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ESTABLISHMENT OF AN EQUILIBRIUM DISTRIBUTION OF NONLOCALIZED EXCITONS IN MAGNETIC CRYSTALS

É. G. PETROV

Institute of Theoretical Physics, Ukrainian Academy of Sciences

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The interaction between nonlocalized excitons and fluctuations of magnetic induction density in magnetic substances is considered and it is shown that exciton-magnon interaction is responsible for the final establishment of an equilibrium exciton distribution. The time required for establishment of the exciton distribution function and the time for levelling out of the spin and exciton system temperatures are calculated by the nonequilibrium density matrix technique. The mechanism of mutual transformation of triplet and singlet excitons into each other is discussed.

1. INTRODUCTION

THE study of collective properties of excitons[1-4], in connection with the possibility of obtaining high exciton densities in crystals (of the order of 10^{18} cm^{-3}) by means of laser sources, became recently of much interest. The excitons are sufficiently accurately described as a boson gas (see[4] and, particularly,[3]) with a known dispersion law $E_f(\mathbf{k})$ (f is the set of exciton quantum numbers, and $\mathbf{k}$ is the wave vector). The equilibrium distribution of an exciton gas

$$n_f^{(0)}(\mathbf{k}) = \left\{\exp\left(\frac{E_f(\mathbf{k})-\zeta}{T}\right)-1\right\}^{-1} \tag{1}$$

(ζ is the chemical potential) is achieved after a certain time τ_0. This time should be much shorter than the exciton lifetime τ_{em} relative to the crystal transition to the ground state (absence of excitons). Thus, in order to be able to talk about exciton distribution functions it is necessary to indicate exactly the processes that lead to the equilibrium distribution $n_f^{(0)}(\mathbf{k})$. The usual mechanism establishing the distribution (1) in nonmagnetic crystals is the exciton-phonon interaction[5]. After a time $\tau_0^{(\text{ef})} \sim 10^{-10}$–$10^{-12}$ sec the exciton achieves thermal equilibrium with the lattice, and its distribution function is determined by Eq. (1), in which T is the lattice temperature.

Since the order of magnitude of the lifetimes of singlet and triplet excitons is 10^{-8} and 10^{-4} sec, respectively, the excitons can be described as a boson gas during a sufficiently long time τ satisfying the condition $\tau_0^{(\text{ef})} < \tau < \tau_{\text{em}}$.

Consider now magnetic crystals (ferrodielectrics and antiferrodielectrics), in which exciton states can be formed[1)] (we discuss only exciton states of the Wannier-Mott type). It is clear, that besides interactwith the phonons excitons interact with the oscillations of the magnetic moments of the magnet. Our problem is the calculation of the establishment time $\tau_0^{(\text{es})}$ of exciton-magnon equilibrium and, consequently, the determination of conditions in which the equilibrium distribution (1) is valid. In view of the fact that the times to reach equilibrium is different for a system of excitons and for a system of ordered spins, the temperatures of the exciton and spin systems may differ from each other in the general case. At high but quite achievable exciton concentrations, a single temperature is established after a time of the same order as $\tau_0^{(\text{es})}$. A study the interactions of excitons with oscillations of the crystal magnetic induction density allows us to investigate effects connected with transitions of triplet exciton states into singlet exciton states. The transition of an exciton system to the equilibrium state is described by the nonequilibrium density matrix technique developed in[8,9] for quantum systems. The dynamic variable operators are conveniently written down in the second quantization representation.

2. THE HAMILTONIAN OF AN ELECTRON-HOLE BOUND STATE IN A MAGNET

As is well-known[10], a nonlocalized Wannier-Mott exciton can be represented in the effective-mass approximation as a bound electrostatically interacting electron-hole pair moving through the crystal. Such a model of the exciton describes quite well all the presently known magnetic effects occurring in a constant magnetic field, like the Zeeman splitting of exciton energy levels and diamagnetism. We use the same exciton model. Let an crystal exciton state be formed in a magnetic dielectric. A characteristic magnetic property of such a dielectric is the magnetic induction at an arbitrary crystal point $\mathbf{R}$, which we denote by $\mathbf{B}(\mathbf{R})$. Thus, since the electron and hole have spins

$$\mathbf{S}_e = \frac{\hbar}{2}\sigma_e, \qquad \mathbf{S}_h = \frac{\hbar}{2}\sigma_h,$$

we have to add to the ordinary interaction Hamiltonian of the electrons and holes

$$\mathscr{H}_0 = -\frac{\hbar^2\nabla_e^2}{2m_e^*} - \frac{\hbar^2\nabla_h^2}{2m_h^*} - \frac{e^2}{\varepsilon R_{eh}}, \tag{2}$$

[1)] A number of experiments on light absorption in antiferrodielectrics [6] have shown that in order to explain several optical effects it is necessary to take into account magnon-exciton interaction. Mott and Phillips [7] even assume that exciton states can be formed in the ferromagnets Fe, Co, and Ni.

which leads to a bound exciton state, the interaction of electron and hole spins with the magnetic induction:

$$\mathscr{H}_{int}=\frac{e\hbar}{2c}\left\{\frac{1}{m_e^*}\sigma_e\mathbf{B}(\mathbf{R}_e)-\frac{1}{m_h^*}\sigma_h\mathbf{B}(\mathbf{R}_h)\right\}. \tag{3}$$

The following notation is introduced in Eqs. (2), (3): m_e^* and m_h^* are the electron and hole effective masses, R_{eh} is their mutual distance, $e = -|e|$ is the electron charge, c is the light velocity, $\hbar$ is Planck's constant, and ϵ is the dielectric constant of the crystal.

Consider now a fluctuation of magnetic induction at point $\mathbf{R}$:

$$\mathbf{b}(\mathbf{R})=\mathbf{B}(\mathbf{R})-\langle\langle\mathbf{B}(\mathbf{R})\rangle\rangle\equiv\mathbf{B}(\mathbf{R})-\mathbf{B}_0,$$

where the brackets $\langle\langle \ldots \rangle\rangle$ denote an average over the equilibrium Gibbs state of the magnet. Such an averaging is meaningful if it is recognized that the establishment of an equilibrium Gibbs distribution in a magnet is due to strong exchange interactions of spins and occurs after a short time $\tau_S \sim 10^{-9}$–10^{-11} sec. We replace in Eq. (3) the induction $\mathbf{B}(\mathbf{R})$ by $\mathbf{b}(\mathbf{R}) + \mathbf{B}_0$. The terms containing the constant induction $\mathbf{B}_0$, which plays the role of a uniform magnetic field, cause effects such as the Zeeman effect. It is, therefore, convenient to combine them with the Hamiltonian $\mathscr{H}_0$. Choosing the direction of the vector $\mathbf{B}_0$ as the z axis, we represent the total Hamiltonian of the electron-hole system in the form

$$\begin{gathered}\mathscr{H}=\mathscr{H}_{0s}+\mathscr{H}_{int},\\ \mathscr{H}_{0s}=\mathscr{H}_0+g_{eff}B_0(\hat{L}_z+\hat{S}_z),\\ \mathscr{H}_{int}=\frac{e\hbar}{2c}\left\{\frac{1}{m_e^*}\sigma_e\mathbf{b}(\mathbf{R}_e)-\frac{1}{m_h^*}\sigma_h\mathbf{b}(\mathbf{R}_h)\right\},\end{gathered} \tag{4}$$

where g_{eff} is the effective gyromagnetic ratio of the pair, $\hat{L}_z$ is the projection of the orbital-momentum operator of the electron and the hole, and $\hat{S}_z$ is the projection of the total spin of the electron and the hole.

The effects due to the constant magnetic field, and described by the Hamiltonian $\mathscr{H}_{0s}$, are by now well known[4], and we shall not discuss them in detail here. g_{eff} is also assumed to be a known parameter. We write down the Hamiltonian $\mathscr{H}$ in the exciton occupation-number representation. Since the energy and wave function corresponding to a certain exciton quantum state are found from the equation

$$\mathscr{H}_{0s}\Psi=E\Psi, \tag{5}$$

where

$$E_f(\mathbf{k})=E_{nm;SS_z}(\mathbf{k})=E_G{}^S-\frac{\mu_{ex}e^4}{2\hbar^2\varepsilon^2n^2}+\hbar g_{eff}B_0(m+\sigma_z)+\frac{\hbar^2k^2}{2(m_e^*+m_h^*)},$$

$$\Psi_{nlm;SS_z;\mathbf{k}}(\mathbf{R},\ \rho;\ \sigma_e,\ \sigma_h)=\frac{1}{\sqrt{V}}e^{i\mathbf{kR}}F_{nlm}(\boldsymbol{\rho})\chi_{SS_z}(\sigma_e,\sigma_h), \tag{6}$$

the Hamiltonian $\mathscr{H}$ is in the second quantization is

$$H=H_0+H_{int}=\sum_\nu E_\nu b_\nu{}^+b_\nu+\sum_{\nu\nu'}W^{\nu'\nu}\overset{+}{b}_{\nu'}b_\nu,\qquad \nu\equiv\{f,\mathbf{k}\}. \tag{7}$$

The following notation is introduced in Eqs. (5) and (6): n, 1, and m are the main, orbital, and magnetic quantum numbers, respectively, μ_{ex} is the exciton effective mass, $\mathbf{R} = (m_e^*\mathbf{R}_e + m_h^*\mathbf{R}_h)/(m_e^* + m_h^*)$ is the center-of-mass coordinate of the bound pair, $\rho = \mathbf{R}_e - \mathbf{R}_h$ is the radius-vector of the electron and hole relative motion, F_{nlm} is the normalized modified hydrogenlike function for $z \to 1/\epsilon$, $\chi_{SS_z}(\sigma_e, \sigma_h)$ is the normalized spin function of the singlet (S = 0) or triplet (S = 1) state, and $E_G^{(S)} = \mu_{ex}e^4/2\hbar^2\epsilon^2n^2$ is the energy of forming a bound pair with spin S. The quantity $W^{\nu'\nu}$ in (7) is a sum of two terms, $W_1^{\nu'\nu}$ and $W_2^{\nu'\nu}$, where[11]

$$\begin{aligned}W_1{}^{\nu'\nu}&=\frac{1}{V}\sum_j\sum_{\sigma_e,\sigma_h}\chi^*_{SS'_z}(\sigma_e,\sigma_h)\hat{\sigma}_1{}^j\chi_{SS_z}(\sigma_e,\sigma_h)\int e^{i(\mathbf{k}-\mathbf{k}')\mathbf{R}}F_{\alpha}{}^*(\rho)\\&\qquad\times A_1{}^j(\mathbf{R},\ \boldsymbol{\rho})F_\alpha(\boldsymbol{\rho})\,d\boldsymbol{\rho}\,d\mathbf{R},\\ W_2{}^{\nu'\nu}&=\frac{1}{V}\sum_j\sum_{\sigma_e,\sigma_h}\chi^*_{S'S'_z}(\sigma_e,\sigma_h)\hat{\sigma}_2{}^j\chi_{SS_z}(\sigma_e,\sigma_h)\int e^{i(\mathbf{k}-\mathbf{k}')\mathbf{R}}F_{\alpha'}{}^*(\rho)\\&\qquad\times A_2{}^j(\mathbf{R})F_\alpha(\boldsymbol{\rho})\,d\boldsymbol{\rho}\,d\mathbf{R},\end{aligned} \tag{8$'$}$$

and

$$\mathbf{A}_1(\mathbf{R},\ \rho)=\frac{e\hbar}{4c}\left\{\left(\frac{1}{m_e^*}-\frac{1}{m_h^*}\right)\mathbf{b}(\mathbf{R})+\frac{1}{m_e^*+m_h^*}\frac{\partial\mathbf{b}(\mathbf{R})}{\partial R^i}\rho^i\right\}, \tag{8$''$}$$

$$\mathbf{A}_2(\mathbf{R})=\frac{e\hbar}{4\mu_{ex}c}\mathbf{b}(\mathbf{R}).$$

The operators $\hat{\sigma}_1 = \hat{\sigma}_e + \hat{\sigma}_h$ and $\hat{\sigma}_2 = \hat{\sigma}_e - \hat{\sigma}_h$ in the expressions for $W_1^{\nu'\nu}$ and $W_2^{\nu'\nu}$ act on the spin functions $\chi_{SS}(\sigma_e, \sigma_h)$ in such a manner that the interaction Hamiltonian

$$\hat{H}_{1int}=\sum_{\nu'\nu}W_1^{\nu'\nu}\,b_{\nu'}{}^+b_\nu$$

produces transitions of triplet excitons from one quantum state into another, and the Hamiltonian

$$\hat{H}_{2int}=\sum_{\nu'\nu}W_2^{\nu'\nu}\,b_{\nu'}{}^+b_\nu$$

produces transitions of triplet and singlet excitons into each other.

We turn now to finding the establishment time of the exciton equilibrium distribution function in a magnet in a given quantum state. We assume at the same time that equilibrium of the excitons with the phonons in the system has already been established ($\tau_0^{(ef)} \sim 10^{-12}$ sec). We consider exciton concentrations such that the mutual interaction of excitons can be neglected, but the Bose commutation relations are still satisfied for excitons[3]. It is also assumed, that the wave functions of the excitons do not overlap, and consequently no correlation capable of leading to exciton "metalization" exists between the excitons.

3. ESTABLISHMENT OF THE EXCITON EQUILIBRIUM DISTRIBUTION FUNCTION

For the study of interactions of excitons with a magnetic system we note that the equilibrium state in the spin system is achieved after a time $\tau_S \sim 10^{-10}$ sec. Therefore, the general density matrix can be represented in the form of a product of the equilibrium density matrix of the spin system ρ_S and the nonequilibrium exciton density matrix ρ_{ex}. At the same time the Hamiltonian of the system of excitons and magnons equals

$$\hat{\mathscr{H}}=\hat{H}_s+\hat{H}=\hat{H}_s+\hat{H}_0+\hat{H}_{int},$$

where $\hat{H}_S$ is the Hamiltonian of the spin system. Using now the results of[9], we write down immediately the kinetic equation for the time variation of the exciton-number density $n_\nu = \mathrm{Tr}(\rho_{ex}\rho_S\hat{n}_\nu)$ and for the quantity $H_S = \mathrm{Tr}(\rho_{ex}\rho_S\hat{H}_S) \equiv \langle\langle \hat{H}_S \rangle\rangle$:

$$\frac{\partial n_\nu}{\partial t}=2\pi\sum_{\nu_1\nu_2}\delta_{\nu\nu_2}I_{\nu_1\nu_2}(\omega_{\nu_1\nu_2})\left\{n_{\nu_1}(1+n_{\nu_2})\exp\left(\frac{\omega_{\nu_1\nu_2}}{T}\right)-n_{\nu_2}(1+n_{\nu_1})\right\}, \tag{9}$$

$$\frac{\partial H_s}{\partial t} = C_s \frac{\partial T}{\partial t} = 2\pi \sum_{\nu_1\nu_2} \omega_{\nu_1\nu_2} I_{\nu_1\nu_2}(\omega_{\nu_1\nu_2}) n_{\nu_2}(1 + n_{\nu_1}). \qquad (9')$$

Equation (9′) describes the exciton action on the spin system and enables us to study the temperature equalization of the exciton and spin systems. The spectral function $I_{\nu_1\nu_2}(\omega_{\nu_1\nu_2})$ characterizing the magnet is determined by the equation

$$I_{\nu_1\nu_2}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau e^{i\omega t} \langle\langle W_\tau^{\nu_2\nu_1} W^{\nu_1\nu_2} \rangle\rangle, \quad W_\tau = e^{i\hat{H}_s\tau} W e^{-i\hat{H}_s\tau}. \qquad (10)$$

Taking into account the determination of the quantities $W^{\nu'\nu}$ (Eqs. (8′) and (8″)), we rewrite relation (10) in the form

$$I_{\nu\nu'}(\omega) = D_{\alpha\alpha'} B^{SS'}_{S_z S'_z}(\mathbf{q}, \omega), \quad \mathbf{q} = \mathbf{k} - \mathbf{k}', \quad \alpha \equiv \{nlm\}.$$

For $\nu \rightarrow \nu'$ transitions occurring without a change of the total spin of the bound pair, we have

$$D_{\alpha\alpha'} = \frac{1}{V}\left(\frac{e\hbar}{4c}\right)^2 \left\{\left(\frac{1}{m_e^*} - \frac{1}{m_h^*}\right)^2 \delta_{\alpha\alpha'} + \frac{a^2 q^i q^{i'} \Phi^i_{\alpha\alpha'} \Phi^{i'}_{\alpha\alpha'}}{(m_e^* + m_h^*)^2}\right\}, \qquad (11)$$

$$B^{SS'}_{S_z S_z'}(\mathbf{q}, \omega) = \delta_{SS'} \sum_{jj'} \langle SS_z' | \hat{\sigma}_1^{\,j} | SS_z \rangle^* \langle SS_z' | \hat{\sigma}_1^{\,j'} | SS_z \rangle \langle\langle b^j b^{j'} \rangle\rangle_{\mathbf{q}\omega},$$

where

$$\langle SS_{z'} | \hat{\sigma}_1{}^j | SS_z \rangle = \sum_{\sigma_e\sigma_h} \chi^*_{SS'_z}(\sigma_e, \sigma_h) \hat{\sigma}_1{}^j \chi_{SS'_z}(\sigma_e, \sigma_h),$$

$$\Phi^i_{\alpha\alpha'} = \frac{1}{a} \int F_{\alpha'}{}^*(\rho) \rho^i F_\alpha(\rho) d\rho, \qquad (12)$$

$$\langle\langle b^j b^{j'} \rangle\rangle_{\mathbf{q},\omega} = \frac{1}{2\pi} \int e^{i(\mathbf{qR} - \omega\tau)} \langle\langle b_\tau{}^j(R) b^{j'}(0) \rangle\rangle \, d\tau \, d\mathbf{R}.$$

Since the correlation function $\langle\langle b^j b^{j'} \rangle\rangle_{\mathbf{q},\omega}$ is proportional to a δ-function of the form $\delta(\omega^2 - \omega_S^2(\mathbf{q}))$ ($\omega_S(\mathbf{q})$ is one of the branches of the spin oscillations), the strongest interaction between excitons and fluctuations of the magnetic induction density will be observed in absorption or emission of spin waves by the excitons, i.e. under the condition

$$E_f(\mathbf{k}) - E_{f'}(\mathbf{k}') = \pm\hbar\omega_s(\mathbf{q}), \qquad \mathbf{q} = \mathbf{k} \mp \mathbf{k}'. \qquad (13)$$

Until now we did not specify the form of the magnetic crystal. Therefore, the equations obtained above are applicable both for ferrodielectrics and for antiferrodielectrics. For further calculations it is necessary to know the explicit form of the correlation function.

We calculate, for example, from Eq. (9), the establishment time $\tau^{(es)}$ of the exciton equilibrium distribution function for an antiferrodielectric of the "easy axis" type. Using Eqs. (10)—(12) and taking the expression for $\langle\langle b^j b^{j'} \rangle\rangle_{\mathbf{q},\omega}$ from[12], we have, in case of weak magnetic fields and a small anisotropy energy for the transitions $S = S'$, $S_z = S_z' = 1$

$$\frac{1}{\tau^{(es)}_{\nu 0}} \approx \frac{16\pi^2}{5} \frac{\mu^2}{a^3\hbar} \frac{\Theta_N}{\Theta_{ex}} \frac{(ak)^2}{\eta} \left[1 + 2\exp\left(-\frac{\hbar\omega_s(0)}{T}\right)\right], \qquad (14)$$

$$\frac{1}{4}\frac{\Theta_N{}^2}{\Theta_{ex}} \ll T \ll \hbar\omega_s(0),$$

$$\frac{1}{\tau^{(es)}_{\nu 0}} \approx \frac{2\pi^2}{3} \frac{\mu^2}{a^3\hbar} \frac{T}{\Theta_{ex}} \frac{ak}{\eta}, \qquad \Theta_N \gg T \gg \hbar\omega_s(0).$$

In case of strong fields we have, from[13], for the same transitions

$$\frac{1}{\tau^{(es)}_{\nu 0}} \approx 8\pi^2 \frac{\mu^2\eta}{a^3\hbar} \left(\frac{\hbar g M_0}{\Theta_N}\right)^2 \frac{T}{\Theta_{ex}} ak, \qquad \Theta_N \gg T. \qquad (15)$$

The following notation was used in Eqs. (14) and (15):

$$\mu = \frac{e\hbar}{2c}\left(\frac{1}{m_e^*} - \frac{1}{m_h^*}\right), \qquad \Theta_{ex} = \frac{\hbar^2}{2(m_e^* + m_h^*)a^2},$$

$$\Theta_N = \hbar g M_0 \sqrt{2\eta(\alpha - \alpha')} \frac{1}{a},$$

where a is the lattice constant, and η, α, and α' are the exchange constants.

Transitions to other spin states with $S = S'$ can be similarly described. Quantum transitions with $S \neq S'$ are generally realized by transfering from the electromagnetic wave to the system the energy necessary to compensate for the large difference $E_G^{(S)} E_G^{(S')}$, which, according to definition (5), arises in the energy conservation law (13).

Let us estimate numerically the values of $\tau^{(es)}_{\nu 0}$ for magnets having low Neel temperatures (for example, for the antiferrodielectric $RbMnF_3$, $\theta_N \sim 82$°K). At T ~ 10°K we have for weak fields $\tau^{(es)}_{\nu 0} \sim 10^{-5}$ sec, and for strong fields $\tau^{(es)}_{\nu 0} \sim 10^{-6}$ sec (the somewhat large values of $\tau^{-1}_{\nu 0}$ for strong magnetic fields is connected with the high sensitivity of antiferrodielectrics to external fields after spin flipping). Thus, the final exciton equilibrium distribution (1) is established in the magnet in a time $\tau^{(es)}_{\nu 0} \sim 10^{-5}$—$10^{-6}$ sec and is maintained up to a time $\tau \sim \tau_{em}$.

Let us dwell briefly on the influence of the excitons on the spin system. At exciton concentrations of the order of 10^{16} cm^{-3}, when the Bose commutation relations still hold, one obtains from Eq. (9′) the following equation for the establishment time of the equilibrium temperatures of the spin and exciton systems (strong fields):

$$\frac{1}{\tau_T} \approx \frac{2\pi}{3} \frac{\mu^2}{a^3\hbar} \frac{\eta\varkappa}{\zeta_s} \frac{(\hbar g M_0)^2}{\Theta_N (\Theta_{ex} T)^{1/2}} \qquad \Theta_N \gg T \gg T_{deg}.$$

Here $\zeta_S = C_S/k_0N$ is the spin heat capacity per crystal cell, $T_{deg} \sim \Theta_{ex}\kappa^{2/3}$, and κ is the relative exciton concentration.) The numerical value of τ_T at T ~ 10°K and $\zeta_S \sim 1$ is about 10^{-5}—10^{-6} sec, which coincides with the establishment time of the distribution $n_\nu^{(0)}$.

4. CONCLUSION

The foregoing scheme for calculating the establishment times of the exciton equilibrium distribution is applicable to any magnet, antiferrodielectric or ferrodielectric, in which exciton excitations of the crystal can occur in addition to spin excitations. Although the case of nonlocalized excitons was considered, the scheme can be generalized also the case of Frenkel excitons (in crystals like $KNiF_3$, K_2NiF_4, NiF_2, MnO, and others[14]). At the same time it may happen that the stronger interaction will be not the interaction of the excitons with the magnetic induction fluctuations, but the exchange interaction between electrons that cause spin excitations of the crystal and electrons that cause exciton excitations of the crystal.

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