Anomalies of Brillouin scattering of light near phase transitions in $KMnF_3$ crystal

V. I. Fomin and Yu. A. Popkov

Physico-technical Institute of Low Temperatures, Ukrainian Academy of Sciences, Kharkov
(Submitted April 24, 1975)
Zh. Eksp. Teor. Fiz. **70**, 123–131 (January 1976)

Measurements were made of the frequencies of longitudinal acoustic phonons in the temperature range 60–300°K along three directions of propagation. Anomalous behavior of the frequencies was observed at 187.6, 102, and 87°K, and band broadening took place at the first phase transition. The observed changes in the frequencies were analyzed on the basis of a model allowing for the interaction between acoustic phonons and soft optical modes Γ_{25}(187.6°K) and M_3(102°K). A determination was made of the elastic moduli of a $KMnF_3$ crystal in the tetragonal phase (in units of 10^{11} dyn/cm^2): $c_{11}=11.21$, $c_{33}=10.46$, $c_{12}=3.8$, $c_{13}=4.55$, $c_{44}=2.34$, and $c_{66}=2.56$. A comparison of the velocity of hypersonic waves in the [100] direction with the ultrasonic data confirmed the relaxation mechanism of the anomlaous Brillouin scattering of light at phase transitions.

PACS numbers: 78.30.Ly, 78.20.Hp, 64.70.−p

INTRODUCTION

A crystal of $KMnF_3$ has the cubic perovskite structure at room temperature, its space group is O_h^1, and it undergoes several phase transitions when its temperature is lowered. The most thoroughly investigated is the first transition at $T_{c1}=186$ °K to the tetragonal phase D_{4h}^{18} [1] accompanied by doubling of the unit cell. This is a first-order phase transition close to a second-order one, and it is due to the instability of the normal vibration mode Γ_{25} [2] at the [111] boundary of the Brillouin zone. Condensation of the soft mode M_3 at the [110] boundary of the Brillouin zone at $T_{c2}=91.5$ °K [3] is responsible for the second phase transition. In addition to structural displacive transitions, antiferromagnetic ordering of a crystal takes place at $T_N=88$ °K. [4] The structure of $KMnF_3$ below T_N has not yet been finally established but the experimental data on the scattering of light [5,6] show that a unit cell contains at least four formula units.

The increase in the unit cell volume as a result of the structural phase transitions results in the location of the points R and M of the cubic phase at the centers of the new Brillouin zones below the temperatures T_{c1} and T_{c2}, respectively, and the soft modes Γ_{25} and M_3 become active in the Raman spectrum. [5–7]

The transition at 186 °K in $KMnF_3$ is analogous to the thoroughly investigated phase transition at 105 °K in $SrTiO_3$, but $KMnF_3$ exhibits a number of distinguishing features. For example, the frequency of the phonon branch at the points R and M in the Brillouin zone of $KMnF_3$ is much lower [2,8] and the width of the soft mode band is comparable with the spectral position of its maximum even at temperatures 40 °K above T_{c1}. [8,9] The relationships between the intensities of the soft mode components in the Raman spectra of $KMnF_3$ and $SrTiO_3$ are quite different.

The reduction in the frequency of a soft mode on approach to a transition temperature increases the rate of scattering of phonons by other phonons and, consequently, reduces the thermal conductivity [10] and increases the absorption of sound. This behavior is due to the interaction between acoustic phonons and soft optical modes. [11–14] Ultrasonic measurements, carried out on $KMnF_3$ in a wide frequency range from 10 to 680 MHz demonstrate an abrupt change in the velocity of sound near T_{c1} accompanied by an anomalous increase in the attenuation. [15–21] Similar behavior of ultrasound is observed also near the second structural phase transition at $T_{c2}=91.5$ °K. [20]

Since phase transitions split a $KMnF_3$ crystal into twins, [7] the measured elastic moduli of the low-temperature phase represent average values. Moreover, the presence of crystallographic twin walls may give rise to an additional absorption of ultrasound, particularly at low frequencies. The Brillouin scattering of light is free from secondary effects and it can be used to investigate acoustic phonons of frequencies two orders of magnitude higher than those which can be studied by ultrasonic methods. The Brillouin scattering in a $KMnF_3$ crystal had been investigated earlier at room temperature. [22,23] The elastic moduli and the velocities of hypersonic waves calculated from the Brillouin scattering data were in good agreement with the results of ultrasonic investigations indicating that there was no dispersion of the velocity of sound in the frequency range from 10^7 to 10^{10} Hz. We investigated the behavior of longitudinal acoustic (LA) phonons traveling along various directions in a wide temperature range. [24]

EXPERIMENTAL METHOD

An He–Ne laser emitting 80 mW at $\lambda=6328$ Å was the source of light in the determination of the Brillouin spectra. Light scattered by a sample at an angle of 90° was directed to a scanning Fabry–Perot interferometer with plates separated by 3 or 4 mm. The scattered light was recorded with a cooled photomultiplier operating as a photon counter. [22] A sample was placed in a cryostat where measurements could be made in the range 30–300 °K without altering the position of the sample relative to the laser beam. This was important because it enabled us to determine the Brillouin spectra at all temperatures in one purest sample selected at room (or some other) temperature. The tempera-

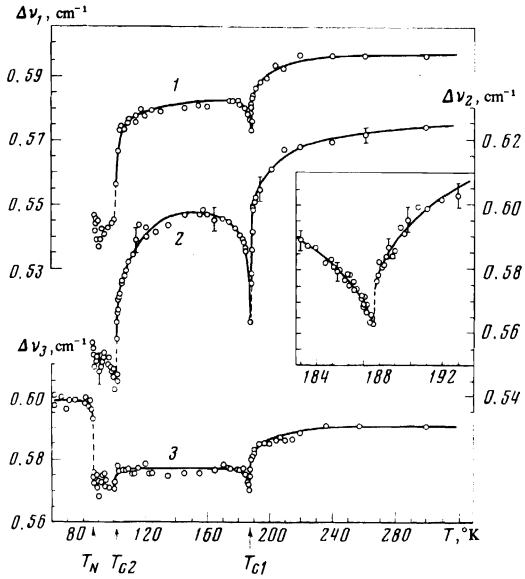

FIG. 1. Temperature dependences of the LA phonon frequency for different experimental geometries: 1) $\mathbf{k} \parallel [100]$, $\mathbf{k}' \parallel [010]$, $\mathbf{q} \parallel [110]$; 2) $\mathbf{k} \parallel [\bar{1}10]$, $\mathbf{k}' \parallel [110]$, $\mathbf{q} \parallel [100]$; 3) $\mathbf{k} \parallel [1\bar{1}0]$, $\mathbf{k}' \parallel [001]$, $\mathbf{q} \parallel [11\sqrt{2}]$. The region of the first phase transition for curve 2 is shown in the inset.

ture of a sample and the absence of a temperature gradient were monitored by copper-constantan thermocouples to within at least 0.03 °K. Before each measurement a sample was kept at a given temperature for at least 30 min.

We used $KMnF_3$ single crystals grown by the Bridgman method.[25] They were oriented by x-ray diffraction to within 1°. Samples were cut in the form of slabs and their faces were carefully polished. We used samples of two orientations with the edges directed along the [100], [010], and [001] axes and along [110], $[1\bar{1}0]$, and [001] axes of the cubic cell. The Raman spectra measurements, reported earlier in[6], were carried out on the same sample.

EXPERIMENTAL RESULTS

The temperature dependences of the LA phonon frequencies are plotted in Fig. 1 for three directions of propagation. The directions of the wave vectors of the incident $\mathbf{k}$ and scattered $\mathbf{k}'$ light, and of sound $\mathbf{q}$ are shown relative to the crystallographic axes of cubic phase. It is clear from Fig. 1 that the Brillouin line frequencies behave anomalously at temperatures 187.6, 102, and 87 °K. At these temperatures there are abrupt changes in the spectrum of the soft phonon modes.[6] When temperature is lowered, the LA phonon frequencies vary continuously except for a sudden change at $T_{c1} = 187.6 \pm 0.1$ °K. Careful measurements indicate that, to within 0.1 °K, the critical temperature is the same irrespective of whether a sample is cooled or heated. The absolute fall in the frequency depends on the direction of propagation of the LA phonons and reaches 10% for $\mathbf{q} \parallel [100]$. When temperature is lowered below T_{c1}, the phonon frequency increases but it becomes saturated at a lower absolute value than before the transition.

Similar behavior is exhibited also at the temperature of the second phase transition $T_{c2} = 102$ °K, where again a smooth variation ends with a sudden change and there is no hysteresis to within 0.2 °K. A strong rise of the intensity of light scattered elastically by optical twin boundaries makes it impossible to carry out measurements below 87 °K. Only in the $\mathbf{q} \parallel [11\sqrt{2}]$ case (this is the direction closest to the space diagonal of the cubic cell) can these measurements be made. The frequency of the LA phonons traveling along this direction changes only slightly at 187.6 and 102 °K but it rises abruptly at 87 °K and it exhibits a hysteresis of the order of 1° at this point. The intensities of the Brillouin satellites decrease approximately linearly with falling temperature and below 60 °K they are so weak that it is no longer possible to measure their frequencies against the background of the much enhanced elastic scattering.

The intensity of light scattered by the transverse (TA) acoustic waves is an order of magnitude lower than the intensity of light scattered by the longitudinal waves, and the frequencies are lower in the transverse case.[22] Therefore, the masking influence of the wing of the scattering line at the excitation frequency affects the transverse waves more strongly and the influence of temperature on these waves cannot be investigated.

In addition to changes in the LA phonon frequency in the region of the phase transition from the cubic to the tetragonal phase, there is also broadening of the Brillouin line. The dependence of the line width (found by averaging several measurements of the Stokes and anti-Stokes components in the $\mathbf{q} \parallel [100]$ direction) on temperature is plotted in Fig. 2. The line width Γ_a is found by subtracting the instrumental width (0.05 cm^{-1}) from that found experimentally. Since the LA phonon wavelength $\Lambda = 0.3$ μ is much less than the typical thickness of crystallographic twins (100–200 μ[7,28]), we may assume that the scattering of phonons by the twin boundaries makes no significant contribution to the observed Brillouin line width.

DISCUSSION OF RESULTS

The anomalous behavior of sound near structural phase transitions is due to the interaction between the long-wavelength acoustic phonons and soft optical modes. Therefore, we shall consider first the behavior of soft modes. We shall confine our discussion to the highest-temperature transition at $T_{c1} = 187.6$ °K because it has

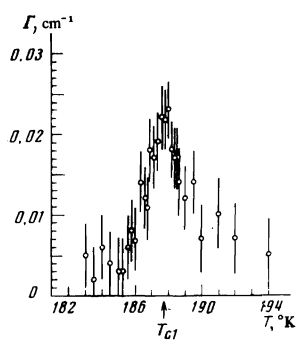

FIG. 2. Temperature dependence of the width Γ_a of the Brillouin line in the region of T_{c1}.

been investigated most thoroughly. This displacive transition is associated with the condensation of the Γ_{25} mode at the point R in the Brillouin zone, corresponding to the rotation of the fluorine ion octahedron about the ⟨100⟩ axis.

The soft phonon modes have been investigated by various methods. The results of inelastic neutron scattering[9] demonstrate that the temperature dependence of the frequency ω_T of the mode Γ_{25} in the range $T > T_{c1}$ is

$$\hbar^2\omega_T^2 = A(T - T_c), \tag{1}$$

where $A = 0.11 \pm 0.02$ meV^2/°K and $T_c = 186 \pm 2$ °K. This dependence is represented by the continuous curve in Fig. 3. Below T_{c1} the unit cell is doubled and the point R is located at the center of the Brillouin zone of the tetragonal phase. The triply degenerate mode Γ_{25} splits into a doubly degenerate one and nondegenerate modes E_g and A_{1g}, which become active in the Raman spectrum. The temperature dependence of the frequency of the mode E_g, based on the Raman data[6] allowing for the dependence such as Eq. (1), is represented by the dashed curve in Fig. 3. In the case of the mode A_{1g} the inelastic neutron scattering data are available only for one temperature: 135 °K (this point is shown in Fig. 3). Lockwood and Torrie[5] observed a 62 cm^{-1} band at 100 °K by the Raman method; this band was interpreted as the soft mode A_{1g}. Our measurements indicated that at this temperature the frequency range near 70 cm^{-1} included a two-magnon scattering band observed earlier at lower temperatures.[26,27] Thus, the available experimental data are insufficient for the reconstruction of the temperature dependence of the frequency of the soft mode A_{1g} so that we shall have to make some estimates.

The structural phase transition in $KMnF_3$ at T_{c1} is a first-order transition close to second order with $\Delta T = T_{c1} - T'_{c1} = 0.5$ °K.[3,29] This small value of ΔT clearly explains the absence of such characteristic features of first-order transitions as latent heat,[30] change in the specific volume,[31] and jump of the transition parameter.[32] The soft mode frequency ω_T decreases considerably on approach to T_{c1} from the high-temerpature side but it remains finite. Clearly, far from the critical point this transition differs little from a second order transition.[33] This allows us to use the results of Thomas and Müller[11] and of Slonczewski and Thomas[13] in estimating the frequency of the soft mode A_{1g} at temperatures below T_{c1}. It follows from the equation of motion that

$$\omega_A(T_c - \delta T)/\omega_T(T_c + \delta T) = \sqrt{2}. \tag{2}$$

The chain curve in Fig. 3 is plotted using the above relationship and the data for the frequency ω_T.[9] It should be pointed out that the expression (2) does not allow for the damping of the critical vibrations. The results of inelastic neutron scattering[9] and of light scattering[6] studies show that the width of the soft mode becomes comparable with its frequency 30–40 °K before the critical temperature. Allowance for the damping complicates considerably Eq. (2). However, we

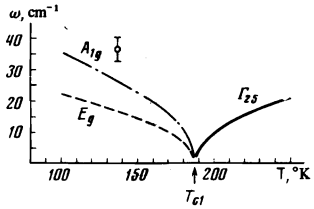

FIG. 3. Behavior of the frequencies of the soft modes in $KMnF_3$. The continuous curve represents the results of inelastic neutron scattering[9] for the Γ_{25} mode; the dashed curve gives the Raman results for the E_g mode[6]; the dash-dot curve is the frequency of the A_{1g} mode calculated using Eq. (3); the point is the result taken from[2].

can see from Fig. 3 that the experimental frequency of the mode A_{1g} at $T = 135$ °K is close to the value calculated without allowance for the damping. Thus, at these temperatures we can ignore the damping and continue our estimates.

We shall now consider the interaction between the soft optical modes and acoustic phonons. In accordance with the symmetry of the crystal at $T > T_{c1}$, the soft mode Γ_{25} is inactive in the Raman spectra and the linear coupling to the acoustic vibrations with $\mathbf{q} \sim 0$ is forbidden. Therefore, we have to allow for the nonlinear interaction, which alters the acoustic phonon energy because of the inelastic scattering by a pair of phonons belonging to a soft optical branch with $\mathbf{q} = \pi/a$. The change in the velocity sound in the cubic phase in the vicinity of the transition, estimated allowing for this interaction, is[12]

$$\Delta V \sim (T - T_c)^{-1/4}. \tag{3}$$

A similar result was obtained by Levanyuk[34,35] by considering the interaction of elastic waves with spatially inhomogeneous fluctuations of the transition parameter.

An exact determination of the critical exponent of the experimentally determined dependence of the change in the frequency of sound (proportional to the velocity) on ΔT near T_{c1} is pointless because the observed phase transition is a first-order transition and the critical temperature is indeterminate. However, it is clear from Fig. 1 that in the range $T > T_{c1}$ the dependence (3) is obeyed qualitatively.

Below the phase transition temperature the soft mode becomes active in the Raman spectrum and now either linear or resonant coupling describes the influence of the soft optical phonon on the acoustic properties of a crystal. Without allowance for relaxation processes, this interaction results in a sudden change in the frequency of the acoustic phonons at $T = T_{c1}$. A treatment of this kind based on the phenomenological model was applied to $SrTiO_3$[13] and it gave results in good agreement with the experimental data. All the calculations are best presented as a change in the elastic moduli of a crystal and then the six moduli of the tetragonal phase are related to the three moduli of the cubic phase by

$$\begin{aligned} &c_{11} = c_{11}^c - D, \quad c_{33} = c_{11}^c - 4D, \quad c_{12} = c_{12}^c - D, \\ &c_{13} = c_{12}^c + 2D, \quad c_{44} = c_{44}^c - E, \quad c_{66} = c_{44}^c, \end{aligned} \tag{4}$$

where

$$D=4\sigma_s^2(c_{11}^c-c_{12}^c)^2/9Q_s^2M\omega_A^2, \quad E=B_t^2Q_s^2/M\omega_E^2, \tag{5}$$

c_{ij}^c are the elastic moduli of the cubic phase, $\sigma_s=c/a-1$ is the change in the lattice parameter T_{c1}, Q_s is the linear displacement of a fluorine atom related to the angle φ of the rotation of the octahedron by $\tan\varphi=2Q_s/a$, $M=2m_F/a^3$, m_F is the mass of a fluorine atom, and B_t is a constant which is independent of temperature.

The elastic moduli of a crystal are related to the measured Brillouin line frequency by

$$X_i=\frac{\rho}{2}\left(\frac{c}{n_i}\frac{\Delta\nu}{\nu}\right)^2, \tag{6}$$

where X_i is the function of the elastic moduli of a crystal, ρ is the density, n_i is the refractive index, and ν is the frequency of the incident light. In the case of the cubic phase of $KMnF_3$ and propagation of the LA phonons along the principal crystallographic directions the relationship between the observed frequencies $\Delta\nu$ of the Brillouin lines and the elastic moduli is given in[22,23] Thus, in the case of propagation of the LA phonons along the cubic axis ($\mathbf{q}\parallel[100]$), the values of X_i in Eq. (6) are simply equal to c_{11}.

All the structural phase transitions are accompanied by the splitting of samples into systems of crystallographic twins.[7,28] Below T_{c1} (phase transition $O_h^1-D_{4h}^{18}$) the c axes are parallel to the initial fourfold axis and mutually perpendicular in neighboring twins, which should result in the splitting of the Brillouin line into two components of frequencies $\Delta\nu=k(c_{11}/\rho)^{1/2}$ and $\Delta\nu=k(c_{33}/\rho)^{1/2}$, where k of Eq. (6) is $k=\sqrt{2}n_i\nu/c$.

The experimental Brillouin spectra exhibit just one line, which shows that the measurements were confined to one twin. For $\mathbf{q}\parallel[100]$ we have $\Delta\nu=0.598$ cm^{-1} (145 °K) which gives the elastic modulus $c_{ii}=10.46\times10^{11}$ dyn/cm^2, which is either equal to c_{11} or c_{33}. Since the direction of the tetragonal axis in the investigated part of the crystal is not known, the difference $c_{ii}-c_{11}^c=1\times10^{11}$ dyn/cm^2 corresponds, in accordance with the system (4) either to D or to $4D$. This indeterminacy can be removed by estimating the value of D in accordance with Eq. (5), using the values of the frequency of the mode A_{1g} calculated earlier (Fig. 3) and employing the experimental values for the other quantities. If the phase transition at $T_{c2}=102$ °K does not yet affect the LA phonon frequency, we find that at $T=145$ °K we have $Q_s=0.3\times10^{-8}$ cm,[1] $\sigma_s=(2.9-3.6)\times10^{-3}$,[31,36] $c_{11}^c-c_{12}^c=(0.741-0.756)\times10^{12}$ dyn/cm^2,[15,22] $M=0.86$ g/cm^3, $\omega_A=24.3$ cm^{-1}. We then find that D is 0.2×10^{11} dyn/cm^2.

The dash-dot curve in Fig. 3 is plotted ignoring the interaction of the soft optical modes with the acoustic mode. However, this interaction has little influence on the value of ω_A so that the above estimate allows us to remove the indeterminacy mentioned earlier. Thus, in the case under consideration the LA phonons travel along the tetragonal axis, i.e., we have $c_{ii}=c_{33}$ and the value of D deduced from the experimental data is 0.25×10^{11} dyn/cm^2.

We shall now determine the value of E using the Brillouin line frequency $\Delta\nu=0.583$ cm^{-1} for $\mathbf{q}\parallel[110]$. In this geometry, we have[23]

$$X_i=\tfrac{1}{4}(c_{11}+c_{33}+2c_{44})+[\tfrac{1}{16}(c_{11}+c_{33}+2c_{44})^2+\tfrac{1}{4}(c_{13}+c_{44})^2-\tfrac{1}{4}(c_{11}+c_{44})(c_{33}+c_{44})]^{1/2} \tag{7}$$

if the c axis of the tetragonal phase is in the scattering plane. The only unknown parameter in Eq. (7) is E and we obtain $E=0.22\times10^{11}$ dyn/cm^2. We can now determine all the elastic moduli of $KMnF_3$ in the tetragonal phase (in units of 10^{11} dyn/cm^2):

$$\begin{aligned} &c_{11}=11.21, \quad c_{33}=10.46, \quad c_{12}=3.8,\\ &c_{13}=4.55, \quad c_{44}=2.34, \quad c_{66}=2.56. \end{aligned} \tag{8}$$

These values can be checked using the third experimental geometry with $\mathbf{q}\parallel[11\sqrt{2}]$. The frequency (Fig. 1) at $T=145$ °K is 0.576 cm^{-1} and a calculation in accordance with the formula[23]

$$X_i=\tfrac{1}{8}(c_{11}+c_{12}+2c_{33}+4c_{44}+2c_{66})+[\tfrac{1}{64}(c_{11}+c_{12}+2c_{33}+4c_{44}+2c_{66})^2+\tfrac{1}{4}(c_{13}+c_{44})^2-\tfrac{1}{8}(c_{11}+c_{12}+2c_{44}+2c_{66})(c_{33}+c_{44})]^{1/2}, \tag{9}$$

which corresponds to this case, gives $\Delta\nu=0.575$ cm^{-1}, i.e., it is in excellent agreement with the experimental results.

Thus, the proposed mechanism of linear coupling of the soft optical modes and the acoustic mode explains satisfactorily the reduction in the LA phonon frequency below the transition. However, it is not possible to carry out a complete calculation of the behavior of the LA phonons throughout this temperature range, including the phase transition and thus it is not possible to determine separately the contribution of the nonlinear interaction. This is due to two reasons: firstly, the exact temperature dependence of the frequency of the soft mode A_{1g} is not available; secondly, it is assumed in the above calculations that the temperature dependence of the transition parameter is given by the simple relationship $\varphi\propto(T_{c1}-T)^{1/2}$. This dependence is indeed satisfied for $KMnF_3$ in the temperature range 120–170 °K but in the immediate vicinity of T_{c1} we find[32] that $\varphi\propto(T_{c1}-T)^{1/3}$.

It is clear from Fig. 1 that the behavior of the Brillouin frequency at the second phase transition ($T_{c2}=102$ °K) is analogous to the tetragonal distortion case discussed above but it is due to the interaction of the LA wave with the soft mode M_3 responsible for this transition. The jump in the Brillouin frequency at 87 °K is either due to a strong magnetoelastic coupling or due to a possible change in the symmetry of the crystal accompanying the transition to the antiferromagnetic state. It should be noted that at $T_N=87$ °K there is also a jump in the soft mode frequencies in the Raman spectrum.[6]

As pointed out earlier, there is no dispersion of the velocity of sound at room temperature in the frequency range from 10^7 to 10^{10} Hz. We shall now compare the

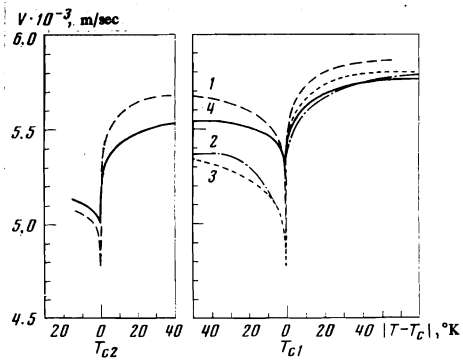

FIG. 4. Temperature dependences of the velocity of the longitudinal sound along the [100] direction of the original cubic structure near phase transitions. Curve 1 is plotted for 11.7 MHz, [20] curve 2 for 30 MHz, [16] curve 3 for 14 MHz, [18] and curve 4 represents our results.

results of the present study with those obtained in ultrasonic measurements near phase transitions. The published data on the velocity of sound are plotted in Fig. 4. The velocity of sound under the Brillouin scattering conditions is found from the simple relationship

$$V_t = \frac{1}{\sqrt{2}} \frac{c}{n_i} \frac{\Delta\nu}{\nu} \qquad (10)$$

where $\Delta\nu$ is the experimentally measured frequency (Fig. 1). It should be pointed out that the absolute values of T_{c1} and T_{c2} obtained by different authors are not in agreement. It is clear from Fig. 4 that ultrasonic measurements reveal quite large changes in the velocity of sound near the critical temperatures, which confirm the relaxation mechanism of the anomalous behavior of sound resulting in dispersion. At temperatures below T_{c1} it is not possible to compare the hypersonic and ultrasonic values because a good agreement is lacking even between the various ultrasonic data. This may be due to the influence of twinning below transition point on the behavior of ultrasound.

We are deeply grateful to V. V. Eremenko for his constant interest and valuable discussions, to K. S. Aleksandrov for his interest in this investigation, and to B. V. Beznosikov for kindly supplying $KMnF_3$ single crystals.

[1]V. J. Minkiewicz, Y. Fujii, and Y. Yamada, J. Phys. Soc. Jpn. 28, 443 (1970).
[2]V. J. Minkiewicz and G. Shirane, J. Phys. Soc. Jpn. 26, 674 (1969).
[3]G. Shirane, V. J. Minkiewicz, and A. Linz, Solid State Commun. 8, 1941 (1970).
[4]A. J. Heeger, O. Beckman, and A. M. Portis, Phys. Rev. 123, 1652 (1961).
[5]D. J. Lockwood and B. H. Torrie, J. Phys. C 7, 2729 (1974).
[6]V. V. Eremenko, V. I. Fomin, Yu. A. Popkov, and N. A. Sergienko, Fiz. Nizkikh Temp. 1, 1030 (1975) [Sov. J. Low Temp. Phys. 1, (1975)].
[7]Yu. A. Popkov, V. V. Eremenko, and V. I. Fomin, Fiz. Tverd. Tela (Leningrad) 13, 2028 (1971) [Sov. Phys.-Solid State 13, 1701 (1972)].
[8]S. M. Shapiro, J. D. Axe, G. Shirane, and T. Riste, Phys. Rev. B 6, 4332 (1972).
[9]K. Gesi, J. D. Axe, G. Shirane, and A. Linz, Phys. Rev. B 5, 1933 (1972).
[10]K. Hirakawa, K. Hamazaki, H. Miike, and H. Hayashi, J. Phys. Soc. Jpn. 33, 268 (1972).
[11]H. Thomas and K. A. Müller, Phys. Rev. Lett. 21, 1256 (1968).
[12]E. Pytte, Phys. Rev. B 1, 924 (1970).
[13]J. C. Slonczewski and H. Thomas, Phys. Rev. B 1, 3599 (1970).
[14]F. Schwabl, Phys. Rev. B 7, 2038 (1973).
[15]K. S. Aleksandrov, L. M. Reshchikova, and B. V. Beznosikov, Phys. Status Solidi 18, K17 (1966).
[16]K. S. Aleksandrov, L. M. Reshchikova, and B. V. Beznosikov, Fiz. Tverd. Tela (Leningrad) 8, 3637 (1966) [Sov. Phys.-Solid State 8, 2904 (1967)].
[17]J. M. Courdille and J. Dumas, Solid State Commun. 9, 609 (1971).
[18]B. Okai and J. Yoshimoto, J. Phys. Soc. Jpn. 34, 837 (1973).
[19]E. R. Domb, T. Mihalisin, and J. Skalyo Jr, Phys. Rev. B 8, 5837 (1973).
[20]K. Fossheim, D. Martinsen, and A. Linz, in: Anharmonic Lattics, Structural Transitions, and Melting (ed. by. T. Riste), Noordhoff Intern. Publ., Leiden, 1974, p. 141.
[21]I. Hatta, M. Matsuda, and S. Sawada, J. Phys. C 7, 299 (1974).
[22]Yu. A. Popkov, V. I. Fomin, and L. T. Kharchenko, Fiz. Tverd. Tela (Leningrad) 13, 1626 (1971) [Sov. Phys.-Solid State 13, 1360 (1971)].
[23]Yu. A. Popkov and V. I. Fomin, Proc. Second Intern. Conf. on Light Scattering in Solids, Paris, 1971 (ed. by M. Balkanski), Flammarion Sciences, Paris, 1971, p. 502.
[24]V. V. Eremenko, Yu. A. Popkov, and V. I. Fomin, Tezisy dokladov 18 Vsesoyuznogo soveshchaniya po fizike nizkikh temperatur (Abstracts of Papers presented at Eighteenth All-Union Conf. on Low Temperature Physics), Physico-technical Institute of Low Temperatures, Academy of Sciences of the Ukrainian SSR, Kharkov, 1974, p. 203.
[25]B. V. Beznosikov and N. V. Beznosikova, Kristallografiya 13, 188 (1968) [Sov. Phys.-Crystallogr. 13, 158 (1968)].
[26]Yu. A. Popkov, V. I. Fomin, and B. V. Beznosikov, Pis'ma Zh. Eksp. Teor. Fiz. 11, 394 (1970) [JETP Lett. 11, 264 (1970)].
[27]V. V. Eremenko, Yu. A. Popkov, and V. I. Fomin, Proc. Second Intern. Conf. on Light Scattering in Solids, Paris, 1971 (ed. by M. Balkanski), Flammarion Sciences, Paris, 1971, p. 372.
[28]K. S. Aleksandrov and L. M. Reshchikova, Kristallografiya 14, 716 (1969) [Sov. Phys.-Crystallogr. 14, 614 (1970)].
[29]M. Furukawa, Y. Fujimori, and K. Hirakawa, J. Phys. Soc. Jpn. 29, 1528 (1970).
[30]V. G. Khlyustov, I. N. Flerov, A. T. Silin, and A. N. Sal'nikov, Fiz. Tverd. Tela (Leningrad) 14, 175 (1972) [Sov. Phys.-Solid State 14, 139 (1972)].
[31]O. Beckman and K. Knox, Phys. Rev. 121, 376 (1961).
[32]F. Borsa, Phys. Rev. B 7, 913 (1973).
[33]V. L. Ginzburg, Usp. Fiz. Nauk 77, 621 (1962) [Sov. Phys.-Usp. 5, 649 (1963)].
[34]A. P. Levanyuk, Fiz. Tverd. Tela (Leningrad) 5, 1776 (1963) [Sov. Phys.-Solid State 5, 1294 (1964)].
[35]A. P. Levanyuk, Zh. Eksp. Teor. Fiz. 49, 1304 (1965) [Sov. Phys.-JETP 22, 901 (1966)].
[36]L. A. Pozdnyakova, A. I. Kruglik, and K. S. Aleksandrov, Kristallografiya 17, 336 (1972) [Sov. Phys.-Crystallogr. 17, 284 (1972)].

Translated by A. Tybulewicz