STIMULATED PHOTON ECHO
&
PHOTON ECHO RELAXATION MEASUREMENTS

Diploma paper by
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ABSTRACT

The first part of this diploma paper gives a survey of a concept for fast high-density optical information storage and processing using the so-called stimulated photon echo.

The second part describes photon echo relaxation measurements made on the $^3H_4-^1D_2$ transition in 0.1% $Pr^{3+}:YAlO_3$ in external magnetic fields. These measurements were performed at the Department of Atomic Physics, Lund Institute of Technology during the first part of 1991 in order to investigate the magnetic field dependence of the photon echo decay in the crystal at liquid helium temperatures.

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1. INTRODUCTION

Storage of information has grown into quite a problem today. The traditional memories such as the ordinary magnetic and the newer opto magnetic ones cannot really, in relative terms of course, hold very much information or provide stored information very fast. In search for better memories, the utilization of light seems to be an attractive way of both storing and transmitting data - it is fast and easily produced. Light produced by a laser which can provide an extremely narrow linewidth, offers the possibilities of addressing individual energy levels in atoms and molecules. Of course if it would be possible to create a memory where, for instance, each excited atom equaled a binary "1" and the ground state equaled a "0", the "ultimate" memory would be born - reading and writing at the speed of light and offering a storage density of 10^{23} bits per cm^3. At this moment such a memory does not seem realistic, but several other approaches have been made where light produced by lasers were used to store data optically.

One suggestion [1,2] is to use Persistent Hole Burning (PHB). To perform PHB a material is needed, e.g. a rare earth doped crystal, which possesses the quality of an absorbing transition accessible by a narrow bandwidth tunable light source, usually a laser, and a possibility of the excited state of the atom decaying into some other state than the ground state. The laser is rapidly tuned in frequency and at the same time amplitude modulated. If the atoms now decay into a state different from the ground state they will not absorb light at the original frequency again. This can subsequently be measured by recording the sample transmission as a function of frequency. At present, though, there are some problems connected with the PHB technique [3]:

1) Rapidly tunable GaAlAs diode lasers necessary for obtaining sufficient data rates for read/write operations only exist in the region 750-850 nm where few suitable materials have their absorption lines.
2) It is difficult to find materials where information can be written and read at sufficient rate and with an acceptable signal-to-noise ratio.

3) Materials that will hold information for an extended time are scarce.

Another and probably more promising way of storing data optically, suggested by [4], is the Stimulated Photon Echo (SPE), sometimes called Coherent Time Domain Optical Memory (CTDOM). In this photon echo approach the information is stored/retrieved by impinging a train of short duration pulses on a rare earth doped crystal. The frequency of the pulses matches the relatively broad absorption line of the crystal and causes interference, constructively and destructively, with the atomic dipoles. The fourier transform of the amplitude modulated light (the incoming pulses) is written into the sample as modulations in the population among the hyperfine levels at the various sites [3,5].
There are some main advantages of using stimulated photon echoes instead of PHB for optical data storage [3]:

1) Fixed frequency lasers can be used.
2) Reading stored information is done simply by sending a single pulse into the sample. This will cause the crystal to "echo" the stored sequence in a specific direction and the information can be detected against a zero background.
3) The photo echo method is 10^3-10^6 times faster than PHB, comparing the data rates possible in the same material.

Because of the fact that storing information with stimulated photon echo is a new technique only estimates of its capacity can be given [5].

Read/write rate	: 10^{10}	bits/sec
Word length	: 10^5	bits/word
Energy/bit	: 10^{-11}	Joule/bit
Bits/address	: 10^5	
Storage density	: 10^{12}	bits/cm^3

These are of course calculated values, but they do suggest that SPE data storage could be a great improvement on all computer memories used today. There are naturally some problems connected with this technique and they will be discussed in chapters 2 and 3.

2. THEORY OF PHOTON ECHOES

The chapter starts with an explanation of the ordinary photon echo (2.1) and the stimulated photon echo in (2.2). Because of the photon echo processes being quite complex, it would be far outside the scope of this paper to give a strictly mathematical treatment of the echoes. The text is written in order to give an understanding of the process and for those who want a detailed description [6,7,8] are recommended. In (2.3) the important relaxation processes involved in photon echoes are treated.

2.1. PHOTON ECHO

A photon echo is a time-delayed burst of coherent radiation and may be pictured as a phase correlated fluorescence confined to a narrow angular cone. Photon echoes have been observed in a large variety of materials here, however, the interest is focused on rare earth doped crystals, e.g. $Pr^{3+}:YAlO_3$ or $Nd^{3+}:YAG$, where the inhomogeneous broadening of a transition is a result of spatial inhomogeneities at the lattice sites giving a spread of individual atomic or ionic resonance frequencies [8]. This broad transition frequency is typically about 20 GHz (FWHM) and, as we shall see later in (3.1), is one of the factors determining the amount of information that can be stored and addressed at a single point of the sample.

When a short pulse from a laser, tuned to the desired optical transition impinges on the crystal a <u>macroscopic</u> dipole moment is induced. This dipole moment decays rapidly, the so-called Free Induction Decay, because of the <u>individual</u> dipoles getting out of phase with respect to one another. This gives us a lot of individual dipoles which are dephased due to the slightly different energy levels in the crystal (the inhomogeneous broadening), <u>but</u> they have one thing in common - the fact that they all started to radiate at the same time, i.e. at the time of the first pulse.

After a time ,τ, the second pulse, which is sent in the same direction as the first one, induces a rephasing of the dipoles, causing the phase relationship between the ground state and the excited state to reverse and after a time, 2τ, the phase of the wavefunction for the upper state dipoles has caught up with those of the lower state for <u>all</u> frequencies (compare with fig.3.), hence resulting in a macroscopic dipole moment which gives the echo copropagating with the first and second pulse. Fig.2.

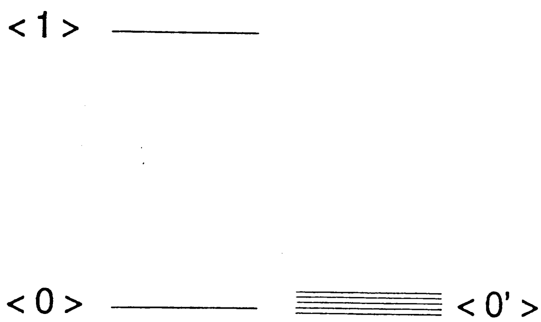


Fig.1. The levels of the optical transition in the crystal. <0> is the lower (ground) state, <1> is the upper (excited> state and <0'> is the hyperfine sublevels of the ground state.

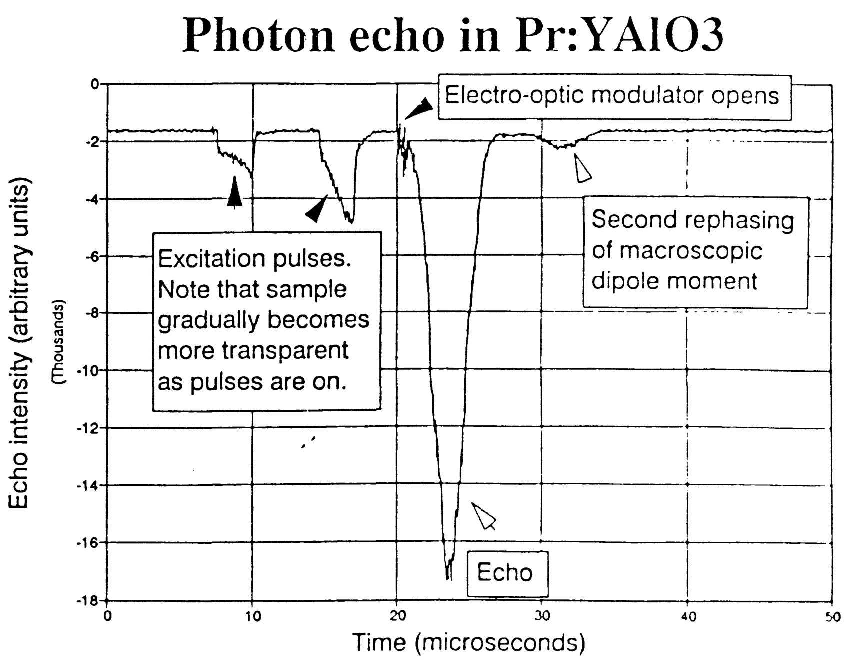


Fig.2. A photon echo recorded in Lund in March 1991.

One way of getting a grasp of what is happening is to study fig.3. This is <u>not</u> a quite true description of what is taking place, but will hopefully give a picture of the process. Each clock symbolizes an ensemble of atoms with a specific transition frequency and a wavefunction, $\Psi_{upper}(t)$ and $\Psi_{lower}(t)$. The macroscopic dipole moment from the ensembles pictured is [8,15]:

$$\sum_{\substack{\text{all transition}\\ \text{frequencies } \omega}} \langle\Psi_u(t)|d|\Psi_l(t)\rangle \qquad (1)$$

where d is the dipole operator. The phase of the oscillating dipole moment for any frequency ensemble is determined by the phase difference between the upper and lower state (i.e. the time difference between the upper and the lower clocks in fig.3.). Thus, only for $t = 2\tau$ (2 tao, see below) will the individual dipole moments for all transition frequencies be in phase. Accordingly, only at $t = 2\tau$ will a macroscopic dipole moment be generated and a signal (the echo) emitted.

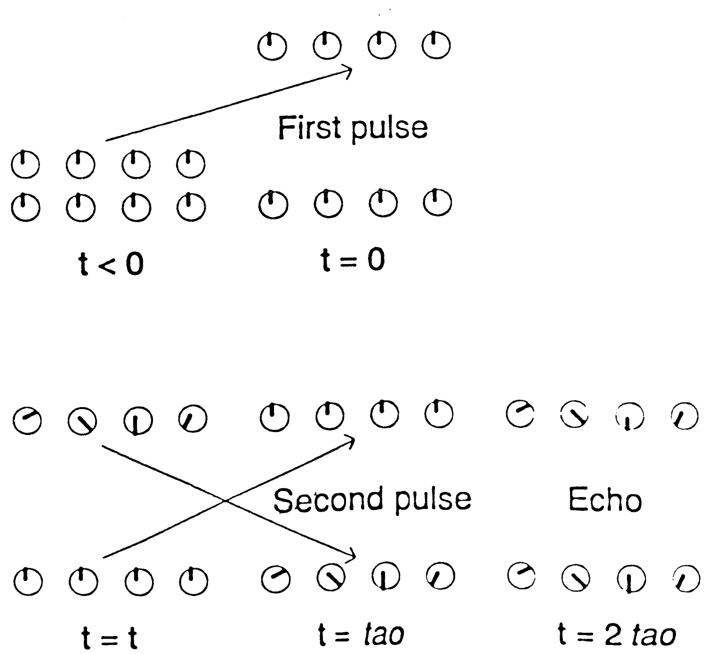


Fig.3. The clocks show the phase of the upper and lower state wavefunctions.

To understand how any information can be stored by sending pulses into a sample we must consider what happens to the population of atoms/ions which at the beginning all are situated in the ground state. Fig.4.

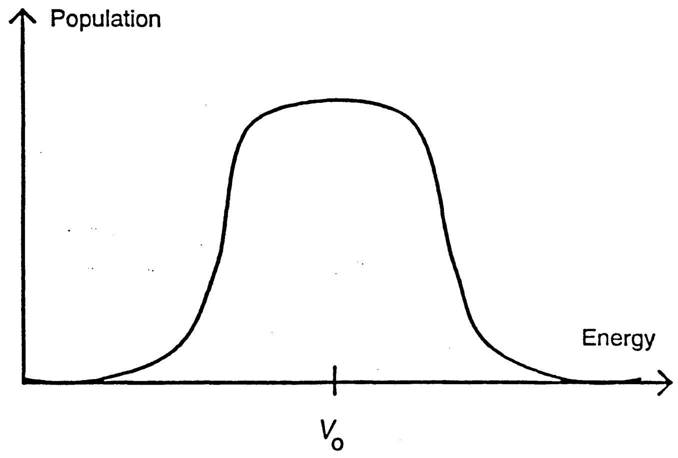


Fig.4. The population in the ground state before the first pulse.

When the second pulse impinges on the crystal it either constructively or destructively interferes with the dipoles created by the first pulse. The dipoles which are in phase with the second pulse, which arrives at the time, τ, are all those with a detuning $\Delta\nu$ from the laser frequency ν_0 where $\Delta\nu x\tau$ equals an integer [5]. This results in stimulated emission of the excited ions and a decreased population level in the excited state. If $\Delta\nu x\tau$ on the other hand equals half an integer the dipoles are out of phase with the second pulse and photons will be absorbed increasing the excited state population.

Of course there are a lot of dipoles which are neither completely in or out of phase with the second pulse, but they are nevertheless affected to some extent by the pulse. The rather simple result of this physically and mathematically quite complicated phenomenon is that a population modulation in the ground state (and in the excited state) is created and the modulation frequency, $\Delta\omega = \tau^{-1}$, is the fourier transformation in frequency of the excitation pulse history in time [5]. Fig.5.

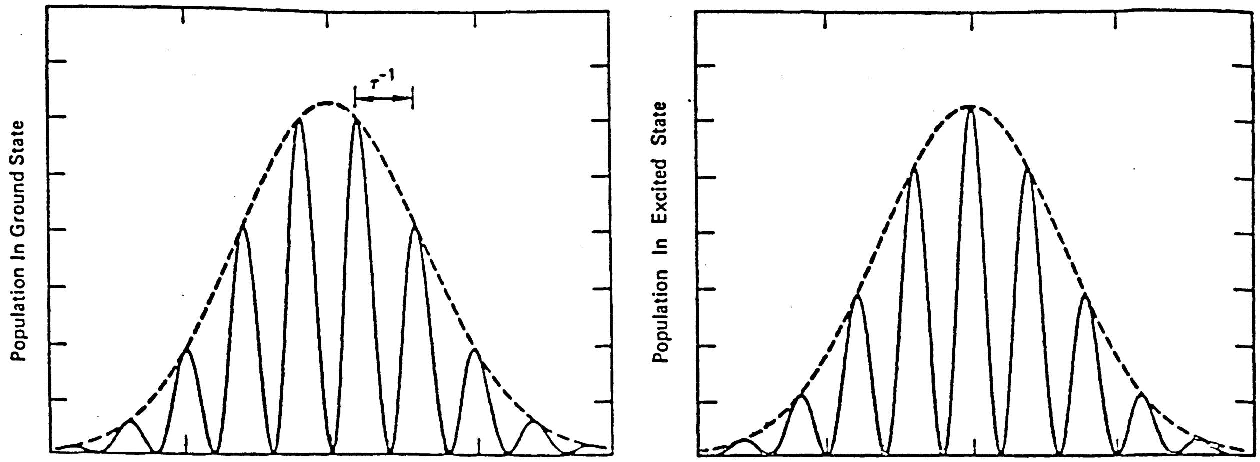


Fig.5. Ground and excited state population modulation resulting from two pulses.

The modulation is given by the expression [7,15]:

$$f_0 \propto \cos^2(\pi.\Delta\nu.\tau) \qquad (2a)$$

$$f_1 \propto \sin^2(\pi.\Delta\nu.\tau) \qquad (2b)$$

where f_0 is the fraction of ions/dipoles in the ground state and f_1 is the fraction in the excited state.

So far we have only discussed photon echoes created by two pulses: the first Write Pulse and the second Data Pulse. Fortunately the data pulse can consist of many pulses. Fig.6. Each one of the data pulses will interfere with the dipoles created by the write pulse and accordingly contribute to the total population modulation. Data sequences up to 10^5 pulses may, theoretically, be stored and addressed at a single spatial location [5]. Data storage is treated in chapter 3.1.

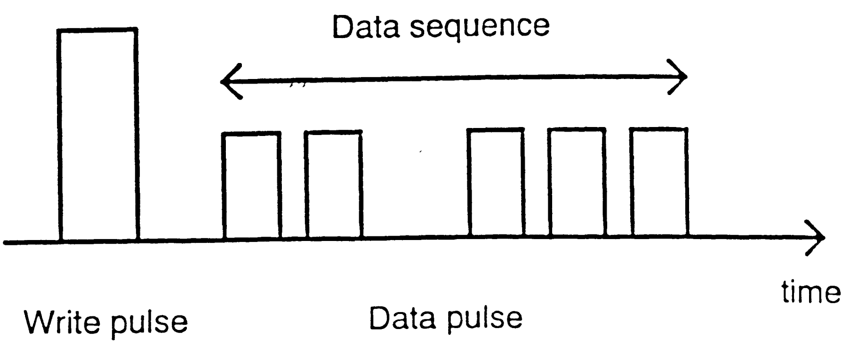


Fig.6. Data sequence.

There is a limit though to how long it is possible to wait between the write and the data pulse. This time must be less than the homogeneous dephasing time of the transition, T_2, and is typically in the micro second regime. If you wait too long the excited ions will deexcite or the dipoles will not only get dephased, but also lose the memory of when they were first excited and then it will not be possible to create the correct population modulation. This limits the time duration of the data pulse sequence, while other criterions governs how much data can be squeezed within the data pulse. These problems will be treated in 3.1.

At this point it might be worthwhile pointing out that the photon echo, as described earlier, will appear at the time, τ, after the second pulse whether the second (data) pulse comprises one or several pulses. So in conclusion, not only do we get the optical echo, but we have stored information about the two pulses. In the next section (2.2) we shall see that a number of pulses can be stored, processed and retrieved.

2.2 STIMULATED PHOTON ECHO

The stimulated photon echo is simply a photon echo forced out of the sample at a time later than the original echo, subsequent to the data pulse. By reexciting the crystal with a single laser pulse, Read Pulse, of the same frequency as used before, the stored information will be emitted. Fig.7.

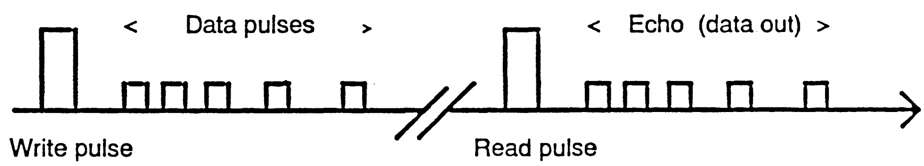


Fig.7. Stimulated photon echo pulses.

To understand how this is possible we return to the time right after the data pulse. As shown in fig.5. a population modulation is created by the second pulse both in the ground state and in the excited state. When the excited state decays it has some probability of decaying into some other state than the original ground state [5]. Fig.8.

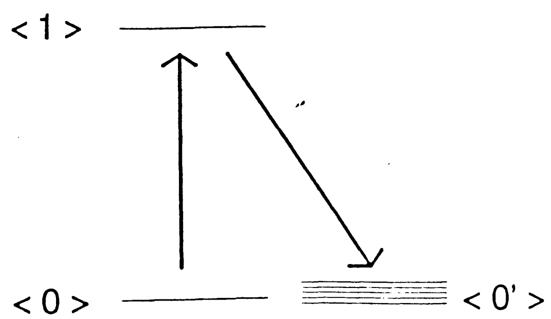


Fig.8. Decay of the excited state.

In the case of rare earth doped crystals this "other state" is hyperfine sublevels of the ground state. The relaxation of these levels is very slow at liquid helium temperatures and it is therefore possible for the population modulation in the ground state to last as long as hours [9].
By sending the read pulse into the sample where the ground state is modulated the excited distribution will be modulated once again with the same distribution in frequency that it had before. The macroscopic dipole moment induced at these frequencies by the read pulse will then generate a replica (echo) of the input data sequence. This stimulated photon echo will be emitted in the same direction as the "ordinary" echo (if the read pulse copropagates with the first pulses) [5].

2.3 RELAXATION PROCESSES

Understanding the relaxation processes involved with photon echoes are quite essential because they govern the performance of any process involving stimulated photon echoes. At this time (Summer 1991) some of the pieces in the relaxation process jigsaw puzzle are still missing and therefore this survey will inevitable have some gaps.

Consider the storage and retrieval of one data bit. Fig.9. The time for the arrival of the first pulse (write pulse) is $t_1=0$, the second pulse (data pulse) occurs at time t_2 and the read pulse at time t_3, thus yielding the stimulated echo at time $t_{32}+ 2t_{21}$. There are two major questions involving data storage with stimulated photon echoes. The first one regards the maximum time allowed between the write pulse and the data pulse, which is the time available for writing bits of information into the sample. This is the so-called homogeneous dephasing time, T_2. The other question deals with the storage time or population relaxation time, T_1. As mentioned earlier in chapter 2.2 the information stored is possible to retrieve as long as the population modulation in the ground state is not ruined by relaxation between the hyperfine levels.

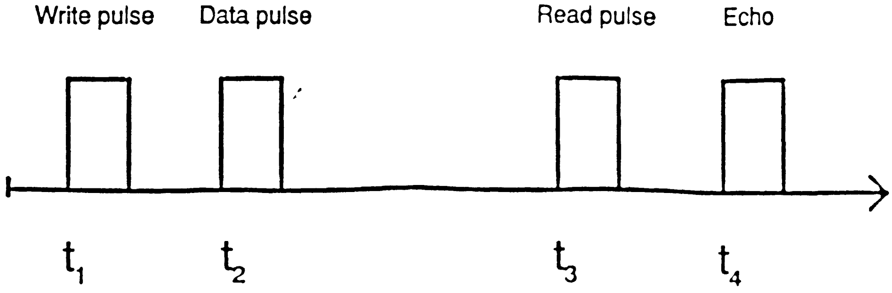


Fig.9. Storage and retrieval of one bit of information.

2.3.1. DEPHASING TIME T_2

The dephasing or homogeneous relaxation time, T_2, is of particular interest because it involves several complicated and to some extent unknown physical actions in the crystal. Another way of expressing T_2 is to use the homogeneous linewidth (FWHM), γ.

$$\gamma = 1/(T_2.\pi) \qquad (3)$$

A typical value of γ is around 10 kHz in rare earth doped crystals. The mechanisms that effect the homogeneous linewidth can be divided into [11]:

$$\gamma(\text{total}) = \gamma(\text{radiation}) + \gamma(\text{phonon}) + \gamma(\text{magnetic}) + \gamma(\text{unknown})$$

γ(radiation) is the natural linewidth or, equally, the radiative lifetime of the excited state.

γ(phonon) is due to phonon interactions in the crystal [11].

γ(magnetic) originates from magnetic dipole induced spin flip-flops of the ligands in the lattice [11].

γ(unknown) is the more or less mysterious contribution which to some extent is pulse energy dependent and seems to be quite different in different materials [11,12].

To study these parts of the homogeneous linewidth we would like to isolate each contribution by suppressing the other unwanted ones. Of course **γ(radiation)** will thanks to Heißenberg always be there, but its contribution is constant and , luckily, small and can therefore be compensated for.
The **γ(phonon)** part is only present at temperatures above 4 K [11] and by cooling the crystal to liquid helium temperatures this effect can be avoided. It might be possible for the pulse to actually heat the crystal

because of the energy absorbed and thereby creating phonons in the sample, but the temperature increase that could be caused by an excitation pulse is less than 0.5 mK even if all the absorbed energy would be transformed into phonons [11]. γ(phonon) is therefore regarded as zero in measurements made at liquid helium temperatures.

γ(magnetic) is considered arising from nuclear spin-flips in the dope atoms/ions induced by nuclear spin-flips in atoms/ions in the lattice. In $Pr^{3+}:YAlO_3$, for example, spin-flips in Pr^{3+} are induced by nuclear spin flips in the Al lattice atoms. Applying an external magnetic field breaks the energy degeneracy of oppositely directed nuclear spins of equal size and thus for sufficiently large magnetic fields the energy needed to induce the mutual spin flips of the Pr atoms is not available in the crystal. [11].

Exactly what **γ(unknown)** arises from is still unclear. Several mechanisms have been suggested [10,11,12,13], among them optical density effects and Instantaneous Spectral Diffusion (ISD).

The dependence of the echo decay time as a function of optical density arises because of the attenuation of the input pulses and the echo as they propagate through the sample [12]. Ions at the end of the crystal will thereby experience a lower excitation energy fluency than the ions situated at the entrance of the crystal.

Instantaneous spectral diffusion is due to a change in the permanent electronic dipole moment of an ion when it is excited (or deexcited). This results in a change of the local crystal field and may cause the transition frequency of a nearby ion to change. If this occurs while the second ion participates in the generation of a photon echo, the rephasing of this second ion will no longer occur at the exactly correct time and the echo signal will decrease. For this mechanism a strong second pulse has a larger effect on the homogeneous dephasing rate than a strong first pulse since a crystal field induced by the first pulse will not destroy the photon echo rephasing [12]. Experiments have shown that ISD contributes to <u>some</u> part of the γ(unknown), but the results from different types of crystals indicate that ISD cannot generally be the only contribution to γ(unknown). For instance in 0.1% $Pr^{3+}:YAlO_3$ neither optical density effects nor ISD is believed to be responsible for the energy dependent homogeneous dephasing time [12].

What causes this energy dependence is unclear, but it has been found [12] that this mechanism is approximately proportional to the density of excited states raised to 1/3 or, equivalently, inversely proportional to the distance between the nearest excited neighbors [15].

2.3.2. POPULATION RELAXATION TIME, T_1

The population relaxation time is the storage time of the crystal and arises from relaxation in the hyperfine levels in the ground state. This time is known to be as long as hours [9].
Recent results have shown that T_2 and T_1 are not independent [10] and this is important when it comes to storing data ,because a long time for writing as much information as possible is wanted (T_2) as well as a long storage time (T_1). The dependence for the $Pr^{3+}:YAlO_3$ has been found to be :

$$T_2(0) \,/\, T_2(T_{1,maximum}) \simeq 4 \qquad (5)$$

The consequence of this is when the maximum time available for storing data is used only 1/4 of the maximum time available for writing data can be utilized.

3. APPLICATIONS

The applications presented here are both fascinating and amazing and shows some of the future possibilities of the stimulated photon echo. Optical data storage as well as image processing have successfully been performed. Pulse shaping/processing and detection of ultra short pulses are fields of applications which are to be investigated in the near future.

3.1. Optical data storage

The probably most obvious way of utilizing the stimulated photon echo is to store bits of data. How data can be stored and retrieved has already been discussed in chapter 2.2, but there are some other features that need to be pointed out. To start with, the choice of material is crucial and today considerable effort is aimed at finding the appropriate rare earth doped crystals or some other suitable material.

A well suited material must have a large inhomogeneous linewidth, while this increases the number of pulses that can be squeezed within the linewidth. The minimum separation between pulses in a data sequence is set by the reciprocal of the inhomogeneous width. This together with the speed of the pulse modulator, which creates the pulses, sets the write (and read) rate.

As an example the absorption linewidth of $Pr^{3+}:LaF_3$ is about 0.53 cm^{-1} (FWHM) [5] which corresponds to a theoretical read/write rate of 16 GHz. Hopefully there will be fast enough tunable laser diodes at a matching wavelength available in the future. This can e.g. mean finding new rare earth doped crystals with suitable transitions at longer wavelengths than now and producing tunable diode laser with shorter wavelengths. (Today tunable visible light is used.)

Already mentioned is the large T_2 and T_1 needed, that is a long time available for writing the data and added to that a long storage time. As explained in the previous section it seems to be impossible to utilize the maximum T_2 and T_1 of a crystal at the same time. This limitation is not as severe a problem as it is an interesting physical phenomenon and once understanding what causes it there might be a way of circumventing it.

The first steps towards storing data using stimulated photon echo in rare earth doped crystals was taken in 1985 by Ravinder Kachru *et al.* at SRI,California and at present a couple of hundred bits have been stored for some hours [9]. This result may not be very remarkable, but the fact is that most of the research that has been done has dealt with the physical aspects and problems of SPE. Accordingly the true potential of rare earth doped crystals as optical storage media will not be seen in the immediate future. For instance nobody knows how close the storage points can be or how an optimal laser scanning device, which addresses the individual locations is best created.

3.2. Image processing

An interesting technique of image processing using backwards stimulated photon echo has been presented and performed by Xu *et al.* [16]. This is done by using a four-wave mixing setup and focusing images on the crystal. The images which impinges on the sample represents the write, data and read pulses. Fig.10. As a result the images are either correlated or convoluted. By letting the data pulse image consist of many different images as many as 10^5 pictures can be stored (theoretically) and subsequently a read pulse/image is convoluted /correlated with <u>all</u> the stored images. This offers a unique possibility of comparing images on a sub nanosecond timescale.

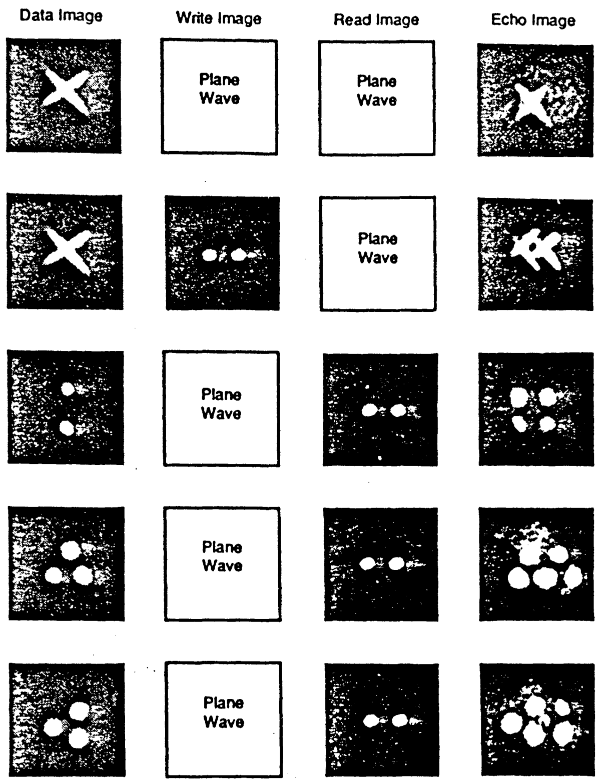
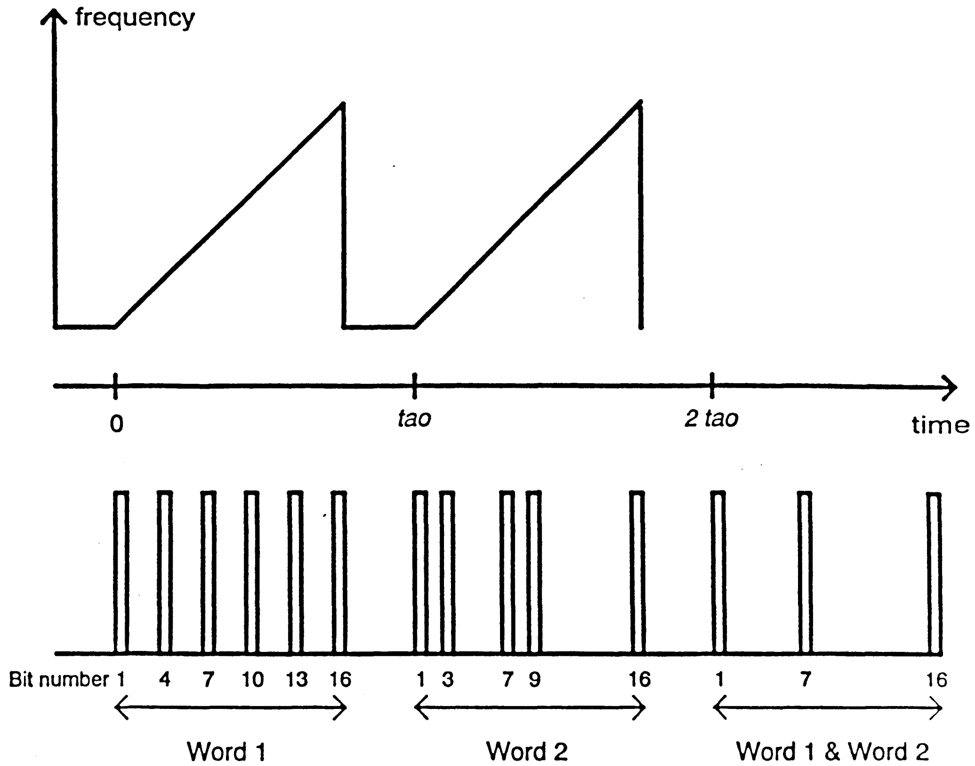


Fig.10. Image processing.

3.3. Pulse shaping and processing

By sweeping the frequency synchronously on two words consisting of a number of bits and letting the first word represent the write pulse and the second word the data pulse, a logic operation can be performed [15]. Fig 11. A number of operations are possible by sweeping the frequency in different ways, e.g. reversing words. This ability to process data together with the fact that SPE offers true parallel processing of images/data opens an interesting way of using rare earth doped crystals as computer processors. Such a processor could handle 10^6 parallel eight bit words with a GHz frequency.

The photon echo concept not only allows spatial but also temporal fourier transformation of pulses [15]. This means that it is possible to manipulate with the shape (in time) of the pulses and could be used to compress high power laser pulses and thereby increasing the power per time unit in the pulse



3.4. Detection of optical pulses.

Because any data pulse sequence with appropriate wavelength will create a frequency distributed population modulation in the ground state it would be possible to record sequences of pulses.

By recording the frequency dependant absorption in the sample the fourier transform of the stored sequence can be read at a slow rate. Fourier transforming the result would recreate the data pulse sequence [15].

As mentioned before in 3.1 how short pulses that can be recorded is governed by the reciprocal of the inhomogeneous linewidth which corresponds to the pico second scale for rare earth doped crystals.

4. PHOTON ECHO RELAXATION MEASUREMENTS

These relaxation measurements on the $^3H_4-^1D_2$ transition in 0.1% $Pr^{3+}:YAlO_3$ were performed at the Department of Physics, Lund institute of Technology, Sweden, during the spring of 1991.

4.1. BACKGROUND

In 1990 Dr. Stefan Kröll started a stimulated photon echo research program at Lund Institute of Technology. This is a program, financed by STUF (NFR) and NFR and aims to clarify the dynamic processes involved in the stimulated echo of rare earth doped crystals and to investigate the prospects for optical storage and processing in these materials. Items to be investigated are e.g. [15]:

1) The magnetic field dependence of the homogeneous relaxation time.
2) The intensity dependence of the homogeneous relaxation time in rare earth doped crystals.
3) Experimentally obtainable S/N in optical storage using SPE.
4) Investigations of the hyperfine level relaxation process.
5) Testing the concept of utilizing SPE for detection of ultra short pulses.
6) Finding the upper limit for the storage density.

This diploma work which mainly has been of experimental character has included the following tasks.

* Aligning the expérimental setup
* Programming the digital delay generators and the communication with the boxcar unit.
* Developing evaluation and data storage programs
* Performing photon relaxation measurements
* Evaluating experiments
* Designing a sample holder

4.2. EXPERIMENTAL SETUP

The experimental setup is shown in fig.12. A Spectra Physics Ar^+ laser pumped a cw ring dye laser, Coherent Radiation CR699-21, which produced a continuous 300 mW beam at 6105.28 Å. A polarizer ensured that the beam impinging on the first of the two Pockels' cells were polarized. By using two Pockels' cells to produce the pulses the continuous background radiation, originating from the imperfect attenuation of the Pockels' cells, could be reduced a factor of 50000.
Theoretically they would have reduced the background by a factor of 2000x1000 = 2000000, but even when we managed to get the first cell to attenuate the beam more than a factor of 1000, the other one would not attenuate more than 50 times. Hence the factor 50000. The reduced attenuation may have been due to the fact that the spatial mode structure of the beam leaving the first cell was a bit distorted.
Before entering the Cryovac 150 cryostat with the 1 mm thick 0.1% $Pr^{3+}:YAlO_3$ sample the pulse passed a filter, to eliminate the straylight from the Ar^+ laser, and a polarization rotator. The crystal was mounted on a holder which could rotate the sample in two directions. This possibility of rotating the crystal was to be used when the different characteristics of the three crystal axis - a,b and c were investigated. Unfortunately it turned out to be impossible to perform these measurements because the external magnetic field was to some extent obscured by the holder. To succeed in these measurements the brass holder has to be exchanged for a plastic one, which does not interfere with the external magnetic field.
A pair of electromagnetic coils placed outside the cryostat generated the magnetic field ranging from 0 to 20 mT. An acusto optic modulator attenuated the strong excitation pulses and was only open when the echo was transmitted. This was registered by a photo multiplier tube connected to a boxcar integrator.
During the initial measurements we used a transient digitizer, Biomation 8100, together with a computer, simulating a boxcar integrator. This had the disadvantage of a low repetition rate and by substituting this setup for an SR 250 boxcar integrator, the repetition rate was increased by a factor five.

The maximum frequency for the measurements was then set by the crystal itself, because if the new set of pulses were to close to the old ones the crystal did not get time enough to relaxate and an unwanted stimulated photon echo would be produced.

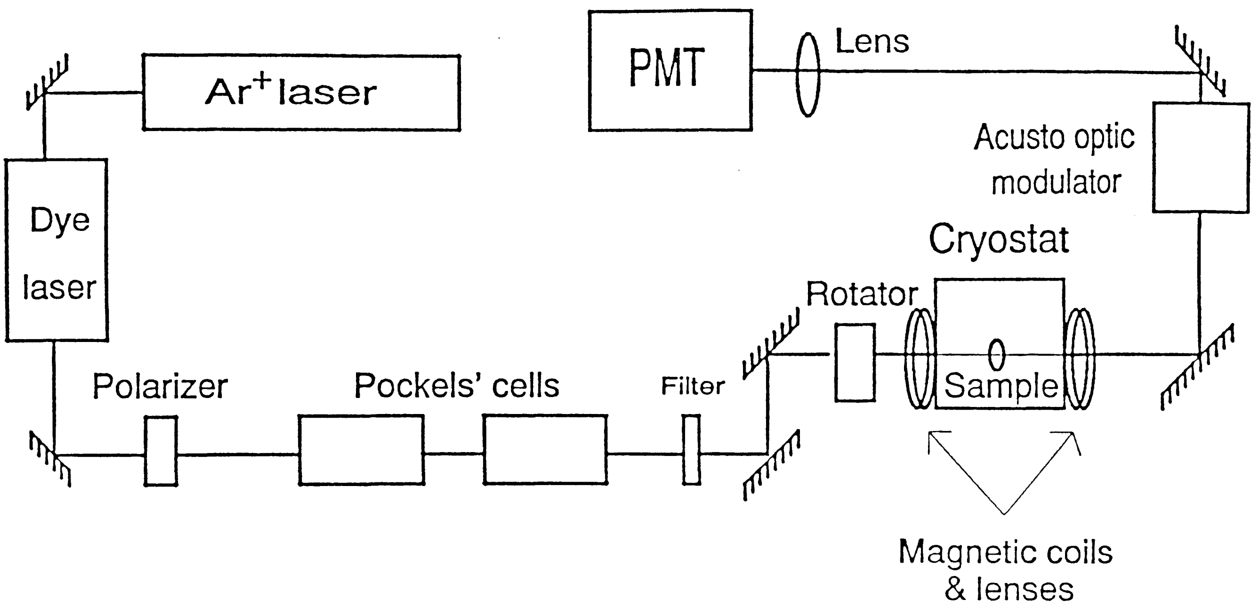


Fig.12. The experimental setup.

4.2.1. Computers and programming

A 386 SX personal computer together with three Digital Delay Generators, (DDG), SRS DG 135 were programmed to produce and time the TTL pulses for the modulators. A program written in Fortran and Assembler handled the communication with the boxcar integrator and the storage and presentation of the experimental data. The three DDG:s were simultaneously triggered at a rate set by the user - the repetition rate. After a time t_0 in the beginning of each cycle the first DDG delivered a pulse, A, lasting #1 seconds. The second pulse, B, lasting #2 seconds was produced at a time, τ, later. These two TTL pulses were amplified and subsequently sent to the Pockels' cells which gave the read pulse (A) and the write pulse (B). To record the photon echo the third DDG produced a pulse, C, at a time $2\tau-\delta$. This pulse caused the acusto optic modulator to open for a time (= Boxcar window) and at the same time trig the boxcar integrator so that the echo signal from the PMT could be recorded and via the SR 245 interface be sent to the computer. Fig. 13. The main program is featured in appendix 1.

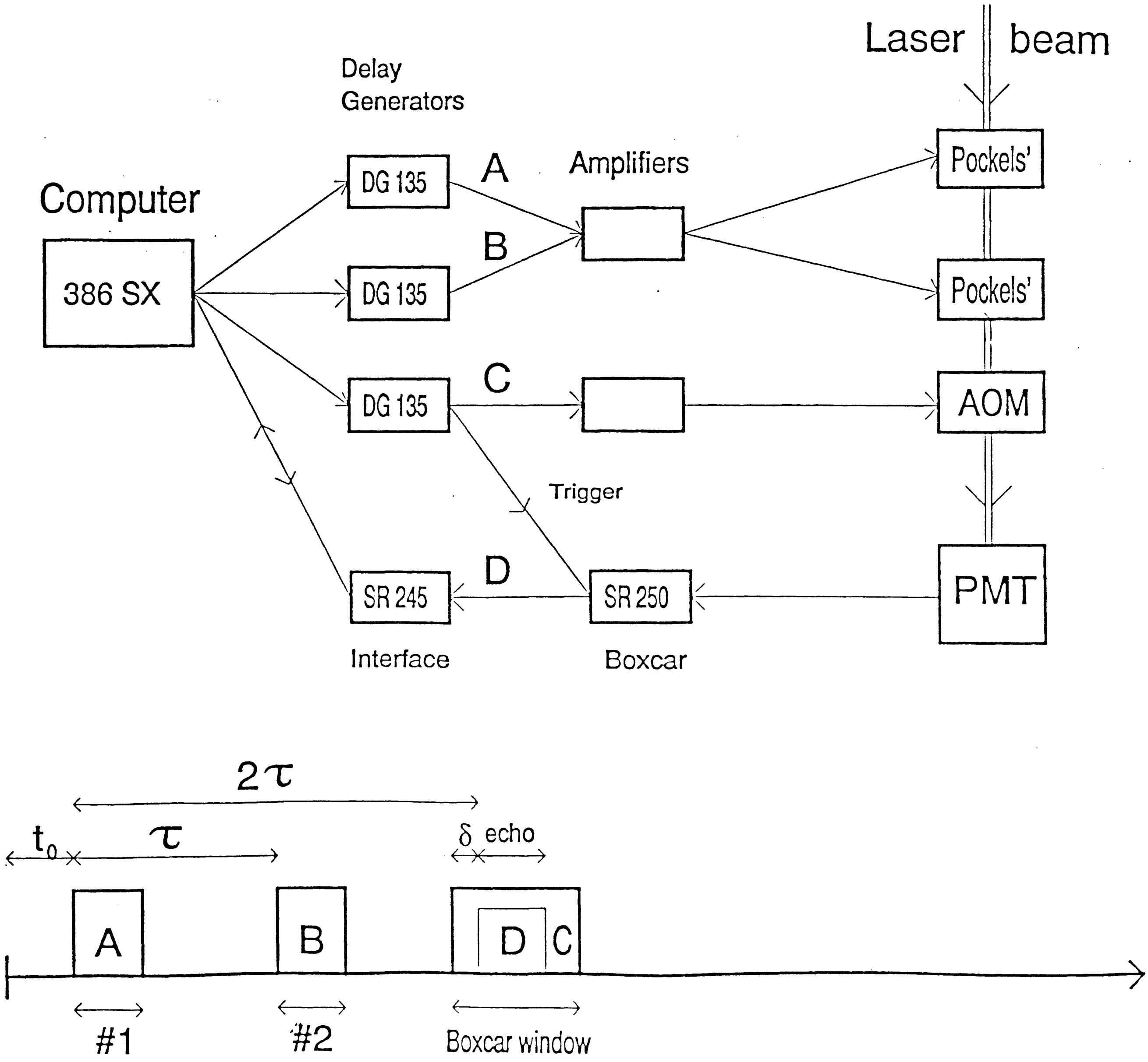


Fig. 13. Computer and electronics.

4.3. MEASUREMENTS AND DISCUSSION

As mentioned earlier (2.3.1.) a part of the homogeneous linewidth is due to the fluctuating magnetic field at the ion site, generated by nuclear spin flip-flops of neighbouring crystal ions, γ(magnetic). Added to this there is another part which has been described as γ(unknown) and to some extent seems to be a result of interaction with the exciting optical field. To obtain further information about the magnetic properties of rare earth doped crystals the $^3H_4-^1D_2$ transition in 0.1% $Pr^{3+}:YAlO_3$ has been studied. The excitation energy dependent photon decay has been investigated as a function of a variable external magnetic field.

Former results [17] have showed that it is possible to "freeze" the flip-flops of the neighbouring atoms by applying an external magnetic field. If the pulse energy is sufficiently low γ(unknown) is essentially eliminated and the effect on the photon echo decay by varying magnetic field can be seen.

If the theory of freezing the flip-flops is correct the decay time should move towards a static value when the magnetic field is sufficiently strong and thus forcing γ(magnetic) to be negligible. Earlier measurements [15] show that this is the case, but also that the decay time <u>decreases</u> at 8 mT - a dip in the rising decay curve occurs. Fig.ww. What causes this dip is not known, but Stefan Kröll has suggested [15] that it might arise from a coincidence between the transition frequencies of the $^3H_4(m_I=-1/2),^1D_2(m_I=1/2)$ and $^3H_4(m_I=1/2),^1D_2(m_I=3/2)$ transitions. If this is true another dip should show at a magnetic field of 13 mT due to a similar coincidence of transition frequencies. Previous decay measurements have not treated fields larger than 10 mT, so in that sense this is a novel experiment.

To find out the decay time, T_2, the pulse area i.e. the energy of the photon echo, was measured. The intensity of the echo as a function of the separation, τ, between the write pulse and the data pulse is [13]:

$$E_e \propto \exp(-4\tau/T_2) \qquad (6)$$

Because of the rather weak photon echo signal an average of some fifty echoes was taken for each τ , which was varied from a couple of microseconds to some hundred micro seconds in steps. Such a measurement is shown in fig.14. These measurements were repeated at different external magnetic fields ranging from 0 to 17 mT. The crystal was placed so that the pulses propagated parallel to the c-axis and the magnetic field was also parallel to the c-axis [15]. Fig.15. shows a decay curve where the magnetic field is 9 mT. Note that the decay is slower than in the zero field and that the magnetic field causes splitting of magnetic levels in the Pr^{3+} ion, which yields the oscillations in the decay curve.

The decay curves were subsequently evaluated by a computer program and the results are shown in fig.16. These are the average results from two different occasions. Comparing our results with the previous measurements of the magnetic field dependence of the decay time shows the same dip at 7-8 mT, <u>but</u> the predicted dip at 13 mT does not appear. We have reached the same result at several different occasions, so there is no reason to believe that the 13 mT dip does exist. Another mechanism than the suggested coincidence must be found to explain the 7 mT dip.

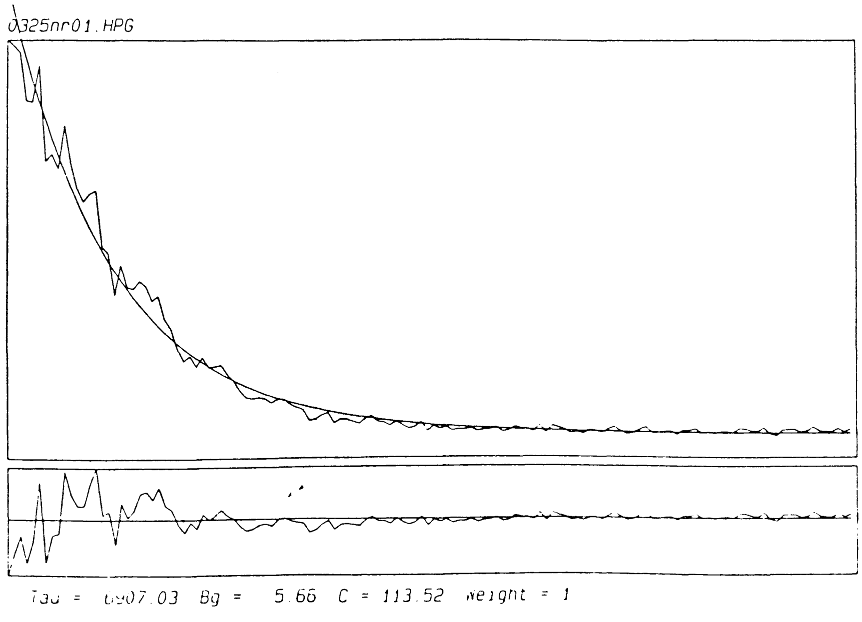


Fig.14. Relaxation measurements made with 0 mT magnetic field. The decay time is 6.9 μs.

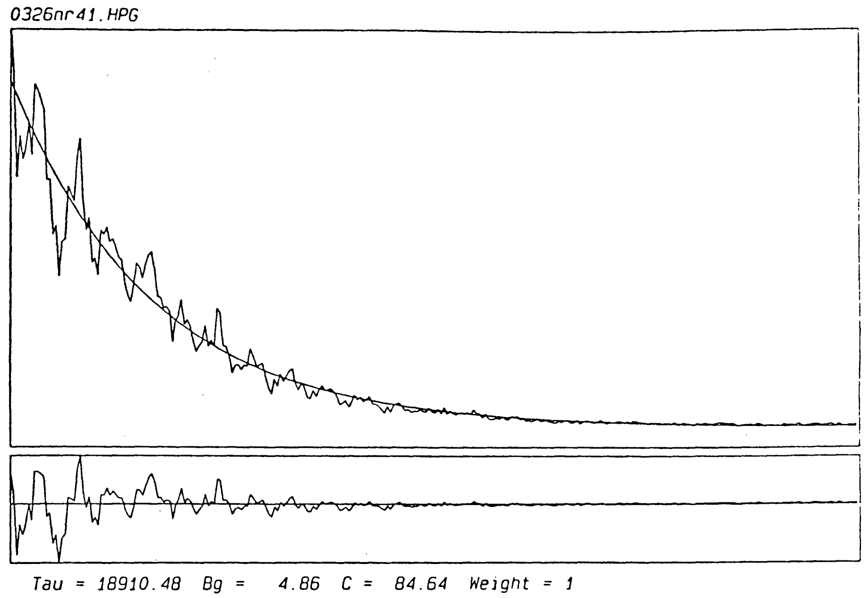


Fig.15. Relaxation measurements made with a 9 mT external magnetic field. The decay time is 18.9 μs.

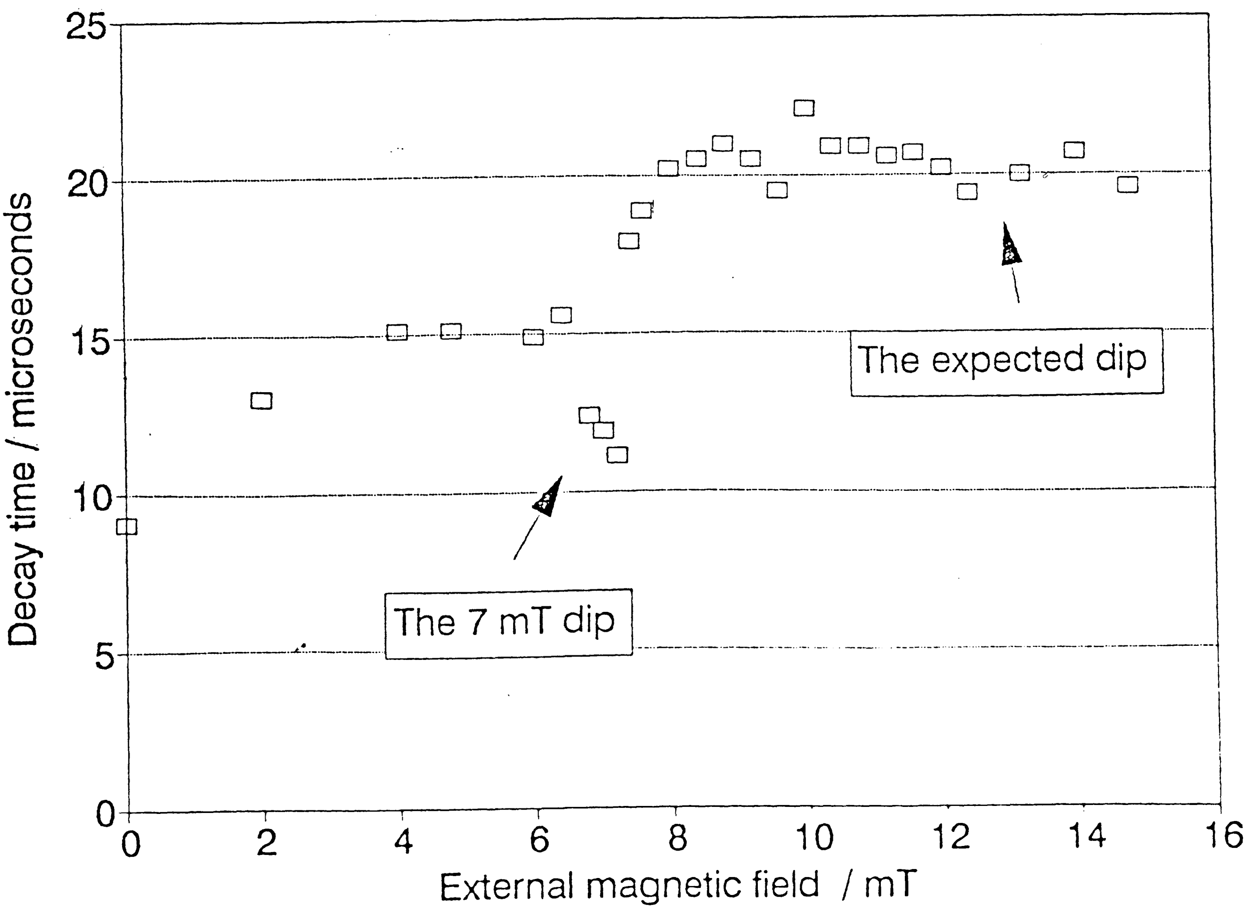


Fig.16. The decay time as a function of an external magnetic field.

In conclusion, we have aligned the experimental setup and for the first time in Scandinavia recorded a photon echo in rare earth doped crystals.

A system for timing the pulses, communication with the boxcar integrator and collecting/storing data has been developed and resulted in an automatic computer system for photon echo measurements.

A theory of coincidences between transition frequencies in external magnetic fields has been investigated by performing photon echo relaxation measurements on the $^3H_4-^1D_2$ transition in 0.1% $Pr^{3+}:YAlO_3$.

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7. APPENDIX

The computer program which controlled the timing of pulses, communication with the boxcar integrator and the storage of data is featured in this appendix.

The main program comprises (Fig.A.):

- Restore - Reads ten sets of stored information about previous measurements.

- Menu - - Submenu -

- Initiate - Initiates the three digital delay generators with the users choice of values.
  - Initunit - Initiates one single unit.
  - Trigrate - Sets the repetition rate of the delay generators.
  - Setupn - Basic initiation made when system starts.
  - Sett0 - Sets the time before the first pulse.
  - Setdelay - Sets the time of the pulses.

- Box - Measurements area.
  - Draw - Draws curves in graphic mode on the screen.
  - Srs245 - Communicates with the boxcar interface.
  - Fread - Reads stored data and settings.
  - Fstore - Stores data and settings.
  - Setdetla - Computes the time to be added between two different τ:s.

- Utilities - Functions and procedures used by other functions /procedures.
  - Openfile - Opens and checks a file.
  - Ok - Waits for a confirmation of a statement.
  - Svar - Yes/No questions answered.

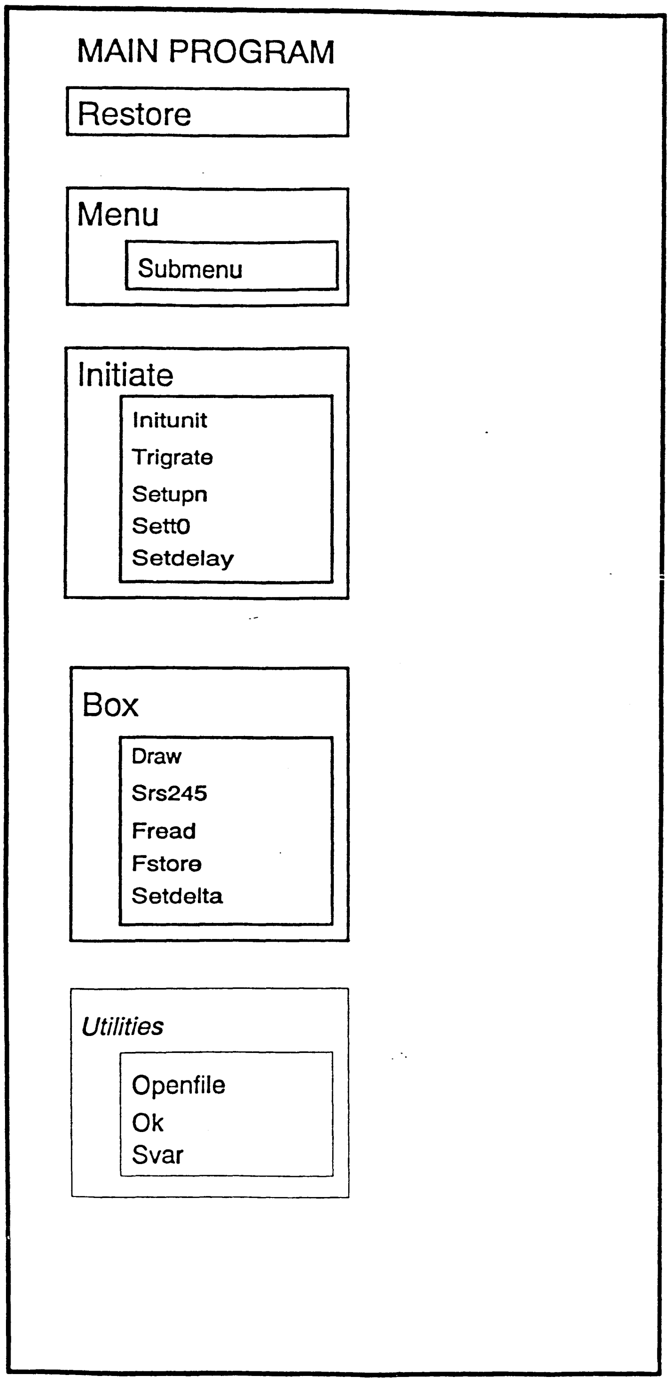
MAIN PROGRAM
Restore
Menu
Submenu
Initiate
Initunit
Trigrate
Setupn
Sett0
Setdelay
Box
Draw
Srs245
Fread
Fstore
Setdelta
Utilities
Openfile
Ok
Svar

Fig.A.