Conditions of Excitation of New Waves (LITW) at Nonlinear Interface and Diagram of Wave States of the System

ALEXANDER E. KAPLAN

Abstract—**We have found the conditions for the excitation of all of the possible waves in a nonlinear medium with a "negative" nonlinearity, in the case when a plane wave is incident from a linear medium upon a plane surface of the nonlinear mediums. We show that these conditions depend on only two independent parameters so that all possible states of the interface can be represented in a two-dimensional diagram. This diagram consists of regions of surface waves (SW), plane waves (PW), and so-called longitudinally inhomogeneous traveling waves (LITW) which were studied recently. The diagram includes two triple points: the first one corresponds to the intersection of boundaries between regions of SW, PW, and LITW states, while the other corresponds to the formation of the hysteresis (i.e., bistable) region between PW and LITW states.**

I. Introduction

In previous works [1], [2] we developed the theory of new effects associated with the reflection and refraction of a strong plane wave from the surface of a nonlinear medium whose refractive index depends on intensity of light. One of these effects is hysteresis (i.e., bistable behavior of the system). It is shown in [1] that the main requirements for observing all possible interface phenomena of this kind in the optical range (i.e., when the nonlinear component of susceptibility is small) are the almost grazing incidence of light and the very small difference between the linear susceptibilities of the interfacing media. This linear difference should be of the same order of magnitude as the nonlinear component. The effects are expected to have different features for different signs of nonlinearity; we will use the term "positive nonlinearity" for the case when the susceptibility of the nonlinear medium increases under the action of light (i.e., the nonlinear component of susceptibility is positive) and the term "negative" for the opposite situation. In the case of positive nonlinearity, hysteresis was recently observed by Smith *et al.* [3] in an experiment where CS_2 was used as a nonlinear medium. Their results show good agreement with our plane wave theory, at least for the reflection coefficient of the system.

In the case of a negative nonlinearity, new features arise which are caused by excitation of nonlinear waves of a new kind, which were considered by us as longitudinally inhomogeneous traveling waves (LITW) [2]. The intensity, as well as the angle of propagation, of these nonlinear traveling waves varies along the direction perpendicular to the interface; LITW are almost similar to a surface wave near the interface, and reduce to a conventional plane wave at infinity. Hysteresis occurs here between LITW states and plane (nonlinear) wave states. Phenomena due to LITW also include an effect of nonlinear self-deflection of refracted rays [2], [4], an effect of self-limitation of transmitted light power [2], [4], [5], and a self-induced transparency of the nonlinear interface [1], [2], [4] (for more details refer to the referenced articles).

For subsequent theoretical study of both the stability of one-dimensional LITW and their possible two-dimensional counterparts (as well as for an experimental search of LITW and the associated effects), it is necessary to predict the required conditions for an excitation of these waves. In other words, it is necessary to distinguish the region of incident field and media parameters where solutions of the kind of LITW exist (at least in the one-dimensional theory), and, specifically, the region of multivalued solutions which result in hysteresis. Let us remember that in the one-dimensional case there are five given parameters: two values of linear parts of susceptibilities of the media ϵ_0 and $\epsilon_0 + \Delta\epsilon_L$ (see below); the nonlinear coefficient of the refracting medium ϵ_2 [see (1)]; the intensity of the incident wave $|E_{in}^2|$; and its glancing angle ψ. This work was not done in the previous articles. The present paper is devoted to this problem, with the results obtained here based on the approach of previous work [2]. (However, in contrast to some of the previous results, in the present calculation no assumption is made about the smallness of the amount of nonlinearity or the glancing angle. This generalization was done because large nonlinearities in optics [7] are now available.)

We believe that an experimental search for LITW, as well as for those effects due to LITW excitation, has become a very interesting and urgent issue because very effective nonlinearities have been discovered in semiconductors such as GaAs [6] and InSb [7]. Especially important for our case is that the nonlinearity in InSb is not only very high but also negative.

Along with the LITW region, we obtain the regions of the rest of the wave states [(plane wave (PW) and surface waves (SW)] in the entire space of parameters. We will show that it is possible to combine all these parameters into two values which completely determine the behavior of the system so that the space of parameters becomes two-dimensional. This simplifies significantly the evaluation of required experimental conditions. The wave model of phenomenon used here implies that the incident wave is plane and that all waves in the system

Manuscript received August 8, 1980; revised November 5, 1980. This work was supported by the United States Air Force Office of Scientific Research under Grant AFOSR-80-0188. A. E. Kaplan is supported by the National Science Foundation.

The author is with the Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, MA 02139.

0018-9197/81/0300-0336$00.75

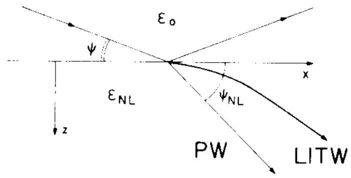

Fig. 1. Schematic diagram of waves in the plane of incidence (PW is plane wave, and LITW is longitudinally inhomogeneous traveling wave).

are one-dimensional [this means that their real amplitude depends only on the direction z, perpendicular to the interface (see Fig. 1)], so that analytical results can be obtained. This model is expected to be close to an experimentally realizable situation if bounded light beams with a broad cross section are applied. But it must be pointed out, in any case, that there are possible experimental situations in which all the equations and results of the one-dimensional wave theory are strictly valid. For example, the phenomenon of reflection of light by the junction between linear and nonlinear single-mode waveguides with metallic walls [8] can be described by the one-dimensional theory.

II. General Equations

Let us assume now that a strong plane wave of amplitude E_{in} is incident from a linear medium with susceptibility ϵ_0 at a glancing angle ψ onto the plane surface of a nonlinear medium whose susceptibility ϵ_{NL} depends on the intensity of light $|E|^2$ at a given point. In the simplest case of quadratic nonlinearity, one can write

$$\epsilon_{\text{NL}}\,(|E|^2) = \epsilon_0 + \Delta\epsilon_L - \epsilon_2 |E|^2 \tag{1}$$

where $\Delta\epsilon_L$ is the intensity-independent linear mismatch. (Here we are dealing only with negative nonlinearity, thus $\epsilon_2 > 0$.) We also assume that all waves in the system have an electric field vector polarized perpendicularly to the plane of incidence. Then the wave equation for the complex amplitude of the field E in the nonlinear medium (the total field is $\frac{1}{2}\,Ee^{-i\omega t}$ + c.c.) can be written

$$\frac{d^2E}{dz^2} + k_0^2 E\left[\frac{\epsilon_{\text{NL}}(|E|^2)}{\epsilon_0} - \cos^2\psi\right] = 0; \quad \left(k_0^2 = \frac{\omega^2\epsilon_0}{c^2}\right). \tag{2}$$

All possible solutions of this equation must satisfy generalized boundary conditions [1], [2] that are valid for linear as well as nonlinear media (including absorptive ones)

$$\frac{i\,dE(0)}{dz} + k_0 \sin\psi\,[2E_{\text{in}} - E(0)] = 0 \tag{3}$$

and generalized "radiation" conditions at infinity [1], [2]

$$|E(z)| \to \text{const} \equiv E_\infty > 0 \qquad \text{if } z \to \infty \tag{4}$$

which means an absence of waves running back from infinity toward the boundary. Boundary conditions also yield the simple formula for the reflection coefficient r:

$$r = E(0)/E_{\text{in}} - 1. \tag{5}$$

Let us now introduce two generalized parameters P and Q by the relationships

$$P = \frac{\epsilon_2 |E_{\text{in}}^2|}{\epsilon_0 \sin^2\psi}; \qquad Q = \frac{\Delta\epsilon_L}{\epsilon_0 \sin^2\psi} + 1 \tag{6}$$

which include all parameters of the field and system, and might be considered, respectively, as "normalized incident intensity" and "normalized linear mismatch." We will see below that they provide the possibility of considering only the two-dimensional space of parameters, rather than the five-dimensional one.

Furthermore, let us represent the unknown field in the nonlinear medium in the form

$$E = \sqrt{\frac{\epsilon_0}{\epsilon_2}} \cdot U(z)\sin\psi \cdot \exp\left[ik_0 \cdot \int_0^z \xi(z)\,dz + i\varphi\right] \cdot \exp\,(ik_0 \cdot x \cdot \cos\psi) \tag{7}$$

where $U(z)$ and $\xi(z)$ are unknown real functions and $\varphi = \text{const}$. Here U is a real "normalized" amplitude of the field, while the function $\xi(z)$ is related to a varied angle ψ_{NL} of the refracted rays (see Fig. 1) by the relationship

$$\tan\psi_{\text{NL}} = \xi/\cos\psi \tag{8}$$

and, under the condition $\psi \ll \Pi/2$, both values ψ_{NL} and ξ become the same; φ is the unknown phase shift of the reflected field relative to the incident one. Substituting the field in form (7) into (2), we obtain the first integral of this equation:

$$\xi U^2 = \text{const} \equiv \xi_\infty U_\infty^2 \tag{9}$$

which expresses the conservation of energy flux in the nonlinear medium (here the values ξ_∞ and U_∞ correspond to the wave at infinity $z \to \infty$), and the equation for real values:

$$\frac{d^2U}{dz^2} + k_0^2 \cdot \sin^2\psi \cdot U\left(Q - U^2 - \frac{\xi^2}{\sin^2\psi}\right) = 0. \tag{10}$$

Further integration of this equation, taking into account condition (4), yields a first-order equation for the real amplitude $U(z)$ which, in the particular case of nonlinearity (1), is as follows:

$$U^2\left(\frac{dU}{dz}\right)^2 = (k_0 \sin\psi)^2\,(U^2 - U_\infty^2)^2 \cdot \left(U_\infty^2 + \frac{U^2}{2} - Q\right). \tag{11}$$

This should describe all possible wave states that satisfy the radiation condition (4).

III. Surface and Plane Waves

It follows from (10) and condition (4) that, under condition $Q < 0$ for $z \to \infty$ (when $d^2U/dz^2 \to 0$), the amplitude U_∞ at infinity should be equal to zero. Then the conservation law (9) yields that $\xi \equiv 0$; thus, for the case $Q < 0$ only, the surface waves are possible (see region SW in Fig. 2), as well as in the linear case. Their profile follows from (11):

$$U(z) = \frac{(-2Q)^{1/2}}{\sinh\,(k_0 \cdot \sin\psi \cdot z\sqrt{-Q} + \text{const})}. \tag{12}$$

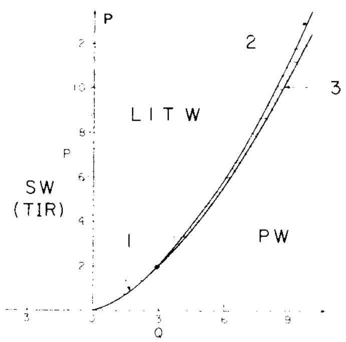

Fig. 2. Diagram of wave states in the space of parameters P and Q. Curve (1) is the boundary between PW and LITW states; curve (2) is the boundary of transition jump PW → LITW; curve (3) is the boundary of reverse transition jump LITW → PW. The space between curves 2 and 3 is the hysteresis region.

In the case $0 < U_\infty^2 < 2Q/3$, only plane homogeneous traveling waves (PW) are possible, which are characterized by the relationships

$$U(z) = \text{const} \equiv U_\infty > 0; \quad \xi(z) = \text{const} \equiv \xi_\infty > 0. \tag{13}$$

For these wave states, resulting from boundary condition (3) and taking into consideration (8), one can obtain "Snell's nonlinear formula" for the refraction angle ψ_{NL}:

$$\left(Q - \frac{\tan^2 \psi_{NL}}{\tan^2 \psi}\right) \cdot \left(1 + \frac{\tan \psi_{NL}}{\tan \psi}\right)^2 = P \tag{14}$$

and "Fresnel's nonlinear formula" for the "normalized" amplitude of the refracted wave U:

$$U^2 (Q - 1 - U^2) = 4\sqrt{P}(\sqrt{P} - U) \tag{15}$$

as well as for the reflection coefficient r (5):

$$(Q - 1)(r + 1)^2 + 4r = P(r + 1)^4. \tag{16}$$

All the formulas obtained (14)-(16) are valid for any value of $\Delta\epsilon_L$, ψ, and $\epsilon_2 |E_{in}^2|$, in contrast to the corresponding results for the same characteristics (U, ϵ_{NL}, r) in [2] which are valid only when these values are small.

IV. LITW and Region of Their Existence

Now if $U_\infty^2 > 2Q/3 > 0$, in addition to plane nonlinear waves (13), excitation of LITW is possible. Integration of (11) yields in this case [2]

$$U^2 = U_\infty^2 \pm 2B^2 \Big/ \begin{matrix}\sinh^2\\ \cosh^2\end{matrix} (Bk_0 \sin\psi \cdot z + C),$$

$$B^2 = \frac{3U_\infty^2}{2} - Q. \tag{17}$$

For $U_\infty^2 = 2Q/3$ (i.e., the minimum possible value of U_∞^2 for LITW) only one (limiting) type of LITW remains:

$$U^2 = \frac{2Q}{3} + \frac{2}{(k_0 z \sin\psi + C)^2}. \tag{18}$$

Here, a very interesting problem, the so-called continuum problem, [2], [4], [5], [8], arises: under given boundary conditions (3) and radiation conditions (4), it is impossible to distinguish a countable set of LITW solutions [i.e., these conditions are inadequate to determine all the unknown constants, U_∞ and C in (17) and φ in (7)]; this problem is specific only for LITW. In this situation, it is necessary to introduce an additional physical factor into the model in order to "remove" the continuum. Taking into consideration the simplest factor, such as small absorption in a nonlinear medium, it is possible to choose a unique, physically realizable type of LITW by taking the limit as the absorption goes to zero.

It was proven analytically in our work, by asymptotic theory [2], [4], and by numerical calculations in [5], [9], that the only kind of LITW that survived under this transition is the simplest (limiting) solution (18). It possesses the only possible value of U_∞, which should be the minimum and equal to $2Q/3$. Therefore, it is now easy to calculate the value of the LITW intensity at the boundary $U_0 = U(z = 0)$ [or equivalently, the values of C in (18)], using boundary condition (3), which yields

$$U_0^2 = Q - 1 \pm [8P - (\sqrt{Q/3} + 1)^3 (\sqrt{3Q} - 1)]^{1/2} \gtrless U_\infty^2 = \frac{2Q}{3}. \tag{19}$$

Thus, the entire solution for the field E is determined by the formula (7) where $U(z)$ is now defined by (18) with

$$C = \sqrt{2}\,(U_0^2 - 2Q/3)^{-1/2}. \tag{20}$$

$\xi(z)$ is determined by (9), where $U_\infty^2 = 2Q/3$ and $\xi_\infty = \sin\psi\sqrt{Q/3}$ [which follows from (10) for $z \to \infty$)]; thus, the integral in (7) can be written as

$$k_0 \int_0^z \xi dz = k_0 \sin\psi \sqrt{Q/3} - \arctan\left[\frac{k_0 z \sin\psi\sqrt{Q/3}}{1 + C(k_0 z \sin\psi + C)(Q/3)}\right] \tag{21}$$

where C is defined by (20).

φ is determined by the boundary condition (3) which yields

$$\cos\varphi = \frac{U_0(\xi_0 + \sin\psi)}{2\sqrt{P} \cdot \sin\psi}; \quad (\tan\varphi < 0) \tag{22}$$

where $\xi_0 = 2(Q/3)^{3/2} U_0^{-2}$. Equations (19)-(22) are also valid now for any value of ψ, $\Delta\xi_L$, and E_{in}^2. The boundary between PW and LITW states in the space of parameters (for nonhysteresic cases) can now be defined by the equality $U_{PW} = U_{0_{LITW}}$. Substituting expression (19) for U_0 into (15) for U, one can obtain the formula for the boundary between PW and LITW states in the plane of parameters P and Q:

$$P = \tfrac{1}{6} Q (1 + \sqrt{Q/3})^2; \quad (P < 2, Q < 3) \tag{23}$$

(see curve 1 in Fig. 2). Under the condition $P < 2$, $Q < 3$, only one wave state should be realized—namely, PW [see (15)] below the curve (23) or LITW [upper sign in (19)] above it—for any given values of P and Q. When P or Q changes, transition from one state to the other is continuous, as can be seen in Fig. 3 (curve 1), where the behavior of the amplitude U_0 as a function of P is shown for $Q < 3$.

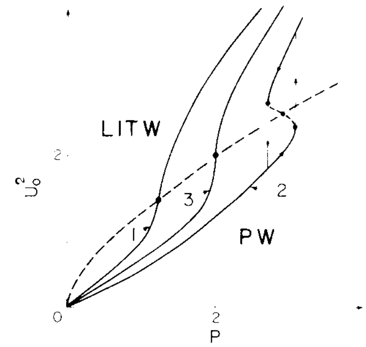

Fig. 3. Behavior of intesnsity U_0^2 of a nonlinear wave at the interface as a function of the parameter P: curve (1) $Q < 3$; curve (2) $Q > 3$; curve (3) $Q = 3$.

But under the conditions

$$P > 2, Q > 3; \quad (2\epsilon_0 \sin^2 \psi < \Delta\epsilon_L < \epsilon_2 |E_{\text{in}}^2|) \tag{24}$$

a region exists in the parameter space where there are three possible states for any given values of P and Q: two LITW [both signs in (19)] and one PW state (or vice-versa). The values of field intensity for PW as well as LITW are given now as before by (15) and (19), respectively, but their features are that, in the region mentioned, the curve of states (as a function of one of the parameters) becomes multivalued (in our case, it is s-shaped, see curve 2 in Fig. 3). Let us assume that those two out of three branches of s-curves are supposed to be stable (inside the region of multistability), which have a continuation into the region of a single state (the solid branches of curve 2 in Fig. 3). It results in an energy criterion for stability of solution [2]:

$$d|E_0^2|/d|E_{\text{in}}^2| > 0. \tag{25}$$

This criterion causes the stable LITW states to be those with the upper sign in (19), and the stable PW states to satisfy $U^2 < \min\{U_{\text{hys}}^2; 2Q/3\}$ where

$$U_{\text{hys}}^2 = \tfrac{1}{8}\,(4Q - 3 + \sqrt{8Q + 1}). \tag{26}$$

The transition jump between stable branches of LITW and PW should take place at points where the number of states changes, i.e., where the characteristic derivative with respect to given parameters is infinite, for instance,

$$dU_0^2/dP = \infty \tag{27}$$

(see curve 2 in Fig. 3). According to this criterion, jumps from the PW to the LITW state occur just along the curve determined by the equality $U^2 = U_{\text{hys}}^2$ [see (26)]. Substituting this value of U^2 into (15) one can obtain the boundary of the hysteresis region in the space of parameters P and Q, which corresponds to these jumps (PW → LITW):

$$P = 2^{-10}\,(\sqrt{8Q + 1} - 1)\cdot(\sqrt{8Q + 1} + 3)^3 \tag{28}$$

(see curve 2 in Fig. 2). The reverse jumps (from LITW to PW states), according to (27) and (19), occur when the square root in (19) goes to zero. This determines the other boundary of the hysteresis region (LITW → PW) in the space of P and Q:

$$P = 2^{-3}\,(\sqrt{Q/3} + 1)^3\,(\sqrt{3Q} - 1) \tag{29}$$

(see curve 3 in Fig. 2). As can be seen from Fig. 2, there are two triple points on the diagrams of states; one is $P = Q = 0$ and the other is $P = 2$, $Q = 3$, which is more interesting for us because of hysteresis. Let us estimate the intensity of light required to reach this point. In the case of InSb, the nonlinear coefficient can be as large as 10^{-2} ESU [7] under some conditions. If we set $\Delta\epsilon_L/\epsilon_0 \sim 10^{-3}$, then the glancing angle ψ, required to achieve the value $Q = 3$, should be of the order of $\sim 1.3°$. Now to get $P = 2$, the required incident field should be $E_{\text{in}} \sim 100$ V/cm. This corresponds to $\sim 134\sqrt{\epsilon_0}$ W/cm^2 which can be achieved by using existing CW lasers.

Qualitative discussion of some problems connected with the influence of actual absorption and nonlinear diffraction (in the case of a bounded beam rather than infinite plane incident wave) was made in [2]. Here we would like to point out that these factors lead to some limitation of effects. For instance, within the bounds of one-dimensional theory, taking absorption into consideration, one can show that at a given value of Q, hysteresis should disappear entirely for any incident power (P) if the absorption reaches some definite threshold value. Let us denote a given length of light absorption in a nonlinear medium by L; it is possible to show that the threshold value of this length is

$$L_{\text{thr}} = \frac{\alpha}{k_0}\left(\frac{\Delta\epsilon_L}{\epsilon_0} + \sin^2\psi\right)^{-1} \tag{30}$$

where α is a numerical coefficient independent of parameters of the problem. A preliminary estimate yields $\alpha \sim 10$–20. If we use the above example and $k_0 = 10^5$ cm^{-1}, L_{thr} should be of the order 0.1 cm, so that the required condition $L > L_{\text{thr}}$ can be satisfied.

Acknowledgment

The author is grateful to P. W. Smith and W. J. Tomlinson for their helpful notes, and to P. L. Kelley, R. L. Aggarwal, and B. Lax for their support.

References

[1] A. E. Kaplan, "Hysteresis reflection and refraction by a nonlinear boundary–A new class of effects in nonlinear optics," *JETP Lett.*, vol. 24, pp. 114–119, Aug. 5, 1976; —, "Theory of hysteresis reflection and refraction of light by a boundary of a nonlinear medium," *Sov. Phys. JETP*, vol. 45, pp. 896–905, May 1977.

[2] —, "Criterion of existence of longitudinally inhomogeneous traveling waves in nonlinear electro-dynamics," *Sov. J. Quantum Electron.*, vol. 8, pp. 95–97, Jan. 1978; —, "Longitudinally inhomogeneous running waves and their role in the nonlinear reflection and refraction of light," *Radiophys. Quantum Electron.*, vol. 22, pp. 229–241, Mar. 1979.

[3] P. W. Smith, J.-P. Hermann, W. J. Tomlinson, and P. J. Maloney, "Optical bistability at a nonlinear interface," *Appl. Phys. Lett.*, vol. 35, pp. 846–848, Dec. 1, 1979; P. W. Smith and W. J. Tomlinson, "Optical properties of nonlinear interfaces," in *Proc. Int. Conf. Optical Bistability*, Asheville, NC, June 3–5, 1980, to be published.

[4] A. E. Kaplan, "Physical effects associated with reflection of light at surface of negative-nonlinear medium," *Dig. Rep. 9th Nat. Conf. Coherent and Nonlinear Opt.*, Leningrad, USSR, June 13–16, 1978, p. 229; —, "Continuum problem for inhomogeneous travelling waves in the negative-nonlinear medium," in *Dig. Rep. 9th Nat. Conf. Coherent and Nonlinear Opt.*, Leningrad, USSR, June 13–16, 1978, p. 232.

[5] O. V. Bagdasaryan and V. A. Permyakov, "Branching of conditions and effect of energy flux limitation of TE mode in medium with ionization nonlinearity," *Radiophys. Quantum Electron.*, vol. 21, pp. 940-947, Sept. 1978.
[6] M. M. Gibbs, S. L. McCall, T.N.C. Venkatesan, A. C. Cossard, A. Passner, and W. Wiegmann, "Optical bistability in semiconductors," *Appl. Phys. Lett.*, vol. 35, pp. 451-453, Sept. 15, 1979.
[7] D.A.B. Miller, M. A. Mozolowski, A. Miller, and S. D. Smith, "Nonlinear optical effects in InSb with C.W. CO_2 laser," *Opt. Commun.*, vol. 27, pp. 133-136, Oct. 1978; D.A.B. Miller, S. D. Smith, and B. S. Wherett, "The microscopic mechanism of third-order optical nonlinearity in InSb," *Opt. Commun.*, to be published.
[8] A. E. Kaplan, "Theory of plane wave reflection and refraction by the nonlinear interface," in *Proc. Int. Conf. Optical Bistability*, Asheville, NC, June 3-5, 1980, to be published.
[9] N. N. Rosanov, "Hybrid conditions for optical reflection and transmission by nonlinear media," *Sov. Tech.-Phys. Lett.*, vol. 3, pp. 239-241, 1977.

Alexander E. Kaplan was born in Kiev, Russia, on June 9, 1938. He received the diploma in Physics from the Moscow Physical Technical Institute, Moscow, Russia, and the Ph.D. degrees in Physics and Mathematics from the Academy of Sciences of the USSR, Moscow, and Gorkii State University in 1961 and 1967, respectively.

From 1963 to 1979 he was a Research Scientist at the Academy of Sciences of the USSR, working on the theory of nonlinear oscillations, radiophysics, quantum electronics, and nonlinear optics. Since December 1979 he has been a member of the Research Staff at the Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge. His present research interests include the theory of interaction of light with nonlinear media and interfaces and its application to optical bistability.

Dr. Kaplan is a member of the American Physical Society.