

Anisotropy of momentum distribution of photoexcited electrons and polarization of hot luminescence in semiconductors

V. D. Dymnikov, M. I. D'yakonov, and N. I. Perel'

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences

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It is shown that the photoexcited electrons produced in interband absorption of light in semiconductors with gallium arsenide band structure have an anisotropic momentum distribution. Excitation with circularly polarized light produces, besides anisotropy of the momentum distribution, also correlations between the directions of the electron momentum and its spin. The hot-photoluminescence polarization characteristics are obtained and found to be completely determined by this anisotropy. As the energy of the electrons relaxes, their distribution function becomes isotropic, as a result of which the degree of polarization decreases with increasing luminescence wavelength. The hot-luminescence polarization spectra can yield interesting information on the energy, momentum, and spin relaxation of the electrons.

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1. INTRODUCTION

Zemskii, Zakharchenya, and Mirlin^[1] have observed unusual polarization phenomena in their experiments on hot luminescence of gallium arsenide and in solid solutions on its basis. The short-wave recombination radiation following interband absorption of linearly polarized light turned out to be also linearly polarized. (Linear polarization of the luminescence in cubic-symmetry crystals was heretofore observed only in the presence of a strain^[2] or of an electric field.^[3,4]) In addition, anomalously high circular polarization of the hot luminescence was observed upon excitation with circularly polarized light.

The purpose of the present paper is to show that the singularities of the hot luminescence polarization can be caused by anisotropy of the momentum distribution of the photoexcited electrons following interband absorption of light in semiconductors having a GaAs band structure. The cause of this anisotropy is the firm connection between the angular momentum and the momentum in the valence band of such semiconductors. As noted in^[5], the transition from the heavy-hole band (angular-momentum projection on the momentum $\pm \frac{3}{2}$) into the conduction band can be set in correspondence with an oscillator oscillating in a plane perpendicular to the momentum. The oscillator corresponding to a transition from a light-hole band (angular-momentum projection on momentum $\pm \frac{1}{2}$) is predominantly (60%) polarized along the momentum direction. Therefore excitation from the heavy-hole band gives rise to carriers having a predominant momentum direction perpendicular to the electric vector \mathbf{e} of the exciting light. On the contrary, excitation from the light-hole band produces carriers with momenta that are for the most part parallel or antiparallel to \mathbf{e} . It is clear that the momentum distribution of the photoexcited carriers will be anisotropic not only if the excitation is by linearly polarized light, but also following excitation by unpolarized or circularly polarized light. In these last two cases the anisotropy axis is the direction of the exciting beam. Since the selection rules are the same in radiative recombination as in the case

of excitation, it is obvious that anisotropy of the momentum distribution leads to a linear polarization of the luminescence.

If the excitation is by circularly polarized light, the produced electrons are oriented along the spin (optical orientation^[6,7]). The degree of the orientation of the electrons, averaged over the momentum directions, amounts to 50% at the instant of excitation, but, as shown earlier,^[8] electrons with different momenta have different polarizations that depend on the angle between the momentum directions and the exciting beam. This correlation must lead to an excess circular polarization of the luminescence.

The anisotropy of the momentum distribution of the photoexcited carriers decays with a characteristic time on the order of the momentum relaxation time τ_p . The customarily observed edge luminescence is therefore due to carriers with an isotropic momentum distribution, since the time τ_p is much shorter than the lifetime. Thus, the anisotropy that is produced at the instant of excitation, as well as the correlation between the electron spins and their momenta, can manifest themselves only in hot luminescence due to electrons whose momentum distribution has not yet become isotropic. In Sec. 2 of the present paper we calculate the momentum-dependent spin-density matrix of the electrons produced in the conduction band when a semiconductor of the GaAs type is illuminated with linearly or circularly polarized light. The spin and momentum anisotropy of this matrix can be characterized by three parameters, the values of which depend on the type of transition (excitations from the heavy- or light-hole band). In the luminescence spectrum, the photon energy is connected with the energy of the recombining electrons. The short-wave edge of the hot luminescence is due to electrons that have not yet managed to lose their energy, spin, and momentum. The radiation of electrons that undergo energy relaxation is shifted to the long-wave region of the spectrum.

In Sec. 3 we present the general form of the density

matrix of the photoexcited electrons under conditions of stationary excitation; this matrix also contains three anisotropy parameters. The dependence of these parameters on the energy determines the variation of the degree of linear or circular polarization over the luminescence spectrum. The degree of polarization is expressed in terms of these parameters under the assumption that the holes have an equilibrium distribution. This assumption corresponds to photoluminescence in a *p*-type semiconductor, in which the non-equilibrium electrons recombine with equilibrium holes.

The energy dependence of the anisotropy parameters is connected with the character of the energy, momentum, or spin relaxation of the hot electrons. This connection is discussed in Sec. 4. It is shown that the spectral dependence of the polarization of the hot photoluminescence is determined directly by the ratios of the momentum and spin relaxation times to the energy relaxation time.

2. DENSITY MATRIX OF PHOTOEXCITED ELECTRONS

The density matrix describing the generation of electrons in the conduction band is given by

$$F_{mm'}(\mathbf{p}) = C(p) \sum_M (\mathbf{p}m | \mathbf{e}d | \mathbf{p}M) (\mathbf{p}m' | \mathbf{e}d | \mathbf{p}M), \quad (1)$$

where C is a function of the electron energy, is proportional to the relative density of states, and is different for transitions from the heavy- and light-electron holes; \mathbf{e} is the unit vector of the polarization of the light; \mathbf{d} is the dipole-moment operator; the subscripts m and m' label the spin-degenerate states of the electrons in the conduction band ($m, m' = \pm \frac{1}{2}$). The summation is over valence-band states characterized by the projection M of the angular-momentum on the direction of the momentum \mathbf{p} (for transitions from the light-hole band the summation includes the values $M = \pm \frac{1}{2}$, while for transitions from the heavy-hole bands it includes the values $M = \pm \frac{3}{2}$). We confine ourselves henceforth to the case when the significant energies in the valence band are less than the spin-orbit splitting Δ . The matrix elements in (1) then depend only on the direction of the momentum \mathbf{p} , but not on its magnitude. For simplicity we neglect the corrugations of the equal-energy surfaces in the valence band.

The most general expression for the density matrix (1) in the case of linear polarization of the exciting light can be written in the form

$$F_{mm'}(\mathbf{p}) = \delta_{mm'} F_0(\varepsilon) \left[1 + \alpha_0 \frac{3(\mathbf{e}\mathbf{v})^2 - 1}{2} \right], \quad (2)$$

where \mathbf{v} is a unit vector along the direction of \mathbf{p} ; $F_0(\varepsilon)$ is a function of the electron energy ε and differs from $C(p)$ only by a constant factor. The coefficient α_0 characterizes the degree of anisotropy of the momentum distribution of the electrons.

In the case of circular polarization of the exciting light, the expression for the density matrix of the produced electrons takes the form

TABLE I. Values of the parameters characterizing the anisotropy of the momentum distribution and the spin orientation of the electrons at the instant of production.

Transition	α_0	S_0	β_0
$h-c$	-1	-1/4	-1
$l-c$	1	-1/4	1

$$\hat{F}(\mathbf{p}) = F_0(\varepsilon) \left\{ \hat{I} \left[1 - \alpha_0 \frac{3(\mathbf{n}\mathbf{v})^2 - 1}{4} \right] + 2S_0(\hat{\sigma}\mathbf{n}) + \beta_0 \frac{3(\hat{\sigma}\mathbf{v})(\mathbf{n}\mathbf{v}) - (\hat{\sigma}\mathbf{n})}{2} \right\}, \quad (3)$$

where \hat{I} is a unit matrix, $\hat{\sigma}$ are Pauli matrices, and \mathbf{n} is a unit vector along the angular momentum of the photon in the exciting beam (the vector \mathbf{n} is parallel or antiparallel to the direction of the light beam, depending on the sign of the circular polarization). The coefficient α_0 coincides with the coefficient in formula (2). $S_0\mathbf{n}$ is the average spin of the electrons at the instant of production, and the coefficient β_0 characterizes the correlation between the electron spin and its momentum. The values of the coefficients α_0 , S_0 , and β_0 , calculated with the aid of formula (1), are listed in Table I. The symbols $h-c$ and $l-c$ denote respectively transitions from the band of heavy holes into the conduction band and from the band of light holes into the conduction band. The negative sign of S_0 means that the average spin of the photoexcited electrons is directed opposite to the angular momentum of the photons in the exciting beam (the remaining part of the angular momentum ($\frac{5}{2}$) is transferred to the photoexcited hole).

With the aid of the density matrix (3) we can calculate the average value of the spin vector of an electron with momentum \mathbf{p} . For transitions from the heavy-hole band we obtain

$$\mathbf{S}(\mathbf{p}) = -\frac{(\mathbf{n}\mathbf{v})\mathbf{v}}{1 + (\mathbf{n}\mathbf{v})^2}, \quad (4)$$

in which case the electron spin is parallel to its momentum. For transitions from the light-hole band we have

$$\mathbf{S}(\mathbf{p}) = \frac{3\mathbf{v}(\mathbf{n}\mathbf{v}) - 2\mathbf{n}}{5 - 3(\mathbf{n}\mathbf{v})^2}. \quad (5)$$

These formulas agree with the corresponding expressions of [6].

We note that when unpolarized light is absorbed, an anisotropy of the momentum distribution of the produced electrons also sets in. This anisotropy is described by formula (3), in which we must put $S_0 = \beta_0 = 0$.

3. INFLUENCE OF ANISOTROPY OF THE MOMENTUM DISTRIBUTION ON THE LUMINESCENCE POLARIZATION

The electrons that are lifted high into the conduction band by the light relax in energy, momentum, and spin. The density matrix under the conditions of stationary

excitation has a general form analogous to expressions (2) and (3):

$$f(\mathbf{p}) = I f(\varepsilon) \left[1 + \alpha \frac{3(\mathbf{e}\mathbf{v})^2 - 1}{2} \right], \quad (6)$$

$$f(\mathbf{p}) = f(\varepsilon) \left\{ I \left[1 - \alpha \frac{3(n\mathbf{v})^2 - 1}{4} \right] + 2S(\hat{\mathbf{g}}\mathbf{n}) + \beta \frac{3(\hat{\mathbf{g}}\mathbf{v})(n\mathbf{v}) - (\hat{\mathbf{g}}\mathbf{n})}{2} \right\}. \quad (7)$$

Formulas (6) and (7) pertain to cases of excitation with linearly and circularly polarized light, respectively. The coefficients α , β , and S in (6) and (7) are determined by the relaxation of the electrons and depend on the energy ε . At $\varepsilon = \varepsilon_i$ (ε_i is the initial energy of the photoexcited electron), these coefficients have the values listed in Table I, and their absolute values decrease with decreasing energy.

The intensity of the recombination radiation with polarization \mathbf{e}_1 is expressed in terms of the density matrix \hat{f} in the following manner:

$$J_{\mathbf{e}_1}(\omega) = C_1(\omega) \int d\Omega_v \sum_{mm'} \left\{ \sum_M (\mathbf{p}m' | \mathbf{e}_1 | \mathbf{p}M) (\mathbf{p}m | \mathbf{e}_1 | \mathbf{p}M)^* \right\}, \quad (8)$$

where $C_1(\omega)$ is a function of the frequency of the light and is determined by the reduced density of state and by the energy distribution of the recombining holes, which are assumed to be in equilibrium. The integration is over the directions of the electron momentum. The summation is over the spin states of the electrons in the conduction band ($m, m' = \pm \frac{1}{2}$) and over the states of the recombining holes, characterized by the index M . This index takes on the values $\pm \frac{1}{2}$ when electrons recombine with light holes and $\pm \frac{3}{2}$ for recombination with participation of heavy holes. The value of $C_1(\omega)$ also depends on the type of transition. The frequency ω is connected in the usual manner with the energy of the recombining electron and the type of the recombining hole. Formula (8) is valid also for recombination of an electron on an acceptor level. In this case the final state is characterized only by the symbol M , which runs through the four values $\pm \frac{3}{2}$ and $\pm \frac{1}{2}$ in accordance with the fourfold degeneracy of the acceptor level.

The expression in the curly brackets in (8) is a matrix in terms of the subscripts m and m' , and differs only by a factor from the matrix (1). It is therefore of the same form as (2) and (3) for linear and circular polarizations, respectively, with the same values of α_0 , β_0 , and S_0 . In formulas (2) and (3) it is necessary in this case to replace the polarization vector \mathbf{e} by \mathbf{e}_1 , and the direction \mathbf{n} by the direction \mathbf{n}_1 of the angular momentum of the radiated photon.

We introduce the degrees \mathcal{P}_l and \mathcal{P}_c of the linear and

TABLE II. Values of the initial degree of linear polarization \mathcal{P}_l for different types of excitation and recombination.

Excitation	Recombination		
	c-h	c-l	c-a
$h-c$	$1/7$	$-3/19$	$1/7$
$l-c$	$-3/19$	$1/7$	$-3/19$

TABLE III. Values of the initial degree of circular polarization \mathcal{P}_c for different types of excitation and recombination.

Excitation	Recombination		
	c-h	c-l	c-a
$h-c$	$5/7$	$-5/19$	$5/7$
$l-c$	$-5/19$	$5/7$	$-5/19$

circular luminescence polarization, respectively:

$$\mathcal{P}_l = \frac{J_{l-} - J_{l+}}{J_{l-} + J_{l+}}, \quad \mathcal{P}_c = \frac{J_{c-} - J_{c+}}{J_{c-} + J_{c+}}.$$

With the aid of (6)–(8) we obtain the following expressions for these quantities:

$$\mathcal{P}_l = \frac{3\alpha\alpha_0}{20 + \alpha\alpha_0}, \quad \mathcal{P}_c = \frac{80SS_0 + 10\beta\beta_0}{20 + \alpha\alpha_0}. \quad (9)$$

The formula for \mathcal{P}_l is applicable when the excitation is with linearly polarized light, $\mathcal{P}_l > 0$ meaning that the luminescence is polarized in the same plane as the exciting light. The formula for \mathcal{P}_c holds for the case of excitation with circularly polarized light at $\mathbf{n} = \mathbf{n}_1$, and $\mathcal{P}_c > 0$ means that the angular momentum of the radiated photon has the same direction as the angular momentum of the absorbed photon.

For electrons with energy equal to the initial energy ε_i , which are as yet unaffected by momentum and spin relaxation, the coefficients α , β , and S in (6), (7), and (9) coincide with α_0 , β_0 , and S_0 (see Table I). The corresponding values of \mathcal{P}_l and \mathcal{P}_c for different types of excitation and recombination are given in Tables II and III. The tables include also data on the luminescence polarization which arises in the case of conduction band—acceptor ($c-a$) transitions. This polarization turns out to be the same as in electron transitions to the heavy-hole band ($c-h$). The reason is that the wave function of the ground state of the hole on the acceptor (at the usual relation $m_l \ll m_h$, where m_l and m_h are the masses of the light and heavy holes) is made up only of wave functions of the heavy-hole subband.^[9]

4. RELAXATION EFFECTS

To the extent that the photoexcited electrons relax in energy, they relax also in momentum and spin, and this leads to a decrease of the degree of polarization with increasing luminescence wavelength. The photoexcited electrons can lose energy by emitting phonons or by colliding with holes. We shall assume the possibility of introducing energy (τ_ε) and spin (τ_s) relaxation times as well as a relaxation time τ_{p2} that characterizes the vanishing of the anisotropy of the momentum distribution and is described by the second Legendre polynomial. We can then write down simple expressions for the time variation of the coefficients α , β , and S during the relaxation

$$\frac{d\alpha}{dt} = -\frac{\alpha}{\tau_{p2}}, \quad \frac{dS}{dt} = -\frac{S}{\tau_s}, \quad \frac{d\beta}{dt} = -\left(\frac{1}{\tau_s} + \frac{1}{\tau_{p2}} \right) \beta. \quad (10)$$

The dependence of α , β , and S on the energy can be obtained by using the relation $d\varepsilon/dt = -\varepsilon/\tau_\varepsilon$. We then obtain

$$S = S_0 \exp\left(-\int_{\varepsilon}^{\varepsilon_0} \frac{\tau_\varepsilon}{\tau_p} \frac{d\varepsilon}{\varepsilon}\right), \quad \alpha = \alpha_0 \exp\left(-\int_{\varepsilon}^{\varepsilon_0} \frac{\tau_\varepsilon}{\tau_{p2}} \frac{d\varepsilon}{\varepsilon}\right), \quad (11)$$

$$\beta = \beta_0 \exp\left[-\int_{\varepsilon}^{\varepsilon_0} \tau_\varepsilon \left(\frac{1}{\tau_\varepsilon} + \frac{1}{\tau_{p2}}\right) \frac{d\varepsilon}{\varepsilon}\right].$$

The first formula of (11) was discussed earlier¹⁾ in^[7], and the spin relaxation of hot electrons was investigated in a number of studies.^[10]

Let us examine the ratio $\tau_\varepsilon/\tau_{p2}$, which characterizes the decrease of the linear polarization of the luminescence with decreasing energy of the recombining electron. For electron scattering by heavy holes, τ_{p2} can be expressed in terms of the ordinary momentum relaxation time τ_p . Since the scattering angles is mostly small, we can write

$$\frac{\tau_{p2}}{\tau_p} = \lim_{\theta \rightarrow 0} \frac{1 - \cos \theta}{1 - P_2(\cos \theta)} = \frac{1}{3}.$$

Therefore, for scattering by heavy holes we have

$$\tau_\varepsilon/\tau_{p2} = 3m_h/2m_e, \quad (12)$$

where m_h and m_e are respectively the masses of the heavy hole and of the electron. The numerical value of this ratio for GaAs is ≈ 12 . To take into account the fact that the momentum relaxation takes place also on charged centers, expression (12) must be multiplied by the factor $(1 + N/n_h)$, where N and n_h are the concentrations of the charged centers and of the heavy holes, respectively. In our case $\alpha = \alpha_0(\varepsilon/\varepsilon_i)^{\tau_\varepsilon/\tau_{p2}}$. According to (9), the degree of linear polarization of the recombination radiation should likewise have practically the same energy dependence.

Another effective energy relaxation mechanism is the emission of optical phonons. An elementary calculation shows that after one optical phonon is emitted the parameter α changes in the following manner:

$$\alpha_1 = \alpha_0 \frac{(3a^2 - 1) \ln b - 3a}{2 \ln b}, \quad a = \frac{p^2 + p'^2}{2pp'}, \quad b = \frac{p + p'}{p - p'}, \quad (13)$$

where p and p' are the electron momenta before and after the phonon emission, respectively, and α_1 is the value of the parameter after emission of one optical phonon. For example, if the energy of the electron is 10 times the phonon energy $\hbar\omega_0$, then it follows from (13) that the emission of one phonon decreases the parameter α , meaning also the degree of linear polarization of the recombination radiation, by a factor 1.6. If we introduce the ratio $\tau_\varepsilon/\tau_{p2}$ for relaxation on optical phonons, then we obtain at $\varepsilon \gg \hbar\omega_0$

$$\frac{\tau_\varepsilon}{\tau_{p2}} = \frac{3}{2} \frac{\varepsilon/\hbar\omega_0}{\ln(4\varepsilon/\hbar\omega_0)}. \quad (14)$$

At $\varepsilon = 10\hbar\omega_0$ we have $\tau_\varepsilon/\tau_{p2} \approx 4$.

The experimental data of^[11] are in qualitative agree-

ment with the results reported above. The degree of linear polarization of the luminescence on the short-wave edge of the spectrum (15%) is close to the 14% that follow from Table II for the $h-c \rightarrow h$ transition. The observed spectral dependence of the degree of linear polarization, however, is weaker than might be expected if the energy relaxation were the result of emission of optical phonons or of electron-hole collisions. The relatively fast decrease of the circular polarization, and also its lower value (50%) at the short-wave luminescence edge in comparison with the theoretical value (71%), may be connected with the mechanism indicated in footnote 1.

It should be noted in conclusion that because of the spin and momentum anisotropies of the photoexcited-electron distribution, an investigation of the polarization spectra of hot photoluminescence can yield interesting information on the energy, momentum, and spin relaxation of the electrons.

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¹⁾The spin-relaxation mechanism connected with the spin splitting of the conduction band of semiconductors without an inversion center was investigated in^[7]. This splitting $\Delta\varepsilon$ can be regarded as the result of the action of a certain effective magnetic field perpendicular to the momentum. The case considered in^[7] $\Delta\varepsilon \ll \hbar/\tau_p$. For hot electrons the opposite limiting case may be realized. Then the spin projection perpendicular to the effective magnetic field vanishes within a time $\hbar/\Delta\varepsilon$. Since, according to (4), the spin of the electron lifted by the light into the conduction band from the heavy-hole subband is directed parallel to the momentum, it follows that the time characterizing the vanishing of the spin orientation of such electrons is equal to $\hbar/\Delta\varepsilon$ and is shorter than the momentum relaxation time. We note that the energy dependence of the parameters S and β is not described in this case by formulas (11).

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Nature of magnetic anisotropy of dysprosium. Paramagnetic susceptibility of dysprosium-yttrium alloys

V. G. Demidov, R. Z. Levitin, and O. D. Chistyakov

Moscow State University

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The magnetic susceptibility of single-crystal dysprosium-yttrium alloys was measured in the basal plane and along the hexagonal axis. It is shown that the susceptibility of these alloys obeys the Curie-Weiss law, the effective magnetic moments along the different directions are equal, and the paramagnetic Curie temperatures are different. The difference between the paramagnetic Curie temperatures in the basal plane and along the hexagonal axis does not depend on the dysprosium concentration in the alloys. This, as revealed by comparison with the theoretical models of magnetic anisotropy demonstrates that the magnetic anisotropy of dysprosium-yttrium alloys is of the one-ion type.

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INTRODUCTION

Important information on the contribution of the two principal mechanisms (one-ion anisotropy and exchange anisotropy) to the magnetic anisotropy of heavy rare earth metals (HREM) can be obtained by investigating alloys of HREM with one another. If the magnetic anisotropy is of one-ion origin, then it is made up additively of the anisotropies of the individual atoms and is a linear function of the alloy-component concentrations. But if the magnetic anisotropy is due to anisotropic exchange, then its magnitude is proportional to the number of pair interactions, i.e., it depends quadratically on the concentration of the alloy components.

The anisotropy of HREM depends strongly on the deviations of the real crystal structure from the ideal hcp structure: in the point-charge model, the coefficient of uniaxial anisotropy (when the anisotropy energy is expanded in harmonic polynomials) is for both anisotropy mechanisms^[1,2]

$$k_z^0 \propto \eta = a^{-3}(1.633 - c/a). \quad (1)$$

To discuss the experimental result it is therefore necessary to take into account the change of the crystal lattices parameters a and c with changing alloy composition.

Starting with the premises set forth above, we have reduced in^[3,4] the experimental data on the magnetic anisotropy of dysprosium-gadolinium alloys in the magnetically ordered state and in the paramagnetic region,

and have shown that the concentration dependence of k_z^0 of these alloys can be described within the framework of the single-ion model. It was therefore concluded in those papers that the magnetic anisotropy of dysprosium is single-ion and is due to the interaction of the anisotropic cloud of $4f$ electrons with the crystal lattice field. Recently, however, Boutron,^[2,5] using essentially the same concept, analyzed the experimental data on the anisotropy of dilute HREM alloys in gadolinium^[6] and of pure HREM,^[7-9] and arrived at the opposite conclusion, that a substantial contribution to the magnetic anisotropy of HREM (and, in particular, dysprosium) is made by the anisotropy due to exchange interaction: according to his calculations, it amounts to more than 50% of the one-ion anisotropy.

To determine more accurately the roles played by the various mechanisms in the magnetic anisotropy of dysprosium, and to ascertain the cause of the fundamental difference between our conclusions concerning the nature of the anisotropy of this metal and those of Boutron, we have investigated the magnetic anisotropy of dysprosium-yttrium alloys in the paramagnetic state. Yttrium has a zero magnetic moment and is a Pauli paramagnet. Therefore its susceptibility in the alloy can be regarded as an additive increment to the susceptibility due to the dysprosium, and this facilitates the calculation of the magnetic anisotropy of the alloy from the experimental anisotropy of the paramagnetic susceptibility. In addition, in dysprosium-yttrium alloys the coefficient η , which characterizes the deviation of the crystal structure of the alloy from the ideal hcp lat-