Critical singularities of the propagation of ultrasonic waves in yttrium iron garnet near the Curie point

I. K. Kamilov and Kh. K. Aliev

Dagestan State University

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We measured the velocity and absorption of ultrasonic waves propagating in yttrium iron garnet along the crystallographic directions [100] and [110] at 5 and 30 MHz. Appreciable anomalies of the velocity and absorption were observed in the vicinity of the Curie point. The critical changes of these parameters in the paramagnetic phase agree with a power-law dependence. The critical exponents characterizing the degree of the temperature dependence of the velocity and absorption near the Curie point are $n=0.52$ and $\eta=0.53$. The equality of the velocity and absorption exponents point to a weak temperature dependence of the relaxation time. It is concluded from this that the main contribution to the anomaly of the elastic properties is made by spin-lattice relaxation.

1. INTRODUCTION

Much attention is being paid of late in investigations of magnetic phase transitions to the study of the kinetic coefficients, with which one can attempt to establish the characteristic features of the magnetic critical state. In particular, study of the propagation of ultrasonic waves near the Curie point enables us to examine the dynamics of the critical behavior of a spin system[1]. The velocity and absorption of ultrasonic waves near the magnetic-transition temperatures exhibit anomalies typical of first-order phase transitions. These anomalies are due to both relaxation and fluctuation processes[2-5].

The numerous theoretical and experimental studies[1-10] of this problem lead to the following principal conclusions:

1. The variation of the absorption coefficient k of ultrasonic waves is approximated by the general formula

$$k\sim\omega^2\varepsilon^{-\eta}, \qquad (1)$$

where ω is the angular frequency of the sound, $\epsilon = (T - T_C)/T_C$ is the relative temperature, and η is a critical exponent characterizing the features of the temperature dependence of k near the Curie point (T_C), η being smaller for antiferromagnets than for ferromagnets.

2. The critical change in the velocity does not, in the main, depend on the frequency, and exhibits a weaker temperature dependence (logarithmic, $\Delta v/v_0 \sim \omega^0 \ln\epsilon$ or power-law $\Delta v/v_0 \sim \omega^0\epsilon^{-n}$). The critical velocity exponent does not depend on the type of ordering of the spin magnetic moments.

3. According to ultrasound absorption data, all magnetic substances can be divided into two groups, magnetic metals (rare-earth elements and nickel) and magnetic insulators (EuO, MnF_2, $RbMnF_3$, etc.). Whereas the absorption anomalies in magnetic metals are due to the interaction between the ultrasonic waves and fluctuations of the order parameter, in magnetic insulators the acoustic waves interact predominantly with fluctuations of the spin energy density[8-11].

Investigation of sound propagation near the critical temperature in rare-earth elements, nickel, and certain insulators has made it possible to observe the regularities listed in items 1—3, whereas the entire group of ferrites, which are nearly magnetic insulators, has been studied little from this point of view, although their exceptional properties (they are antiferromagnets with respect to the ordering of the spin magnetic moments and ferromagnets with respect to their macroscopic properties) make these substances interesting objects.

We have started in this connection to investigate the equilibrium and kinetic parameters systematically near the ferrimagnet-paramagnet transition temperature, for the purpose of revealing the main regularities in the behavior of the spin system in the critical region. These investigations are also of interest from the point of view of the recently considered question of isomorphism of different phase transitions[12].

We report here the results of measurements of the velocity and absorption of longitudinal ultrasound waves with frequencies 5 and 30 MHz in $Y_3Fe_5O_{12}$ in the crystallographic directions [100] and [110]. The sound absorption was measured by the usual echo-pulse procedure, while the velocity was measured by two precision methods: the Aleksandrov method[13] and the McSkimin phase-comparison method[14]. The errors did not exceed 0.06% and 5% in the velocity and damping measurements, respectively. The temperature was measured with a chromel-alumel thermocouple with an absolute error 0.5°C. The temperature stabilization was no worse than 0.01°C.

2. RESULTS AND DISCUSSION

In the ferrimagnet-paramagnet transition-temperature region, the longitudinal-wave velocity ($v_{||}$) reveals an anomalous behavior typical of second-order phase transitions. At the Curie point (275°C, Fig. 1), $v_{||}$ goes through a minimum, and the absorption maximum occurs at the same temperature (the absorption results were published earlier[15]). The slight shift of the velocity minimum toward lower temperatures relative to the absorption peak that was observed in pure nickel[6], was not observed by us in yttrium iron garnet, this being apparently connected with the low temperature resolution and with difficult-to-monitor distortions of the iron-garnet crystal structure.

Both the velocity and the absorption of the ultrasound waves have an anisotropic behavior at the Curie point, in particular

$$\left[\frac{\Delta v}{v_0}\right]_{[110]}\Big/\left[\frac{\Delta v}{v_0}\right]_{[100]}=1.28$$

(v_0 is the velocity at the initial measurement temperature). The anisotropy of the velocity and of the absorp-

tion in $Y_3Fe_5O_{12}$ may be due to the contribution of the one-ion spin-lattice coupling mechanism.

The critical change in the velocity does not depend on the sound frequency, whereas the critical absorption changes when the frequency varies near T_C. Figure 2 shows data on the absorption at frequencies 5 and 30 MHz for the [110] direction. The temperature at which the absorption peak occurs at 5 MHz coincides with the Curie point. The absorption peak at 30 MHz is shifted on the temperature scale toward higher temperatures relative to that at 5 MHz. This shift lies within limits of the temperature-measurement error and is therefore not discussed. Although all the theories of ultrasound absorption predict a quadratic dependence on its frequency in the low-temperature limit, we observed in yttrium iron garnet a lower critical absorption at 30 MHz than at 5 MHz. In this case, the condition $\omega\tau \ll 1$ is probably not satisfied and the relaxation theory of sound absorption holds true. If we assume that the main coupling responsible for the critical effects in the yttrium iron garnet is that between the acoustic waves and the spin energy density, then, following [10], we can write

$$k \sim \frac{q^2 BC\Sigma\tau_i^{-1}}{\omega^2+(\Sigma\tau_i^{-1})^2} \qquad (2)$$

where q is the wave vector of the sound, C is the magnetic specific heat, B is the bulk-magnetostriction coupling constant, and τ_i is the relaxation time. According to our estimate, the relaxation time in $Y_3Fe_5O_{12}$ is of the order of 10^{-8} sec, and the frequency region satisfies the condition $\omega\tau \sim 1$. With further increase of the frequency it is possible that the absorption peak will vanish completely, because the frequency region ~100 MHz and higher satisfies the condition $\omega\tau \gg 1$. It follows then from (2) that k does not diverge in this frequency region. A similar relation was observed in $Y_3Fe_5O_{12}$ in the measurement of the complex susceptibility of the paraprocess [16]. Such a frequency dependence of the sound absorption and of the susceptibility of the paraprocess can be attributed to the appearance of spin-correlation formations near the Curie point.

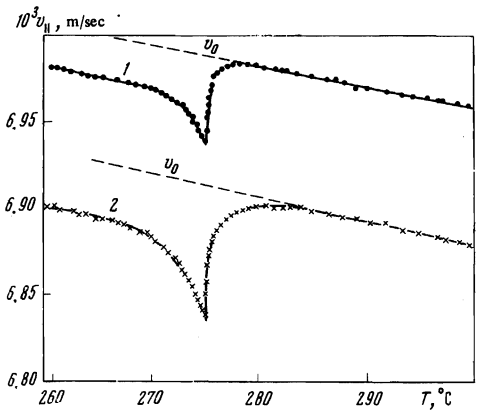

FIG. 1. Temperature dependence of $v_{\parallel}$: 1–for the direction [100], 2–for [110], $T_C = 275°K$.

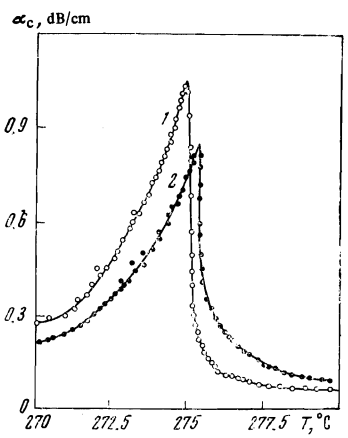

FIG. 2. Critical absorption in the [110] direction: 1–at 5 MHz; 2–30 MHz.

3. DETERMINATION OF THE CRITICAL EXPONENTS

A good test of the idealized theoretical premises is the critical exponent. Experimental determination of the critical exponents encounters certain difficulties connected with the determination of the normal change in the velocity in the critical region and of the transition temperature. In most cases, the normal change of the velocity in the critical region is obtained by extrapolating the temperature dependence of $\Delta v/v_0$. We regard as more reliable determination of the $v_0(T)$ dependence by linearly extrapolating the absolute value of the velocity from the paramagnetic phase. Using this method of determining the normal change in the velocity, we found that in the temperature interval $5 \times 10^{-4} \leq \epsilon \leq 10^{-2}$ the experimental points agree well with the power-law dependence $\Delta v/v_0 \sim \omega^0 \epsilon^{-n}$ with a critical exponent $n = 0.52$ (see Fig. 3).

In view of the absence of a rigorous expression for the regular part of the sound absorption near the Curie point, the absorption background is usually established from experimental data. Recognizing that the temperature interval of anomalous sound absorption is quite narrow, it can be assumed that the background absorption near the Curie point does not vary with temperature, and we choose the value of the background far from the transition, approximately 10–20°C above T_C. Indeed, the sound absorption hardly changes above this temperature interval. An estimate of the background value of the sound absorption makes it possible to separate the anomalous part of the absorption and, consequently, to establish the value of its critical exponent. The corresponding reduction of the experimental data (Fig. 4) shows that for absorption the critical exponent is $\eta = 0.53$ in the interval $1.3 \times 10^{-3} \leq \epsilon \leq 10^{-2}$. It should be noted that whereas the impurities strongly affect the absorption and since the temperature dependence of k near T_C is given by the formula $k \sim e^{-\gamma\epsilon}$, the impurities do not affect the velocity down to $\epsilon = 5.0 \times 10^{-4}$.

Identification of the transition temperature with the absorption peak or with the velocity minimum always introduces a certain error owing to their shift on the tem-

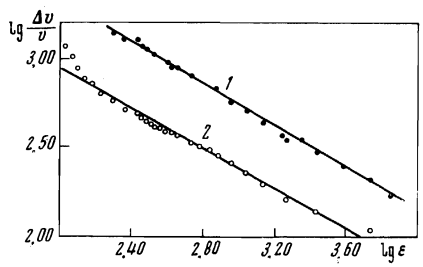

FIG. 3. Doubly-logarithmic plots of $\Delta v/v_0$ against ϵ: 1–for [100], 2–for [110]. $T > T_C$; $n = 0.52 \pm 0.04$.

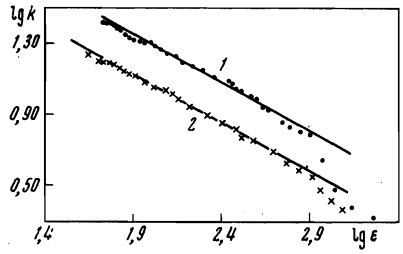

FIG. 4. Doubly-logarithmic plot of k against ϵ: 1–for [100], 2–for [110] (at 5 MHz), $T > T_C$; $\eta = 0.53 \pm 0.1$.

perature scale[6]. It is therefore preferable to measure this temperature by an independent method. We determined the T_C of $Y_3Fe_5O_{12}$ from magnetic measurements by the thermodynamic-coefficient method. The Curie point determined in this manner is 275°C and agrees well with the data obtained from the complex susceptibility of the paraprocess and from diffraction of polarized neutrons[16,17]. It is possible to determine the critical exponent of the velocity without connecting this determination either with the normal change in the velocity or with the transition temperature[18]. Using the derivatives $v''(T)$ and $v'''(T)$ we determined from the formula

$$v''/v''' = (T - T_c)/(n+2)$$

not only the value of n from the slope of the straight line, but also T_C, which is the intercept of the line on the abscissa axis. This method yields $n = 0.50$ and $T_C = 275°C$.

A comparison of the experimental value of n with the known theories shows that the critical exponent of the velocity agrees well with results that follow from Bennet's theory[7]. According to Bennet, the values of n lie in the interval $0.0 \leq n \leq 0.66$. The lower limit was obtained from the assumption that

$$(\chi F)^{1/n} \sim C_v(F) \sim \ln \varepsilon + a,$$
$$(\chi' A)^{1/n} \sim C_v(A) \sim \ln \varepsilon + b,$$

where χ and χ' stand for the static susceptibility, while F and A are the exchange integrals for ferromagnets and antiferromagnets, respectively. The upper limit was obtained from scaling theory[12], which postulates $\chi F = \chi' A \sim \epsilon^{-4/3}$.

As to the critical exponent of absorption, the experimental value of the exponent for $Y_3Fe_5O_{12}$ is much smaller than the values predicted theoretically. A similar situation is observed in magnetic insulators[9,10]; by the same token we can state that the dynamics of the critical behavior of the spin system in yttrium iron garnet is similar to the behavior of magnetic insulators. In this case, as noted above, the interaction of the sound waves with the fluctuations of the spin energy density is the principal mechanism of anomalous absorption of high-frequency sound.

At $T < T_C$, the experimental values of ultrasound absorption satisfy a logarithmic relation, and in the case of the velocity we have a power law with a critical exponent $n = 0.06$.

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