Antiferromagnetic resonance and parametric excitation of spin waves in $CsMnCl_3$

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High-frequency properties have been studied in a single crystal of the easy-plane antiferromagnet $CsMnCl_3$ ($T_N = 67$ K, space group D^5_{3d}). The measurements were made over the frequency range $\nu = 9$–150 GHz, in magnetic fields H up to 50 kOe, at temperatures $T = 1.2$–4.2 K. The low-frequency branch of the antiferromagnetic resonance was investigated. The experimental results agree with the theoretical formula $(\nu/\gamma)^2 = H^2 + H_\Delta{}^2$ for the following values of the constants: $\gamma = 2.8$ GHz/kOe, $H_\Delta{}^2 = 11.4T^{-1}$ kOe^2. Parametric excitation of spin waves of the lower branch of the spectrum was accomplished by the method of parallel pumping. From the value of the threshold field, the damping of spin waves in $CsMnCl_3$ was calculated. It is shown that the principal mechanisms of relaxation under the conditions of this experiment are: scattering of magnons by domain walls, absorption at the specimen boundaries, decay of a magnon into a magnon and a phonon, and three-magnon relaxation. When the power greatly exceeded the threshold value, $P/P_c \gtrsim 70$, oscillations of the beyond-threshold susceptibility were observed; these disappeared subsequently, at $P/P_c \gtrsim 200$.

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Parametric excitation of electronic spin waves has so far been brought about and thoroughly studied in two antiferromagnetic materials (AFM): $MnCO_3$ and $CsMnF_3$. The present paper is devoted to an investigation of the high-frequency properties [antiferromagnetic resonance (AFMR) and parametric excitation of spin waves (SW)] in still another antiferromagnetic dielectric, $CsMnCl_3$, for the purpose of determining the parameters of the SW spectrum and explaining the mechanism of SW relaxation.

The structure of $CsMnCl_3$ can be described by a hexagonal crystal lattice with parameters $a_H = 7.288$ Å and $c_H = 27.44$ Å, or by a rhombohedral with parameters $a_R = 10.07$ Å and $\alpha = 42°26'$[1,2] (space group D^5_{3d}). From neutron-diffraction investigations[1] it is known that at temperature $T < T_N = 67$ K, $CsMnCl_3$ is an easy-plane antiferromagnet (EPAFM) in which the ferromagnetic plane is perpendicular to the high-order axis, and the elementary magnetic cell is obtained by doubling of the crystallographic and contains 18 Mn^{2+} ions.

If $H \ll (2H_AH_E)^{1/2}$ and $|\mathbf{k}| \ll \pi/a$, the two lower branches of the spin-wave spectrum (the quasiferromagnetic and the quasiantiferromagnetic) for an EPAFM can be described as follows:

$$(\nu_{1k}/\gamma)^2 = (H\cos\theta)^2 + H_\Delta{}^2 + \alpha_\perp{}^2 k_\perp{}^2 + \alpha_\parallel{}^2 k_\parallel{}^2, \quad (1)$$

$$(\nu_{2k}/\gamma)^2 = 2H_AH_E + (H\sin\theta)^2 + \alpha_\perp{}^2 k_\perp{}^2 + \alpha_\parallel{}^2 k_\parallel{}^2. \quad (2)$$

Here ν_{ik} and $\mathbf{k}$ are the frequency and the wave vector of the spin wave, H is the static magnetic field, γ is the gyromagnetic ratio, H_A is the plane-axis anisotropy field, H_E is the exchange field, α is the nonuniform-exchange constant, θ is the angle between the direction of H and the basal plane of the crystal, and H^2_Δ is the gap in the spectrum caused by hyperfine interaction or magnetoelastic interaction.

Study of $CsMnCl_3$ was also the subject of Refs. 3 and 4. Ref. 3 reports observation of AFMR ($k = 0$). The data of that paper corroborate that $CsMnCl_3$ belongs to the EPAFM. In addition, in Ref. 3 there was observed 180° anisotropy of the resonance field on rotation of the static field in the basal plane of the crystal. Ref. 4 investigated the static magnetic properties of $CsMnCl_3$. Values were obtained for the flip field in the basal plane, $H_{\text{flip}} = 526$ Oe (180° anisotropy) and for $(2H_AH_E)^{0.5} = 7.4$ kOe. According to formula (2), the latter quantity determines the gap in the spectrum of the quasiantiferromagnetic SW branch. Thus according to the data of Ref. 4, $\nu_{20} = 20.7$ GHz.

SPECIMENS AND METHOD OF MEASUREMENT

The single crystals of $CsMnCl_3$ were grown in the Institute of Physics, Siberian Branch, Academy of Sciences, USSR, by V. B. Beznosikov. The crystals were grown from the melt by the Bridgman method. The specimens that we used were optically transparent, of red color, with linear dimensions 1 to 3 mm. Orientation of the specimens was accomplished with x rays. Crystals of $CsMnCl_3$ are hygroscopic; therefore during storage they were placed in transformer oil, and during the measurements they were covered with Zapon lacquer.

For investigation of AFMR and of parametric excitation of spin waves, we used a microwave spectrometer similar to that described in Ref. 5. The specimen was placed at an antinode of the microwave magnetic field of a shorted waveguide or of a copper resonator with quality factor $Q \approx 2000$–12 000. The microwave power absorbed by the specimen was measured as a function of the value of the magnetic field and the temperature, at fixed pumping frequencies ν_p. The experiments were done over the frequency range $\nu_p = 9$–150 GHz, in a magnetic field up to 50 kOe, at temperatures $T = 1.2$–4.2 K.

ANTIFERROMAGNETIC RESONANCE

In the investigation of single crystals of $CsMnCl_3$, we detected resonance absorption corresponding to the low-frequency branch of the AFMR spectrum. The AFMR

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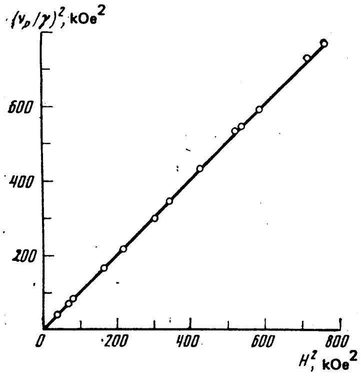

FIG. 1. Spectrum of the quasiferromagnetic branch of AFMR: $H \perp C_3$, $T=4.2$ K.

linewidth was 100–500 Oe. Figures 1 and 2 show the experimental relations $\nu_p(H)$ and $\nu_p(T)$ for the case in which the magnetic field **H** is parallel to the plane of easy magnetization (EP). When the static field was moved out of the EP, the resonance absorption line was displaced to higher fields, such that the projection of **H** on the EP remained constant. When **H** was rotated in the EP, no change of the position of the resonance line was observed. This means that H_{flip} does not exceed 0.5 kOe (see Ref. 4).

The experimental results (Figs. 1 and 2) are well described by formula (1) for $k=0$ with the following values of the constants: $\gamma=2.8$ GHz/kOe, $H_\Delta^2=(11.4\pm0.5)T^{-1}$ kOe^2. As was shown in Ref. 6, the inverse dependence of H_Δ^2 on temperature is characteristic of hyperfine interaction. In this case the value of H_Δ^2 can be expressed in terms of the effective field H_N of hyperfine interaction as follows:

$$H_\Delta^2=2H_EH_N. \quad (3)$$

From the value of the static susceptibility of $CsMnCl_3$, $\chi_\perp=2.0\times10^{-2}$ cgs emu/mol,[1)] we calculated the exchange field $H_E=700\pm70$ kOe, and then the effective field of hyperfine interaction $H_N=(8.1\pm1)T^{-1}$ Oe. This value is close to the values of H_N measured on other materials containing Mn^{2+} ions: $H_N=9.15T^{-1}$ Oe in $CsMnF_3$,[7] $H_N=9.1T^{-1}$ Oe in $MnCO_3$,[8] and $H_N=8.1T^{-1}$ Oe in $RbMnCl_3$.[9]

The high-frequency branch of AFMR in $CsMnCl_3$ was not detected at liquid-helium temperature, up to $\nu_p=150$ GHz. This deviates from the predictions made in Ref. 4 but does not contradict the data obtained in Ref.

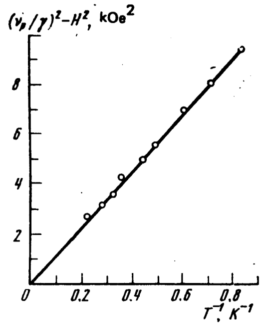

FIG. 2. Variation of the gap $H_\Delta^2=(\nu_p/\gamma)^2-H^2$ with temperature, in coordinates H_Δ^2 and $1/T$: $H\perp C_3$, $\nu_p=8.8$ GHz.

1. In Ref. 1, the dipole-anisotropy field was calculated theoretically and found to be $H_A=3$ kOe. By using the value of H_E given above, one can obtain an estimate of the gap of the high-frequency branch of the spectrum:

$$\nu_{20}=(2H_AH_E)^{1/2}\approx180 \text{ GHz}. \quad (4)$$

This value exceeds by a factor of about one and one half ν_{20} in the spectra of $CsMnF_3$ and $MnCO_3$.

PARAMETRIC EXCITATION AND RELAXATION OF SPIN WAVES

We succeeded in exciting spin waves in $CsMnCl_3$ by the method of parallel pumping.

1. The parametric excitation had a hard character: the onset of instability occurred when the amplitude of the microwave field exceeded a threshold value h_{c1}, the disappearance of instability at a field h_{c2}, where $h_{c1}>h_{c2}$. The order of magnitude of h_{c1} and h_{c2} is the same as for $MnCO_3$[10] and $CsMnF_3$[11], that is $h_{c1},h_{c2}\approx0.01-0.5$ Oe; but the following rule is observed: the ratio h_{c1}/h_{c2} remains practically constant at constant temperature, but with change of temperature from 1.2 to 4.2 K it decreases from 1.4 to 1.05.

2. The relaxation frequencies $\Delta\nu$ of excited SW were calculated from the value of the threshold field h_{c1} by formulas derived by Ozhogin.[12] The characteristic relation $\Delta\nu(H, T)$ is shown in Fig. 3. By comparison with the relaxation of SW in $CsMnF_3$[11] and $MnCO_3$,[5] we observe here a weaker dependence of $\Delta\nu$ on the magnetic field and the temperature; this is apparently due to the following circumstances.

As is well known, in $CsMnF_3$ and $MnCO_3$ a substantial contribution to the scattering of magnons at a temperature above 1.5 K is made by a three-magnon relaxation process ($\Delta\nu_{3M}$): fusion of two magnons of the lower branch of the spectrum, with formation of a magnon of the upper branch.[13] In $CsMnCl_3$, the larger gap in the spectrum of the second branch makes this process less effective, and therefore other relaxation processes play a greater role. The data are insufficient for accurate theoretical calculation of the relaxation frequency of SW in $CsMnCl_3$; in particular, there are no values of ν_{20} and of the magnon-phonon interaction interaction constant. But it is possible to distinguish the relaxation mecha-

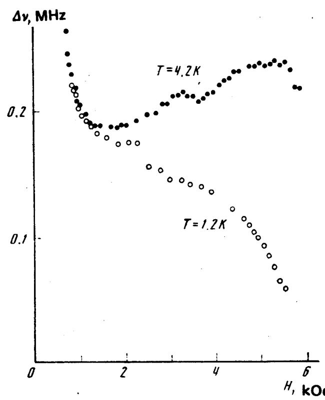

FIG. 3. Relaxation frequency of SW as a function of field and temperature: $\nu_p=35.8$ GHz.

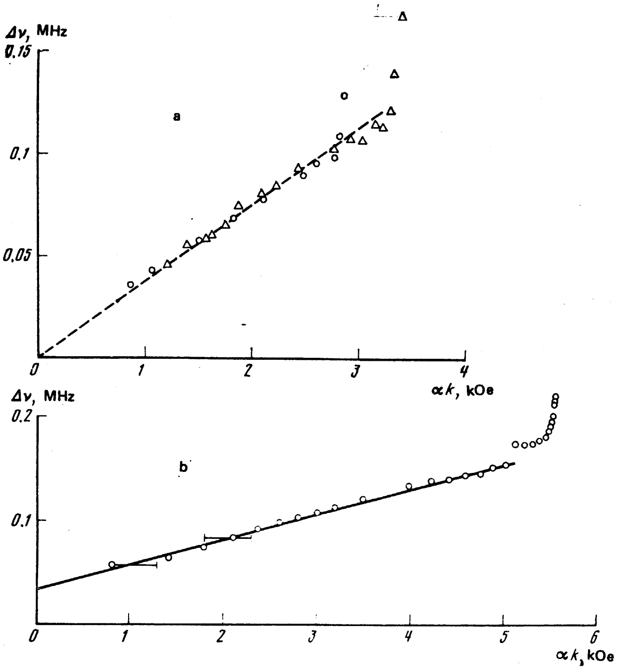

FIG. 4. Relaxation frequency of SW as a function of the value of αk: a) $T=1.2$ K, $\nu_p=35.8$ GHz; b) $\nu_p=23.5$ GHz, $T=1.2$ K for points ○, $T=2.13$ K for points Δ.

nisms on the basis of the different dependencies of $\Delta\nu$ on T, H, and k predicted by the theory (of course each of the processes can occur only in a certain range of variation of the parameters, which is determined by the laws of conservation of energy and momentum of the quasiparticles).

1) On decrease of H below 1.5 kOe, an abrupt rise of the relaxation frequency is observed. This may be due to the fact that at small fields the specimen is not saturated, and scattering of SW by domain walls makes an appreciable contribution. From a comparison of Figs. 3 and 4a it follows that the increase of $\Delta\nu$ is determined by the value of the magnetic field, and not by the value of the wave number k. This also agrees with the assumption made about scattering of SW by domain walls.

2) Figure 4 shows the variation of the damping frequency with the value of k. At low temperatures ($T < 3.0$ K), there is a considerable interval of k in which $\Delta\nu(k)$ can be described by a linear function. At these temperatures, apparently, an appreciable contribution to the relaxation is made by absorption of spin waves at the boundaries, $\Delta\nu_{\mathrm{def}}$. The proportionality of $\Delta\nu_{\mathrm{def}}$ to the value of k is easily obtained[10] by prescribing the free-path length L of a magnon:

$$\Delta\nu_{\mathrm{def}}=\frac{\partial\nu_k/\partial k}{L}=\frac{2\gamma^2\alpha^2}{\nu_p L}k. \qquad (5)$$

Knowing H_E and the lattice parameters, we can calculate[14] the values of $\alpha_\parallel$ and $\alpha_\perp$:

$$\alpha_\parallel=3.18\cdot10^{-5}\ \mathrm{kOe\cdot cm},\quad \alpha_\perp=2.08\cdot10^{-5}\ \mathrm{kOe\cdot cm}, \qquad (6)$$

and then, by using the experimental data and the relation (5), can obtain the free-path length of a magnon. In our case, $L\approx2$ to 4 mm; that is, it is comparable with the linear dimensions of the specimen.

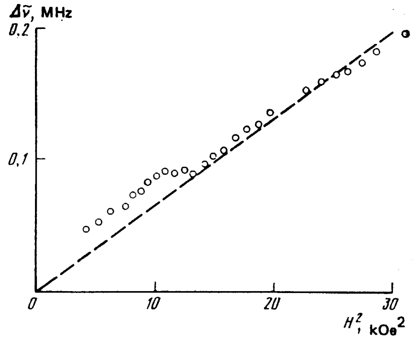

FIG. 5. Variation of the value of $\Delta\tilde{\nu}$ with H^2: $T=4.2$ K, $\nu_p=35.8$ GHz.

3) At higher temperatures, $\Delta\nu(k)$ departs noticeably from the linear law, and this departure increases with increase of H and of T. Figure 5 shows the variation of the additional contribution

$$\Delta\tilde{\nu}=\Delta\nu(T=4.2\ \mathrm{K})-\Delta\nu(T=1.2\ \mathrm{K})$$

with the value of the magnetic field, for the highest temperature in our experiment, $T_{\max}=4.2$ K. The quadratic dependence of the relaxation frequency on H is characteristic of three-magnon attenuation.[13] Also characteristic of the three-magnon relaxation mechanism is an exponential decrease of $\Delta\nu_{3M}$ with temperature. A deviation of the $\Delta\nu(k)$ relation from a linear law is clearly observable in the narrow temperature interval 3.5–4.2 K, and therefore the accuracy of our experiment was not sufficient for determination of the index of the exponential. Theoretical estimates give, for example, the following result: for an EPAFM with $H_\Delta^2=11.4\ T^{-1}$ kOe^2 and $\nu_{20}=180$ GHz, at $\nu_p=35.8$ GHz and $H=5$ kOe, the value of $\Delta\nu_{3M}$ decreases by a factor ≈11.5 on decrease of temperature from 4.2 to 3.0 K, and under the conditions of our experiment it should amount to no more than 7% of $\Delta\nu$ at $T=3.0$ K. Our experimental results are not contradictory to this statement.

4) From Fig. 5 it is evident that the plotted $\Delta\tilde{\nu}(H^2)$ relation cannot be described by just one three-magnon relaxation process. The most important deviation from $\Delta\nu_{3M}$ is observed at field $H_u=3.6$ kOe. This singularity, which has the form of a step, is clearly evident also in Figs. 3 and 4b: at a certain value H_u, the relaxation changes discontinuously by an amount $\Delta\nu_u=0.02\pm0.01$ MHz. If the specimen temperature changes at constant pumping frequency, the position of the step (the value of H_u) changes in such a way that k_u of a magnon remains constant. This phenomenon can be explained by supposing that some magnon relaxation process is permitted at large k and forbidden at $k<k_u$. Analysis of the laws of conservation of energy and momentum for the various processes of relaxation of SW in EPAFM shows that this property may be possessed by processes in which phonons participate:

a) If the velocity of sound u is larger than the limiting velocity of spin waves $v=2\pi\alpha\gamma$ ($u>v$), then fusion of a magnon with a phonon, with formation of a phonon, $m+ph\to ph$ (I), will occur from $H=0$ to H_{uv}, where H_{uv} is the field at which the SW spectrum intersects the phonon

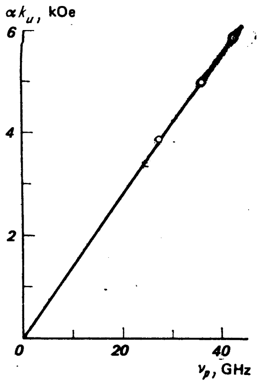

FIG. 6. Variation of αk_u with ν_p.

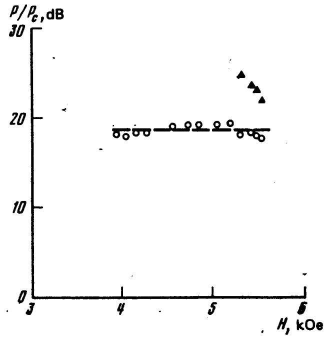

FIG. 8. The excess of the microwave power above the threshold, at which oscillations appear (○) and disappear (▲), as a function of the magnetic field H; $T=1.2$ K, $\nu_p=35.8$ GHz.

spectrum for a given value of ν_p.

b) If $u<v$, then the magnon and phonon spectra do not intersect, but the required property is possessed by the disintegration process $m \to m+ph$ (II). The limit of this process is determined by the condition

$$u=v_{\lim}=2\pi\partial\nu_k/\partial k, \qquad (7)$$

process II is permitted when $u<v_{\lim}$.

In order to establish which of these cases occurs, it is obviously necessary to know the velocities of phonons and magnons. But certain facts enable us to give preference to disintegration II, despite the absence of data on the velocity of sound. The participants in fusion I are thermal phonons, whose number depends sharply on temperature; consequently the relaxation due to process I should also rise rapidly with temperature. In our experiments the temperature changed by a factor 3.5, but the value $\Delta\nu_u=0.02\pm0.01$ MHz was independent of temperature.

It may thus be assumed that in $CsMnCl_3$, $u<v$ and the step in $\Delta\nu_u$ is due to process II. The condition (7) can be written in the form

$$u=\frac{4\pi\alpha\gamma^2}{\nu_p}\alpha k_u. \qquad (8)$$

Figure 6 shows experimental values of the quantity αk_u as a function of the pumping frequency ν_p. The straight line drawn through them corresponds to $u=2.9\times10^5$ cm/sec in the case $\alpha=\alpha_\perp$ and to $u=4.4\times10^5$ cm/sec if $\alpha=\alpha_\parallel$.

5) The contribution to SW relaxation that is independent of H and k may be attributed to magnon-phonon interaction $m+ph\to m$ (III). The amplitude of this process in the case $u<v$ was calculated in Ref. 15. Estimates of $\Delta\nu_{m-ph}$ give the value 0.01 MHz at $T=1.2$ K and $\nu_p=35.8$ GHz.

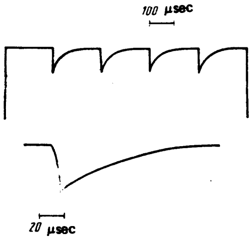

FIG. 7. Schematic representation of the oscillations of the beyond-threshold susceptibility on the microwave pulse passing through the resonator. Below, the oscillations on an enlarged scale.

Approximately the same result is obtained when the experimental $\Delta\nu(\alpha k)$ data (Fig. 4a) are extrapolated by a straight line to $k=0$. Unfortunately we were unable to observe any regularity in the change of behavior of this part of the relaxation with pumping frequency and temperature. Perhaps this damping depends on mechanical stresses in the crystal, caused by attachment of the specimens to the substrate. We made no special investigation of this process. Thus it cannot be asserted that the process observed in our experiments was actually (III).

3. We also carried out an investigation of the dependence of the threshold microwave field on the direction of the field **H** in the EP at $\nu_p=25$ GHz. In these experiments, the parallel-pumping condition $\mathbf{h}\parallel\mathbf{H}\perp C_3$ was satisfied, and the crystal was rotated about the C_3 axis. In contrast to $CsMnF_3$,[16] the value of the threshold field in $CsMnCl_3$ did not depend on the rotation of the crystal. For each particular specimen (of any form), $h_c(\varphi)=\mathrm{const}$ with accuracy 2%.

4. When the threshold microwave power is greatly exceeded ($P/P_c>70$), the oscillogram of the microwave pulse passing through the resonator shows oscillations of the signal (Fig. 7) similar to the oscillations in CsMnF3.[17] The measurements showed that at constant temperature, the appearance of oscillations occurs at the same supercriticality, independently of the value of the static magnetic field. If, at constant field H, the ratio P/P_c is increased, the amplitude of the oscillations and the period of their succession τ increase. The value of τ varies over the range 100–300 μsec. When $P/P_c>200$, there appears on the microwave pulse a new split (similar to that at field h_{c1}), and the oscillations disappear (Fig. 8). The cause of the occurrence of oscillations of the beyond-threshold susceptibility in parametric excitation of spin waves is at present unknown. A discussion of possible mechanisms of this phenomenon is contained in Ref. 17.

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[1)]The static susceptibility of $CsMnCl_3$ was measured at our request by A. N. Bazhan at the Institute of Physical Problems, Academy of Sciences, USSR. The authors express their deep gratitude to A. N. Bazhan.

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Nonphonon branches of the Bose spectrum in the *B* phase of systems of the He^3 type

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We investigate all 18 branches of the Bose spectrum in the He model, which are of the form $E^2 = \Omega^2 + \alpha k^2$. The frequencies $\Omega = 0$, $(8/5)^{1/2}\Delta$, $(12/5)^{1/2}\Delta$, and 2Δ and the coefficients α are calculated for all branches. It is shown that the branches with $\Omega = 2\Delta$ have complex dispersion coefficients α.

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1. INTRODUCTION

The Bose spectrum in the superfluid phases of He^3 consists of 18 branches (18 is the number of degrees of freedom of the complex tensor order parameter $A_{ij}, i,j=1,2,3$). In the B phase of He^3 there are four phonon (Goldstone) branches-one acoustic, one longitudinal-spin-wave, and two transverse-spin-wave. The remaining 14 branches have in the B phase a gap at $k=0$ and constitute different oscillations modes of the self-consistent field. At small k these branches are of the form

$$E^2(\mathrm{k}) = \Omega^2 + \alpha k^2. \tag{1.1}$$

The frequencies Ω were calculated in Refs. 1–5. The case of nonzero k was considered in Ref. 1 with the aid of the kinetic equation and in Ref. 2 using the Bethe-Salpeter equation. The branches with $\Omega = 2\Delta$, however, as well as some branches with $\Omega = (8/5)^{1/2}\Delta$, $(12/5)^{1/2}\Delta$ were not investigated accurate to k^2.

In this paper we investigate all the nonphonon branches of the Bose spectrum at small k in the B phase of the He^3 model. The model is defined by a "hydrodynamic action" functional obtained by functional integration with respect to the Fermi fields.[6] In first-order approximation, the spectrum of the Bose excitations is given by the quadratic part of the functional. Investigation of this part makes it possible to calculate the frequencies Ω and the coefficients α in (1.1) for all 14 nonphonon branches. We take special notice of the fact that the coefficients α corresponding to the four branches with $\Omega = 2\Delta$ turn out to be complex (and different for the different branches).

The plan of the paper is the following. In Sec. 2 we