AN OPTICAL RESONATOR FOR A LASER WITH A LIQUID ACTIVE SUBSTANCE

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A theoretical discussion is given of the principal problems involved in the construction of lasers in which the active substances are solutions of complex compounds of rare-earth elements. A concrete design of the optical resonator is proposed, and the oscillatory modes arising in it are investigated. The effect of inhomogeneity arising in the liquid due to intense optical pumping on the operation of the resonator is also investigated.

AT the present time solutions of europium chelates are used as the active substance in liquid lasers.[1] These solutions have an especially intense absorption in the ultraviolet region, where the pumping is carried out ($\epsilon \approx 6 \times 10^4$ liter/mole cm).

The liquid state of the active substance requires a cell that is transparent in the pumping region (quartz, sapphire) and provision for the possibility of expansion of the liquid due to heating during operation.

The intensity of absorption of a liquid is related to the concentration of the solution of the active substance. Estimates made on the basis of the generation condition[2] show that the concentration of active molecules in the solution must be 5 to 6 $\times 10^{18}$ molecule/cm^3 (10^{-2} mole/liter). At such a concentration the absorption in the middle of the pump band is 600 cm^{-1}. This means that the intensity of pump radiation is already down by a factor of e at a depth $\delta \approx 0.02$ mm. Thus, in cells of significantly greater dimensions than the depth of penetration of this radiation into the liquid, the intense absorption in the middle of the pump band precludes effective pumping in the deeper layers of the liquid. There the pumping will occur only in the wings of the absorption band, where the intensity of absorption falls sharply. At the same time, the greater heating of the surface layers of the liquid can lead to the appearance of inhomogeneities in the liquid, lowering the quality of the optical resonator. Thus, intense absorption in the middle of the pump band should be thought of as a negative effect, rather than a positive one.

Another situation is created in thin cells with a thickness comparable to δ. Here intense absorption is a helpful effect. This can be understood qualitatively from the following considerations. In a nearly ideal illuminating system, the flow of pump energy through a unit surface of the sample is πB_ω, where B_ω is the energetic spectral brightness of the pumping source. (The pumping source is assumed to obey Lambert's law.) This energy is absorbed in a layer of thickness $\sim \delta$. Therefore the density of pumping energy (i.e., the energy per unit volume) in the top surface layers is $\pi B_\omega/\delta$. Thus, for constant source brightness, the smaller δ, the larger is the density of pump energy. Since in complex compounds δ is a hundred times less than, for example, in ruby, the density of pumping energy is correspondingly larger.

On the other hand, in cells of thickness comparable to δ, the dispersion of energy over the volume will be rather uniform and so the disturbance of optical homogeneity will not be too strong.

However, one may think that a reduction in the transverse dimensions of the cell will lead to an increase in diffraction losses.

In order to eliminate the increase in diffraction losses, i.e., to keep the radiation in the liquid, it is proposed to use reflection from a liquid-dielectric interface (refractive indices respectively n_1 and n_2, $n_1 < n_2$). Actually, we are interested in the rays propagating along the liquid layer. Such rays are incident on the liquid-dielectric boundary at an angle close to $\pi/2$. It is known that at such angles of incidence the reflection coefficient tends toward unity.[3]

These considerations lead to the design of the cell shown in Fig. 1. A dielectric transparent in the ultraviolet region (quartz, sapphire) permits pumping of the liquid. The liquid, which does not completely fill the cell, has room to expand upwards when it heats up.

A characteristic feature of this resonator is the presence of "internal" mirrors, i.e., mirrors in contact with the liquid. This arrangement of the mirrors eliminates the outflow of radiation from

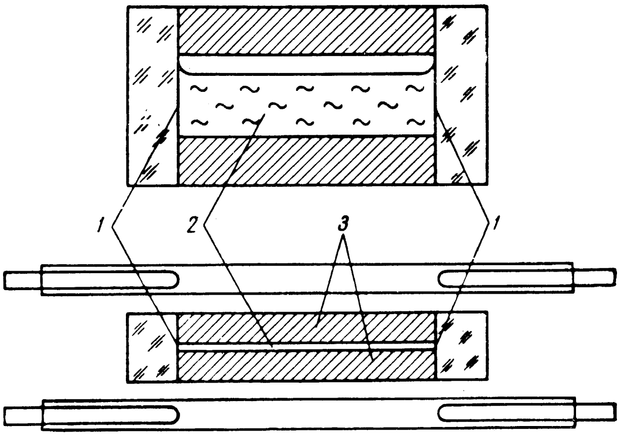

FIG. 1. Diagram of a resonator for a laser with a liquid active substance: 1 – mirror, 2 – liquid, 3 – dielectric.

the generating layer and the reverse influx of it into this layer together with the light losses that accompany both these processes.

Since they are outside the scope of the present paper, we shall leave aside certain constructional questions, e.g., the preparation of the mirrors exposed to contact with the liquid and condensation in the cell.

The oscillations in such resonators are most simply treated from the viewpoint of geometrical optics, supplemented by quantum conditions.[4] The natural frequencies of this resonator are determined by the relation

$$\omega = \pi c\left(\frac{p^2}{l^2}+\frac{q^2}{d^2}\right)^{1/2}, \tag{1}$$

where c is the velocity of light in the liquid, l and d are respectively the length and thickness of the resonator, and p and q are integers characterizing the longitudinal and transverse oscillations, whereby p is a large number and q = 1, 2, 3, ...

Besides frequency, the oscillations are characterized by the angle which the ray makes with the resonator axis. This angle

$$\alpha_q \cong \lambda q / 2n_l d \tag{2}$$

depends on q (λ is the wavelength).

There are three forms of loss in the resonator: radiation into the dielectric, absorption of light in the liquid, and absorption in the mirrors. Correspondingly, we can introduce three Q factors, which are equal to

$$Q_1 = 2\pi n_l{}^3 d^3\sqrt{n_0^2-1} / k^2\lambda^3, \tag{3}$$

where n_0 is the refractive index of the dielectric referred to that of the liquid,

$$Q_2 = 2\pi L n_l / \lambda, \tag{4}$$

where L is the thickness of the liquid layer in which the light intensity at the working wavelength falls by a factor e (L ~ 2 to 3 × 10^2 cm),

$$Q_3 = 2\pi n_l l / \lambda(1-R), \tag{5}$$

where R is the coefficient of reflection from the mirrors. Substitution of numerical values into these formulas shows that all three Q's have approximately the same value. This means that the proposed resonator will store light rather effectively.

As the liquid is irradiated with light from the pump, it will become inhomogeneous, hence the treatment presented above is valid only at the initial moment of the pump pulse.

Since the intensity of the pumping light changes significantly in depths of the order of the thickness of the cell, the liquid layer next to the dielectric will heat up more strongly than layers in the center of the cell. Since the coefficient of refraction of a liquid at constant pressure decreases with increasing temperature, the distribution of the refractive index at some moment of the pump pulse will be as shown in Fig. 2. It is easy to see that such a medium possesses focusing properties. The rays in it will not be propagated in straight lines, but along curves, not escaping, however, outside of the two bounding planes, which serve as caustic surfaces.

Calculations show that for temperature drops of 5–10°C in the liquid several transverse modes, together with the caustics, will lie entirely within the liquid. For less inhomogeneity, the caustics do not form, even though the rays may be bent. In Fig. 2 this case is shown by dashes. It is as if the caustics were located outside the liquid.

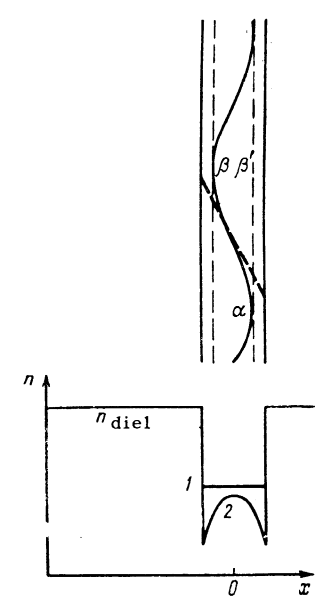

FIG. 2. Path of the rays and distribution of the refractive index in the resonator: 1 – without pumping, 2 – with pumping.

If the pump is pulsed, then at the beginning of a pulse the liquid is homogeneous and the natural frequencies of the resonator are as considered above. Thereafter the homogeneity of the liquid is destroyed somewhat. The rays are bent, but the caustics have not yet formed. It is easy to see that in this case the rays are incident on the liquid-dielectric interface at more glancing angles than in the case of a homogeneous liquid. In addition the difference between the refractive indices of the liquid and dielectric is somewhat increased. Both factors lead to an increase in the coefficient of reflection from the liquid-dielectric boundary, and consequently to an increase in Q.

When the liquid inhomogeneity grows to such an extent that the caustics form, the loss of light energy at the liquid-dielectric boundary can in general be neglected. Other losses will play the main role. Thus the calculated Q is the minimum one.

This treatment permits us to draw several conclusions relative to the design of a liquid laser using a solution of complex compounds:

a) The laser resonator should consist of a thin layer of the active liquid bounded by dielectric plates that are transparent in the pumping region.

b) The presence of liquid-dielectric boundaries enables the effective storage of light in the working volume.

c) The destruction of the homogeneity of the liquid in thin layers, in the first place, is not so great as in wide cells, hence the dispersion of pump power in a thin cell occurs approximately over the entire volume; in the second place, as a consequence of the difficulty of mixing up a liquid in a thin layer, the disturbance of homogeneity takes place in a regular fashion, and not randomly; finally, in the third place, this disturbance is of such character as to increase the Q of the resonator.

d) When thin layers are used, intense absorption in the middle of the pumping region becomes a favorable factor, since the pumping occurs in a wide spectral interval, and not only in the "wings" of the absorption band. But the main thing is that the density of pumping is increased. Compared to the ruby laser, the pumping density is increased a hundred-fold, hence one can hope to obtain laser action by illuminating a liquid with a continuous source. In conjunction with the possibility of cooling the liquid by means of pumping it through the working volume, this circumstance will make it possible to build a cw liquid laser.

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