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STARK INDUCED OPTICAL TRANSIENTS IN RUBY

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ABSTRACT

Extension of the Brewer-Shoemaker technique of Stark induced optical transients to a solid, ruby, is reported. Optical free induction decay and photon echoes have been obtained as well as the first observation of an optical edge echo.

STARK INDUCED OPTICAL TRANSIENTS IN RUBY

In this paper we report on the first application of the Brewer-Shoemaker[1] technique of Stark induced optical transients to a solid extending our earlier[2] preliminary observations. Our motivations in doing this experiment are (1) to extend the Stark technique to solids which, as will be discussed later, allows studies not accesible to the conventional pulsed beam method and (2) to further study coherence times in ruby using new techniques with a view towards examining the order of magnitude discrepancy in the measured[3] photon echo decay times using pulsed and gated CW ruby lasers.

The use of lasers to study homogeneous linewidth in gases and solids has produced much new information on the interactions which determine linewidth. Homogeneous linewidths in solids have been observed in two general ways; (1) by observation in the frequency domain, e.g. fluorescence line narrowing[4] and absorption hole burning[2,5,6] and (2) by observation in the time domain, e.g. coherence time (T_2) measurements using photon echoes[7] or free induction decay[8] (FID). The techniques are presumably complementary (although as yet rigorous equivalence has not been demonstrated for solids) and for certain linewidth regions each has its particular advantages. For example in dilute ruby at 4°K and zero magnetic field, the homogeneous linewidth of the R_1 line is about 60 MHz (FWHM) as measured by fluorescence line narrowing[4] and hole burning techniques[5,6]. This corresponds to $T_2 = 6$ nsec. This short decay time makes accurate measurements difficult and the frequency domain approach seems preferred. On the other hand, for 0.005% ruby in 6 K gauss,

$T_2 \approx 13$ μsec (ref.3) corresponding to a linewidth of 25 kHz which would be difficult to measure in the frequency domain, although sideband absorption[6] or Stark shifting[2,5] could in principle measure this width if a stable laser is available and spectral diffusion does not occur.

The sample ruby (Czochralski) was a slab 1.5 mm thick with a 0.03 wt % Cr_2O_3 concentration. The sample temperature was 4.2 to 5°K with a magnetic field of ∿ 1 K Gauss applied along the C axis. Transparent conducting coatings were applied to the ruby to allow propagation of a laser beam and application of an pulsed electric field parallel to the c axis. The circularly polarized laser source was a CW ruby laser oscillator-amplifier combination[9] producing a single mode output of up to 100 mw. The laser was thermally tuned to the peak of one of the Cr^{3+} $\bar{E} \rightarrow (\pm 1/2)^4A_2$ transitions near 693.4 nm. The R_1 line Stark shift was measured by fluorescence line narrowing[5] and found to differ substantially (∿ 25%) from the initial work[10] on Stark effects in ruby and to be in good agreement with the later results of Cohen and Bloembergen.[11] The Stark shift is 0.110 ± 0.008 MHz/volt cm^{-1}. The heterodyne beat between the transmitted beam and transients was detected by a silicon photodiode and averaged by a boxcar integrator using a 1 sec time constant. The large noise present due to laser spiking was greatly suppressed by subtracting diode signals observed before and after the sample. An aperture selected ∿ 10% of the beam in the centre. The repetition rate of the Stark pulses was in the range 500 - 2000 Hz.

Examples of optical FID and photon echoes are shown in Figs. 1 and 2. The observed beat period for the FID is 28.3 ± 0.2 nsec in excellent agreement with the calculated value of 27.3 ± 2.2 nsec. The period was observed to track the pulse voltage as expected. The behaviour of the FID following a square pulse could be easily seen in real time on an oscilloscope as the pulse length was varied. As the pulse width t_p was increased from short values, the FID amplitude and decay time increased until a maximum was reached near $t_p \sim 1 \mu\text{sec}$ (Fig. 1). The decay time in this region is clearly controlled by the pulse width (i.e. bandwidth of excitation). As the t_p was further increased the FID amplitude and decay time decreased reflecting the gradual onset of power broadening[8] (the power broadened decay rate for infinite t_p for the conditions of Fig. 1 and 3 is 23 nsec). In the initial stages of the hole burning, coherence effects remain and the induced polarization contains frequency structure due to excitation of off resonance dipoles. When these dipoles radiate, interference effects arise leading to the modulated FID shown in Fig. 3. This behaviour at high turning angles is similar to the edge echo phenomena observed[12] in NMR. We believe this is the first clear observation of optical edge echoes of which some experimental indications were reported earlier by Brewer and Shoemaker[13] as well as theoretical studies by Hopf and Scully.[14]

Our measurements of photon echo decay show an exponential shape over three orders in echo power. The measured T_2 for the present concentration and magnetic field is 2000 ± 500 nsec corresponding to a homogeneous linewidth of 160 kHz. The large indicated fluctuation represents variations seen in different measurements, presumably arising from T_2 spatial fluctuations in the sample. The echo decay shape and rate obtained here agree with earlier[3] studies (T_2 = 2800 nsec) using a gated CW laser.

We had also reported earlier,[2] observations of optical nutation. For reasons not understood as yet, the nutation results are not reproducible for different samples and position in the sample.[15] We can reproducibly obtain a oscillation of the absorption vs time, however we do not always see a negative absorption at the reversal. This may be related to residual absorption effects due to pairs or impurities.[16]

The echo results further emphasize the order of magnitude discrepancy[3] observed between decay times measured using flashlamp pulsed and CW lasers. This is a startling difference, as yet unexplained, which raises serious doubts as to the validity of coherence time measurements using transient decay methods. A possible determining factor may be laser frequency jitter or chirps. For FID, the decay rate will obviously be affected by laser jitter in the preparation stage. For photon echoes however, it is not clear what effect jitter will have on the decay rate, in particular where the second pulse is a delayed version of the first. The gated CW results of Liau and Hartmann[3] suggest that the short time jitter of the CW ruby laser is as low as 25 kHz. We have measured[9] an instrument limited laser linewidth of 100 kHz for short times (∿ 100 μsec) using a heterodyne technique.

Finally we discuss some advantages of the Stark technique of inducing transients vs pulse methods. A number of these have been listed by Brewer and Shoemaker.[1] A distinct advantage is experimental simplicity in that optical shutters for sequentially pulsing the laser and blocking the photodetector are not required. In addition we are not limited to low pulse repetition frequencies often demanded by such shutters. This, of course allows improved signal to noise ratios. On the other hand more elaborate preparation of the sample is required, although this could be obviated using frequency switching methods.[8] Another advantage is that frequency jitter can be readily simulated to study its effects on coherence decay times. Such studies as well as other frequency sweeping experiments will be reported later.

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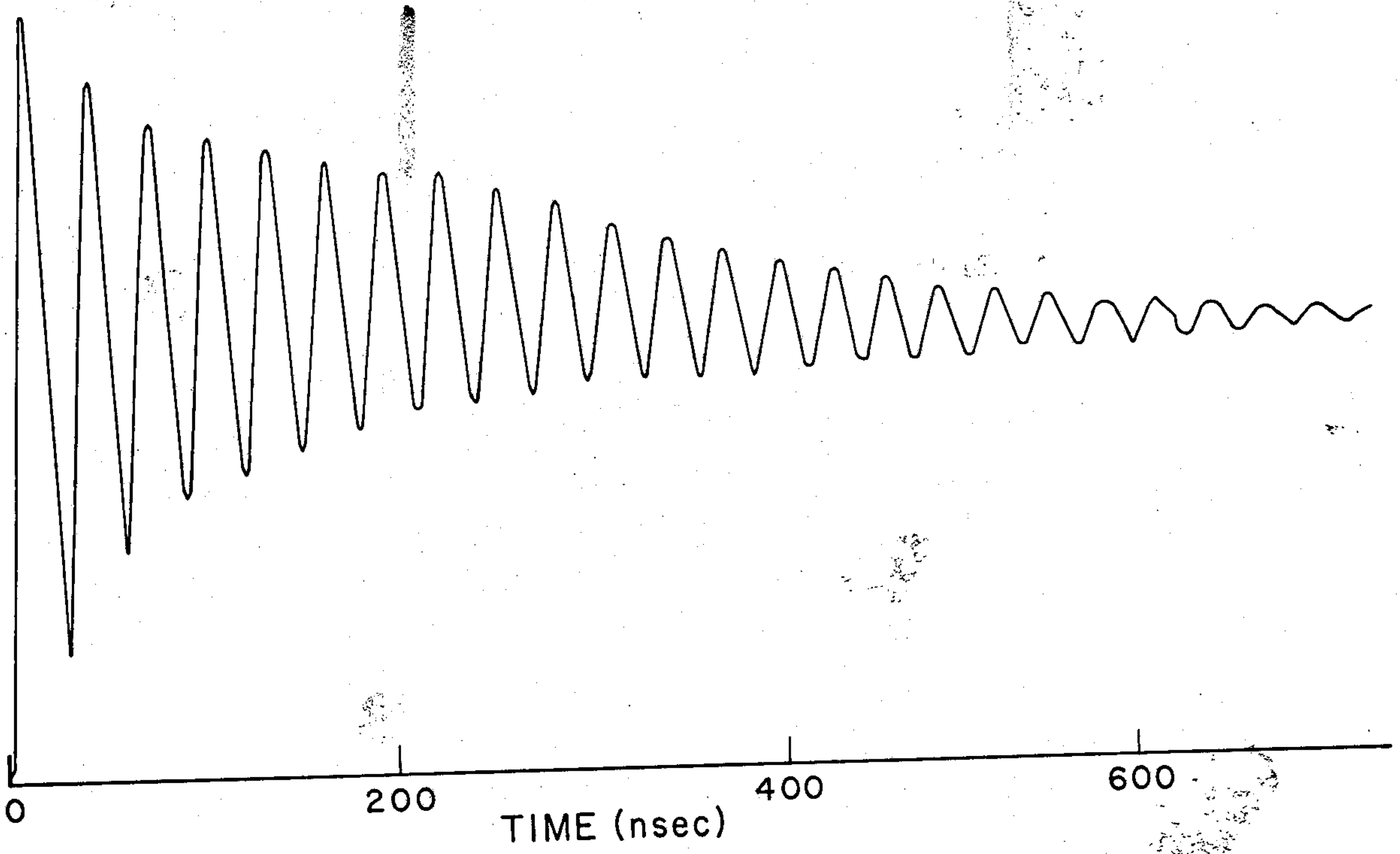
FIGURE CAPTIONS

Fig. 1. Optical free induction decay in ruby following a 0.9 μsec 50 volt Stark pulse of angle = 0.53π. A magnetic field of 1.5 K gauss is applied along the c axis.

Fig. 2. Photon echo in ruby. The arrow indicates the expected[1] position of the echo. A magnetic field of 1.0 K gauss is applied along the c axis.

Fig. 3. Optical free induction decay in ruby showing edge echo behaviour following a 7 μsec, 50 volt Stark pulse of angle ∼ 4π . A magnetic field of 1.5 K gauss is applied along the c axis.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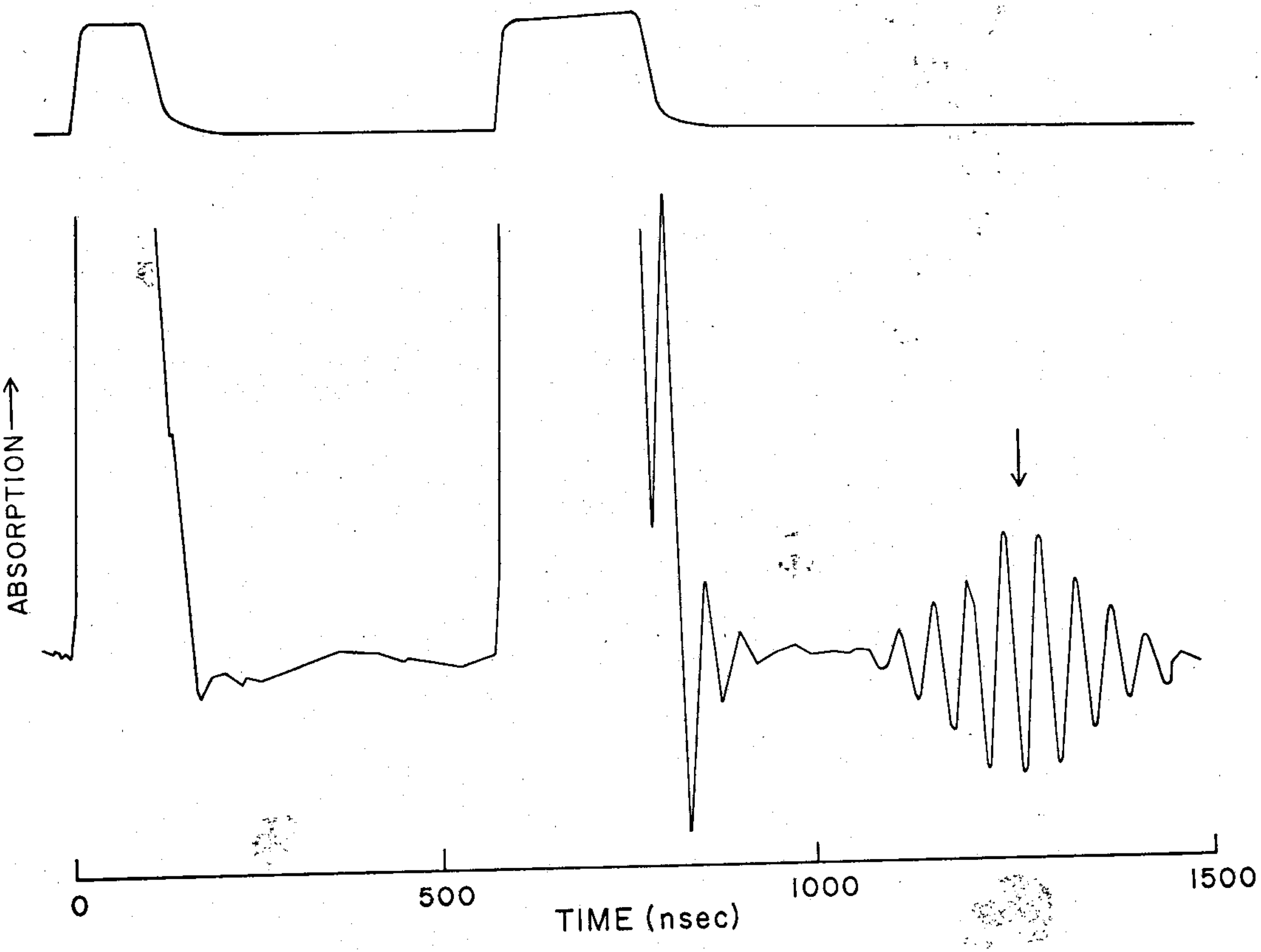
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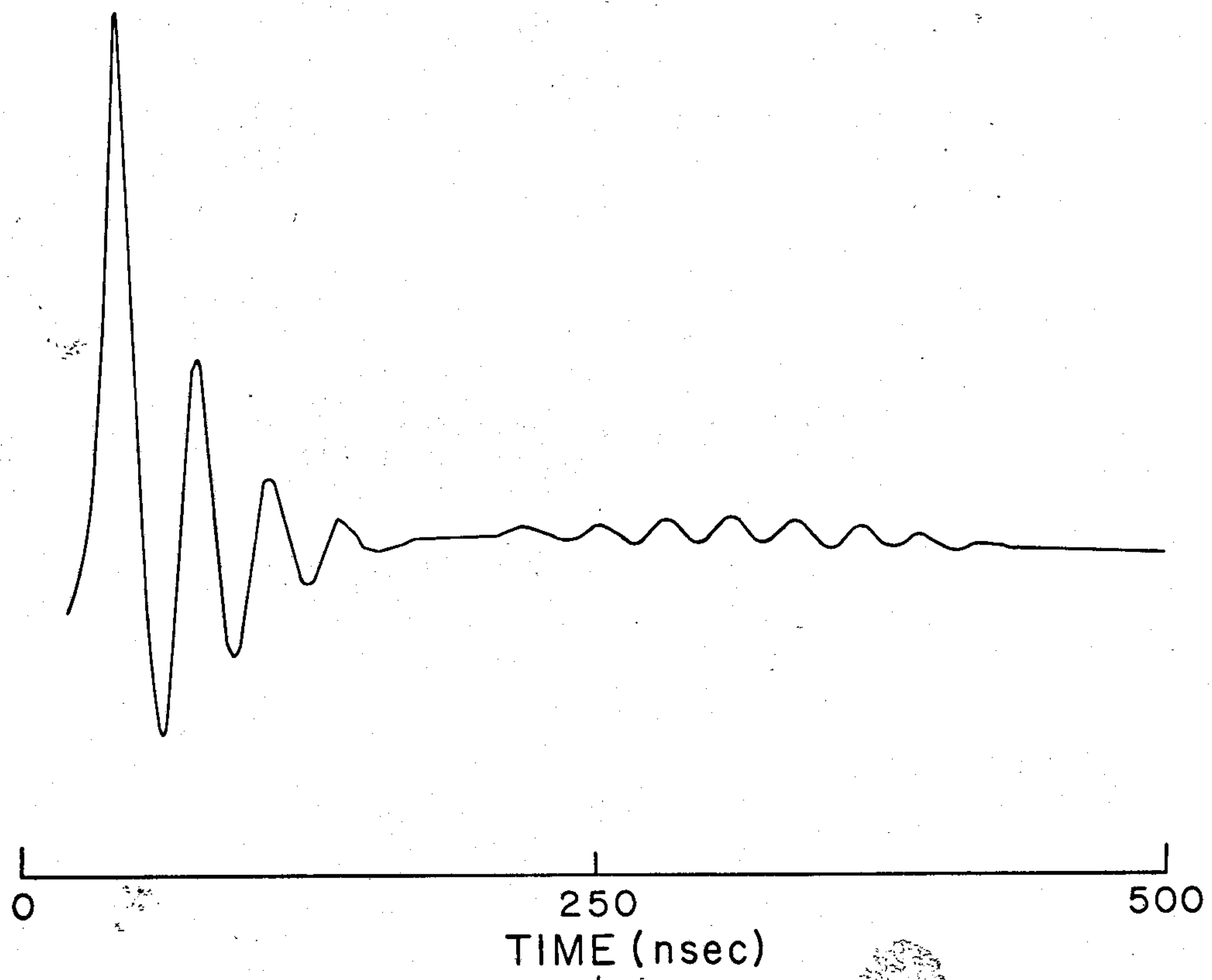
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