Parametric excitation of nuclear spin waves in antiferromagnetic $MnCO_3$ crystals

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Parametric excitation of nuclear spin waves in the antiferromagnetic crystal $MnCO_3$ is studied at 1080 MHz by the parallel pumping technique. Two threshold processes are observed in the experiments. One refers to spin wave excitation in a nuclear magnetic system and the other to excitation of magnetoelastic waves. The post-threshold sample susceptibility in such processes is studied. A very pronounced overheating of the nuclear magnetic system of the sample with respect to the lattice is observed after the second threshold. The nature of these overheating phenomena shows that two magnetoelastic oscillation branches are excited in the second threshold process. The dependence of the threshold field on the wave vector is more complicated in a small magnetic field because of magnetization processes in the sample. In a large magnetic field, complications are produced by the magnetoelastic coupling.

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INTRODUCTION

The study of parametric excitation of spin waves in magnetically ordered systems by the method of parallel pumping is of definite interest. This is one of the few methods which allows us to study spin waves over a wide range of wave vectors with simply operated apparatus. In several cases, it has been possible to obtain absolute values of the wave vectors of the magnons, for example, in $CsMnF_3$ and $MnCO_3$, [1,2] where the intersection of the spectrum of antiferromagnetic magnons with the phonon branch is observed. In the present work, parallel pumping has been studied in a system of nuclear spin waves of an antiferromagnet with weak ferromagnetism, $MnCO_3$. The first idea of nuclear spin waves, which follows from the Suhl-Nakamura interaction, [3,4] was advanced in the work of de Gennes *et al.* [5] The observation of parametric excitation of nuclear spin waves was carried out in the works of Adams, Hinderks and Richards [6] in $CsMnF_3$ and $RbMnF_3$, and of Yakubovskiĭ [7] in $MnCO_3$.

The object of this investigation is $MnCO_3$, a well-studied antiferromagnet with weak ferromagnetism. Manganese carbonate has a rhombohedral structure; the plane perpendicular to the threefold axis is the easy plane for the magnetic system of the sample. The spectra of electron antiferromagnetic and nuclear resonances under the conditions of strong Suhl-Nakamura interaction have been studied in the works of Borovik-Romanov, Kreĭnes and Prozorova [8] and Borovik-Romanov and Tulin. [9,10] The low-frequency branches corresponding to these resonances are of the form

$$\omega_{ek}^2=\gamma_e^2[H(H+H_D)+H_\Delta^2+\alpha^2k^2], \qquad (1)$$
$$\omega_{nk}^2=\omega_{n0}^2(1-\gamma_e^2H_\Delta^2/\omega_{ek}^2), \qquad (2)$$

where γ_e is the gyromagnetic ratio for the electron, ω_{ek} and ω_{nk} are the frequencies of the electron and nuclear spin waves, respectively, H is the external magnetic field, $H_D = 4.4$ kOe is the effective Dzyaloshinskiĭ interaction field, H_Δ^2 is the gap in the spectrum of the electron system, due to the hyperfine interaction α^2k^2 is the spatial dispersion term, and $\omega_{n0}/2 = 640$ MHz is the frequency of nuclear magnetic resonance in the hyperfine field of electrons in the absence of dynamic coupling.

As is seen from expression (2), the shift in the frequency of nuclear spin wave from ω_{n0} for certain k is determined by the frequency of the corresponding oscillation of the antiferromagnetic system. For $k=0$, the shift in frequency of the nuclear system will be a maximum in a constant magnetic field; when $k\to\infty$ the frequency shift approaches zero in view of the relative smallness of the term $\gamma_e^2H_\Delta^2$.

EXPERIMENT

As is seen from the foregoing, the spectrum of nuclear spin waves is located in the frequency range below 640 MHz. The minimum frequency corresponds to $k=0$ and is determined by the external magnetic field. To observe the parametric excitation of spin waves it is necessary to apply a high-frequency magnetic field at double the frequency of the spin-wave resonance. The polarization of the high-frequency field should be parallel to the constant magnetic field. In the experiment, the susceptibility of the $MnCO_3$ sample at a frequency of 1080 MHz was studied in the direction parallel to the constant magnetic field. The samples of $MnCO_3$ were single crystals, with well-pronounced faceting, in the form of plates, the plane of which coincides with the easy plane of the anisotropy. The high-frequency and static magnetic fields were parallel to this plane ($\mathbf{H} \parallel \mathbf{h} \perp C_3$). A pass-through spectrometer was used with an absorption cell in the form of a helical resonator, made of aluminum wire. The sample was placed inside the helix in the antinode of the magnetic field. The coupling was provided by posts extending to the ends of the helix. The power passed through the resonator to a decimeter-band receiver. The power fed to the resonator was regulated by an attenuator. At the input of the receiver there was another attenuator. In the course of operation, these two attenuators were used to vary the power fed to the resonator and to maintain the power to the receiver constant. Thus, in the steady state, the susceptibility parallel to the magnetic field at 1080 MHz was studied as a function of the magnetic field at various levels of the high-frequency field in the resonator.

The traditional pulse method did not allow us, under our conditions, to observe a number of details in the parametric excitatiom. Therefore, it was used sporadically to study the post-threshold oscillations of the susceptibility.

All the experimental results were obtained at a temperature of 1.5 °K.

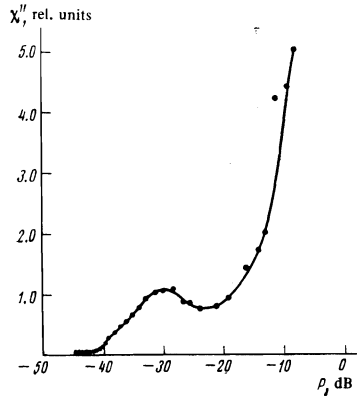

FIG. 2. Dependence of the imaginary part of the post-threshold susceptibility of the sample on the value of the damping of the input attenuator; $T = 1.5$ °K, $H = 1320$ Oe.

EXPERIMENTAL RESULTS

The characteristics of the passage of power through the resonators were measured as functions of the magnetic field for different power levels. With account of the filling factor, these characteristics are connected with the absorption of high-frequency power in the sample. Several absorption curves are shown in Fig. 1, the increase in the number of the curves corresponding to increase in the high-frequency power in the resonator. The curves are displaced along the vertical axis for convenience. Curve 1 was obtained for a small value of power. A monotonic increase of the absorption is observed in it upon decrease in the magnetic field H, owing to the processes of magnetization reversal of the antiferromagnetic system of the sample. This form of absorption is evidently a linear process and it is a characteristic of antiferromagnets with weak ferromagnetism. Curve 3 reveals the appearance of additional absorption in the magnetic field, corresponding to the resonance field at half the pump frequency. This value of the power will correspond in what follows to threshold for the magnetic field near $H = H_{res}$. Further development of the process consists in the propagation of the absorption into the region of smaller values of the magnetic field and in a rapid increase in the susceptibility of the sample (curves 4, 5). The susceptibility passes through a maximum and then decreases. The character of the change in the imaginary part of the susceptibility in a magnetic field $H = 1320$ Oe is shown in Fig. 2. Upon further increase in the power of the high-frequency field in the region of H_{res}, still another absorption appears (curve 6, Fig. 1); the range of this absorption broadens rapidly and a corresponding increase in the susceptibility results. Beginning with curve 7 of Fig. 1, when the absorption of the high-frequency power in the sample is extended through the entire range of the magnetic field from H_{res} to zero, the absorption curve takes on a hysteretic character upon increase and decrease of the magnetic field. The hysteresis curve is shown in Fig. 3. Thus, two threshold processes are observed in the experiment, the beginnings of which correspond to the curves 3 and 6 of Fig. 1. The distinguishing feature of these processes is the fact that on passing through the first threshold process (corresponding to the smallest pump power) no significant extension of the absorption is observed into the region of fields greater than H_{res}, while after the second process the absorption spreads significantly into the region of large fields. The presence of hysteresis in this second process is in some sense accidental and is determined by the value of the absorption of power in the sample. Further increase in the power leads to a splitting of the maximum of the absorption near H_{res}, and the location of the maximum of the absorption in the smallest magnetic field depends strongly on the amplitude of the high-frequency field (Fig. 4). The next stage of increase in the power leads to a smearing out of all the singularities and to the appearance of instability (sharp jumps in the absorption), which is evidently connected with the disruption of the

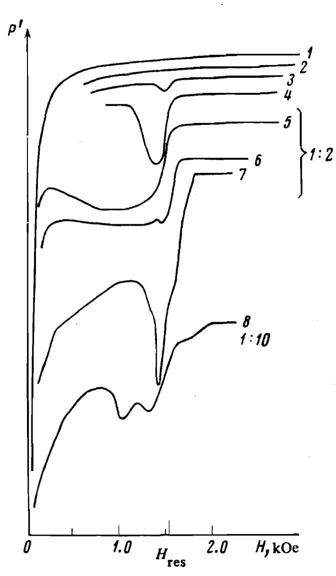

FIG. 1. Passage of the high-frequency power through a resonator with the sample. P'—power fed to the input of the receiver. Curves 1–8 correspond to the following values of the damping of the input attenuator: 1)—46, 2)—44.2; 3)—43.5; 4)—39.35; 5)—28.1; 6)—23.8; 7)—16.2; 8)—7.65 dB.

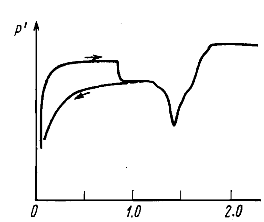

FIG. 3. Absorption of high-frequency power in the sample as a function of the magnetic field in the case of the development of the phenomenon of overheating of the nuclear system. P'—power applied to the input of the receiver. The arrows indicate the direction of the sweep of the field.

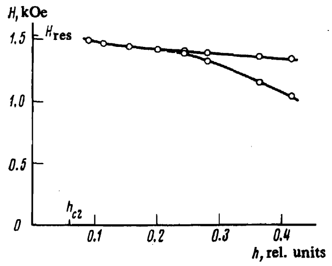

FIG. 4. Absorption of the maxima of the absorption of high-frequency power as a function of the amplitude of the high-frequency field on the sample after the second threshold.

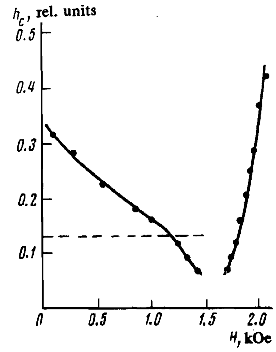

FIG. 6. Dependence of the critical amplitude of the high-frequency field on the static magnetic field for the second threshold process. The horizontal dashed line indicates the value of the amplitude of the high-frequency field above which the phenomena of overheating are observed.

heat transfer regime between the sample and the liquid helium bath.

From the observed curves of the transmission of high-frequency power as a function of the external magnetic field (Fig. 1), we can construct the dependence of the amplitude of the threshold field h_c on the value of the static magnetic field H. For this purpose, the values of the magnetic field which limit the excess absorption curve are found on the experimental curves and, as against these values we plot the amplitude of the high-frequency field corresponding to the given curve. The results of the construction are shown in Figs. 5 and 6. The points referring to the second threshold in small magnetic fields (above the dashed straight line in Fig. 6) were constructed according to the location of the jump in the absorption on the curves similar to those of Fig. 3. The interpretation of these curves must be made carefully, since in the case of advanced overheating the increase in the absorption in the sample can be due to some other phenomenon, as is observed in the excitation of homogeneous resonance at large detunings.[10]

DISCUSSION OF RESULTS

Parallel pumping in the system of nuclear magnons of $MnCO_3$ exhibits a number of singularities. The frequency range of its observation is bounded from above at double the frequency of the undisplaced nuclear resonance: $2\omega_{n0}/2 = 1280$ MHz. The lower bound is at double the resonance frequency of the nuclear system, for which the resonance magnetic field is comparable with the real width of the NMR line. The existing inhomogeneous NMR line broadening extends also to spin waves, i.e., for the same energy of spin waves there is a wide choice of wave vectors **k**. In this case, we can assign any convenient value to the wave vector of the spin wave. In the present work, we used a pump frequency of 1080 MHz; the resonance field corresponding to one half the frequency amounted to 1520 Oe in the case of a line width of the order of 100 Oe.

Still another feature is associated with the phenomenon of overheating in the nuclear system, namely, the frequency of the nuclear spin waves depends strongly on the temperature of the nuclear magnetic subsystem of the sample, which is not difficult to overheat by means of easily achievable power.[10] A third feature is the strong magnetoelastic interaction in $MnCO_3$ at frequencies in the investigated range (see the work of Gakel'[11]).

In view of the presence of the latter feature, the spectrum of interacting magnons and phonons takes the form represented in Fig. 7 by the continuous curves. This corresponds to the formation of shifted magnetoelastic branches of the spectrum which, far from the point of intersection, represent as before almost pure magnons and phonons, and in the intersection region, magnetoelastic waves.

In parallel pumping, when the acting force is the magnetic field, it is natural to assume that the magnons are easily excited and the magnetoelastic waves with a small admixture of magnetic oscillations more difficult. Then the qualitative dependence of the amplitude of the critical field on the value of the static field for the branches shown in Fig. 7 will have the form shown in Fig. 8 (curves 1, 2). There is a strong inhomogeneous broadening of the spectrum of nuclear spin waves in the investigated samples. This is tenta-

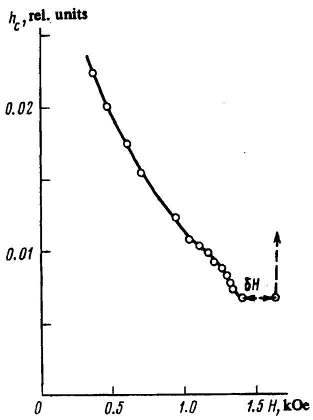

FIG. 5. Dependence of the critical amplitude of the high-frequency field on the static magnetic field for the first threshold process. H—order of the inhomogeneous NMR broadening.

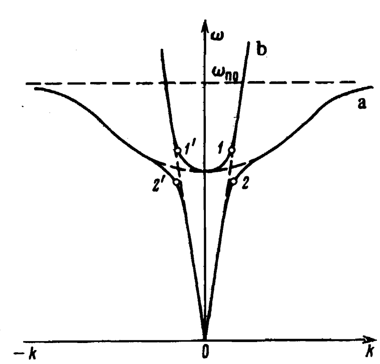

FIG. 7. Form of the spectra of coupled magnetoelastic oscillations of the nuclear magnons (a) and a single branch of phonons (b). The points 1, 1′ and 2, 2′ are pairs of quasiparticles with equal but oppositely directed wave vectors k and −k.

tively represented in Fig. 8 in the form of a set of plots of h_c as a function of H with different values of the resonance fields H_{res}. Such a representation enables us to explain the peak in the critical field h_c in the interaction of magnons with any nonmagnetic systme, and the presence of such a peak indicates the existence in this region of still another threshold process with a large value of the critical field. Near the point of interaction in a neighborhood of the order of the inhomogeneous broadening, the character of the dependence of the post-threshold susceptibility on the pump power $\chi''(P)$ will bring into the process newer and newer regions of the crystal. Thus the process of parametric excitation that we have observed can describe both the excitation of purely magnon parts of the two branches of the spectrum shown in Fig. 7 (the first threshold) and the excitation of the magnetoelastic portions of these branches in the region of intersection (the second threshold).

Figure 9 represents the dependence of the amplitude of the critical high-frequency field on the quantity $\mathscr{H} = [H_{res}(H_{res} + H_D) - H(H + H_D)]^{1/2}$, which is proportional to the wave vector of the spin wave for the first threshold, which we shall connect with the excitation of magnons. It is seen from this drawing that the inhomogeneous broadening corresponds to a wide range of wave vectors (the value of the wave vector of the first point is $k \approx 10^5$ cm^{-1}), i.e., even at the excitation frequency of 1080 MHz a large indeterminacy remains in the wave vector of the nuclear spin wave. A similar indeterminacy remains also for the relaxation frequency of the magnons, if we reduce the curve by means of the formula

$$h_c^{nn} = \frac{4\eta_{nk}\omega_{ek}^2}{\gamma_e^2(H_D+2H_0)\omega_{nk}}\left(\frac{\omega_{ek}^2}{\gamma_e^2 H_\Delta^2} - 1\right),$$

where η_{nk} is the relaxation frequency of nuclear spin waves. [7,12] Attention is called to the fact of the similarity of the dependence of the critical field amplitude in the case of parallel pumping in a system of nuclear magnons (Fig. 5 in this work) and the dependence of the critical field amplitude of the excitation of nuclear resonance in $MnCO_3$ at large detunings. [10] The con-

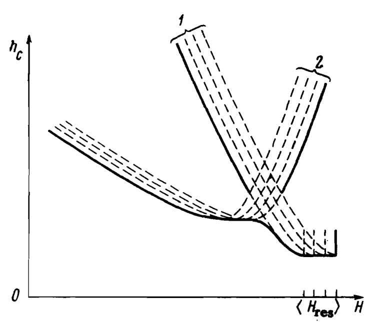

FIG. 8. Qualitative picture of the behavior of the amplitude of the critical high-frqeuency field as a function of a constant magnetic field. The dashed curves refer to the magnetoelastic branches in the absence of inhomogeneous broadening, but with different H_{res}. The solid curves correspond to the measured values of the threshold field in the case of inhomogeneous broadening of the spin wave spectrum.

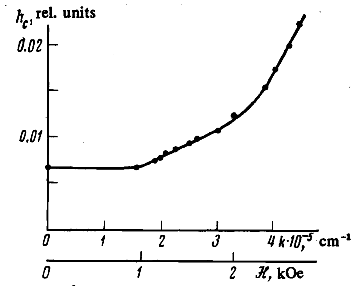

FIG. 9. Dependence of the critical high-frequency field of parametric excitation of magnons (first threshold) on the quantity $\mathscr{H} = [H_{res}(H_{res} + H_D) - H(H + H_D)]^{1/2} \sim k$. The scale of k is referred to the velocity of antiferromagnetic spin waves $v = 1.07 \times 10^5$ cm/sec.

clusion can be drawn that the excitation of homogeneous resonance at large detuning can be realized via the nuclear spin wave.

As the susceptibility of the nuclear system in parallel pumping grows and the amplitude of the high-frequency field increases, the nuclear system begins to overheat relative to the lattice. Significant overheating is observed in the range of amplitudes of the second critical field, above the dashed straight line of Fig. 6. The experimental manifestations of overheating in the case of parallel pumping are similar to those in the case of homogeneous resonance. [10] The post-threshold susceptibility is observed in the interval of fields from H_{res} to $H = 0$ and the relation $\chi''(H) \sim P'$ has a strong hysteresis when the direction of the sweep of the field is changed (Fig. 3).

The overheating of the nuclear magnetic system takes place at amplitudes of the high-frequency field for which a strong maximum of the post-threshold susceptibility is observed near H_{res}. The behavior of this maximum with overheating allows us to confirm the fact that the second threshold corresponds to the excitation of magnetoelastic waves of two branches of the spectrum (Fig. 7). It is seen from Fig. 8 that simultaneous excitation of the magnetoelastic waves of the two branches takes place in the region of intersection. It turns out that these parts of the branches behave in different fashion upon overheating.

The heating of a nuclear magnetic system leads to a shift in the spin-wave spectrum of the magnetic system that does not interact with the phonons along the frequency axis to ω_{n0}. For magnetoelastic waves at the points 1 and 1' in Fig. 7, the heating brings these points close to the magnon branch, which leads to a decrease in the threshold and to an increase in the post-threshold susceptibility at a fixed frequency and pumping level. This produces additional power absorption in the nuclear system and a greater degree of its heating, which in turn increases the absorption of power in the form of an increase in the susceptibility (there is a positive inverse connection between the absorption and heating). The maximum absorption in this case is transferred to a smaller field in this case, corresponding to an increase in the temperature of the nuclear system. At points 2 and 2' the situation is reversed; the heating leads to the result that these points approach the purely phonon part of the spectrum, with corresponding decrease in the susceptibility in view of the increase in the threshold. The inverse coupling in this case will be negative and the location of the maximum absorption will be stable. The experimentally observed behaviors of the absorption maximum—splitting and the dependence of the location of the split maxima on the amplitude of the high-frequency field (Fig. 4)—confirms these considerations.

Thus, the rather complicated properties observed in parallel pumping in $MnCO_3$ at a frequency near 1000 MHz are qualitatively described by the excitation of nuclear magnons and magnetoelastic waves with account of the phenomenon of overheating of the nuclear magnetic system of the sample.

[1]M. H. Seavey, Phys. Rev. Lett. 23, 132 (1969).
[2]V. V. Kveder, B. Ya. Kotyuzhanskiĭ and L. A. Prozorova, Zh. Eksp. Teor. Fiz. 63, 2205 (1972) [Sov. Phys. JETP 36, 1165 (1972)].
[3]H. Suhl, Phys. Rev. 109, 606 (1958).
[4]T. Nakamura, Prog. Theor. Phys. 20, 542 (1958).
[5]P. G. de Gennes, P. Pincus, F. Hartmann-Boutron and T. M. Winter, Phys. Rev. 129, 1105 (1969).
[6]B. T. Adams, L. W. Hinderks and P. M. Richards, J. Appl. Phys. 41, 931 (1970).
[7]A. Yu. Yakubovskiĭ, Zh. Eksp. Teor. Fiz. 67, 1539 (1974) [Sov. Phys. JETP 40, 766 (1975)].
[8]A. S. Borovik-Romanov, N. M. Kreĭnes and L. A. Prozorova, Zh. Eksp. Teor. Fiz. 45, 64 (1963) [Sov. Phys. JETP 18, 46 (1964)].
[9]A. S. Borovik-Romanov and V. A. Tulin, Pis'ma Zh. Eksp. Teor. Fiz. 1, No. 5, 18 (1965) [JETP Lett. 1, 134 (1965)].
[10]V. A. Tulin, Zh. Eksp. Teor. Fiz. 55, 831 (1968) [Sov. Phys.-JETP 28, 431 (1969)].
[11]V. R. Gakel', Zh. Eksp. Teor. Fiz. 67, 1827 (1974) [Sov. Phys. JETP 40, 908 (1975)].
[12]V. I. Ozhogin and A. Yu. Yakubovsky, Phys. Lett. 43A, 505 (1973).

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The role of drag effects in pure superconductors

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The effect of drag processes on the thermal conductivity of superconductors is investigated. It is shown that for normal metals and for superconductors in the region $\beta < 1$, the correction to the phonon distribution function is completely determined by drag effects. In the region $\beta > 1$, the solution is identical with that obtained by Gurevich and Krylov {Zh. Eksp. Teor. Fiz. 68, 1337 (1975) [Sov. Phys. JETP 41, 665 (1975)}. The influence of boundaries, impurities and defects is considered, and it is shown that drag effects are always a correction to the total thermal conductivity in the parameter Θ_D/ϵ_F.

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1. THE KINETIC EQUATION FOR EXCITATIONS

For relaxation processes in pure superconductors, the principal role is played by the electron-phonon interaction. In this case the exact solution of the problem requires the consideration of a system of kinetic equations for phonon and electron excitations. Account of drag processes corresponds to account of nonequilibrium corrections to the phonon distribution function in the kinetic equation for electron excitations, and of corrections to the electron distribution function in the equation for the phonons. The electron thermal conductivity $\varkappa_e$, which is connected with the scattering of the electrons by phonons without account of drag effects, was considered earlier by a number of authors.[1,2] The phonon thermal conductivity $\varkappa_{ph}$, which is connected with the scattering of phonons by electrons ($\varkappa_{ph\,e}$) for the case of dirty superconductors, when the phonon thermal conductivity can be comparable with the electron conductivity not only near $T=0$, was considered in Ref. 3. However, the account of drag effects in the case of pure superconductors can bring about a significant contribution to $\varkappa_e$ as $T \to 0$ and to $\varkappa_{ph}$ at $T \sim T_c$; in this case, however, the contribution of drag effects to the total thermal conductivity is small, because of the smallness of the ratio Θ_D/ε_F (or s/v_F). It can be shown that the Umklapp processes make a small contribution and can be disregarded.

In a recent work, Gurevich and Krylov[4] showed that account of nonequilibrium phonons begins to play a decisive role in the determination of the electron distribution function at sufficiently low temperatures, and the possibility of experimental testing of this fact is predicted. The analysis was carried out in an approximation in which the nonequilibrium contributions to the electron distribution function in the kinetic equation for phonons were not taken into account. However, the discarded terms are not small and are decisive in the case of normal metals and in a whole range of temperatures for superconductors. Moreover, it should be noted that the total momentum of the electrons and phonons should be conserved. Mathematically, this