

**Critical thermal diffusivity in anisotropic magnets**

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(Received 3 February 2003; revised manuscript received 24 June 2003; published 17 September 2003)

We derive nonasymptotic expressions for the frequency- and temperature-dependent thermal diffusivity near a critical point in an Ising-type magnet. We consider a dynamic model which takes also into account the longitudinal sound mode. The asymptotic scaling function of the thermal diffusion constant at long wavelengths is given within the renormalization group formalism in one-loop order. A relation connecting longitudinal sound velocity and thermal diffusivity in a broad range of frequency and reduced temperature is obtained.

DOI: 10.1103/PhysRevB.68.094416

PACS number(s): 05.70.Jk, 75.40.Gb, 62.65.+k, 64.60.Ht

**I. INTRODUCTION**

The critical phenomena associated with heat transport at phase transitions have been extensively studied using the renormalization group method.<sup>1</sup> In the case of anisotropic magnets (with conserved energy density), the critical dynamics can be explained in terms of model C (Ref. 2) and the thermal conductivity  $\kappa$  has been found there to be finite at the critical point. This does not mean that there is no singularity in the heat transport at the critical temperature  $T_C$ . In the zero-frequency limit, the thermal diffusivity  $D_T = \kappa/C_p$ , where  $C_p$  is the constant-pressure specific heat per unit volume, converges to zero because the specific heat critical exponent is positive for Ising-like systems. Recently a sharp dip in  $D_T$ , due to a critical slowing down, was found experimentally in a uniaxial antiferromagnet  $\text{FeF}_2$  (Ref. 3) and also in  $\text{Cr}_2\text{O}_3$  (Ref. 4) (the uniaxial behavior was obtained in this weakly anisotropic antiferromagnet very close to  $T_C$ ) for very low frequencies. However, the detailed nature of this singularity as well as the possible frequency dependence of  $D_T$  is still unclear from both experimental and theoretical points of view.

In this paper we follow the phenomenological-hydrodynamical approach to critical dynamics.<sup>1</sup> In such an approach the critical effects in the transport properties near a continuous phase transition are usually described by a set of stochastic equations neglecting the sound mode. Such an approximation is not generally true for nonasymptotic behavior, so here we study not only the effects of critical fluctuations in the spin system on the thermal diffusivity but also include the effect of a sound mode coupled to both spin and energy fluctuations. The dynamic model considered here is an extension of the model C.<sup>1</sup> A general expression for  $D_T$  valid also at finite frequencies is obtained within the dynamic renormalization group approach in one-loop order for  $T \geq T_C$ . We show that additional terms are present in the thermal diffusivity due to the sound mode. Subsequently we discuss two limits of this expression corresponding to isothermal and adiabatic propagation of the related sound mode, respectively. An interesting relation connecting the “low-frequency” and “high-frequency” diffusivities, as well as the isothermal and adiabatic sound velocities, is obtained which is a generalization of the static relation (known for fluids) between the ratio of the adiabatic and isothermal compress-

ibilities and the ratio of specific heats at a constant pressure and volume, respectively.

This paper is organized as follows. In Sec. II our model and method are explained. In Sec. III the temperature- and frequency-dependent thermal diffusivity is derived. Two limiting cases are discussed and a relation between them is obtained. The summary and discussion are given in Sec. IV.

**II. MODEL**

To study the hydrodynamics of a fluid we need only to consider densities of conserved quantities which decay slowly on long-length scales. There are five such conserved densities for three-dimensional pure fluid: momentum density (three components), mass density, and energy density. In a solid, there are three additional broken-symmetry hydrodynamic variables  $u_i$ . The number of independent degrees of freedom determines the number of modes,<sup>5</sup> so eight modes are expected in monomolecular nonmagnetic crystals. These are two longitudinal sound modes (propagating modes always occur in pairs), composed essentially of longitudinal momentum and longitudinal displacement, and two pairs of transverse sound modes associated with transverse components of displacement and momentum. In addition there is one diffusive mode for thermal conductivity and one corresponding to the diffusion of vacancies.<sup>5,6</sup> The vacancy diffusion is very slow, so it is often ignored as we did in our analysis. In Ising-like magnets near a critical point we must add to this set the slow relaxational mode of the order parameter. In general, there are eight modes for an Ising-like magnetic crystal with no vacancies. However, it is known that in magnets the dominant interaction between the sound and spin fluctuations is the volume magnetostriction.<sup>7</sup> In an isotropic solid, which is considered here for simplicity, the transverse sound decouples from the other modes and will be neglected. Thus we need only to consider a model with four degrees of freedom in which the longitudinal sound, energy, and order-parameter modes are mutually coupled to each other.

**A. Statics**

We start from a Hamiltonian describing an Ising-type magnet on a  $d$ -dimensional elastic solid:<sup>8</sup>

$$H = H_{\text{OP}} + H_{\text{el}} + H_{\sigma} + H_{\text{int}}, \quad (1)$$

where  $H_{\text{OP}}(\Phi)$  is the Ginzburg-Landau part for the one-component order parameter  $\Phi(\mathbf{x})$ ,

$$H_{\text{OP}} = \frac{1}{2} \int d^d x \left[ r \Phi(\mathbf{x})^2 + (\nabla \Phi)^2 + \frac{\lambda}{2} \Phi(\mathbf{x})^4 \right], \quad (2)$$

and

$$H_{\text{el}} = \frac{1}{2} \int d^d x \left\{ B u_{ii}(\mathbf{x})^2 + 2\mu \left[ u_{ij}(\mathbf{x}) - \frac{1}{d} \delta_{ij} u_{ll}(\mathbf{x}) \right]^2 \right\} \quad (3)$$

is the elastic contribution in the harmonic approximation, with  $u_{ij}(\mathbf{x})$  denoting the strain tensor related to the displacement vector components  $u_i(\mathbf{x})$  by

$$u_{ij}(\mathbf{x}) = \frac{1}{2} (\nabla_i u_j + \nabla_j u_i).$$

The first term in Eq. (3) describes the contribution due to the volume changes [in the absence of vacancies  $u_{ii}(\mathbf{x})$  is proportional to the density fluctuation] and the second due to the shear distortions.<sup>6</sup>  $B$  and  $\mu$  are the (bare) bulk and shear modulus (we have assumed  $k_B T = 1$ ), respectively. For simplicity, we have assumed the solid to be isotropic. The Hamiltonian

$$H_\sigma = \frac{1}{2C_V^0} \int d^d x [\sigma(\mathbf{x})^2] \quad (4)$$

describes the entropy (per mass) fluctuations  $\sigma(\mathbf{x})$  (being the linear combination of the energy and density fluctuations<sup>6</sup>) with  $C_V^0$  being proportional to the specific heat at a constant volume. Finally, the interaction Hamiltonian is given by

$$H_{\text{int}} = \int d^d x [g u_{ii}(\mathbf{x}) \Phi(\mathbf{x})^2 + w u_{ii}(\mathbf{x}) \sigma(\mathbf{x}) + f \sigma(\mathbf{x}) \Phi(\mathbf{x})^2], \quad (5)$$

where the first term describes the volume magnetostriction with the bare coupling constant  $g$ . We have taken also into consideration the entropo-elastic interaction with the coupling constant  $w$ . This interaction is responsible for the fact that the adiabatic longitudinal sound velocity differs from the isothermal one also for nonmagnetic systems. The last term in Eq. (5) is responsible for the critical behavior of the specific heat.<sup>2</sup>

In the first step a given elastic configuration  $u_{ij}(\mathbf{x})$  can be separated<sup>9,10</sup> into a homogenous deformation and the constant-volume phonon part:

$$u_{ij}(\mathbf{x}) = u_{ij}^0 + \frac{1}{2V} \sum_{\mathbf{k} \neq 0} i [k_i u_j(\mathbf{k}) + k_j u_i(\mathbf{k})] \exp(i\mathbf{k} \cdot \mathbf{x}), \quad (6)$$

where  $V$  is the volume of the system at equilibrium. We decompose the displacement vector  $\mathbf{u}(\mathbf{k})$  into longitudinal and transverse parts defined via

$$\mathbf{u}(\mathbf{k}) = \hat{\mathbf{k}} u_L(\mathbf{k}) + \mathbf{u}_T(\mathbf{k}), \quad (7)$$

where  $u_L(\mathbf{k}) = \hat{\mathbf{k}} \cdot \mathbf{u}(\mathbf{k})$ ,  $\mathbf{u}_T(\mathbf{k}) = \mathbf{u}(\mathbf{k}) - \hat{\mathbf{k}} u_L(\mathbf{k})$ , and  $\hat{\mathbf{k}} = \mathbf{k}/|\mathbf{k}|$ . Only the longitudinal part  $u_L$  is coupled, in our model, to the order parameter fluctuations, so we integrate over the transverse modes as well as over homogenous deformations  $u_{ij}^0$  in the partition function. This is equivalent to considering the system under a fixed external pressure.<sup>11</sup> As a result we get a new Hamiltonian whose elastic part takes a simple form

$$H_{\text{el}} = \int \frac{d^d k}{(2\pi)^d} k^2 c_0^2 |u_L(\mathbf{k})|^2,$$

with  $c_0^2 = B + 2(d-1)\mu/d$  as the velocity square of the longitudinal sound mode of the noninteracting system.

Next with the aid of a static transformation

$$[R_{ij}^{\text{st}}] = \begin{bmatrix} i k c_0 \cos \varphi_a & -C_V^{-1/2} \sin \varphi_T \\ i k c_0 \sin \varphi_a & C_V^{-1/2} \cos \varphi_T \end{bmatrix}, \quad (8)$$

where  $\sin \varphi_a = g/(v_+ c_0)$ ,  $\cos \varphi_T = f \sqrt{C_V^0}/v_+$ , and

$$v_+^2 = \frac{g^2 c_0^{-2} + f^2 C_V^0 - 2w g f c_0^{-2} C_V^0}{1 - w^2 c_0^{-2} C_V^0},$$

we transform the variables  $(u, \sigma)$ , where  $u$  is the longitudinal displacement component, into new ones  $(m_1, m_2)$  of which only the second is coupled to the order parameter:

$$H(m_1, m_2, \Phi) = \frac{1}{2} \int \frac{d^d k}{(2\pi)^d} [m_1(\mathbf{k})^2 + m_2(\mathbf{k})^2 + 2v_+ m_2(\mathbf{k}) \Phi_{-\mathbf{k}}^2] + H_{\text{OP}}^{\text{eff}}(\Phi), \quad (9)$$

where the effective order-parameter Hamiltonian  $H_{\text{OP}}^{\text{eff}}(\Phi)$  takes the Ginzburg-Landau form with the parameters  $r$  and  $\lambda$  shifted and  $\Phi_{\mathbf{k}}^2 = \int (d^d p/2\pi) \Phi(\mathbf{p}) \Phi(\mathbf{k} - \mathbf{p})$ . We neglect here a small nonanalyticity (with respect to the wave vector) of the coupling constants; as in magnets, the resulting instability can be neglected in the experimentally accessible temperature range.<sup>10,12</sup>

## B. Dynamics

On the grounds of hydrodynamics and renormalization group arguments, the following system of dynamic equations, appropriate for the disordered phase, is considered:

$$\frac{\partial}{\partial t} \Phi = -\gamma \frac{\delta H}{\delta \Phi} + \theta_\Phi, \quad (10)$$

$$\frac{\partial}{\partial t} \sigma = \kappa \nabla^2 \frac{\delta H}{\delta \sigma} + \theta_\sigma, \quad (11)$$

$$\frac{\partial}{\partial t} u = P, \quad (12)$$

$$\frac{\partial}{\partial t} P = -\frac{\delta H}{\delta u} + \Theta \nabla^2 \frac{\delta H}{\delta P} + \theta_P, \quad (13)$$

where we have introduced the longitudinal momentum  $P$  conjugated with  $u$  (we assume a unitary mass density). This set of equations is an extension of the model C.<sup>1</sup> The Fourier components of the Gaussian white noise  $\theta_i$  ( $i = \Phi, \sigma$ , and  $P$ ) have variances related to the bare damping terms  $\gamma, \kappa k^2$ , and  $\Theta k^2$  through the usual Einstein relations. Here,  $\kappa$  denotes the thermal conductivity,  $\Theta k^2$  is responsible for the noncritical sound dumping, and  $\gamma$  is a relaxation coefficient of the order parameter.

It is convenient to analyze the dynamics of the model in terms of the equivalent functional form<sup>13,14</sup> with a Lagrangian  $L(\Phi, \sigma, u, P; \tilde{\Phi}, \tilde{\sigma}, \tilde{u}, \tilde{P})$ , where auxiliary ‘‘response’’ fields  $\tilde{\Phi}, \tilde{\sigma}, \tilde{u}$ , and  $\tilde{P}$  are introduced.

### III. TEMPERATURE- AND FREQUENCY-DEPENDENT THERMAL DIFFUSIVITY ABOVE $T_C$

The relations between the dynamic vertex functions, in our model, and the hydrodynamic transport coefficients are obtained by comparing the coefficient determinant of the linearized hydrodynamic equations  $\Delta_H$  with the determinant of the dynamic two-point vertex functions  $\Delta_{th} = \det[\Gamma_{\alpha_i \tilde{\alpha}_j}(k, \omega)]$  with  $\alpha_i = \{\Phi, m_1, m_2, m_3\}$ .<sup>14-16</sup> From the hydrodynamics we get

$$\Delta_H = (-\omega^2 - i\omega D_s k^2 + c_s^2 k^2)(-i\omega + D_T k^2) \times [-i\omega + \gamma(k^2 + \xi^{-2})], \quad (14)$$

where  $c_s$  and  $D_s$  are the longitudinal sound velocity and damping coefficient, respectively. The thermal diffusivity

$$D_T = \kappa / C_p(\tau) \quad (15)$$

is given by the ratio of the thermal conductivity and specific heat (per unit volume) at a constant pressure. The last factor in Eq. (14) describes the relaxation of the order parameter, with  $\xi$  as the correlation length. On the other hand, from perturbation theory we obtain

$$\begin{aligned} \Delta_{th} = & \{i\omega^3 - \omega^2[\Theta k^2 + (\sin^2 \varphi_T + \cos^2 \varphi_T \Gamma_{m_2 m_2}) D_T^0 k^2] \\ & - i\omega [c_0^2 k^2 (\cos^2 \varphi_a + \sin^2 \varphi_a \Gamma_{m_2 m_2}) \\ & + (\sin^2 \varphi_T + \cos^2 \varphi_T \Gamma_{m_2 m_2}) D_T^0 \Theta k^4] \\ & + c_0^2 D_T^0 k^4 \cos^2(\varphi_a - \varphi_T) \Gamma_{m_2 m_2}\} \Gamma_{\Phi \tilde{\Phi}}(k, \omega), \quad (16) \end{aligned}$$

where  $D_T^0 = \kappa / C_V^0$  and

$$\Gamma_{m_2 m_2}(\tau, \omega) = \frac{1}{\kappa} \frac{\partial}{\partial k^2} \Gamma_{m_2 \tilde{m}_2} |_{k=0}$$

is a frequency-dependent extension of the static two-point vertex function  $\Gamma_{m_2 m_2}^{(st)} = \langle m_2 m_2 \rangle^{-1}$ .<sup>14</sup>

Our main interest is the thermal transport which is determined by the temperature and frequency dependence of thermal diffusivity  $D_T(\tau, \omega)$ , where  $\tau = (T - T_C) / T_C$  is the reduced temperature. From Eq. (16) a general expression for the complex diffusivity ( $D_T = \text{Re } \hat{D}_T$ ) can be obtained:

$$\hat{D}_T(\tau, \omega) = \frac{\hat{D}_T^{(LF)}(\tau, \omega) + i\zeta \hat{D}_T^{(HF)}(\tau, \omega)}{1 + i\zeta}, \quad (17)$$

where  $\zeta = \omega(\hat{D}_T^{(LF)} - \Theta) / \hat{c}_{(ad)}^2$  is a frequency parameter. The coefficients  $\hat{D}_T^{(LF)}$  and  $\hat{D}_T^{(HF)}$  are limiting values of the complex thermal diffusivity in two regimes and

$$\hat{c}_{(ad)}^2(\tau, \omega) = c_0^2 [\cos^2 \varphi_a + \sin^2 \varphi_a \Gamma_{m_2 m_2}(\tau, \omega)] \quad (18)$$

is the (complex) adiabatic sound velocity.<sup>8,17</sup>

For (strongly damped) thermal waves<sup>19,20</sup> with real frequency and complex wave vector  $\tilde{k} = k_{th \text{ wav}}(\omega) + i\mu^{-1}(\omega)$ , where  $k_{th \text{ wav}}(\omega) = \mu^{-1}(\omega) = 2D_T / \omega$ , the parameter  $\zeta(\omega)$  has a simple interpretation: neglecting the correction from the bare sound dumping coefficient  $\Theta$ , it can be rewritten as

$$\zeta(\omega) \approx \frac{\omega D_T}{c_{(ad)}^2} = \frac{1}{2} \frac{\omega^2}{c_{(ad)}^2 \left( \sqrt{\frac{\omega}{2D_T}} \right)^2} = \frac{1}{2} \frac{\omega^2}{\omega_{\text{sound}}^2 [k = k_{th \text{ wav}}(\omega)]} \quad (19)$$

$$= 2\pi^2 \left[ \frac{\mu(\omega)}{\lambda_{\text{sound}}(\omega)} \right]^2. \quad (20)$$

So it is (to the unimportant factor) the square of the ratio of the frequency of the thermal wave to the sound frequency with the same wave vector  $k_{th \text{ wav}}(\omega)$  or, according to the second line in this equation, it is the square of the thermal diffusion length  $\mu(\omega)$  by the wave length  $\lambda_{\text{sound}}(\omega) = 2\pi c_{(ad)} / \omega$  of the sound mode with given frequency  $\omega$ .

For  $\zeta \ll 1$  (low frequency) the sound mode can be treated as an ‘‘annealed’’ variable in the process of heat diffusion. One can also say that the related sound mode propagates adiabatically. In this low-frequency limit we have

$$\hat{D}_T(\tau, \omega) \rightarrow \hat{D}_T^{(LF)}(\tau, \omega) = D_T^0 \frac{\cos^2(\varphi_a - \varphi_T) \Gamma_{m_2 m_2}(\tau, \omega)}{\cos^2 \varphi_a + \sin^2 \varphi_a \Gamma_{m_2 m_2}(\tau, \omega)}, \quad (21)$$

which is a finite-frequency generalization of the expression  $D_T = \kappa / C_p$ , where  $C_p \sim \tau^{-\alpha}$  is the static specific heat at a constant pressure giving a weak  $\tau^\alpha$  singularity of  $D_T$ . It can be shown that at zero frequency the second factor in Eq. (21) is proportional to the inverse of the isobaric specific heat. There is no perturbation contribution to the thermal conductivity in this model.<sup>2</sup>

The coefficient

$$\hat{D}_T^{(HF)}(\tau, \omega) = D_T^0 (\sin^2 \varphi_T + \cos^2 \varphi_T \Gamma_{m_2 m_2}) \quad (22)$$

corresponds to the high-frequency limit  $\zeta \gg 1$ , where the longitudinal sound variables are ‘‘frozen’’ (the related sound propagates isothermally). It can be shown that at frequencies much lower than the characteristic frequency of the order-parameter fluctuations the following holds:

$$D_T^{(HF)} = \kappa / C_V(\tau) \sim \text{const} + \tau^\alpha, \quad (23)$$

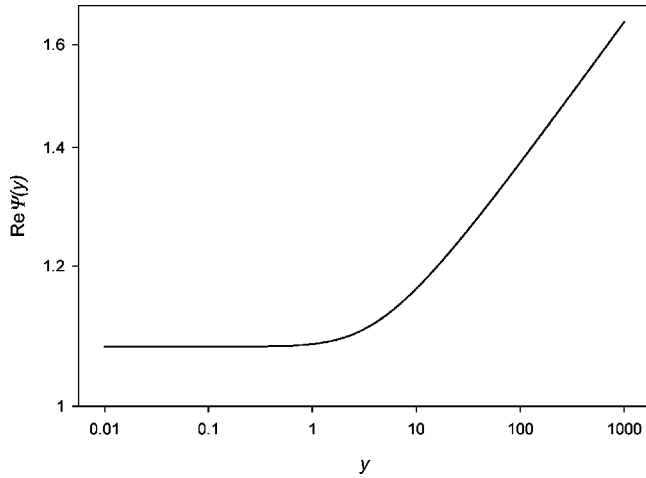


FIG. 1. The scaling function  $\text{Re } \Psi(y)$  from Eq. (24) vs the scaling variable  $y = \omega/\omega_c$  for  $\alpha = 0.110$ ,  $\nu = 0.630$  ( $n=1$ ), and  $z = 2 + \alpha/\nu = 2.175$  (the dynamic model C).

where  $C_V(\tau)$  is the constant-volume specific heat of the system, which is finite at  $T_C$ .<sup>21</sup>

The frequency-dependent vertex function  $\Gamma_{m_2 m_2}$  obeys the scaling relation

$$\Gamma_{m_2 m_2}(\tau, \omega) = \tau^\alpha \Psi(y)$$

in the asymptotic regime, where  $\Psi$  is a scaling function and  $y = \omega/\omega_c$  is the reduced frequency with characteristic frequency  $\omega_c \sim \tau^{z\nu}$  and  $\alpha$ ,  $\nu$ ,  $z$  as critical exponents. We have calculated  $\Psi$  at one-loop order:

$$\Psi(y) = \left[ 1 + \left( \frac{y}{2} \right)^2 \right]^{\alpha/2z\nu} \left\{ 1 + \frac{\alpha}{\nu} \left[ \frac{1 - iy/2}{y} \arctan \frac{y}{2} + i \frac{\ln[1 + (y/2)^2]}{2y} \right] \right\}. \quad (24)$$

We have neglected here the corrections to scaling due to the departure of the couplings  $\lambda$  and  $v_+$  from the asymptotic values.

Usually, we have  $\zeta \ll 1$  and asymptotically near the critical point the frequency-dependent thermal diffusivity behaves as

$$D_T^{(\text{LF})}(\tau, \omega) \sim \tau^\alpha \text{Re } \Psi(y).$$

The scaling function  $\text{Re } \Psi(y)$  is plotted against  $y$  in Fig. 1. In the limit  $y \rightarrow 0$  it goes to a constant, so  $D_T^{(\text{LF})}(\tau, 0) \sim \tau^\alpha$ , converging slowly to zero. For a finite frequency, the thermal diffusivity saturates, when approaching  $T_C$  ( $y \rightarrow \infty$ ), at a value  $D_T^{(\text{ad})}(0, \omega) \sim \omega^{\alpha/z\nu}$ . So it is always finite for finite frequencies. This crossover from hydrodynamic ( $y \ll 1$ ) to critical ( $y \gg 1$ ) behavior may be difficult to be detected in experiments as frequencies of the order of  $\omega_c$  are required. In magnets usually we have  $\omega_c \sim 10^{11} \tau^{z\nu} > 10^6 \text{ s}^{-1}$ , where the last inequality is obtained for the reduced temperatures  $\tau \sim 10^{-4}$ , so angular frequencies of the order of  $10^6 \text{ s}^{-1}$  and higher are needed.

The literature does not give many experimental data on the critical diffusivity in Ising-type magnets. However, recently high-resolution measurements of  $D_T$  in the uniaxial antiferromagnet  $\text{FeF}_2$  (Ref. 3) and in the weakly uniaxial antiferromagnet  $\text{Cr}_2\text{O}_3$  (Ref. 4) were reported. The frequencies used in these experiments were of the order of  $10^2 \text{ s}^{-1}$ . To be able to observe the crossover from hydrodynamic to critical behavior at so low frequencies, one would need extremely small reduced temperatures which are now outside the experimental capabilities. However, the present work can throw also some light on the detailed nature of the thermal diffusivity singularity in the hydrodynamic regime in these systems. For low frequency  $\Gamma_{m_2 m_2}(\tau, \omega)$  goes to its static limit  $\Gamma_{m_2 m_2}^{(\text{st})}(\tau) = (1 + v_+^2 \langle \Phi_0^2 \Phi_0^2 \rangle)^{-1}$ , where the angular brackets denote the static average. Inserting it into Eq. (21) we obtain

$$D_T(\tau) = D_T^{(\text{LF})}(\tau, 0) = \frac{D_T^0 \cos^2(\varphi_a - \varphi_T)}{1 + \cos^2 \varphi_a v_+^2 \langle \Phi_0^2 \Phi_0^2 \rangle} = \frac{\tilde{V}}{1 + \tilde{U} \tau^{-\alpha} (1 + F \tau^\Delta)}, \quad (25)$$

where for the ‘‘energy operator’’ correlation function  $\langle \Phi_0^2 \Phi_0^2 \rangle$  we have used the renormalization group nonasymptotic expression  $\langle \Phi_0^2 \Phi_0^2 \rangle = A \tau^{-\alpha} (1 + F \tau^\Delta) + B$  (Ref. 22) with the leading nonanalytic correction exponent  $\Delta \approx 0.53$  for the Ising system.  $\tilde{V}$ ,  $\tilde{U}$ ,  $A$ ,  $B$ , and  $F$  are nonuniversal amplitudes and analytic corrections have been neglected. Near the critical point the diverging term dominates the numerator and Eq. (25) can be expanded, giving

$$D_T(\tau) = U \tau^\alpha (1 + G \tau^\alpha + H \tau^{2\alpha} + \dots + F \tau^\Delta), \quad (26)$$

where  $U \sim 1/A$ ,  $G$ , and  $H$  are some constants. It is seen from this equation that the dominant correction exponent for the thermal diffusivity is  $\alpha \approx 0.110 < \Delta$ . Unfortunately, the measurements of thermal diffusivity in  $\text{FeF}_2$  (Ref. 3) and  $\text{Cr}_2\text{O}_3$  (Ref. 4) were fitted with the expression

$$D_T(\tau) = V + E \tau + U \tau^{-b} (1 + F \tau^\Delta), \quad (27)$$

with  $b < 0$ . Apart from the unimportant analytic correction in Eq. (27) also a constant-background term  $V$  appears, which makes the thermal diffusion coefficient finite at  $T_C$ . Equation (27) differs from Eq. (26) mainly by this constant term and the exponent of the leading correction. Fortunately, for the ideal Ising system  $\text{FeF}_2$ , this constant term was found to be extremely small.<sup>3</sup> It may be a result of the calibration procedure used by the authors, as the photopyroelectric technique gives only relative values of thermal diffusivity and specific heat. Measurements over a wider reduced temperature interval would be necessary to check the usefulness of the corrections predicted in our work:  $\tau^\alpha$ ,  $\tau^{2\alpha}$ ,  $\dots$ . The theory presented confirms also the expectation<sup>3</sup> that the critical amplitude ratio for diffusivity in the Ising system is equal to the inverse of the amplitude ratio for specific heat,  $U/U' = A'/A$ . For  $\text{FeF}_2$  the Ising-like exponent



$-b = \alpha = 0.11 \pm 0.02$  and  $U/U' = 1.97 \pm 0.08$  were found,<sup>3</sup> whereas the theory<sup>23</sup> predicts  $A'/A \approx 1.96$ .

Unfortunately, no conclusive interpretation of the measurements of  $D_T$  in  $\text{Cr}_2\text{O}_3$  is possible in terms of our theory. First, a large background term  $V$  was found<sup>4</sup> and  $b = -0.09 \pm 0.01$  for the fits with the fitting function (27). Having in mind that  $\text{Cr}_2\text{O}_3$  is almost an isotropic system, a formula similar to Eq. (27) is obtained, with a constant term, by expanding Eq. (25) (which is also valid for the Heisenberg model) in powers of  $\tau^{-\alpha_H}$ , where  $\alpha_H$  denotes the negative Heisenberg model specific heat exponent. Then we obtain essentially Eq. (27) with  $b = -\alpha_H$  and a simple relation for the amplitude ratios,  $U/U' = (A/A')_H$ , where the subscript ‘‘H’’ denotes the Heisenberg point. Thus, the substantial constant term would rather suggest Heisenberg behavior than an Ising one. However, the situation is additionally complicated by the fact that Marinelli *et al.*<sup>4</sup> obtained also a quite good fit with the truly Ising behavior described by Eq. (25) with the exponent describing the divergence of the denominator equal to  $0.11 \pm 0.02$ . Good fits with different functions in the case of  $\text{Cr}_2\text{O}_3$  may be explained by the fact that the reduced temperature interval chosen for the fitting procedure was much narrower than that used for  $\text{FeF}_2$  as a consequence of the assumed crossover from Heisenberg to Ising behavior. Thus, the situation here is still unclear and measurements over a wider reduced temperature range would be highly desirable.

The question also arises as to the extent to which the sound modes influence the heat mode. For the phonon and thermal diffusion modes uncoupled ( $g = w = 0$ ) we have  $\cos^2 \varphi_a = \cos^2 \varphi_T = \cos^2(\varphi_a - \varphi_T) = 1$  and from Eqs. (17), (21), and (22) one can see that there is no difference between the low- and high-frequency thermal diffusivity, which is then equal to  $D_T^0 \Gamma_{m_2 m_2} \sim \tau^\alpha$ . The presence of phonons changes the overall coefficient in front of  $\Gamma_{m_2 m_2}$  in Eq. (21) and introduces a singular correction in the denominator.

Comparing Eqs. (14) and (16) we have also obtained an interesting relation between the complex low-frequency and high-frequency diffusivities and longitudinal sound velocities:

$$\begin{aligned} \hat{D}_T^{(\text{LF})}(\tau, \omega) \hat{c}_{(\text{ad})}^2(\tau, \omega) &= \hat{D}_T^{(\text{HF})}(\tau, \omega) \hat{c}_{(\text{is})}^2(\tau, \omega) \\ &= c_0^2 D_T^0 \Gamma_{m_2 m_2}(\tau, \omega) \cos^2(\varphi_a - \varphi_T) \\ &\sim \tau^\alpha \Psi(y), \end{aligned} \quad (28)$$

where

$$\hat{c}_{(\text{is})}^2(\tau, \omega) = c_0^2 \frac{\cos^2(\varphi_a - \varphi_T) \Gamma_{m_2 m_2}(\tau, \omega)}{\sin^2 \varphi_T + \cos^2 \varphi_T \Gamma_{m_2 m_2}(\tau, \omega)} \quad (29)$$

is the isothermal sound velocity. The expressions (18) and (29) for the sound velocities are found from analogous considerations of the acoustic mode.<sup>8,17,18</sup> The relation (28) is a finite-frequency generalization of the known thermodynamic formula<sup>19</sup>  $(\partial p / \partial \rho)_\sigma C_v = (\partial p / \partial \rho)_T C_p$ .

#### IV. SUMMARY AND DISCUSSION

We have performed a detailed analysis of the critical behavior of the thermal diffusion mode in Ising-type magnets for  $T \geq T_C$ . The coupling to the sound mode has been fully taken into account in the prediction of the temperature and frequency dependence of the thermal diffusivity. We were able to express  $D_T(\tau, \omega)$  in terms of the vertex function  $\Gamma_{m_2 m_2}(\omega)$  of the idealized phonon-free model C. The last quantity can be relatively easily calculated by the renormalization group method. The dynamic scaling function is calculated here in the one-loop approximation. The finite spin-phonon coupling leads to additional nonasymptotic effects in addition to the ones related to the deviation of the four-spin coupling constant  $\lambda$  from the asymptotic value. The most important new effect is the existence of the two frequency regimes in the behavior of thermal diffusivity with distinct asymptotic singularities as the reduced temperature approaches zero.

In the low-frequency limit the heat conduction mode is much slower than the related sound mode with the same wave vector. Even in this limit, the temperature and frequency behavior of  $D_T(\tau, \omega)$  shows nonasymptotic corrections due to the finite values of the spin-phonon and entropy-phonon couplings as shown in Eq. (21). Otherwise, in the high-frequency limit the sound variables can be treated as frozen. In principle, in each frequency limit one can distinguish the hydrodynamic region  $\omega \ll \omega_c$ , where  $\omega_c$  is the characteristic frequency of the order-parameter fluctuations, as well as the critical region  $\omega \gg \omega_c$ . However, the temperature crossover from hydrodynamic to critical behavior, which takes place as a result of a critical slowing down, may not be observed in experiments for very low frequencies as very small reduced temperatures are then required. In order to test the frequency-dependent formulas obtained in our paper new high-resolution measurements of  $D_T$  performed at higher frequencies are highly desirable.

As a by-product of our analysis we have obtained a relation between the low- and high-frequency thermal diffusivities and isothermal and adiabatic velocities of the longitudinal sound, which is a finite-frequency generalization of the thermodynamic relation connecting the isothermal and adiabatic compressibilities as well as the specific heats at a constant pressure and at a constant volume, respectively.

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