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LASER PULSES  
BY DIFFERENT ORDER PHOTOELECTRIC EFFECTS



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MEASUREMENT OF MODE-LOCKED ULTRASHORT LASER  
PULSES BY DIFFERENT ORDER PHOTOELECTRIC EFFECTS

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#### ABSTRACT

A simple experimental method consisting of simultaneous oscilloscopic observation of current signals induced by linear and nonlinear photoelectric effects is given for the determination of the duration and intensity of ultrashort pulses.

#### РЕЗЮМЕ

Описывается простой метод экспериментального определения длительности и интенсивности ультракоротких лазерных импульсов. Метод основан на одновременном осциллоскопическом наблюдении сигналов, исходящих от нелинейных фотоэмиссионных процессов различных порядков, вызванных лазерными импульсами.

#### KIVONAT

Ultrarövid laserimpulzusok időtartamának és intenzitásának meghatározására szolgáló, egyszerű kísérleti módszert ismertetünk. A módszer lényegét különböző, lineáris illetve nemlineáris fotoeffektusból származó elektromos impulzusok megfigyelése képezi.



## INTRODUCTION

The widespread application of mode-locked lasers in quantum electronic research requires the measurement of the time duration and intensity of the pulses in the train. The duration of these pulses varies between the natural bandwidth limited  $10^{-13}$ - $10^{-12}$  sec [1,2] and  $10^{-9}$  sec corresponding to an artificially narrowed spectral width [3]. The time duration can be measured by

- 1./ direct electronic methods /photocell [3,4] and streak camera [2,5] / or by
- 2./ indirect nonlinear methods /two-photon fluorescence [6], harmonic generation [7,8], auto Kerr effect [9]/.

With the help of the direct electronic methods  $\sim 70$  psec resolution was achieved by using photocells and about 5 psec resolution by streak cameras. The latter is nearly sufficient for the observation of pulses in the picosecond range, but the camera is rather complicated and expensive. The photocell is very convenient but can be used only above 100 psec pulse duration. Two-photon fluorescence /TPF/ is the most general indirect method but it gives only an average value for the whole train without any information about the duration of the individual pulses, which varies from pulse to pulse. Other nonlinear methods [7, 8, 9] combine effects of different order nonlinearity to avoid the inadequacy of the TPF method.

The method described here is a technically very simple indirect nonlinear method in which the problem is solved by the simultaneous oscilloscopic registration of current signals induced by linear and nonlinear photoelectric effects which are detected using a linear and a nonlinear photocell and two oscilloscopes /or, if necessary, only one/.



PRINCIPLE OF THE MEASUREMENT

It is well known that in the case of an  $n^{\text{th}}$  order photoelectric effect the dependence of the photocurrent  $j$  on the light intensity  $I$  is

$$j \sim I^n$$

In log-log coordinates this expression gives a straight line with a slope  $n = \left[ \frac{\text{work function}}{\text{photon energy}} + 1 \right]_{\text{int}}$ , as confirmed by experiments [10, 11].

It was noted in our paper on the nonlinear photoelectric effect /NLP/ experiment, however, that a marked asymmetry was observed in the ascendent and descendent halves of the laser pulse train: linear signals  $V_L$  of the same magnitude but from different halves of the train corresponded to nonlinear signals  $V_{NL}$  of very different magnitudes /see Fig. 1/. This anomaly was explained by the lengthening of successive pulses of the train, which is indicated by both theory and experiment [7, 11, 12].

The experimental arrangement shown in Fig. 2. is similar to that used for the NLP and field emission measurements [11, 12]. The time integrals of signals  $V_L$  and  $V_{NL}$  registered on oscilloscopes<sup>+</sup>  $O_1$  and  $O_2$  are proportional to the numbers of electrons  $N$  and  $M$  emerging from the linear and nonlinear photodetectors, respectively; these numbers are at the same time proportional to the first and the  $n^{\text{th}}$  power of the intensity  $I(t)$ . The numbers of electrons induced by the  $i^{\text{th}}$  pulse of the mode-locked laser train are:

$$N_i = \frac{a}{R_1} \int_{-\infty}^{\infty} V_{iL}(t) dt = \alpha \int_{-\infty}^{\infty} I_i(t) dt \quad /1/$$

$$M_i = \frac{a}{R_2} \int_{-\infty}^{\infty} V_{iNL}(t) dt = \beta_n \int_{-\infty}^{\infty} I_i^n(t) dt \quad /2/$$

where  $\alpha$  and  $\beta_n$  are the yield constants of the linear and nonlinear photoelectric effect,  $a^{-1} = (1,6 \cdot 10^{-19})$  [coulomb],  $R_1$  and  $R_2$  are the photocell resistances. The real intervals for integration do not reach the neighbouring pulses and, in general, the whole calculation is valid only for pure mode-locked pulses without any satellites.

Assuming that the time dependences of  $I(t)$  and  $V(t)$  are of the form  $\exp[-(t/\tau)^2]$  and  $\exp[-\frac{t}{T}]$  respectively:

$$N_i = \frac{a}{R_1} V_{iL}(0) T_{O1} = \sqrt{\pi} \cdot \alpha I_i(0) \tau_i \quad /3/$$

$$M_i = \frac{a}{R_2} V_{iNL}(0) T_{O2} = \sqrt{\pi} \cdot \beta_n I_i^n(0) \frac{\tau_i}{\sqrt{n}} \quad /4/$$

<sup>+</sup>  $V_L$  and  $V_{NL}$  can be observed on the same oscilloscope by delaying one of the signals with respect to the other by a portion of the pulse separation  $2L/c$  in the mode-locking train.



where  $T_{O1}$  and  $T_{O2}$  are the oscilloscopic resolutions and  $\tau_i$  is the half width of the  $i^{\text{th}}$  light pulse.

From the two equations

$$\tau_i = \frac{1}{\sqrt{\pi}} \left[ \frac{N_i^n}{M_i} \frac{1}{\sqrt{n}} \frac{\beta_n}{\alpha^n} \right]^{\frac{1}{n-1}} \quad /5a/$$

$$I_i = \left[ \frac{M_i}{N_i} \sqrt{n} \frac{\alpha}{\beta_n} \right]^{\frac{1}{n-1}} \quad /5b/$$

while the relative values are

$$\frac{I_j}{I_i} = \left[ \frac{M_j N_i}{M_i N_j} \right]^{\frac{1}{n-1}} = \left[ \frac{v_{jNL}(o) v_{iL}(o)}{v_{iNL}(o) v_{jL}(o)} \right]^{\frac{1}{n-1}} \quad /6a/$$

$$\frac{\tau_j}{\tau_i} = \left[ \frac{N_j^n M_i}{M_j N_i^n} \right]^{\frac{1}{n-1}} = \left[ \frac{(v_{jL})^n v_{iNL}}{v_{jNL}(o) (v_{iL}(o))^n} \right]^{\frac{1}{n-1}} \quad /6b/$$

Thus, by measuring  $M_i$  and  $N_i$ ,  $\alpha$  and  $\beta_n$  being known [10, 11, 12], the absolute values of  $\tau_i$  and  $I_i$  can be obtained from equ. 5, or alternatively, without knowing  $\alpha$  and  $\beta_n$  relative values for  $I_j/I_i$  and  $\tau_j/\tau_i$  can be simply calculated from equ. /6/.

From the ratio  $\theta_i = \tau_{i+1}/\tau_i$  and the difference  $\Delta\tau_i = \tau_{i+1} - \tau_i$  the pulse duration is

$$\tau_i = \frac{\Delta \tau_i}{\theta_i - 1} \quad /7/$$

The value of the pulse lengthening  $\Delta\tau_i$  can be easily estimated with the help of the following simplified model. Let us assume that one ultrashort pulse is of the minimum duration  $\tau_o$  permitted by the bandwidth  $\Delta\Omega$  of a mode-locked Nd glass laser. Taking into account that for two frequencies separated by  $\Delta\Omega$  the transit times on a distance  $x$  in a refractive medium are different, the lengthening of the pulse will be

$$\Delta\tau = \frac{x}{c} \left[ \frac{\partial\eta}{\partial\Omega} \Delta\Omega + \frac{1}{2} \frac{\partial^2\eta}{\partial\Omega^2} (\Delta\Omega)^2 \right] = \frac{x\Delta\Omega}{c} \left[ \frac{\partial\eta}{\partial\Omega} + \frac{\Delta\Omega}{2} \frac{\partial^2\eta}{\partial\Omega^2} \right] \quad /8/$$

where  $\eta$  is the index of refraction and  $c$  is the velocity of light in vacuum. For pulses of the same bandwidth the dispersion lengthening will be



the same, regardless of their original time durations. (If the lifetime of the dye is comparable to or shorter than the pulse duration, the dispersion lengthening will be limited by the dye, as was shown recently by the experiments of Babenko et al. [13].)

From /7/ and /8/ we can get

$$\tau_i = \frac{x\Delta\Omega}{C} \frac{1}{\theta_i - 1} \left[ \frac{\partial\eta}{\partial\Omega} + \frac{\partial^2\eta}{\partial\Omega^2} \frac{\Delta\Omega}{2} \right] \quad /9/$$

The intensity dependence of the refractive index  $\eta = \eta_0 + \eta_2 E^2$  may also cause modifications in the shape of the pulse, though these are negligible compared with the dispersion lengthening at the power values used. It should be noted, however, that the  $\eta(E^2)$  dependence may alter the time substructure of a given single picosecond pulse by self-phase modulation [1, 14, 15, 16]. Furthermore, the second term in  $\eta$  could modify the diameter of the beam [15] /which was assumed to be constant/, the power density being above the self-focusing threshold [17]. Considering, however, that in our laser TEM<sub>00</sub> mode distribution was achieved and filaments were never observed, we may safely assume that the change of the beam cross section is small, all the more because the variation of the beam diameter would be of opposite sign in the ascendant and descendant halves of the mode-locking train, whereas the experiment shows a continuous increase in duration from pulse to pulse /Fig.4/.

## RESULT

From our former measurements [11, 12] with a Nd glass laser and Au target, where the order of the NLP was  $n=4$ , for  $\beta_4$  we obtained the value  $\sim 4 \cdot 10^{-97}$  [electron/cm<sup>2</sup>] / [(photon/cm<sup>2</sup> sec)<sup>4</sup> sec] while the efficiency  $\alpha$  /including the transmission of the filters/ was about  $10^{-8}$  electron/photon. Typical values for N and M were  $\sim 10^{10}$  and  $4 \cdot 10^8$  electrons, respectively. Thus using /5a/

$$\tau \sim 8 \cdot 10^{-12} \text{ sec}$$

It should be emphasized that  $\beta_4$  was determined by TPF method, so that our  $\tau$  determination is an indirect, calibrated time duration measurement only.

In Fig. 1 the  $V_L - V_{NL}$  pulse pairs of a mode-locking train are plotted in log-log coordinates. Note that at the beginning and end of the train the different  $V_{NL}$  signals correspond to  $V_L$  signals of the same magnitude. The asymmetry is due to the fact that the envelope of the  $V_L$  monitor signal train differs from the calculated /equ. /6a/ / real intensity envelope. An



average of normalized monitor trains and the corresponding calculated intensity variation can be seen in Fig. 3.

Fig. 4 shows the relative changes of pulse duration calculated from linear-quadratic /Cs<sub>3</sub>Sb cathode, n = 2 / and linear-fourth order /Au-cathode, n=4 / photoelectric effect measurements. The greater errors of the second order measurements are due to the non - TEM<sub>00</sub> mode of operation and poorer reproducibility. From Fig. 4 :

$$\theta_1 = \frac{\tau_2}{\tau_1} \approx 1,1$$

In our experiment  $x \sim 2(\ell+d) \sim 50\text{cm}$ , where  $\ell$  and  $d$  are the lengths of the rod and the dye cell, respectively,  $\Delta\Omega \sim 2 \cdot 10^{13}\text{Hz}$  and  $\left[\frac{\partial\eta}{\partial\Omega} + \frac{\partial^2\eta}{\partial\Omega^2} \frac{\Delta\Omega}{2}\right] \sim 10^{-17}\text{sec}$  [12, 18], so for  $\tau$  with the use of /9/ we obtain

$$\tau_1 = [3 \div 4] \cdot 10^{-12} \text{ sec}$$

while

$$\tau_{10} = [6 \div 8] \cdot 10^{-12} \text{ sec}$$

This value is in agreement with the value of  $\sim 8$  psec obtained from /5a/ and with the  $[5 \div 10] \cdot 10^{-12}$  sec duration obtained with TPF method [11,12]. This agreement shows that our assumptions are valid.

The absolute intensity from /5b/ is

$$I \sim 10^{29} \frac{\text{photon}}{\text{cm}^2 \text{ sec}} \sim 20 \text{ GW/cm}^2$$

## CONCLUSION

The described method enables us

- a./ to determine the variation of the real peak intensity and pulse duration in a mode-locked train<sup>FF</sup>
- b./ to estimate the absolute peak intensity and pulse duration from the linear and nonlinear photoelectric signals/  $\alpha$  and  $\beta_n$  being known/
- c./ to calculate the pulse duration/without  $\alpha$  and  $\beta_n$ / from the bandwidth, dispersion and relative lengthening.

As the value of  $\tau$  may change in a wide range for different trains,  $\theta = \tau_{i+1} / \tau_i$



varies from 1,03 to 1,30 explaining the observed change in the degree of asymmetry /see Fig. 2/. For a relatively greater initial pulse duration  $\tau \sim 10^{-11}$  sec the asymmetry vanishes, or at least becomes less than the experimental error.<sup>333</sup>

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<sup>33</sup> Recently the variation of pulsewidth in a mode-locking train was also observed by direct electronic method [09P] , and gave similar results [19].

<sup>333</sup> In our experiments on the NLP [10, 11, 12] only these measurements of small asymmetry were taken into account, and the pulse - lengthening problem was avoided by automatic selection of trains containing  $\tau \sim 10^{-11}$  sec pulses only.



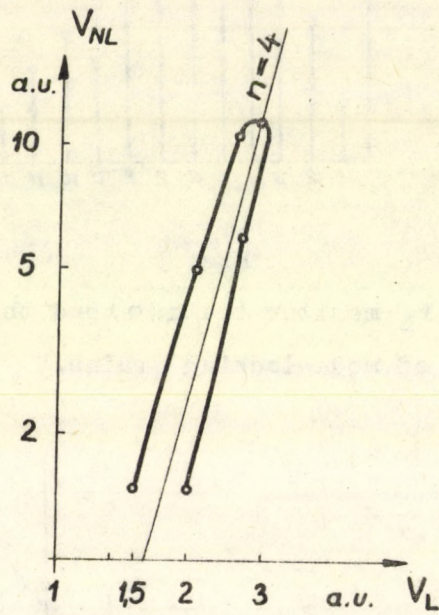


Fig. 1

The asymmetry in the variation of the  $V_{NL}$  nonlinear signal as a function of the signal  $V_L$ .

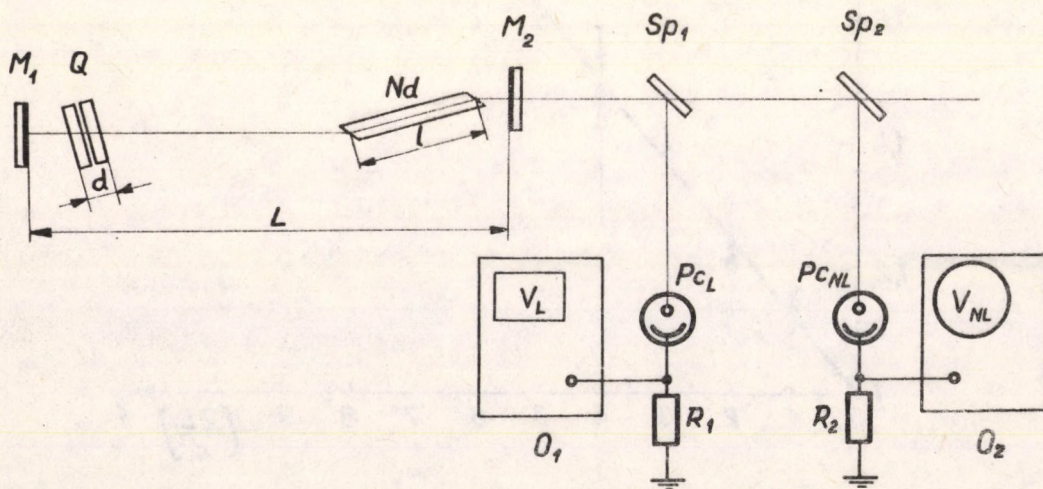


Fig. 2

The experimental arrangement:  $M_1$  and  $M_2$  mirrors;  $Q$ -bleachable dye /N<sup>o</sup> 3955/;  $Nd$ -Brewster-ended  $Nd$  glass;  $Sp_1$ ,  $Sp_2$  -splitters;  $Pc_L$  and  $Pc_{NL}$  -linear and nonlinear photocells;  $R_1$ ,  $R_2$ -resistances;  $O_1$  and  $O_2$  -oscilloscopes with  $V_L$ ,  $V_{NL}$  signals, respectively.



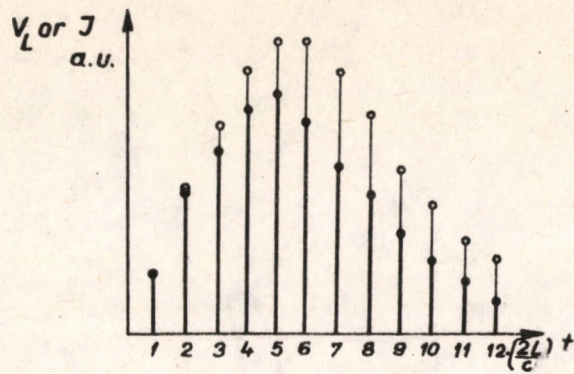


Fig. 3

Variation of the  $V_L$  monitor train:(o) and the calculated real intensity  $I$ :(•) of mode-locking trains.

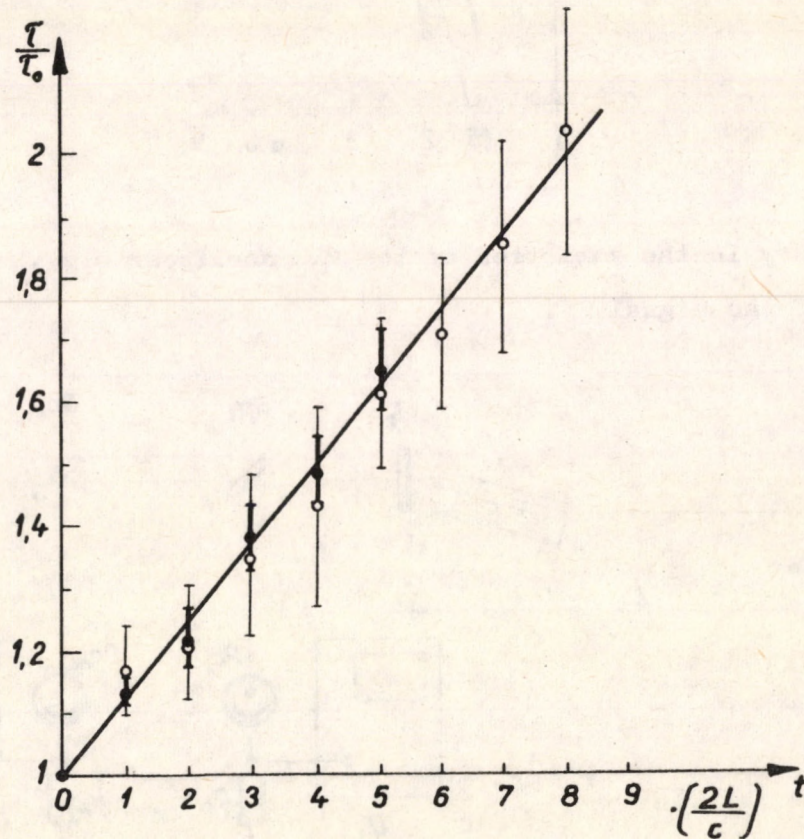


Fig. 4

The change of relative pulse duration from pulse to pulse in the linear-quadratic:(o) and the linear-fourth order:(•) photoelectric effect.



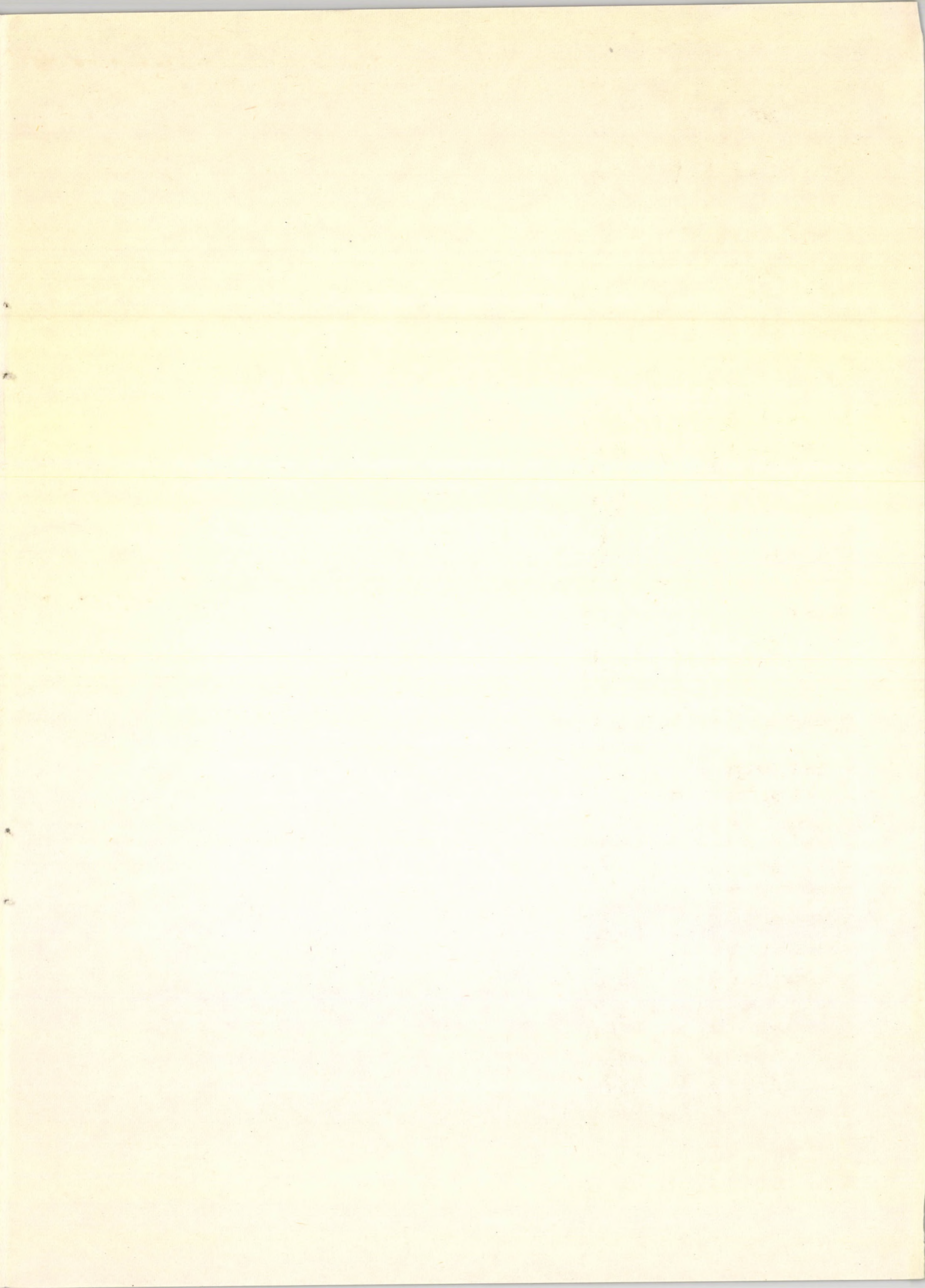
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