

## Excitation Energy (arbitrary units)

FIG. 2. A plot of area under fluorescence decay curves for  $R_1$  and 7010 A in the same red ruby at 77°K as a function of excitation flash energy.

is not sufficiently greater than the threshold number needed for stimulated emission to exhibit the degree of lifetime shortening observed in pink ruby  $[compare$  Fig. 1(a) and Fig. 1(b).

These experimental results also yield information on the excitation processes of the coupled ions. Since more than half of the single ions are in the ground state even at high excitation, direct excitation via the broad absorption bands is not likely for those ion pairs near the center of the rod. On the other hand, thermal transfer of excitation between single ions and paired ions is ruled out by the saturation behavior of the 7010A line. One possibility which seems promising is radiative excitation by trapped 6934A light. As a preliminary check we have observed absorption lines in the red sample which overlap the 6934A line and have also observed the 7010A fluorescence when the red rod is excited by a pulse of 6934A light from a pink sample. Further experiments to clarify the nature of the excitation processes of exchange-coupled ions are in progress.

'I. Wieder, Proceedings Ann Arbor Conference on Optical Pumping, June, 1959 (unpublished).

<sup>2</sup>A. L. Schawlow, Proceedings of the Quantum Electronics Conference, Shawanga, New York, September, 1959 [Quantum Electronics, edited by C. H. Townes (Columbia University Press, New York, 1960)].

 $3I.$  Wieder, Phys. Rev. Letters  $3.468$  (1959).

4S. Geschwind, R. J. Collins, and A. L. Schawlow, Phys. Rev. Letters 3, 545 (1959).

 $5T.$  H. Maiman, Phys. Rev. Letters  $4, 564$  (1960). 6T. H. Maiman, Nature 187, 493 (1960); British

Communications and Electronics 7, 674 (1960).  ${}^{7}R.$  J. Collins et al., Phys. Rev. Letters 5, 303

(1960). <sup>8</sup>A. L. Schawlow, D. L. Wood, and A. M. Clogston,

Phys. Rev. Letters 3, 271 (1959).

 $A$ . L. Schawlow and C. H. Townes, Phys. Rev. 112, 1940 (1958).

## SIMULTANEOUS OPTICAL MASER ACTION IN TWO RUBY SATELLITE LINES

A. L. Schawlow and G. E. Devlin Bell Telephone Laboratories, Murray Hill, New Jersey (Received December 19, 1960)

We have obtained pulsed maser oscillations simultaneously at wavelengths of 7009 A and 7041 A in concentrated ruby. The lines involved are two of the strongest of the satellite or neighbor lines,<sup>1</sup> which have been known and studied for many years but are not yet fully analyzed. $2-5$ It is known<sup>5</sup> that these lines disappear at low concentrations, but appear strongly in emission at chromium concentrations of the order of 0.5%. In absorption, they are weak and disappear entirely at low temperatures.

Since these lines terminate on states which are far enough above the ground states to be empty at low temperatures and since their upper states can be populated easily by white light illumination, it was proposed that the strongest of them be used for an optical maser. $6$  A very low estimate of the fluorescence efficiency  $(0.1\%)$  was used, but with the advantage of an empty lower state, pulsed operation seemed possible at low

temperatures. Nom that the fluorescence efficiency for the main  $R$  lines is known to be nearly ciency for the main  $\kappa$  fines is known to be fountly,<sup>7</sup> it is apparent that much less pumping light is needed. Moreover, recent investigations of the 7009A line in absorption have shown that its lower state is about  $100 \text{ cm}^{-1}$  above the ground state. Thus liquid hydrogen or neon temperatures mould be sufficiently low to depopulate this lower state.

However, operation should also be possible at higher temperatures. This is expected because even at  $77^\circ$ K the line is so strong in emission, but so weak in absorption. It seems possible that the very numerous individual chromium ions absorb the pumping radiation and transmit it to the much rarer paired ions which emit the  $N$  lines.<sup>6</sup> Thus even if we have to invert the population of the N-line levels by pumping to empty the lower level, this can be accomplished with only enough pumping light to excite a small fraction of the isolated ions.

Maser operation was sought first in a ruby rod containing about  $0.5\%$  Cr<sub>2</sub>O<sub>3</sub>, having a diameter of 2 mm and a length of 4 cm. The ends of the rod mere silvered, one of them completely, and the other to a transmission of about 1%. The rod was immersed in liquid nitrogen in a small clear Dewar flask, with its upper end supported in a cone which allowed emergence only of light coming through the silver film. Pumping radiation was supplied by a General Electric  $FT524$ xenon flash tube. When a 400- microfarad capacitor bank was charged to 3500 volts and discharged through the flash tube, the relative intensity of the 7041A line was greatly enhanced over the other lines in the spectrum. At about 3700 volts it was joined by the 7009A line. Figure 1 shows these two lines photographed in a single flash at about 3900 volts. For comparison, part of the spectrograph slit was covered by a concentrated ruby at room temperature. When the latter mas illuminated by a microscope lamp, it gave a comparison spectrum similar to that of the ruby below the maser threshold except for a small wavelength shift of all lines.

The observation of enhancement thresholds is a strong indication of maser action.<sup>8</sup> Further confirmation was obtained by observation of parallelism in the output radiation, as shown by a sharply defined spot on a distant surface. The beam width, well above the pumping threshold energy, mas about 1 degree. This approximate parallelism implies considerable spatial coherence in the output.<sup>9</sup>



FIG. 1. (a) Spectrum of a concentrated ruby at room temperature with a 10-second exposure. (b) Spectrum of a ruby rod at 77'K showing N-line maser oscillations as described in the text, with a single flash lasting about 1/2000 second. All lines are shifted slightly because of the temperature difference between (a) and (b). The wavelength scale is in angstroms.

It may seem surprising that optical maser action occurs at two frequencies simultaneously. If the upper levels of the tmo lines are identical, or relax rapidly to each other, as with the  $\overline{E}$  and  $2\overline{A}$  levels of the isolated  $Cr^{3+}$  ions, this will not happen. In such a case, when maser action starts to empty one of the levels rapidly, it prevents the additional buildup of population needed for maser action from the other level. However, it had already been suspected that the 7041A line has an origin not closely connected with the 7009A and other neighbor lines. The evidence for this different origin is that the relative intensity of the two lines varies erratically from sample to sample. The intensity of the 7009A line varies with concentration in much the same way as most of its neighbors, whereas the 7041A line is usually weaker, but occasionally brighte<br>than the 7009A line.<sup>10</sup> Occurrence of simultathan the 7009A line.<sup>10</sup> Occurrence of simulta neous maser action is confirmation of their separate origin.

The pumping power needed for this sample is no less than that required for  $R_1$ -line maser action in dilute ruby. $8,9$  There are many ways in which the power required can be reduced. Of these, the simplest is to cool it further, and this is being studied. Preliminary attempts produced too much heat in the sample, so that its temperature rose far above that of the bath. Indeed, heating may have increased the threshold

power even when the sample was in liquid nitrogen. It is believed that more careful filtering of the light source to remove all but the useful pumping radiation will permit low- temperature operation at greatly reduced pump power.

It has thus been shown that optical maser operation at the wavelength of the 7009A line  $(N_2)$ does occur in concentrated ruby as predicted.<sup>6</sup> Moreover, simultaneous maser oscillations at 7041 A  $(N_1)$  show that these two lines are not closely related, and probably come from different kinds of chromium ion pairs.

We wish to thank W. L. Bond for polishing the ruby rod, D. F. Nelson for lending us the flash tube mount and power supply, and E. M. Kelly for silvering the ends of the rod. We are also indebted to W. S. Boyle for pointing out the importance of sample heating by the flash, and to A. M. Clogston and D. L. Wood for helpful discussions on the origin and properties of the satellite lines.

<sup>1</sup>Deutschbein labelled these lines  $N_1$  and  $N_2$ , where N stands for Nebenlinien. This usage has advantages now that the word "satellite" is used so often in its astronomical sense —even in connection with masers.  ${}^{2}C$ . E. Mendenhall and R. W. Wood, Phil. Mag. 30, 316 (1915).

<sup>3</sup>O. Deutschbein, Ann. Physik 14, 712 (1932); 14, 729 (1932); 20, 828 (1932).

4S. F. Jacobs, thesis, John Hopkins University, Baltimore, 1956 (unpublished) .

<sup>5</sup>A. L. Schawlow, D. L. Wood, and A. M. Clogston, Phys. Rev. Letters 3, 271 (1959).

 $6A$ . L. Schawlow, talk at Quantum Electronics Conference, Shawanga, New York, September 16, 1959 [Quantum Electronics, edited by C. H. Townes (Columbia University Press, New York, 1960)].

 $T$ . H. Maiman, Phys. Rev. Letters 4, 564 (1960).  ${}^{8}$ T. H. Maiman, Nature 187, 493 (1960); British Communications and Electronics, 7, 674 (1960).

 ${}^{9}R.$  J. Collins, D. F. Nelson, A. L. Schawlow, W. Bond, C. G. B. Garrett, and W. Kaiser, Phys. Rev. Letters 5, 303 (1960).

D. L. Wood and R. A. Ford (private communications) .

## INTERPRETATION OF THE  $\rm Fe^{57}$  ISOMER SHIFT

L. R. Walker, G. K. Wertheim, and V. Jaccarino Bell Telephone Laboratories, Murray Hill, New Jersey (Received January 3, 1961)

We have made a systematic study of the Mössbauer effect<sup>1</sup> of  $Fe<sup>57</sup>$  in di- and tri-valent iron compounds and in  $d$ -group metals. The observed "isomer shift" measures the total s-electron density at the nucleus. The shift in compounds is shown to depend mainly upon the  $3d$  configuration of iron involved and to a lesser extent upon the "chemical" bond. The Hartree-Fock calculations of Watson<sup>3</sup> on the various  $3d$  configurations of iron are combined with the data on the shifts in the most ionic compounds to obtain a calibration of the shift in terms of s-electron density. This enables us to estimate the difference in charge radius of the ground state and isomeric state of Fe<sup>57</sup>. We associate the shift of an Fe solute ion in a  $d$ -group metal with the addition of some fraction of a 4s electron to an Fe  $3d^7$  configuration; an estimate of the 4s-electron wave function density at the nucleus from the Fermi-Segrè-Goudsmit (FSG) formula<sup>4</sup> enables this fraction to be determined.

Kistner and Sunyar<sup>5</sup> first observed in the recoilfree emission and resonant absorption of the 14.4-kev nuclear gamma ray of  $Fe<sup>57</sup>$ , that the

energies of the emitted and absorbed gamma rays were noticeably different if the emitter and absorber were two dissimilar lattices containing iron. The origin of this effect is as follows.

The ground and isomeric levels of the nucleus have different effective charge radii; the electrostatic interaction with the electronic charge is then different in the two states and the gammaray energy is consequently changed (relative to its value for a point nucleus) by an amount proportional to the s-electron density at the nucleus. If the s-electron density is different for the absorber and emitter, the difference in gammaray energies,  $E_a - E_e$ , is defined as the isomer shift.

$$
E_{a} - E_{e} = \frac{2}{5}\pi Z e^{2} [R_{is}^{2} - R_{gr}^{2}] [\|\psi(0)_{a}\|^{2} - \|\psi(0)_{e}\|^{2}], (1)
$$

where  $R_{\rm \, is}$  and  $R_{\rm \, gr}$  are the radii of the isomeric and ground states and  $|\psi(0)_a|^2$  and  $|\psi(0)_e|^2$  are the total s-electron densities at the nucleus for absorber and emitter, respectively. $6$  It is to be noted that the sign of the shift has its origin in the fact that the level energy is lower the more



