Theory of spontaneous and stimulated electroluminescence of ZnS: Mn Films

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A new theoretical interpretation is given of the stimulated electroluminescence emitted by ZnS: Mn films placed between totally reflecting and semitransparent mirrors. It is assumed that photons are accumulated in an intermediate mode which is characterized by a high Q factor. This makes it possible to achieve laser emission for a small value of the gain. The proposed interpretation removes several contradictions inherent in the earlier theory. All the electromagnetic modes of the multilayer system in question are calculated rigorously. The frequency and angular dependences are calculated for the intensities of the spontaneous and stimulated luminescence. The optical gain is computed. An estimate is given of the threshold value of the population inversion corresponding to the beginning of laser emission.

Electroluminescence of Mn-doped ZnS films has been investigated experimentally by various workers, and some of the results are reported in^[1,6]. A luminescent film was placed in^[4] between two metal mirrors, one of which was semitransparent. It was found that the electroluminescence band became narrower with increasing intensity of the excitation of the manganese impurities by electron impact; at the same time the intensity of this band increased by several orders of magnitude and the angular distribution of the intensity became narrower. These observations were attributed in^[4] to a population inversion and to laser emission in the resonator formed by the two mirrors covering the film. It was assumed that the light traveling across and not along the film was amplified. This assumption was supported by the following observations.

a) No light was emitted from the edges of the film.

b) A stimulated emission beam passing through the semitransparent mirror formed coaxial cones with their axes perpendicular to the film. Inside the dielectric film the vertex half-angles β of the cones satisfied the condition [see Eq. (3) in^[3]]

$$\frac{2nd}{\lambda}\cos\beta + \frac{1}{\pi}(\delta + \delta') = j, \qquad (1)$$

where λ is the wavelength of light in vacuum; n is the refractive index of the dielectric; d is the thickness of the dielectric film; j is an integer; δ and δ' are the phase shifts due to reflection of waves from the surfaces of the metal mirrors (this point is discussed later). The condition (1) was identical with the condition for the maximum of the intensity of the light transmitted by a Fabry-Perot interferometer. The angles β were in the range $0-24^{\circ}$ for different values of d and λ .

c) The threshold pumping level (the applied electric field) decreased with increasing d.

d) A monochromatic beam falling normally on the film subjected to an electric field (this field excited the manganese impurity ions) was amplified in the film and the gain deduced from this experiment was of the same order of magnitude as the gain required for the stimulated emission of light at right-angles to the film.^[4]

However, there were many important considerations which were in conflict with the proposed interpretation of the experimental observations. Since the reflection coefficient of the semitransparent mirror used in [3,4]

was 0.65, the laser action could take place in the 1- μ thick dielectric film only if the gain α was at least 3×10^3 cm⁻¹. However, this value of α was improbably high and incompatible with the absorption coefficient which was 13 $cm^{-1[7]}$ at the maximum of the absorption band corresponding to the luminescence band in question. This absorption was measured at approximately the same concentrations of Mn (of the order of 10^{21} cm⁻³) as in^{$\lfloor 4 \rfloor$} but at T = 77°K. The Franck-Condon emission and absorption transitions occurred in different ion configurations and could therefore be associated with different matrix elements. The populations of the initial energy levels could also be different, because of the differences between the vibrational frequencies in those cases when the electrons were in the ground and exciton states. For all these reasons and because of the difference between the temperatures at which the absorption and the luminescence measurements were carried out, the gain α could be different from the absorption coefficient at the band maximum. However, this difference should not amount to three orders of magnitude.

The gain $\alpha = 3 \times 10^3$ cm⁻¹ would correspond to a spontaneous emission power of the manganese impurities several orders of magnitude higher than the electric power supplied to the phosphor.¹⁾ Therefore, it became necessary to carry out a detailed theoretical analysis of the phenomena described above bearing in mind the special features of the geometrical configuration in which the dimensions of the optical resonator were comparable with the wavelength of light and the Q factor of the resonator was very low. Some of the results of this theoretical analysis are given below. We shall propose a different interpretation of the observed phenomena in which the stimulated emission begins at much lower values of α and which, therefore, removes all the contradictions mentioned above. Moreover, the new interpretation explains all the experimental observations.

We shall simplify quantization of the electromagnetic field by assuming that the system is conservative, i.e., we shall ignore the absorption of light in the dielectric and in the metal electrodes. We shall therefore assume that the permittivities ϵ'_m and ϵ_m of the two metals are real and negative so that the refractive indices of these metals are purely imaginary. This assumption describes quite well the real situation in the investigated range of frequencies.^[8] The half-space x < 0 is assumed to be filled by a metal, the dielectric occupies a layer defined

by 0 < x < d, the next layer d < x < d + a is the semitransparent metal, and the rest of the space d + a < x < L is occupied by vacuum (Fig. 1).

The permittivity $\epsilon(x)$ is thus a step-like function of x as shown in Fig. 1.

The electromagnetic field will be described by the vector potential

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{f}(x) \exp\{i\mathbf{k}_t \mathbf{r} - i\omega t\},\tag{2}$$

where \mathbf{k}_t is a two-dimensional wave vector lying in the yz plane. The potential **A** is defined by the equations

$$\Delta \mathbf{A} - c^{-2} \mathbf{\hat{e}} \mathbf{\hat{A}} = 0, \quad \text{div } \mathbf{A} = 0.$$
 (3)

The substitution of Eq. (2) in these equations gives

$$d^{2}f / dx^{2} + (\varepsilon \omega^{2} / c^{2} - k_{t}^{2})f = 0, \qquad (4)$$

$$f_t = ik_t^{-1}df_x / dx.$$
(5)

Here, and later the subscript t of a vector denotes the projection of this vector along the direction \mathbf{k} .

For given values of ω and \mathbf{k}_{t} Eqs. (4) and (5) have two mutually perpendicular solutions:

1) the solution $f_s(x)$, in which the vector f has the direction s perpendicular to the plane of incidence (the plane passing through 0x and k_t); we shall call this solution the s-polarized wave;

2) the other solution is the p-polarized wave for which the vector f lies in the plane of incidence and has the projections $f_x(x)$ and $f_t(x)$.

In the laser theory the space between the planes x = 0and x = d (Fig. 1) is usually regarded as the resonator. However, in the present case such a resonator would have a very low Q factor because of the small value of d and the generated modes would be strongly nonmonochromatic. Therefore, we shall assume that a third totally reflecting mirror is located in the plane x = L, where L is very large (Fig. 1). Thus, the resonator, or (more exactly) the field quantization region is assumed to be the space between the planes x = 0 and x = L.

The solutions of Eqs. (4) and (5) are known for all values of x subject to the continuity of the tangential projections of the electric and magnetic fields in the planes x = 0, x = d, x = d + a, and x = L. The corresponding solution of Eq. (3) can be represented by

$$\mathbf{A}_{\mathbf{v}}(\mathbf{r})e^{-i\omega t}.$$
 (6)

The serial number of the electromagnetic mode ν is a multicomponent index consisting of ω , \mathbf{k}_{t} , and the polarization index (s or p). The cyclic conditions with a large period L apply along the directions 0y and 0z. The modes are orthonormalized in the following manner:



$$\int_{0}^{L}\int_{0}^{L}\int_{0}^{L}\int_{0}^{L} (\mathbf{A}_{\mathbf{v}}^{*}, \mathbf{A}_{\mathbf{v}}^{*}) \varepsilon(x) dx dy dz = \frac{2\pi c^{2}\hbar}{\omega_{\mathbf{v}}} \delta_{\mathbf{v}\mathbf{v}^{*}}.$$
 (7)

We shall not give $\mathbf{A}_{\nu}(\mathbf{r})$ in full but we shall consider some of its properties and the relevant parameters which will be needed later. In the interval 0 < x < d the projections of $\mathbf{f}(x)$ are sinusoidal and their amplitudes $\mathbf{f}_{\mathbf{S}}(x)$ and $\mathbf{f}_{\mathbf{t}}(x)$ will be denoted by $\mathbf{B}_{\mathbf{S}}$ and $\mathbf{B}_{\mathbf{t}}$, respectively. In the interval d + a < x < L there are two types of projection $\mathbf{f}(x)$:

A) if $k_t^2 < \omega^2/c^2$, the functions $f_S(x)$ and $f_t(x)$ are also sinusoidal; the corresponding amplitudes are D_S and D_t ;

B) if $k_t^2 > \omega^2/c^2$, all the projections of f(x) decrease exponentially with increasing x in proportion to $\exp\left[-(k_t^2 - \omega^2/c^2)^{1/2}x\right]$; these solutions correspond to total internal reflection of light into the dielectric.

A function
$$F(\omega, \vartheta)$$
 is important in solutions of type A:

$$F(\omega, \vartheta) \equiv \begin{cases} |B_s/D_s|^2, & s\text{-polarization,} \\ B_l/D_l^2\cos^2 \vartheta, & p\text{-polarization.} \end{cases}$$
(8)

Here, ϑ is the angle between the 0x axis and the direction of the light beam in vacuum, defined by the relationships

$$|k_t| = c^{-1}\omega\sin\vartheta, \quad 0 < \vartheta < \pi/2.$$
(9)

For a fixed value of ϑ the function F varies periodically with ω and it consists of a series of equidistant very and high and narrow peaks. If ω is replaced by the dimensionless frequency w defined by

$$w = \frac{2d}{\lambda} \overline{\gamma}_{\varepsilon_0 - \sin^2 \vartheta} + \frac{2}{\pi} \delta, \quad \delta = \operatorname{arctg} \frac{n}{\overline{\gamma} - \varepsilon_m}$$
(10)

 $(n = \sqrt{\epsilon_0}, \epsilon_0 \text{ and } \epsilon_m \text{ are the permittivities of the dielectric and of the semitransparent metal mirror, as shown in Fig. 1), the period of the function F in respect of the argument of w is unity. The peaks of this function are 1 located close to integral values of w.$

We shall assume that $\epsilon^{-2\kappa a} \ll 1$, where $\kappa = 2\pi\lambda^{-1}\sqrt{-\epsilon_m}$ is the damping coefficient of a light wave in a nonabsorbing metal. In the specific systems studied experimentally we have $\exp(-2\kappa a) = 0.044$. In this case the value of $F(\omega, \vartheta)$ in the vicinity of a peak can be represented approximately by

$$F(\omega, \vartheta) = \frac{e^{2\kappa\omega}\cos\vartheta}{n} \frac{\Gamma/2\pi}{(\xi + \xi_0)^2 + \frac{4}{4}\Gamma^2},$$
 (11)

where

$$\xi = e^{2\kappa a} (w - j), \qquad (12)$$

$$\xi_{0} = \frac{\sin 2\delta}{\pi} \frac{1-Q^{2}}{1+Q^{2}}, \qquad \Gamma = \frac{4}{\pi} \frac{Q}{1+Q^{2}} \sin 2\delta,$$
(13)

$$Q = \frac{1}{\sqrt{-e_m}} \begin{cases} \cos \vartheta & s \text{-polarization,} \\ 1/\cos \vartheta & p \text{-polarization.} \end{cases}$$
(14)

In Eq. (12) the symbol j denotes a positive integer (the number of the peak). In the region of a peak the value of w is very close to j. The function (11) is Lorentzian in ξ or w and Γ is the half-width of the peak of the variable ξ . In terms of the argument of w, the half-width of the peak is

$$\Delta w = e^{-2\kappa a} \Gamma. \tag{15}$$

For a fixed value ϑ the function $F(\omega, \vartheta)$ can be represented as a series of nonequidistant peaks along the axis of ϑ . In terms of the argument of $\cos^2 \vartheta$ (for $\omega = \text{const}$), the half-width of a peak is

$$\Delta \cos^2 \vartheta = (\lambda / d) \sqrt{\epsilon_0 - \sin^2 \vartheta} \Delta w.$$
 (16)

It follows from Eq. (11) that the maximum of a peak is not located exactly at w = j, i.e., the maximum does not correspond to $\xi = 0$ but to $\xi = -\xi_0$. For the systems under consideration the half-width given by Eq. (15) is of the order of 0.01-0.02.

Between the peaks the function $F(\omega, \mathfrak{s})$ is of the order of

$$\frac{4Q^2\cos^2\delta}{1+Q^2}e^{-2\alpha a}\begin{cases} 1, & s-\text{polarization,}\\ \cos^2\vartheta, & p-\text{polarization,} \end{cases}$$

i.e., it is hundreds of times smaller than in the region of a peak.

We shall stop here our review of the properties of $\mathbf{A}_{\nu}(\mathbf{r})$ and $\mathbf{F}(\omega, \mathbf{s})$ and consider the physical consequences If the vector potential of the electromagnetic field $\mathbf{A}(\mathbf{r}, t)$ is expanded as a complete system of basis functions $\mathbf{A}_{\nu}(\mathbf{r})$ and the coefficients of the expansion are subjected to the usual quantization, it is found that the photons are not described by the usual exponential waves with an amplitude which is constant in space but by waves of the type given by Eq. (6). For type A waves, i.e., the waves which are not damped in vacuum, the amplitude in vacuum is, according to Eq. (7):

$$|D_{s,t}| = \sqrt{\frac{4\pi c^{2}\hbar}{L^{3}\omega}} \begin{cases} 1, & s\text{-polarization} \\ \cos \vartheta, & p\text{-polarization,} \end{cases}$$

i.e., it depends weakly on ω and ϑ and is almost the same for all the photons. On the other hand, it follows from Eq. (8) that the amplitude of the wave in the dielectric film

$$|B_{i,t}| = [4\pi c^2 \hbar L^{-s} \omega^{-1} F(\omega, \vartheta)]^{t/4}$$
(17)

is an abrupt peak-like function of ω and ϑ . For most of the values of ω and ϑ we have $|B_{s,t}| \ll |D_{s,t}|$, i.e., the photons corresponding to these values hardly penetrate into the dielectric, do not interact with the luminescence centers, and are not emitted from the dielectric. However, at selected values of ω and ϑ , when $w \approx j$, we have $|B_{s,t}| \gg |D_{s,t}|$. The corresponding photons now interact very strongly with the centers and the probability of their emission is much higher than in the case of an infinite dielectric.

The interaction of an impurity center with a few tens or hundreds of the surrounding ions is important in an optical transition. The linear dimensions of a complex formed by the impurity center and these ions are much smaller than the film thickness d. Therefore, the electron-vibrational wave function of such a complex is the same as in the case of a single center in an infinite dielectric. The ratio of the matrix elements of an optical transition in the system being considered to the matrix elements for the same transition in an infinite dielectric is equal to the ratio of the amplitudes $|B_{s,t}|$ in these two cases. This applies to any optical transition (dipole, quadrupole, magnetic dipole, etc.). Thus, we can obtain directly the relationship between the probabilities of optical transitions in these two cases.

Let us assume that in the case of an excited impurity center in an infinite dielectric the probability of emission of a photon per unit time in the frequency interval $d\omega$ is isotropic in direction and is the same for both polarizations. This probability, summed over all the directions of emergence of the photon, can be written in the form

$$dW = \tau^{-i} \varphi(\omega) d\omega. \tag{18}$$

If $\varphi(\boldsymbol{\omega})$ is normalized by the expression

$$\int \varphi(\omega) d\omega = 1, \qquad (19)$$

it is found that τ is the average lifetime of an excited center in an infinite dielectric under spontaneous emission conditions.

Bearing in mind the aforementioned relationship between the emission probabilities in the system being considered and in an infinite dielectric, we can show that in our system the probability of spontaneous emission of a photon of one of the polarizations per unit time in the frequency interval $d\omega$ and in the solid angle $d\Omega$ is

$$P(\omega, \vartheta) d\omega d\Omega = \frac{\varphi(\omega) F(\omega, \vartheta)}{4\pi\tau \sqrt{\varepsilon_0}} d\omega d\Omega.$$
 (20)

Here, d Ω is the solid angle of the directions of the rays in vacuum. The formula (20) shows that, in contrast to an infinite dielectric, the probability of emission in our system is strongly anisotropic, depends slightly on the polarization [Eqs. (13) and (14)], and has a completely different frequency dependence. The average lifetime of an excited state also differs from τ .

The maxima of the probability (20) coincide with the peaks of the function $F(\omega, \vartheta)$, i.e., they occur for $\omega \approx j$. This condition is exactly identical with Eq. (1) because $\sin \vartheta = n \sin \beta$ (in our calculations we have assumed that $\delta' = \delta$). This makes it possible to explain the experimental results obtained in^[3] for the spontaneous emission.

In the theoretical discussion of the stimulated emission it is necessary to determine the probability of emission of a photon per unit time and in a definite mode of index ν . This probability is

$$P_{\mathbf{v}} = \frac{\pi c \varphi(\omega)}{4 \gamma_{\varepsilon_{0} \tau} \hbar \omega} (n_{\mathbf{v}} + 1) \begin{cases} |B_{\mathbf{v} \varepsilon}|^{2} & s \text{-polarization,} \\ |B_{\mathbf{v}}|^{2} \cos^{-2} \beta, & p \text{-polarization.} \end{cases}$$
(21)

Here, n_{ν} is the number of photons emitted in the specified mode. If we ignore unity compared with n_{ν} , we obtain the logarithmic increment of the number of photons in this mode:

$$\frac{\dot{n}_{v}}{n_{v}} = \frac{\pi c \varphi(\omega)}{4 \sqrt{\epsilon_{u}} \tau \hbar \omega} NL^{2} d \begin{cases} |B_{v_{u}}|^{2}, & s \text{-polarization,} \\ |B_{v_{l}}|^{2} \cos^{-2}\beta, & p \text{-polarization.} \end{cases}$$
(22)

Here, N is the excess of the concentration of the excited impurity centers over the concentration of the unexcited centers.

For waves of type A it follows from Eq. (17) that the increment (22) should be proportional to 1/L, i.e., it should be negligibly small. Therefore, the photons cannot be accumulated in the mode ν in a finite time and the stimulated emission cannot be obtained. In the case of type B waves (the waves which are damped in vacuum), it follows from Eq. (7) that

$$|B_{s,t}^{0}|^{2} \approx \frac{4\pi c^{2}\hbar}{L^{2}\omega\varepsilon_{0}d} \begin{cases} 1, & s-\text{polarization,} \\ \cos^{2}\beta, & p-\text{polarization.} \end{cases}$$
(23)

In this case the increment (22) is independent of L, i.e., it is finite and the accumulation of photons in the specified mode is possible. The quantities referring to the type B mode will be denoted by the index 0. In contrast to the type A modes, the values of ω and ϑ cannot be specified independently for the B modes: these quantities are related by the dispersion law

$$w(\omega, \vartheta) = j, \quad j = 1, 2, 3, \ldots$$

Here, we still have Eq. (10) for w but j should be so

small that $\sin^2 \vartheta$ should be greater than unity. The reflection coefficient of these waves from the edge of the dielectric film is very close to unity.^[9] This gives rise to a high Q factor of the mode and explains why the emission is not observed from the film edges. Thus, in the case of a type B mode the conditions for the accumulation of photons are very favorable and this makes it possible to achieve laser action for relatively low values of the gain.

The observed radiation emitted by the phosphor across the semitransparent mirror obviously consists of type A and not type B modes. Therefore, we must assume that photons generated by stimulated emission in a type B mode ν_0 are transformed, by intermode transitions, into type A modes of index ν . These intermode transitions may occur because of the presence of defects and optical inhomogeneities in the dielectric film, roughness and surface relief of the metal mirrors, etc. All such defects can be included in the analysis by assuming that the permittivity of the dielectric is a function of the coordinates. The function can be represented by ϵ_0 $+ \epsilon_1(\mathbf{r})$. Having identified the additional terms which appear in the energy operator of the electromagnetic field because of introduction of the term $\epsilon_1(\mathbf{r})$, we can regard them as a perturbation and the cause of the intermode transitions. The value of ω does not change in these transitions.

Our calculations indicated that the probability (per unit time) of a transition of a photon from a mode ν_0 to one of the type A modes ν in the case of a beam confined within the solid angle d Ω in vacuum is given by

$$P_{\mathbf{v}\mathbf{v}_0}d\Omega = \frac{\omega^{\mathbf{s}} n_{\mathbf{v}_0} L^3}{32\pi^{\mathbf{s}} \hbar^2 c^{\mathbf{v}}} |\varepsilon_{\mathbf{i}\mathbf{v}\mathbf{y}_0}|^2 d\Omega,$$
(24)

where

$$\varepsilon_{ivv_0} = \int (\mathbf{A}_{v}^*, \mathbf{A}_{v_0}) \varepsilon_i(\mathbf{r}) dx dy dz, \ \left| \varepsilon_{ivv_0} \right|^2 = G_{vv_0}(\omega, \vartheta) F(\omega, \vartheta).$$
(25)

Here, the form of the function $G_{\nu\nu_0}(\omega, \mathfrak{s})$ depends on $\epsilon_1(\mathbf{r})$ but in all cases $G_{\nu\nu_0}$ is a smooth function compared with $F(\omega, \mathfrak{s})$. Therefore, the maxima of the photon emission probability in vacuum given by Eq. (24) coincide with the maxima of the function $F(\omega, \mathfrak{s})$ i.e., they occur for $w \approx j$. This explains the relationship (1) which now applies to the stimulated emission.

The laser emission threshold and the threshold value of the gain can be found if we know the total probability of transition of a photon from a mode v_0 to any of the type A modes. This probability can be found by calculating the integral of the probability (24) with respect to $d\Omega$. It is assumed that when ϑ increases from zero to $\pi/2$ the function $F(\omega, \vartheta)$ has only one peak. In estimating the permittivity of Eq. (25) it is assumed that $\epsilon_1(\mathbf{r})$ does not vanish in isolated nonoverlapping defects whose concentration is N_d and that the positions of these defects are completely random and not correlated at all with the periods of the functions $\mathbf{A}_{\nu}(\mathbf{r})$ and $\mathbf{A}_{\nu}(\mathbf{r})$. If $\epsilon_1(\mathbf{r})$ in the bulk of the dielectric film is approximated by various simple functions such as $\epsilon_1^0 \sin gx \sin gy \sin gz$, a Π -like function, etc., the values of $\epsilon_{1\nu\nu_0}$ obtained in this way are similar.

Estimates of this kind yield the following equation for the increment in the number of photons in the working mode ν_0 :

$$\dot{n}_{v_0} = n_{v_0} L^2 d |B^{\circ}_{s_0, t_0}|^2 \left[\frac{\pi c N \varphi(\omega)}{4 \sqrt{\tilde{e_0}} \tau \hbar \omega} \Xi - \frac{\omega^5 \varepsilon_1^{\circ 2} \lambda N_d}{8 \pi^2 \hbar c^5 \chi^6 d} \right],$$
(26)

where $\Xi = 1$ for an s mode and $\Xi = \cos^{-2} \beta_0$ for a p mode. Here, the first term of the right-hand side follows from Eq. (21), the second term represents the loss of photons from the working mode because of intermode transitions, and χ is the greater of the two quantities $\sqrt{\epsilon_0} \omega/c$ and g.

The laser action begins when the quantity defined in Eq. (26) becomes positive, i.e., when

$$\frac{\varphi(\omega)}{\tau} \Xi > \frac{\omega^{6} \overline{\gamma} \overline{\epsilon_{0}} \epsilon_{1}^{02} \lambda}{2\pi^{3} c^{6} \chi^{6} d} \frac{N_{d}}{N}.$$
(27)

This criterion is most stringent in the case when $\chi \approx \sqrt{\epsilon_0} \omega/c$. In this case Eq. (27) reduces to

$$\frac{\varphi(\omega)}{\tau} \Xi > \frac{\varepsilon_1^{o_2} N_d \lambda}{2\pi^3 \varepsilon_0^{s/2} dN}.$$
(28)

In the case of our ZnS: Mn phosphor we find that the maximum value of φ is $1.6 \times 10^{-15} \text{ sec.}^{\lfloor 10-12 \rfloor}$ For an infinite dielectric we have $\tau = 2 \times 10^{-4} \text{ sec.}^{\lfloor 13 \rfloor}$ If we assume, with some exaggeration, that $\epsilon_1^{\text{CN}} N_{\text{d}} \sim 10^{12} \text{ cm}^{-3}$, $\epsilon_0 = 5.5$, $\lambda = 5850$ Å, $d = 10^{-4}$ cm, we find that the inequality (28) begins to be satisfied from population inversions N $\sim 10^{19}$ cm⁻³. However, we must bear in mind that the stringency of the criterion (27) has been overestimated twice in an arbitrary manner. Therefore, in practice the laser emission may begin at population inversions N much lower than the concentration of impurity ions (10^{21} cm⁻³).

The optical gain, governed only by the first term on the right-hand side of Eq. (26), is

$$\alpha = N\lambda^2 \varphi(\omega) / 4\epsilon_0 \tau. \tag{29}$$

If we substitute $N = 10^{19} \text{ cm}^{-3}$ in Eq. (29), we find that $\alpha = 0.012 \text{ cm}^{-1}$. Thus, in the proposed laser emission mechanism, when photons are accumulated in an intermediate mode, the laser action may begin at a value of α which is five orders of magnitude smaller than that suggested in^[4]. This removes the contradiction between the spontaneous luminescence power and electric power supplied to the phosphor.

In the maximum inversion limit we have $N = 10^{21} \text{ cm}^{-3}$ and $\alpha = 1.2 \text{ cm}^{-1}$. In contrast to^[4], this value of α is not in conflict with the absorption coefficient of light in an excited dielectric.

We mentioned at the beginning that one of the experimentally established observations was the decrease in the threshold pumping level with increasing thickness d. It follows from Eqs. (26) and (27) that the threshold inversion N and the thickness d should be inversely proportional. However, it is difficult to say whether this inverse proportionality can explain qualitatively the experimental observations.

The right-hand side of the rate equation (26) should be supplemented by a term which makes allowance for other (so far ignored) forms of the loss of the photons from the working mode such as the absorption in the metal mirrors. This absorption may be considerable but it is much weaker than in the case of type A modes. Consequently, the threshold values of N and α may increase.

Equation (26) should apply to all the type B modes. Under steady-state conditions the right-hand side of all such equations should be negative or zero. This means that at one or several frequencies corresponding to the maximum value of terms in the square brackets, the sum of these terms should be zero. This is ensured by the steady-state value of N which is independent of the pumping level.²⁾ At other values of the frequency the sum of the terms in the square brackets in Eq. (26) is negative. Consequently, in this case $n_{\nu_0} = 0$. Thus, stimulated emission of monochromatic photons occurs in the intermediate mode. Their subsequent intermode transitions and escape into vacuum occur without any further change in the frequency. This explains the spectral narrowing of the stimulated electroluminescence found experimentally.^[4]

The angular distribution of the stimulated luminescence is governed by the formulas (24) and (25), i.e., it is governed mainly by the factor $F(\omega, \vartheta)$. Consequently, this distribution is independent of the pumping level. The experimental results on the phosphor under consideration indicated that the lobes of the polar distribution of the intensity become narrower with increasing pumping level although the narrowing is considerably less than for the conventional crystal lasers. The explanation of this observation and of the experiments involving injection of light from outside will be given in a separate paper. needed and this calculation must allow for the departure from thermal equilibrium in the population of the vibrational levels of the excited impurity centers.

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Note added in proof (December 1, 1972). A similar theory can easily be developed for the superluminescence of thin anthracene films-see M. D. Galanin, Sh. D. Khan-Magometova, Z. A. Chizhikova, ZhETF Pis. Red. 16, 141 (1972) [JETP Lett. 16, 97 (1972)].

¹⁾Our attention to this point was drawn by Yu. M. Popov. ²⁾At high stimulated emission intensities a more rigorous calculation is