PHASE CONJUGATION BY DEGENERATE FOUR WAVE MIXING IN DISODIUM FLUORESCEIN SOLUTION IN METHANOL*

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ABSTRACT

Organic dyes are known to show resonant type of non-linear optical properties, including phase conjugation. In our present work, disodium fluorescein in methanol is used as an organic non-linear medium for degenerate four wave mixing at 532 nm to see the intensity dependence of the phase conjugate signal at different concentrations of the solution. It is observed that the maximum reflectivity of the signal occurs in a concentration range of 5×10^{-3} g/cm^3 to 1.2×10^{-2} g/cm^3. It is also observed that the intensity of the signal drops suddenly to less than half of its maximum outside the concentration range mentioned above.

An investigation of the phase conjugate signal intensity by changing the delay time between probe signal and the forward pump signal is also examined.

Briefly discussed is the possibility of population grating in dye liquids as a source of enhancing the third order susceptibility besides the other techniques mentioned in reference[1].

The experiment is done by beam splitting the second harmonic (532 nm) of Nd:YAG laser, Q-switched at 20-pulses/sec. (pulse width ~8ns and ~200 mJ per pulse).

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Introduction

Phase conjugation by degenerate four wave mixing (DFWM) has shown considerable promise as a technique of generating high fidelity phase conjugate signals. The only difficulty which limits its use is its low efficiency. Because of that, a great deal of research work is concentrated on finding the proper and most efficient nonlinear media with the highest possible third order susceptibilities to improve the efficiency.

In an earlier experiment[6], Caro and Gower obtained a DFWM signal which decreased and vanished if the path difference was greater than the coherence length, showing that their signal was purely due to DFWM. On the other hand, Basov et. al[8] obtained stimulated Brillouin Scattering from two pump beams even with a path difference much larger than the coherence length of the laser. Seeded Brillouin Scattering has also resulted in phase conjugation of two incoherent beams[9].

Recently, considerable interest has been generated in the study of organic dye solutions as efficient nonlinear media. A class of highly efficient organic dyes with a wide and flexible range of operating conditions has become known.

The purpose of our experiment is to pick up an organic dye as a nonlinear medium for our DFWM experiments to investigate the possible mechanisms which are responsible for phase conjugation. Disodium fluorescein was chosen among several other organic dyes as a candidate for our experiment based on the idea that it showed a relatively strong phase conjugate signal, using a frequency doubled Nd:YAG laser (532nm), Q-switched at 20 pulses/sec (pulse width ~8 ns and ~200 mJ/pulse), although its absorption spectrum in ethanol shows only a small tail beyond 525 nm.

Mechanisms for DFWM:

When mutually coherent laser beams interfere in an absorbing medium, the interference pattern heats the medium nonuniformly. The nonuniform heating causes density variations in the medium, consequently an index of refraction grating, which Bragg scatters a read beam into the conjugate wave signal, is formed.

It is believed[1] that in the initial few picoseconds after the energy has been deposited, the medium has not yet had time to expand, the density is thus constant and any index change is due to the intrinsic variation in the index of refraction with temperature at constant density. Subsequently, sound waves produced by the nonuniform heating propagate across the medium and begin the expansion process. Finally, the sound waves damp out and the usual thermally induced grating[2] becomes dominant.

Besides the thermal grating mechanism, there are other possible mechanisms which are also responsible for producing nonlinear susceptibility like saturable absorption[3] and the optical Kerr effect[4].

In addition to these different mechanisms, one may also examine the energy level structure of a typical dye molecule, shown in Fig. 1[5], where the first excited singlet state is generally higher than the lowest triplet state. Since dye molecules are large, the vibrational and rotational levels in any electronic state are too numerous and too closely spaced, and the spread of these levels is large. Further, the excited singlet states are not well separated from one another. All the dye molecules in the excited states relax quickly by nonradiative interactions to the lowest singlet state S_1. This is the only state which has been observed to fluoresce and has, in most of the cases, lifetimes of the order of a few nanoseconds. The upper singlet states are not known to fluoresce and they relax to the S_1 state in times of the order of picoseconds. There are two other competing processes for relaxation of the molecules from S_1 in addition to the spontaneous emission to the ground state. Although singlet to triplet state transitions are generally forbidden, relaxation to triplet levels is still possible. The triplet state T_1 has a lifetime of a few microseconds which is large compared to the life time of the S_1 state.

The possible excitation to the triplet state T_1, which is of a relatively longer lifetime, may result in a population grating which should be considered as an extra mechanism which will contribute to the third order nonlinearity and Bragg scattering of the phase conjugate signal.

Experiment

The experimental set up is as shown in Fig. 2 in which the frequency doubled Q-switched Nd:YAG laser (532 nm) is split to obtain ~20% as a probe beam A_3 and the rest as two equally intense and counter propagating pump beams A_1 and A_2. The phase conjugate signal is seen as a beam counter propagating to A_3, the intensity of which is first observed at different concentrations of the disodium fluorescein in methanol in a 1 cm thick cuvette. The experimental results obtained are as expected (Fig. 3). At relatively low concentrations the number of the dye molecules are not enough to form a strong grating to reflect the signal. As the concentration increases to the optimum one, $\sim 7 \times 10^{-3}$ g/cm^3 in this specific case, the grating is good enough to reflect a relatively powerful phase conjugate signal. At higher concentrations up to 20×10^{-3} g/cm^3 the curve shows a weaker phase conjugate signal.

The decay in the phase conjugate signal at high concentration can be attributed to two factors: (1) The fluorescence quenching where the fluorescence emitted by some molecules are absorbed by others in the medium. This contributes to heating the medium quickly. The rise in temperature inhibits the formation of the grating and hence the medium becomes less efficient. (2) The heat increase in the cell causes the medium to boil. The hydrodynamic motion of the liquid tends to wash out the grating and reduces its efficiency.

The dependence of the phase conjugate signal on the delay time between the probe and the forward pump is shown in Fig. 4. Since the phase conjugate signal is thought to be due mainly to the grating which is formed by the interference pattern of the two coherent beams, A_1 and A_3, one expects that if the path difference between the beams is larger than the coherence length of the laser, which is ~1 cm (0.03 ns delay time), the signal should vanish. Surprisingly, the phase conjugate signal is detected even for a path difference of more than 360 cm (12.0 ns delay time) (Fig. 5). This result is probably attributable to the presence of stimulated scattering. Similar results are obtained for the same sample in a smaller cell of 1 mm thickness. Preliminary experiments with a single beam show that stimulated back-scattering signals can be obtained from pure ethanol, acetone and methanol. Experiments on stimulated scattering in dye solutions are now in progress to clarify this situation.

In summary, we discussed a population grating as a mechanism for the index grating in liquids, along with the previously mentioned ones. In addition, our experimental results show that the phase conjugate signal through DFWM could be superimposed on a signal due to stimulated scattering.

Acknowledgments

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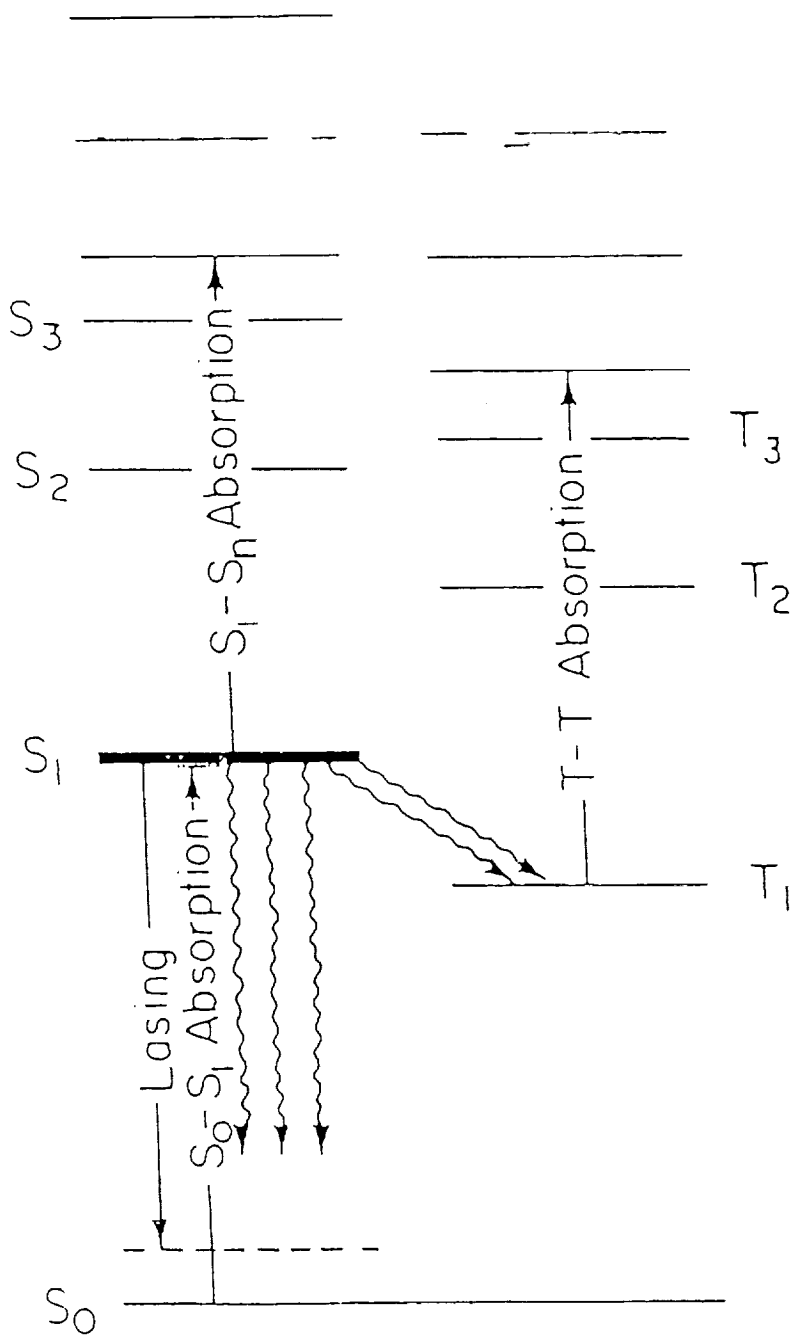

Figure 1. Energy level diagram of a dye molecule.

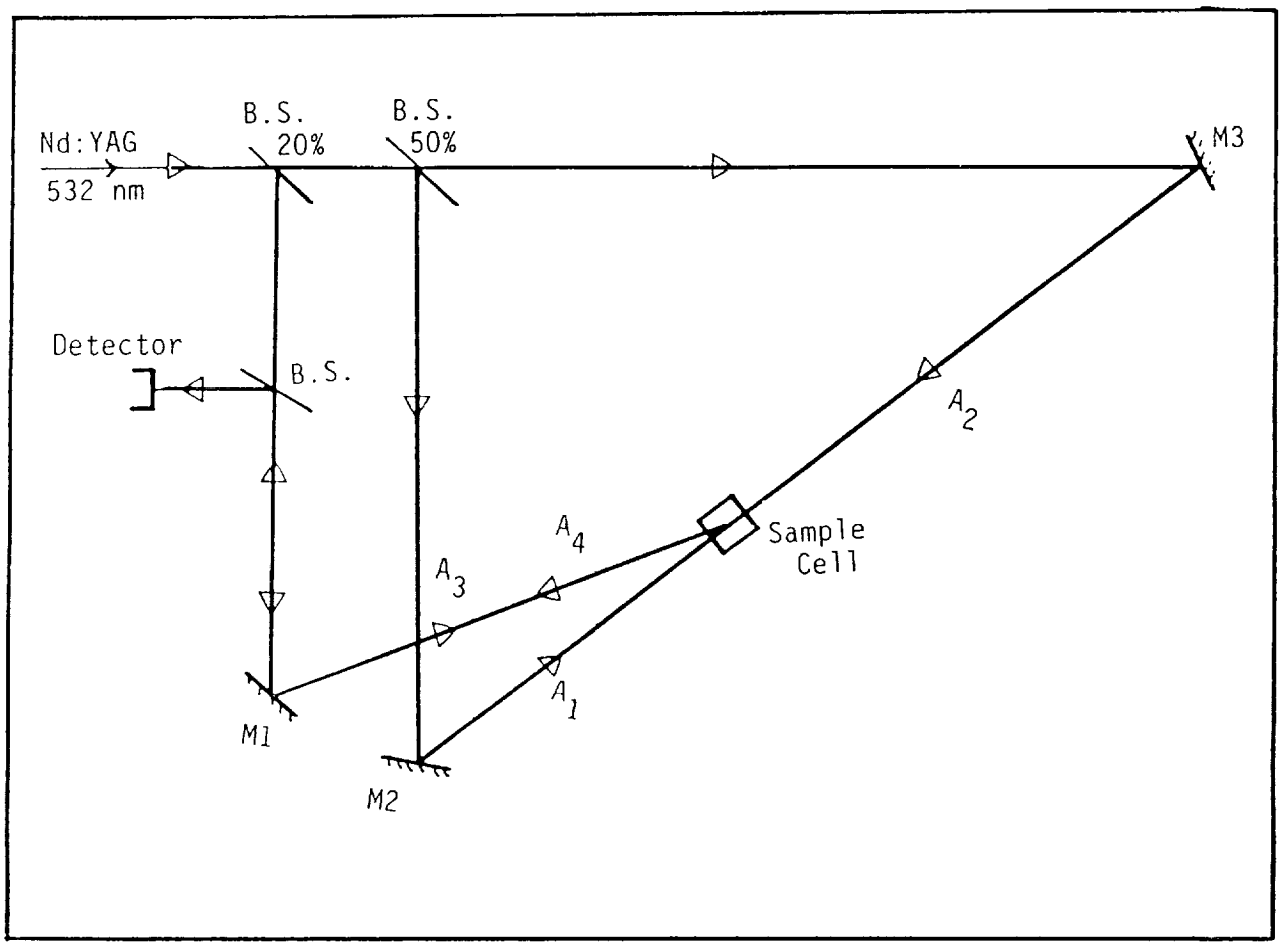

Figure 2. Experimental Layout: BS: Beam splitters M: Mirrors
A_1, A_2: Pump Beams, A_3: Probe, A_4: Phase Conjugate.

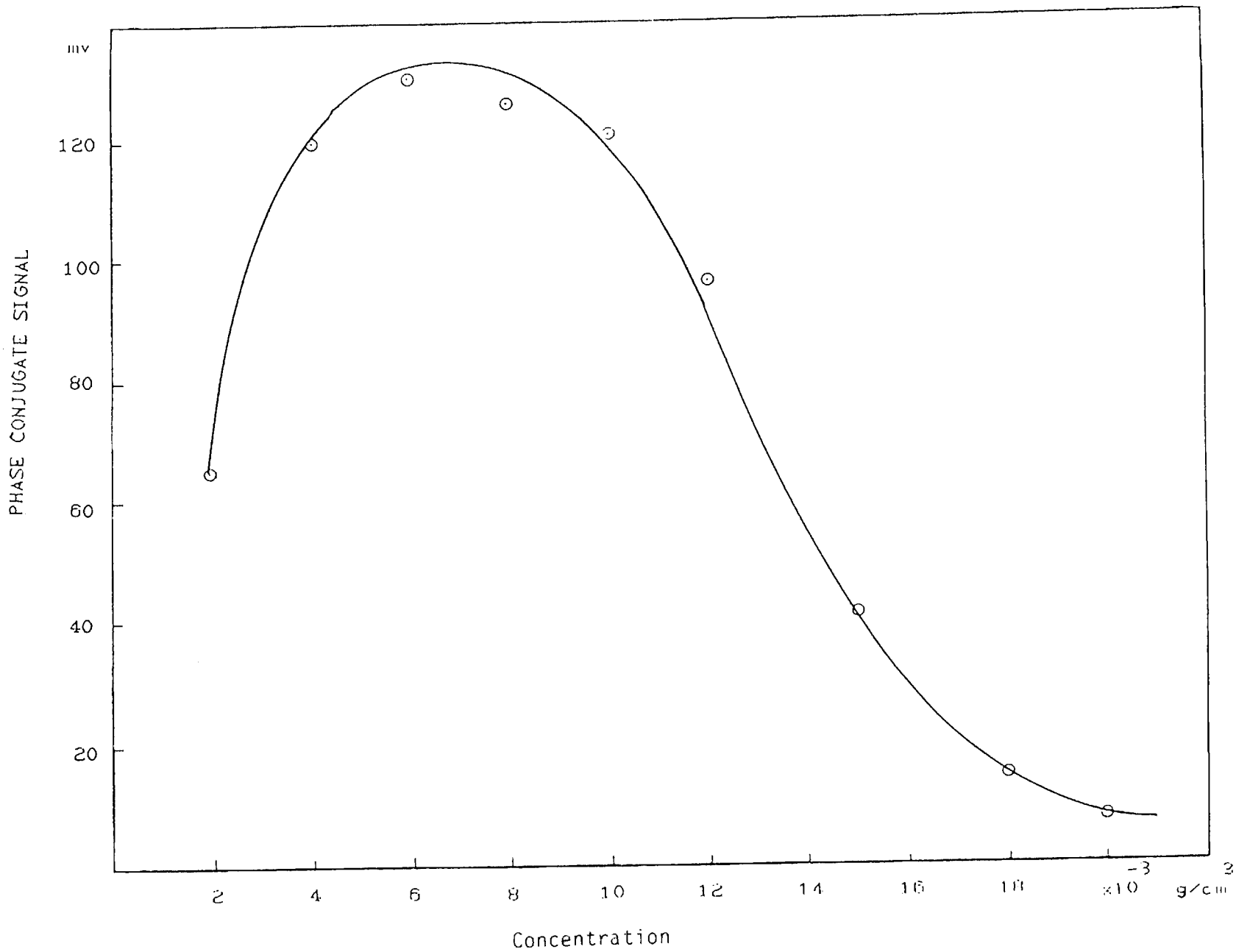

Figure 3. Dependence of phase conjugate signal on concentration of disodium fluorescein.

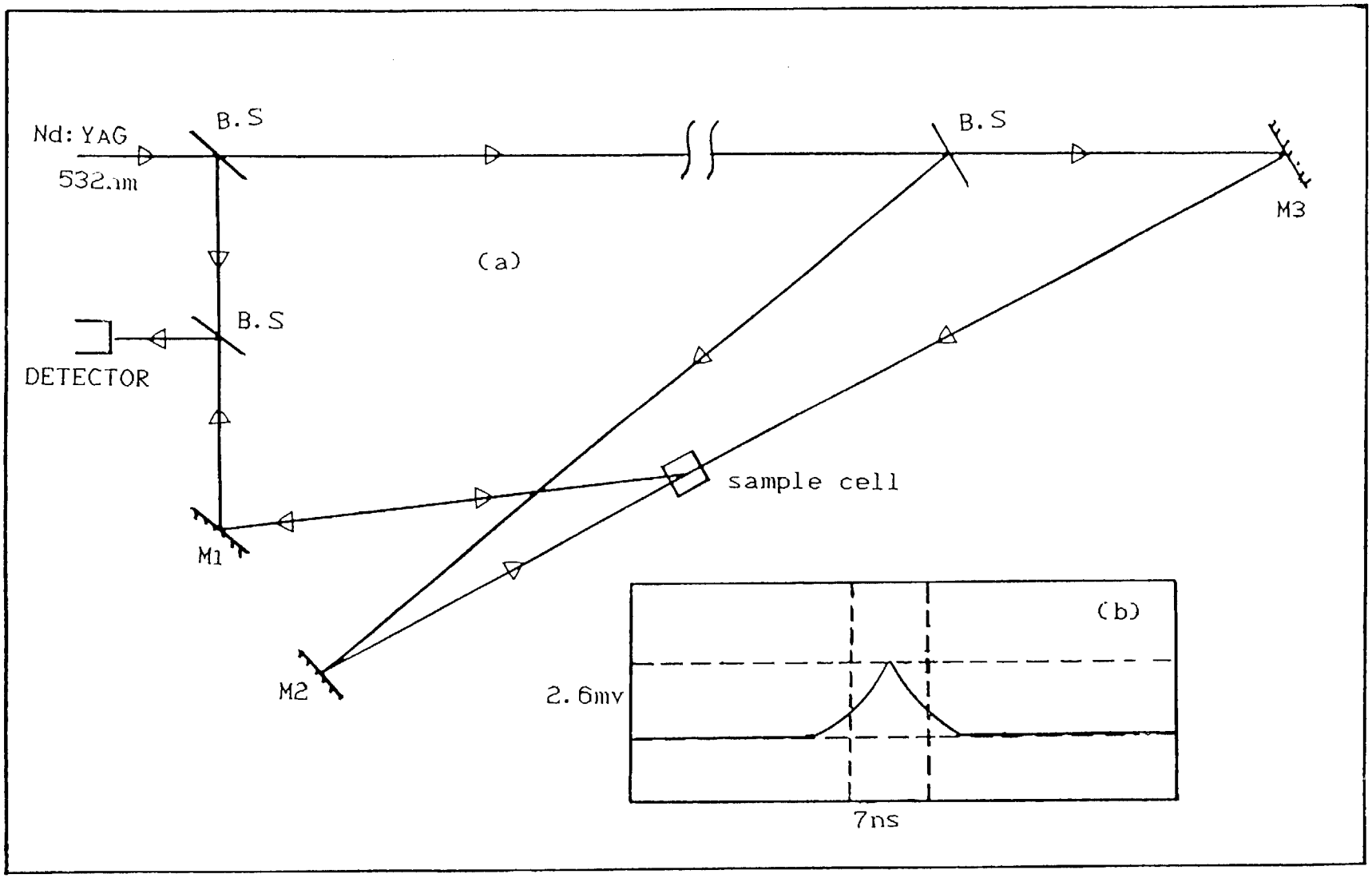

Figure 4. (a) The experimental layout with the pump beam #1 (A_1) delayed.
(b) The phase conjugate signal obtained at 12.2ns delay with respect to the probe (A_3)

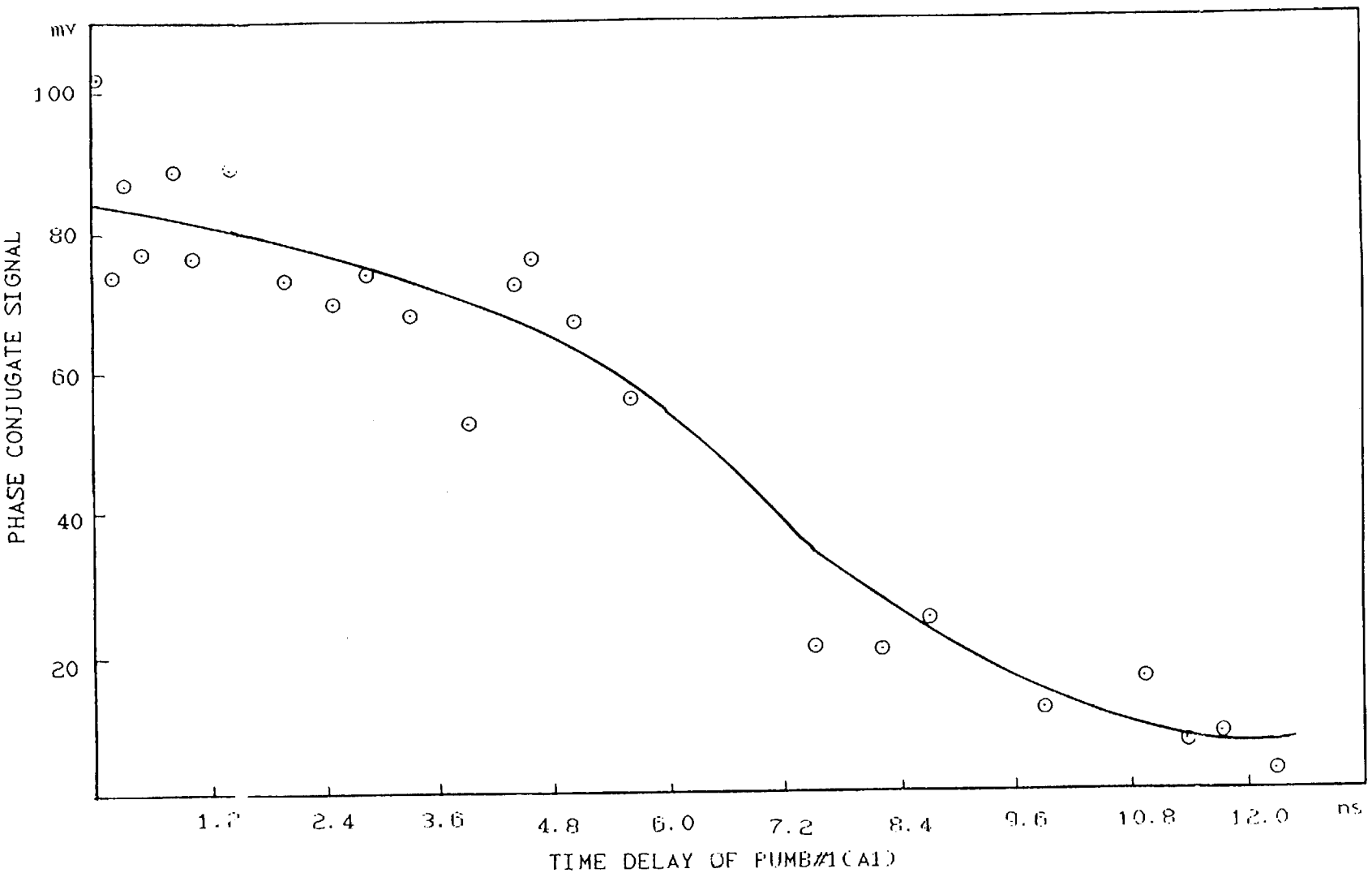

Figure 5. Dependance of the phase conjugate signal on the delay time of the pump beam (A_1).