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Summary of Dissertation

Optical Pumping of Ruby by Laser Resonance Radiation

by Alexander Szabo

工学博士学位論文内容要旨
学位論文題目

レーザー共鳴放射による ルビーの光ポンピング

アレキサンダー・サァボ

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Title : Optical Pumping of Ruby by Laser Resonance Radiation

Author : Alexander Szabo

A Introduction

Since introduction of the term "optical pumping" by Kastler,[1] its meaning has broadened somewhat, especially with the advent of the quantum electronics era and the laser. Generally optical pumping is a method of creating and maintaining a non-thermal population distribution of atoms in different quantum states. While early experiments were concerned with optical pumping of gases with resonance radiation, more recently the technique has also been applied to solids. In particular broad band pumping is now commonly used to excite solid state lasers. In this thesis, the resonant optical pumping of the ruby transition ${}^4A_2 \rightarrow {}^2E$ (R_1 line) by a ruby laser beam is examined. The study is focussed on two applications of such a resonant interaction, (1) laser pumping of microwave masers and (2) self-Q-switching of ruby lasers. Specific objectives in (1) were to measure the laser induced change, δ, in the ground state fine structure populations in ruby and to compare these with available theories and to construct new theoretical models where necessary. In particular hole-burning was incorporated into the theory for the first time and a new experimental technique introduced which directly demonstrates the inhomogeneous nature of the ruby R_1 line at low temperatures. In (2) a new technique of laser Q switching is described and good agreement between saturable absorber theory and experiment demonstrated.

B Laser-Pumped Ruby Maser

There are two primary advantages of optical pumping of microwave masers as first outlined by Hsu and Tittel[2] in the case of CW pumping, (1) low noise maser operation is possible at bath temperatures much higher than the usual $42^{o}K$ value which is required when microwave pumping is used and (2) maser operation can be extended to higher frequencies. In the case of ruby at $78^{o}K$ say, the latter analysis shows that CW low noise maser operation is not possible, essentially because of a low ratio, f/w, of optical to microwave relaxation rate. An extension of the analysis into the time domain is introduced in Chapter B2.1 which shows that, under pulsed conditions, low noise operation becomes possible during a time approximately equal to the microwave relaxation time following application of the pump. The theory was verified experimentally using the apparatus shown in Fig.1. A spin temperature $T_s = -12^{o}K$ was measured with ruby laser pumping at a microwave ruby temperature of $78^{o}K$. Since the maser noise temperature[3] $T_n \simeq |T_s|$, this demonstrated that low noise maser operation at high bath temperature was indeed possible[4,5] as predicted by the time dependent theory.

The feasibility of CW high frequency (300 GHz) operation of a ruby microwave maser using optical pumping is discussed in chapter B 2.2. Calculations indicate that the pump power requirements are quite reasonable ($\simeq$ 5 Watts/cm^2). Another advantage of optical pumping is pointed out, namely that because of of the spin selection rules, 3 level maser operation using only the ground state Zeeman levels is not possible at the high magnetic fields required for mm wave operation. However optical transitions to the ^2E electronic state are allowed and in fact constitute the only way to achieve maser action in ruby at mm wavelengths.

In chapter B 3, a new technique (internal optical pumping) is described for overcoming an unfavourable f/w ratio by combining laser and maser action in the same crystal. Unlike the external pumping scheme of Hsu and Tittel,[2] internal pumping allows the attainment of a low $|T_s|$ in ruby, at 78°K for example, under CW conditions. The calculated pump power for such operation in ruby however is probably impractically high and other materials such as F centres in alkali halides which have much longer microwave relaxation times at higher temperatures are suggested as being more promising.

C Optical-Microwave Double Resonance Studies of Ruby

Comparison of experiment and the simple theory of chapter B showed that an order of magnitude larger pump energy was required in practice than predicted by theory. Such a discrepancy had also been previously noted by Devor et al[6] and was ascribed to a lack of frequency match between the ruby laser and absorption in the microwave ruby. To test this idea, a careful measurement of the laser and microwave ruby line positions was undertaken using direct optical spectroscopy as well as an optical-microwave double resonance technique[7]. This technique and theoretical extensions arising from these experiments are summarized below.

In the theoretical extension, consideration is given to three main factors neglected in the simple theory. These are, the finite optical linewidth which results in pumping of all the ground state levels in varying degrees depending on the laser frequency, the effect of hole-burning in the inhomogeneously broadend R_1 line at low temperatures and finally the fact that the microwave ruby is optically thick, that is the pump power varies with distance in the crystal. The apparatus used is shown in Fig. 1 with a modification which allowed variation of the sample temperature from 4.2 to 100°K. The cavity resnance frequency was 9.4 GHz. When the microwave ruby is irradiated by the pulsed laser beam, a change in the cavity reflection coefficient occures due to a change, δ, in the population difference in the energy levels. Experimental and theoretical values of this change are shown in Fig. 2 for the $1/2 \leftrightarrow -1/2$ transition (magnetic field is parallel to the crystal axis) as a function of the crystal temperature. Since the optical energy level spacing is temperature dependent, δ goes through positive and negative values as the laser pumps the upper or lower microwave levels. Of particular interest are the zero crossing points at 46°K and 87°K at which points one of the two lasers lines lies midway between the optical transitions from the +1/2 and -1/2 $^4A_2$ levels. The measurement of these zero crossing points allowed a precise determination of the relative positions of the laser and absorption lines. These positions were in good agreement with direct measurements using a large (35 foot) grating spectrograph. The solid line in Fig. 2 is a theoretical calculation valid for low pump powers. Agreement with experiment is reasonable except at higher temperatures where spin-lattice relaxation effects become important. At higher pump energies (x in Fig. 2), the hole-burning theory agreed with experiment within 20 % at 4.2°K.

However the large change in δ which occurs in the temperature range 10 to 40°K was quite unexpected, since in this range, negligible change in the absorption frequency occurs.[8] Factors such as temperature-dependent spectral diffusion and transparency effects were theoretically examined, however they were found to be too small to explain the results. This anomalous behaviour was finally explained as due to a thermalization effect in the ^2E levels which, under saturation conditions, allows additional ions to be pumped into the excited state. Calculations of this effect were in good quantitative agreement with experiment.[7]

D Laser Induced Fluorescence Line Narrowing in Ruby

An important conclusion of the previous chapter was that under optical saturation conditions, "hole-burning" occurs in the R_1 line when it is excited by a laser whose frequency width is less than the R_1 inhomogeneous linewidth. In other words, only those ions which are resonant with the laser will saturate, while the rest of the line is unaffected. The evidence however is indirect as indeed are other observations such as the temperature dependence of linewidth.[8] In this chapter, the inhomogeneous nature of the R_1 line at low temperatures is directly demonstrated using a technique of laser-induced fluorescent line-narrowing.[9]

The apparatus used is shown in Fig. 3. A liquid nitrogen cooled ruby laser resonantly excites the R_1 line, after which the fluorescence is frequency-analyzed by a high-resolution scanning Fabry-Perot interferometer. Initial results showed a narrowing from the inhomogeneous width of 2.2 GHz to a value of 600 MHz. More recent results[9] have resulted in values as low as 60 MHz, which is approaching the homogeneous width of ≃ 10 MHz implied by photon-echo studies in zero magnetic field.[10] The latter width arises from the Cr-Al^{27} electron-nuclear spin dipole interaction. An interesting aspect of the observations is that no spectral diffusion occurs for times up to 10 msec after excitation. These results support Birgeneau's[11] recent calculations which show that earlier postulated multipolar interactions do not produce single-ion to single-ion energy transfer in concentrated ruby but rather that the interaction must be of a short range type such as exchange.

E Self-Q-Switching of Ruby Lasers

A novel technique for producing high power optical pulses from a ruby laser is described.[12] It is shown that when Cr ions in an unpumped portion of a ruby laser rod resonantly interact with laser radiation produced by the pumped portion of the rod, then rapid saturation of the unpumped ions occurs resulting in a corresponding rapid decrease of the optical cavity loss. As is well known, such a loss switching mechanism in lasers results in a form of operation called Q-switching. In the present case, the term self-Q-switching is introduced since the usual optical switch such as a Kerr or Pockel cell external to the laser rod is not required. An essential feature of the self-Q-switching mechanism is that a non-uniform cavity photon density is required to obtain saturable absorber switching[13] by unpumped laser atoms. This was achieved by the use of a rooftop laser rod in which the photon density is doubled in the volume contained in the rooftop because of folding of the beam on itself. When the rooftop was shielded from the pump, a Q-switched laser pulse of peak power 200 KW and width 6 nsec was produced. This operation only occurred at liquid nitrogen temperatures, ceasing at temperatures above 150°K. The Q-switching behaviour and its temperature dependence are quantitatively explained by saturable absorber giant pulse theory.[13] A plot of theoretical and experimental pulse shapes is shown in Fig. 4. The theoretical curve was adjusted to the experimental points by variation of the theoretical parameters. A comparison of the latter parameters with values deduced from experimental conditions showed good agreement.[12]

F Conclusions

The main conclusions and accomplishments of this work are listed below.

(1) Theoretical prediction that laser pumping of microwave masers could produce spin temperatures (absolute value) much less than the bath temperature was experimentally verified. For a ruby system, a spin temperature of -12°K was measured[4,5] at a bath temperature of 78°K.

(2) A difficulty which arises with ruby is that at higher temperatures, the optical to microwave relaxation time ratio becomes unfavourable for CW maser action using optical pumping. A method of circumventing this problem was suggested and a detailed theory outlined.

(3) Quantitative comparison between theory and experiment of the effects of laser pumping on the ground state populations of ruby showed that several factors not considered in previous work[6] required consideration. The most important of these were line-overlap, hole-burning and thermalization in the ^2E levels. In addition a double-resonance

technique was developed to precisely locate the laser frequency relative to the absorption frequencies.[7]

(4) A limiting factor in the resolution of sharp lines in gases and solids by conventional spectroscopy is inhomogeneous broadening. Doppler inhomogeneous broadening in gases has recently been overcome by a laser saturation technique.[14] In this thesis a laser technique was described which, for the first time has overcome inhomogeneous broadening effects in a solid.[9]

(5) A new technique of Q-switching ruby lasers was reported which involved saturation of an unpumped portion of the ruby rod.[12]

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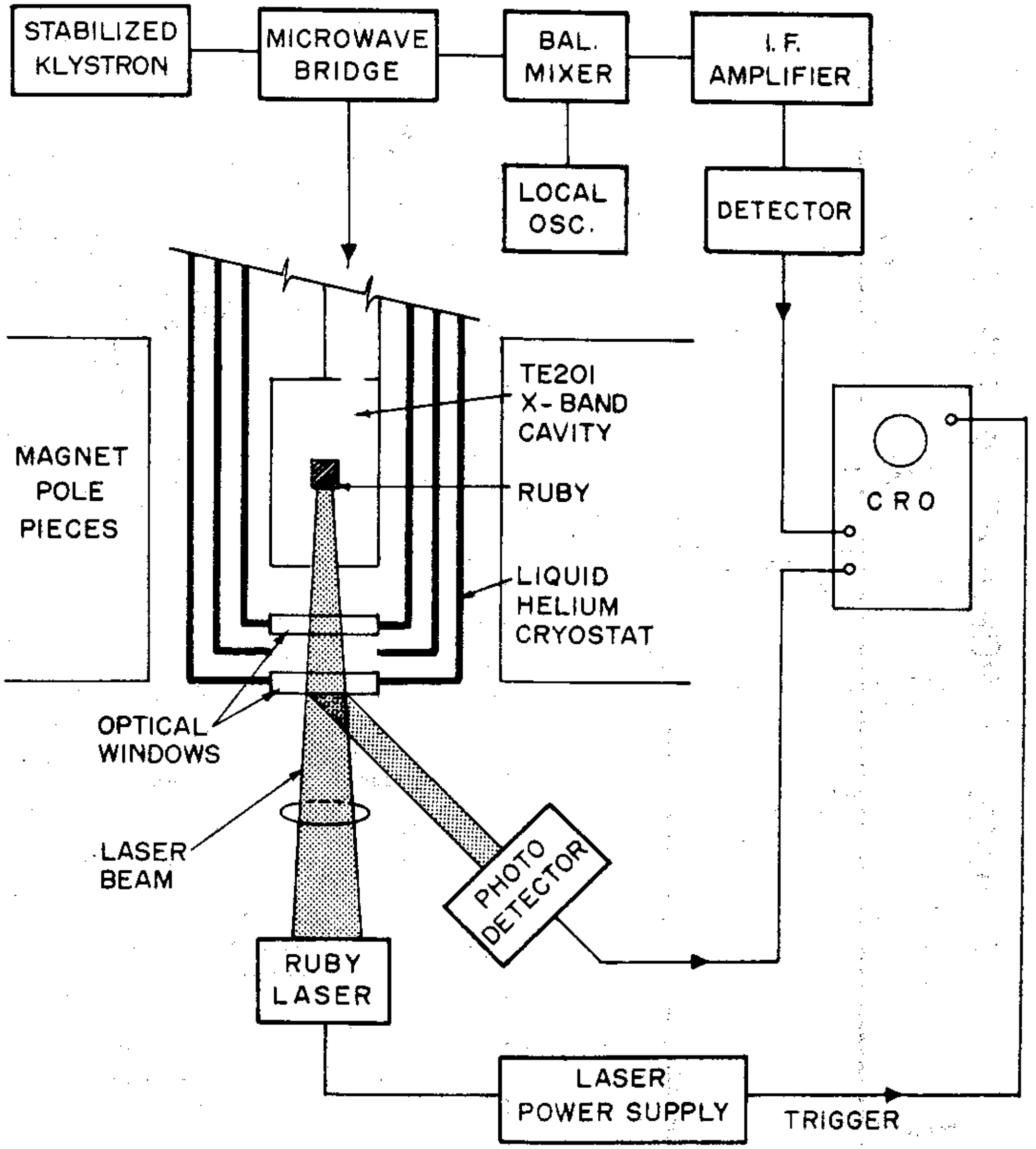

Fig.1 Schematic of apparatus used for experiments on laser-pumped maser and optical microwave double resonance studies of ruby.

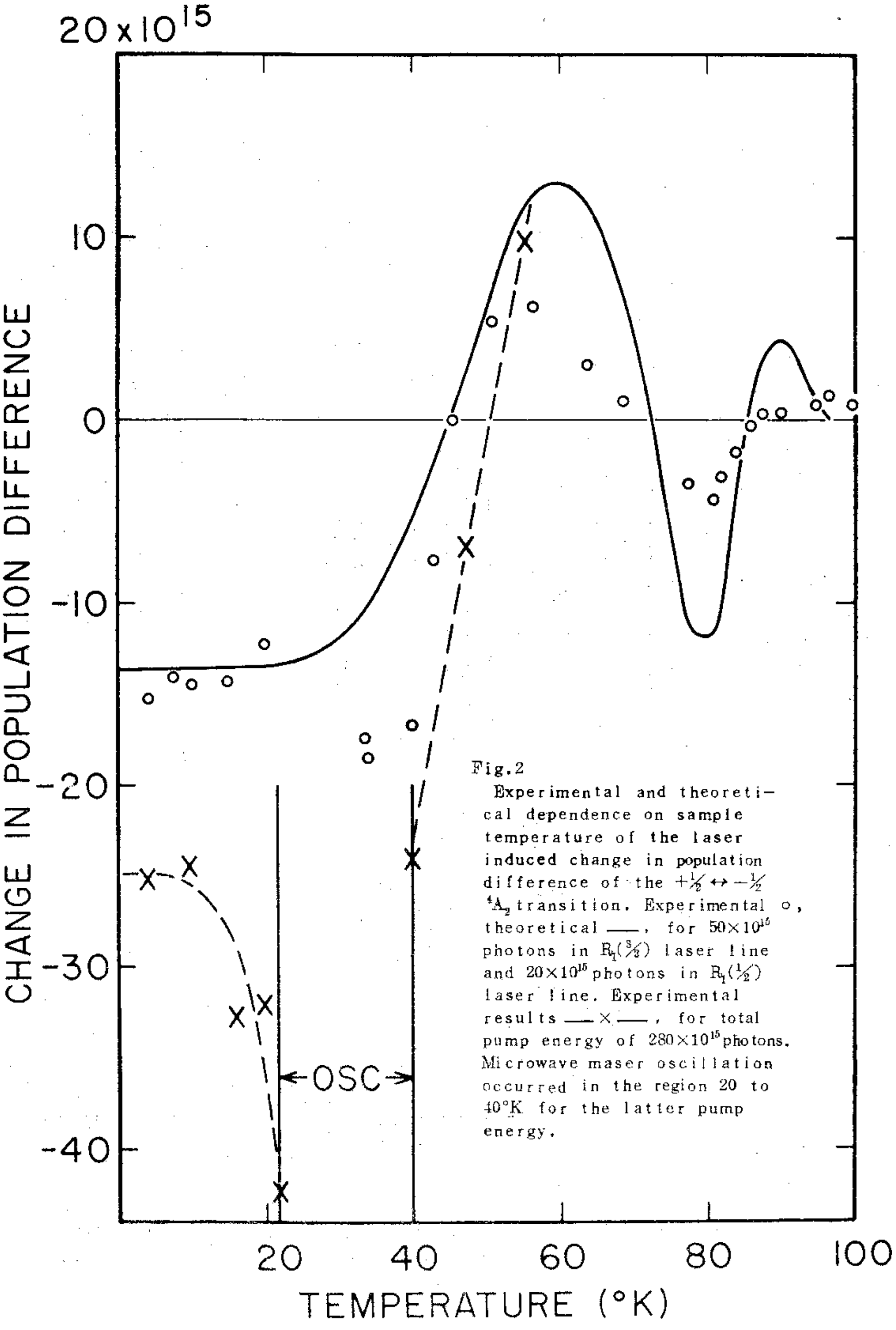

Fig.2

Experimental and theoretical dependence on sample temperature of the laser induced change in population difference of the $+\frac{1}{2} \leftrightarrow -\frac{1}{2}$ $^4A_2$ transition. Experimental ○, theoretical ——, for 50×10^{15} photons in $R_1(\frac{3}{2})$ laser line and 20×10^{15} photons in $R_1(\frac{1}{2})$ laser line. Experimental results —×—, for total pump energy of 280×10^{15} photons. Microwave maser oscillation occurred in the region 20 to 40°K for the latter pump energy.

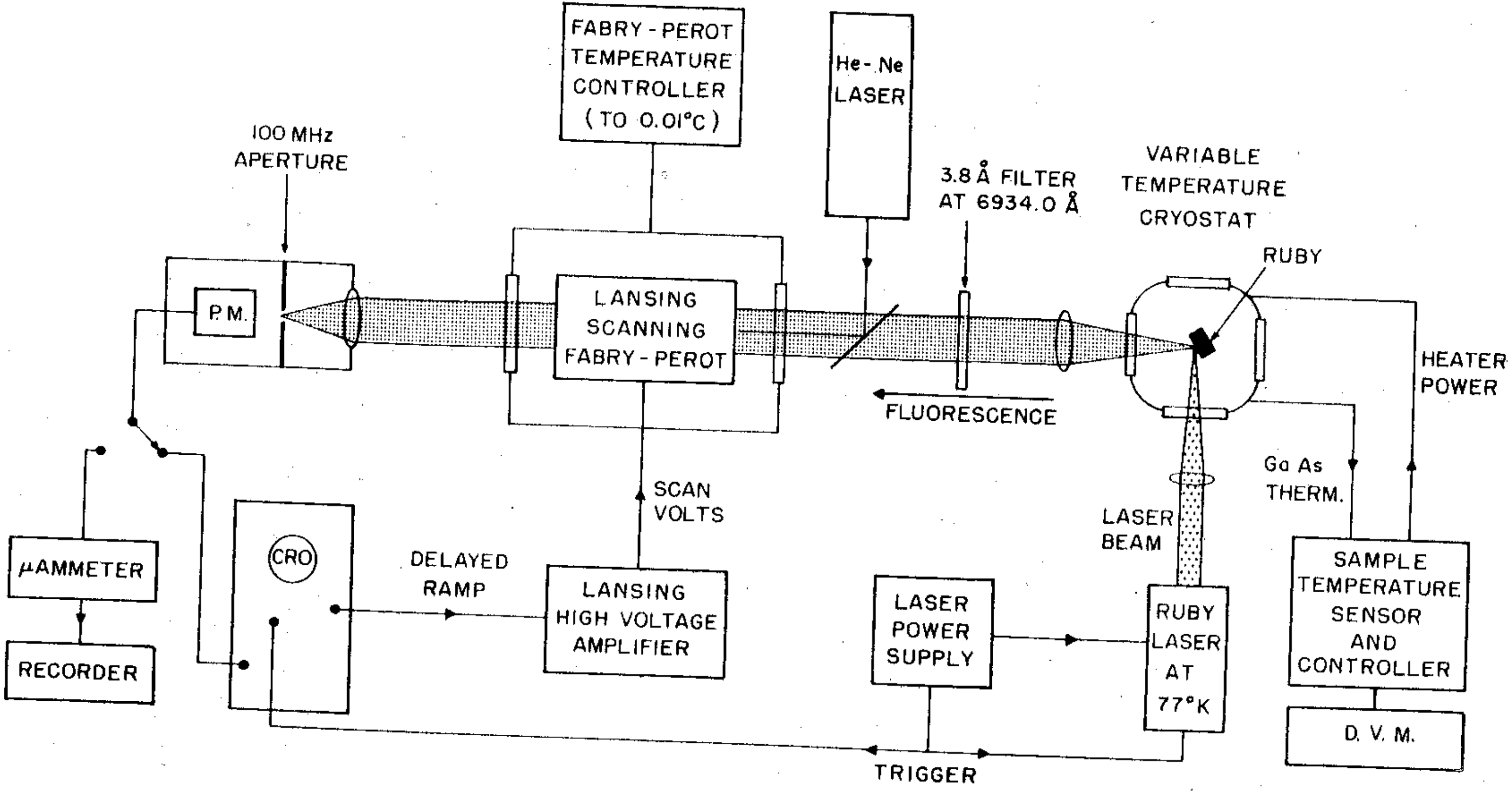

Fig.3 Schematic of apparatus used for laser induced fluorescent line narrowing experiments.

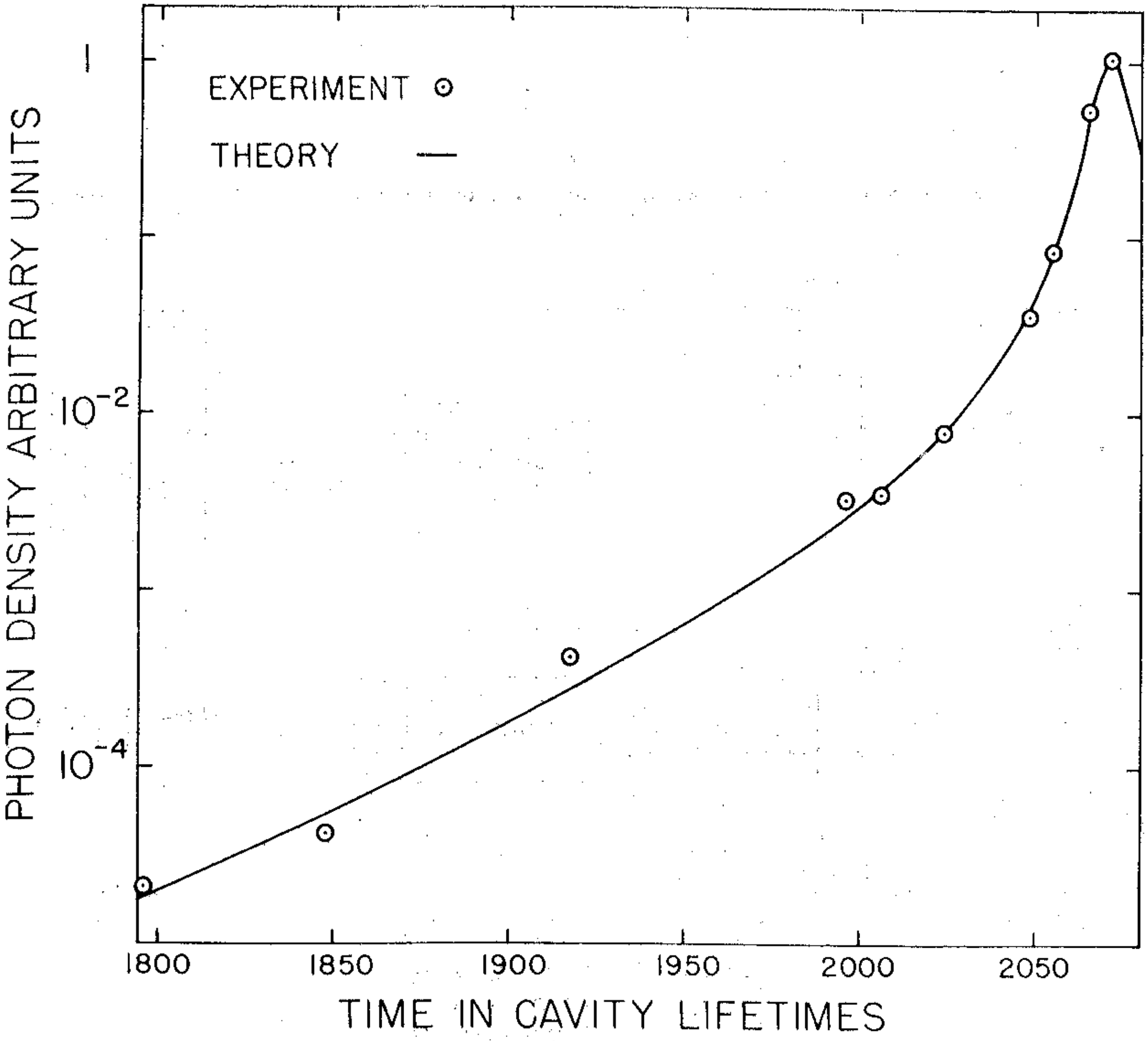

Fig.4 Experimental and theoretical time development of self -Q- switched laser pulse.