Coherent backscattering from anisotropic scatterers

L. V. Kuzmin, V. P. Romanov, and L. A. Zubkov
Department of Physics, St. Petersburg State University, Petrodvoretz, St. Petersburg, 198904, Russia
(Received 5 September 1995)

The intensity of coherent backscattering from pointlike anisotropic scatterers is calculated. The polarized component has a peak in the backward direction, whereas the depolarized component does not exhibit a backscattering enhancement unlike the depolarized component for the isotropic scatterer case. These results agree with the measurement data on a disordered nematic liquid crystal. [S1063-651X(96)02208-8]

PACS number(s): 42.25.−p

Coherent backscattering [1–10] manifests itself as a sharp enhancement of light scattered backward in a narrow angular interval $\theta \sim \lambda / l_{\text{ext}}$ where λ is the wavelength and l_{ext} is the extinction length. The effect is observed in highly opalescing systems in which the extinction length is significantly less than the linear size of the system. It has been studied in latex suspensions [1–3,5,11,12], ceramics [4], porous glasses [13], etc.

The physical mechanism underlying coherent backscattering is quite simple. The coherent plane waves incident upon a turbid system become generally incoherent due to the multiple scattering from randomized inhomogeneities except for the waves which pass some sequence of scatterers in opposite directions. These waves can be coherent. However their interference is important only for backward direction. Such an interference is quite general for any wave process and it was first anticipated and discovered in the multiple scattering of conductance electrons in disordered metals [14,15].

The backscattering problem was solved analytically for pointlike scatterers in Refs. [6,7,9,16] for a scalar field and in Refs. [7,8,17,18] for an electromagnetic field. Stephen and Cwilich [7] took into consideration the polarization effects. They showed that the backscattering peak occurs in both polarized and depolarized components. The magnitude of the peak of the polarized component is five to seven times higher than that of the depolarized component, and its form is close to triangular while the form of the peak of the depolarized component is close to Lorentzian. These results are confirmed in numerous light scattering experiments in latex suspensions [1–3,5,11,12].

The coherent backscattering problem may be important not only for scalar scatterers but also for tensor ones since this effect is studied for various systems such as ceramics [4], porous glasses [13], polycrystals and liquid crystals [4,11], etc. In these systems the permittivity anisotropy may be significant. The coherent backscattering from anisotropic pointlike scatterers was briefly considered in [7]. However one cannot test experimentally results obtained there because an expression for the scattered light intensity, obligatorily real, contains an imaginary part for the case of totally anisotropic fluctuations.

This paper presents the study of coherent backscattering from the pointlike anisotropic scatterers. We find that the depolarized component does not practically exhibit the backscattering enhancement unlike the depolarized component for the isotropic scatterer case. The backscattering measurements performed in a disordered nematic verify this theoretical result.

The wave equation describing the monochromatic electromagnetic wave in a nonmagnetic dielectric medium with random inhomogeneous permittivity $\epsilon(\mathbf{r})$ can be presented in the integral form

$$\mathbf{E}_{fl}(\mathbf{r}) = \mathbf{E}(\mathbf{r}) + \int d\mathbf{r}_1 \hat{T}(\mathbf{r}-\mathbf{r}_1)\, \frac{\Delta\epsilon(\mathbf{r}_1)}{4\pi}\, \mathbf{E}_{fl}(\mathbf{r}_1), \qquad (1)$$

where $\mathbf{E}_{fl}(\mathbf{r})$ is the electric field in the randomly fluctuating medium, $\mathbf{E}(\mathbf{r})$ is the average field in a homogeneous medium with permittivity ϵ, $\hat{T}(\mathbf{r})$ is the electromagnetic field propagator

$$\hat{T}(\mathbf{r}) = \frac{k^2}{\epsilon r}\left(\hat{I} - \frac{\mathbf{r}\times\mathbf{r}}{r^2}\right)\exp(ikr). \qquad (2)$$

$\hat{I}$ is the unit matrix, $k = \omega n/c$ is the wave number, ω is the circular frequency, n is the average refractive index, and c is the light velocity in vacuum. We omit factor $\exp(i\omega t)$ describing the field temporal dependence assuming fluctuations of $\epsilon(\mathbf{r})$ to be static. Parameter $\Delta\epsilon(\mathbf{r}) = \epsilon(\mathbf{r}) - \epsilon$ describes the fluctuations of the permittivity where $\epsilon = \langle \epsilon(\mathbf{r})\rangle$ is the permittivity average.

The integral light scattering intensity is defined as $I = \langle \mathbf{E}_{fl}\mathbf{E}^*_{fl}\rangle - \mathbf{E}\mathbf{E}^*$. Iterating Eq. (1) and substituting it into intensity I one obtains the intensity in the form of series in orders of the $\Delta\epsilon$ fluctuations. We assume that the Gaussian approximation for averages of the form $\langle \Delta\epsilon^n\rangle$ is valid, i.e., these averages can be presented as all possible products of the binary correlation functions $\langle \Delta\epsilon(\mathbf{r}_1)\Delta\epsilon(\mathbf{r}_2)\rangle$. The correlation length r_c which determines the spatial range of this correlation function is assumed to be negligible as compared with the wavelength λ: $r_c \ll \lambda$, and the correlation function can be presented in the pointlike form [7]

$$\langle \Delta\epsilon(\mathbf{r}_i)\Delta\epsilon(\mathbf{r}_j)\rangle = (4\pi)^2 g_0 \delta(\mathbf{r}_i - \mathbf{r}_j). \qquad (3)$$

Neglecting the scattering from the isotropic part of the permittivity fluctuations we present the correlation function in the form of the traceless tensor

$$\langle \Delta\hat{\epsilon}(\mathbf{r}_i)\Delta\hat{\epsilon}(\mathbf{r}_j)\rangle = \delta(\mathbf{r}_i - \mathbf{r}_j)\langle \Delta\hat{\epsilon}\Delta\hat{\epsilon}\rangle, \qquad (4)$$

where

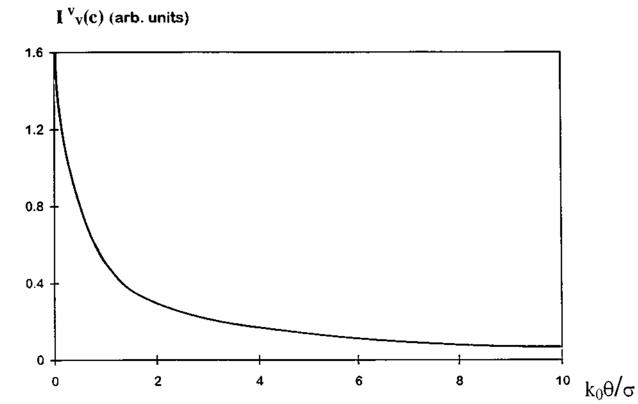

FIG. 1. The cyclic diagram contribution into the polarized component of the backscattering intensity for anisotropic scatterers.

$$\langle \Delta\epsilon_{\alpha\beta}\Delta\epsilon_{\gamma\delta}\rangle = \frac{a_0}{2}\,(\delta_{\alpha\gamma}\delta_{\beta,\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma} - \tfrac{2}{3}\,\delta_{\alpha\beta}\delta_{\gamma\delta}). \quad (5)$$

This function describes the correlation of orientation fluctuations in an isotropic medium.

To reveal the main features of coherent backscattering from purely anisotropic fluctuations we first consider the scattering strictly backward within the double scattering approximation. The ladder diagram contributes

$$I^{\beta}_{2\alpha}(L) = \int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}'_1 d\mathbf{r}'_2 T_{\alpha\alpha_1}(\mathbf{r}_0 - \mathbf{r}_2) T^*_{\alpha\alpha_2}(\mathbf{r}_0 - \mathbf{r}'_2) \times \delta(\mathbf{r}_2 - \mathbf{r}'_2)\langle \Delta\epsilon_{\alpha_1\nu_1}\Delta\epsilon_{\alpha_2\nu_2}\rangle \times T_{\nu_1\gamma_1}(\mathbf{r}_2 - \mathbf{r}_1) T^*_{\nu_2\gamma_2}(\mathbf{r}'_2 - \mathbf{r}'_1)\,\delta(\mathbf{r}_1 - \mathbf{r}'_1) \times \langle \Delta\epsilon_{\gamma_1\beta}\Delta\epsilon_{\gamma_2\beta}\rangle E_{\beta}(\mathbf{r}_1) E^*_{\beta}(\mathbf{r}'_1), \quad (6)$$

We present the product of a pair of propagators in the form

$$T_{\phi\mu}(\mathbf{r}) T^*_{\gamma\rho}(\mathbf{r}) \equiv \Lambda_{\phi\gamma,\mu\rho}(\mathbf{r}) = \frac{k_0^4 \exp(-\sigma r)}{r^2}\left(\delta_{\phi\mu} - \frac{r_\phi r_\mu}{r^2}\right) \times \left(\delta_{\gamma\rho} - \frac{r_\gamma r_\rho}{r^2}\right), \quad (7)$$

where $\sigma = l^{-1}_{\mathrm{ext}}$. Using Eqs. (4) and (6) we rewrite the double scattering intensity as follows

$$I^{\beta}_{2\alpha}(L) = A\int d\mathbf{r}_1 d\mathbf{r}_2 (\Lambda_{\nu\nu,\gamma\gamma} + \tfrac{1}{3}\Lambda_{\alpha\alpha,\gamma\gamma} + \tfrac{1}{3}\Lambda_{\nu\nu,\beta\beta} + \tfrac{1}{9}\Lambda_{\alpha\alpha,\beta\beta}), \quad (8)$$

where $\hat{\Lambda} = \hat{\Lambda}(\mathbf{r}_1 - \mathbf{r}_2)$, $A = [(k_0^4|E|^2)/r_0^2](a_0^2/4)$, r_0 is the distance from the system to the observation point. The contribution of the cyclic diagram into the double scattering intensity in the backward direction is

$$I^{\beta}_{2\alpha}(C) = A\int d\mathbf{r}_1 d\mathbf{r}_2 [\,\delta_{\alpha\beta}(\Lambda_{\beta\gamma_1,\gamma_1\alpha} + \Lambda_{\nu_1\beta,\alpha\nu_1} + \Lambda_{\nu_1\gamma_1,\gamma_1\nu_1} - \tfrac{2}{3}\Lambda_{\nu_1\alpha,\beta\nu_1} - \tfrac{2}{3}\Lambda_{\alpha\gamma_1,\gamma_1\beta}) - \tfrac{4}{3}\Lambda_{\beta\alpha,\beta\alpha} + \tfrac{13}{9}\Lambda_{\beta\beta,\alpha\alpha}]. \quad (9)$$

Calculating integrals in case of a medium occupying a half-space we obtain for the polarized I^V_V and depolarized I^H_V components

$$I^V_{2V}(L) = I^V_{2V}(C) = \frac{1252}{135}\,AVk_0^4\pi l_{\mathrm{ext}},$$
$$I^H_{2V}(L) = \frac{1228}{135}\,AVk_0^4\pi l_{\mathrm{ext}}, \quad I^H_{2V}(C) = -\frac{236}{135}\,AVk_0^4\pi l_{\mathrm{ext}}, \quad (10)$$

where V is the scattering volume.

As is seen from Eq. (10) the contributions of the ladder and cyclic diagrams to the polarized component are equal. It means that the polarized component exhibits the coherent backscattering peak. As for the depolarized component the cyclic diagram contribution is small as compared with that of the ladder one and even negative. It means that at least within the double scattering approximation there is no back-

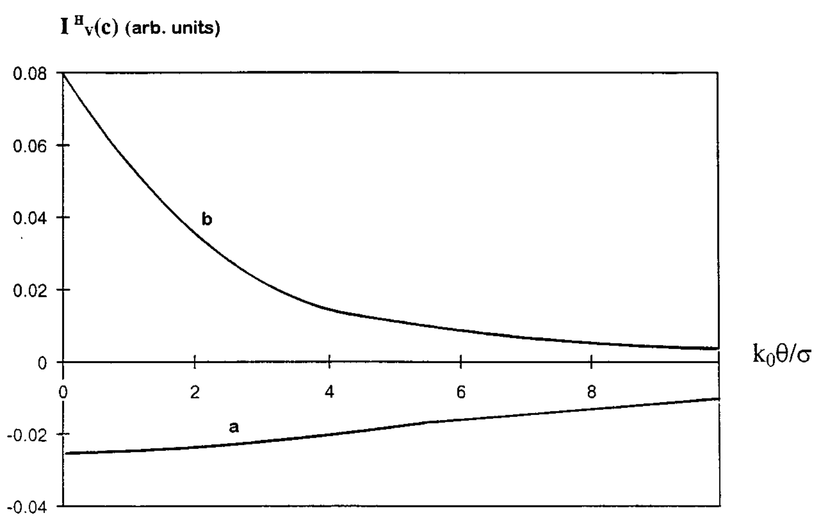

FIG. 2. The cyclic diagram contribution into the depolarized component: (a) anisotropic scatterers, (b) isotropic scatterers.

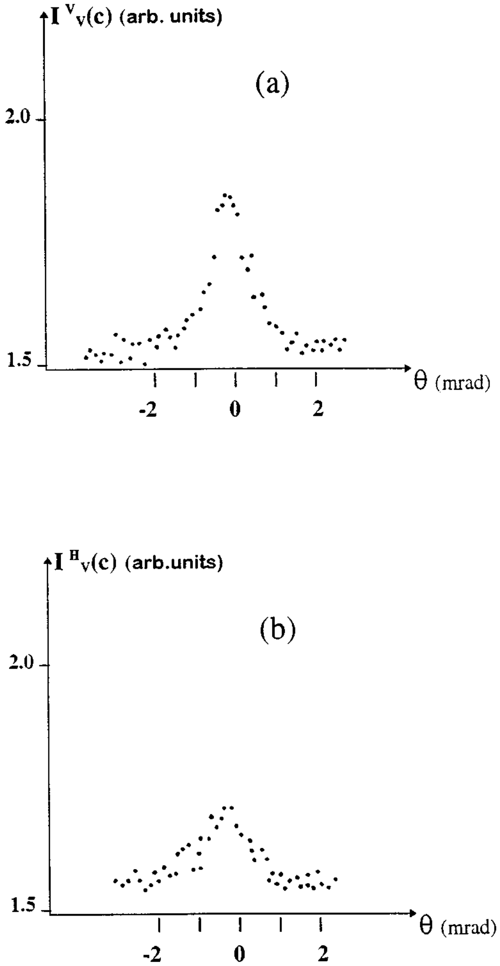

FIG. 3. The coherent backscattering for the polarized (a) and depolarized (b) components in the 0.55-mkm-diam latex suspension.

scattering enhancement for the depolarized component. The negative sign of the depolarized component of the cyclic diagram does not contradict the requirement for the intensity to be positive as a whole.

In the general case one has to sum a series in scattering orders. This sum can be presented in the operator form

$$\hat{S}=\hat{N}+\hat{M}\hat{N}+\hat{M}\hat{M}\hat{N}+\cdots=\sum_{n=0}^{\infty}\hat{M}^{n}\hat{N}, \qquad (11)$$

where tensors $\hat{N}$ and $\hat{M}$ are defined as

$$N_{\mu_1\mu_2,\alpha_1\alpha_2}(\mathbf{r}_1-\mathbf{r}_2)=\langle\Delta\epsilon_{\mu_1\gamma_1}\Delta\epsilon_{\mu_2\gamma_2}\rangle\Lambda_{\gamma_1\gamma_2,\beta_1\beta_2}(\mathbf{r}_1-\mathbf{r}_2)\times\langle\Delta\epsilon_{\beta_1\alpha_1}\Delta\epsilon_{\beta_2\alpha_2}\rangle, \qquad (12)$$

$$M_{\mu_1\mu_2,\alpha_1\alpha_2}(\mathbf{r}_1-\mathbf{r}_2)=\langle\Delta\epsilon_{\mu_1\gamma_1}\Delta\epsilon_{\mu_2\gamma_2}\rangle\Lambda_{\gamma_1\gamma_2,\alpha_1\alpha_2}(\mathbf{r}_1-\mathbf{r}_2).$$

Summing series (11) one obtains a closed equation for the radiation transfer propagator

$$S_{\alpha\beta,\gamma\delta}(\mathbf{r}_1,\mathbf{r}_2)=N_{\alpha\beta\gamma\delta}(\mathbf{r}_1-\mathbf{r}_2)+\int d\mathbf{r}_3M_{\alpha\beta,\mu\nu}\times(\mathbf{r}_1-\mathbf{r}_3)S_{\mu\nu,\gamma\delta}(\mathbf{r}_3,\mathbf{r}_2). \qquad (13)$$

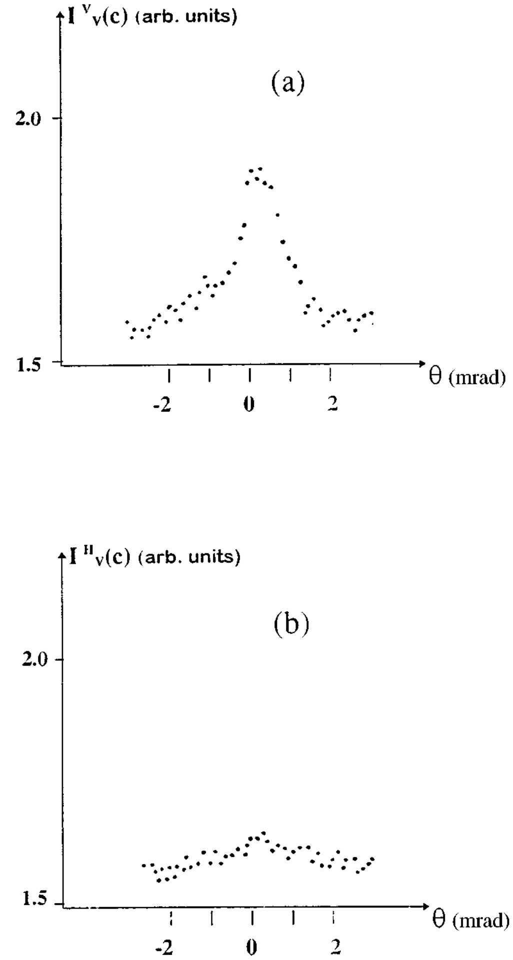

FIG. 4. The coherent backscattering as a function of angle in the disordered liquid crystal: (a) I_V^V component, (b) I_V^H component.

Considering the problem of scattering from the medium occupying half-space $z\geqslant 0$, where z is the Cartesian coordinate, $\mathbf{r}=(x,y,z)$, one has explicitly to take into account boundary conditions. In accordance with Ref. [7] we require propagator $\hat{S}$ to be zero at the boundary $z=0$. Using the image method one finds the solution of Eq. (16) in the form

$$\hat{S}(\mathbf{r}_1,\mathbf{r}_2)=\hat{S}_0(\mathbf{r}_1-\mathbf{r}_2)-\hat{S}_0(\mathbf{r}_1-\mathbf{r}^{(s)}), \qquad (14)$$

where $S_0(\mathbf{r})$ is the corresponding solution for an infinite homogeneous system and $\mathbf{r}_2^{(s)}$ is the mirror image with respect to $\mathbf{r}_2$ [$\mathbf{r}_2^{(s)}=(x_2,y_2,-z_2)$]. The radiation transfer propagator $\hat{S}(\mathbf{r}_1,\mathbf{r}_2)$ given by Eq. (14) turns out to be zero if at least one of two points $\mathbf{r}_1$ and $\mathbf{r}_2$ lies in the $z=0$ plane. For a homogeneous system propagator $\hat{S}_0$ is easily obtained by means of a Fourier transformation of Eq. (13).

Assuming that the wave vector of the scattered light lies in the x,y plane the separate inputs to the scattered intensity components can be presented in the form

$$I_V^V(L)=B\int_{-\infty}^{\infty}dq_zF(q_z)\widetilde{S}_{yy,yy}(q_z), \qquad (15)$$

$$I_V^H(L)=B\int_{-\infty}^{\infty}dq_zF(q_z)\widetilde{S}_{xx,yy}(q_z),$$

$$I_V^V(C)=B\int_{-\infty}^{\infty}dq_zF(q_z)\widetilde{S}_{yy,yy}(\sqrt{q_z^2+k_0^2\theta^2}),$$

$$I_H^V(C)=B\int_{-\infty}^{\infty}dq_zF(q_z)[\widetilde{S}_{xy,yx}(\sqrt{q_z^2+k_0^2\theta^2})\cos^2\phi$$
$$+\widetilde{S}_{xz,zx}(\sqrt{q_z^2+k_0^2\theta^2})\sin^2\phi],$$

where

$$B=\frac{k_0^4s|E|^2}{2\pi r_0^2},\quad F(q_z)=\frac{2q_z^2}{(\sigma^2+q_z^2)^2},$$

$\phi=\arctan(k_0\theta/q_z)$, $\tilde{\tilde{S}}$ is the Fourier transform of the radiation transfer propagation, θ is the scattering angle measured from the backward direction, and s is the illuminated area.

We calculate numerically the scattered light intensities formed by the series of the ladder as well as cyclic diagrams. The calculated backscattering intensity of the polarized component is plotted against the scattering angle in Fig. 1. The form of the backscattering peak is close to triangular and its height is approximately equal to the intensity of the background. The behavior of the depolarized component is quite different. Input $I_V^H(C)$ appears to be negative, and its absolute value is in 75 times less than the value of the corresponding ladder input $I_V^H(L)$. This negative contribution of the sum of cyclic diagrams to the intensity is plotted in Fig. 2. As is seen, this contribution vanishes significantly slower than that of the polarized component. The angular dependence of the depolarized component for the case of backscattering from isotropic permittivity fluctuations is also shown in this figure exhibiting a more rapid angular decrease.

We test these results experimentally. A continuous mode He-Ne gas laser is used as a source of radiation. The principal optical element of the setup is a beam splitter. A part of the incident beam is reflected from the splitter and hits the cell, whereas the second part is used for adjusting the optical system. The backscattered light passing through the splitter enters a detecting photomultiplier and afterwards comes to a multichannel analyzer. The cell is filled with a disordered nematic liquid crystal, the $MBBA+EBBA$ mixture, at a temperature 25° C. Such a liquid crystal is thought to consist of small domains of the order of several microns with random local ordering. These domains exhibit no sharp boundaries changing their orientations continuously. Therefore the light is assumed to be scattered by the ordering fluctuations whereas the depolarization ratio for the single scattering is said to be close to 3/4.

We measure the polarized I_V^V and depolarized I_V^H components in the disordered liquid crystal. Since the peak height of the I_V^V component is quite moderate special attention was paid to remove noises caused by a diffusive reflection as well as scattering from all elements of the setup. To control the data obtained and improve their validity we perform test measurements in some well-studied systems. To this aim first we fill the cell with a solution of the black water color. The absorption coefficient of the solution is in the range $\sim10^4$–10^5 cm^{-1}. The diffusive intensity does not exceed 2% of the total scattering intensity for scattering angle $\theta=180°$. We also measure the backscattering from a 0.55 mkm diam latex particle suspension. We choose a concentration of the suspension so the backscattering peak value can be close to that of the disordered nematic. The scattered light I_V^V and I_V^H components are plotted in Fig. 3 for the 2.3% latex suspension. The points present the intensity averages of the scattered light measured by the multichannels analyzer.

The intensities of backscattering from the disordered $MBBA+EBBA$ mixture are plotted in Fig. 4. As is seen the I_V^V component exhibits the peak in the backward direction. On the contrary, the depolarized component does not depend practically on the angle. The small peak, of order of 3%, is probably produced by the diffusive scattering. Nevertheless if this peak has the physical origin its height is at least several times less than that in the latex, i.e., in the scalar scatterer system.

[1] M. P. Van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985).
[2] P. E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).
[3] S. Etemad, R. Thompson, and H. J. Andreiko, Phys. Rev. Lett. **57**, 575 (1986).
[4] H. Kaveh, M. Rosenbluh, I. Edrei, and I. Freund, Phys. Rev. Lett. **57**, 2049 (1986).
[5] Y. Kuga and A. J. Ishimaru, J. Opt. Soc. Am. A **8**, 831 (1984).
[6] E. Akkermans, P. E. Wolf, and R. Maynard, Phys. Rev. Lett. **56**, 1471 (1986).
[7] M. J. Stephen and G. Cwilich, Phys. Rev. B **34**, 7564 (1986).
[8] E. Akkermans, P. E. Wolf, R. Maynard, and G. Maret, J. Phys. (Paris) **49**, 77 (1988).
[9] Yu. N. Barabanenkov and V. D. Ozrin, Zh. Eksp. Teor. Fiz. **94**, 56 (1988) [Sov. Phys. JETP **67**, 30 (1988)].
[10] G. Maret and P. Wolf, Z. Phys. B **65**, 409 (1987).
[11] D. V. Vlasov, L. A. Zubkov, N. V. Orekhova, and V. P. Romanov, Pisma Zh. Eksp. Teor. Fiz. **48**, 86 (1988) [JETP Lett. **48**, 91 (1988)].
[12] A. Almamuri, D. V. Vlasov, L. A. Zubkov, and V. P. Romanov, Zh. Eksp. Teor. Fiz. **99**, 1431 (1991) [Sov. Phys. JETP **72**, 798 (1991)].
[13] S. Kawato, T. Hattori, T. Takemori, and H. Nakatsuka, Phys. Rev. B **49**, 90 (1994).
[14] G. Bergmann, Phys. Rep. **107**, 1 (1984).
[15] P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[16] A. A. Golubentsev, Zh. Eksp. Teor. Fiz. **86**, 47 (1984) [Sov. Phys. JETP **59**, 26 (1984)].
[17] V. L. Kuzmin, V. P. Romanov, and L. V. Kuzmin, Opt. Spektrosk. **72**, 227 (1992) [Opt. Spectrosc. (USSR) **72**, 125 (1992)].
[18] V. L. Kuzmin, V. P. Romanov, and L. V. Kuzmin, Opt. Spektrosk. **73**, 376 (1992) [Opt. Spectrosc. (USSR) **73**, 220 (1992)].