SELF-TRAPPING OF OPTICAL BEAMS*

R. Y. Chiao, E. Garmire, and C. H. Townes Massachusetts Institute of Technology, Cambridge, Massachusetts (Received 1 September 1964)

We shall discuss here conditions under which an electromagnetic beam can produce its own dielectric waveguide and propagate without spreading. This may occur in materials whose dielectric constant increases with field intensity, but which are quite homogeneous in the absence of the electromagnetic wave. Such self-trapping in dielectric waveguide modes appears to be possible in intense laser beams, and to produce marked optical and physical effects.

A crude description of the phenomenon suggested here can be obtained by considering the diffraction of a circular optical beam of uniform intensity in material for which the index of refraction may be expanded in terms of field strength as

$$n = n_0 + n_2 E^2 + \cdots$$
 (1)

If the beam diameter is D, the beam might be expected to expand by diffraction with an angular divergence of $\theta \approx 1.22\lambda/n_0 D$. But if the term $n_2 E^2$ produces a dielectric constant within the beam which is so high that the critical angle for total internal reflection at the beam's boundary is greater than θ , then spreading by diffraction will not occur. For $\theta \ll 1$, this requires

$$P = \frac{\pi D^2}{4} \frac{n_0 \dot{E}^2 c}{8\pi} \ge (1.22\lambda)^2 \frac{c}{64n_2}, \qquad (2)$$

where P is the total beam power. This simple approximation indicates that a beam above a

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certain critical power level may be trapped at any arbitrary diameter and not spread, and that this power level decreases with λ^2 . For normal dielectric materials, the constant n_2 is such that the critical power for trapping is within one or two orders of magnitude of 10⁶ watts for visible light, a power level commonly obtained in laser beams. For radio waves, the longer wavelength makes the critical power for such materials unattainable at present.

The nonlinear coefficient n_2 may be associated with high-frequency Kerr effects involving molecular orientation, with electrostriction, or with nonlinearities due to electronic polarizability of the type which generates third-harmonic waves in optical materials. For liquids, the first two effects are of comparable size and the third much smaller, as indicated in Table I. For solids, in which molecular rotation is frozen out, electrostrictive effects dominate. Each effect has its own inertial and resistive characteristics, but we shall consider primarily a steadystate condition in which these are unimportant, a valid approximation for many interesting cases.

It is instructive to note also that, on the basis of geometric optics, the nonlinear dielectric response implies that light cannot be focused to a point, but is rather focused into a line. Consider a plane wave passing through a lens which would converge the light to a point. A ray of the beam which initially makes an angle θ_0 with the

Material	$n_2 \times 10^{13}$ (Kerr effect) ^a		$n_{0} \times 10^{13}$	P (electrostriction)
	A	В	$(electrostriction)^{b}$	(MW)
Carbon disulfide	180	18	18	0.2
Benzene	49		13	0.25
Water		0.13	2	1
Air (1 atm)			0.041	80
(100 atm)			4.1	0.8
Glass (heavy silicate flint)			0.9	4
Calcite			0.8	4
Sapphire			0.2	20

Table I. Coefficients for nonlinear indexes of refraction n_2 and the critical power levels P for self-trapping. Index of refraction $= n_0 + n_2 E^2$, where E is in esu.

^aJ determined from: (A) G. Mayer and F. Gires, Compt. Rend, <u>258</u>, 2039 (1964); (B) P. D. Maker, R. W. Terhune, and C. M. Savage, Phys. Rev. Letters 12, 507 (1964).

 $^{
m b}\gamma$ and B determined from International Critical Tables (McGraw-Hill Book Company, Inc., New York, 1929).

axis can be shown, assuming $n = n_0 + n_2 E^2$, not to follow a straight line but the hyperbola

$$r - (8n_2 P/n_0^2 c r) = -\theta_0 z,$$

where P is the power, r is the ray's distance from the axis, and z is measured along the lens axis from the normal focal point z = 0. This expression makes the approximation that the light path is not affected by terms higher than first order in P. Thus the focused beam never reaches a focus, but approaches the axis asymptotically. However, by the time the beam's diameter has become as small as a few wavelengths, the approximation of geometric optics is no longer valid, and wave properties must be allowed for.

The electromagnetic wave equation is modified in the presence of nonlinearities to become

$$\nabla^2 \vec{\mathbf{E}} - \frac{\epsilon_0}{c^2} \frac{\partial^2 \vec{\mathbf{E}}}{\partial t^2} - \frac{\epsilon_2}{c^2} \frac{\partial^2}{\partial t^2} (E^2 \vec{\mathbf{E}}) = 0, \qquad (3)$$

where $\epsilon = \epsilon_0 + \epsilon_2 E^2$. Consider now solutions of the form $\vec{E} = \vec{E}_t(x, y) \cos(k_z z - \omega t)$, which we will call steady-state solutions, since there is no variation of the field at any point other than the regular oscillation of optical frequency ω . We shall also assume now that the nonlinear dielectric response is much slower than the optical oscillation, which can be true for electrostrictive effects and for some types of Kerr effects, but not for electronic polarization which produces optical harmonics. For such conditions, one may average E^2 over time, and (3) becomes

$$\frac{\partial^2}{\partial x^2} \vec{\mathbf{E}}_t + \frac{\partial^2}{\partial y^2} \vec{\mathbf{E}}_t - \Gamma^2 \vec{\mathbf{E}}_t + \frac{\epsilon_2}{2} k_0^2 E_t^2 \vec{\mathbf{E}}_t = 0, \qquad (4)$$

where $\Gamma^2 = k_z^2 - k^2$ and $k = n_0 k_0 = n_0 \omega/c$. In the case where E_t depends only on y, and under the assumption of linear polarization,

$$\frac{d^2}{dy^2}E_t(y) - \Gamma^2 E_t(y) + \frac{\epsilon_2}{2}k_0^2 E_t^3(y) = 0.$$
 (5)

If E_t represents a slab-shaped beam, confined in the y direction, the boundary conditions are $E(y) \rightarrow 0$ as $y \rightarrow \infty$ and dE/dy = 0 at y = 0. This excludes periodic solutions, so that $\Gamma^2 > 0$. A mechanical analog of (5) is a particle in a doublewell quartic potential-energy function. It is immediate from consideration of this analog that there is a unique solution which is not oscillatory, namely $E_t(y) = E_t(0)/\cosh\Gamma y$, where Γ must equal $\frac{1}{2}\epsilon_2^{1/2}k_0E_t(0)$. Note that, given a certain size of the beam $(\sim 1/\Gamma)$, the field inside the beam must attain a value $E_t(0)$ for trapping.

A more interesting, though somewhat more complex, case is a cylindrical beam. For this case, and with the assumption of circular polarization, (3) becomes

$$\frac{d^2 E^{*}(r^{*})}{dr^{*2}} + \frac{1}{r^{*}} \frac{dE^{*}(r^{*})}{dr^{*}} - E^{*}(r^{*}) + E^{*3}(r^{*}) = 0, \quad (6)$$

if $r^* = \Gamma r$, $E^*(r^*) = b^{1/2} E_f(r^*)$, and $b = \frac{1}{2} \epsilon_2 (k_0 / \Gamma)^2$. This shows that the steady-state solutions can be scaled via the factor \sqrt{b} to give any arbitrary size for the beam cross section. Equation (6) appears to have no simple analytical solution; a solution obtained by numerical integration is shown in Fig. 1.

The critical power for trapping of a cylindrical beam is given by

$$P = \frac{c}{4\pi} n_{\text{eff}} 2\pi \int E^{*2}(r^*) r^* dr^* (\Gamma^2 b)^{-1},$$

where $n_{\text{eff}} = k_z/k_0 = [n_0^2 + (\epsilon_2/2b)]^{1/2}$. Numerical integration of (7) gives the critical power *P* = $5.763\lambda^2 cn_{\text{eff}}/8\pi^3 n_2 n_0$, which is just equal to that given by the simple Eq. (2) for large beams. This integration also allows evaluation of b as a function of E(0) and hence gives $n_{\text{eff}} = [n_0^2 + \epsilon_2 E^2(0)/9.72]^{1/2}$. The sole dependence of power on beam diameter is through n_{eff} , which is usually unimportant except for beams with diameters as small as a few wavelengths. This, plus other nonlinearities in the dielectric material which have not been included in this approximation, will make the actual diameter of the trapped beam depend somewhat on power.

The dynamics of beam formation and changes in the dielectric material are not yet well understood. This includes questions about initiation of trapping, perturbations to which the cylindrical trapped beam is stable, and the beam's behavior if the power is appreciably above threshold. During an intense laser pulse which lasts about 10^{-8} sec, there must be time for change of the dielectric constant for trapping to develop. For electrostrictive effects, the time required is that for sound transmission across the beam's diameter, or 10^{-9} sec for a filament a few wavelengths in diameter. Time constants for Kerr effects associated with molecular rotation are of order 10^{-11} sec and are independent of beam diameter. During the response times of the dielectric material, its inertial and resistive properties will have important effects on the dynamic behavior of the light beam. To initiate a very small trapped beam, focusing a laser beam in optical material appears a natural technique,



FIG. 1. Calculated radial distribution of the electric field in a self-trapped electromagnetic wave.

since the steady-state result of the geometric optics approximation is not a focal point, as shown above, but a thin axial beam. If a broad beam is considerably above threshold, presumably it breaks up into several beams of threshold power, but in the case of slab-shaped beams of a single frequency such solutions do not seem to occur stably, and the situation is not clear for more complex shapes.

It is clear that there can be weak modulation of the steady-state beams discussed above if the modulation is fast compared with the response time of the nonlinearity. Because the beam when once trapped establishes a waveguide of appropriate characteristics for its own conduction, any weak wave of higher frequency can also easily be shown to be conducted, but not one of lower frequency. The dielectric properties of the waveguide are undisturbed to first order in the weak field as long as the beat frequency between it and the initial wave is too high for the dielectric to respond. If the beat frequency is lower, then one has a waveguide of modulated dielectric constant, and solutions for the two simultaneous waves are much more complex.

It does seem clear that two waves whose frequency difference is too high for the dielectric response are more stably trapped than is a wave of a single frequency. This is because the increase in dielectric constant of the waveguide produced by one wave which helps form the waveguide is relatively unaffected by small perturbations of the second wave and vice versa.

Table I gives values of n_2 for Kerr and for elec-

trostrictive effects, and the critical power calculated for electrostrictive effects alone. For Kerr effects, $n_2 = \frac{2}{3}\lambda J$, where J is the high-frequency Kerr constant due to molecular rotation. For electrostriction, $n_2 = \gamma^2 / 16\pi n_0 B$, where γ $=\rho dG/d\rho$, ρ is the density, and B is the bulk modulus. The critical power for trapping is calculated for electrostriction along, partly because the theory given above holds rigorously only for a circularly polarized beam, to which molecular rotation would make only a partial contribution. However, for a plane-polarized beam, the Kerr effect contributes fully to n_2 as well as to electrostriction, and it is reasonable to assume that this will result in a comparable equation for threshold power.

The most striking present experimental evidence for trapped optical beams are the extremely thin, long streaks of ionization spots and damage which sometimes occur in optical materials in which an intense laser beam is focused. These were first reported by Hercher¹ as thin threads of damage in glass and other materials, and are fairly easily demonstrated in glass by focusing a ruby-laser beam greater than a few megawatts inside good optical glass. Usually, though not always, there is extensive damage near the focal point and beyond the focal point a long straight filament of small bubbles and damage along the lens axis, accompanied by ionization. This filament may be as long as several centimeters and at the same time have a diameter of only a few wavelengths. This diameter is in some cases two orders of magnitude smaller than the focal

diameter, assuming linear optics.¹

Without trapping and a major increase in index of refraction it seems very difficult to explain such a long straight path of concentrated light energy, since the diameter of only a few wavelengths would imply rapid diffraction spreading. Any acoustic or shock waves, which might be considered an alternate source of damage, would travel a distance of only about 10^{-3} cm during the laser pulse, and would have to be of such high frequency that they would be highly attenuated.

If the laser beam is trapped in such small diameters, as is indicated, the field intensity would be of the order of 10^8 volts per centimeter, and the increase in index of refraction due to nonlinearities of the order of unity. It is hence easy to see that important optical, mechanical, and field effects can take place in the filament. The low power levels at which self-trapping can occur theoretically in liquids indicate that this phenomenon may be related to some of the unusual characteristics of stimulated Raman emission in liquids.

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NUCLEAR INTERACTIONS AND ROTATIONAL MOMENT OF F_2^{\dagger}

Irving Ozier, Lawrence M. Crapo, James W. Cederberg,* and Norman F. Ramsey Lyman Laboratory of Physics, Harvard University, Cambridge, Massachusetts (Received 9 September 1964)

The molecule F_2 is closely similar to H_2 in the following respects: Both are homonuclear molecules, both have nuclear spins of $\frac{1}{2}$ and consequently no quadrupole interaction, and both have large nuclear magnetic moments. Since measurements of the nuclear magnetic interactions and rotational moments of H_2 have been particularly valuable in studies of molecular structure and nmr relaxation processes, we have made similar measurements on F_2 by the molecularbeam magnetic-resonance method using an apparatus previously described by Baker et al.¹

The nuclear transition spectrum was initially observed in the high-field limit. A typical run in an external field H of 3735.8 gauss is shown

in Fig. 1. High-field perturbation theory shows that, for each value of the rotational magnetic quantum number, m_J , there should be a different line separated from the central $m_J = 0$ line by approximately cm_J , where c is the spin-rotation interaction constant. The structure of the individual lines can be attributed to the spin-spin interaction and second-order spin-rotation effects. Lowering H to 1864.2 gauss shifted the positions of the lines slightly and broadened them appreciably by increasing second-order effects. From the splitting of the lines |c| can be measured, and from their shapes the spin-spin interaction constant d can be deduced. The precise definitions of c and d are the same as used in the cor-



FIG. 1. Nuclear spectrum of F_2 . This spectrum was observed with a 5-turn radio-frequency coil 1.5 inches long under the following conditions: external magnetic field = 3735.8 gauss, source temperature = 77°K, rf current = 0.85 A, time constant = 50 sec. The lines above the spectrum are frequency markers spaced 20 kc/sec apart. The number beneath each spectral line is its m_J value, found by using high-field perturbation theory and the sign of c determined from the low-field experiment.