Experiments on Nonlinear Optics

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Name of Contractor: The Regents of the University of California

Project Scientist: Y. R. Shen

Abstract

Preliminary results on self-trapped filaments in different liquids and liquid mixtures are reported. It is shown that stimulated Raman and Brillouin scattering is probably not responsible for the filament formation. Further work is needed to determine whether Kerr effect or field-induced phase transition is the principal mechanism for self-trapping.

Difference-frequency generation in the submillimeter region from quartz and $LiNbO_3$ by beating of two ruby laser beams is reported. The difference-frequency is tunable through thermal tuning of the laser frequencies. This is the first coherent, tunable, far-infrared pulse generator ever constructed.

I. Introduction

During the past six months, much of our effort has been spent in trying to understand the physical mechanisms responsible for the small-scale self-trapped filaments in liquids. The motives for carrying out experiments on self-trapped filaments are manifold. (1) From our previous experience in experiments on stimulated Raman and Brillouin scattering in self-focusing liquids,[1] we found that without understanding of the self-trapped filaments, it is basically impossible to understand the stimulated scattering process in these liquids. (2) The theory of Chiao et. al.[2] on self-trapping due to optical Kerr effect cannot fully explain the observed characteristics of the filaments.[3] (3) We proposed that the filaments might be the result of a possible field-induced phase transition in the liquid medium.[4] Experiments are needed to check the validity of such a radical assumption.

We have measured the characteristics of self-trapped filaments in different liquids and liquid mixtures under various conditions. While the results are not conclusive in favoring either of the existing theories, we do gain much insight to the possible physical mechanisms responsible for self-trapping Further experiments, much more carefully designed, are necessary for better understanding of the problem.

We have also succeeded in detecting far-infrared difference-frequency generation from quartz and $LiNbO_3$ using two ruby lasers operated at different temperatures, but Q-switched simultaneously. Our system is the first coherent, tunable, far-infrared pulse generator which has ever been constructed.

II. Self-trapped Filaments. (Michael M. T. Loy)

Our experimental set-up on the investigation of self-trapped filaments is similar to that of Chiao et. al., but precaution was taken to operate the laser in a single mode. Normally, above the threshold for self-focusing, only a single filament appeared in the self-focused beam. Two or three filaments were observed occasionally when the laser intensity was high. The characteristic of these filaments were similar to those observed by others[3]. For intensity well above the self-focusing threshold, the size of the filaments in a particular liquid was roughly a constant. There was no change in the filament size even if the laser intensity was varied by a factor of three. We measured through the densitometer trace in pure CS_2, nitrobenzene, and toluene a filament diameter of 5 ± 1, 20 ± 2, 10 ± 1 microns respectively. No correction on the measurements due to resolution of the microscope was made. The filaments were linearly polarized. The energy content in each filament in a particular liquid was also roughly a constant. The duration of a filament was typically less than 0.5 nsec., which is the limit of our detection system.

We also studied the problem of spectral broadening in a filament in pure CS_2. We found that if the laser output was of a single mode, then within the resolution limit, there was no broadening in either the laser or the Raman spectrum. When the laser was adjusted deliberately to put out two or more modes, a spectral broadening of around 100 cm^{-1} showed up readily. We therefore believe that spectral broadening must result from interaction of two or more laser modes with the liquid medium.

We have suspected that a field-induced phase transition could be responsible for the observed filaments.[4] In order to see whether molecular correlation, or possibly phase transition, plays an important role in the filament formation, we measured the characteristics of filaments in liquid mixtures as a function of relative concentration. We studied the mixtures of CS_2-toluene, CS_2-nitrobenzene, and CS_2-CCl_4.

The variation of the filament size with concentration of CS_2 in the three mixtures is shown in Figure 1. In all cases, the size change is more appreciable at low concentrations of CS_2. The intensity of the filament also increases with the increase of the filament size. The results show that intermolecular interaction is important in determing the filament characteristics. However, the results are certainly not sufficient to conclude whether there exists a field-induced phase transition. In Figure 2, we compare the experimental results for CS_2-CCl_4 with the

filament sizes calculated by assuming the refractive index change $\Delta n = n_2|E|^2$, where the values of n_2 for the mixture were taken from the work of Takatsuji.[5] No agreement is found as one would expect. Higher order terms in the Δn expansion must be important for filament formation.

In the course of investigation on liquid mixtures of CS_2-toluene and CS_2-CCl_4, we found that above certain critical concentration, a first-order anti-Stokes ring of about 20μ in diameter appears around the filament. There was also strong anti-Stokes at the center of the filament. While the anti-Stokes in the filament has been observed earlier and can be qualitatively explained as due to coupling between Stokes and anti-Stokes, the anti-Stokes ring mentioned here has never been reported. Since the ring is fairly sharp, it must have come from a semi-point source. Our tentative explanation is that the anti-Stokes could be originated from the self-focusing point, and could be partially trapped by reflections and then amplified among the filament. The partial transmission through the filament boundary near the end face of the liquid cell leads to the appearance of the anti-Stokes ring together with the anti-Stokes which remains trapped in the filament.

In self-focusing liquids, the stimulated Raman and Brillouin processes are usually inseparable from self-focusing and self-trapping. This raises the question whether stimulated Raman and Brillouin scattering could be the cause of filament formation. In fact, Townes suggested that the population redistribution among

the vibrational levels through stimulated Raman process could yield a sufficiently large change of refractive index Δn to account for the observed filaments. To eliminate the effect of stimulated Raman and Brillouin scattering on self-trapping, we used a toluene cell, followed by a 2 mm section of liquid mixture CS_2-nitrobenzene or CS_2-CCl_4. We found no detectable Raman radiation generated from the short section. The Brillouin radiation should be weak likewise. We noticed however that the filament in toluene, after propagating into the short section of mixture, turned into a filament of the mixture immediately. In CS_2-nitrobenzene, they appeared different as shown in Figure 3. This shows that the stimulated Raman and Brillouin scattering in nitrobenzene mixture may indeed affect the formation and the characteristics of the self-trapped filaments.

We are continuing measurements on the filaments using the two-cell method. In particular, we wish to find out experimentally the actual relationship between Δn and $|E|^2$, from which we can then conclude which mechanism is responsible for the formation of self-trapped filaments.

III. Tunable Far Infrared Pulse Generator (D. Faries)

The generation of difference-frequency in the far infrared through beating of two laser frequencies in a nonlinear crystal has been reported earlier.[6] However, in those experiments, the laser frequencies are fixed, and therefore the difference

frequency generated cannot be changed. We have now succeeded in constructing a tunable far-infrared pulse generator by adjusting the laser frequencies in the difference-frequency generation.

Our system consists of two ruby lasers with a common rotating prism for Q-switching. The two lasers are operated at different temperatures, so that the lasing frequencies are different. A difference-frequency around 10 cm^{-1} is being used in our experiments. The nonlinear crystals we have investigated are quartz (1 cm thick) and $LiNbO_3$ (1/3 mm thick). In both cases, we detected a far-infrared pulse with a peak power of few hundred microwatts around the phase matched direction. The far-infrared generation fell off as the crystal was tuned away from the phase-matched direction. This gave us the clear evidence that the signal from our far-infrared detector was indeed from the difference-frequency generation in the crystal.

More work is however needed to complete our investigation. In particular, the far-infrared signals should be normalized against the sum-frequency generation to reduce fluctuations, and the nonlinear susceptibilities for difference-frequency generation in different crystals should be measured.

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Caption of Figures

Fig. 1 Variation of the filament size with respect to the relative concentration (in volume fraction) in a mixture.

Fig. 2 Comparison of filament sizes measured in CS_2-CCl_4 with those calculated from $\Delta n = n_2|E|^2$

Fig. 3 Comparison of filament sizes measured in CS_2-Nitrobenzene with and without a toluene cell in the front.

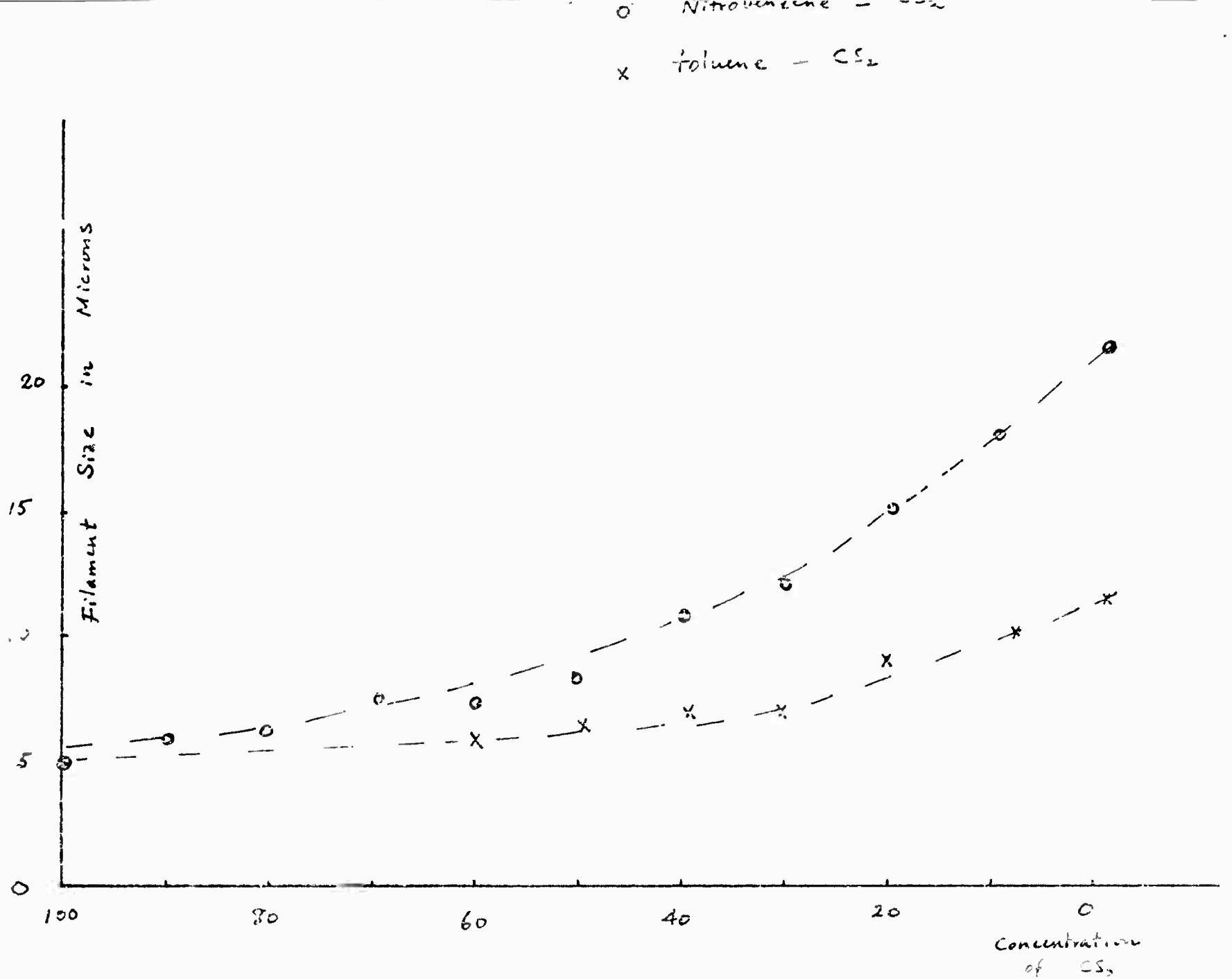
o Nitrobenzene – CS_2
x Toluene – CS_2
Filament Size in Microns
0
5
15
20
100
80
60
40
20
0
Concentration of CS_2

Fig. 1

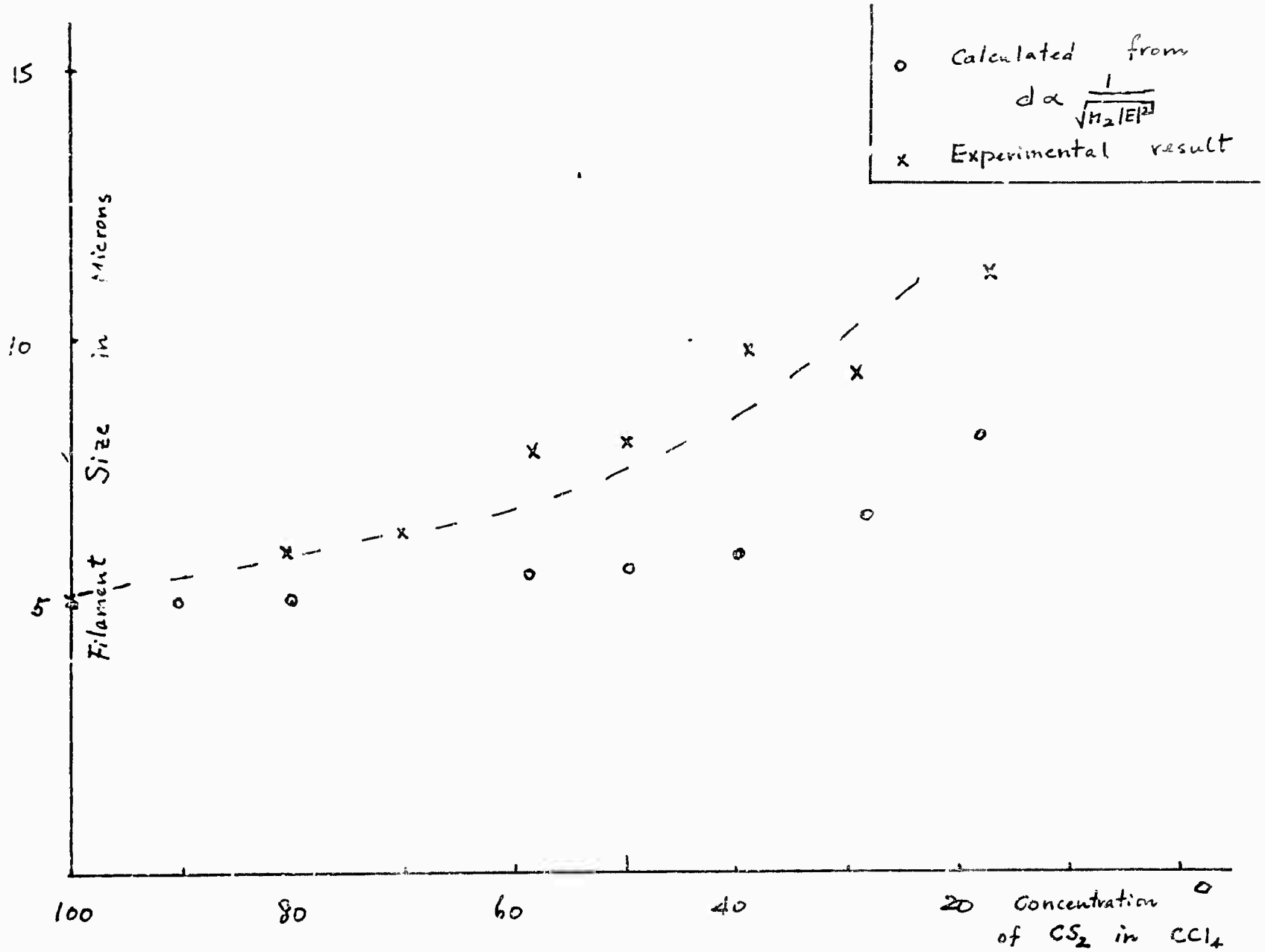
o Calculated from
$d \propto \frac{1}{\sqrt{H_2 |E|^2}}$
x Experimental result
15
10
5
Filament Size in Microns
100
80
60
40
20
0
Concentration of CS_2 in CCl_4

Fig 2

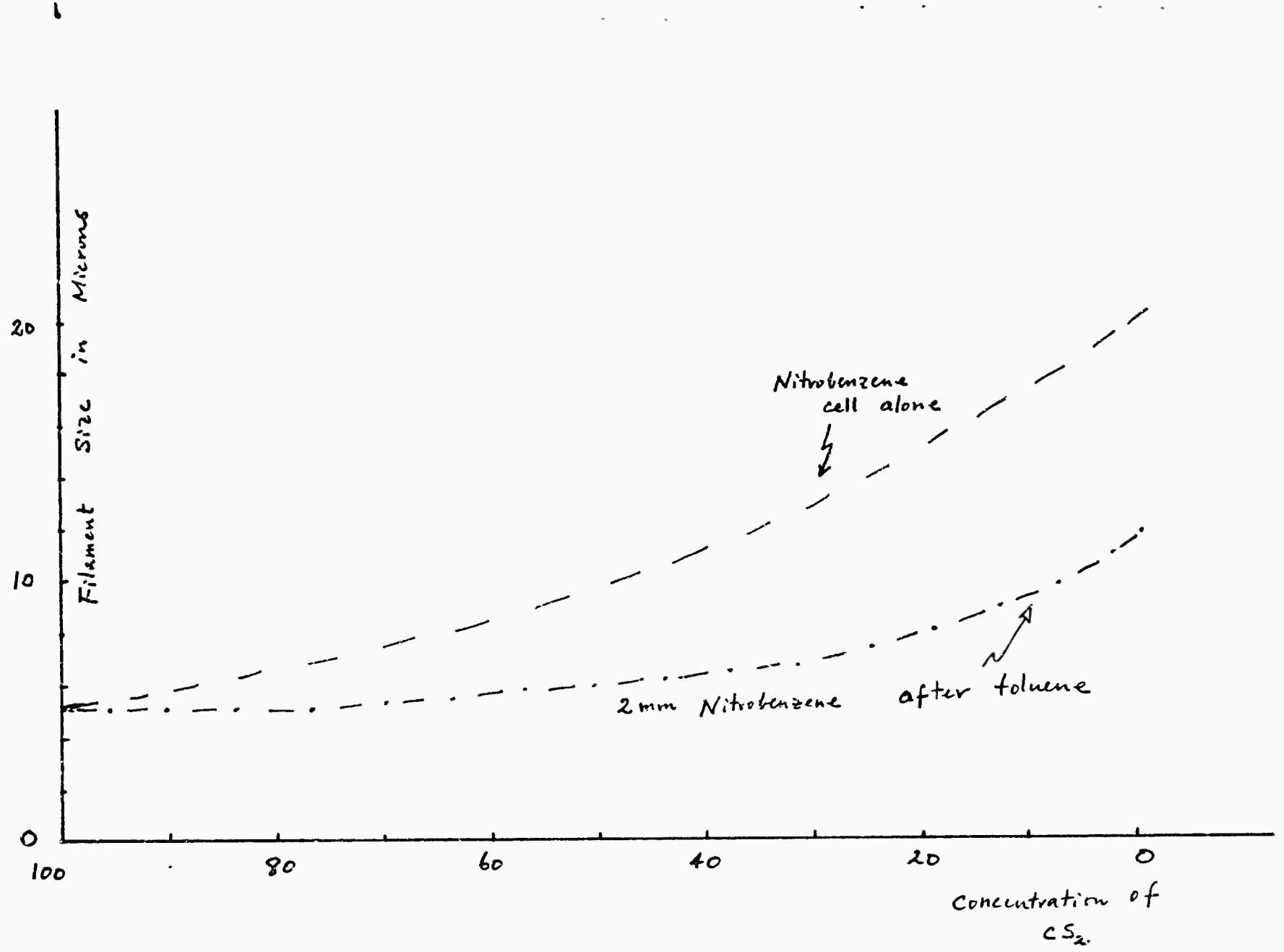
Filament Size in Microns
20
10
0
Nitrobenzene
cell alone
2mm Nitrobenzene
after toluene
100
80
60
40
20
0
Concentration of
CS_2.

Fig. 3

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Far-infrared pulse generator						