LASER MODULATION AT THE ATOMIC LEVEL

Monthly Report No. 8

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LASER MODULATION AT THE ATOMIC LEVEL

Purpose

Research on methods of influencing internally the radiating centers of active laser materials in order to achieve laser modulation is the principal objective of the work carried out under this contract.

Summary

Spectroscopic study of YAG:Nd^{3+} has continued, resulting in measurement of the temperature dependence of energy levels involved in laser emission. The separation of the levels of the $^4F_{3/2}$ state is found to be 84.5 ± 0.1 cm^{-1}, independent of temperature. Integrated intensities of the eight fluorescent lines between 1.05 and 1.08 μ have been measured between 180°K and 400°K. Apparent linear dependence of linewidth on temperature is noted, but is subject to refinement of data. Temperature dependence of laser threshold has been measured and is discussed on the basis of spectroscopic data.

Man-Hours Worked

The total number of man-hours worked during this reporting period is 327.

TECHNICAL DISCUSSION

A. Study of YAG:Nd^{3+} Spectrum

1. Temperature Dependence of Energy Levels Relevant to Laser Oscillation

The energy levels relevant to the present discussion are the upper and lower levels of the $^4F_{3/2}$ term and the three lowest levels of the $^4I_{11/2}$ term. The positions of the centers of these levels have been determined by measuring the absorption from the lowest of the $^4I_{9/2}$ ground state levels to the $^4F_{3/2}$ levels, and the fluorescence emitted during transitions from $^4F_{3/2}$ to $^4I_{11/2}$ levels. It is found that the energy, in wavenumbers relative to the ground state, can in all cases be represented to within experimental error by quadratic equations of the form:

$$\nu = \nu_o + aT + bT^2.$$

The values of the parameters ν_o, a, and b for the levels that we are considering are shown in Table I. The lower and higher levels of $^4F_{3/2}$ are labeled L and H respectively, while the levels of the $^4I_{11/2}$ term are labeled A, B, C... in order of increasing energy. Figure 1 presents the

TABLE I

Parameters Specifying Temperature Dependence of Energy Levels

Level	ν_o (cm^{-1})	a ($cm^{-1}/°K$)	b ($cm^{-1}/°K$)
A	2003.9	-1.3×10^{-3}	-----
B	2031.4	-7.4×10^{-3}	-5.1×10^{-6}
C	2110.4	-1.3×10^{-3}	-----
L	11429.7	-1.28×10^{-2}	-2.3×10^{-5}
H	11514.2	-1.28×10^{-2}	-2.3×10^{-5}

experimental data in graphical form. The separation of L and H remains $84.5 \pm 0.1\ cm^{-1}$ over the entire temperature range, but the temperature dependence of the entire $^4F_{3/2}$ term is the principal source of laser thermal tuning. The temperature dependence of levels A and C was too slight to permit useful accuracy in the determination of the b terms, so they have been omitted from Table I. The greater temperature dependence of level B than level C is responsible for reducing the separation of emission lines 4 and 5 (see Monthly Report No. 7 for line notation) as temperature increases.

2. Population Distribution in the $^4F_{3/2}$ Term

Measurements of the intensities of the eight emission lines in the 1.05 to 1.08μ range were made by scanning the spectrum with an S-1 photomultiplier at 8 Å resolution. This resolution was adequate to resolve all lines except 4 and 5, but resulted in an instrument limited linewidth. The peak intensity recorded under these conditions closely approximates the integrated intensity of the line. The data obtained from this experiment are shown in Fig. 2. If we assume that the radiative transition probabilities between states are temperature insensitive, then the ratio of intensity from an upper state initial level to that from a lower state initial level is set by the Boltzmann distribution of the populations of the states:

$$\beta_j I_{H,j}/I_{L,j} = (1 + e^{+\gamma})/(1 + e^{-\gamma}),$$

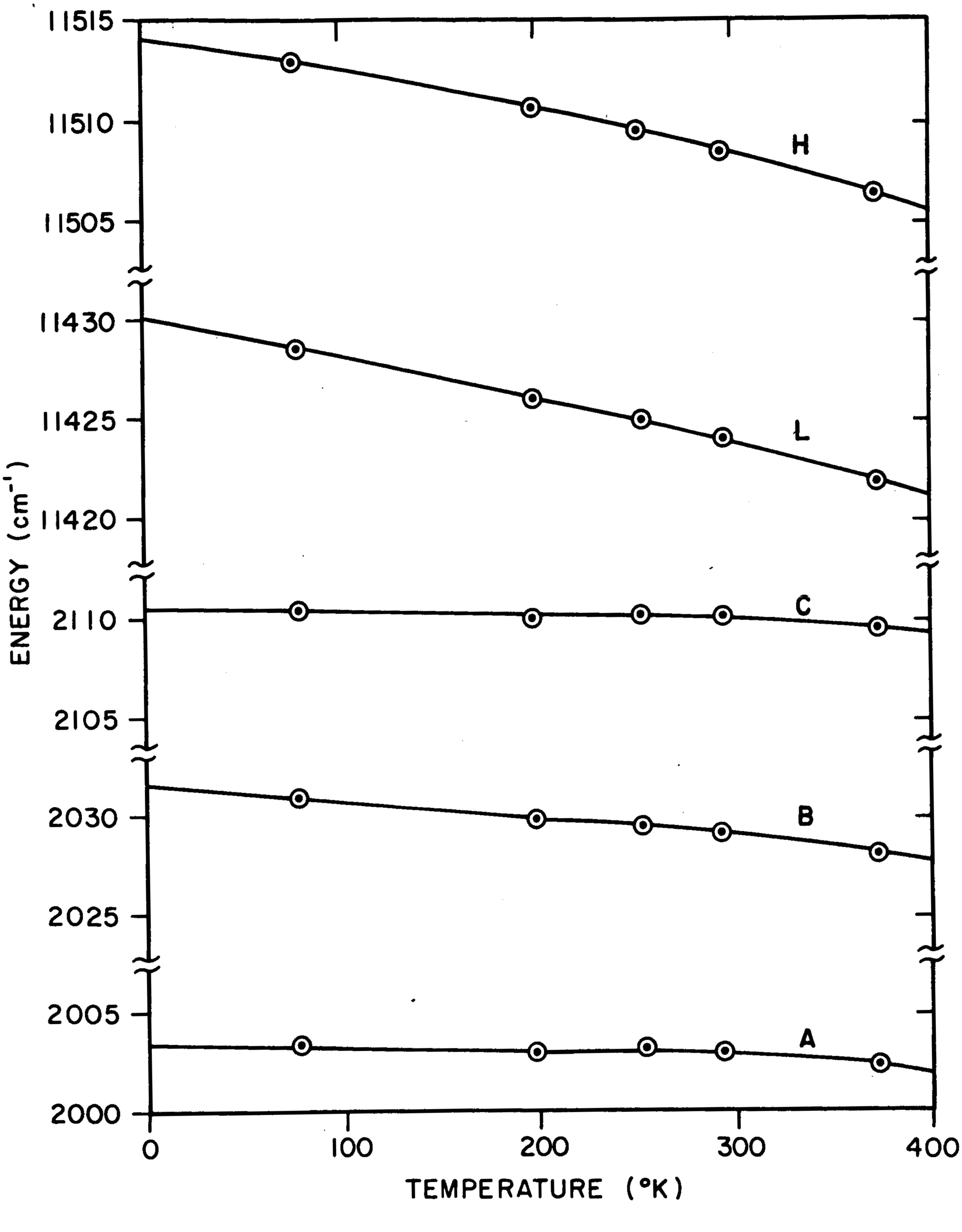

Fig. 1. Temperature dependence of energy of the YAG:Nd^{3+} levels involved in laser emission.

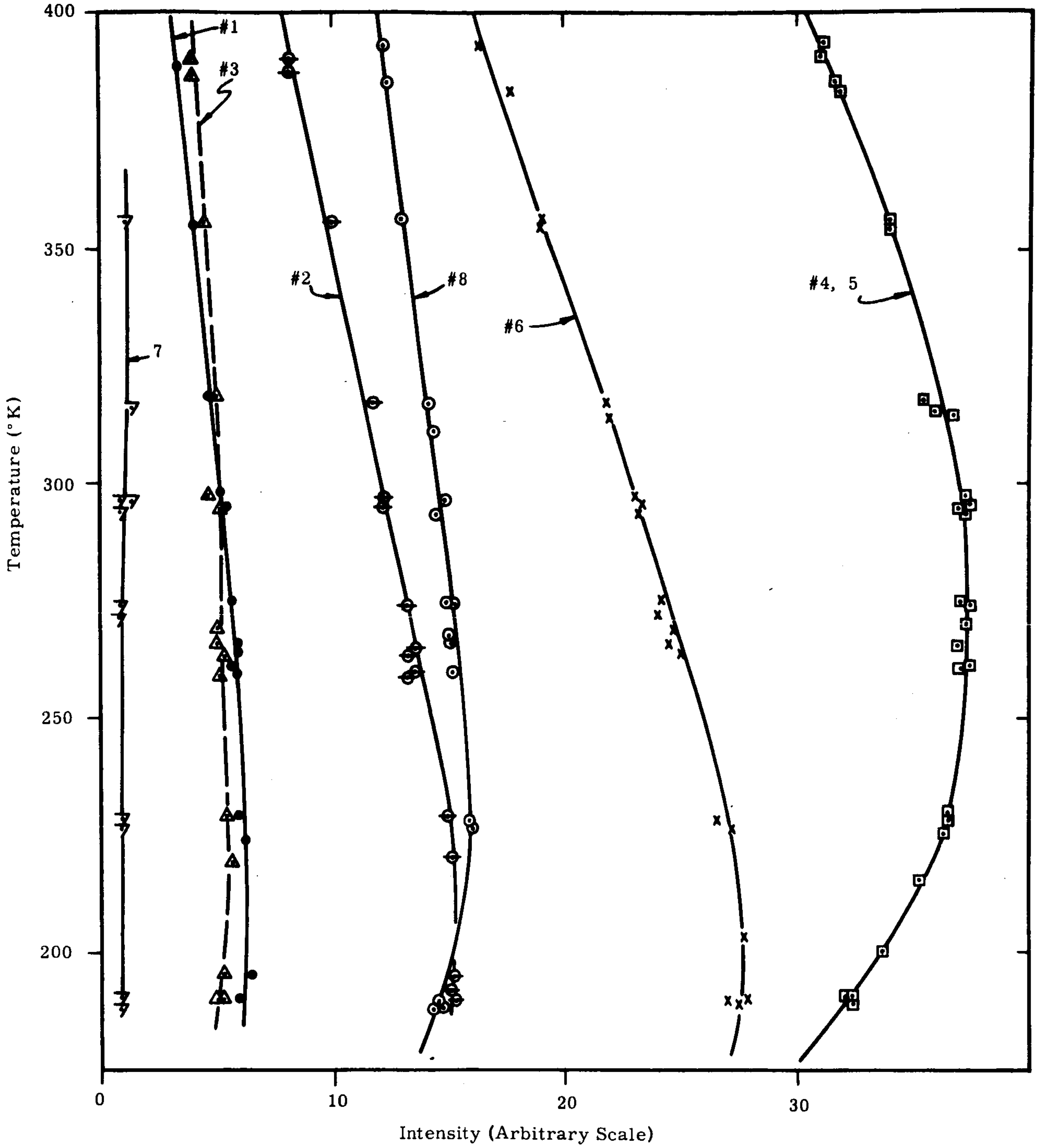

Fig. 2. Integrated intensity of YAG:Nd^{3+} fluorescent lines as a function of temperature.

where β depends on the relative transition probabilities to the same terminal state, j, and $\gamma = hc\,\Delta\nu/kT$. The theoretical ratios, normalized for $T = \infty$, are shown in Fig. 3, along with the experimental ratios obtained for lines 6 and 8 and lines 1 and 3 between 200°K and 400°K. The agreement is within the experimental error for the intensity measurements, which were taken from data presented in Fig. 2.

A general tendency toward lower fluorescence intensity in all lines as temperature increases is indicated by Fig. 2. This is probably due to the increase in phonon assisted fluorescence as temperature increases. The broad phonon assisted fluorescence spectrum has been noted on some of our long exposure plates, but it has not been studied quantitatively.

3. Linewidth

The linewidth measurements that we have made are subject to considerable error because the spectroscopic 1-Z plates were not exposed in a way which would provide a gray scale, so the plate gamma could only be roughly estimated. Over the temperature range where line broadening is mainly due to homogeneous interaction with lattice vibrations rather than strain, the linewidth closely approximates a power law relationship (e.g., McCumber and Sturge[1] for Cr^{3+} in Al_2O_3). The data are plotted on a log $(\Delta\nu)$ vs. log T graph in Fig. 4. Straight lines fitting the points have a slope of 1.0, indicating a simple linear relationship, $\Delta\nu = bT$, where for

1. D. E. McCumber and M. D. Sturge, J. Appl. Phys., 34, 1682 (1963).

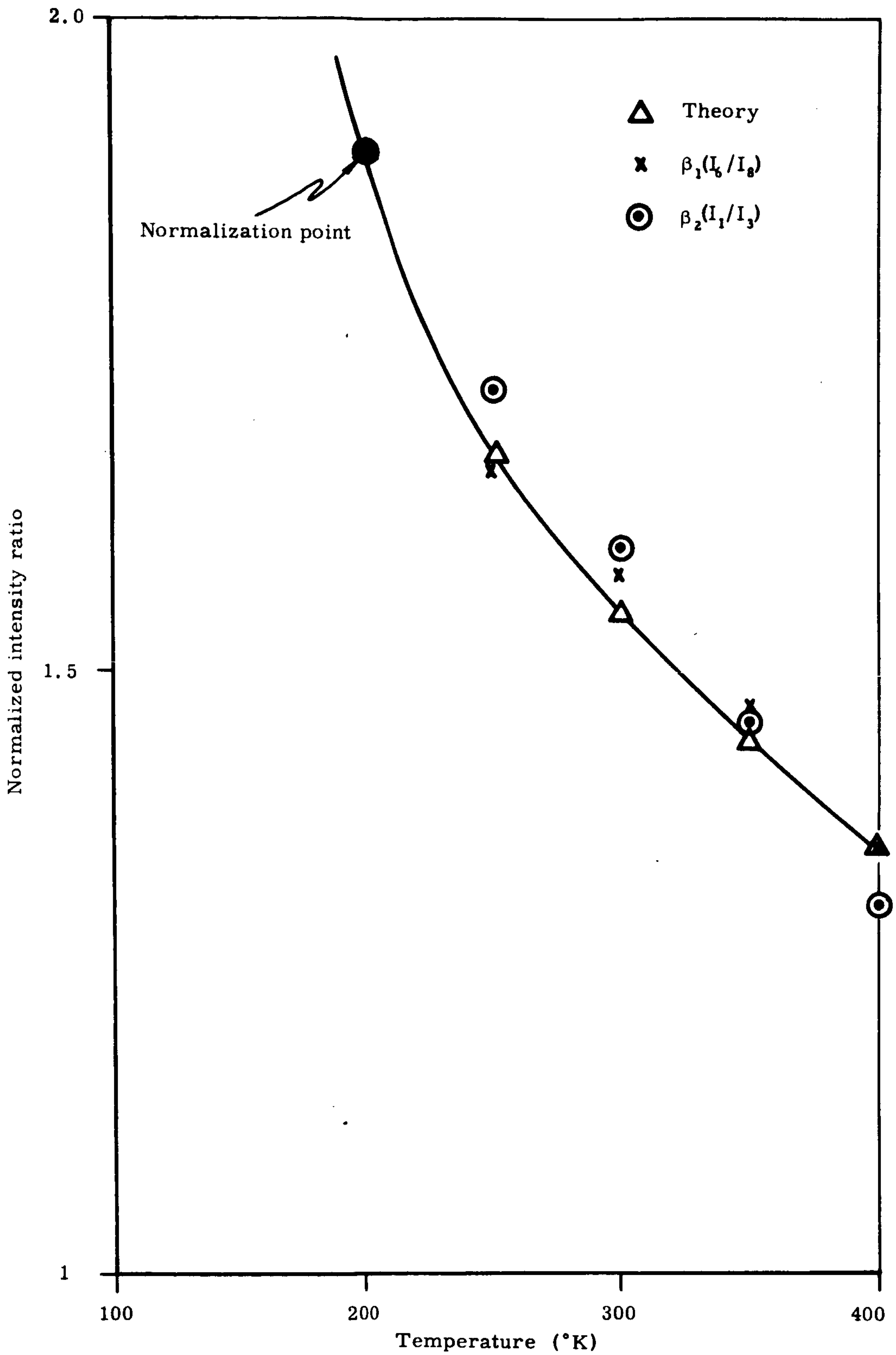

Fig. 3. Ratio of intensities of fluorescent lines originating from upper and lower levels of $^4F_{3/2}$ state. Experimental curves adjusted to match theory at 200 °K.

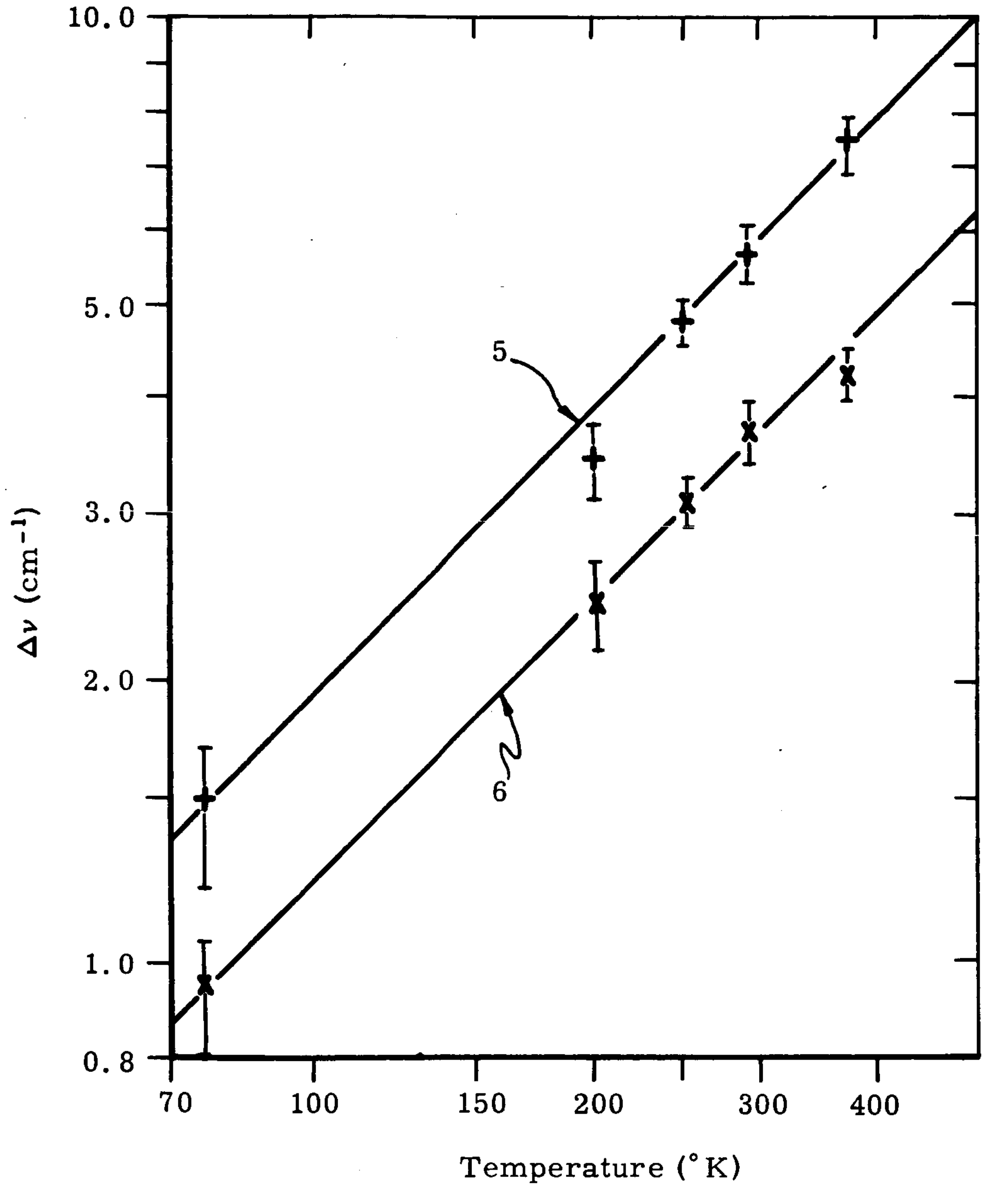

Fig. 4. Fluorescent line width as a function of temperature for $YAG:Nd^{3+}$ lines 5 and 6.

line 5, $b = 1.95 \times 10^{-2}\ (\mathrm{cm}\,^\circ\mathrm{K})^{-1}$; and for line 6, $b = 1.17 \times 10^{-2}\ (\mathrm{cm}\,^\circ\mathrm{K})^{-1}$. However, measurements of greater precision over a larger temperature range would be required before one could consider such a relationship to be well established.

B. Calculation and Measurement of Laser Threshold Temperature Dependence

With the spectroscopic data now available, laser threshold at constant pumping efficiency is predictable. Since we are dealing with a four-level laser system with negligible population in the terminal state, laser gain is proportional to initial level population and the peak intensity of the fluorescent line. The optical resonator characteristics which determine the gain required to reach threshold are temperature insensitive, so we consider the case where the same gain is required at all temperatures in order to achieve threshold. Constant pumping efficiency implies that the population of the $^4F_{3/2}$ term, N_F, is proportional to the pump energy, E_p. Assuming a Boltzmann distribution in population within the $^4F_{3/2}$ term, the pump energy required to maintain constant gain when the initial state is the upper state (e.g., line 5) is:

$$E_p = C\,\Delta\nu\,(1 + \exp\gamma),$$

and when the initial state is the lower level (e.g., line 6) it is:

$$E_p = C'\,\Delta\nu\,[1 + \exp(-\gamma)],$$

where C and C' are temperature independent constants inversely proportional to the radiative transition probabilities and γ has been defined above.

Experimental measurements of laser threshold between 120°K and 400°K were made using a xenon flashlamp of 300 μsec flash duration as pump. At each temperature the integrated laser emission energy per pulse was measured as a function of input pump energy. The laser energy was then linearly extrapolated to zero in order to obtain threshold pump energy. The results of these measurements are shown in Fig. 5. The threshold calculated for line 5 and line 6 on the basis of the above equations are also shown, the values of C and C' having been adjusted so that all three curves meet at T = 222°K.

It is interesting to note that the experimental threshold rises much more slowly with temperature between 300°K and 400°K than is calculated. Since the calculated threshold did not take into account reduction in sharp line fluorescent intensity due to phonon assisted fluorescence, this effect is even more remarkable. It is due to the reduction in separation between lines 4 and 5 in addition to increasing overlap due to line broadening. At low temperatures the threshold for line 6 does not decrease as rapidly as calculations predict. This effect is due to a reduction in fluorescent intensity at low temperature, which is probably due to the narrowing of pump absorption lines, which reduces the efficiency of coupling to the broadband pumping lamp spectrum.

The negative temperature coefficient of threshold near room temperature implied in Monthly Report No. 7 was based on inadequate measurements and should be considered to be superceded by the data presented here.

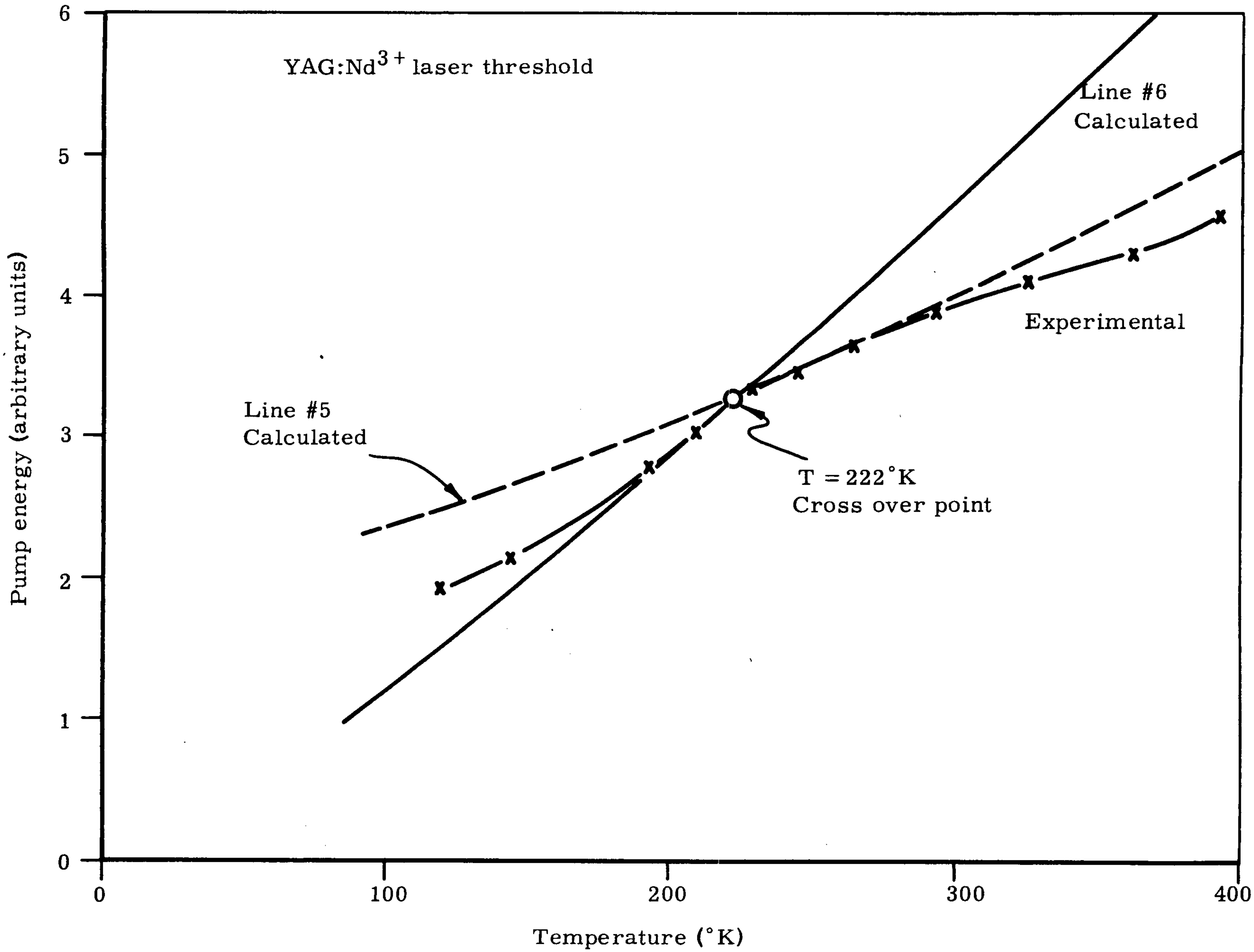

Fig. 5. Observed and calculated thresholds for YAG:Nd $^{3+}$ laser oscillation.

C. Work In Progress

The magnetic field tuning indicated in Report No. 7 needs to be investigated with greater precision. A series of experiments is being undertaken wherein the peak field is varied to obtain field dependence, rather than using the time dependence of a single high-field pulse as in previous experiments. A CW pumping cavity which will fit in the pole gap of a dc electromagnet is being designed. This will permit determination of transverse magnetic field tuning with greater precision than is possible with pulsed fields.

Consideration of E. P. R. spectroscopy of the metastable states in YAG:Nd^{3+} (and Al_2O_3:Cr^{3+}) is continuing. This technique could be a considerable help in analyzing Zeeman splitting of optical transitions originating from such levels.