $\alpha(H,T)$ by a single curve can be represented in the

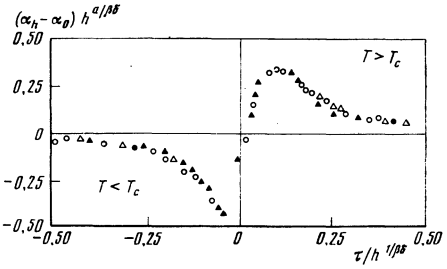

FIG. 3. Magnetic equation of state for α in the critical region of $Y_3Fe_5O_{12}$: △) $H=100$ Oe; •) $H=300$ Oe; ○) $H=1140$ Oe; ▲) $H=3000$ Oe.

form

$$[\alpha_h(\tau,h)-\alpha_h(\tau,0)]h^{a/\beta\delta}=g(\tau h^{-1/\beta\delta}), \qquad (7)$$

where g is some undetermined function; $h=\mu_0H/kT_c$ is the reduced magnetic field; μ_0 is the spontaneous magnetization at 0°K; k is the Boltzmann constant; $\alpha_h(\tau,h)$ and $\alpha_h(\tau,0)$ are the normalized thermal expansion coefficients in the presence of a magnetic field and in $H=0$, the normalization being made to a value of α calculated using the Debye function $D(\Theta/T=1)$.

The experimental data on the influence of the field H on α near T_c of $Y_3Fe_5O_{12}$, needed to construct the equation of state (6), are presented in Fig. 2. A characteristic feature of the influence of H is a reduction of α below T_c and an increase above T_c, as the magnetic field is increased. The specific heat behaves similarly.[10] The change of α in a magnetic field, like the change in C_m, is due to suppression of critical fluctuations and a corresponding change in the curvature of the temperature dependence of the magnetization. It should be pointed out that below T_c in fields $H<100$ Oe there is an anomalous change in α, which is clearly related to the kink (shape) effect and, therefore, these data were not used in plotting the magnetic equation of state in Fig. 3. We can see from Fig. 3 that the experimental data for the thermal expansion coefficient in magnetic fields (Fig. 2) can be described by a single curve in terms of the reduced coordinates $\Delta\alpha_h h^{\alpha/\beta\delta}$ and $\tau/h^{-1/\beta\delta}$.

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Magnetic "order-order" and "order-alien disorder" phase transitions

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For an isotropic magnetic material with non-Heisenberg exchange, the possibility is demonstrated of a new type of "order-disorder" phase transition, in which the short-range order above the transition point is of a different type from the long-range order below the transition point (an "order-alien disorder" phase transition). Thus the paramagnetic Curie temperature may be positive in an isotropic antiferromagnet and negative in a ferromagnet. The reason for this is the different temperature dependence of the Heisenberg and of the non-Heisenberg exchange, as a result of which the high-temperature properties may be determined by the former and the low-temperature properties by the latter. For the same reason, phase transitions of the "ferromagnet-antiferromagnet" type are possible; and such a purely magnetic mechanism of "order-order" phase transitions is in many cases much more realistic than the Kittel exchange inversion. In particular, this mechanism permits the occurrence of a whole series of phase transitions between commensurable collinear structures (from a two-sublattice antiferromagnet to a three-sublattice ferrimagnet to a four-sublattice antiferromagnet to a paramagnet), observed in EuSe. By analysis of the experimental data for EuSe it is shown also that the last of these transitions is of the "order-alien disorder" type. If a ferromagnetic state is possible, then three-spin exchange enhances the singularity of the susceptibility at T_C as compared with a Heisenberg magnet.

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INTRODUCTION

It is usually considered self-evident that upon heating, an isotropic antiferromagnet (AF) transforms to a paramagnetic (PM) state with a negative PM Curie temperature Θ, and a ferromagnet (FM) to a state with a positive Θ. The sign of Θ is the same as that of the binary spin correlators of neighboring atoms, which describe the short-range magnetic order. Therefore a negative sign means that after breakdown of the long-range AF order in the crystal, there remains in it short-range AF order; a positive Θ means that the short-range order is ferromagnetic. Similarly in other cases also in which we have to do with order-disorder phase transitions (PT), it is supposed that the short-range order above the transition point is of the same type as the long-range order below it.

One of the basic results of our paper is the demonstration of the existence of a new type of order-disorder phase transition, for which the short-range order above the transition point is of a different type from the long-range order below it. For example, if such a transition occurs in an isotropic antiferromagnet, its PM Curie temperature is not negative but positive[1)]; if in a ferromagnet, then Θ is negative. Such order-strange disorder phase transitions are necessarily of first order, whereas PT with retention of the type of short-range order (order-ordinary disorder) may be either of first or of second order.

The question of the nature of the short-range order above the point where the long-range order disappears is very important, since many properties of crystals are determined not by the long- but by the short-range order. One example has already been given: the PM susceptibility of strong magnets. This includes also the electrical and optical properties. In particular, the position of the optical absorption edge in magnetic semiconductors and insulators depends very strongly on the short-range order; and this fact is used by us below, together with other experimental data, to establish the occurrence of FM short-range order after disappearance of long-range AF order in EuSe. Thus the paper presents not only a theoretical but also an experimental demonstration of the existence of an order-alien disorder phase transition.

The theoretical demonstration of the existence of an order-alien disorder PT is given for a model of a magnet with isotropic many-spin exchange.[2)] What has been investigated so far in the literature is the effect on an order-disorder phase transition of biquadratic exchange[2,3] $[\sim(\mathbf{S}_1\mathbf{S}_2)^2]$ and of four-spin exchange[4] $[\sim(\mathbf{S}_1\mathbf{S}_2)\times(\mathbf{S}_3\mathbf{S}_4)]$. In these papers it was established that addition of such terms to the Heisenberg Hamiltonian may produce a discontinuous phase transition from the FM state to the PM. But as will be clear from what follows, a Hamiltonian with biquadratic exchange[2,3] permits only an order-ordinary disorder PT. A four-spin Hamiltonian, for a certain choice of its parameters, allows one to obtain an order-alien disorder PT, but this possibility went unnoticed earlier.[4]

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