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Transient Self-Focusing in a Nematic Liquid Crystal in the Isotropic Phase

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ABSTRACT

We have made the first quantitative experimental study of transient self-focusing in a Kerr liquid by propagating Q-switched laser pulses in an isotropic nematic substance which has a long orientational relaxation time. Our results show good qualitative, and occasionally quantitative, agreement with the existing theoretical calculations.

Self-focusing of light in a Kerr liquid has been a subject of extensive investigation.[1] It gives rise to the observed intense streaks and many other related phenomena. Quasi-steady-state self-focusing is now well understood. Most of the observed results can be explained satisfactorily by the picture of moving foci.[2,3] Transient self-focusing is however still in a state of confusion. There already exist a number of detailed numerical calculations on the subject,[4-6] but no quantitative experimental results are yet available to check these calculations. The reason is obvious. Transient self-focusing occurs when the laser pulsewidth W is shorter than or comparable with the relaxation time τ of the field-induced refractive index Δn. Since in ordinary Kerr liquids, τ is in the picosecond range, picosecond pulses must be used in the transient studies.[7] Unfortunately, picosecond pulse

technology is still in such a primitive stage that quantitative measurements are extremely difficult and expensive. Experiments on transient self-focusing would be so much easier if nanosecond pulses can be used, but then the medium should have a relaxation time τ in the 50-nsec. range. This happens to be the case for liquid crystalline material in the isotropic phase.[8] In this paper, we would like to report the preliminary results of our experimental investigation on transient self-focusing in such a medium. Our results are in good agreement with the existing theoretical calculations.[5,6]

Let us first have a brief review on the theory of transient self-focusing. The phenomenon is presumably governed by the equations

$$[\nabla^2 - (\partial^2/c^2\partial t^2)(n_o + \Delta n)^2]E = 0 \tag{1}$$

$$(\tau\partial/\partial t + 1)\Delta n = \Delta n_o \tag{2}$$

where n_o is the linear refractive index and $\Delta n_o \cong n_2|E|^2$, n_2 being a constant. The solution of Eqs. (1) and (2) has been obtained numerically.[4-6] It can be described qualitatively from the following physical reasoning. If the input pulse is short, then Δn can never reach its steady-state value. We can write

$$\Delta n(z,\zeta) = (1/\tau)\int_{-\infty}^{\zeta} n_2|E(z,\eta)|^2 \exp[-(\zeta - \eta)/\tau]d\eta \tag{3}$$

where $\zeta = t - zn_o/c$ is the local time. For an input pulsewidth comparable to τ, Eq. (3) shows that the later part of the pulse may see a larger Δn. Consequently, different parts of the pulse propagate differently in the medium as shown in Fig. 1.[6] The front part (a,b) of the pulse sees little induced Δn and diffracts accordingly. It however leaves a sufficiently large Δn in the first section of the medium to cause the lagging part (c-f) to first self-focus and then diffract. Because of the transient response, both self-focusing and diffraction are expected to be gradual.

Figure 1 shows that the spatial distribution of the input pulse first deforms into a horn shape and then propagates on without much further

change.[6,9] If one probes the pulse at a point sufficiently deep in the medium, one would see a horn-shape pulse pass through; the pulse describing the variation of the on-axis intensity with time appears distorted as compared to the input pulse. The minimum diameter of the self-focused beam should be a sensitive function of the input power since it depends strongly on the magnitude of the induced Δn. However, it turns out that other nonlinear processes may set in to limit the diameter.[10-12] When this happens, we may find that the minimum diameter of the self-focused beam remains nearly unchanged for an appreciable section of the pulse, or in other words, the horn-shape pulse propagates over a finite distance with little change in its neck diameter. The numerical results of transient self-focusing, expressed in terms of the normalized parameters such as T/τ, etc., are given in Refs. 5 and 6.

In our experimental study, we used the liquid crystalline material MBBA in its isotropic phase(above T_K = 42.5°C) as the nonlinear medium. This material has a large steady-state optical Kerr constant(about 70 times that of CS_2 at 50°C) and an orientational relaxation time which varies with temperature from ~ 40 to > 800 nsec.[8] It is therefore ideal for study of transient self-focusing with Q-switched laser pulses. Our experimental set-up was the same as the one described earlier.[2,3] The input single-mode laser pulse had a diameter of 210 μm a pulsewidth W of 10 nsec. and a maximum peak power of 50 KW. The cell containing MBBA was sealed under N_2 atmosphere, and was thermally controlled to ±0.03°C in the range between 40 and 53°C. Self-focusing was readily observed in such a medium.

We first photographed the self-focused beam at the end of a 9.5cm cell and used the densitometer traces to measure the reduced beam diameter at different input power levels. The results for MBBA at 50°C are shown in Fig. 2 on a semilog plot. When the input peak power increases, the

beam diameter first decreases almost exponentially and then approaches a limiting diameter. As seen in Fig. 2, the nearly exponential decrease agrees very well with the theoretical curve extrapolated from the curves calculated by Shimizu.[6] At $P/P_{cr} = 20$ ($P_{cr} = 0.12$ Kw), some other nonlinear process presumably sets in to limit the beam diameter. At this power level, the peak intensity at the focus is several orders of magnitude below the avalanche breakdown threshold[12] and therefore breakdown cannot be the limiting mechanism. We also found that stimulated Brillouin scattering and stimulated Raman scattering do not appear until the input power reaches appreciably higher values as indicated in Fig. 2, and therefore they also cannot be responsible for the limiting diameter.[11] We suspect that two-photon absorption is probably the limiting mechanism in this case because of the near UV absorption band of MBBA. Anyway, this suggests that different nonlinear processes must be responsible for the limiting diameter of the self-focused beam in different cases.

The results at different temperatures are nearly the same after the correction due to scattering loss. The reason is as follows. In our case, $W \ll \tau$, and hence from Eq. (2), $\Delta n(t) \cong \int_{-\infty}^{t} \Delta n_o(t')dt'/\tau$ in the first-order approximation. Both Δn_o and τ due to molecular reorientation are proportional to $(T - T_c)^{-1}$ where T_c is the temperature at which a second-order isotropic-nematic transition would occur.[8] As a result, $\Delta n(t)$ is nearly independent of temperature except for the change of scattering loss with temperature. At 50°C or higher, the effect of scattering loss on self-focusing in our case is negligible.

We also measured the on-axis intensity variation of the self-focused beam with time at the end of the cell. In Fig. 3, we show the results. As the input power increases, the peak of the output intensity pulse first

appears (Fib. 3b) appreciably delayed from the peak of the input pulse (Fig. 3a) and then gradually moves (Fig. 3c-e) towards it. At sufficiently high input power, weak oscillation starts to develop on the lagging part of the pulse (Fig. 3d) and gets gradually more pronounced (Fig. 3e). At even higher input power, stimulated Brillouin scattering begins to show up. The above results agree qualitatively very well with the predictions derived physically from Fig. 1, and also with the numerical calculations of Refs. 5 and 6 (which also show oscillation in the output pulses at high input powers). Quantitative comparison between theory and experiment is presently not possible since no numerical calculation corresponding to the experimental parameters we used is available.

We also calculated from Fig. 3 the temporal variation of the self-focused beam diameter at the end of the cell by assuming the shape of the intensity profile remains nearly unchanged. The results are shown in Fig. 4a-d, which correspond to the intensity pulses of Fig. 3b-e respectively. It is seen that the incoming pulse does gradually develop into a horn shape as described qualitatively in Fig. 1. In particular, the neck diameter of the horn remains almost constant over a fairly long section, although at high input power, it shows some weak oscillation which correlates with the oscillation on the output intensity pulse.

We have also made measurements at different cell lengths. The results are similar to what we have already described, indicating that after the initial period of deformation through self-focusing, the deformed horn-shapedpulse does propagate on with little further change in shape. The self-focused beam showed practically no spectral broadening since the phase modulation rate here is expected to be small.

Finally, we should briefly comment on the recent work of Rao and Jayaraman.[13] on self-focusing in isotropic MBBA. In analyzing their data,

they have neglected the facts that self-focusing is transient and that the scattering loss is non-negligible in their 30 cm. cell, especially at temperatures close to the transition temperature. It is also difficult to understand how they could obtain a self-focusing threshold P_{th} (defined as the input power at which the self-focused beam at the end of the cell reaches a limiting diameter) which decreases as $(T - T_c)^{0.16}$ with temperature. Our results show that P_{th} increases as $(T - T_c)$ descreases. This is what one should expect because the scattering loss increases as[14] $\exp[A/(T - T_c)]$.

In conclusion, we have presented the results of the first quantitative experimental study on transient self-focusing. They agree qualitatively, and also quantitatively when comparison can be made, with the earlier theoretical predictions.[5,6] The limiting diameter in the present case is neither due to stimulated Raman and Brillouin scattering nor due to avalanche breakdown.

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FIGURE CAPTIONS

Fig. 1 Schematic drawing showing how an input pulse gets deformed through self-focusing into a horn-shaped pulse which then propagates on with little change in shape.

Fig. 2 Radius of the self-focused beam at the end of a 9.5 cm MBBA cell at 50°C as a function of the incoming peak power. The reduced radius is normalized against the radius of the incoming beam, and P_{cr} = 0.12 kW. The solid curve is a theoretical curve extrapolated from the calculations in Ref. 6. The arrows B and F indicate where stimulated Brillouin and Raman scattering set in.

Fig. 3 On-axis intensity variation with time at the end of a 9.5 cm MBBA cell at 50°C with different input peak powers: (a) the input pulse; (b) P = 1.6 kW; (c) P = 3 kW; (d) P = 3.2 kW; (e) P = 3.5 kW. I_{max} is in MW/cm^2. Note that the baseline is shifted for each pulse.

Fig. 4 Temporal variation of the self-focused beam radius at the end of the cell. (a)-(d) correspond respectively to the cases (b)-(e) in Fig. 3. Note the shift of the baseline for different cases.

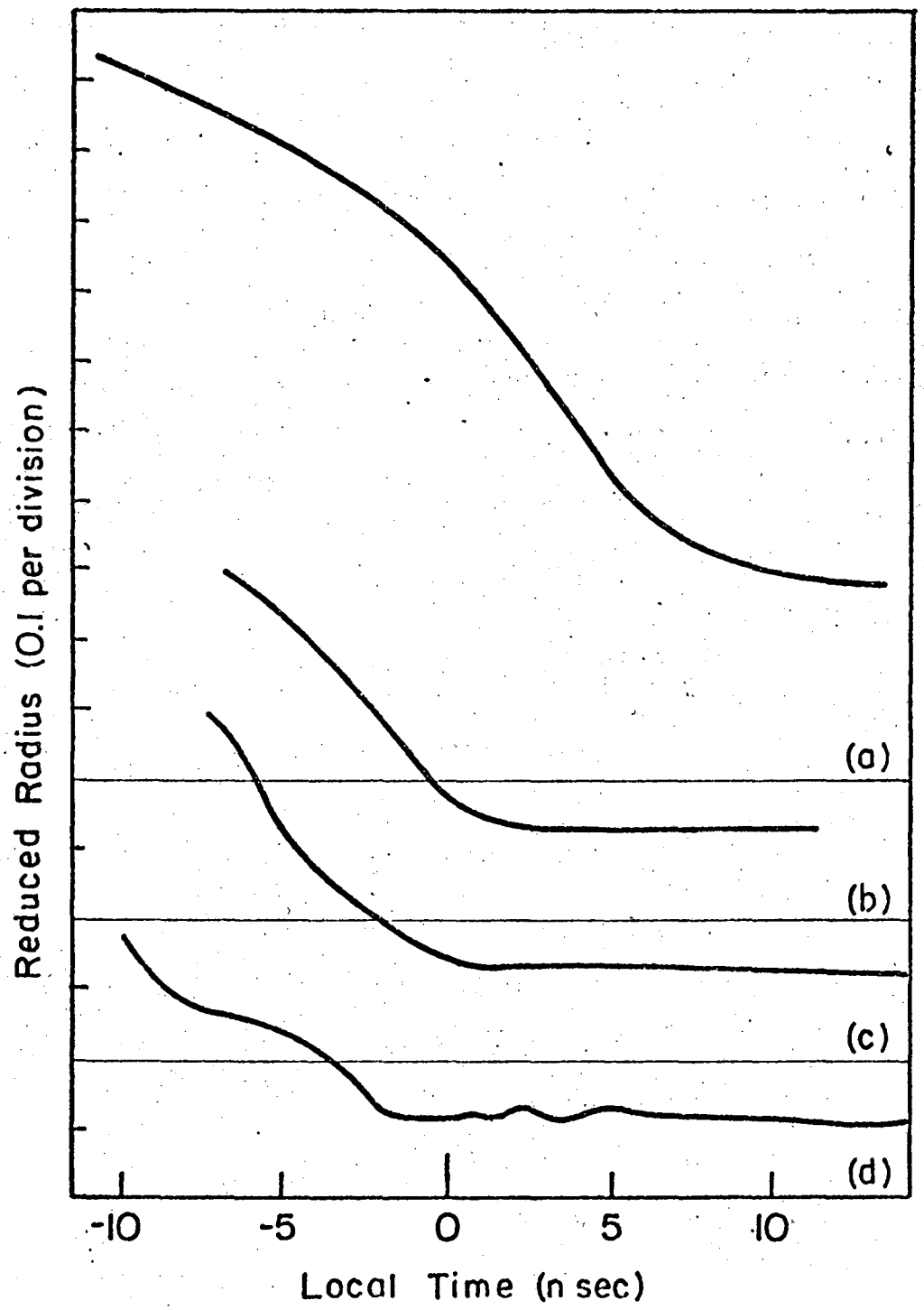

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Fig. 1

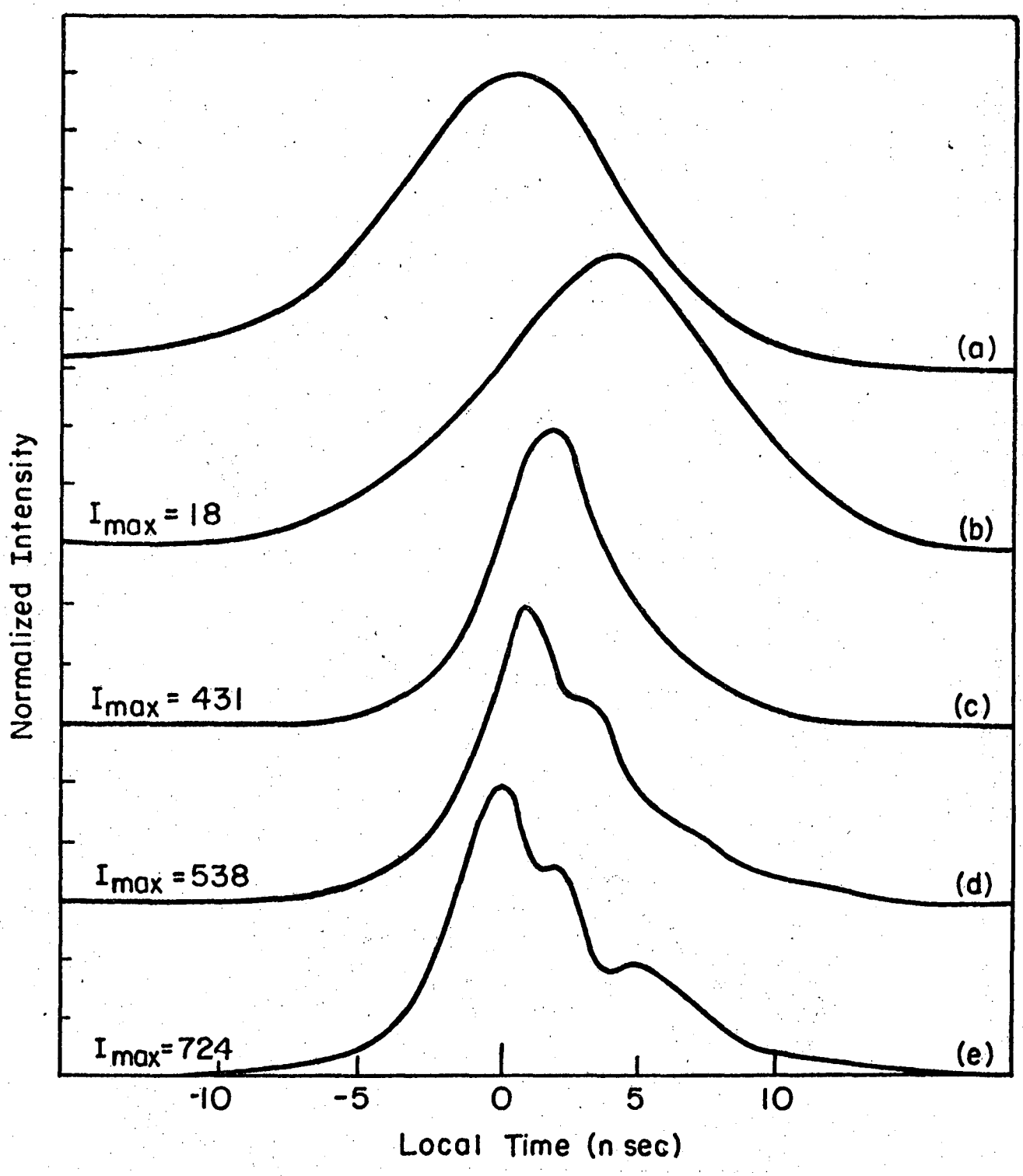

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Fig. 2

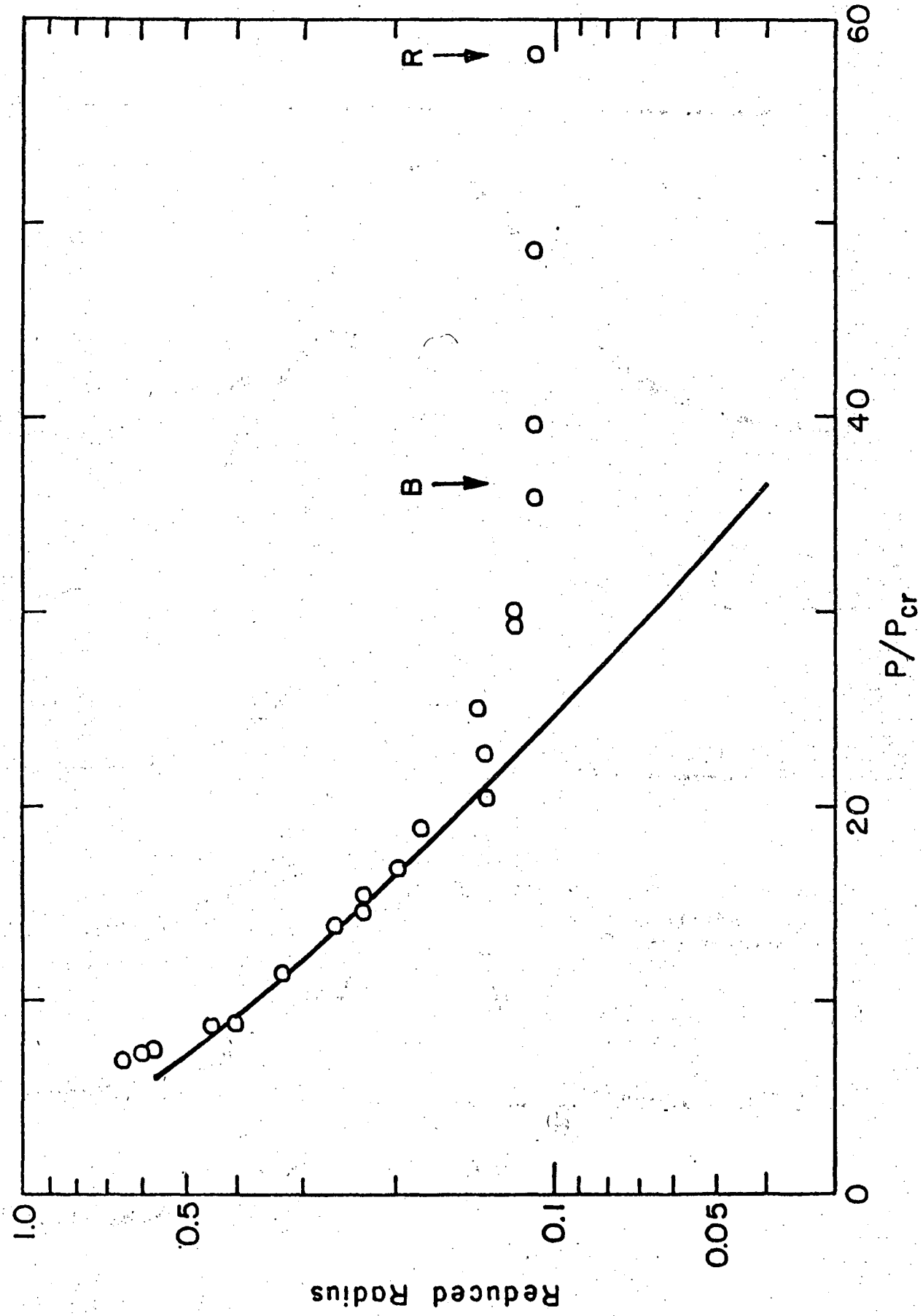

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Fig. 3

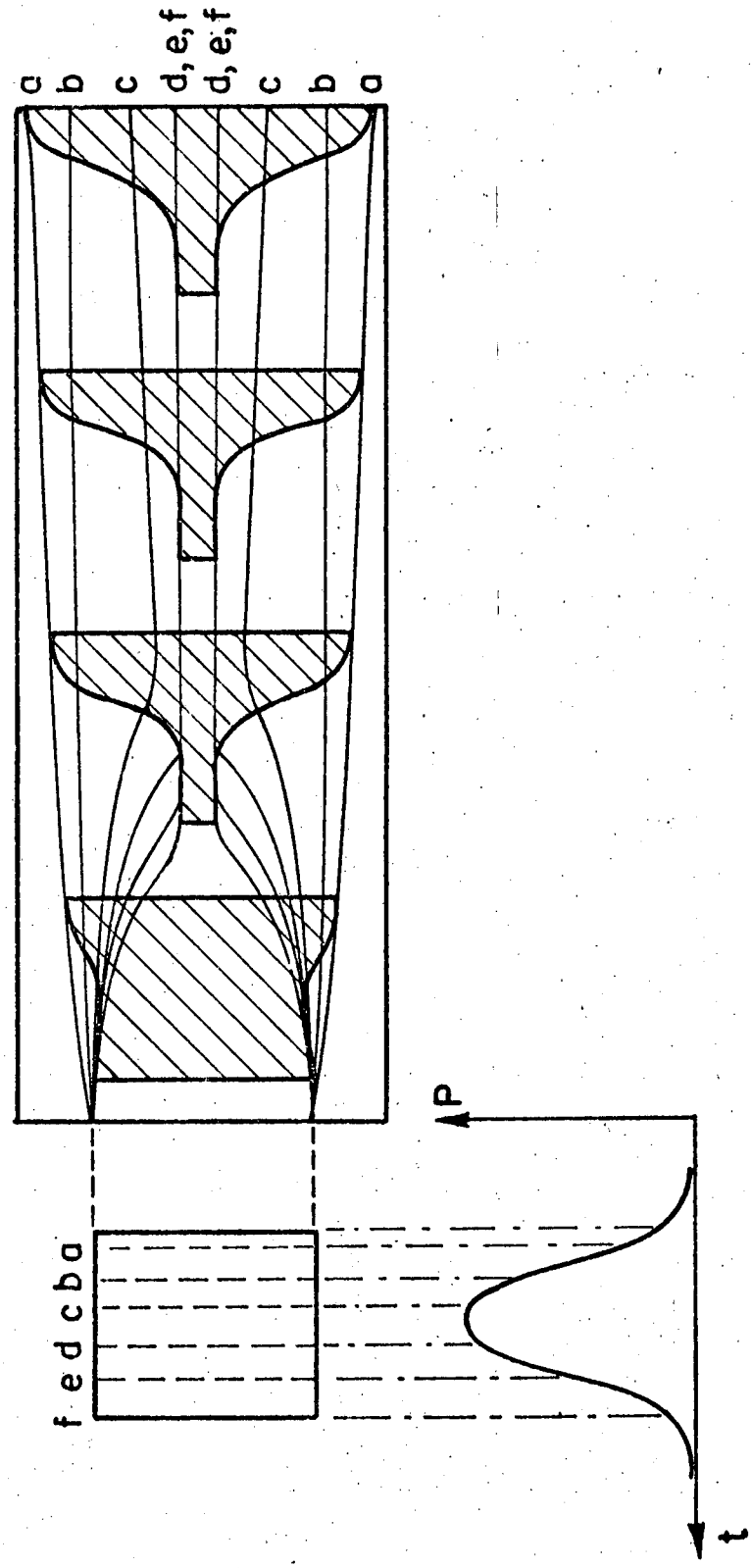

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Fig. 4

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