MANIFESTATION OF ELECTRON-PHONON INTERACTION IN THE LASING OF A CRYSTAL WITH Nd $^{3+}$ 

- D.N. Vylegzhanin and A.A. Kaminskii Institute of Crystallography, USSR Academy of Sciences Submitted 17 April 1970; resubmitted 15 May 1970 ZhETF Pis. Red. <u>11</u>, No. 12, 569 - 573 (20 June 1970)
- 1. The increase of the generation energy  $(E_{\rm g})$  with increasing temperature (T) has been observed in lasers based on many crystals activated with Nd³+ ions, at excitation energies  $(E_{\rm exc})$  greatly exceeding the threshold energy  $E_{\rm thr}$ ). This effect is attributed to activation of the processes of electron-phonon interaction, which are manifest in the acceleration of the decay of the final state  $^4I_{11/2}$  for the stimulated emission (SE). We present below a brief description of the effect and its interpretation with  $Y_3Al_5O_{12}:Nd³+$  as an example.
- 2. In the analysis of the functional characteristics of lasers based on media with Nd³+ ions with the aid of the kinetic equations, it is usually assumed for the purpose of simplifying the analysis that the probabilities of nonradiative transitions  ${}^{t}F_{5/2}$ ,  ${}^{2}H_{9/2} \rightarrow {}^{t}F_{3/2}$  and  ${}^{t}I_{11/2} \rightarrow {}^{t}I_{9/2}$  greatly exceed  $\sigma_e^pI$  (here  $\sigma_e^p$  is the peak value of the cross section of the transition, and I is the radiation density at the generation wavelength  $\lambda_g$ ). This is the condition for maintaining the necessary inversion of the working levels. An investigation of the SE parameters in the interval 77 1000°K shows that such assumptions are valid for the practically attainable  $E_{\rm exc}$  in neodymium lasers only in a region of T lying usually above 300°K. For lower T, at  $E_{\rm exc}$  greatly exceeding  $E_{\rm thr}$  as well as in certain special generation regimes, the SE intensity can increase to such an extent that the rate of deactivation of the state  ${}^{t}F_{3/2}$ , which is determined mainly by the value of  $\sigma_e^pI$ , becomes commensurate with the probability  $w_{21}$  if the nonradiative transition  ${}^{t}I_{11/2} \rightarrow {}^{t}I_{9/2}$ , and  $E_g$  will be limited by the narrowness of the relaxation channel  ${}^{t}I_{11/2} \rightarrow {}^{t}I_{9/2}$ .
- 3. The experiments were performed with  $Y_3Al_5O_{12}:Nd^{3+}$  crystals ( $^{\circ}O.3$  wt.%, l=30 mm, diameter 5.5 mm) using the customary experimental technique. The main investigated characteristic was the parameter  $E_g$  and its dependence on  $n=E_{\rm exc}/E_{\rm thr}$  at different T (see Figs. 1 and 2). In the experiments we took into account the following: the exact dependence  $E_{\rm thr}(T)$ , the influence of the effect of "ultraviolet turning-off of the SE" [1], the excitation regime, and many other conditions. The most interesting facts that follow from the foregoing results are the increase of  $E_g$  with increasing T at fixed n, and also the deviation of the dependences of  $E_g(n)$  at  $n\stackrel{>}{\sim} 30$  from linearity and their tendency to saturation at large n (>100).
- 4. The obtained regularities allow us to assume that the effect of saturation is connected with the narrowness of the channel of the nonradiative relaxation  ${}^4I_{11/2} \rightarrow {}^4I_{9/2}$ , and that the temperature increase of the generation energy is due to the increase of  $w_{21}$ . In this case the analysis of the experimental data makes it possible to estimate the course of the  $w_{21}(T)$  dependence.

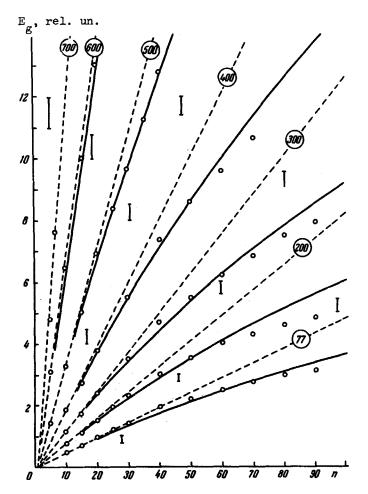


Fig. 1. Plots of  $E_g(n)$ ; the circles indicate the temperature in degrees K.

From the solution of the kinetic equations in the stationary approximation we find that  $w_{21}$  and the limiting value of  $E_g$  at saturation ( $E_g^{\rm sat}$ ) are connected by the relation

$$w_{21} = \frac{E_{g}^{sat}}{Nh \nu_{g} \kappa f_{g}} \left(1 + \frac{b_{4}}{b_{2}}\right) + \frac{1}{r_{lum}} \frac{b_{4}}{b_{2}} ,$$

where N is the number of Nd³+ ions,  $\kappa$  is the ratio of the output loss to the total loss, b =  $\sum_{j}(N_{ij}/N_{i1})$ ,  $N_{ij}$  is the population of the j-th Stark component, i is the number of the multiplier (i = 2 for  $^4I_{11/2}$  and i = 4 for  $^4F_{3/2}$ ),  $\tau_{lum}$  is the lifetime relative to radiative transitions  $^4F_{3/2} \rightarrow ^4I_{11/2-15/2}$ , tg is the duration of the SE, and hvg is the photon energy at  $\lambda_g$ . The value of  $E_g^{sat}$  was determined in the following manner. It can be shown that

$$E_{g}(n,T) = \operatorname{tg} \beta_{o} \frac{n-1}{1+n\sigma} , \qquad (1)$$

where  $\tan \beta_0 = E_g(n + n')/(n' - 1)$ at n'a << 1, and <u>a</u> is the saturation parameter, which can be determined by considering at different values of T the course of the function

$$\frac{E_{g}(n)}{E_{g}(n')} = \frac{E_{g}}{E'_{g}} = \frac{n-1}{n'-1} \frac{1}{1+(n-n')a}.$$

In our case we obtained for <u>a</u> the following approximate relation: a =  $(3.5 \pm 1.0) \times 10^{-4} \, \mathrm{T}^{1/2}$ . From (1) we obtain the relation  $\mathrm{E}_{\mathrm{g}}^{\mathrm{sat}} = \mathrm{E}_{\mathrm{g}}^{\mathrm{i}}/\mathrm{a(n'-1)}$ , making it possible to determine  $\mathrm{E}_{\mathrm{g}}^{\mathrm{sat}}$  from the value of  $\mathrm{E}_{\mathrm{g}}$  at small n, which are perfectly attainable in experiment even at relatively high T (in our case up to  $\sim 700^{\circ}\mathrm{K}$ ).

5. The  $w_{21}(T)$  dependence obtained on the basis of the foregoing experimental results is shown in Fig. 3. This dependence is satisfactorily approximated by the expression [2]

$$w_{21}(T) = 7 \cdot 10^4 \left( \frac{\exp\left[ (400 \text{ cm}^{-1}) h c / k T \right]}{\exp\left[ (400 \text{ cm}^{-1}) h c / k T \right] - 1} \right)^3 \left( \frac{1 + \exp\left[ - (23 \text{ cm}^{-1}) h c / k T \right]}{b_2} \right),$$

which corresponds to nonradiative decay of the state 4I11/2, occurring via

emission of three phonons with energy of approximately 400 cm $^{-1}$  in transitions from its lower two levels (2002 and 2025 cm $^{-1}$ ) to the upper component ( $^{8}50$  cm $^{-1}$ ) of the  $^{4}I_{9/2}$  multiplet.

The described method of estimating the absolute value of  $w_{21}$  is quite indirect and therefore cannot claim high accuracy. In particular, the possible value of  $w_{21}(0)$ , in our opinion, should be limited to the interval  $7 \times 10^{(4\pm1)}$  sec<sup>-1</sup>. The relative change of  $w_{21}(T)$  is determined by the dependence of  $E_g(n')$  on the

temperature and is measured with higher accuracy. The low value of  $w_{2\,1}$  offers evidence of the weakness of the electron-phonon interaction for the ions  $\mathrm{Nd}^{3\,+}$  in  $\mathrm{Y}_3\mathrm{Al}_5\mathrm{O}_{1\,2}$ , and agrees with such known facts as the relatively small temperature broadening of the lines and the weak manifestation of the electron-vibrational structure in the optical spectra of  $\mathrm{Nd}^{3\,+}$ . We note that

in accordance with the data of [3], the phonon spectrum of  $Y_3Al_5O_{12}$  extends to ~860 cm<sup>-1</sup>. An analysis of the modes of the lattice oscillations, carried out in [3], shows that the part of this spectrum lying above ~620 cm<sup>-1</sup> belongs to the complex  $(AlO_4)^5$ , whereas the oscillation frequencies in which the Y <sup>3+</sup> ion takes part range up to ~580 cm<sup>-1</sup>. Bearing this in mind, the small value of the "effective" Debye phonon, which follows from our analysis, can be attributed to the fact that the Nd <sup>3+</sup> ions, which replace Y <sup>3+</sup>, interact weakly with the oscillations of the complex  $(AlO_4)^5$ . The "effective" phonon obtained by us agrees well with the data of [4], in which  $T_D$ , determined from the temperature shift of the Nd <sup>3+</sup> lines in  $Y_3Al_5O_{12}$ , turns out to be equal to ~600°K.

6. From the point of view of applications, the observed phenomenon also seems quite important to us, since the temperature increase of  $w_{21}$  makes it possible to increase  $E_g$  at a specified excitation power ( $P_{\rm exc}$ ) by choosing the optimal T ( $T_{\rm opt}$ ). Thus,  $T_{\rm opt} \simeq 550^{\rm o}{\rm K}$  for  $P_{\rm exc} \simeq 300$  kW.

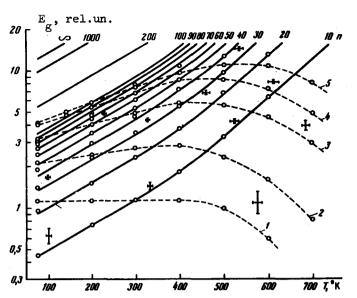


Fig. 2. Plots of  $E_g(T)$  at different n (solid lines) and at the following excitation powers (dashed lines): 1-35, 2-75, 3-150, 4-220, 5-300 kW.

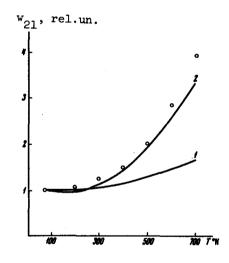


Fig. 3. Plots of  $w_{21}(T)$ : 1 and 2 - theoretical curves for two- and three-phonon processes and relaxation, respectively.

As seen from Fig. 2 (curve 5), in this case the high-temperature regime yields a  $\sqrt{30\%}$  increase of laser efficiency compared with T =  $300^{\circ}$ K. Under other SE conditions, especially in the Q-switched mode, it is possible to obtain an even greater efficiency increase by choosing  $T_{\rm opt}$ . The data of the foregoing in-

vestigations show that the existence of a narrow phonon channel can be one of the causes of the low efficiency of "low-temperature" lasers based on media with  ${\rm Nd}^{3+}$  ions.

A.A. Kaminskii, Phys. Stat. Sol. (a)  $\underline{1}$ , 573 (1970). L.A. Riseberg and H.W. Moos, Phys. Rev.  $\underline{174}$ , 429 (1968).

G.A. Slaek, D.W. Oliver, R.M. Chrenko, ans S. Roberts, Phys. Rev. 177, [3] 1308 (1969).

[4] T. Kushida, Phys. Rev. <u>185</u>, 500 (1969).

INTERACTION OF LASER RADIATION WITH YTTRIUM IRON GARNET IN PARAMETRIC EXCITA-TION OF SPIN WAVES

I.A. Deryugin, V.I. Mykityuk, A.A. Solomko, and V.N. Radchik Kiev State University Submitted 21 April 1970 ZhETF Pis. Red. 11, No. 12, 573 - 576 (20 June 1970)

We report the results of an investigation of spin waves excited parametrically by parallel pumping in an yttrium iron garnet using laser radiation ( $\lambda = 1.15 \mu$ ).

It is known that in alternating fields h  $\parallel$  H $_{0}$  exceeding the critical value, the longitudinal magnetization decreases by an amount  $\Delta M_{_{\rm Z}}$  and an alternating component of the magnetization appears, having double the pump frequency [1]. In our experiments we investigated the rotation change, proportional to  $\Delta M_{7}$ , of the plane of polarization of laser radiation by the Faraday effect.

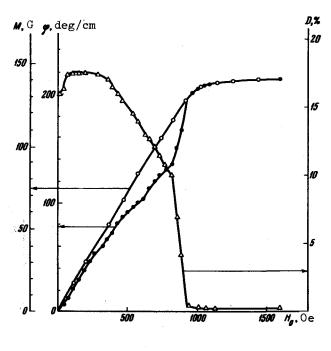


Fig. 1

The sample was a disc of 4.1 mm diameter and 1.6 mm thickness and was placed in the antinode of the magnetic field of a resonator operating in the  $H_{102}$  mode ( $v_H = 9470 \text{ MHz}$ ). A constant field Ho | h was applied along the disc axis. The laser beam propagated parallel to the applied external field. We measured in the experiment the magnetization-induced rotation of the plane of polarization of the laser beam, averaged over a **ce**ntral area of the sample with dimensions  $0.4 \times 0.7$  mm, and determined by the dimensions of the photosensitive area of the receiver.

In the absence of pumping, we measured the dependence of the rotation of the plane of polarization of the laser beam  $\phi$  and its depolarization D on the constant field (Fig. 1). We note that the  $\phi(H_0)$  dependence does not coincide with the results obtained in [2], or with the results of our measurements of the magnetization of the sample  $M(H_0)$ with the magnetometer (Fig. 1). constant fields weaker than the satura-

tion fields, two regions are seen on the  $\phi(\mathsf{H}_{\mathsf{0}})$  plot, characterized by different slopes of the  $\phi(H_0)$  curve. This is also clearly seen on the plot of the depolarization vs. the constant field. Such a  $\phi(H_0)$  plot can be attributed to the fact that the internal field in the sample is inhomogeneous (the sample is a disc).

The results of the investigation of the decrease of the rotation of the plane of polarization  $\phi_s(H_0)$  of the laser beam, determined by the value of  $\Delta M_{\pi}$ ,