Measurement of the Laser Transition Cross Section for Nd^{+3} in Yttrium Aluminum Garnet*

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This paper reports measurements of the room-temperature laser transition cross section of Nd^{+3} in yttrium aluminum garnet at 1.06 μ. One method used is based on the measured upper-state lifetime and a determination of the fluorescence branching ratio for this state. The value thus determined is 3.5×10^{-19} cm^2. The second method involves measurement of the ratio of fluorescent intensities from the upper laser level to the ground state and to the lower laser level, and measurement of the cross section in absorption of the transition which involves the ground state. The second method yields a value of 2.7×10^{-19} cm^2. The difference represents the contribution of fluorescence to high-lying levels (not included in the determination of branching ratio) and nonradiative transitions. The probable experimental accuracy in both methods is $\pm15\%$.

INTRODUCTION

SINCE the first report of the operation of YAG:Nd^{+3} lasers[1] and indications that YAG (yttrium aluminum garnet) is a very suitable host for the Nd^{+3} ion, there has been considerable interest in the details of the energy-level structure of neodymium in this material. Koningstein and Geusic[2] have presented a comparison between crystal-field calculations and the energy levels deduced from absorption and fluorescence spectra. However, to date there has been no reported measurement of transition probabilities. A knowledge of the laser transition probability would be particularly useful. It is necessary to know this quantity to be able to determine analytically such laser system parameters as optimum mirror transmission, maximum output power to be expected, or maximum gain available.[3] In this paper we report the values of the room-temperature laser transition cross section as determined by two different methods.

In a three-level laser system such as ruby the cross section is easily determined by measuring the absorption coefficient at the laser wavelength and dividing by the number of atoms/cm^3 in the lower state of the transition. In ruby at room temperature this latter number is merely the Cr^{+3} concentration, since all of the population is in the two lowest levels, which are not resolved in the absorption line. However, in the four-level system of YAG:Nd^{+3} the laser transition at 1.065 μ terminates in a state ~2000 cm^{-1} above the ground state. Since the Boltzmann population of this state at room temperature is only $\sim e^{-10}$ of the ground-state population, no absorption can be observed at the laser wavelength. Therefore, less direct methods of determining the cross section must be found. One method used in this paper is based on a determination of the branching ratio for transitions from the upper level. Another method involves measurement of the ratio of fluorescent intensities from the upper laser level to the ground state and to the lower laser level, and measurement of the cross section in absorption of the transition which involves the ground state.

METHOD 1

If the radiative lifetime of the laser transition is known, then the transition cross section may be determined from the equation

$$\sigma=\lambda^2/8\pi^2\epsilon\tau_l\Delta\nu, \qquad (1)$$

where λ is the wavelength; σ is the transition cross section; τ_l is the radiative fluorescence lifetime associated with the laser transition; ϵ is the dielectric constant; $\Delta\nu$ is the linewidth. In the case of YAG:Nd^{+3}, determination of τ_l is complicated by the fact that the upper

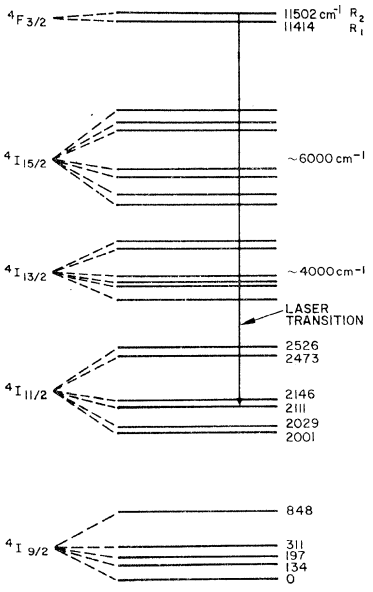

FIG. 1. Schematic diagram of energy levels of Nd^{3+} in YAG. Not to scale. In measurement of the branching ratio, transitions to the $^4I_{15/2}$ and $^4I_{13/2}$ multiplets were neglected.

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[3] J. K. Neeland and V. Evtuhov (to be published).

laser level $R_2(^4F_{3/2})$ is thermally coupled to the $R_1(^4F_{3/2})$ level 88 cm^{-1} away, and both levels decay to the $^4I_{15/2}$, $^4I_{13/2}$, $^4I_{11/2}$, and $^4I_{9/2}$ multiplets. This situation is shown schematically in Fig. 1. However, the radiative lifetime of the transition of interest may be computed from the branching ratio and the total radiative lifetime. The branching ratio may be determined by measuring the photon rates (intensity divided by frequency, denoted henceforth by I') for the various lines in the fluorescence spectrum.

Experimentally the measurement of branching ratio is complicated by the wide spread in wavelength of the fluorescence spectrum, from ~8700 Å to ~1.8 μ. Fortunately the transitions to the $^4I_{13/2}$ and $^4I_{15/2}$ multiplets are believed to be rather weak.[4,5] Therefore, for a first estimate the spectrum was analyzed only to 1.12 μ, which includes all of the $^4I_{11/2}$ and $^4I_{9/2}$ multiplets. The measurements were made using a cooled RCA 7102 photomultiplier and a Jarrell-Ash 0.5-m monochromator with a 15 000 line/in. grating blazed for 1 μ. Relative system sensitivity over the spectral region of interest was determined by use of a tungsten source of known temperature. Proper corrections were made for the emissivity of tungsten as a function of wavelength.

Assuming strong coupling between the R_1 and R_2 levels, the rate equation for the two levels together (neglecting nonradiative transitions) may be written

$$\frac{d(N_1+N_2)}{dt}=-N_1\sum_i S_{1i}-N_2\sum_{i\neq l} S_{2i}-N_2S_{2l}, \quad (2)$$

where the sum of S_{ai} represents the total transition probability for all transitions originating at state a, and the laser transition is displayed separately as N_2S_{2l}. If the R_1 and R_2 levels are in thermal equilibrium then

$$N_1=N_2/\kappa, \quad (3)$$

where $\kappa=e^{-\Delta E/kT}$ is the Boltzmann factor between the R_1 and R_2 levels (0.65 at room temperature). The rate equation for level 2 only is

$$\frac{dN_2}{dt}=-N_2S_{2\,\mathrm{tot}}, \quad (4)$$

where $S_{2\,\mathrm{tot}}$ is the reciprocal of the upper-state radiative lifetime (τ_{tr}). Combining Eqs. (2)–(4), we have

$$N_2(1+1/\kappa)S_{2\,\mathrm{tot}}=-N_1\sum_i S_{1i}-N_2\sum_{i\neq l} S_{2i}-N_2S_{2l}. \quad (5)$$

As each of these terms represents a photon rate I', we may write

$$\frac{N_2S_{2l}}{N_2(1+1/\kappa)S_{2\,\mathrm{tot}}}=\frac{I_l'}{I_{\mathrm{tot}}'} \quad (6)$$

or

$$\tau_l=\frac{\kappa}{1+\kappa}\frac{I_{\mathrm{tot}}'}{I_l'}\tau_{\mathrm{tr}},$$

where $S_{2l}=1/\tau_l$, $S_{2\,\mathrm{tot}}=1/\tau_{\mathrm{tr}}$, and I_t'/I_l' is the ratio of the photon rate of all transitions originating from either R_1 or R_2 to the photon rate for the laser transition only. In integrating the spectrum to obtain I_t'/I_l', the phonon-assisted transitions lying close in wavelength to the purely radiative transitions are automatically taken into account.

We have measured the lifetime of the R_2 level as ~236 μsec which agrees well with Geusic's value of 230 μsec.[5] If nonradiative transitions from $^4F_{3/2}$ are negligible, then this lifetime will be the total radiative lifetime τ_{tr}. The results of the relative line intensity measurements yield a value of 0.158 for I_l'/I_{tot}'. From Eq. (7) with $\tau_{\mathrm{tr}}=236$ μsec, $\tau_l=590\times10^{-6}$ sec.[6] Taking $\Delta\nu=7$ cm^{-1}, Eq. (1) gives $\sigma=3.5\times10^{-19}$ cm^2 at room temperature. The probable experimental accuracy is ±15%, excluding, of course, the limitations introduced by the initial assumptions.

It should be emphasized that the value above is really an upper bound on σ since in this method of measuring cross section, we are unable to include the effects of nonradiative transitions, or even to estimate their magnitude. Also, because of experimental difficulties, we neglected transitions to the $^4I_{13/2}$ and $^4I_{15/2}$ multiplets, as mentioned before.

METHOD 2

Because of the possible inaccuracies of the previous approach, a second method of determining σ was devised. In this method, the absorption coefficient for the transition from the ground state to the upper laser level (8700 Å) was measured on a Cary spectrophotometer. Dividing by the ground-state population (~½ Nd^{+3} concentration at room temperature) gives the cross section for this transition. This cross section was found to be 1.97×10^{-20} cm^2. Note that a knowledge of the dopant concentration is necessary here, but was not needed in the previous method. The value used here [Nd^{+3} in (1.0±0.1)% of Y^{+3} sites] is an average of the values quoted by the crystal supplier and determined by spectrographic analysis. Measurement of the ratio of peak photon rates between fluorescence from the upper laser level to the ground state and to the lower laser level gives the ratio of cross sections for these two transitions. In measuring the fluorescence ratio, care was taken to avoid self-absorption. Thus, a sample only 1 mm thick was used. Using the equipment described under Method 1, the ratio $\sigma_{1.06\,\mu}/\sigma_{0.87\,\mu}$ was found to be 13.8. This gives a value of 2.7×10^{-9} cm^2 for the laser

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[5] J. E. Geusic (to be published).
[6] A value of 560 μsec has been used in some calculations by J. E. Geusic, H. M. Marcos, and L. C. Van Uitert, in *Physics of Quantum Electronics*, edited by P. L. Kelly, B. Cox, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 725.

transition (with the same experimental accuracy as in Method 1). As this result is about 78% of the previously determined value, it suggests that nonradiative transitions plus transitions to the $^4I_{15/2}$ and $^4I_{13/2}$ multiplets make up about one-fifth of the total drain from the $^4F_{3/2}$ levels. The radiative laser-transition lifetime corresponding to the above cross section is 758 μsec.

Either of the above two techniques can also be used with other four-level systems. The second method, while undoubtedly more accurate, requires that a reasonable percentage of the upper-state population decay directly to the ground state (or to some state which has a reasonable Boltzmann population).

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Mössbauer Effect Following Coulomb Excitation in the Even-Even Isotopes of Ytterbium*

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Mössbauer levels in Yb^{172}, Yb^{174}, and Yb^{176} have been observed following the population of the first excited states by Coulomb excitation. Using oxide and metallic targets and absorbers, the quadrupole energy splittings and relative quadrupole moments were determined for each isotope. Coulomb-excitation cross-section data were used to determine the electric-field gradient at the nucleus for Yb^{3+} in Yb_2O_3.

I. INTRODUCTION

RECENT experiments have shown that Mössbauer effects can be observed when suitable nuclear levels are populated by Coulomb excitation[1–3] and by direct nuclear reactions.[4,5] The successful observation of the Mössbauer effect by accelerator-produced reactions significantly enlarges the list of possible nuclear levels in which the Mössbauer effect can be observed. The requirement that a nuclear level be populated in the decay of a radioactive parent with a suitably long half-life need no longer apply.

The rare-earth region contains many stable isotopes with suitable Mössbauer levels which are not accessible by β decay. Figure 1 shows seven stable ytterbium isotopes from Yb^{168} to Yb^{176} with eight possible levels for recoilless emission. Only a few of these states[6–8] have been studied by the conventional method using a radioactive parent. Of these seven isotopes, Yb^{171}, Yb^{172}, Yb^{174}, and Yb^{176} are sufficiently abundant in the naturally occurring ytterbium compounds to make use of enriched absorbers unnecessary. All of the possible ytterbium Mössbauer levels shown in Fig. 1 are collective, and most of them possess large Coulomb-excitation cross sections.[9]

Of special interest is the variation of the nuclear quadrupole moment and gyromagnetic ratio as one proceeds from one isotope to another of the same element. The present article deals with the observation of Mössbauer hyperfine spectra in the even-even isotopes Yb^{172}, Yb^{174}, and Yb^{176}, and the variation of the electric quadrupole moment among these isotopes. These data were compared with the Mössbauer spectrum for Yb^{170} which was obtained following β decay of Tm^{170}. Both oxide and metallic targets and absorbers were used.

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