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SPECTRUM OF LASER BASED ON ELECTRON-VIBRATIONAL TRANSITIONS IN A $CaF_2:Sm^{2+}$ *CRYSTAL*

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Results are reported of an experimental investigation of the temperature dependence (T = 77—300°K) of the luminescence spectrum of a $CaF_2:Sm^{2+}$ crystal and of the absorption coefficient at the characteristic frequencies. More detailed information is presented on the structure of the generation spectrum of a laser based on electron-vibrational transitions, which shifts discretely by 700 cm^{-1} when the temperature of the $CaF_2:Sm^{2+}$ crystal is changed.

1. INTRODUCTION

THE development of lasers using $CaF_2:Sm^{2+}$ crystals has stimulated intense investigations of the optical spectrum of the latter, as a result of which the electronic energy-level scheme has been constructed[1-5] (Fig. 1). At low temperatures, ~20°K, the spectrum contains only one narrow intense 14,118 cm^{-1} luminescence line, at which a laser based on the four-level scheme operates. Raising the temperature influences strongly the luminescence spectrum of the crystal. The broadening of the 14,118 cm^{-1} line and the decrease of its integral intensity are accompanied by an increase of the integral intensity of the electron-vibrational transitions, followed by a temperature quenching of the luminescence[6]. The use of a ruby laser as a pump source has made it possible to broaden greatly the region of operating temperatures and to attain generation at electron-vibrational transitions[7,8].

In the present article we report the results of an investigation of the mechanism whereby a ruby laser excited luminescence in a $CaF_2:Sm^{2+}$ crystal, and of the temperature dependence of the absorption coefficient at several characteristic frequencies. We investigate in detail the contour of the electron-vibrational luminescence band in the temperature interval 77—300°K. More detailed information is presented on the generation spectrum at electron-vibrational transitions, which shifts discretely towards the long-wave side by 700 cm^{-1} with increasing temperature.

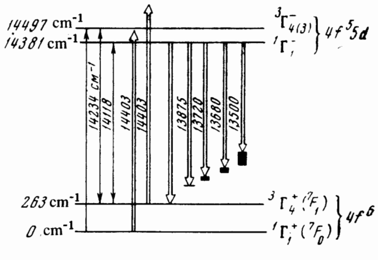

FIG. 1. Energy level scheme of $CaF_2:Sm^{2+}$ crystal. The thin lines denote electronic transitions, the double lines denote excitation with the ruby laser and generation.

2. CRYSTALS

The main measurements were made on samples grown by the Stockbarger method in graphite crucibles in a fluoriding atmosphere[9]. The required amount of SmF_3 was introduced into a charge of natural fluorite, and CeF_3 was added (see the table). Eight single crystals were grown simultaneously. A polariscope was used to locate in the obtained raw crystals (120 mm long) sections with uniform coloring, from which laser samples of cylindrical form (8 mm diameter, 20 mm long) were prepared. The lateral surfaces were polished. The end faces of 32 samples were polished simultaneously at one setting in the mount. The end surfaces were checked for plane-parallelism with a goniometer, accurate to 5″. The flatness of the end faces, determined with the aid of cover glasses, was 0.1 λ. The optical volume homogeneity of the samples was monitored by interference of parallel beams of light from a helium-neon laser, reflected from the end faces. The interference pattern consisted usually of one or two fringes of irregular form. Only one sample gave an even field. It can be stated that an increase of the impurity concentration leads to a larger distortion of the optical homogeneity. The samples were not retouched. Small-angle scattering was monitored on two batches of single crystals. For most samples, the equivalent scattering coefficient was 0.03 cm^{-1}.

After performing the aforementioned investigations, the samples were exposed to an x-ray dose of 10^8 r. The colorless samples were colored green as a result of the conversion of the Sm^{3+} into Sm^{2+}[10]. From the oscillator strengths of the 14,497 and 14,118 cm^{-1} absorption lines, measured in[1], the absolute concentration of the Sm^{2+} ion (see the table), was determined by a photographic method. If we use the value of the oscillator strength for the 1.6×10^4 cm^{-1} band, determined in[11], then the concentration is somewhat larger. Dielectric mirrors with reflectances $r_1 = 99$ and $r_2 = 93\%$ were sputtered simultaneously on the end faces of the samples intended for the investigation of lasing.

3. EXCITATION

To be able to calculate the excitation parameters of a crystal in a wide temperature interval, we measured

Dependence of the concentration of Sm^{2+} in the $CaF_2:Sm^{2+}$ crystal on the concentration of SmF_3 and CeF_3 (wt.%) after an x-ray dose of 10^8 r.

SmF_3	CeF_3 (wt.%)					
	—	0,03	0,05	0,1	0,2	0,5
0,03		$1,7\cdot10^{18}$				
0,1	$0,4\cdot10^{18}$		$1,8\cdot10^{18}$	$2,2\cdot10^{18}$	$2,3\cdot10^{18}$	
0,5	$1,2\cdot10^{18}$				$2,6\cdot10^{18}$	$2,8\cdot10^{18}$

the absorption coefficient of ruby radiation $\alpha_r(T)$ by two methods. In the first method the coefficient was determined from the attenuation of the beam of a Q-switched ruby laser in a $CaF_2:Sm^{2+}$ plate at probing-beam energies 10^{-3}—0.5 J. The probing signal had the same waveform as the reference signal, and no distortions connected with saturation of the transition were observed. The absorption coefficient was also measured by transmission of incandescent-lamp light, cut off from the short-wave side at $\lambda = 660$ Nm, through the crystal and by subsequent separation of the necessary spectral component with a MDR-2 monochromator with dispersion 40 Å/mm. The spectral width of the gap was 0.8 Å (1.6 cm^{-1}).

The results for a sample with Sm^{2+} concentration 1.8×10^{18} cm^{-3} are shown in Fig. 2. The temperature of the crystals was varied by slowly heating or cooling the samples in liquid-helium and nitrogen vapor. The temperature was measured with the aid of a copper wire-wound resistor[12] with accuracy better than 1°K. The crystals were placed in a glass Dewar or in a foamed-plastic cryostat, in which the sample was clamped between quartz rods serving as light pipes. The inactive absorption was measured with a $CaF_2:Sm^{3+}$ sample cut out from the same stock piece, but not exposed to x-rays. In one of the samples, this absorption was less than 0.01 cm^{-1} at the ruby-laser frequency.

Let us examine the mechanism of the optical excitation of the $CaF_2:Sm^{2+}$ crystal by ruby-laser emission. The existing classification of the energy levels in this crystal[5] indicates that the first allowed electronic transition from the ground state to the upper electronic states is the transition ${}^1\Gamma_1^+({}^7F_0) \rightarrow {}^3\Gamma_4^-$ from the $4f^6$ shell to the $4f^55d$ shell (Fig. 1). The 14,497 cm^{-1} absorption line corresponding to this transition is anti-Stokes relative to the 14,403 cm^{-1} ruby-laser line.

In the temperature region 4.2—300° under consideration, and at not too high an intensity of the incident light (to exclude two-photon transitions), the ruby-emission absorption can be attributed to several causes. The first is the absorption by the wings of the 14,497 cm^{-1} line or lines from the stronger 1.6×10^4 cm^{-1} band. It is due to the electron-vibrational transitions from the ground state, with absorption of one or several lattice phonons. However, the most intense apparently is the transition, allowed by the symmetry rules, from the ground state ${}^1\Gamma_1^+({}^7F_0)$ to the level ${}^3\Gamma_4^-$ with absorption of a phonon of 94 cm^{-1} frequency. The symmetry rules allow also an electron-vibrational transition from the ground state directly to the ${}^1\Gamma_1^-$ level with emission of two phonons. The last obvious cause of excitation of the luminescence are electronic transitions from the level ${}^3\Gamma_4^+({}^7F_1)$ to the higher electronic states of the $4f^55d$ shell.

Figure 2 shows also the temperature dependence of the absorption coefficient measured by us at a frequency of 14118 cm^{-1} of the electronic transition ${}^3\Gamma_4^+({}^7F_1) \rightarrow {}^1\Gamma_1^-$. In a broad temperature interval, the main contribution to the absorption is apparently made by the same processes that are essential for $\alpha_r(T)$, but in the temperature region 50—130°K there appear weakly pronounced resonant properties of this transition[1]. The absorption coefficient at the frequency 14497 cm^{-1} of the electronic transition ${}^1\Gamma_1^+({}^7F_0) \rightarrow {}^3\Gamma_4^-$ has an entirely different dependence. It is large already at 4.2°K (1.7 cm^{-1}), and increases slowly with increasing temperature to 78°K, after which it goes jumpwise to ~ 3 cm^{-1}. In the temperature interval 80—130°K, the absorption coefficient decreases exponentially to 1.8 cm^{-1}, and with further increase of temperature it behaves like a continuous absorption near this line at the frequency 14620 cm^{-1}. It should be noted that according to our measurements the width of the absorption line at 78°K amounts to 12.5 cm^{-1}, whereas other measurements[1-3] quote 20 cm^{-1} at 20°K. In other words, this absorption line

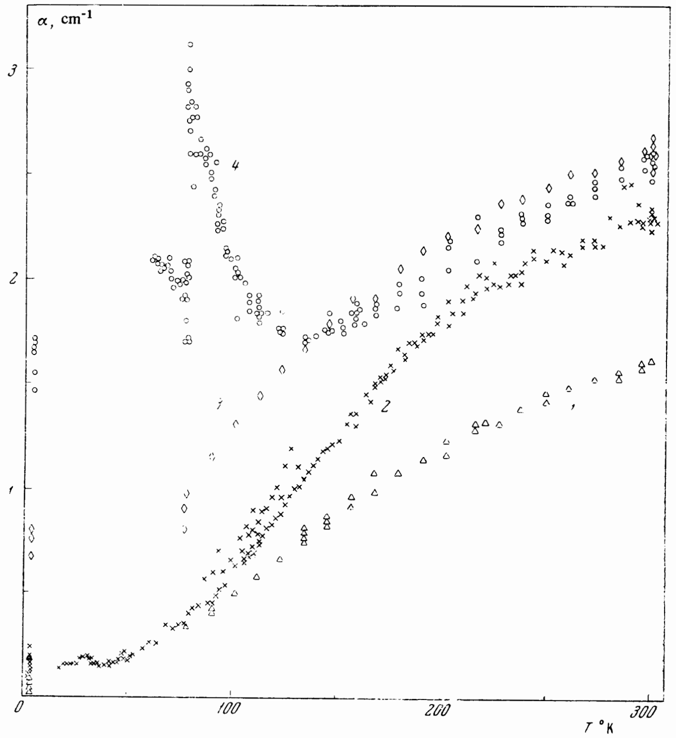

FIG. 2. Temperature dependence of the absorption coefficient of the $CaF_2:Sm^{2+}$ crystal at several frequencies (in cm^{-1}): 1—14118, 2—14403, 3—14620, 4—14497. Sm^{2+} concentration—1.8×10^{18} cm^{-3}.

does not broaden when the temperature is increased from 20 to 78°K, and measurements at higher temperatures show that it becomes broader by several times. Thus, this is similar to a clearly pronounced resonance process. It is too early to speak of its nature as yet. We can assume, however, that it is due to the transition ${}^1\Gamma_1^+({}^7F_0) \to {}^1\Gamma_1^-$ with emission of two phonons. The distance between the levels ${}^1\Gamma_1^-$ and ${}^3\Gamma_4^-$ is 116 cm^{-1}, and the temperature at which resonance occurs is 55°K. It is possible that the levels begin to interact strongly at this temperature, leading to a broadening of the ${}^3\Gamma_4^-$ level.

It should be noted, however, that the experiments have a preliminary character, since the measurements made in the intermediate temperature region 65—130°K may be distorted by unaccounted for factors such as settling of absorbing and scattering films on the optical surfaces, although attention was paid to this. Special complications may arise at a result of the transfer of the measurements from the liquid nitrogen to its vapor.

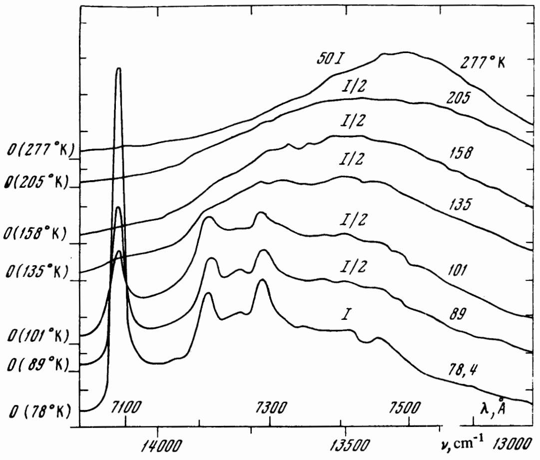

FIG. 3. Temperature dependence of the contour of the luminesence band in $CaF_2:Sm^{2+}$ crystal (I—intensity, 0(T°K)—level of zero intensity at T°K).

LUMINESCENCE

We made a thorough study of the luminescence spectrum of the $CaF_2:Sm^{2+}$ crystal in the temperature region 65—300°K, using a photographic method with an ISP-51 spectrograph with dispersion ~ 50 Å/mm. The most suitable for the indicated spectral interval were the ORWO I-750 photographic plates. The samples were slowly heated in nitrogen vapor and when the required temperature was reached they were exposed to a pulse from a Q-switched ruby laser or from the commercial "Razdan-2a" apparatus, which generated pulses of 0.2 msec duration.

It can be stated on the basis of the performed measurements that the spectrum is identical with that obtained by excitation with ordinary light sources. An interesting fact is the excitation of the luminescence line 14,497 cm^{-1}, which is anti-Stokes with respect to the ruby laser frequency. In the observation of this line, self-absorption must be eliminated. The intensities of the sharp lines 14118, 14234, and 14497 cm^{-1} at 77°K are related as 1 : 0.057 : 0.0018, which coincides at the experimental accuracy with the previously measured values[1,3]. These results offer evidence that thermal equilibrium is established between the upper levels within a time shorter than the luminescence lifetime $\tau_l = 2 \times 10^{-6}$ sec.

In our investigation we measured, by the heterochromic photometery method, the temperature dependence of the contour of the luminescence continuum, which is shown in Fig. 3 for several temperatures. At temperatures up to 200°K a luminescence spectrum with normal photographic density was obtained following one ruby-laser flash. In the temperature-quenching region, the number of flashes was increased. It should be noted that the spectral sensitivity of the photographic plates was measured against the black-body spectrum at an exposure of one minute, and no corrections were introduced for the possible change of the sensitivity at an illumination with a 10^{-4} sec pulse.

A characteristic feature of the luminescence in the crystal is also the fact that, besides the electronic lines 14497, 14234, and 14118 cm^{-1}, it contains a broad (~ 1000 cm^{-1}) and intense structure band—a continuum due to the electronic-vibrational transitions. At a temperature 4.2°K, the continuum is adjacent on the long-wave side to the 14114 cm^{-1} line[3]. It follows therefore that at this temperature it is due to electron-vibrational transitions between the levels ${}^1\Gamma_1^- \to {}^3\Gamma_4^+({}^7F_1)$, with emission of lattice phonons; these transitions are symmetry-allowed in the dipole approximation. The continuum has a structure and its different maxima (quasilines) broaden with increasing temperature and merge with the continuum, the first to drop out being the shortest-wavelength components including the 14118 cm^{-1} line. It can be apparently assumed that the quasilines of the continuum are due to the pseudolocal oscillations of the Sm^{2+} impurity center[13].

A theoretical investigation, made for example by Rebane[14] in the adiabatic approximation using the Franck-Condon principle and the harmonic-oscillator wave functions, shows that the integrated emission intensity of the electron-vibrational band in an impurity crystal does not depend on the temperature. In our case this approximation is apparently valid at temperatures lower than those at which temperature quenching of the luminescence sets in, T = 150°K[6]. The integrated intensity of the electronic luminescence line I, which appears as a result of the transition not accompanied by emission of a vibrational quantum (quantumless quasiline) decreases with increasing temperature:

$$I \sim |M|^2 \exp\left\{-\frac{P}{\hbar\omega_v}\,\mathrm{cth}\,\frac{\hbar\omega_v}{2kT}\right\}.$$

Here M—electronic matrix element, ω_V—average frequency of the local oscillations, and P—total Stokes energy losses.

We performed relative measurements of the integrated intensity of the 14118 cm^{-1} electronic line, i.e., of the ratio of the area under the line to the area under the entire contour. The results are shown in Fig. 4. The same figure shows the theoretical curve plotted with parameters $\omega_V = 110$ cm^{-1} and P = 460 cm^{-1}, determined from the experimental points. It is seen that the assumptions used in plotting the curve are perfectly

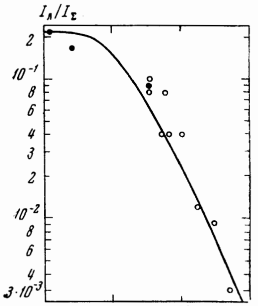

FIG. 4. Temperature dependence of integrated intensity of the 14118 cm^{-1} luminescence line. The dark points are taken from [1].

justified. The plot shows that if T ≳ 80°K the integrated intensity of the 14118 cm^{-1} luminescence line decreases exponentially with increasing temperature, and this is apparently the cause of the cessation of the generation of this line[15].

5. GENERATION

We investigated experimentally the generation spectrum of the $CaF_2:Sm^{2+}$ crystal excited with a Q-switched ruby laser having a radiation energy ~ 0.5 J. In accordance with the temperature dependence of the luminescence-band contour, the generation spectrum varies in the working-temperature range 65—210°K. At low temperatures, the generation is on the electronic 14118 cm^{-1} line. An increase of the temperature gives rise to additional broad lines in the generation spectrum, coinciding in position with the luminescence quasilines[1)]. These lines have a structure, with a characteristic distance ~ cm^{-1} between components. The position and the form of the components were determined with a Fabry-Perot resonator, made up of mirrors coated on the end surfaces of the crystal. The distance between two longitudinal modes in the investigated samples was 0.18 cm^{-1}.

The present study is devoted principally to the coarse structure of the generation spectrum and to its temperature dependence. A theoretical consideration of this question is given in[16,17] for an impurity center having two systems of levels. These papers contain calculations of the gain function g(ω, T) at frequency ω and temperature T as a function of the parameters of the substance:

$$g(\omega, T) = \frac{Z_2}{Z_1}\left[N_2 - N_1 \exp\left(\frac{\hbar(\omega-\omega_0)}{kT}\right)\right]\left[\frac{2\pi c}{\omega n(\omega)}\right]^2 f(\omega, T);$$

Here Z_1 and Z_2 are the partition functions corresponding to the ground and excited states of the ion; N_1 and N_2 are the total populations of the final and initial systems of the laser-transition levels. It is assumed that thermodynamic equilibrium is established inside each system. Further, n(ω) is the refractive index and f(ω, T) is a function describing the contour of the luminescence band in terms of the number of photons emitted per second in a unit spectral interval at the frequency ω. Thus, by specifying the population of the level systems taking part in the laser emission, and knowing the luminescence contour, it is possible to determine, for a given temperature, the spectral region in which the generation takes place. This phenomenon was investigated experimentally in the $CaF_2:Sm^{2+}$ excited with a ruby laser.

In accordance with the arguments advanced in Secs. 2 and 3, it is assumed that the laser transition occurs between the electronic states $^1\Gamma_1^-$ and $^3\Gamma_4^+(^7F_1)$, the energy gap between which is equal to $\hbar\omega_0$, with emission of one or several vibrational quanta. To determine the spectral region of the generation at a given temperature and pump, it is sufficient to know the relative value of $g(\omega, T_{fix})$, and to confine oneself to a relative measurement of the quantities that depend explicitly on the temperature. The quantity N_2 is determined from the ruby-emission absorption, the impurity concentration N_0 is known (see the table), $N_1 = (N_0 - N_2)\exp[-\Delta E/kT]$, where $\Delta E \geq 263$ cm^{-1}, and f(ω, T) (in relative units) is obtained from measurements of the luminescence contour.

Figure 5 shows the results of such calculations as well as microphotograms of the generation spectra. It is obvious that the generation frequencies coincide with the maxima of the gain function. This result differs from that obtained by Johnson et al.[18] for a $MgF_2:Ni^{2+}$ crystal, possibly because the experiment with monochromatic pumping is cleaner than with excitation by a flash lamp, and the temperature and luminescence spectrum are measured more accurately. It should be noted, however, that the generation lines sometimes do not coincide with the absolute maxima of g(ω, T) at high temperatures. This may be due to errors (on the order of several percent) in the measurement of the contour of the continuum, and also to the frequency dependence

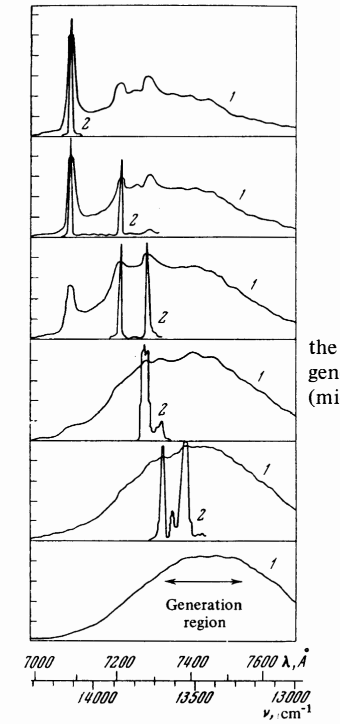

FIG. 5. Temperature dependence of the gain function g(ω, T)(1) and of the generation spectrum of the $CaF_2:Sm^{2+}$ (microphotograms)(2).

[1)] The assumption [15] that the appearance of the 0.73 nm generation line can be attributed to the transition $^3\Gamma_{4^-} \rightarrow \Gamma_5(^7F_2)$ seems incorrect to us.

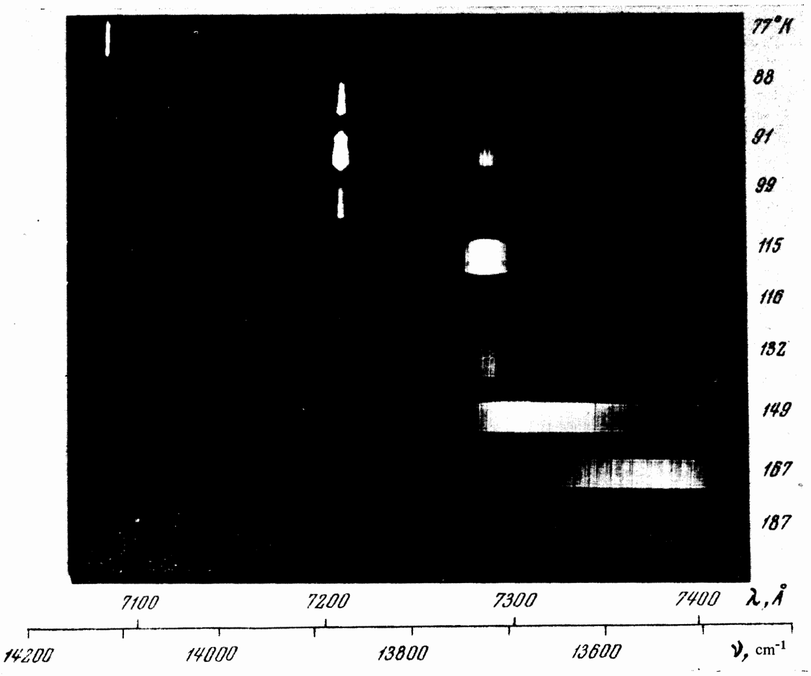

FIG. 6. Structure of generation spectrum of $CaF_2:Sm^{2+}$ crystal as a function of the temperature.

of the reflectance of the mirrors.

The coarse structure of the generation spectrum changes significantly in the temperature region 65–210°K. The highest temperature at which the generation spectrum was recorded was 210°K. It was not the purpose of the present work to obtain generation at a higher temperature. At a temperature below 77°K, generation exists only on the 14118 cm^{-1} (708 nm) electronic transition (Fig. 5). An increase of the temperature leads to a broadening of the generation line, in agreement with the data of Anan'ev et al.[19]. In the region $T > 77°K$, new lines appear in the generation spectrum, the positions of which coincide with the quasilines of the continuum. An increase of the temperature causes generation to stop at the short-wave lines, and to be replaced by long-wave lines with ever increasing width. In the temperature range under consideration, the total width of the generation region changes from several to several hundred cm^{-1}. The maximum wavelength at which generation was obtained was 745 nm (13420 cm^{-1}). It can be assumed that the generation spectrum shifts discretely by ~700 cm^{-1} only as a result of the change of the temperature of the $CaF_2:Sm^{2+}$ crystal in the 65–210°K range. Data on the generation kinetics[8] confirm that these lines are due to generation and not to stimulated Raman scattering, although the latter may be present. It is interesting to note that the generation is excited also when temperature quenching of the luminescence takes place[6]. This indicates that the probability of stimulated electron-vibrational transitions in this temperature region prevails over the non-radiative decay of the metastable level.

The temperature dependence of the structure of the generation spectrum of the $CaF_2:Sm^{2+}$ crystal was investigated at high resolution with a DFS-13 spectrograph with dispersion 4 Å/mm. The spectra obtained with one of the investigated samples are shown in Fig. 6. It is seen from them that each of the lines, including the electronic one, consists of many spectral components with a characteristic distance ~1 cm^{-1}, which sometimes is maintained with a high degree of accuracy, and in other cases constitutes a random set of narrow lines. The width of many of these components is narrower than the distance between the nearest longitudinal modes. Such a discreteness indicates that each of the luminescence quasilines is broadened inhomogeneously and it is impossible to cause the excitation energy to become concentrated in one line by simple methods. The distances between the structure components make it possible to estimate the spectral region of the homogeneous broadening at ~1 cm^{-1}[20].

6. CONCLUSION

The investigations performed in this work show that the use of a ruby laser as a pump source is quite fruitful for the study of the temperature dependence of the luminescence and generation spectra of the $CaF_2:Sm^{2+}$ crystal. The luminescence spectrum as a whole is an inhomogeneously broadened band. The luminescence quasilines are likewise inhomogeneously broadened. A determination of the region of the homogeneous broadening can be based on an investigation of the mode structure of the generation spectrum[21]. From measurements made by Anan'ev and Sedov[22] on the 14118 cm^{-1} generation line it follows that this region can be confined to 4×10^{-3} cm^{-1} (2×10^{-3} Å).

In this connection, the problem of smooth tuning of the generation frequency and of effective utilization of the excitation energy calls for identification of the vibrational structure of the electronic states and for an investigation of the relaxation processes. Great help may be rendered in this case by spectral-kinetic investigations of generation and luminescence with resolution time 10^{-10}–10^{-11} sec.

A quantitative explanation of the temperature dependence of the absorption coefficient at the characteristic frequencies, excluding the 14497 cm^{-1} line, is possible on the basis of the available information. However, to calculate $\alpha(T)$ at the frequency of the ${}^{1}\Gamma_{1}^{+}({}^{7}F_{0}) \rightarrow {}^{3}\Gamma_{4}^{-}$ transition it is necessary to have additional facts.

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