Quantum-limit laser-cyclotron pumping of spin waves in the europium chalcogenides assisted by Landau electrons

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The theory of the excitation and amplification of spin waves via free carriers in the europium chalcogenides in the presence of a quantizing magnetic field is presented. For electromagnetic radiation polarized transverse to the magnetic field the process of free-carrier absorption of laser radiation merges into that of cyclotron resonance and we find that, in the ultraquantum limit where only the lowest Landau level is fully occupied, the magnon system may reach instability as the cyclotron frequency of the free carriers approaches the laser frequency.

I. INTRODUCTION

The europium chalcogenides EuX are, respectively, antiferromagnetic (X = Te), ferrimagnetic (X = Se), and ferromagnetic (X=O,S) semiconductors.¹ All EuX compounds crystallize in the rocksalt structure, i.e., the Eu sublattice is fcc. The magnetic properties are caused by the half-filled 4f shell of Eu^{2+} . Because of negligible overlap of the 4f wave functions of adjacent Eu^{2+} ions, these 4f electrons form quasilocalized magnetic moments $(S = \frac{7}{2})$. The undoped EuX compounds are thought to be good realizations of the three-dimensional Heisenberg model. Still more spectacular than their purely magnetic properties are their magneto-optic properties, which result from an interaction between the localized 4f moments and the itinerant electrons of the 5d conduction band. The drastic red shift of the optical absorption edge, observed in the ferromagnetic compounds when the temperature decreases below T_c , is a very frequently investigated effect in this connection. It can be explained almost quantitatively by an intra-atomic s-f (or d-f) exchange interaction between the Eu 4f spin and the conduction-electron spins. This interaction provides a relatively strong spin splitting of the conduction and valence bands in the ordered phase and even in the paramagnetic phase where it leads to a complicated band structure. Furthermore, it is a very interesting fact that the physical properties of the magnetic and semiconducting EuX compounds strongly depend on the concentration of free carriers, created, for instance, by doping with suitable impurities (Gd^{3+}) .^{2,3} The presence of these free carriers can be observed mainly in connection with intraband light-absorption experiments⁴⁻⁶ below T_c in which the magnons provide an additional channel to assist the indirect intraband absorption processes. The absorption takes place with the simultaneous emission or absorption of magnons.^{7,8} These magnons constitute welldefined excitations in these materials at the same time that their interaction with the free carriers may constitute

a fundamental mechanism in the damping and growth of spin waves. In fact, as recently discussed,⁸ the magnonassisted radiation absorption by a free carrier in a magnetic semiconductor such as $CdCr_2Se_4$ can, under certain conditions, give rise to a net emission of magnons rather than absorption and the magnon population grows with time.

Under the additional presence of a strong magnetic field, an increase in the absorption of radiation by free carriers in EuO has also been observed at low temperatures,⁴ and interpreted as the consequence of an electronic transition between Landau levels when the laser-cyclotron resonance condition is reached. On the other hand, it is natural to expect that external fields which change the spectrum and the occupation number of the electron states will considerably influence the spectrum and damping of spin waves. This has recently been discussed in Ref. 9, where it was shown that the presence of a quantizing (strong) magnetic field can significantly change the magnon damping in a magnetic semiconductor.

In this paper we are interested in extending the theory of the excitation and amplification of spin waves in a magnetic semiconductor developed previously⁸ to take into account the presence of quantizing magnetic fields. As is well known, in the presence of a strong magnetic field, the electronic energy levels in a band are split into subbands of discrete Landau levels. Laser-cyclotron resonance absorption occurs when a photon has sufficient energy to take an electron from one Landau level to the adjacent one (transitions between other than neighboring Landau levels in semiconductors are forbidden by selection rules in semiconductors having parabolic energy bands¹⁰), and this transition is accompanied by the emission or the absorption of a magnon. It is therefore important to consider the question of how the carrier-assisted laser excitation of magnons⁸ may be affected by the presence of a quantizing magnetic field. Our calculation here will be restricted to the $4f^7$ electronic configuration of

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magnetic ions $(S = \frac{7}{2})$ applicable to the physical properties of EuO.¹ A brief description of this work has been published elsewhere.¹¹

The paper is organized as follows. In Sec. II, we present the theory of spin-wave excitation by a photon field in the presence of a quantizing external magnetic field. The results are then specialized to the quantum limit where the carrier concentration is such that only the Landau state with the lowest oscillator quantum number is fully occupied. In Sec. III the results are presented and discussed.

II. THEORY

We consider a monochromatic beam of photons from a laser source of frequency ω_L incident on our magnetic semiconducting crystal. The Hamiltonian of the system is

$$H = H_E + H_M + H_R + H_{\rm EM} + H_{\rm ER} , \qquad (1)$$

where H_E is the Hamiltonian for the many-electron system, H_M is that of the magnon system, H_R is the Hamiltonian of the photon field, $H_{\rm EM}$ represents the electronmagnon interaction energy, and $H_{\rm ER}$ is the interaction between the radiation and the electron system. In Eq. (1) we have not included the direct photon-magnon coupling as we are interested only in investigating the role of the carriers in the magnon pumping. The crystal is assumed to be in the presence of a static magnetic field **B**, with associated vector potential, given in the Landau gauge, $A_0 = (-yB, 0, 0)$. We work within the effective-mass approximation and take the one-electron wave functions to be the Landau states¹² $|v=n,k_x,k_z\rangle$, where *n* is the Landau-level quantum number. The corresponding oneelectron energies are

$$E_{K,n,\downarrow} = (n + \frac{1}{2})\hbar\omega_c + (\hbar k_z)^2 / 2m^*$$

$$\mp \frac{1}{2} (J_{s-f}S + \hbar\omega_c) , \qquad (2)$$

where k_z is the component of the electron wave vector along the magnetic field direction and $\omega_c = eB/m^*c$ is the carrier cyclotron frequency. The electron-magnon interaction, when the *s*-*f* exchange interaction is the dominant interaction mechanism, is given by

$$H_{\rm EM} = \sum_{\mathbf{v}, \mathbf{v}, \mathbf{q}'} \left(V_{\rm EM} \langle \mathbf{v} | \exp(i\mathbf{q} \cdot \mathbf{r}) | \mathbf{v} \right) \\ \times C_{\mathbf{v}\uparrow}^{\dagger} C_{\mathbf{v}\downarrow} b_{\mathbf{q}}^{\dagger} + {\rm c.c.} \right), \qquad (3)$$

where $V_{\rm EM} = -J_{s-f}(S/2N)^{1/2}$, J_{s-f} being the exchangeinteraction parameter, v, v' are Landau states, and the sign \uparrow (\downarrow) refers to the electrons in the spin-up (-down) subband. The Hamiltonian which gives the interaction between the photon field and the electrons is



FIG. 1. (a) and (b) magnon emission and (c) and (d) absorption in the lowest and the first excited Landau levels. The electron states are represented by full lines, the magnons by wavy lines, and the photons by dashed lines. Time increases from left to right.

$$H_{\rm ER} = \sum_{\mathbf{v}, \mathbf{v}, \mathbf{K}_L} \left\{ V_{\rm ER} \langle \mathbf{v}' | \exp(i\mathbf{K}_L \cdot \mathbf{r}) \hat{\mathbf{e}}_{\lambda} \cdot \left[\mathbf{K} - (e/c) \mathbf{A}_0 \right] | \mathbf{v} \rangle C_{\mathbf{v}}^{\dagger} C_{\mathbf{v}} a_{\mathbf{K}_L} + \text{c.c.} \right\},$$
(4)

where $V_{\text{ER}} = (1/m^*)(2\pi e^2 \hbar / v\omega_L)^{1/2}$, $\mathbf{K} = (\hbar/i)\nabla$, $\hat{\mathbf{e}}_{\lambda}$ is the unit polarization vector for a photon \mathbf{K}_L propagating parallel to the magnetic field and polarized along the x direction, and v is the crystal volume. Also, in Eqs. (3)

and (4), C_{ν} , b_{q} , and $a_{K_{L}}$ are second-quantization operators for electrons, magnons, and photons, respectively.

In the following we present the quantum-mechanical formulation for the excitation of magnons by the laser field via the Landau electrons (here a Landau electron represents an electron in the presence of a quantizing magnetic field). This is very much similar to the photon conversion by charged particles encounted in plasma physics. In lowest order of perturbation theory,¹³ the photon-magnon conversion processes assisted by Landau electrons would be analogous to the Compton scattering of quantum-field theory as shown in Fig. 1. The excitation rate $\gamma(\mathbf{q})$ may be calculated from the rate of change of the magnon population $N(\mathbf{q})$, $dN(\mathbf{q})/dt$, where $dN(\mathbf{q})/dt$ is given by the sum of the magnon probabilities for transitions in which magnons are emitted, minus a similar sum of the probabilities for transition in which magnons are absorbed, averaged over the initial states, and summed over the final states. The emission or absorption of a magnon is accompanied by the absorption of a photon of laser field.

The transition proabilities are to be calculated by the usual methods of time-dependent perturbation theory¹³ in terms of the second-order matrix elements as

$$W_{i} = \left[\frac{2\pi}{\hbar}\right] \sum_{f} |\langle f | M | i \rangle|^{2} \delta(E_{i} - E_{f}), \qquad (5)$$

where M_{fi} is the second-order matrix element, and E_i and E_f are the initial and final energies. The second-order matrix element in Eq. (5) is given by

$$M_{fi} = \sum_{\beta} \frac{\langle f | H | \beta \rangle \langle \beta | H | i \rangle}{E_i - E_{\beta} + i\eta} \quad (\eta \to 0) .$$
 (6)

In Eq. (6) $\langle \beta | H | i \rangle$ and $\langle f | H | \beta \rangle$ are the appropriate vertices for the electron-photon and electron-magnon interactions and the sum is over all the intermediate states β . Also in Eq. (6), $\hbar\omega_L$ and $\hbar\omega_M$ are the photon and magnon energies, respectively. To evaluate $\langle f | M | i \rangle$, we need to know the overlaps $\langle v' | e^{i\mathbf{q}\cdot\mathbf{r}} | v \rangle$ and

$$\langle v' | e^{i\mathbf{K}_L \cdot \mathbf{r}} \hat{\mathbf{e}}_{\lambda} \cdot [\mathbf{K} - (e/c)\mathbf{A}_0] | v \rangle$$

With the use of the Landau wave functions¹² it can be seen that^{9,14}

$$\langle \nu' | e^{i\mathbf{q}\cdot\mathbf{r}} | \nu \rangle = \delta_{k'_x,k_x+q_x} \delta_{k'_z,k_z+q_z} \chi_{n',n} , \qquad (7)$$

with

$$\chi_{n',n} = \delta_{n',n} e^{-\rho/2} L_n(\rho) + \Theta(n'-n) \left[\frac{n!}{(n')!} \right]^{1/2} e^{i(n'-n)\phi} e^{-\rho/2} \rho^{(n'-n)/2} L_n^{n'-n}(\rho) + \Theta(n-n') \left[\frac{(n')!}{n!} \right]^{1/2} e^{i(n-n')\phi} e^{-\rho/2} \rho^{(n-n')/2} L_n^{n-n'}(\rho) .$$
(8)

For a photon propagating along the z axis and polarized parallel to the z direction we obtain

$$\langle \mathbf{v}' | e^{i\mathbf{K}_{L}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\lambda} \cdot [\mathbf{K} - (e/c)\mathbf{A}_{0}] | \mathbf{v} \rangle$$

= $\delta_{k_{x}',k_{x}} \delta_{k_{z}',k_{z}} + K_{L} \left[\frac{m\hbar\omega_{c}}{2} \right]^{1/2}$
 $\times [(n+1)^{1/2} \delta_{n',n+1} - n^{1/2} \delta_{n',n-1}]$ (9)

Here, $L_n(\rho)$ are the associated Laguerre polynomials, Θ is the unit step function,

$$\Theta(n-n') = \begin{cases} 1 & \text{if } n > n' \\ 0 & \text{if } n \le n' \end{cases}, \\ \rho = \hbar q_{\perp}^2 / 2m^* \omega_c, \quad \phi = \tan^{-1}(q_y / q_x) \end{cases}$$

and q_{\perp} is the component of the magnon wave vector **q** perpendicular to the magnetic field. The expression for $\chi_{n',n}$ in (8) is very complicated for arbitrary *q* because, in general, both the $n \rightarrow n$ and $n \rightarrow n'$ $(n \neq n')$ transitions are possible. However, because of the selection rules for semiconductors having parabolic energy bands,¹⁰ only transitions between neighboring Landau levels $(n'=n\pm 1)$ are allowed. For simplicity, we shall now consider only up-

ward n'=n+1 electron transitions. In other words we assume the quantum limit in which the carrier concentration is such that only the n=0 Landau level is completely occupied. Under the foregoing considerations, Eq. (8) reduces to

$$\chi_{n',n} = \left[\frac{n!}{(n')!}\right]^{1/2} e^{i(n'-n)} e^{-\rho/2} \rho^{(n'-n)/2} L_n^{n'-n}(\rho)$$

$$(n' > n) . \quad (10)$$

By making use of the properties of the Laguerre polynomials,¹⁵ Eq. (10) can be drastically simplified in the case when $\rho \ll 1$. Under these conditions ($\rho \ll 1$)

$$L_n^r(\rho) \approx \frac{(n+r)!}{n!r!} - \frac{(n+r)!}{(n-1)!(r+1)!}\rho .$$
 (11)

Using Eq. (11) in Eq. (10), and taking into account the ultraquantum limit (n=0), Eq. (10) reduces to

$$\chi_{1,0} \simeq e^{i\phi} \rho^{1/2}$$
 (12)

The condition $\rho \ll 1$ can easily be obtained by increasing the magnetic field strength ($\rho \propto 1/B$) for a fixed magnonpropagation direction. Hence, using Eqs. (6), (7), (9), and (12) we can write $\langle \nu' | M | \nu \rangle$ as

$$\langle v' | M | v \rangle = V_{\rm EM} V_{\rm ER} (m^* \hbar \omega_c)^{1/2} e^{i\phi} \rho^{1/2} F$$

$$F = \frac{1}{E_{0,k_z,1} - E_{1,k_z + K_{L,1}} + \hbar\omega_L} + \frac{1}{E_{0,k_z,1} - E_{1,k_z - q_z,1} - \hbar\omega_M} = \frac{K_L q_z / m^*}{(\omega_L - \omega_c)^2}$$
(13)

Having obtained $\langle v' | M | v \rangle$, we can now look at the rate of change of the magnon population due to the scattering processes assisted by the Landau electrons (Fig. 1),

$$(\hbar\omega_L)_{\mathbf{K}_L} + e_{\nu=0,k_x,k_z,\downarrow} \rightarrow (\hbar\omega_M)_{\mathbf{q}} + e_{\nu=1,k_x-q_x,k_z-q_z+k_L,\uparrow},$$

where a Landau electron (hole) in an initial state $|v=0,k_x,k_z,\downarrow\rangle$ absorbs a photon $\mathbf{K}_L = K_L \hat{\mathbf{z}}$ and collides with a magnon **q**, leaving in a final state $|v'=k_x+q_x,h_z+K_L-q_z,\uparrow\rangle$, namely

$$\frac{\partial N_q}{\partial t} = \frac{2\pi}{\hbar} \sum_{k_x, k_z} |\langle v' | M | v \rangle|^2 [N_L(N_q + 1)f_{vl}(1 - f_{v'\dagger}) - (N_L + 1)N_q f_{v'\dagger}(1 - f_{vl})] \delta(E_{v'\dagger} - E_{vl} + \hbar\omega_M - \hbar\omega_L) + \gamma', \qquad (14)$$

where γ' is the irrelevant spontaneous emission term, f_v is the electron-distribution function, and N_L is the number of photons with wave vector \mathbf{K}_L . Assuming now that N_q and N_L are large compared with unity, one obtains, from (14),

$$\partial N_q / \partial t = \gamma_q N_q + \gamma' , \qquad (15)$$

with

$$\gamma_{q} = \frac{2\pi}{\hbar} N_{L} \sum_{k_{x},k_{z}} |\langle v' | M | \omega \rangle|^{2} (f_{v\downarrow} - f_{v'\uparrow}) \\ \times \delta(E_{v\downarrow} - E_{v'\uparrow} + \hbar\omega_{L} - \hbar\omega_{M}) . \quad (16)$$

It follows immediately from (16) that if the photonmagnon system is at resonance $(\omega_L = \omega_M)$, $E_{\nu\uparrow} = E_{\nu\downarrow}$, and $\gamma_q = 0$. In other words, for $\omega_L = \omega_M$ there is no exchange of energy between the bosonic systems having the carrier as spectators; the carrier-assisted magnon excitation is zero. On the other hand, if ω_L is near ω_M but not necessarily at resonance (as is the case considered here), the carrier final energy differs slightly from its initial energy and the approximation for the carrier-distribution function is assumed to be

$$f(E_{\nu\uparrow}) = f(E_{\nu\downarrow}) + \hbar(\omega_L - \omega_M) \frac{\partial f(E_{\nu\downarrow})}{\partial E_{\nu\downarrow}} .$$
 (17)

Inserting this result into (16), we obtain the expression for the magnon excitation rate, namely,

$$\gamma_{q} = 2\pi N_{L}(\omega_{M} - \omega_{L}) \sum_{k_{x}, k_{z}} |\langle v' | M | v \rangle|^{2} \frac{\partial f(E_{vi})}{\partial E_{vi}} \times \delta(E_{vi} - E_{v'i} + \hbar\omega_{L} - \hbar\omega_{M})$$
(18)

It will follow from Eq. (18) that if γ_q is positive, the magnon population will be amplified at the expense of the laser field, whereas if γ_q is negative it is damped. Actually, a net growth of the magnon population is only achieved provided γ_q is greater than the losses η_q due to other processes than magnon emission or absorption by electrons.¹⁶ We now assume the electron gas to be nondegenerate so that the electron-distribution function is given by the Maxwell-Boltzmann distribution,¹⁷ in the presence of the quantizing magnetic field, namely,

$$f(\mathbf{k},n,\sigma) = 2n(T) \left[\frac{2\pi\hbar^2}{m^* k_B T} \right]^{1/2} \frac{\sinh(\hbar\omega_c/2k_B T)e^{\sigma\Delta/k_B T}}{\cosh(\Delta/k_B T)} \exp\left[-\left[(n+\frac{1}{2})\frac{\hbar\omega_c}{k_B T} + \frac{\hbar^2 k_z^2}{2m^* k_B T} \right] \right],$$
(19)

where n(T) is the carrier concentration at temperature T, $\Delta \equiv J_{s-f}S$ is the energy gap between spin subbands, and $\sigma = \pm 1$ (+ for \uparrow , - for \downarrow spins). The sums in Eq. (18) can now be performed easily with the help of the δ function. Hence, upon taking the degeneracy in k_x , given by $m^*\omega_c L^2/2\pi\hbar$, γ_q takes, in the ultraquantum limit where $\hbar\omega_c \gg k_B T$, the particularly simple form

$$\gamma_{q} = (\omega_{L} - \omega_{M}) \frac{N_{L} n (T) \Delta^{2} e^{2} q_{1}^{2} q_{z} \hbar^{2}}{NS (m^{*})^{2} c^{2} \omega_{L}^{3} (m^{*} k_{B} T)^{3/2}} \frac{\lambda}{(1 - \lambda)^{4}} \frac{\exp(\Delta/k_{B} T)}{\cosh(\Delta/k_{B} T)} \exp(-K_{0}^{2}/K_{T} 2), \quad \lambda \equiv \omega_{c}/\omega_{L}$$
(20)

provided $q_z \gg K_L$. In Eq. (19), K_T is the thermal wave number of the free charge carriers $K_T = (2m^*k_BT/\hbar^2)^{1/2}$,

$$K_0 = (\Delta + \hbar \omega_L - \hbar \omega_M) \frac{m^*}{\hbar^2 q_z} - \frac{\hbar^2 q_z^2}{2m^*} ,$$

and $q_{\perp} = q \sin \theta$, where θ is the angle that q makes with the magnetic field direction, the z direction.

III. DISCUSSION

Equation (20) is the expression for the magnon excitation rate by a photon field via Landau electrons in the ultraquantum limit where only Landau states with the lowest oscillator quantum number are fully occupied. It indicates that the magnon population grows with time provided $\omega_L > \omega_M$ and that it may be enhanced as com-

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pared with the zero-field case, whenever the cyclotron frequency of the charge carrier approaches the laser frequency. Physically the condition $\omega_L > \omega_M$ means that a pumping laser field of frequency ω_L cannot generate magnons of frequency higher than its own frequency. The growth rate is no longer isotropic, being maximum for magnons propagating in a direction such that $q_1 = q_z = q/\sqrt{2}$. Furthermore, upon taking into account the condition for the electron wave number K_0 in Eq. (20) and the fact that in intraband processes we always have $q \gg K_L$ (i.e., the electron-magnon scattering process is essential for the electron to gain the necessary momentum) the excited magnons are restricted to a relatively very narrow band of q values in the interval

$$\left[\frac{2m^*}{\hbar^2}(\Delta+\hbar\omega_L-\hbar\omega_M)\right]^{1/2}-K_T\leq q\leq \frac{\pi}{a_0},$$

 a_0 being the lattice constant. The upper limit is that given by the end of the Brillouin zone and the lower limit was set by the condition $K_0 \leq K_T$ for which the growth rate [Eq. (20)] does not vanish exponentially. For $K_0 > K_T$, γ_a decreases very rapidly and can be seen to be very small compared with the linear losses η_q , in order to get an actual growth of the magnon population. By the time an instability is obtained when the condition $\gamma_q \ge \eta_q$ is satisfied the magnon population grows with time and saturates because of the nonlinear relaxation mechanisms (different other than magnon absorption or emission by electrons), which should become effective in stabilizing these amplified magnons. The condition $\gamma_q \ge \eta_q$ determines the laser threshold intensity for an actual magnon amplification. To obtain an order-of-magnitude estimate of the critical value of N_L/V (photon density) we assume the following values for the physical parameters of a Gddoped EuO sample illuminated by a CO₂ laser ($\lambda = 10.8$ μ m) in the magnetic field of 60 kG: $m^* \simeq 10^{-28}$ g, $T = 50k_B, a_0 = 5.141$ Å, $s = \frac{7}{2}, J = 0.17$ eV, $h\omega_M \simeq g\mu_B H$ (dispersion is neglected), $n(T) \approx 10^{16} \text{ cm}^{-3}$, $N/V = 10^{23}$ cm⁻³. Hence, assuming η_q is typically¹⁵ of the order of 10^{10} sec^{-1} one gets $N_L/V = 5.0 \times 10^{17} \text{ cm}^{-3}$ for magnons propagating in a direction such that $q_{\perp} = q_z = q/\sqrt{2}$ with $q = 5 \times 10^7$ cm⁻¹ and $\omega_c = 1.0 \times 10^{13}$ sec⁻¹. This, in turn, entails a laser intensity of about 3.0×10^8 W/cm², well within the experimental capabilities.

In conclusion, we have discussed in this paper the conditions of the excitation and amplification of spin waves in nondegenerate EuO in a quantizing magnetic field by free carrier in the presence of a laser radiation. We found that, for the laser beam polarized perpendicular to the magnetic field direction (where the process of free-carrier absorption is that of cyclotron resonance), the magnon amplification can be obtained whenever the laser frequency approaches the cyclotron frequency of the carriers. This strong departure of the magnon population from its equilibrium value obtains when the photoexcited electrons cascade down the neighboring Landau levels emitting magnons. Furthermore, the excited magnons are restricted to a very narrow band of magnon wave numbers, thus showing a selective mechanism for magnon excitation and amplification. On the other hand, for the laser beam polarized along the dc magnetic field the dependence of the free-carrier absorption as well as of the amplification coefficient on the applied magnetic field only appears when the quantization of the electronic energy levels in the magnetic field becomes important, which occurs when the separation between adjacent Landau levels, $\hbar\omega_c$, is greater than either the collisional broadening \hbar/τ or the thermal broadening $k_B T$ of these Landau levels. In this particular configuration the resonance condition $\omega_L = \omega_c$ is no longer a singular point in the amplification coefficient. This problem is being considered for a forthcoming paper.

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distribution function, $f(\mathbf{k}, n, \sigma)$, Eq. (19) must be a nonequilibrium (nonthermal) one. However, in order to simplify the problem we assume the laser field to be a pulsed one of duration τ_0 such that $\tau_0 \ll \tau$ (τ is the electron relaxation time), i.e.,

a quasistationary equilibrium state is reached in the electron system. Under this condition the electrons do not have sufficient time to become "hot" and this makes it possible to assume $f(\mathbf{k}, n, \sigma)$ to be a quasiequilibrium distribution.