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SPONTANEOUS APPEARANCE OF PICOSECOND PULSES IN RUBY AND Nd:GLASS LASERS

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Picosecond light pulses have been observed to appear spontaneously in ruby and Nd:glass lasers Q-switched by means of a rotating mirror. This striking phenomenon is explained on the basis of coherence effects. The implication is that lasers with broad linewidths will quite generally tend to oscillate as pulse-regenerative oscillators.

We have observed the spontaneous appearance of picosecond (10^{-12} sec) duration light pulses from a ruby laser and a neodymium:glass laser Q-switched at room temperature by means of a rotating mirror. Two-photon fluorescence pulse collision and speed-up techniques[1,2] have been used to display these pulses directly. Figure 1(a) is a photograph of the blue fluorescence produced in a 1.8-cm-long cell of dibenzanthracene in benzene when a beam of picosecond pulses at 0.53 μ, entering the cell from the left, is reflected upon itself by a mirror located at the right end of the trace. The green light pulses at 0.53 μ are derived from the 1.06-μ output of the Nd:glass laser by phase-matched second harmonic generation in KH_2PO_4. The individual pulses each have a duration of less than 1.0 psec; they emerge from the laser in the form of trains of 5-10 pulses 67 psec apart, giving rise to the uniformly spaced spots in Fig. 1, as in the experiment of Giordmaine, Rentzepis, Shapiro and Wecht.[1] Brighter fluorescence spots occur where the picosecond pulses reflected from the mirror overlap the oncoming ones. The spot width is a direct measurement of the pulse duration. The 67-psec spacing of the pulses corresponds to twice the width of an interferometer flat used as the output reflector of the laser. The flat had a 80% reflective dielectric coating on the side facing the laser rod. Note that for our laser the round-trip transit time $2L/c$ was 3.3 nsec, where L is the effective length of the cavity and c the speed of light.

Similar but longer fluorescence spots were also produced in a benzene solution of 9,10-diphenylanthracene by the output beam of a Q-switched ruby laser constructed in exactly the same manner as the Nd:glass laser described above. The ruby laser pulses were found to have an average duration of approximately 10 psec.

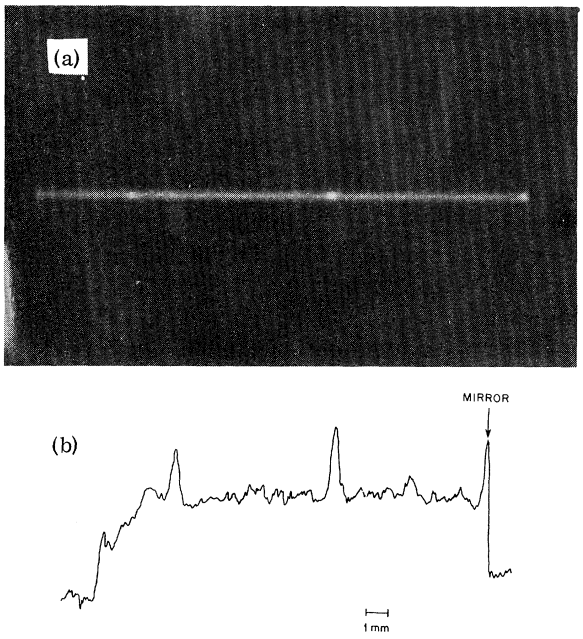

FIG. 1. (a) Fluorescence trace produced by a train of picosecond pulses spaced 67 psec, entering the cell from the left, and reflected upon itself by a mirror (not visible) located at the right end of the track. The pulses are derived from a Nd:glass laser, Q-switched by a rotating mirror. (b) Photodensitometer tracing of the fluorescence tracks. Numerical data on the fluorescent intensities observed will be found in Ref. 1.

The output power waveforms as observed with a fast (0.3-nsec risetime) photodiode and Tektronix 519 oscilloscope (overall system risetime 0.5 nsec) were found to have the usual "hill" shape,[3] about 20 nsec at half-height with a generally small ripple at the cavity round-

trip frequency. The oscilloscope waveform gave little evidence of picosecond pulsing. However, the two-photon fluorescence displays showed the picosecond spots for every firing of the ruby and Nd:glass laser. Our interpretation is that several groups of picosecond pulses are traveling back and forth in the cavity at any given time, and are not resolved by the oscilloscope.

Picosecond light pulses have previously been seen in Nd:glass laser Q-switched by the Kodak dye No. 9740. Contrary to previous speculations,[4,5] it is now very clear from our experiments that the dye is not essential to the appearance of picosecond pulses. We believe that the development of short high-power pulses is intrinsic to the active laser medium, and that these pulses appear in all Q-switched lasers. This means that most Q-switched ruby and glass lasers used in the past, with the exception of those especially constructed to sustain a single mode, have been producing picosecond pulses for years, unbeknownst to the experimenters!

We believe that the appearance of picosecond pulses in ruby and Nd:glass lasers has to do with coherent dynamic effects, and our explanation for this phenomenon follows naturally from the calculations of Wittke and Warter[6] and others.[7-9] It is in the same spirit as the suggestion by Fox and Smith[10] that the 0.5-nsec pulses occurring in the He-Ne self-locked laser are 180° pulses. We refer to effects which take place in a two-level system when a strong coherent light pulse of duration short compared to T_1 and T_2 is incident upon the system, where T_1 and T_2 are respectively the longitudinal and transverse atomic relaxation times. For both the ruby and the Nd:glass systems, T_1 is approximately 1 msec and T_2, the inverse homogeneous linewidth, is of the order of 3 psec at room temperature.[11,12]

The geometric representation of the Schrödinger equation adapted by Feynman et al.[13] from nuclear magnetic resonance to the general two-level problem is very useful in visualizing the effect of coherent pulses. The vector $\vec{r}$ (or $\vec{R}$) is a measure of the population difference between upper and lower laser states. For a fully pumped system, $\vec{r}$ is initially a unit vector along the axis III [see Fig. 2(a)].

Under the action of a light pulse, the motion of $\vec{r}$ for an atom on resonance is a precession about a vector $\vec{\omega}$ directed along the axis II (see Fig. 2), the rotation angle being proportional to the time integral of the electric field strength.

The rapid growth of picosecond pulses in Q-switched lasers can be understood qualitatively on the basis of the following simple argument. As laser oscillation builds up from noise, parts of the complex wave form are likely to be shorter and more intense than other parts. Now, as shown in Fig. 2, the action of a pulse shorter than T_2 is to rotate $\vec{r}$ by a certain angle. On the other hand a pulse longer than T_2 will have its action on $\vec{r}$ interrupted and hindered by the random phase changes (rotations about III) caused by phonon processes. The shorter pulse therefore stimulates more atoms to radiate and grows faster. In a very long amplifying column the pulses would grow until they became 180° pulses.[6,7] In our lasers their growth is limited by the losses at the mirrors and the gradual depletion of the inverted population. The Nd:glass and ruby rods were 7.6 cm long and 0.95 cm in diameter. The maximum energy in the 1-psec pulses of the Nd:glass laser was about 0.5×10^{-3} J, and about 1.0×10^{-3} J for the 10-psec ruby laser pulses. Taking into account the 20% transmission of the output mirror and strong inhomogeneities in the laser beam due to filament formation in the rod, we estimated that the peak power densities inside the cavity were of the order of 2 GW/cm^2 for the ruby laser and 10 GW/cm^2 for the Nd:glass laser. Using radiative lifetimes of 5 msec for the ruby and 1 msec for the Nd transitions, and formula (7) of Ref. 13, one finds that the largest pulses seen caused rotations on the $\vec{r}$ vector of the order of 20° for

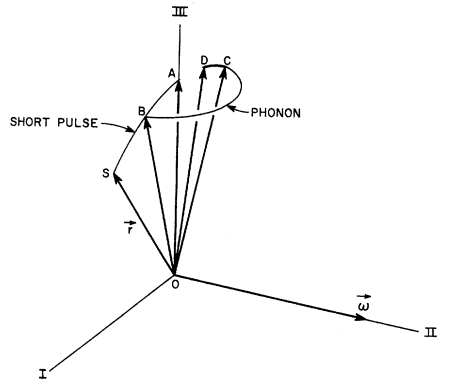

FIG. 2. The behavior of $\vec{r}$ under a short pulse is a precession from A to S about $\vec{\omega}$. For a long pulse a phonon process interrupts the precession at B and suddenly moves $\vec{r}$ to C, whereupon $\vec{r}$ precesses to D.

both lasers.

Spectra were taken of the output beams and showed a linewidth of about 3 cm^{-1} and 100 cm^{-1} for the ruby and Nd:glass lasers respectively; these linewidths are compatible with the 10-psec and 1-psec pulses seen in the ruby and Nd:glass lasers, respectively.

We believe that the conclusion to be drawn from these observations and the theory[6-9] is that lasers with broad linewidths will quite generally tend to produce short pulses. Also, it is clear that many experiments, especially nonlinear experiments, done in the past with "ordinary" Q-switched lasers, have to be re-evaluated in terms of picosecond pulses of very high peak powers.

We wish to express our gratitude to Dr. C. G. B. Garrett and Dr. D. E. McCumber for very helpful discussions, and to Mr. K. W. Wecht for technical assistance.

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EXCHANGE AND THE "10/3 EFFECT" IN $Cu(NH_3)_4SO_4 \cdot H_2O$ †

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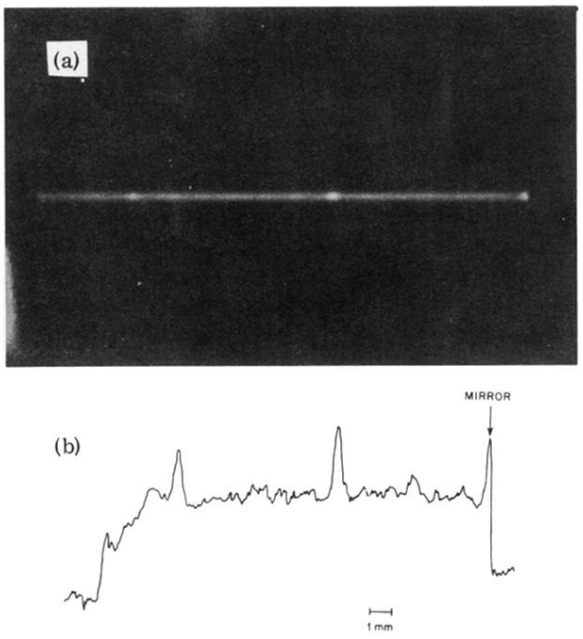
Observations and calculation of the "10/3 effect" in $Cu(NH_3)_4SO_4 \cdot H_2O$ confirm a non-Gaussian form for one-dimensional spin time correlation functions.

"The esoteric phenomenon of the 10/3 effect"[1] has been a part of the literature of electron paramagnetic resonance (epr) since the original discussion by Anderson and Weiss in 1954.[2] Only recently, however, has a quantitative experimental verification of the effect been published (for measurements in $K_2CuCl_4 \cdot 2H_2O$).[3] In this Letter, observation and calculation of the effect in the magnetic linear chain material $Cu(NH_3)_4SO_4 \cdot H_2O$[4] are reported. The resulting agreement gives strong evidence that there exists a marked distinction between the one-dimensional spin time correlation functions and the commonly assumed Gaussian form.

The "10/3 effect" (more accurately described as "nonsecular line broadening"), in brief, refers to the increase in magnetic-resonance linewidth that occurs in an exchange-narrowed material due to the inclusion of nonsecular spin-spin terms when the Larmor frequency becomes of the order of or less than the effective "exchange frequency." Using the theory of Kubo and Tomita,[5,6] the frequency dependence of the dipolar linewidths of an exchange-narrowed paramagnetic salt may be written approximately as

$$\Delta H = \Delta H(\infty)[1 + \beta_1 j(\omega) + \beta_2 j(2\omega)], \quad (1)$$

where $\Delta H(\infty)$ is the infinite-frequency (secular) value of the linewidth, β_1 and β_2 give the relative nonsecular contributions (i.e., $\Delta M = \pm 1$ and $\Delta M = \pm 2$) to the linewidth (for CTS, $\beta_1 = 0.69$ and $\beta_2 = 0.65$),[5,7] and $j(\omega)$ is obtained from the time correlation function of the exchange-mod-



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