

Influence of intermediate order on the binary correlation function of a random field

V. D. Iskra

Ukrainian Academy of Agriculture

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The binary correlation function of the random field of a heteropolar semiconductor is calculated by using very simple model representations that take the presence of intermediate order into account. It is shown that the character of the random self-field, which is a Coulomb field in the absence of intermediate order, changes in the presence of the latter, and can be regarded as smooth in a wide range of the intermediate-order parameter $y = r_c/r_0$ (here r_0 is the screening radius and r_c is the correlation radius, i.e., the distance in which the binary correlation function of the atoms becomes effectively equal to zero).

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§1. INTRODUCTION

Many properties, particularly electric and optical, of amorphous semiconductors can be interpreted on the basis of the concept of a random force field acting on the carriers.

In a number of problems it is important to know the behavior of the correlation functions that describe the random field, and primarily the binary correlation function

$$\Psi(\mathbf{r}, \mathbf{r}') = \langle \delta V(\mathbf{r}) \delta V(\mathbf{r}') \rangle.$$

Here

$$\delta V(\mathbf{r}) = V(\mathbf{r}) - \langle V(\mathbf{r}) \rangle,$$

$V(\mathbf{r})$ is the potential energy of the electron in the random field, and the symbol $\langle \rangle$ means averaging over the random field. We confine ourselves to macroscopic, homogeneous, and isotropic systems, for which it is known that $\Psi(\mathbf{r}, \mathbf{r}') = \Psi(|\mathbf{r} - \mathbf{r}'|)$. The objects of our investigations are heteropolar materials (such as, e.g., a -Si:H).

As shown in Ref. 1, a binary correlation function can be expressed in terms of atomic pseudopotentials that describe the structure of the compound:

$$\Psi(\mathbf{r}) = \frac{1}{(2\pi)^3} \sum_{a,a'} (\Omega_a \Omega_{a'})^{-1/2} \int d\mathbf{q} S_{aa'}(\mathbf{q}) v_a(\mathbf{q}) v_{a'}(\mathbf{q}) e^{-i\mathbf{q}\mathbf{r}}. \quad (1)$$

The indices a and a' number here the types of atoms (or ions), Ω_a is the atomic volume, and $v_a(\mathbf{q})$ is the screening pseudopotential of the ion in the empty-core approximation,

$$v_a(\mathbf{q}) = 4\pi Z_a \cos q R_a / \epsilon \Omega_a (q^2 + r_0^{-2}), \quad (2)$$

where Z_a is the charge of the ion of the a -th type in units of the modulus of the electron charge e , r_0 is the screening radius, ϵ is the inertialess dielectric constant of the substance, R_a is the radius of the empty core (it will be shown later that the region $r < R_a$ does not play a substantial role, and we therefore assume from the outset that $R_a = 0$). The quantity $S_{aa'}(\mathbf{q})$ in the right-hand side of (1) is the well known interference function, which describes the intensity of the elastic scattering of x rays, electrons, and neutrons with change of the wave vector by \mathbf{q} :

$$S_{aa'}(\mathbf{q}) = \delta_{aa'} + (n_a n_{a'})^{-1/2} \int d\mathbf{r} [g_{aa'}(\mathbf{r}) - 1] e^{i\mathbf{q}\mathbf{r}}; \quad (3)$$

$n_a = \Omega_a^{-1}$ is the average concentration of the atoms of the given type, $\delta_{aa'}$ is the Kronecker symbol, and $g_{aa'}(\mathbf{r})$ is the binary correlation function of the atoms.

In amorphous substances we have $g_{aa'}(\mathbf{r}) - 1$ as $r \rightarrow \infty$ and it is possible to introduce the concept of the correlation radius $r^{aa'}$ defined as the distance over which the quantity $[g_{aa'}(\mathbf{r}) - 1]$ effectively vanishes. We shall use hereafter a single correlation radius r_c , chosen to be largest of the lengths $r^{aa'}$. The ratio $y = r_c/r$ will play the role of the dimensionless parameter of the theory (the intermediate-order parameter). In Ref. 1, $\Psi(\mathbf{r})$ was calculated under the assumption $y \ll 1$. In this paper this restriction is lifted and the correlation function is calculated for the values of the parameter y in the interval $0 < y < \infty$. We calculate $\Psi(\mathbf{r})$ by invoking very simple model representations that reflect the main features of systems with intermediate order.

The presence of intermediate order in certain amorphous materials (e.g., in amorphous Se and Al_2O_3) has been established in experiment.² The recent serious interest in it is connected, in particular, with investigations of the atomic-matrix oscillation frequencies.³ In the present paper, the intermediate order is considered in general form with an aim at determining its influence on the character of the "intrinsic" random field in amorphous semiconductors.

§2. THE CORRELATION FUNCTION $\Psi(r)$ AT SMALL CORRELATION RADII

We consider the case when the correlation radius r_c is comparable with $\Omega_a^{1/3}$. This situation is realized in a number of compounds—the binary correlation function has several clearly pronounced peaks that attenuate over a length of the order of several interatomic distances.⁴ In this case the function $g_{aa'}(\mathbf{r})$ can be approximately represented in the form

$$g_{aa'}(\mathbf{r}) = \begin{cases} g_{aa'}^0(r), & r \leq r_c, \\ 1, & r > r_c, \end{cases} \quad (4)$$

where $g_{aa'}^0(r)$ is the exact value of the function $g_{aa'}(\mathbf{r})$ in the interval $0 \leq r \leq r_c$.

With (4) taken into account, the interference function (3) takes the form

$$S_{aa'}(q) = \delta_{aa'} + \frac{4\pi}{q} (n_a n_{a'})^{1/2} \int_0^{r_0} \sin qr [g_{aa'}^*(r) - 1] r dr.$$

We note that by virtue of (2) the main contribution to the integral in the right-hand side of (1) is made by values of q that satisfy the condition

$$q \leq r_0^{-1}. \quad (5)$$

Since the relation $r_0 \gg \Omega_0^{1/3}$, where Ω_0 is the largest of the Ω_a , is satisfied as a rule in the materials considered by us, the inequality $y = r_c/r_0 \ll 1$ is certainly satisfied. In this case we have $qr_c \ll 1$, for the values of q that satisfy the condition (5), hence

$$S_{aa'}(q) \approx \delta_{aa'} + 4\pi (n_a n_{a'})^{1/2} \int_0^{r_0} [g_{aa'}^*(r) - 1] r^2 dr = S_{aa'}(0). \quad (6)$$

Using this expression and changing in (2) to the usual units for the correlation function (1), we obtain

$$\Psi(r) \approx \frac{2\pi r_0 e^4}{\varepsilon^2} \sum_{a, a'} \frac{Z_a Z_{a'}}{(\Omega_a \Omega_{a'})^{1/2}} S_{aa'}(0) F(R), \quad (7)$$

$$F(R) = \frac{4}{\pi R} \int_0^{\infty} \frac{x \sin Rx}{(1+x^2)^2} dx = e^{-R}, \quad R = \frac{r}{r_0}. \quad (8)$$

Expression (7) is formally the correlation function of a random Coulomb field produced by pointlike ions randomly distributed in space and having a concentration n_i given by

$$n_i = \sum_{a, a'} \frac{Z_a Z_{a'}}{(\Omega_a \Omega_{a'})^{1/2}} S_{aa'}(0). \quad (9)$$

We note that by going over in (7) and (9) to extremely small correlation radii, and recognizing that according to (6)

$$\lim_{r_c \rightarrow 0} S_{aa'}(0) = \delta_{aa'},$$

we arrive at the result of Ref. 1.

§3. SIMPLEST MODEL THAT ACCOUNTS FOR THE PRESENCE OF INTERMEDIATE ORDER

In the case of an ideal crystal with a "rigid" lattice, the binary correlation function $g_{aa'}(r)$ in (3) is of the form

$$g_{aa'}(r) = \Omega_{a'} \sum_n' \delta(r - a_n), \quad (10)$$

where a_n are the lattice vectors. The summation in (10) is over all sites of type a' , and the prime on the summation sign denotes omission of the term corresponding to the site located at the origin. In this case relations (1) and (3) yield, as expected

$$S_{aa'}(q) = 0, \quad \Psi(r) = 0.$$

The (probably) simplest function $g_{aa'}(r)$ that takes the presence of intermediate order into account can be written in the form

$$g_{aa'}(r) = b_{aa'} \sum_{|a_n| < r_c} \delta(r - a_n) + \theta(r - r_c), \quad (11)$$

where $b_{aa'}$ is a constant. The interference function (3)

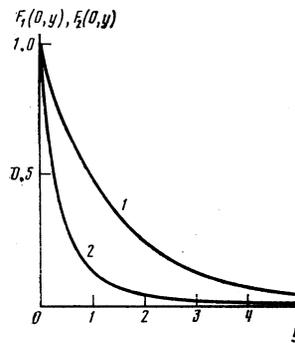


FIG. 1. Plots of the functions $F_1(R, y)$ (curve 1) and $F_2(R, y)$ (curve 2), defined by Eqs. (14) and (20), respectively, vs the intermediate-order parameter y at $R = 0$.

corresponding to this expression is

$$S_{aa'}(q) = \delta_{aa'} + (n_a n_{a'})^{1/2} \left\{ b_{aa'} \left[\sum_{|a_n| < r_c} e^{iqa_n} - \delta_{aa'} \right] - \int_{r < r_c} e^{iqr} dr \right\}. \quad (12)$$

We consider the case $r_c \gg \Omega_0^{1/3}$. Changing in (12) from summation over the lattice sites to integration with respect to r (this is valid for q values satisfying the condition $q\Omega_0^{1/3} \ll 1$), and expressing the constant $b_{aa'}$ with the aid of (12), we obtain

$$S_{aa'}(q) \approx 3S_{aa'}(0) \left[\frac{\sin qr_c}{q^2 r_c^3} - \frac{\cos qr_c}{q^2 r_c^2} \right].$$

For the correlation function (1) we obtain accordingly

$$\Psi(r) \approx \frac{2\pi r_0 e^4}{\varepsilon^2} \sum_{a, a'} \frac{Z_a Z_{a'}}{(\Omega_a \Omega_{a'})^{1/2}} S_{aa'}(0) F_1(R, y). \quad (13)$$

Here

$$F_1(R, y) = \frac{12}{\pi R} [y^{-2} I_1(R, y) - y^{-2} I_2(R, y)], \quad (14)$$

$$I_1(R, y) = \int_0^{\infty} \frac{\sin Rx \sin yx}{x^2 (1+x^2)^2} dx = \begin{cases} \frac{\pi}{4} \{2R - \exp(-y) [(y+3) \text{sh } R - R \text{ch } R]\}, & R \leq y, \\ \frac{\pi}{4} \{2y - \exp(-R) [(R+3) \text{sh } y - y \text{ch } y]\}, & R \geq y, \end{cases} \quad (15)$$

$$I_2(R, y) = \int_0^{\infty} \frac{\sin Rx \cos yx}{x (1+x^2)^2} dx = \begin{cases} \frac{\pi}{4} \exp(-y) [(y+2) \text{sh } R - R \text{ch } R], & R \leq y, \\ \frac{\pi}{4} \{2 - \exp(-R) [(R+2) \text{ch } y - y \text{sh } y]\}, & R \geq y. \end{cases} \quad (16)$$

It follows from (14)–(16) that at $y \ll 1$ the function $F_1(R, y)$ with $R \geq y$ practically coincides with the function $F(R)$ [see (8)], and as $y \rightarrow 0$ we again arrive at expression (7) for the correlation function $\Psi(r)$. As $y \rightarrow \infty$ (which corresponds to a transition to a crystal), $F_1(R, y)$ tends to zero at all R .

A plot of the function $F_1(r, y)$ at $R = 0$

$$F_1(0, y) = 3y^{-2} [2 - \exp(-y) (y^2 + 2y + 2)],$$

is shown in Fig. 1 (curve 1). Figures 2 and 3 show plots of the function $F_1(R, y)$ and plots of the relative

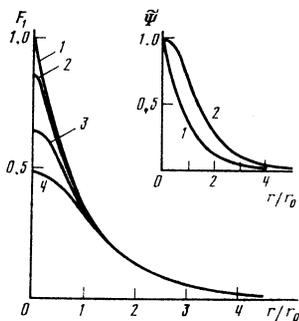


FIG. 2. Plots of the function F_1 , defined by (14), at $y = 0$ (curve 1), $y = 0.2$ (2), $y = 0.6$ (3) $y = 1$ (4) and of the relative correlation function $\tilde{\Psi} = F_1(R, y)/F_1(0, y)$ at $y = 0$ (1), $y = 1$ (2).

correlation function $\tilde{\Psi} = \Psi(r)/\Psi(0)$ at different values of the parameter y .

§4. MODIFICATION OF THE SIMPLE MODEL

The model (11) considered in §3, which leads to reasonable physical results for the correlation function $\Psi(r)$, contains nonetheless an unphysical discontinuity at $r = r_c$ [as a result, in particular, $S_{aa}(q)$ oscillates and takes on a number of negative values]. This discontinuity can be eliminated by replacing the factor $\theta(r_c - r)$ in the expression

$$g_{aa'}(r) - 1 = \left[b_{aa'} \sum_n \delta(r - a_n) - 1 \right] \theta(r_c - r) \quad (17)$$

by a continuous function that takes into account that the quantity $[g_{aa'}(r) - 1]$ tends to zero at large distances. We choose this function to be, for simplicity, the exponential $\exp(-r/r_c)$. In addition, we replace the δ -function under the summation sign in (17) by some arbitrary δ -like function, such as the normalized Gaussian

$$\pi^{-1/2} \sigma_n^{-1} \exp\left[-(r - a_n)^2 / \sigma_n^2\right],$$

where σ_n is a constant. Accordingly, the unity in the right-hand side of (17) should be replaced by a constant (which we designate by b) determined from the condition $g_{nn'}(0) = 0$. As a result, (17) is transformed into

$$g_{aa'}(r) - 1 = \left\{ b_{aa'} \sum_n \pi^{-1/2} \sigma_n^{-1} \exp\left[-\frac{(r - a_n)^2}{\sigma_n^2}\right] - b \right\} \exp\left(-\frac{r}{r_c}\right). \quad (18)$$

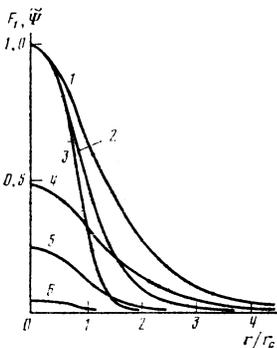


FIG. 3. Plots of the function F_1 (curves 4, 5, and 6) and of the relative correlation function $\tilde{\Psi}$ corresponding to F_1 (curves 1, 2, and 3) at different values of the intermediate-order parameter $y \geq 1$: $y = 1$ (curves 1, 4), $y = 2$ (2, 5) and $y = 3$ (3, 6).

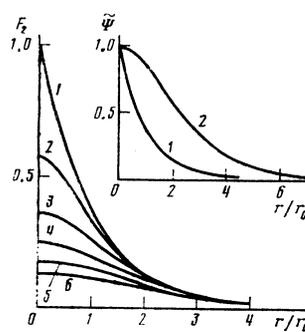


FIG. 4. Plots of the function F_2 defined by relation (20) at $y = 0$ (curve 1), $y = 0.2$ (2) $y = 0.4$ (3) $y = 0.6$ (4) $y = 0.8$ (5) $y = 1$ (6) and of the relative correlation function $\tilde{\Psi} = F_2(R, y)/F_2(0, y)$ at $y = 0$ (1), $y = 1$ (2).

The function $g_{nn'}(r)$ obtained in this manner satisfies at any finite r_c the requirement

$$\lim_{r \rightarrow \infty} g_{nn'}(r) = 1,$$

and goes over into (10) as $\sigma_n \rightarrow 0$ and $r_c \rightarrow \infty$ (in this case $b \rightarrow 1$ and $b_{nn'} \rightarrow \Omega_a$).

The quantity σ_n should generally speaking be regarded as increasing with increasing distance a_n from the given site to the origin, in accord with the fact that the peaks of the experimentally determined binary correlation function broaden continuously with increasing distance. For simplicity we put nevertheless

$$\sigma_n = \sigma = \text{const},$$

assuming at the same time that $\sigma \ll \min\{\Omega_a^{1/3}\}$. Repeating next the same sequence of operations as in the calculation of the interference function $S_{nn'}(q)$ in §3, we obtain under the same assumptions concerning the values of r_c and q

$$S_{aa'}(q) \approx \frac{S_{aa'}(0)}{(1 + q^2 r_c^2)^2}.$$

The correlation function (1) corresponding to this expression is of the form

$$\Psi(r) = \frac{2\pi r_0 e^i}{\varepsilon^2} \sum_{\alpha, \alpha'} \frac{Z_\alpha Z_{\alpha'}}{(\Omega_\alpha \Omega_{\alpha'})^{1/2}} S_{aa'}(0) F_2(R, y), \quad (19)$$

where

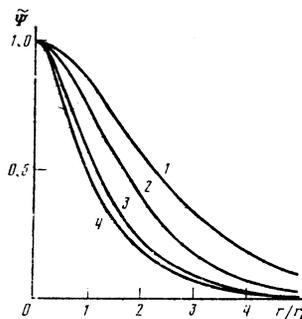


FIG. 5. Plots of the relative correlation function $\tilde{\Psi} = F_2(R, y)/F_2(0, y)$ at different values of the intermediate-order parameter $y \geq 1$: $y = 1$ (curve 1), $y = 2$ (2), $y = 5$ (3), $y = 8$ (4).

$$F_2(R, y) = \frac{4}{\pi R} \int_0^{\infty} \frac{x \sin Rx}{(1+x^2)^2(1+y^2x^2)^2} dx$$

$$= \begin{cases} \frac{1}{R(1-y^2)^2} \{ [(1-y^2)(R-1) + 1-5y^2] e^{-R} \\ + y[(1-y^2)(R-y) + 5y-y^3] e^{-R/y} \}, & y \neq 1, \\ \frac{1}{24} (R^2 + 3R + 3) e^{-R}, & y = 1. \end{cases} \quad (20)$$

It follows from (20) that at $y \ll 1$ the function $F_2(R, y)$ can be approximated by the function $F(R)$; we arrive thus again at the result obtained in §2. As $y \rightarrow \infty$ the function $F_2(R, y)$ tends to zero for any R . A plot of the function $F_2(0, y) = (1+y)^{-3}$ is shown in Fig. 1 (curve 2). Figures 4 and 5 show plots of the function $F_2(R, y)$ and of the relative correlation function $\tilde{\Psi} = \Psi(r)/\Psi(0)$ at different values of the parameter y .

§5. CONCLUSIONS

The results of §§2-4 lead to general conclusions concerning the influence that can be exerted by the presence of intermediate order on the binary correlation function $\Psi(r)$.

First, an increase of the intermediate-order parameter y leads to a decrease of the mean squared fluctuation of the potential energy $\Psi_1 = \Psi(0)$, which vanishes as $y \rightarrow \infty$, i.e., on going to the limit of long-range order (see Fig. 1). Of course, the character of the dependence of Ψ_1 on the intermediate-order parameter is somewhat more complicated than that shown in Fig. 1, since the effective density n_e also depends on y via the quantities $S_{aa}(0)$ that must be determined from experiment. This, nevertheless, does not change the conclusion that Ψ_1 decreases with increasing intermediate-order parameter y .

Second, with change of the intermediate-order parameter, a change takes place in the very character of the random field. Thus, at extremely low values of the intermediate-order parameter, $y \ll 1$, the random self-field in a heteropolar material can be approximately regarded as a Coulomb field¹ and described in terms of the theory of strongly doped semiconductors. Within the framework of the models considered in §§3 and 4, however, the random field is, strictly speaking, not Coulomb even at any nonzero value of the parameter y .

In the interpretation of a number of experiments⁵⁻¹⁰ it was found to be quite convenient to use the concept of a smooth random field, introduced in Ref. 11. It seems important therefore to check on the conditions under which the presence of intermediate order makes it possible to regard the random self-field in a heteropolar semiconductor as smooth.

It is known that a field is called smooth if the function $\Psi(r)$ has continuous derivatives with respect to the coordinates r at zero, and these derivatives decrease successively with increase in their order. Usually the condition of "smoothness" of a random field is of the form

$$\hbar^2 \Psi_2 / 4m \Psi_1^{3/2} \ll 1, \quad (21)$$

where m is the effective mass of the carrier (defined as

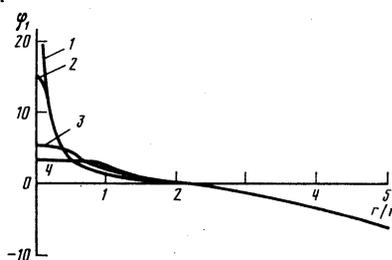


FIG. 6. Plots of the function φ_1 defined by relation (24) at different values of the intermediate-order parameter $y \leq 1$: $y = 0$ (curve 1), $y = 0.2$ (2); $y = 0.6$ (3); $y = 1$ (4).

in Ref. 1), and Ψ_2 is half the mean squared gradient of the potential energy.

$$\Psi_2 = 1/2 \langle [\nabla \delta V(\mathbf{r})]^2 \rangle.$$

For a Coulomb random field, the last quantity diverges and the "smoothness" condition (21) does not hold.

We shall also use a more general smoothness condition

$$\frac{\hbar^2 |\nabla_r \nabla_{r'} \Psi(|\mathbf{r}-\mathbf{r}'|)|}{8m [\Psi(|\mathbf{r}-\mathbf{r}'|)]^{3/2}} \ll 1, \quad (22)$$

which goes over into (21) as $r' \rightarrow r$.

Taking (9), (13), and (19) into account, the conditions (21) and (22) are transformed into

$$C |\varphi_i(0, y)| \ll 1, \quad (21a)$$

$$C |\varphi_i(R, y)| \ll 1, \quad (22a)$$

where

$$C = \frac{\hbar^2 e}{8(2\pi)^{3/2} m e^2 r_0^{3/2} n_e^{1/2}}, \quad (23)$$

$$\varphi_i = - \left[\frac{d^2 F_i}{dR^2} + \frac{2}{R} \frac{dF_i}{dR} \right] F_i^{-3/2}, \quad i=1, 2, \quad (24)$$

and $F_1 = F_1(R, y)$ and $F_2 = F_2(R, y)$ are the functions defined by Eqs. (14) and (20).

The functions φ_1 and φ_2 were calculated with a computer for different values of the parameter y . The differentiation of the functions F_1 and F_2 at $R \geq 0.01$ was carried out approximately—the differential of the argument was replaced by its finite increment $\Delta R = 10^{-4}$. The values of $\varphi_i(0, y)$ were calculated from the exact formulas

$$\varphi_1(0, y) = 3e^{-y} \{ 3y^{-3} [2 - e^{-y}(y^2 + 2y + 2)] \}^{-3/2} y^{-1}, \quad (25)$$

$$\varphi_2(0, y) = (1+y)^{3/2} y^{-1}, \quad (26)$$

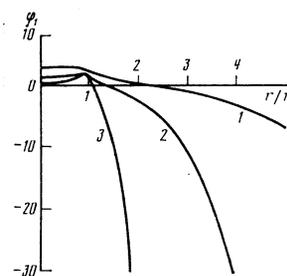


FIG. 7. Plots of the function φ_1 at different values of the intermediate-order parameter $y \geq 1$: $y = 1$ (curve 1), $y = 2$ (2), $y = 5$ (3).

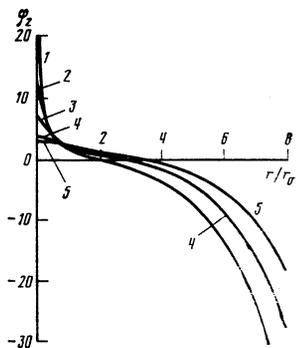


FIG. 8. Plots of the function φ_2 defined by relation (24) at different values of the parameter $y < 1$: $y = 0$ (curve 1), $y = 0.1$ (2), $y = 0.2$ (3), $y = 0.6$ (4) $y = 0.8$ (5).

obtained from (14) and (20). Some of the calculation results are shown in Figs. 6–9.

For an approximate estimate of the coefficient C we put in (23)

$$r_0 = 10^{-4} \text{ cm} \quad (27a)$$

(we note that for many heteropolar materials this quantity is undervalued; we thus obtain an overestimate). We next set the effective concentration n_t equal to 10^{21} cm^{-3} , using an estimate given in Ref. 1 and corresponding to $y \rightarrow 0$; the effective mass m is set equal to the free-electron mass. At $\epsilon = 10$ we then obtain

$$C \approx 0.6 \cdot 10^{-9}. \quad (27b)$$

It follows from (21a), (25), and (26) that within the limits of the validity of the estimates [27(a), (b)] at low values of the intermediate-order parameter, $y \ll 1$, which are compatible with the requirement $r_c = yr_0 \gg \Omega_0^{1/3}$, the smoothness condition [21(a)] is satisfied. At $y \gg 1$ the satisfaction of the inequality [21(a)] becomes less obvious [at least in the case (26)], for as $y \rightarrow \infty$ the effective concentration n_t decreases and consequently the coefficient C increases.

The smoothness condition [22(a)] is definitely violated at large distances. Calculation shows, however, that this violation takes place at distances such that the relative correlation function $\bar{\Psi} = \Psi(r)/\Psi(0)$ is negligibly small compared with unity. For the intermediate-order parameters considered by us in the interval $0.01 \leq y \leq 10$ the condition [22(a)] was found to hold for values $R = r/r_0$ corresponding to variation of $\Psi(r)/\Psi(0)$ in the interval $1 \geq \Psi(r)/\Psi(0) \geq 0.001$ even when the coefficient C in (23) was estimated using an effective concentration n_t smaller by 4–6 orders than that used to obtain

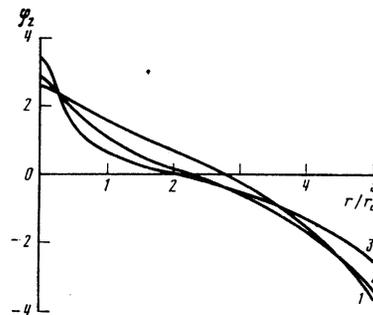


FIG. 9. Plots of the function φ_2 at different values of the parameter $y > 1$: $y = 2$ (curve 1), $y = 4$ (2), $y = 8$ (3).

the estimate [27(b)]. A more detailed estimate calls for experimental data, primarily concerning the values of $S_{aa}(0)$.

In calculations of a number of properties of a disordered semiconductor (such as the state density in the band gap, the position of the Fermi level, and others), the character of the behavior of the correlation function $\Psi(r)$ at large distance does not play a major role. In this case, as follows from all the foregoing, the random self-field in a heteropolar semiconductor with intermediate order at $r_c \gg \Omega_0^{1/3}$ in a wide range of the intermediate order parameter $y = r_c/r_0$ can be regarded as smooth, then the smooth-field model can be used for the interpretation of the experimental data.

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