

# Amorphization of Heisenberg magnets

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Amorphization is investigated for crystalline, magnetically ordered structures, each of which is characterized by two exchange parameters of different magnitudes and signs. The amorphous magnet is treated within the framework of the lattice model with fluctuating exchange bonds, by use of the coherent-potential approximation. Conditions are found under which the amorphized magnetic can be described as an effective ferromagnet; the coherent exchange parameter and the modified density of magnon states are also found. It is shown that amorphization of antiferromagnets may, under certain conditions, be accompanied by a change of the type of magnetic order, an increase of the temperature of the magnetic phase transition, and the appearance of singularities in the density of magnon states.

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## 1. INTRODUCTION

A broad class of magnetically ordered structures is known, which is characterized by anisotropy of the distribution of exchange bonds: for example, quasi-one-dimensional and quasi-two-dimensional magnets.<sup>[1]</sup> In the simplest case, such structures are described by two exchange parameters; the type of magnetic order and the temperature  $T_C$  of the magnetic phase transition in the crystal are determined by the weak exchange that binds the magnetic chains or planes. Amorphization of such structures may be accompanied by a considerable increase of the temperature of the magnetic phase transition and also by a change of the type of magnetic order, since in this case the exchange is averaged and it is rather this averaged exchange that determines the type and temperature of magnetic ordering.<sup>[2,3]</sup> Consequently, amorphization of this class of magnetically ordered structures may prove to be an important method of obtaining new magnetic materials.

The magnetic state of nonconducting magnets is described sufficiently well by the spin Hamiltonian<sup>[4]</sup>

$$\mathcal{H} = - \sum_{i,m} A_{im} S_i S_m, \quad A_{ii} = 0, \quad (1)$$

where  $A_{im}$  are the effective exchange parameters. Because of the short-range character of exchange, it may be supposed that the  $A_{im}$  differ from zero only for nearest neighbors (NN). In ideal crystals, because of translational invariance,  $A_{im} \rightarrow A_0(\mathbf{f} - \mathbf{m}) = A_0(\mathbf{h})$ . In general the exchange parameter  $A_0(\mathbf{h})$  depends on the orientation of the exchange bond in the crystal. For example, for quasi-low-dimensional crystals, in the simplest case, it is necessary to introduce two exchange parameters,  $A_0(\mathbf{h}_1) \equiv J$  and  $A_0(\mathbf{h}_2) \equiv K$ . In a crystal this automatically leads to anisotropy of the distribution of exchange bonds  $J$  and  $K$ . The anisotropy of the distribution of exchange is characterized by the parameter  $\lambda = K/J$ . In quasi-low-dimensional magnets,  $|\lambda| \ll 1$ , and the temperature  $T_C = T_C(|\lambda|)$ ;  $T_C(0) = 0$ .

The magnetic properties of an amorphous material can be described approximately within the framework of the lattice model<sup>[5]</sup> with the Hamiltonian (1), where

now the  $A_{im}$  are fluctuating exchange parameters, different from zero only for NN. In general the  $A_{im}$  fluctuate in magnitude and in sign. One of the methods for theoretical investigation of amorphous magnets is the coherent-potential approximation (CPA). In this method, the amorphous magnet is approximated by a certain ideal crystal, in which the exchange between NN is represented by a coherent exchange parameter  $A_c(\mathbf{f} - \mathbf{m}, E)$ . This parameter is found from the condition for absence, on the average, of spin-wave scattering, and it is a function of the excitation energy  $E$ .

The problem of the present paper is the development of a theory of amorphization of magnetically ordered crystals with anisotropy of the distribution of exchange bonds. In a strict formulation, this problem is insolubly complicated. Therefore we shall use here the lattice model of an amorphous magnet and the CPA. For the case of amorphization of a ferromagnet, the solution of the problem was given in a previous paper.<sup>[3]</sup> The generalization of the theory to the case when the exchange parameters  $J$  and  $K$  have different signs turns out to be significantly nontrivial, and it is this case that is the principal goal of the present article.

## 2. GREEN'S FUNCTIONS AND CHARACTERISTICS OF IDEAL CRYSTALS WITH ANISOTROPICALLY DISTRIBUTED EXCHANGE BONDS

The equation for Green's function

$$G_{im}(E) = \langle\langle S_i^+ | S_m^- \rangle\rangle_E$$

with use of Tyablikov's<sup>[4]</sup> decoupling, has the form

$$\sum_{\mathbf{m}} [E \delta_{im} - H_{im}] G_{m\mathbf{n}}(E) = \sigma_i \delta_{i\mathbf{n}}, \quad (2)$$

where

$$H_{im} = \delta_{im} \left( \sum_{\mathbf{b}} A_{i,i+\mathbf{b}} \sigma_{i+\mathbf{b}} \right) - \sigma_i A_{im}, \quad \sigma_i = \langle 2S_i^+ \rangle_i$$

$\langle \dots \rangle$  is a thermodynamic average. The representation (2) is correct for description both of ideal crystals

and of disordered magnets (the lattice model). For definiteness we shall consider a simple cubic lattice (SCL) with  $z = 6$  NN and with spin  $S = \frac{1}{2}$ . In this case the components of  $S_i$  are expressed in terms of Pauli operators:

$$S_i^+ = b_i, \quad S_i^- = b_i^\dagger, \quad 2S_i^z = 1 - 2b_i^\dagger b_i.$$

To facilitate comparison of the properties of amorphous and of ideal materials, we shall first consider the principal characteristics of ideal magnetically ordered crystals. For an ideal crystal, the matrix  $\tilde{H}_{fm}$  becomes translationally invariant, and the Green's function is found by Fourier transformation of equation (2). In the most general case of an anisotropic distribution of exchange bounds in a SCL,

$$A_0(\mathbf{h}_i) = A_0(-\mathbf{h}_i) = A_i, \quad \mathbf{h}_i = \mathbf{e}_i a, \quad (3)$$

where  $\mathbf{e}_i$  are unit vectors in the directions  $i = x, y, z$ , and  $a$  is the lattice parameter.

Ferromagnetic ordering occurs when  $A_i > 0$ . In this case  $\sigma_f = \sigma$ ,

$$G_{\mathbf{m}-\mathbf{n}}^0(E) = \frac{1}{N} \sum_{\mathbf{k}} e^{i\mathbf{k}(\mathbf{m}-\mathbf{n})} \frac{\sigma}{E - \sigma E_0(\mathbf{k})}, \quad E_0(\mathbf{k}) = \sum_i 2A_i(1 - c_i) \quad (4)$$

( $c_i = \cos k_i a$ ), and the Curie temperature  $T_C^0$  is estimated by the relation<sup>[4]</sup>

$$\frac{1}{2T_C^0} = \frac{1}{N} \sum_{\mathbf{k}} \frac{1}{E_0(\mathbf{k})} = \int \frac{g_0(E)}{E} dE, \quad (5)$$

where  $g_0(E)$  is the density of spin-wave states corresponding to the dispersion law  $E_0(\mathbf{k})$ .

If  $A_1 = A_2 = A_3 = A$ , the spectrum is isotropic:

$$E_0(\mathbf{k}) = 6A \epsilon_{\mathbf{k}}, \quad \epsilon_{\mathbf{k}} = 1 - \gamma_{\mathbf{k}}.$$

In the presence of symmetry in a plane, when  $A_1 = A_2 = J$  and  $A_3 = K$ , the Curie temperature depends significantly on the ratio  $K/J = \lambda$  or  $J/K = \eta$ . The limiting cases of an anisotropic distribution of exchange interactions,  $\lambda \ll 1$  and  $\eta \ll 1$ , are called, respectively, quasi-two-dimensional and quasi-one-dimensional ordering.

It is easily shown that for  $K \rightarrow 0$  ( $\lambda \rightarrow 0$ ) or  $J \rightarrow 0$  ( $\eta \rightarrow 0$ ) the integral (5) diverges; that is,  $T_C^0(\lambda = 0) = T_C^0(\eta = 0) = 0$ , which indicates the impossibility of plane or one-dimensional ferromagnetic (FM) ordering, in agreement with the theorem of Mermin and Wagner.<sup>[6]</sup> The function  $T_C^0(\lambda)$  is given by Lines.<sup>[7]</sup>

Antiferromagnetic (AF) ordering in SCL is possible when (a)  $A_i < 0$ , (b)  $A_1 > 0$ ,  $A_2 > 0$ ,  $A_3 < 0$  (FM planes coupled antiferromagnetically), or (c)  $A_1 < 0$ ,  $A_2 < 0$ ,  $A_3 > 0$  (AF planes coupled ferromagnetically). In AF ordering the crystal splits into two sublattices:  $F$  ( $f \in F$ ) and  $M$  ( $m \in M$ );  $N_F = N_M = N/2$ , and

$$\sigma_f = \frac{1}{N/2} \sum_i \langle 2S_i^z \rangle_i = \sigma > 0, \quad \sigma_m = \frac{1}{N/2} \sum_i \langle 2S_i^z \rangle_i = -\sigma.$$

All cases of AF ordering can be treated in a single form by introducing the quantities

$$\alpha_i^{(F)} = \begin{cases} 1, & A_i > 0, \\ 0, & A_i < 0, \end{cases} \quad \alpha_i^{(M)} = \begin{cases} 0, & A_i > 0, \\ 1, & A_i < 0, \end{cases} \quad (6)$$

which enable us to establish the membership of sites

$f + \mathbf{h}_i$  and  $m + \mathbf{h}_i$  in sublattices  $F$  and  $M$ . On performing a Fourier transformation by sublattices, we find the matrix Green's function:

$$\begin{pmatrix} \langle \langle b_F(\mathbf{q}) | b_{F^+}(\mathbf{q}) \rangle \rangle_E \langle \langle b_M(\mathbf{q}) | b_{M^+}(\mathbf{q}) \rangle \rangle_E \\ \langle \langle b_M(\mathbf{q}) | b_{F^+}(\mathbf{q}) \rangle \rangle_E \langle \langle b_M(\mathbf{q}) | b_{M^+}(\mathbf{q}) \rangle \rangle_E \end{pmatrix} = \frac{\sigma}{E^2 - \sigma^2 E_0^2(\mathbf{q})} \begin{pmatrix} E + \sigma \omega_1(\mathbf{q}) & \sigma \omega_2(\mathbf{q}) \\ \sigma \omega_2(\mathbf{q}) & -E + \sigma \omega_1(\mathbf{q}) \end{pmatrix}. \quad (7)$$

Here  $\sigma E_0(\mathbf{q})$  is the spectrum of AF spin waves:

$$E_0^2(\mathbf{q}) = \omega_1^2(\mathbf{q}) - \omega_2^2(\mathbf{q}) \geq 0, \quad (8)$$

$$\omega_1(\mathbf{q}) = \sum_i 2A_i [(1 - c_i) \alpha_i^{(F)} - \alpha_i^{(M)}], \quad \omega_2(\mathbf{q}) = \sum_i 2A_i c_i \alpha_i^{(M)}.$$

By using the spectral theorem, we obtain by the standard method an equation for the magnetization  $\sigma$  of a sublattice and for the estimate of the Néel temperature  $T_N^0$ :

$$\frac{1}{2T_N^0} = \frac{1}{N/2} \sum_{\mathbf{q}} \frac{\omega_1(\mathbf{q})}{E_0^2(\mathbf{q})} = \int g_0^{\text{AF}}(E) \frac{\omega_1(E)}{E^2} dE. \quad (9)$$

In the case of plane symmetry,  $A_1 = A_2 \equiv J$ ,  $A_3 \equiv K$ , the Néel temperature depends substantially on  $|\lambda| \equiv |K|/J$  (or on  $|\eta| \equiv J/|K|$ ) for all the cases enumerated, and it vanishes for  $|\lambda| \rightarrow 0$  ( $|\eta| \rightarrow 0$ ), which indicates the impossibility of AF ordering in two-dimensional (one-dimensional) systems. Thus quasi-two-dimensional,  $|\lambda| \ll 1$ , or quasi-one-dimensional,  $|\eta| \ll 1$ , ordering of ideal magnets is characterized by small values of the temperature of the magnetic phase transition.

### 3. THE PROBLEM OF THE GROUND STATE OF AN AMORPHIZED MAGNET

In a description of the amorphized state of a material, we shall start from the following postulates:

- 1) amorphization of the crystal leads to complete isotropy of the macroscopic properties of the material;
- 2) in the amorphized material, on the average, the short-range order characteristic of the original ideal crystal is retained.

The plausibility of these postulates is confirmed by experimental facts.<sup>[8,9]</sup> Investigations show that the amorphous structure contains coherent regions of dimensions  $\sim 5 \text{ \AA}$  and that it is not characterized by closest packing of the atoms. Since the exchange interactions that lead to long-range magnetic order at  $T < T_C$  are, in magnetic dielectrics, short-ranged and consequently sensitive to a local change of the short-range crystallochemical order, it is natural to expect, as a consequence of amorphization of nonconducting magnets, not only mixing of different exchanges but also fluctuations of them. Besides this, there occur fluctuations of the effective spin at the sites, for example as a result of fluctuations of the density of the material.

As has already been mentioned in the Introduction, we shall describe an amorphous magnet within the framework of the lattice model with the spin Hamiltonian (1), with fluctuating exchange parameters  $A_{fm}$ .

We shall discuss the character of the ground state ( $T=0$ ) of an amorphous magnet.

I. If in an amorphous magnet the fluctuating exchange parameters are of a single sign for an arbitrary pair of NN ( $\mathbf{l}, \mathbf{m}$ ), then when  $A_{lm} > 0$  the ground state is ferromagnetic, and when  $A_{lm} < 0$  antiferromagnetic.

This statement is completely obvious. The first of these cases was treated in Ref. 3; the generalization to the second case is trivial. In the general case, when the exchange parameters fluctuate in magnitude and in sign, the magnetic ground state will depend on the relation of the magnitudes and numbers of positive and negative exchange bonds.

II. On an alternating lattice, let there be prescribed a random isotropic distribution of antiferromagnetic ( $K < 0$ ) and ferromagnetic ( $J > 0$ ) bonds between NN, in concentrations  $\nu < \nu_{cr}(|\lambda|, z)$ , where  $|\lambda| = |K|/J$ , the ground state of the system possesses long-range FM order, with relative magnetization  $\bar{\sigma} < 1$ .

We shall prove this statement. In a lattice consisting of  $N$  sites, the total number of bonds is  $N_b = N/2$ , where  $z$  is the number of NN. At each site of the lattice there is a spin of magnitude  $S = \frac{1}{2}$ ; in the ground state,  $S_z^* = +\frac{1}{2}$  or  $-\frac{1}{2}$ . We shall consider the energy  $E_{am}$  of the amorphous material, for a prescribed random isotropic distribution of  $J$  and  $K$  bonds, as a function of the relative magnetization

$$\bar{\sigma} = (N_+ - N_-)/N, \quad N_+ + N_- = N,$$

where  $N_{\pm}$  is the number of atoms with projections  $S_z^* = \pm \frac{1}{2}$ .

For complete FM ordering,

$$\frac{E_{am}(\bar{\sigma}=1)}{S^2 N_b} = \varepsilon_{am}(1) = -J(1-\nu) + |K|\nu = -J[1-\nu - |\lambda|\nu], \quad (10a)$$

all the  $J$  bonds are energetically advantageous ("right"), and all the  $K$  bonds "wrong." Since in an alternating lattice AF ordering is possible, in this case

$$\varepsilon_{am}(0) = +J[1-\nu - |\lambda|\nu], \quad (10b)$$

with all the  $K$  bonds right and  $J$  bonds wrong. Only for an ideal crystal with an anisotropic translation-invariant distribution of bonds does the energy reach the absolute minimum for AF ordering of the spins:

$$\varepsilon_{id}(0) = -J[1-\nu_0 + |\lambda|\nu_0], \quad (10c)$$

where  $\nu_0 = \frac{1}{3}$  for FM ordering in the  $xy$  plane (case 10b)) or  $\frac{2}{3}$  for AF ordering in the  $xy$  plane (case 10c) in SCL. For such ideal AF, all bonds are right.

The further proof of the statement divides into two cases.

1. When  $1 - \nu - |\lambda|\nu > 0$  or

$$|\lambda| < (1-\nu)/\nu, \quad (11)$$

then  $\varepsilon_{am}(0) > \varepsilon_{am}(1)$ . In the state with  $\bar{\sigma} = 1$ , it is possible for the spins on certain lattice sites to reverse, tending to decrease the concentration of wrong

$K$  bonds. In general this leads to the appearance of additional wrong  $J$  bonds. The energy of a state with  $\bar{\sigma} < 1$  is

$$\varepsilon_{am}(\bar{\sigma}) = -J[\nu_J^0 - n_J + |\lambda|(\nu_K^0 - n_K)], \quad (12)$$

where  $\nu_J^0$  and  $\nu_K^0$  are the concentrations of right  $J$  and  $K$  bonds;  $n_J$  and  $n_K$  are the concentrations of wrong  $J$  and  $K$  bonds, and

$$\nu_J^0 + n_J = 1 - \nu, \quad \nu_K^0 + n_K = \nu. \quad (13)$$

It is obvious that in an amorphous material  $0 \leq n_J \leq 1 - \nu$ ,  $0 \leq n_K \leq \nu$ .

The statement reduces to a proof of the correctness of the inequality  $\varepsilon_{am}(1) > \varepsilon_{am}(\bar{\sigma})$ , which is equivalent to

$$|\lambda|\nu > n_J + |\lambda|n_K. \quad (14)$$

If short-range order is retained in the amorphous material, i.e.,  $\nu = \nu_0$ , then the energy difference is

$$\varepsilon_{am}(\bar{\sigma}) - \varepsilon_{id}(0) = \Delta E = 2J[n_J + |\lambda|n_K] > 0.$$

But the energy minimum of an amorphized material is attained at minimum concentrations of wrong  $J$  and  $K$  bonds:  $n_J = (n_J)_{min}$  and  $n_K = (n_K)_{min}$ . Such a material will be an effective FM with  $\bar{\sigma} \leq 1$ . On combining (11) and (14), we find that the inequality

$$\varepsilon_{am}(\bar{\sigma}) < \varepsilon_{am}(1) < \varepsilon_{am}(0)$$

is satisfied if the following one is:

$$\frac{n_J}{1-n_K} < |\lambda| < \frac{1-\nu}{\nu}. \quad (15)$$

The limiting case  $|\lambda| \rightarrow 0$  ( $n_J \rightarrow 0$ ) corresponds to breaking of the AF  $K$  bonds. It is known from percolation theory<sup>[10]</sup> that long-range FM order occurs only when  $\nu_J^0 > \nu_{cr}^F$ , where  $\nu_{cr}^F \approx 2/z$  is the "ferromagnetic percolation" concentration. Thus

$$\nu_{cr}(|\lambda|=0) = 1 - \nu_{cr}^F \approx 1 - 2/z.$$

The magnetization at  $|\lambda| = 0$ , as a function of  $\nu$ , behaves thus:

$$\bar{\sigma}(\nu, |\lambda|=0) = \begin{cases} 1 - P_s(z-1) - P_s(z), & \nu < 1 \\ 0, & \nu \geq \nu_{cr} \approx 1 - 2/z \end{cases}. \quad (16)$$

Here

$$P_s(m) = \frac{z!}{m!(z-m)!} \nu^m (1-\nu)^{z-m}.$$

2. When  $1 - \nu - |\lambda|\nu < 0$ , which corresponds to

$$|\lambda| > (1-\nu)/\nu, \quad (17)$$

the energy  $\varepsilon_{am}(0)$  of the AF amorphous state  $< \varepsilon_{am}(1)$ . If in such an AF state we reverse the spins at certain sites of the lattice in order to decrease the concentration of wrong  $J$  bonds, this will in general lead to the appearance of additional wrong  $K$  bonds. It is clear that if  $\nu$  is less than the "antiferromagnetic percolation" concentration, then an effective FM order will be established in the medium, with  $\bar{\sigma} \neq 0$ . Here  $\varepsilon_{am}(0) > \varepsilon_{am}$

$\bar{\sigma}$ ), which, if we take account of (10b), 12, and (17), corresponds to the inequality

$$\frac{1-\nu}{\nu} < |\lambda| < \frac{1-\nu-n_K}{n_K}. \quad (18)$$

We shall consider the limiting case  $|\lambda| \rightarrow \infty$  ( $n_K \rightarrow 0$ ). At small  $\nu_K^0 \ll 1$ , it is advantageous to retain the AF spin arrangement only within the bounds of individual sites and their nearest neighborhood. It is energetically advantageous to polarize the rest of the spins; that is, to form a FM matrix. With increase of the concentration  $\nu_K^0$ , the process of polarization becomes more and more difficult; and when

$$\nu_K^0 = \nu_{cr}^{AF} \approx 2/z \equiv \nu_{cr}(|\lambda| = \infty),$$

corresponding to the "AF percolation" concentration, the formation of a FM matrix is impossible and  $\bar{\sigma} = 0$ .

For  $|\lambda| = \infty$ , the magnetization as a function of  $\nu$  behaves thus:

$$\bar{\sigma}(\nu, |\lambda| = \infty) = \begin{cases} 1-2\nu, & \nu \ll 1 \\ 0, & \nu \geq \nu_{cr} \approx 2/z. \end{cases} \quad (19)$$

As is evident, the character of the variation of  $\bar{\sigma}$  with  $\nu$  is strongly dependent on the value of  $|\lambda|$ . The behavior of  $\bar{\sigma}(\nu, |\lambda|)$  for  $|\lambda| = 0$  and for  $|\lambda| = \infty$  is shown in Fig. 1. For  $|\lambda| \rightarrow \infty$  and  $\nu \geq \nu_{cr}(\infty)$ , the amorphized material remains AF. We note that when  $|\lambda| = (1-\nu)/\nu$  there is degeneracy:  $\varepsilon_{am}(0) = \varepsilon_{am}(1)$ . For  $\nu_0 = \frac{1}{3}$ , the corresponding value of  $|\lambda|$  is 2.

Thus we may draw the completely general conclusion that on amorphization of a magnetically ordered crystal, when the fluctuating exchange parameters have different signs, the amorphous magnet under certain conditions will represent an effective FM with  $\bar{\sigma} \neq 0$ . In the following sections, on the basis of a solution of the self-consistent problem of an effective FM exchange parameter in an amorphous magnet with a prescribed exchange distribution function, the region of existence of ferromagnetism on the  $(\nu, \lambda)$  diagram will be found, and also the density of magnon states and  $\bar{\sigma}(T)/\bar{\sigma}(0)$ . The question of the value of  $\bar{\sigma}(0)$  requires consideration of a concrete model, with inclusion of numerical combinatorial calculations.

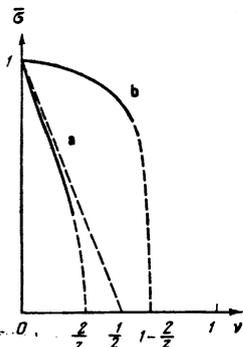


FIG. 1. Variation of the relative magnetization  $\bar{\sigma}$  with the concentration  $\nu$  of antiferromagnetic  $K$  bonds: a, when  $|\lambda| = |K|/J = \infty$ ; b, when  $|\lambda| = 0$ .

#### 4. DESCRIPTION OF AMORPHOUS MAGNETS BY THE COHERENT-POTENTIAL METHOD

The coherent-potential method is based on representation of a disordered material as a certain effective translation-invariant medium, in which the exchange interactions are brought about by one or more self-consistent potentials.<sup>[11-13]</sup> In the preceding section it was shown that under certain conditions the ground state of an amorphized AF may turn out to be ferromagnetic. With application to this case, we shall describe the amorphous material as an effective translation-invariant isotropic ferromagnet with  $\bar{\sigma}(0) \leq 1$ , in which the exchange interactions are determined by a coherent parameter  $A_c(f-m)$ . The method of finding  $A_c$  has been set forth in detail in a previous paper of the authors.<sup>[3]</sup> Here we shall discuss the key aspects of the CPA.

On introducing the fluctuations with respect to the coherent-potential parameter

$$A_{fm} = A_c(f-m) + [A_{fm} - A_c(f-m)] = A_c(f-m) + \xi_{fm} \quad (20)$$

and using the definition (3), where  $\sigma_f = \bar{\sigma}$ , we get

$$\begin{aligned} H_{fm} &= \bar{\sigma} [H_c(f-m) + V_{fm}], \\ H_c(f-m) &= \delta_{fm} \sum_n A_c(h) - A_c(f-m), \\ V_{fm} &= \delta_{fm} \sum_n \xi_{f,h} - \xi_{fm}. \end{aligned} \quad (21)$$

Equation (2) takes the form

$$\sum_m \left[ \frac{E}{\bar{\sigma}} \delta_{fm} - H_c(f-m) - V_{fm} \right] G_{m\alpha}(E) = \delta_{f\alpha} \quad (22a)$$

or in matrix form ( $\Omega = E/\bar{\sigma}$ ;  $\hat{I}$  is the unit matrix)

$$(\Omega \hat{I} - \hat{H}_c - \hat{V}) \hat{G}(\Omega) = \hat{I}. \quad (22b)$$

The zeroth approximation corresponds to the translation-invariant matrix  $\hat{H}_c$  and is described by the Green's function

$$G^c(\Omega) = (\Omega \hat{I} - \hat{H}_c)^{-1},$$

which in the site representation has the form

$$G_{m-n}^c(\Omega) = \frac{1}{N} \sum_k \frac{e^{i\mathbf{k}(m-n)}}{\Omega - E_c(\mathbf{k}, \Omega)}, \quad E_c(\mathbf{k}, \Omega) = z A_c(\Omega) \varepsilon_{\mathbf{k}}. \quad (23)$$

The solution of (22b) can be expressed in terms of a scattering matrix  $\hat{T}$ :

$$\hat{G} = \hat{G}^c + \hat{G}^c \hat{T} \hat{G}^c; \quad (24)$$

$\hat{T}$  is determined by the equation  $\hat{T} = \hat{V} + \hat{V} \hat{G}^c \hat{T}$ .

On carrying out a configurational average of (24), we get

$$\langle \hat{G} \rangle = \hat{G}^c + \hat{G}^c \langle \hat{T} \rangle \hat{G}^c;$$

$\langle \hat{G} \rangle$  describes the properties of the effective crystal that reproduces, on the average, the amorphous material. If  $\langle \hat{T} \rangle = 0$ , the amorphous material is modeled by an ideal crystal described by the Green's function

$\hat{G}^c$ . Since the values of the matrix elements of the  $\hat{T}$  matrix depend on  $G^c$ ,  $\langle \hat{T} \rangle = 0$  is the equation that determines the coherent exchange parameter  $A_c = A_c(\Omega)$ . In its physical meaning,  $A_c(\Omega)$  is the self-consistent exchange parameter that insures, on the average, no scattering of spin waves by the fluctuations.

In the approximation of statistical independence of the fluctuations of exchange bonds,

$$\hat{T} \approx \sum_{\alpha} \hat{t}_{\alpha},$$

where  $\hat{t}_{\alpha}$  is a partial scattering matrix; and the requirement  $\langle \hat{T} \rangle = 0$  is satisfied when  $\langle \hat{t}_{\alpha} \rangle = 0$  for all pairs  $\alpha = (f, m)$  of NN. The matrix  $\hat{t}_{\alpha}$  can be calculated exactly, and the equation for the coherent parameter  $A_c$  has the form<sup>[3]</sup>

$$\rho(A) \frac{A - A_c}{1 - 2(A - A_c)[G_0^c(\Omega) - G_1^c(\Omega)]} dA = 0, \quad (25)$$

where  $\rho(A)$  is the isotropic distribution function of the fluctuations and where

$$G_i^c(\Omega) = z^{-1} \sum_n G_n^c(\Omega).$$

We shall consider amorphization of a FM with  $J > 0$ ,  $K > 0$  ( $\lambda = K/J > 0$ ) and of an AF with  $J > 0$ ,  $K < 0$  ( $\lambda < 0$ ). Both cases can, from a formal point of view, be considered in the same manner, because under certain conditions the amorphous material will be ferromagnetic. Let  $\nu_0$  be the fraction of  $K$  bonds (along the  $z$  axis) in the original ideal crystal. Since the short-range order is on the average retained in the amorphous magnet, it is reasonable to suppose that  $\nu \sim \nu_0$ ,  $\lambda \sim \lambda_{am}$ . Then in accordance with Sec. 3,  $\bar{\sigma}(0) = 1$  for  $\lambda > 0$ , and  $\bar{\sigma}(0) = F(|\lambda|, \nu)$  for  $\lambda < 0$ ; that is, on amorphization of the AF it is necessary to indicate the range of the parameters  $\lambda$  and  $\nu$  that insures the existence of ferromagnetism. This existence region, within the framework of the CPA, will be determined from the condition  $A_c(0) > 0$ .

In accordance with the postulates of Sec. 3, the distribution function  $\rho(A)$  must reflect an isotropic distribution of  $J$  and  $K$  bonds in the AM, and also retention of short-range order. If we take into account only the effect of mixing of  $J$  and  $K$  bonds, then

$$\rho(A) = \nu \delta(A - K) + (1 - \nu) \delta(A - J), \quad \int \rho(A) dA = 1. \quad (26)$$

By use of (25) and (26), we get the equation

$$2J[G_0^c(\Omega) - G_1^c(\Omega)] = \frac{1 - x - (1 - \lambda)\nu}{(1 - x)(\lambda - x)}, \quad x = \frac{A_c}{J}. \quad (27)$$

Since

$$G_0^c(\Omega) - G_1^c(\Omega) = \frac{1}{N} \sum_{\mathbf{k}} \frac{1 - \gamma_{\mathbf{k}}}{\Omega - zA_{\mathbf{k}}\varepsilon_{\mathbf{k}}} = \frac{1}{zJ} \frac{1}{N} \sum_{\mathbf{k}} \frac{\varepsilon_{\mathbf{k}}}{\omega - x\varepsilon_{\mathbf{k}}},$$

where  $\omega = \Omega/zJ$ , (27) takes the form

$$\frac{1}{N} \sum_{\mathbf{k}} \frac{\varepsilon_{\mathbf{k}}}{\omega - x\varepsilon_{\mathbf{k}}} = \Psi(x; \lambda, \nu), \quad \Psi = \frac{z}{2} \frac{1 - x - (1 - \lambda)\nu}{(1 - x)(\lambda - x)}. \quad (28)$$

The left member of (28) must be interpreted as a

principal value. Then the dimensionless coherent parameter  $x = A_c/J$  is a function of the dimensionless frequency  $\omega = \Omega/zJ$ ; that is,  $x = x(\omega)$ .

From (28) one can find the value of  $x(0)$  that are solutions of the equation

$$-1/x(0) = \Psi(x(0); \lambda, \nu).$$

After elementary transformations, it reduces to a quadratic equation with the solutions

$$x_{\pm}(0) = \frac{z}{2(z-2)} \left[ P \pm \left( P^2 + 8\lambda \frac{z-2}{z^2} \right)^{1/2} \right], \quad (29)$$

$$P = \left( 1 - \frac{2}{z} - \nu \right) - \lambda \left( \frac{2}{z} - \nu \right).$$

When  $\lambda > 0$ , there is always a real positive solution  $x_+(0) > 0$ ; that is, the ground state of the amorphous magnet is ferromagnetic if the fluctuating exchange parameters are positive ( $J > 0$ ,  $K > 0$ ).

When  $\lambda = 0$  (breaking of the  $K$  bonds),  $x_+(0) > 0$  only when  $\nu < 1 - 2/z$ , whereas for  $\nu \geq 1 - 2/z$  the solution is  $x_+(0) = 0$ ; this indicates the disappearance of ferromagnetism. The value  $\nu_c = 2/z$  corresponds precisely to the concentration that insures "percolation" of the ferromagnetic bonds.

When  $\lambda < 0$ , the condition for existence of real positive solutions of (29) has the form

$$P = \left( 1 - \frac{2}{z} - \nu \right) + |\lambda| \left( \frac{2}{z} - \nu \right) > 0, \quad P^2 - 4|\lambda| \frac{2}{z} \left( 1 - \frac{2}{z} \right) \geq 0. \quad (30)$$

It follows from (30) that there exist critical values  $|\lambda_c|$ , functions of  $\nu$ , which determine the regions of existence of ferromagnetism. These regions are shown in Fig. 2. Since the CPA is hardly valid when  $\nu \ll 1$  (i.e., in the "impurity" range), the corresponding critical value  $|\lambda_c| = (1 - 2/z)(2/z)^{-1}$  at  $\nu = 0$  will not be discussed here. These results obtained in the CPA agree completely with the results of Sec. 3.

In our case of amorphization of a plane AF with the CPA and with  $z = 6$ , the fraction of  $K$  bonds is  $\nu = 2/z = 1/3$ . And it follows from (30) that the approximation of the amorphous material by an effective ferromagnet is possible when  $|\lambda| \leq 1/8$ . Then the solutions of (29) take the form

$$x_{\pm}(0) = 1/4 [1 \pm (1 - 8|\lambda|)^{1/2}]. \quad (31)$$

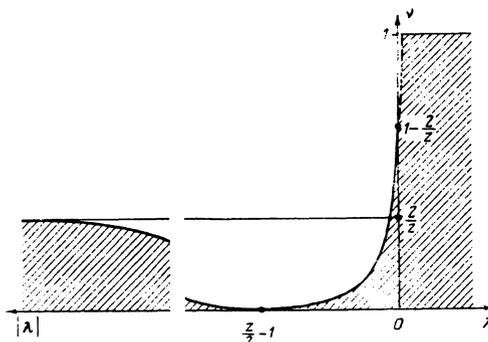


FIG. 2. Region of existence of ferromagnetism (shaded) on amorphization of an ideal magnet with an anisotropic distribution of exchange bonds.

We turn now to analysis of the real solutions  $x(\omega) > 0$  of equation (28) when  $\omega \neq 0$ . The equation can be expressed in the form

$$\frac{\omega}{x} G_0\left(\frac{\omega}{x}\right) = 1 + x\Psi(x), \quad (32)$$

where  $G_0(u)$  is the Green's function of the ideal isotropic FM:

$$G_0(u) = \frac{1}{N} \sum_{\mathbf{k}} \frac{1}{u - \varepsilon_{\mathbf{k}}} = \int \frac{g_0(\varepsilon) d\varepsilon}{u - \varepsilon};$$

$g_0(\varepsilon)$  is the density of magnon states corresponding to the dispersion law  $\varepsilon_{\mathbf{k}} = 1 - \gamma_{\mathbf{k}}$ . The density of states  $g_0(\varepsilon)$  is nonzero on the interval  $[0, 2]$  and is conveniently approximated by the half-ellipse

$$g_0(\varepsilon) = \frac{2}{\pi} [\varepsilon(2-\varepsilon)]^{1/2}, \quad \int_0^2 g_0(\varepsilon) d\varepsilon = 1. \quad (33)$$

By use of (33), the principal value ( $\mathcal{P}$ ) of Green's function on the interval  $0 \leq u \leq 2$  is

$$\mathcal{P}G_0(u) = 2(u-1)$$

and Eq. (32) takes the form

$$2\frac{\omega}{x}\left(\frac{\omega}{x} - 1\right) = 1 + x\Psi(x), \quad 0 \leq \frac{\omega}{x} \leq 2. \quad (34)$$

After elementary transformations, it reduces to a fourth-degree equation ( $z = 6, \nu = \frac{1}{3}$ )

$$x^4 - (\omega + 1/2)x^3 + [\omega^2 + (1+\lambda)\omega - 1/2\lambda]x^2 - \omega[(1+\lambda)\omega + \lambda]x + \lambda\omega^2 = 0. \quad (35)$$

The results of a computer solution of this equation are shown graphically in Fig. 3.

Analysis of (34) shows that when  $\lambda > 0$  and  $|\lambda| < \lambda_{cr} \approx 1/34$ , the solutions are unique. When  $|\lambda| > \lambda_{cr}$ , in the range  $0 \leq \omega \leq \omega_s$ , three real solutions appear. The physically admissible choice is the unique solution with

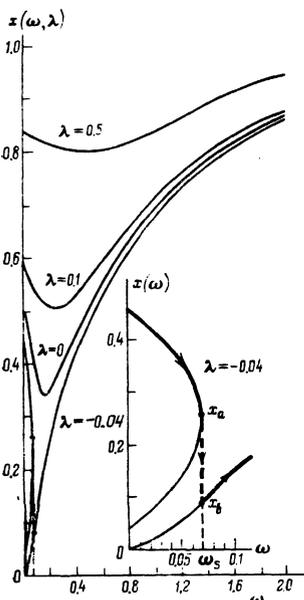


FIG. 3. Variation of the dimensionless coherent exchange parameter  $x = A_c/J$  with  $\omega = E/[\bar{\sigma}(0) \cdot 6J]$ , when  $\nu = 1/3$ , for several values of  $\lambda$ .

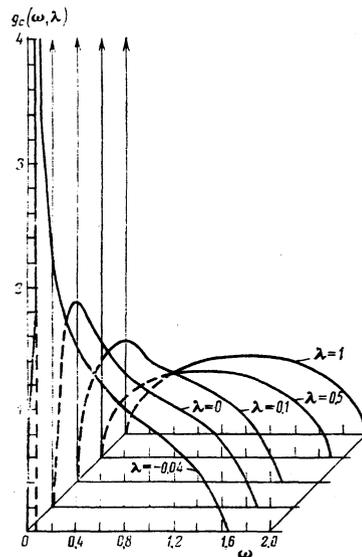


FIG. 4. Density of spin-wave states  $g_c(\omega)$  of an amorphous isotropic ferromagnet, in the CPA, for several values of  $\lambda$  ( $\nu = 1/3$ ).

a break at the point  $\omega_s$ , as is shown in Fig. 3 for the solutions with  $|\lambda| = 0.04$  ( $\omega_s \approx 0.07$ ). When  $|\lambda| \rightarrow \frac{1}{8}$ , we have  $\omega_s \rightarrow 0$ ; and at  $|\lambda| = \frac{1}{8}$  there remain two solutions for the coherent potential: the isolated point  $x(0) = \frac{1}{4}$  and the lower branch of the solution  $x_-(\omega)$ , for which  $x_-(0) = 0$ .

## 5. DENSITY OF SPIN-WAVE STATES OF AN AMORPHOUS FERROMAGNET, AND CURIE TEMPERATURE

Knowing the frequency dependence  $x = x(\omega)$  of the coherent potential, we can find the spin-wave spectrum and the density of states in an amorphous FM. Since the Green's function of an amorphous FM in the CPA has the form  $G_{\mathbf{k}}^c(\omega) = [\omega - x(\omega)\varepsilon_{\mathbf{k}}]^{-1}$ , the spin-wave spectrum is determined by the equation

$$\omega - x(\omega)\varepsilon_{\mathbf{k}} = 0. \quad (36)$$

In order to calculate the thermodynamic characteristics of an amorphous magnet, it is convenient to introduce the density of states  $g_c(\omega)$ <sup>[13]</sup>:

$$g_c(\omega) = -\frac{1}{\pi} \text{Im} G_0^c(\omega + i0) = \int g_0(\varepsilon) \delta(\omega - x(\omega)\varepsilon) d\varepsilon = \frac{1}{x(\omega)} g_0\left(\frac{\omega}{x(\omega)}\right). \quad (37)$$

Figure 4 shows the densities of states calculated with formula (37) for several values of  $\lambda$ . When  $|\lambda| > \lambda_{cr}$ , the presence of a finite discontinuity of the coherent parameter at the point  $\omega = \omega_s$  leads to the appearance of a finite discontinuity of the density of states. The limiting point  $\omega_m$  of the spectrum is determined by the relation  $\omega_m/x(\omega_m) = 2$ . The discontinuity of the coherent parameter leads to the occurrence of a finite flat section in the dispersion law  $\varepsilon_{\mathbf{k}}$  at frequency  $\omega_s$ . These singularities will presumably be smoothed out if a "smeared out"  $\delta$  function is used in the distribution (26).

With use of the relation  $\bar{\sigma} = \bar{\sigma}(0) - 2\langle b^+b \rangle_T$ , the equa-

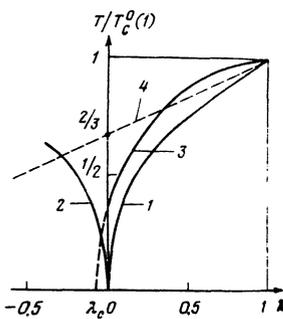


FIG. 5. Ratio of the magnetic ordering temperature to the Curie temperature  $T_C^0(\lambda=1)$  of an ideal isotropic FM, as a function of  $\lambda$ , when  $\nu=1/3$ . 1,  $T_C^0(\lambda)/T_C^0(1)$ , where  $T_C^0(\lambda)$  is the Curie temperature of an ideal FM,  $\lambda \geq 0$ . 2,  $T_N^0(\lambda)/T_C^0(1)$ , where  $T_N^0(\lambda)$  is the Néel temperature of an ideal AF,  $\lambda \leq 0$ . 3,  $T_C^{\text{am}}(\lambda)/T_C^0(1)$ , where  $T_C^{\text{am}}(\lambda)$  is the Curie temperature of an amorphous FM in the CPA (solid line, amorphization of an ideal FM; dotted, amorphization of an ideal AF). 4,  $\bar{T}_C(\lambda)/T_C^0(1)$ , where  $\bar{T}_C(\lambda)$  is the Curie temperature of an amorphous FM in the approximation of a mean exchange parameter  $\bar{A} = \frac{2}{3}J(1 + \frac{1}{2}\lambda)$ .

tion that determines the temperature dependence  $\bar{\sigma} = \bar{\sigma}(T)$  of the magnetization, in the variables  $\omega = E/\bar{\sigma}Jz$  and  $\tau = T/Jz$ , takes the form

$$\frac{\bar{\sigma}(0)}{\bar{\sigma}} = 1 + 2 \int \frac{d\omega}{\exp(\bar{\sigma}\omega/\tau) - 1} g_c(\omega). \quad (38)$$

At low temperatures a deviation from the  $\frac{3}{2}$ -power law may occur if the behavior of  $g_c(\omega)$  at small  $\omega$  differs significantly from a square root. The presence of a discontinuity of the coherent potential at  $\omega = \omega_s$  leads to a finite discontinuity of  $g_c$  at  $\omega = \omega_s$  and, as a consequence, to a singularity in the temperature behavior of the magnetization at  $\tau \sim \omega_s$ .

When  $\tau \rightarrow \tau_c = T_c/Jz$ , where  $T_c$  is the Curie temperature,  $\bar{\sigma} \rightarrow 0$ , and the Bose distribution function in (38) can be expanded as a series in the quantity  $\bar{\sigma}\omega/\tau_c \ll 1$ . Neglecting unity in comparison with  $1/\bar{\sigma}$ , we get an estimate of the Curie temperature:

$$\frac{\bar{\sigma}(0)}{2\tau_c^{\text{am}}(\lambda)} = \int_0^{\omega_m} \frac{d\omega}{\omega} g_c(\omega) = I(\lambda). \quad (39)$$

In the relation (39),  $g_c(\omega)$ ,  $\bar{\sigma}(0)$ , and  $\omega_m$  are functions of the number of antiferromagnetic bonds  $\nu$ , the coordination number  $z$ , and  $\lambda = K/J$ .

In amorphization of a FM ( $\lambda > 0$ ,  $\bar{\sigma}(0) = 1$ ), one may consider the whole range of variation of the parameter  $\lambda$  ( $0 \leq \lambda \leq 1$ ). Figure 5 shows the variation with  $\lambda$  of the Curie temperatures (in units  $J$ ) of an ideal FM (according to Lines<sup>[7]</sup>) and of an amorphous isotropic FM. It follows from the figure that amorphization of a low-dimensional FM ( $\lambda \ll 1$ ) leads to a significant increase of the Curie temperature:  $T_C^{\text{am}} \gg T_C^0$ .

In amorphization of an AF ( $\lambda < 0$ ), the range of existence of an amorphous FM in the case considered ( $\nu = \frac{1}{3}$ ,  $z = 6$ ) is bounded by the values  $0 \leq |\lambda| \leq \frac{1}{6}$ . The value of the integral (39) is 4.3 for  $\lambda = 0$  and 7.8 for  $|\lambda| = 0.04$ . But because the value of  $\bar{\sigma}(0)$  is unknown, it is impossible to give a numerical estimate of the Curie temperature.

It may be assumed that  $\bar{\sigma}(0) \leq 1$  when  $|\lambda| \rightarrow 0$  for  $\nu = \frac{1}{3}$ , since this concentration of broken  $K$  bonds is considerably smaller than the percolation concentration  $\nu_{\text{cr}}(\lambda = 0) = \frac{2}{3}$ . Then  $\tau_c(\lambda = 0) \leq I(0)/2$ . On the other hand, we may expect a very rapid decrease of  $\bar{\sigma}(0)$  even at small  $|\lambda| > 0$  (ferromagnetism disappears when  $|\lambda| = \frac{1}{6}$ ). Therefore the function  $\tau_c(|\lambda|)$  will be principally determined by the behavior of  $\bar{\sigma}(0, |\lambda|)$ . Figure 5 shows the assumed  $\tau_c(|\lambda|)$  relation, from which it is evident that amorphization of a quasi-low-dimensional AF with  $|\lambda| \ll 1$  can lead (under certain conditions) to formation of an amorphous FM with  $T_C^{\text{am}} > T_N^0$ . If the quasi-low-dimensionality is not so clearly expressed, then amorphization leads to a change of the magnetic order (AF  $\rightarrow$  FM), but  $T_C^{\text{am}} < T_N^0$ .

## CONCLUSION

The analysis carried out above determines the behavior of magnets characterized by the presence of two different exchange parameters  $J$  and  $K$  when the magnets are amorphized. We have shown that it is necessary to distinguish two cases of amorphization: a)  $J > 0$ ,  $K > 0$ ; b)  $J > 0$ ,  $K < 0$ . In the first case, the ground state of the original crystal is FM. Amorphization leads to averaging of the exchange interactions; and when  $\lambda = K/J \ll 1$  (a quasi-two-dimensional FM), the resulting amorphous FM may have a Curie temperature appreciably exceeding the Curie temperature of the crystal. The situation is considerably more complicated in the case of amorphization of an AF ( $\lambda < 0$ ). In this case it is found that there exists only a bounded region in the  $(\nu, |\lambda|)$  plane within which the amorphous magnet can be described as an effective FM with  $\bar{\sigma}(0) \neq 0$ . Because within the framework of the CPA method it is not possible to calculate  $\bar{\sigma}(0)$  for given  $\nu$  and  $|\lambda|$ , there is, correspondingly, no possibility of constructing a closed theory, for example in the sense of a determination of  $T_c$ . Qualitative analysis, however, shows that in this case  $T_c$  decreases rapidly with increase of  $|\lambda|$ , and therefore amorphization of a quasi-low-dimensional AF with  $J > 0$  and  $K < 0$  can lead to an increase of  $T_c$  only at very small values of  $|\lambda|$ . The presence of negative exchange also expresses itself very strongly in the character of the density of states. Thus under certain conditions a characteristic is the appearance of a sharp peak of  $g_c(\omega)$ , of the localized-state type. This must lead to anomalies of the spectrum of spin-wave states and of the thermodynamic characteristics. The approximate nature of the theory makes it extremely desirable to perform experiments to reveal these anomalies.

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## Long-wave secondary radiation in polar semiconductors

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A new type of secondary radiation is predicted upon excitation of a semiconductor with light of frequency  $\omega_i$  in the region of intrinsic absorption. The radiation is the result of resonant Raman scattering of light, wherein an electron-hole pair and a certain number of longitudinal optical phonons are present in the final state of the crystal. Since the kinetic energies of the electron and hole in the final state can be different, the secondary radiation spans the band  $0 \leq \omega_s \leq \omega_{s,\max}$  in the long-wave band. The scattering cross section is calculated with allowance for the dispersion of the LO phonons. It is shown that the dependence of the cross section of the process in question on the frequencies  $\omega_i$  and  $\omega_s$  has a steplike character. Each higher step corresponds to turning on a process in which the number of emitted LO phonons increases by unity. It is shown that the scattering cross section is a quantity of zero order in the Fröhlich coupling constants of the electrons or holes with the LO phonons. The reason is that the secondary radiation is the result of a sequence of real transitions, each of which is accompanied by the emission of one LO phonon.

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### 1. INTRODUCTION

With the appearance of high-power laser sources of light, and with increasing receiver sensitivity, the capabilities of modern experiments on registration of secondary radiation of condensed media have greatly increased. This progress gives grounds for hoping that study of secondary radiation will become a reliable method of measuring a number of parameters of solids. The experimental research in this field has stimulated theoretical investigations (see, e.g., Refs. 1 and 2).

In this paper we predict a new type of secondary radiation of polar semiconductors. The radiation is the result of the following set of real transitions: primary radiation with frequency  $\omega_i$  produces an electron-hole pair (EHP), the electron (or hole) emits in succession  $k$  longitudinal optical (LO) phonons (we neglect interactions with acoustic phonons), and finally the electron (or hole) emits a quantum  $\hbar\omega_s$  of secondary light, accompanied by still one more optical phonon. No annihilation of the electron and hole takes place; the electron and hole, having lost an energy  $\hbar\omega_s + (k+1)\hbar\omega_{LO}$ , can retain part of their kinetic energy. Indirect emission of the quantum  $\hbar\omega_s$  accompanied by the LO phonon, by the electron, is a process inverse to indirect absorption of light by free carriers that interact with LO phonons.<sup>3</sup>

It follows from the energy conservation law that when an electron emits  $k+1$  LO phonons the secondary radiation spans the frequency band

$$0 < \omega_s < (1 + m_e/m_h)^{-1}(\omega_i - E_g/\hbar) - (k+1)\omega_{LO}, \quad (1.1)$$

where  $m_e(m_h)$  is the effective mass of the electron (hole), and  $E_g$  is the width of the forbidden band. In fact, if the kinetic energy of the electron after emission of secondary radiation is equal to zero, then

$$\omega_s = \omega_i - E_g/\hbar - (k+1)\omega_{LO} - E_{oh}/\hbar, \quad (1.2)$$

where  $E_{oh}$  is the kinetic energy of the hole produced by light  $\omega_i$ . Since the equality

$$E_{oh} = (\hbar\omega_i - E_g)(1 + m_h/m_e)^{-1} \quad (1.3)$$

is satisfied in direct production of EHP, it follows that by substituting (1.3) in (1.2) we obtain the upper bound from (1.1). The lower bound  $\omega_s = 0$  corresponds to the maximum residual kinetic energy of the electron. If secondary radiation with participation of  $k+1$  LO phonons is to take place at all, it is necessary that the right-hand side of (1.2) be larger than zero, i.e., that the following condition hold:

$$\omega_i > E_g/\hbar + (1 + m_e/m_h)(k+1)\omega_{LO}. \quad (1.4)$$

If a hole rather than an electron participates in the radiation, then the mass  $m_e$  in (1.1)-(1.4) must be replaced by  $m_h$ , and vice versa. It follows from (1.4) that the frequency  $\omega_i$  should lie in the region of the intrinsic absorption of the semiconductor.

An examination of the electron energy distribution