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LASER MODULATION AT THE ATOMIC LEVEL

Monthly Report No. 7

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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

21308

The emission of 1 wt% Nd³⁺ in yttrium aluminum garnet (YAG) in the wavelength range 1.05 to 1.08 μ was studied as a function of temperature between +100°C and -196°C, giving a clearer picture of the reasons for the unusual reduction of YAG:Nd³⁺ laser threshold with lowering of temperature near room temperature.

The shift of laser emission with homogeneous pulsed magnetic fields was also studied over the same temperature range by time resolved spectroscopy. For the transition responsible for laser emission above -50°C the highest gain Zeeman component has an apparent "g" value of -2.51, with a weaker +2.51 component observable at the low temperature end of the range over which this transition oscillates. The highest gain component of the low temperature laser transition has a "g" value of +1.94.

Author

Man-Hours Worked

The number of man-hours worked during this report period was 352.

TECHNICAL DISCUSSION

A. Spectroscopic Study of Infrared Nd³⁺ Fluorescence from YAG:Nd³⁺

The only spectroscopic data available to us on Nd³⁺ in YAG are contained in the paper of Koningstein and Geusic¹ which has been referred to in previous reports. These data are sketchy and only of moderate resolution. In order to clarify the behavior of the YAG:Nd³⁺ laser, we have undertaken a more detailed study of the infrared fluorescence and absorption spectrum of this crystal.

The first step was to obtain the temperature dependence of the fluorescence in the 1.06 μ to 1.07 μ range, which is responsible for the laser emission. The Bausch and Lomb dual grating spectrograph was used with Kodak type 1-Zinfrared sensitive spectroscopic plates. An ARL "Spectroline" scanner was used for reading the plates. The procedure used permitted measurement of sharp lines to $\pm 0.03 \text{ \AA}$, using quadratic interpolation from higher order mercury discharge reference lines. The temperature was regulated with a nitrogen gas stream as in our previous experiments. The measurement at liquid nitrogen temperature was performed with a stream of liquid nitrogen cooling the sample.

A spectrum of the eight emission lines in the vicinity of the laser emission obtained from the densitometer trace of emission from the sample at a temperature of -20°C is shown in Fig. 1. This spectrum is

1. J. K. Koningstein and J. E. Geusic, Phys. Rev., 136, A711 (1964).

labeled with the transition numbers given in Reference 1, which will be used in the subsequent discussion of the spectrum. Segments of the spectra obtained from the densitometer trace, showing the laser transitions (lines 4, 5, and 6) are presented in Fig. 2. The shift of transition 6 (low temperature laser) against the superposed mercury arc reference lines can be clearly seen. The plot of peak wavelength and linewidth versus temperature for lines 4 and 5 in Fig. 3 demonstrates one reason for the lowering of threshold with increasing temperature in the range near room temperature. The slightly greater temperature coefficient of line 5 ($-2.5 \text{ cm}^{-1}/^{\circ}\text{K}$) than that of line 4 ($-1.3 \text{ cm}^{-1}/^{\circ}\text{K}$), combined with the broadening of the lines, causes the total gain at the peak of the compound line to increase with temperature. In addition, line 5 originates from the upper level of the $^4\text{F}_{3/2}$ state, so that increasing population of that level with increasing temperature raises the integrated intensity of that transition. The combination of increasing overlap and increasing intensity for line 5, competing with increasing broadening of both lines, should lead to a maximum gain (minimum laser threshold) in the range $+30^{\circ}\text{C}$ to $+100^{\circ}\text{C}$. The location of this maximum is important for optimum operation of CW lasers made of this material. Laser measurements to determine the temperature of maximum gain are being made and will be reported next month. The temperature dependence of line center and line width for line 6 is shown in Fig. 4. In order to determine the temperature shifts associated with the separate energy levels, temper-

ature shifts of the ${}^4F_{3/2} \leftrightarrow {}^4I_{9/2}$ transitions are being studied to determine the temperature shifts in the upper and lower levels of the laser transitions independently.

B. Magnetic Field Shift of YAG:Nd³⁺ Laser Emission

1. Experimental Procedure.

The influence of a homogeneous magnetic field on the emission frequency of the YAG:Nd³⁺ laser was measured by utilizing the technique used for ruby lasers, as reported in Monthly Report #6. The Helmholtz coil laser pumping cavity shown in Fig. 3 of Report #2 was used. The active length of the laser rod was only 23 mm, which is 0.72 times the coil radius. From Fig. 4 of Report #2 we note that the field at the ends of the rod is only 1.4% lower than at the center in this case, which is a negligible inhomogeneity. The magnetic field obtained by discharging 90 μ f through the loops has a time dependence of the form: $H(t) = H_0 e^{-\gamma t} \sin \omega t$, where $\gamma = 3.003 \times 10^4 \text{ sec}^{-1}$ and $\omega = 3.015 \times 10^5 \text{ sec}^{-1}$. This relation was used to convert the time dependent spectra to magnetic field dependence, using the magnetic field calibration obtained with the probe coil.

The etalon flats for these experiments have dielectric coatings with 98% reflectivity at 1.06 μ . A subsidiary reflection peak in the blue permitted adjustment of parallelism using a blue line from a cadmium arc lamp. Analysis of the errors encountered in this method of time resolved spectroscopy showed that it is preferable to reduce magnification

of the ring pattern so as to include at least one fringe beyond the zero angle point in order to determine y_0 more accurately.

2. Experimental Results

At room temperature the laser emission is produced by line 5 (see Fig. 1 for spectrum) with a shoulder on the long wavelength side due to line 4. The magnetic field causes the emission frequency to shift to longer wavelengths with an apparent "g" value of -2.51, where "g" = $\Delta\nu/\beta H$ (see Fig. 5). The free ion "g" value for this transition, assuming $\Delta m = 1$, is $g_{\text{calc}} = \pm 1.81$ (see Report #5). As the temperature is lowered, another component with $g = +2.5$ is observed between -40°C and -55°C , which is the low temperature limit for laser oscillation on line 5. The oscillation takes place either in the $+\Delta\nu$ or $-\Delta\nu$ mode, but not both simultaneously under the conditions of our experiment. Furthermore, when the rod is pumped near threshold, the magnetic field quenches oscillation during the first quarter period of magnetic field, indicating a removal of degeneracy, and hence reduction in gain, by the magnetic field. This effect becomes more pronounced as the temperature is lowered because of reduction in linewidth. Since measurements at -50°C indicate that the $+\Delta\nu$ and $-\Delta\nu$ transitions have approximately the same gain, the dominance of the $-\Delta\nu$ transition at room temperature can be attributed to the enhancement of gain on the low frequency side of line 5 line by line 4, as shown by the marked fluorescence line assymetry (see Fig. 2).

At temperatures lower than -55°C the gain in line 6 exceeds that in line 5, and the laser emission occurs at a frequency corresponding to the peak of line 6. The magnetic field also quenches oscillation in this transition during the initial quarter cycle, indicating that there is removal of degeneracy. Unlike the emission from line 5, however, only a component with a positive frequency shift is observed, having a value $g = +1.94$. (See Fig. 6.) The value of g predicted for the free ion for this transition (see Report #5) is $g_{\text{calc}} = \pm 0.683$. The discrepancy between the free ion g_{calc} values and the observed g values is due to the crystal field state mixing. As mentioned in Report #6, the crystal field terms should introduce angular dependence into g as well as distinguishing three different types of Nd^{3+} sites. Aside from gain reduction under the influence of the magnetic field, which merely implies removal of state degeneracy, no splitting due to different Nd^{3+} sites has been observed. More detailed studies, possibly including paramagnetic resonance spectroscopy, would be required to further elucidate the Zeeman effect in this material.

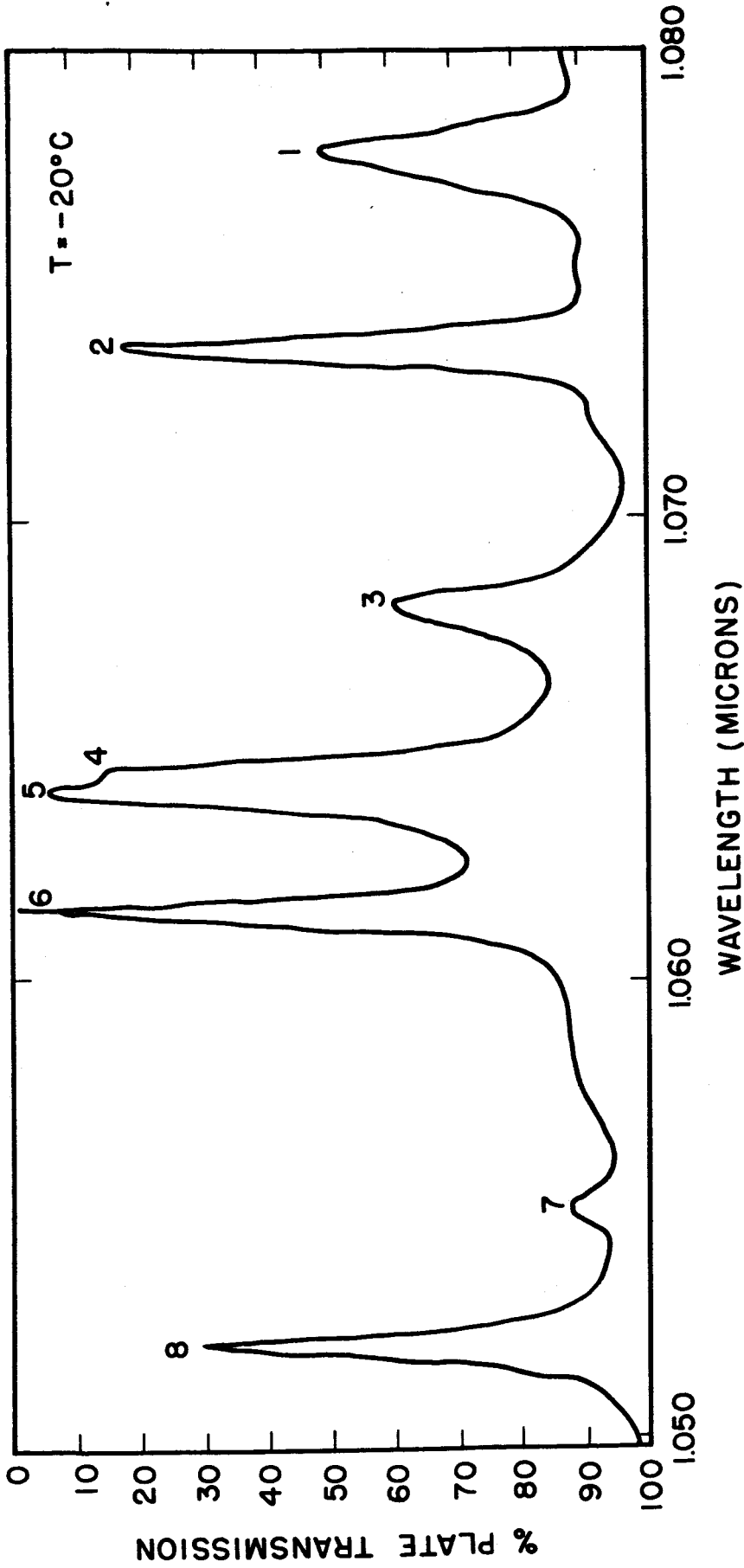


Fig. 1. Emission spectrum of Nd³⁺ in YAG at -20°C in the range 1.05 to 1.08 microns.

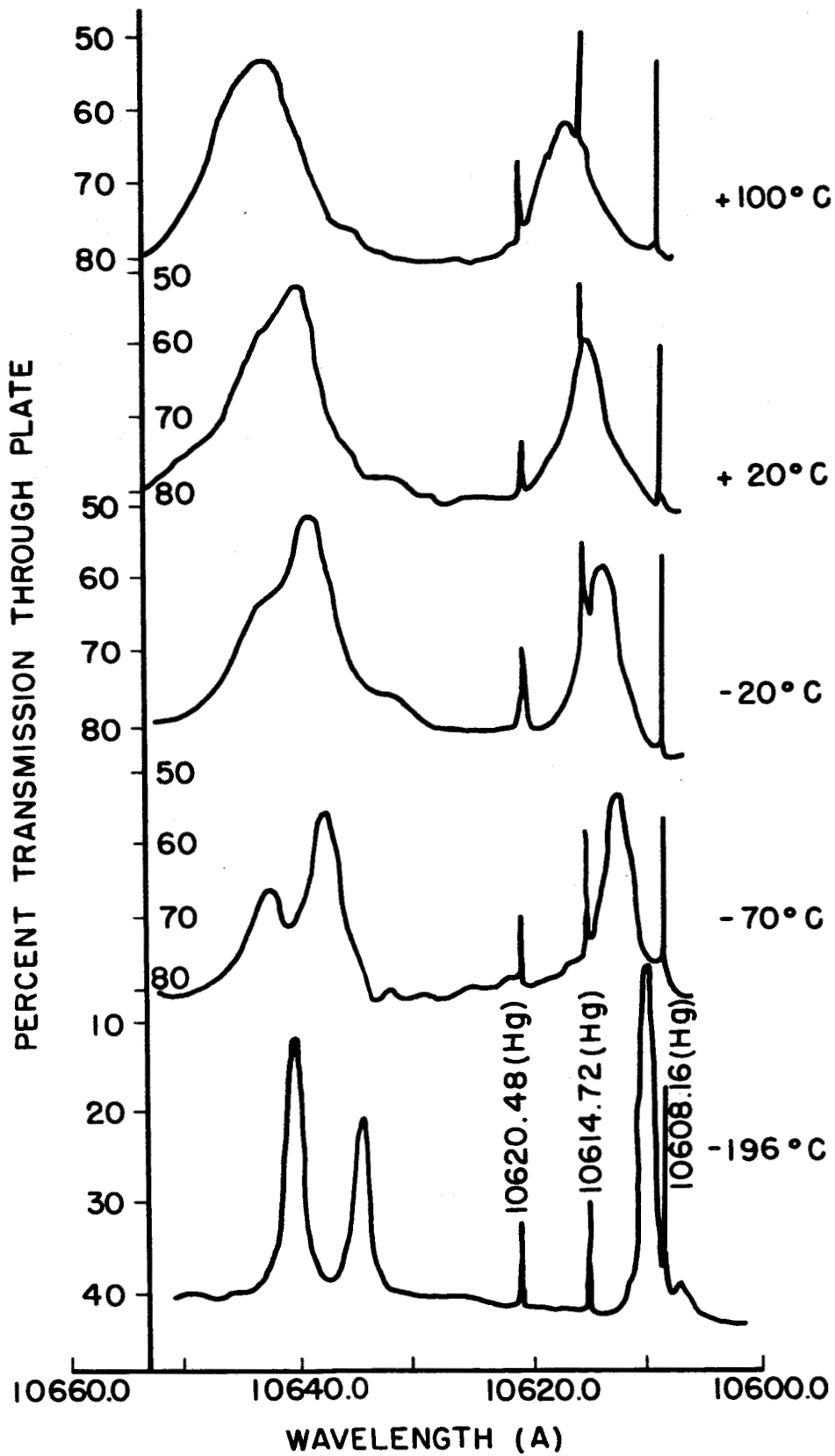


Fig. 2. Emission spectrum of Nd³⁺ in YAG for lines 4, 5, and 6 as a function of temperature. All exposures were identical except that at -196° C, which was considerably longer. Microdensitometer traces are from type 1-Z spectroscopic plates.

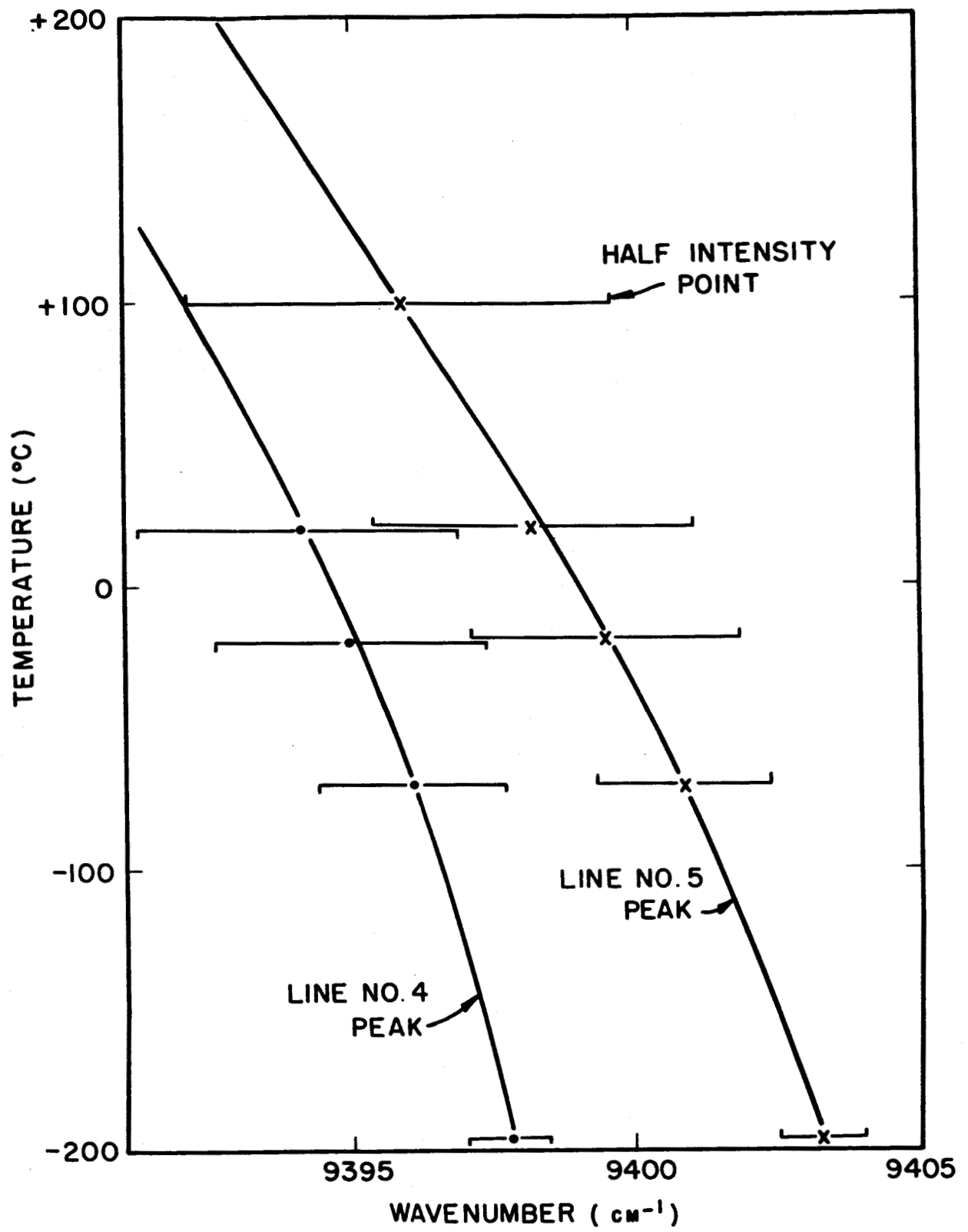


Fig. 3. Temperature dependence of emission peak and linewidth of lines 4 and 5.

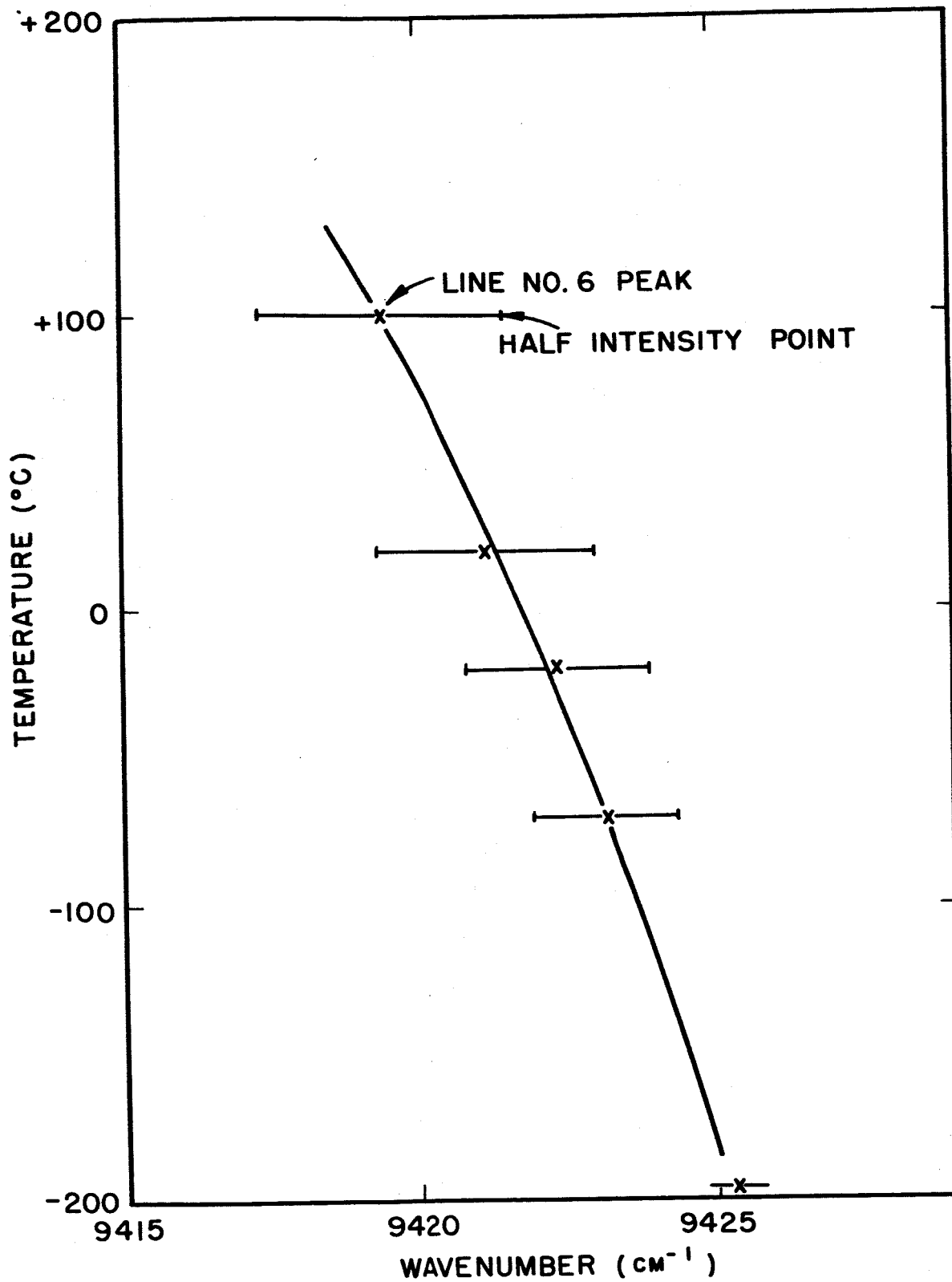


Fig. 4. Temperature dependence of emission peak and linewidth of line 6.

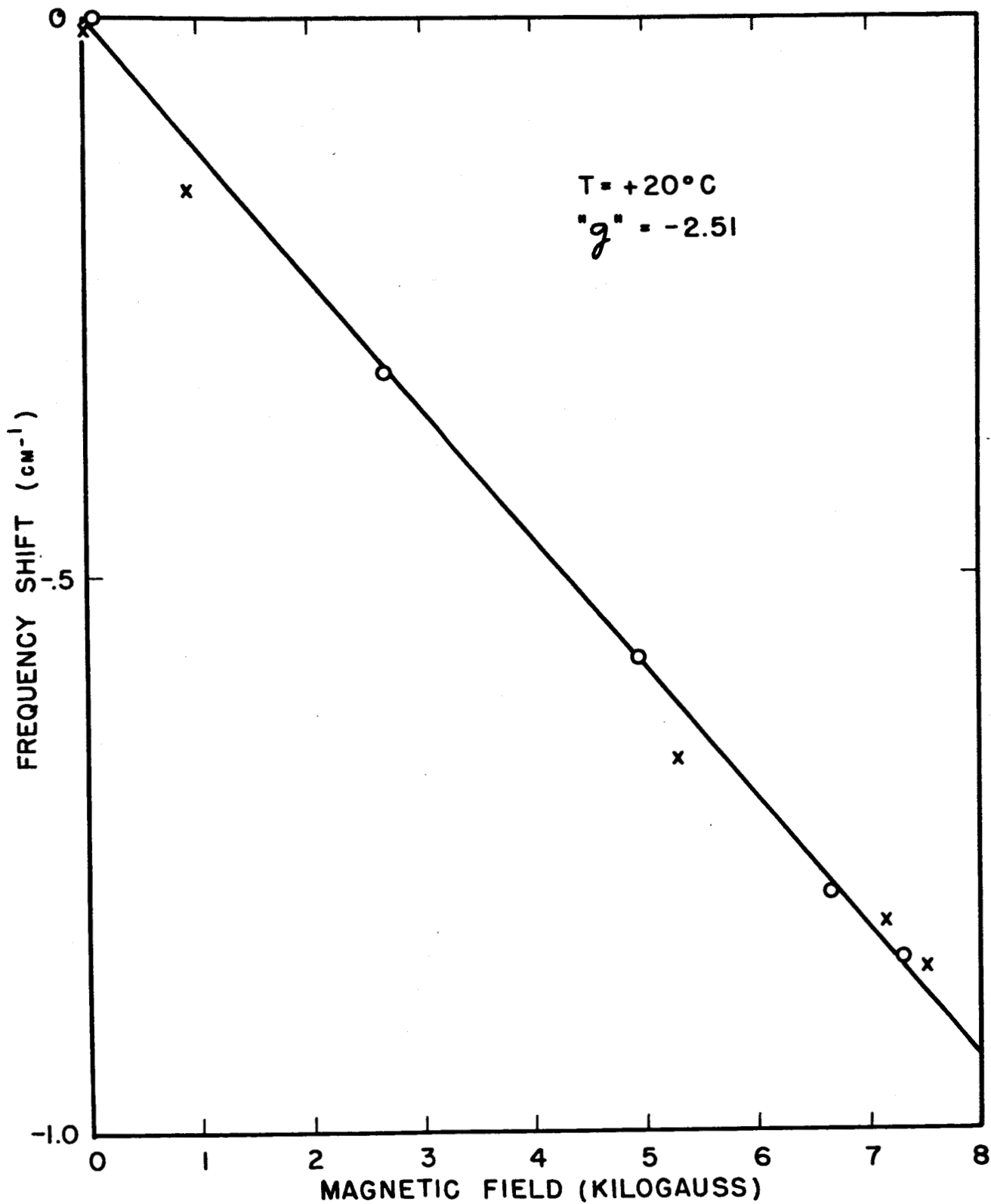


Fig. 5. Magnetic field shift of laser emission frequency for emission from line 5 at +20°C.

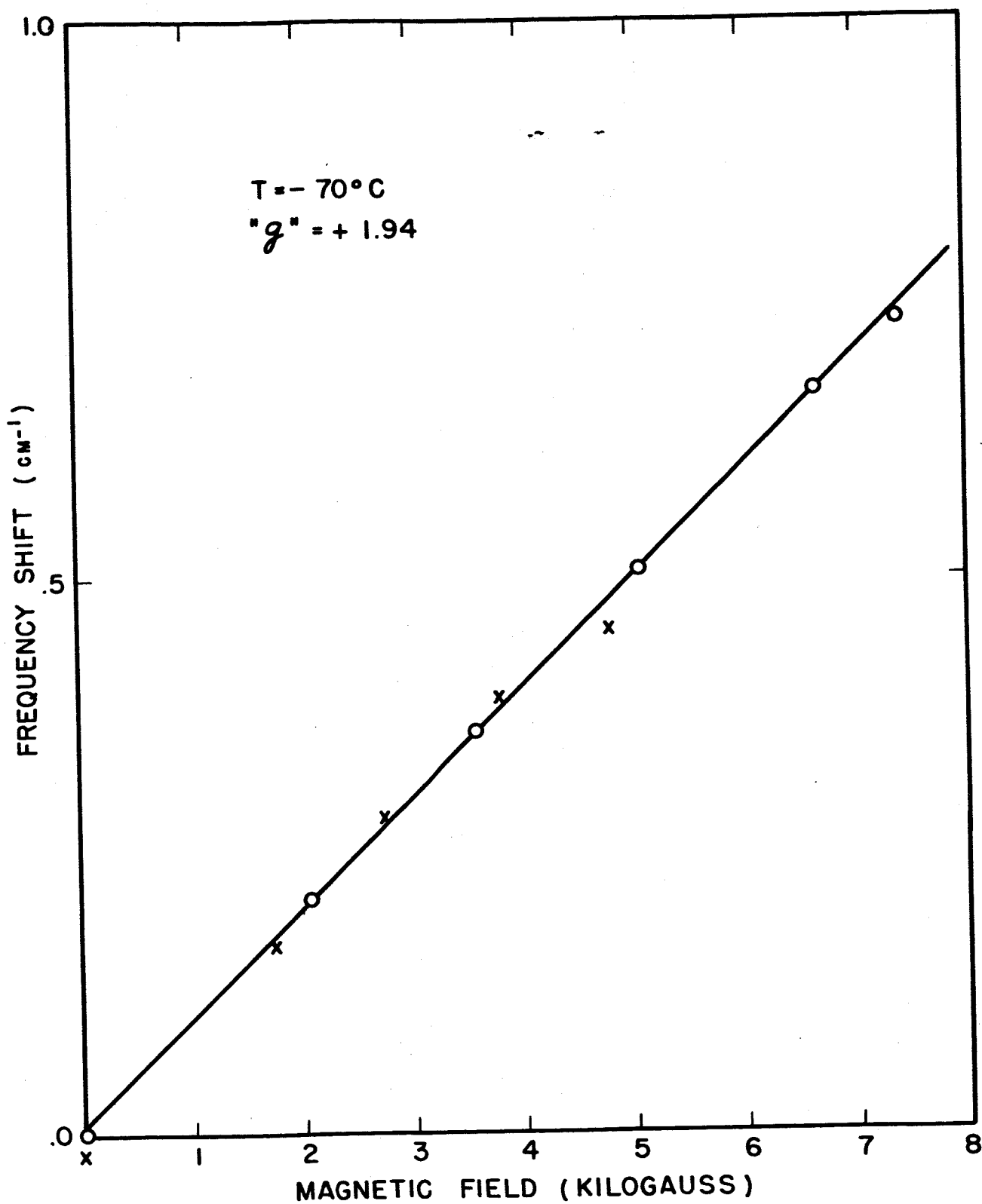


Fig. 6. Magnetic field shift of laser emission frequency for emission from line 6 at -70°C.