United Aircraft Research Laboratories

November 8, 1968

BMDR
Room 2B, 263
The Pentagon
Washington, D.C. 20301

Attention: Major Glenn Sherwood


References: (a) ONR letter 42l:CES:jdz dated 2 August 1967

Enclosures: (A) Eight (8) copies of UAC Research Laboratories Report G-920479-9

Gentlemen:

In accordance with SECTION E of the subject contract SCHEDULE and with the instructions presented in the Reference (a) letter, we are transmitting herewith eight (8) copies of the subject report, Enclosure (A), for the period from August 1, 1968, through October 31, 1968.

Very truly yours,

UNITED AIRCRAFT CORPORATION
Research Laboratories

Harlan D. Taylor
Manager of Physics Laboratories

cc: Head, Physics Branch
Physical Sciences Division
Office of Naval Research
Washington, D.C. 20360
Attention: Mr. Frank E. Isakson (3 copies)
Gentlemen:

1. The Research Laboratories of the United Aircraft Corporation are conducting under the subject contract an experimental and theoretical investigation using ultrashort laser pulses to measure the lifetimes of excited states in the time regime $10^{-8}$ to $10^{-12}$ sec and to determine the transient response of quantum systems in solids, liquids, and gases.

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"This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research, and the Department of Defense."
2. Introduction
   (a) Background
   Investigations conducted at United Aircraft Research Laboratories have successfully demonstrated that saturable absorbers with fast relaxation time can be used to simultaneously Q-switch and mode-lock lasers and, therefore, obtain optical pulses having peak powers in excess of $10^9$ watts and time durations of the order of $10^{-12}$ sec. The applications of such pulses to research on optically generated plasmas, optical radar, optical computers, high speed photography and holography, optical and vibrational spectroscopy of liquids, solids, and gases, nonlinear optical properties of materials, transient response of atomic and molecular systems, velocity of light measurements, etc., appear very promising.
   (b) Objectives
   The objectives of this program are to conduct experimental and theoretical investigations using ultrashort laser pulses to measure the lifetimes of excited states in the time regime of $10^{-8}$ to $10^{-12}$ sec and to determine the transient response of quantum systems in solids, liquids, and gases.
   (c) Assignment of Responsibility
   Dr. Anthony J. DeMaria has overall technical responsibility for this project. Drs. W.H. Glenn, Jr., G.L. Lamb, Jr., E.B. Treacy, and M. E. Mack are directly responsible for the theoretical and experimental progress.

3. The following investigations were performed during this period:

   (a) MODE-LOCKING OF ORGANIC DYE LASERS

   A large number of organic dye solutions exhibit laser action when excited with a short duration, high intensity pump pulse.\(^1\) Pump pulses from Q-switched lasers and from specially constructed flashlamps have been employed. The output spectra from such dye lasers are quite broad, extending in some cases over a range of a few hundred angstroms. This suggests the possibility of mode-locking an organic dye laser to produce very short (picosecond or shorter) pulses over a wide range of the visible spectrum. The availability of ultrashort pulses with any desired wavelength would greatly increase their applicability to studies of nonlinear and transient optical effects, spectroscopy and lifetime measurements.

   It has been demonstrated and reported previously under the present contract that an organic dye laser can be mode-locked by pumping it with another mode-locked laser.\(^2\) Because the fluorescent decay time of the dye is rapid, the pumping by a series of short pulses produces a modulation of the gain of the dye laser. If the frequency of this modulation corresponds to the mode spacing of the dye laser then mode-locked operation should result. Experimentally, this requirement is simply that the optical cavity lengths of the dye and pumping laser be equal. Mode-locked operation can also be achieved with the dye laser cavity equal to a submultiple of the pumping laser since in this case the modulation waveform will have harmonic components at the mode spacing of the dye laser.
Such a dye laser has been constructed using the 5300 Å second harmonic of a mode-locked Nd-glass laser as the pump and a solution of Rhodamine 6G in alcohol as the dye laser medium. Mode-locked operation was readily achieved and typical spectral and temporal outputs have been reported previously.\(^{(2)}\)

It has been reported that the very broad spectral output of a dye laser pumped with a Q-switched laser or a flashlamp can be very efficiently narrowed through the use of a selective reflector in the cavity.\(^{(3)}\) If a diffraction grating is used in a Littrow arrangement as one of the reflectors, spectral narrowing occurs and the output of the laser can be tuned over the original broad spectrum.

It was of interest to see if this tuning could be achieved when the dye laser was mode-locked. The spectral narrowing in such a case would limit the ultimate shortness of a pulse that could be obtained. A bandwidth of \(1Å\) however at 5000Å is sufficient to produce a pulse of duration less than \(10^{-11}\) sec. A dye laser was set up as shown in Fig. 1. A \(10^{-4}\)M solution of Rhodamine 6G was used. The grating was ruled with 600 lines/mm. and was blazed at 5000Å. The rulings of the grating were perpendicular to the plane of the figure. The rotation about the axis lying in the plane of the figure was adjusted using a helium neon laser and the alignment about the other axis was achieved by shining a white light on the grating and rotating it so that the desired color, in this case the center of the yellow region, returned upon the incident beam. The output of the laser was taken off the second order from the grating.

The laser was pumped with the second harmonic from a mode-locked Nd:glass laser generated in a 1.2 cm length of KDP. It was necessary to decrease the beam diameter to increase the second harmonic power generated. A Galilean telescope of approximately 2.5 power was used for this purpose. The conversion efficiency in the KDP was quite small due to the fact that the beam divergence of the pumping laser, approximately 3 mrad, was considerably greater than the acceptance angle of the KDP as determined by the phase matching requirement. The telescope reduces the beam diameter but increases the beam divergence by the same amount. The phase matching angle however is extremely critical only in one direction, however, so a net gain can be realized. The dye laser cavity was adjusted to one-half of the length of the pumping laser, primarily for experimental convenience.

Typical spectral outputs that were obtained from the laser as shown in Figure 2. A mercury spectrum is shown for reference. The grating was rotated by approximately 1.3 mrad between each of the firings shown. The output was found to be tunable over slightly more than 300Å from the green through orange.
The appearance of the output pulse train was generally similar to those that have been reported previously. The output was well mode-locked and returned to zero between pulses. The pulses were detector limited in width. The envelopes of the pulse trains were somewhat more irregular than those produced when a mirror used in place of the diffraction grating.

It would be desirable to determine the actual pulse duration of the pulses produced by the dye laser. The two photon absorption-fluorescence technique can be used for this purpose provided that a sufficient amount of power can be obtained from the dye laser. At the present time, however, the power level from the laser has been insufficient to produce an observable fluorescence in chloronapthalene, a liquid which has a fairly strong two-photon absorption at 5300 Å.

Sorokin has analyzed the problem of the conversion efficiency of a dye laser pumped by a short duration pulse from a flashlamp or Q-switched laser. He finds that the efficiency increases as the duration of the pumping light pulse decreases. The efficiency of conversion can exceed 80% in some cases for the pulse duration of the order of $10^{-8}$ sec. The conversion efficiency of the mode-locked dye laser was not measured, but was certainly orders of magnitude lower than the predicted value for a short, Q-switched pump pulse. It is felt that the reason for this is the fact that when the laser is running in the mode-locked region, a light pulse is only present in the dye cell for a very small fraction of the time. The fluorescent decay time is comparable to the transit time of the cavity. As a consequence, unless the dye laser pulse complete depletes the overpopulation as it passes through the cell, the energy stored in the overpopulation will be lost in non useful fluorescence by the time the pulse returns to the cell. This will lead to very poor conversion efficiency. An analytical model to describe this effect and to calculate the efficiency to be expected is being set up. The model and results of the calculation will be described in a subsequent report.

In order to increase the conversion efficiency then it is necessary to have a longer fluorescence lifetime or to increase the power level of the pulses so that they are capable of depleting the overpopulation on a single pass. The former possibility is not promising since most dyes of interest have fluorescent decay times of the order of $10^{-8}$ sec. The latter requires more pumping power in order to operate the dye laser far above threshold. To do this, it would be desirable to eliminate the inefficient second harmonic conversion and pump the laser directly with the fundamental beam from a mode-locked laser. Experiments have been initiated using a mode-locked ruby laser and mode locked operation of a 3,3'-diethylthiatricarbocyanine iodide (DITC iodide) laser has been achieved. Two photon absorption has been detected in Rhodamine 6G and it is expected that with some minor modifications, measurements of the pulse widths can be made.
(b) OPTICAL PULSE PROPAGATION IN AN INHOMOGENEOUSLY BROADENED MEDIUM

A method for treating the propagation of ultrashort optical pulses in an inhomogeneously broadened medium of two level systems is summarized. The method is used to determine the area under the envelope of the first photon echo which may develop behind two pulses passing through such a medium.

The interaction of an ultrashort pulse of coherent light with a system of inhomogeneously broadened two level systems may be described by the equations

\[ \frac{\partial \xi}{\partial \tau} = \pm \int_{-\infty}^{\infty} dq(t) P(t, \xi) \]

\[ \frac{d}{d\tau} \begin{pmatrix} P \\ Q \\ H \end{pmatrix} = \begin{pmatrix} \xi H + fQ \\ -fP \\ -\xi P \end{pmatrix} \]

where \( Q(t, \xi, \tau) \) is the amplitude of the polarization that is in phase with the electric field of carrier frequency \( \omega_0, f = (\omega - \omega_0)/\Omega \) is a dimensionless frequency variable determining the extent to which an individual two level system is off resonance and \( g(t) \) is the spectrum of such inhomogeneous broadening (assumed symmetric about \( f = 0 \)). The plus (minus) sign in Eq. (1) is used depending upon whether the medium is initially in the upper (lower) level. The other terms are as defined previously.

As noted previously (2) Eqs. (2) have the same structure as the Serret-Frenet equations (5) and one may introduce two functions \( \phi \) and \( \psi (z = i/\phi^*) \) defined by

\[ \phi = i(1 + \phi\psi)/(\phi - \psi), \quad \psi = (1 - \phi\psi)/(\phi - \psi) \]

which are each found to satisfy a Riccati equation. Setting \( \phi = \exp(i\mu) \) the equation for \( \phi \) becomes

\[ \dot{\phi} = i \mu \sin \mu \xi. \]

Choosing \( \xi(\xi, \tau) = \theta(\xi) \delta(\tau) \) the state of a system immediately after passage of the pulse is found to be related to that before the pulse by the equation

\[ P(t^+) = R_Q(\theta)P(t^-) \]

where \( P(t) \) is the column vector \((\phi, Q, H)\) of Eq. (2) and \( R_Q(\theta) \) is the 3 x 3 matrix representing a rotation about the \( Q \) axis through an angle \( \theta \). At time \( \tau \) after the pulse the system is described by a rotation about the \( H \) axis through an angle \( f \tau \) i.e.

\[ P(t) = R_H(f\tau)R_Q(\theta)P(t^-). \]

When the \( P \) component is calculated and substituted into Eq. (1), one finds

\[ \frac{d\theta}{d\xi} = \pm \sin \theta \]
which is the area theorem. (6)

The apparent inconsistency of using a delta function in the slowly varying envelope $\xi$ does no violence to the theory. It merely provides a convenient device for obtaining solutions to Eqs. (1) and (2) in the short pulse limit.

This technique may be used repeatedly to determine the state of the system when a series of pulses interacts with it. In particular, if a pulse having area $\theta_1$ interacts with a system in its ground state at $T=0$ followed by a pulse of area $\theta_2$ at $T=T$, one finds that an echo pulse of area, say, $\theta_3$, may develop at $T=2T$. Carrying out the appropriate matrix multiplications and using Eq. (1) one finds that the area functions satisfy

$$
\frac{d}{d\xi} \begin{pmatrix} \theta_1 \\ \theta_2 \\ \theta_3 \end{pmatrix} = \begin{pmatrix} -\sin \theta_1 \\ -\cos \theta_1 \sin \theta_2 \\ \sin \theta_1 \sin^2 \frac{\theta_1}{2} \cos \theta_3 - \cos \theta_1 \cos \theta_2 \sin \theta_3 \end{pmatrix}.
$$

The first two of Eqs. (5) have solutions $\tan \frac{\theta_1}{2} = e^{-\xi+a}$, $\tan \frac{\theta_2}{2} = \beta \sech (\xi-a) = \beta \sin \theta_1$, where $\theta_2 = \tan \frac{\theta_2(0)}{2}$ and $\beta = \tan \frac{\theta_2(0)}{2} \csc \theta_1(0)$. If $\theta_1(0) = \pi/2$, $\theta_2(0) \approx \pi$, then $|\beta| >> 1$ and from the solution for $\theta_2$ one sees that $\theta_2$ remains $\approx \pi$ until $\theta_2 \sim 1/\beta$.

Until this final stage in the pulse evolution one may set $\theta_2 \approx \pi$ in the last of Eqs. (5) which may then be transformed to

$$
\frac{d\theta_3}{d\theta_1} + \cos \theta_3 + \cot \theta_1 \sin \theta_3 = 0.
$$

The solution which satisfies $\theta_3 = 0$ for $\theta_1 = \pi/2$ (i.e. $\xi = 0$) is found to be (7,8)

$$
\tan \frac{\theta_3}{2} = -k' \frac{d}{dk'} [K(k') + K(k')] / [K(k) + K(k')]
$$

where $K(k)$ is the complete elliptic integral of modulus $k = \sin(\theta/2)$ and $k' = \cos(\theta/2)$ is the complementary modulus. The spatial evolution of the various area functions is shown in Figure 3.
Since the last report period a number of experiments have been performed on "light amplification in saturable absorbers." These experiments have shown that the experimental observations are the result not of a single effect but of two separate effects, one being that discussed previously under the above title. The new effect is a kind of thermal light scattering much like "stimulated thermal Rayleigh scattering".\(^9\) The relative importance of these two effects in saturable absorbers has not yet been established. However, from theoretical considerations the thermal effect would be expected to dominate at the high intensities used for these experiments, while at lower intensities the saturation effects should dominate. Experimental evidence described below supports this prediction. The most important difference between the two is that the thermal scattering is the result of a non-linear refractive index change while the saturation effect is, of course, the result of a non-linearity in the absorbivity. The thermal scattering can be observed in linear absorbers as well as saturable absorbers.

The experimental apparatus used to study the new effect is the same as used in the saturable absorber experiments. This is shown in Fig. 4. As in the case of saturable absorbers both electronic and photographic detection were used. Figure 5 shows photographic data taken for a variety of absorbing solutions. In all cases the absorption is linear. The incident probing beam appears to the left of the central spot in the photographs shown. It should be noted that in non-absorbing liquids the probing beam is not sufficiently intense to expose the film so that only central spot appears. Of all the liquids examined to date quinoline exhibits the largest effect. Up to nine generated beams have been observed with this liquid, which, incidentally, requires no additional absorbers since, it, itself, is absorbing. Only water shows little or no effect, a result, which prevented its discovery in earlier experiments. This is true regardless of the absorber used. Most liquids give an effect intermediate between these two extremes. As with water the magnitude of the effect appears characteristic of the liquid rather than the absorber.

The patterns illustrated in Fig. 5 are highly reminiscent of diffraction by a transmission grating. This suggests that by some means the two beams set up a grating such that the 1st order scattering of the strong beam is in the direction of the weak incident beam and in the direction of the 1st generated beam. The increased intensity in these directions would further increase the strength of the grating which would in turn further increase the scattering. That is to say, one has a gain effect. The higher order beams represent a higher order scattering. Either a phase grating such as occurs in "light by light scattering"\(^{(10)}\) and in stimulated thermal Rayleigh scattering \(^{(9)}\) or an amplitude grating as occurs in light amplification by saturable absorption \(^{(11,12)}\) could be developed.
In the present instance the latter possibility can be ruled out by the experiment shown in Fig. (6a) Here, four beams, two at the fundamental frequency and two at the second harmonic are incident on the solution, in this instance cryptocyanine in methanol. For the case of an amplitude grating, the fundamental beams, should have no effect on the weak second harmonic beams since they are outside any absorption band for the dye. In the case of a grating in the refractive index the second harmonic beams would still be scattered. Figures 6b and 6c show the experimental results. The results demonstrate that at these intensities even in saturable absorbers phase grating effects predominate over amplitude effects. As noted earlier the reverse should be true at lower intensities. It is worth noting that the angular spacing of the orders in fig. 6c is half that in fig. 6b.

This is just the result expected from the difference in frequency.

There are a number of mechanisms which could give rise to a refraction grating. The fact that the pure liquid shows no gain and that the gain in the solution is characteristic of the solvent indicates that the absorbing dye transfers an excitation to the liquid which in turn affects its refractive index. Eventually this excitation will take the form of a density wave as in stimulated thermal Rayleigh scattering. However, it would be impossible for such wave to form within the 2 picosecond pulse duration. One might envision the gradual buildup of a density wave during the pulse train, however, an electronic determination of the gain indicates that in linear absorbers the gain follows the peak power of the pulse regardless of its position in the train. One is lead to conclude that the transferred internal excitation in the solvent is, itself, responsible for the change in refractive index. Roughly speaking this may be regarded as a change in the internal temperature of the solvent molecules. Thus, the refractive index is of the form

\[ n = n_0 + \left( \frac{\partial n}{\partial T} \right)_p T. \]  

(1)

Because of the short pulse duration the field equations given by Herman and Gray simplify to

\[ \rho_0 c \nabla \frac{\partial T}{\partial t} = \frac{1}{4\pi} n c \alpha \vec{E}^2 \]  

(2A)

\[ - \nabla^2 \vec{E} + \frac{n_0^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = - \frac{2n_0}{c^2} \left( \frac{\partial n}{\partial T} \right)_p \frac{\partial^2}{\partial t^2} (TE). \]  

(2b)
These equations presuppose an instantaneous heat transfer from the absorbing molecules to the liquid. For a pulse lacking phase coherence these equations can be solved following the same procedure used by Herman and Gray. Because of the coupling, one finds a non-exponential growth for the off-axis waves. If these couplings are ignored, a power gain coefficient of

\[ G \approx \frac{n_0 a \omega \lambda^2}{4 \pi \rho C_v} \left( \frac{\partial n}{\partial T} \right)_\rho \frac{\omega_w - \omega_l}{\left( \omega_w - \omega_l \right)^2 + \left( \frac{1}{2} \Gamma_L \right)^2} \]  

is found for the weak wave. This result indicates a Stokes shift in the amplified wave of half the laser linewidth. Following the arguments of Herman and Gray a Stokes shift would be expected, since \( (\partial n/\partial T)_\rho > 0 \).

The gains calculated from (3) are found to be much smaller than measured experimentally (with \( \alpha \approx 0.2, \ G \approx 3 \)), Moreover, within experimental error (\( \sim \frac{1}{5} \Gamma_L \)) there is no frequency shift in either the amplified incident wave or in the 1st generated wave. The latter discrepancy can be attributed in part at least to the neglect of the coupling. However, it must be noted that the expression given above in (3) is not valid for a pulse which is phase coherent as one from a mode locked laser would be. Formally, the convolution integral indicated by Herman and Gray becomes an integral over both the frequencies in the strong pulse and also those in the amplified weak pulse so that the equation is an integral equation for the scattered field. The analysis of this problem is not yet complete. However, the fact that a coherent pulse would yield a greater gain than an incoherent incident pulse can be understood from the grating picture. The incoherent pulse leads to a grating which is "smeared out" spatially except over the coherence length. On the other hand, the coherent pulse gives a well defined grating structure over its entire length. A qualitative indication of the correctness of the theory can be obtained by comparing the results shown in Fig. 2 with \( \frac{1}{\rho_0 C_v} (\partial n/\partial T)_\rho \) which is tabulated in Table I.

The origin of the thermal scattering suggests other experiments which may be of interest. Two fundamental beams can be used to create a grating in the liquid and as before two second harmonic beams used to probe it. If the harmonic beams are advanced in time ahead of the fundamental beams, the harmonic beams can be used to investigate the transfer of excitation from the absorber to the solvent. Similarly, by delaying the harmonic beams with respect to the fundamental beams the decay of this excitation into translational motion can be studied.

Additional theoretical and experimental work in this area is in progress.
(d) OTHER PROGRAMS

During the reporting period work has continued on the adiabatic rapid passage experiment that has been described previously. Results of this work will be reported in a subsequent report. In the previous report (2) a technique for the detection and compensation of a frequency sweep in the pulses produced by a mode-locked Nd: glass laser was described. This achievement was the result of another program on the study of ultrashort laser pulses being conducted at the United Aircraft Research Laboratories.* It was found possible to compress the output pulses to approximately 0.4 picoseconds, close to the theoretical limit as determined by the laser bandwidth. This extremely short pulse can be used as a scanning pulse in a two photon absorption-fluorescence system to observe the pulse shape of other short pulses. This technique should be extremely useful for the study of the propagation characteristics of ultrashort pulses, and experiments to study these characteristics are being planned.

4. It is anticipated that during the next reporting period experimental and theoretical work will continue on the propagation characteristics of ultrashort pulses, the mode-locking of organic dye laser and the stimulated scattering processes in absorbing liquids in addition to the programs discussed in (d) above.

* The work was supported by the Air Force Cambridge Research Laboratories Office of Aerospace Research under Contract F19628670075.

Very truly yours,

UNITED AIRCRAFT CORPORATION
Research Laboratories

Anthony J. DeMaria
Senior Principal Scientist
Quantum Physics Laboratory
REFERENCES


7. E. Kamke, Differentialgleichungen Losungsmethoden und Losungen, Band I (Chelsea, New York, 1959), Nos. 1.79 and 2.68.


| Liquid       | $\rho_0$  | $n_0^{(0)}$ | $(\frac{dn}{dT}|\rho_0)_{0K} \times 10^5$ | $C_p \times 10^{-7}$ | $\gamma$ | $\frac{n_0 L}{(\rho_0 C_p)} \times \frac{dn}{dT}|\rho_0 \frac{a}{d\rho}^2$ |
|--------------|-----------|-------------|---------------------------------|-----------------|------|--------------------------------------|
| Quinoline    | 1.10      | 1.62        | 16                              | 1.47            | --   | >16.0                                 |
| Benzene      | 0.88      | 1.50        | 9                               | 1.7             | 1.40 | 12.5                                  |
| Carbon       | 1.60      | 1.46        | 7                               | 0.83            | 1.46 | 11.2                                  |
| Tetrachloride|           |             |                                 |                 |      |                                       |
| Acetone      | 0.79      | 1.36        | 7                               | 2.1             | 1.40 | 8.0                                   |
| Ethyl Ether  | 0.71      | 1.35        | 4                               | 2.3             | 1.39 | 4.6                                   |
| Methanol     | 0.79      | 1.33        | 4                               | 2.5             | 1.21 | 3.3                                   |
| Water        | 1.00      | 1.33        | 0.5                             | 4.2             | 1.01 | 0.2                                   |
TUNABLE MODE-LOCKED ORGANIC DYE LASER

FROM MODE-LOCKED Nd:GLASS LASER

DIFFRACTION GRATING

FIRST ORDER

OUTPUT IN 2ND ORDER

DETECTOR

DYE CELL

95% R ABOVE 5500 Å
20% R BELOW 5400 Å

FIG. 1
TUNABLE MODE-LOCKED DYE LASER SPECTRA

GRATING ROTATION INCREMENT 1.33 mrad
$\theta_1(0) = \pi/2$

$\theta_2(0) = \pi - 0.10$
FIG. 4

EXPERIMENTAL ARRANGEMENT FOR OBSERVING GAIN IN SATURABLE ABSORBERS

MIRROR

GROUND GLASS

GROUND GLASS

FAST PHOTODIODE

DYE CELL

(1cm PATH LENGTH)

WEDGE

WEDGE

GLASS BEAMSPLITTER

NODE-LOCKED RUBY LASER BEAM

O D C Q

O D C >

O D C =

O D C <

O D C X
THERMAL LIGHT SCATTERING FROM VARIOUS LIQUIDS

QUINOLINE (QUINOLINE)

CARBON TETRACHLORIDE (IODINE)

ACETONE (IODINE)

METHANOL (COPPER SULFATE)

WATER (COPPER SULFATE)
FIG. 6

PHASE GRATING EXPERIMENT

FRESNEL BIPRISM

CRYTOCYNINE IN METHANOL

CS-2-58 FILTER

RUBY LASER BEAM

ADP ORIENTED FOR PHASE MATCHING

FUNDAMENTAL

HARMONIC
This report covers work under Contract N00014-66-C0344 for the period 1 August to 31 October 1968. Topics discussed include analytical results on the propagation of ultrashort optical pulses, experimental results on tunable mode-locked organic dye laser and investigations of stimulated scattering process in absorbing liquids.
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