

Nonequilibrium states in ferromagnetic substances upon nonresonant excitation of spin waves by an alternating magnetic field

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A ferromagnet subjected to the action of a parametrically nonresonant high-frequency alternating magnetic field ($\tau^{-1} \ll \Omega < 2\varepsilon_0 \varepsilon_k$ is the energy of a magnon with wave vector \mathbf{k} , Ω is the frequency of the field, and τ is the magnon relaxation time) applied parallel to the magnetization axis is considered. It is shown that such a field leads to suppression of the magnetization of an ideal ferromagnet. The imaginary part of the high-frequency susceptibility is calculated. This susceptibility is due to both magnon-magnon and magnon-impurity collisions.

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Great interest has appeared recently in solid state and plasma physics in the investigation of the nonequilibrium states of quasiparticles, when the disequilibrium is produced by action on the system of a strong nonresonant high-frequency field (see, for example, Refs. 1-5). In the work of Zel'dovich and Raizer,¹ a kinetic equation was written for the electrons in the field of a spatially homogeneous high-frequency electromagnetic wave, when the electrons are scattered elastically by the atoms. In the work of Épshtein² and Mel'nikov,³ quantum kinetic equations were introduced in the case of a strong electromagnetic field acting on an electron-phonon system. The nonequilibrium states of the electronic excitations in a superconducting film subjected to the action of a microwave electromagnetic field, when the basic mechanism of stochastization of the electrons is produced by electron-impurity collisions, were considered by Eliashberg.⁴ In the work of Kas'yanov and Starostin,⁵ a quantum kinetic equation was written for the case of inelastic electron-ion collisions in the plasma. However, in the theory of magnetism, studies analogous to Refs. 1-5 have not been made to date, so far as we know. This raises the question of the nonequilibrium states of magnons in a ferromagnetic substance located in a strong alternating magnetic field.

It is well known that at $\Omega > 2\varepsilon_k$ (case of parametric resonance, see, for example, Ref. 6), at a sufficiently intense alternating magnetic field, an instability develops relative to processes of the type $\Omega \rightarrow \varepsilon_k + \varepsilon_{-k}$, leading to the nonequilibrium states of the magnons. In the case $\Omega < 2\varepsilon_k$, there are no direct processes which lead to the absorption of energy of the external field¹ and the spin system can be excited only because the magnons interact with one another, with impurities, and so on.

In this work, we have considered nonequilibrium stationary states of magnons, due to magnon-magnon and magnon-impurity interactions, in the case $2\varepsilon_0 > \Omega > \tau^{-1}$.

We consider a ferromagnetic substance (one-dimensional sample, $H > 4\pi M_0$) located in an alternating magnetic field parallel to the static field H and to the equilibrium direction of magnetization M_0 . The Hamiltonian of the system of magnons can be written in the form⁷

$$\begin{aligned} \mathcal{H} = & \sum_p \varepsilon_p c_p^\dagger c_p + \sum_p h_p(t) c_p^\dagger c_p + \sum_{123} (V_{1,23} c_1^\dagger c_2 c_3 + \text{H.c.}) \\ & + \sum_{1234} W_{12,34} c_1^\dagger c_2^\dagger c_3 c_4 + \sum_{1234} (W'_{1,23} c_1^\dagger c_2 c_3 c_4 + \text{H.c.}) \\ & + \sum_{12} U_{12} c_1^\dagger c_2 + \sum_{12} (V_{12} c_1 c_2 + \text{H.c.}) \end{aligned} \quad (1)$$

where

$$\begin{aligned} \varepsilon_p &= (A_p^2 - [B_p]^2)^{1/2}, \quad A_p = \Theta_c (ap)^2 + \varepsilon_0 + m \sin^2 \theta_p, \\ B_p &= m \sin^2 \theta_p \exp(-2i\varphi_p), \quad \varepsilon_0 = 2\mu(H + \beta M_0), \\ h_p(t) &= h_p^0 \cos \Omega t, \\ h_p^0 &= \mu h_0 \frac{\Theta_c (ap)^2 + \varepsilon_0 + m \sin^2 \theta_p}{[[\varepsilon_0 + \Theta_c (ap)^2]^2 + 2[\varepsilon_0 + \Theta_c (ap)^2] m \sin^2 \theta_p]^{1/2}}. \end{aligned}$$

Here h_0 is the amplitude of the alternating magnetic field; $\mu = g\mu_0$, μ_0 is the Bohr magneton, Θ_c is the Curie energy; a is the lattice constant; θ_k and φ_k are the polar angles in the wave-vector space; c_k^\dagger and c_k are the creation and annihilation operators of magnons with wave vector p ; the laws of momentum conservation are assumed to be incorporated in the corresponding designations of the amplitudes (see Ref. 7) of the interactions $V, W, W', m \equiv 4\pi\mu M_0$.

In the Hamiltonian (1) we have limited ourselves to only purely nonresonant components of the interaction of the particles with the field, omitting the component of type

$$H_p(t) c_p^\dagger c_{-p} + \text{H.c.}$$

This is caused by the fact that, under the condition $\Omega > 2\varepsilon_0$ ($\varepsilon_0 \equiv \varepsilon_k, k=0$) the processes of excitation of the spin system in second-order perturbation theory in the field are forbidden by the law of energy conservation, while account of such terms in higher order perturbation theory gives much smaller effects than those already taken into account in (1) through the parameter

$$(M_0/H)^2 (\Omega/\varepsilon_0)^2 \ll 1. \quad (2)$$

To construct the kinetic equation for the magnon distribution function f_p we can, for example, carry out a canonical transformation of the Hamiltonian (1) with the aid of the unitary operator

$$U = \exp \left\{ -i \sum_p \frac{h_p^0}{\Omega} \sin \Omega t c_p^\dagger c_p \right\}, \quad (3)$$

thus transforming the time dependence [realized through

$h_p(t)$] into a Hamiltonian of the relatively weak interaction of the particles with one another and with the impurities.² In such a procedure, the field can be taken into account exactly, and the perturbation theory is developed in terms of the weak magnon-magnon and magnon-impurity interactions. Limiting ourselves to second order perturbation theory in the interaction, we get the following kinetic equation for the zeroth harmonic of the distribution function ($\Omega\tau \gg 1$):

$$\begin{aligned} \frac{\partial f_p}{\partial t} = & 4\pi \sum_{n=-\infty}^{\infty} \sum_{123} |V_{1,23}|^2 (\delta_{p_3} + \delta_{p_2} - \delta_{p_1}) \\ & \times J_n^2 \left(\frac{h_1^0 - h_2^0 - h_3^0}{\Omega} \right) \{ (1+f_2) (1+f_3) f_1 - (1+f_1) f_2 f_3 \} \delta(\varepsilon_1 - \varepsilon_2 - \varepsilon_3 + n\Omega) \\ & + 16\pi \sum_{n=-\infty}^{\infty} \sum_{1234} |W_{12,34}|^2 \delta_{p_1} J_n^2 \left(\frac{h_1^0 + h_2^0 - h_3^0 - h_4^0}{\Omega} \right) \\ & \times \{ (1+f_1) (1+f_2) f_3 f_4 - f_1 f_2 (1+f_3) (1+f_4) \} \delta(\varepsilon_1 + \varepsilon_2 - \varepsilon_3 - \varepsilon_4 + n\Omega) \\ & + 12\pi \sum_{n=-\infty}^{\infty} \sum_{1234} |W'_{1,234}|^2 J_n^2 \left(\frac{h_1^0 - h_2^0 - h_3^0 - h_4^0}{\Omega} \right) (\delta_{p_1} + \delta_{p_2} + \delta_{p_3} - \delta_{p_4}) \\ & \times \{ f_1 (1+f_2) (1+f_3) (1+f_4) - f_2 f_3 f_4 (1+f_1) \} \delta(\varepsilon_1 - \varepsilon_2 - \varepsilon_3 - \varepsilon_4 + n\Omega) \\ & + 2\pi n_i \sum_{n=-\infty}^{\infty} \sum_{p'} |U_{pp'}|^2 J_n^2 \left(\frac{h_p^0 - h_{p'}^0}{\Omega} \right) (f_{p'} - f_p) \delta(\varepsilon_p - \varepsilon_{p'} + n\Omega) \\ & + 2\pi n_i \sum_{n=-\infty}^{\infty} \sum_{p'} |V_{pp'}|^2 J_n^2 \left(\frac{h_p^0 + h_{p'}^0}{\Omega} \right) (1+f_p + f_{p'}) \delta(\varepsilon_p + \varepsilon_{p'} + n\Omega), \end{aligned} \quad (4)$$

where $J_n(x)$ is a Bessel function and n_i is the concentration of the impurities.

In the derivation of Eq. (4), we took it into account that the oscillating part of the distribution function f_p is small (see the Appendix). Quantum kinetic equations similar to (4) have been studied previously¹⁻⁵ for other systems under the condition $\Omega\tau \gg 1$. In the Appendix, and also in Ref. 8, a kinetic equation is given for a ferromagnetic substance, which enables one to consider arbitrary harmonics of the distribution function. Equation (4) makes it possible to consider a wide circle of problems in which account of the nonequilibrium stationary states is important.

We shall be interested below in the relatively weak field

$$\mu h_0 \ll \Omega,$$

when we can restrict ourselves (in the quadratic approximation in the field) to single-quantum processes only.

Equation (4) shows that the external field induces processes that proceed with nonconservation of the number of magnons. This means that the magnetization of the ferromagnet, which is defined by the equality

$$M(h, T) \approx M_0 - \mu \sum_p f_p, \quad (5)$$

changes in the nonequilibrium state in comparison with the thermodynamic equilibrium value that corresponds to the given temperature T . Since, as a rule, the probability of triple magnon-magnon interactions is greater than the probability of quadruple interactions accompanied by a change in the number of magnons,⁷ we analyze only the contribution connected with processes of the type $\varepsilon_1 + \varepsilon_2 = \varepsilon_3 \pm \Omega$. As to the other interactions, which

are described by the Hamiltonian (1), they, conserving the number of magnons, do not have any effect on the magnetization but of course also determine the absorption of the energy of the external field, which leads to a redistribution of the magnons in p space.

With account of the remarks made above, we solve Eq. (4) in the stationary case in the τ approximation. Substituting the solution in (5) we obtain the following expression for the relative change in the magnetization:

$$\delta m = -\frac{\mu}{M_0} \sum_p \delta f_p = \frac{\mu^2 \tau}{16\pi^2} \left(\frac{\mu h_0}{2\Omega} \right)^2 \frac{M_0 T^2}{\Theta_c^2 v_0} J(\omega, x_0), \quad (6)$$

where δf_p is the nonequilibrium correction to the Bose distribution function of the magnons, v_0 is the volume of the elementary cell, $\omega \equiv \Omega/T$, $x_0 \equiv \varepsilon_0/T$,

$$\begin{aligned} J(\omega, x_0) = & I(\omega, x_0) + I(-\omega, x_0), \\ I(\omega, x_0) = & \int_{3x_0+\omega}^{\infty} dx \int_x^{\infty} dx' \frac{e^x (1-e^{-\omega})}{(e^x-1)(e^{x'}-1)(e^{x-x'-\omega}-1)}, \\ & x_{\pm} = 1/2(x-\omega) \pm 1/2[(x-x_0)(x-3x_0-2\omega)]^{1/2}. \end{aligned}$$

As is seen, a noticeable effect should be expected upon satisfaction of the inequality $T \gg \varepsilon_0$, i.e., under conditions in which ferromagnets are usually studied experimentally. In the case $T \gg \varepsilon_0 \gg \Omega$ we get

$$\delta m \sim \frac{1}{2^2 \pi^2} \frac{\mu}{M_0 v_0} \left(\frac{\mu h_0}{\Omega} \right)^2 \left(\frac{\mu M_0}{\Theta_c} \right)^2 \frac{T}{\Theta_c} T \tau. \quad (7)$$

As is seen from formula (7), for the experimental observation of the effect of suppression of the magnetization, and also of the phenomena connected with it, the most preferable magnets are those with, first, a small exchange energy (more or less of the order of several dozen degrees Kelvin) and, second, with small activation energies ε_0 (for example, films magnetized in a plane). Moreover, very pure samples are desirable, in which the characteristic time of spin-spin relaxation is relatively small ($\tau \sim 10^{-6} - 10^{-7}$ sec). We regard also of interest the situation in which the ferromagnetic material is close to a second order phase transition of the spin-flip type. In this case, there is a strong lowering of the magnon mode $\varepsilon_0 \sim \mu (|H_c - H| H + \Delta_0^2)^{1/2}$ (H_c is the spin-flip field, Δ_0 is the gap due to the magnetostrictive interaction), and the nonequilibrium states of the magnons should have a significant effect on the transition fields.

As has already been pointed out above, the energy of the external alternating field goes into maintaining the nonequilibrium state of the magnon system. We now calculate the value of the absorption due to triple magnon-magnon and magnon-impurity interactions. The absorption of nonresonant energy of the alternating field was considered earlier⁹ in the case $\Omega\tau \ll 1$; here we shall be interested in the opposite limiting case. Defining the energy Q absorbed by the ferromagnet per unit time as $T\dot{S}$, where S is the entropy of the nonequilibrium gas of magnons, and noting that, on the other hand,

$$Q = \frac{h_0^2}{8\pi} \Omega \chi'',$$

we obtain the following result for the imaginary part of the high-frequency magnetic susceptibility χ'' due to triple magnon-magnon interactions at $T \gg \varepsilon_0 \gg \Omega$:

$$\chi'' \approx \frac{\nu}{2^2 \pi^2 \nu_0 \Omega} \left(\frac{\mu M_0}{\Theta_c} \right)^2 \left(\frac{T}{\varepsilon_0} \right) \left(\frac{T}{\Theta_c} \right), \quad \nu \approx 10^{-2}. \quad (8)$$

At $\mu M_0 \sim 0.1$ K, $\varepsilon_0 \sim 2$ K, $T \sim 300$ K, $\Theta_c \sim 500$ K, $\Omega \sim 10^7$ sec $^{-1}$, we have $\chi \approx 10^{-2}$ which, as is seen, can manifest itself significantly in the experiment. We note that our result (8) differs from that given by Lutovinov and Chechetkin,¹⁰ owing to their not entirely correct calculation of the effective vertices, as is easy to prove.

A larger value of the absorption can be obtained by considering a ferromagnet with impurities. The quantity χ'' due to magnon-impurity collisions, can be represented in the form

$$\chi'' \approx \frac{1}{16\pi^2} \left(\frac{\mu^2 n_i}{\Omega} \right) \left(\frac{4\pi\mu M_0}{\Theta_c} \right) \left(\frac{U}{\Theta_c} \right)^2 \left(\frac{T}{\varepsilon_0} \right) \quad (9)$$

under the conditions $M_0 \ll H$, $T \gg \varepsilon_0 \gg \Omega$. We now estimate the order of the absorption in the given case. At $n_i \sim 10^{21}$ cm $^{-3}$, $\Theta_c \sim 500$ K, $m \sim 1$ K, $\varepsilon_0 \sim 2$ K, $t \sim 300$ K, $U \sim \Theta_c$, $\Omega \sim 10^7$ sec $^{-1}$, the value of χ'' amounts to 10^{-1} in order of magnitude. Thus, the interaction of the magnons with the impurities, which leads to the possibility of parametric nonresonant absorption of the alternating magnetic field, can result in a significant absorption of energy. Experimental verification of the quantities considered in the present work would, in our opinion give interesting information on the mechanisms of nonresonant excitation of spin waves in the nonequilibrium state.

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APPENDIX

When the frequency of the alternating magnetic field $\Omega \ll \tau_0^{-1} \equiv \langle \varepsilon \rangle$ ($\langle \varepsilon \rangle$ is the average energy of the magnon, for example, of the order of T) at times $t \gg \tau_0$, the kinetics of the magnons can, as is known, be written with the help of the distribution function f_p , "smoothed" over intervals $\Delta t \gg \langle \varepsilon \rangle^{-1}$. In the case of weak coupling, it can be shown⁸ that the kinetic equation for the system described by the Hamiltonian (1) has the form

$$\frac{\partial f_p}{\partial t} = 4 \sum_{1,2,3} |V_{1,23}|^2 K_{123}(h, \Omega, t) (\delta_{p_3} + \delta_{p_2} - \delta_{p_1}) \{ (1+f_2)(1+f_3)f_1 - (1+f_1)f_2f_3 \} + 2n_i \sum_{p'} |U_{pp'}|^2 K_{pp'}(h, \Omega, t) (f_p - f_{p'}), \quad (A.1)$$

where

$$\begin{aligned} \bar{K}_i(h, \Omega, t) &= \int_{-\infty}^{\infty} dt e^{i\Omega t} \cos \{ E_i \tau + \lambda_i [\sin \Omega(t+\tau) - \sin \Omega t] \} \\ &= \frac{\pi}{2} \sum_{n, n' = -\infty}^{+\infty} (-1)^n e^{i m \Omega t} \{ \delta_+(E_i - n\Omega) + (-1)^m \delta_-(E_i + n\Omega) \} J_n(\lambda_i) J_{m-n}(\lambda_i), \\ \delta_-(x) &= \delta_+(x) = 1/\pi(\eta + ix), \quad \eta \rightarrow +0, \\ i &= \{ (pp'), (1,23) \}, \quad E_{pp'} = \varepsilon_p - \varepsilon_{p'}, \quad E_{1,23} = \varepsilon_1 - \varepsilon_2 - \varepsilon_3, \\ \lambda_{pp'} &= (h_p^0 - h_{p'}^0)/\Omega, \quad \lambda_{1,23} = (h_1^0 - h_2^0 - h_3^0)/\Omega. \end{aligned}$$

This equation is valid at any value of the parameter $\Omega\tau$ (but, of course, in the case $\Omega\tau_0 \ll 1$). We now show that at $\Omega\tau \gg 1$ the oscillations of the distribution function can be neglected and thus Eq. (A.1) goes over into Eq. (4) for the zeroth harmonic.

For brevity, we consider only the impurity part of the

generalized collision integral. Representing f_p in the form of the sum

$$f_p^{(1)} + f_p^{(1)} e^{i\Omega t} + f_p^{(1)*} e^{-i\Omega t},$$

where $f_p^{(n)}$ is a function that changes slowly over intervals Ω^{-1} , and carrying out the substitution in Eq. (A.1), we obtain an equation of the following form for the first harmonic of $f_p^{(1)}$ in the τ approximation

$$\frac{\partial f_p^{(1)}}{\partial t} + i\Omega f_p^{(1)} = -\frac{1}{\tau} f_p^{(1)} + \Phi_p^{(1)}, \quad (A.2)$$

where

$$\begin{aligned} 1/\tau &= 2\pi n_i \sum_{p'} |U_{pp'}|^2 \sum_n J_n^2(\lambda) \delta(E - n\Omega), \\ \lambda &= (h_p^0 - h_{p'}^0)/\Omega, \quad E = \varepsilon_p - \varepsilon_{p'}, \\ \Phi_p^{(1)} &= \pi n_i \sum_{p'} |U_{pp'}|^2 \sum_{m=-\infty}^{\infty} J_m(\lambda) J_{m-1}(\lambda) (f_p^{(0)} - f_{p'}^{(0)}) \\ &\quad \times [\delta_+(E - m\Omega) - \delta_-(E + m\Omega)]. \end{aligned}$$

Using the definition of τ we see that, in absolute value,

$$\Phi_p^{(1)} \ll \Lambda_p(\lambda) \tau^{-1} f_p^{(0)}$$

[here Λ_p is determined by the ratio of Φ_p to $1/\tau(p)$].

Thus, we have from (A.2) at $\Omega\tau \gg 1$,

$$f_p^{(1)} \ll \Lambda_p(\Omega\tau)^{-1} f_p^{(0)}. \quad (A.3)$$

The function Λ_p has a rather complicated dependence both on λ and on p . We give estimates for $\varepsilon_p \sim \varepsilon_0$ when, in the case of equilibrium in the system, the function $f_p^{(0)}$ is maximal (and consequently, $f_p^{(1)}$ is also maximal). In the case of strong fields, when $\lambda \gg 1$, after rather un-complicated calculations, we obtain

$$\Lambda_0 \approx \frac{1}{\lambda} \sum_{m>0} m^2 J_m^2(\lambda) / \sum_{m>0} m^2 J_m^2(\lambda), \quad \lambda \gg 1.$$

The case $\lambda \ll 1$ is the simplest, since here we can carry out the expansion in (A.2) and obtain

$$f_p^{(1)} \sim (\Omega\tau)^{-1} f_p^{(0)}, \quad \lambda \ll 1.$$

In conclusion, let us make one more remark. In the case $\Omega\tau \leq 1$, as is seen from the investigation that has been carried out, consideration of the kinetic equation in the form A (A.1) is required from first principles. In this case, for a real experimental situation, fields are easily achieved in which $\mu h_0 \gg \Omega$, i.e., when the external field enters in sufficiently nonlinear fashion in the equation. It should also be noted that at $\Omega\tau \gg 1$ a reasonable condition is $\mu h_0 \gg \Omega$ under multiquantum processes must be taken into account in the equation for the zeroth harmonic of the distribution function.

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Effect of electron-lattice interaction on the formation of the equilibrium structure of a metal

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The calculation of the derivatives of the total energy of a metal with respect to the lattice parameters is investigated. Since direct differentiation of the perturbation-theory series leads to infinite corrections when the Fermi surface is tangent to the Bragg plane, a program for the direct calculation of the derivatives is sought. Particular attention is paid to the tangency of the Fermi surface to an edge of the Brillouin zone. It is found that differentiation with respect to the parameter $\gamma = c/a$ that describes the oblateness of the lattice is finite, and that the correction for the intersection of the edge is of the order of $W^{5/2}$ (W is the electron-lattice interaction constant). The derived equations are used in the analysis of the stability of the strongly anisotropic structure of metallic hydrogen. It is shown that upon closer analysis the simple hexagonal structure predicted within the framework of ordinary perturbation theory to be metastable at zero pressure can be metastable only under pressure.

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The pseudopotential method is now used successfully to calculate the cohesion energy of metals; many papers are devoted to the principles of the method, to the selection of the pseudopotential, and to the calculation of the properties of various metals.^{1,2} A number of questions, however, call for additional study, particularly the question of the correct assessment of the role of the electron-lattice interaction in the formation of the anisotropic metallic structure. The point is that the electron-lattice interaction constant of a number of metals cannot be regarded as small and it becomes necessary to take into account higher orders of perturbation theory, the third³ and even the fourth.⁴⁻⁶ The situation is made more complicated by the fact that when this constant increases energy considerations favor the formation of highly anisotropic structures⁷ for which it is important to take correct account of the intersection of the Fermi surface with the faces and edges of the Brillouin zone. It is known that perturbation theory leads to infinite corrections for the band energy $\varepsilon(\mathbf{k})$ if the wave vector \mathbf{k} is close to a Bragg plane. If, however, this energy is integrated over the states within the Fermi sphere, the result is finite and, as shown by a detailed analysis, this method can be used to calculate the correction to the total energy in the second^{1,2} and third⁴ orders of perturbation theory.

The search for the energywise most favored structure at a given volume includes also the variation of the anisotropy parameter $\gamma = c/a$, but the equations of the

simple perturbation theory no longer permit calculation of the derivatives of the total energy with respect to γ . The derivative calculated in this manner becomes infinite when the Fermi sphere is tangent to a Brillouin-zone face or edge. Calculation of the energy for one Bragg plane without the use of perturbation theory² leads, of course, to a finite derivative.

The purpose of the present paper is the calculation of the total energy and its derivative with respect to the parameter γ in the case when the Fermi surface is tangent to an edge of the Brillouin zone. To this end we consider in detail the singularity of the produced in the state-density curve by the presence of the edge, and determine the parameters of the Van Hove singular point. It is shown that the derivative with respect to the parameter γ has a finite correction of the order of $W^{5/2}$ (W is the electron-lattice interaction constant), and the question of the stability of the spectrum in the case of a tangent edge is determined by the competition between the various contributions to the total energy.

The obtained procedure is used to analyze the stability of a simple hexagonal (SH) structure of metallic hydrogen, in view of the mentioned difficulties encountered in the perturbation-theory calculations. The electron-lattice interaction parameter of metallic hydrogen is large because there are no ionic cores, and at low pressures the strongly oblate anisotropic SH structure is favored.^{7,8} It is shown that tangency of the