Surface solitons due to second order cascaded nonlinearity

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Abstract

In this paper we show theoretically that cascaded surface solitons can exist on the surface of a medium with the bulk inversion symmetry center. Although in the bulk of such media the second order nonlinearity vanishes, $\chi^{(2)} = 0$, the optical quadratic nonlinearity appears due to the contribution of spatial derivatives of the fields to the nonlinear response. This nonlinearity is localized at the surface and leads to the cascaded $\chi^{(2)}$ surface solitons. © 1999 Published by Elsevier Science B.V. All rights reserved.

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Propagation of solitons in media with cascaded second order nonlinearity has recently attracted a considerable attention (see, e.g., a review article [1]). Along with the theoretical studies the experiments demonstrated also the existence of such type of solitons [2,3]. In the present paper we would like to attract attention to the possibility of the analogous phenomena at a plane surface of media with bulk inversion symmetry. Although in such media the bulk value of second order nonlinearity vanishes, $\chi^{(2)} = 0$, the optical quadratic nonlinearity appears due to the contribution of spatial derivatives of the fields to the nonlinear response. Such a nonlinearity is localized mainly at the interface between two media with different dielectric constants and leads, e.g., to the second harmonic generation in reflection of light from metals (see, e.g., Refs. [4,5]). In metals the dielectric constant can have large absolute values at frequencies sufficiently low compared to the plasma frequency. The normal component of the electric field changes drastically across the interface (for example, between metal and vacuum) which contributes strongly to the nonlinearity under discussion. However, not only fields but material constants undergo also drastic changes within a transitional layer, and we have to take into account their first spatial derivatives within the same accuracy of approximation. Therefore, the new quadratic terms become possible [6,7], which contribute to the surface nonlinearity on equal footing.

In view of the fact that the nonlinearity in the case under discussion is localized at the surface, one may expect that different kinds of nonlinear waves can exist at the surface of such media and, particularly, parametric solitons analogous to those in media with second order nonlinearity. In this article we show that this is the case indeed. Note that optical solitons at a surface have been studied theoretically for many years; for example, in Ref. [8] the existence of surface self-induced transparency solitons has been shown, and in Ref. [9] solitons on a surface of a medium with Kerr nonlinearity have been investigated. Here we shall consider solitons of quite a different origin, using the nonlinear boundary conditions (BC) for the fields. These boundary conditions can be constructed (as was done in Refs. [6,7]) on the basis of the above
mentioned quadratic nonlinear constitutive relations between the displacement vector $\mathbf{D}$ and the electric field $\mathbf{E}$.

We consider a homogeneous and isotropic medium which possesses inversion symmetry, so that the dipole contribution to the bulk nonlinear polarization is absent ($\chi^{(2)} = 0$). The surface of the medium coincides with the $x,y$ plane, so the half-space $z > 0$ corresponds to vacuum (the dielectric constant $\varepsilon = 1$) and the half-space $z < 0$ to the medium, characterized by the frequency dependent dielectric function $\varepsilon = \varepsilon(\omega)$. The surface nonlinearity is localized in the medium–vacuum interface layer and can be of intrinsic or artificial origin. The intrinsic nonlinearity arises due to the presence of the surface itself. The presence of the surface breaks the inversion symmetry, and since both the electromagnetic fields and material constants vary rapidly at the surface, their gradients give rise to the optical nonlinearity of the surface [4,5]. This type of surface nonlinearity is especially strong in the case of metal surfaces, whose dielectric function can be large in its absolute value. The nonlinear transition layer can be created artificially by adsorption of a thin layer of optically nonlinear molecules on the surface. In both cases the nonlinearity is localized in the medium–vacuum interface layer that has a finite thickness on the microscopic scale.

Consequently, it can be taken into account through the nonlinear terms in the Maxwell equations for the electromagnetic fields, which can be obtained by integrating the Maxwell equations across the interface layer and subsequently passing to the limit of a vanishing thickness of the layer [6]. As is usually done in the theory of cascaded solitons in the bulk [10–18], we take into account only the fields with fundamental $\omega$ and second harmonic $2\omega$ frequencies $\mathbf{F}(\mathbf{r},t) = \mathbf{F}_1(\mathbf{r})\exp(-i\omega t) + \mathbf{F}_2(\mathbf{r})\exp(-2i\omega t) + c.c.,$ where $\mathbf{F} = \mathbf{E},\mathbf{D},\mathbf{H}$. The boundary conditions for the fundamental and harmonic fields can be derived in the way described in Ref. [6] and have the form:

$$H_{1x}^\omega - H_{1y}^\omega = \frac{i\omega}{c} \left[ E_{1x}^\omega D_{2z}^\omega \psi_1 + E_{2x}^\omega D_{1z}^\omega \psi_1 \right],$$

$$H_{2x}^\omega - H_{2y}^\omega = \frac{2i\omega}{c} E_{1x} D_{1z} \psi_1,$$  

$$E_{1x}^\omega - E_{1y}^\omega = -\frac{\partial}{\partial x} \left[ E_{1x}^\omega E_{2}\mu_1 + D_{1z}^\omega D_{2z}^\omega 2\mu_{1z} \right],$$

$$E_{2x}^\omega - E_{2y}^\omega = -\frac{\partial}{\partial x} \left[ E_{1x}^\omega \mu_2 + D_{2z}^\omega \mu_{1z} \right].$$

where the asterisk denotes complex conjugate, and the nonlinear coefficients appearing in the boundary conditions are the phenomenological parameters characterizing the linear and nonlinear optical properties of the interface layer, and are to be determined from either experiments or microscopic calculations. When the nonlinear response is associated with a layer of non-centrosymmetric molecules adsorbed on the surface the nonlinear coefficients are expressed through the nonlinear polarizability of the molecules.

For the plane wave solutions of the Maxwell equations

$$E_1(z > 0) = \left( \frac{c\kappa_1}{\omega} \varepsilon_x + ick \varepsilon_y \right) A_1 \exp(ikx - \kappa_1 z).$$

$$E_2(z > 0) = \left( \frac{c\kappa_2}{\omega} \varepsilon_x + ick \varepsilon_y \right) A_2 \exp(2ikx - 2\kappa_2 z).$$

$$E_1(z < 0) = \left( -\frac{c\kappa_1}{\omega} \varepsilon_x + ick \varepsilon_y \right) \tilde{A}_1 \exp(ikx + \kappa_1 z),$$

$$E_2(z < 0) = \left( -\frac{c\kappa_2}{\omega} \varepsilon_x + ick \varepsilon_y \right) \tilde{A}_2 \exp(2ikx + 2\kappa_2 z).$$

where $\kappa_{1,2}^2 = k^2 - \varepsilon_{1,2} \omega^2/c^2,$ $\tilde{\kappa}_{1,2}^2 = k^2 - \varepsilon_{1,2} \omega^2/c^2,$ we obtain from the nonlinear boundary conditions (1)–(4) the following equations

$$F_1(k,\omega) A_1 = -\frac{ick}{\omega} \psi_1 A_1^2,$$

$$F_2(k,\omega) A_2 = -\frac{2ick}{\omega} \psi_2 A_2^2,$$

where

$$F_{1,2}(k,\omega) = \varepsilon_{1,2} \tilde{\kappa}_{1,2} + \tilde{\varepsilon}_{1,2} \kappa_{1,2},$$

$$\psi_1 = 2\mu_1 \tilde{\varepsilon}_{1,2} \kappa_{1,2} + 2\mu_{1z} e_1 \varepsilon_{1,2} k^2 + \varepsilon_{1,2} \kappa_1$$

$$- v_1 e_1 \tilde{\kappa}_{1,2},$$

$$\psi_2 = \mu_2 \tilde{\varepsilon}_{1,2} \kappa_{1,2} - \mu_{1z} e_2 \varepsilon_{1,2} k^2 + v_2 e_2 \tilde{\kappa}_{1,2}.$$  

These equations are the amplitude-dependent dispersion laws for the surface electromagnetic waves. Equating to zero their left-hand sides leads to the well-known dispersion laws for the linear surface waves [19]:

$$k_{1,2}(\omega) = \frac{\omega}{c} \sqrt{\frac{\varepsilon_{1,2} \tilde{\varepsilon}_{1,2}}{\varepsilon_{1,2} + \tilde{\varepsilon}_{1,2}}}. $$

Let us first consider the temporal soliton solutions. In this case the amplitudes $A_{1,2}$ are slowly varying functions of $x$ and $t$. For the linear waves $E_1 \propto A_1 \exp(ikx - i\omega t), E_2 \propto A_2 \exp(2ikx - 2i\omega t)$ we have $F_1(k,\omega) = 0, F_2(k,\omega) = 0,$ and these equations are equivalent to the linear differential equations

$$F_1 \left( -\frac{i}{c} \frac{\partial}{\partial x} , i \frac{\partial}{\partial t} \right) E_1 = 0, \quad F_2 \left( -\frac{i}{2} c \frac{\partial}{\partial x} , i \frac{\partial}{\partial t} \right) E_2 = 0.$$

We seek the solution of these linear equations in the form

$$E_1 = A_1(x,t) \exp(ik_1 x - i\omega_0 t),$$

$$E_2 = A_2(x,t) \exp(i2k_2 x - 2i\omega_0 t),$$

where $\omega_0$ is the frequency of the linear wave. The equations for the amplitudes $A_1(x,t), A_2(x,t)$ can be solved by separating of variables, and the solutions are described by the Airy functions (see Ref. [20]).
so that \( F_1 (k_1, \omega_1) = 0 \) and \( F_2 (k_2, \omega_2) = 0 \). Assuming that the amplitudes change little on one wavelength and one period we obtain the following equations for the slowly varying envelope amplitudes \( A_{1,2} \):

\[
\begin{align*}
- \frac{\partial A_1}{\partial x} - ik'_1 \frac{\partial A_1}{\partial t} + \frac{k''_1}{2} \frac{\partial^2 A_1}{\partial t^2} & = \frac{x_1}{F_{1k}} A_1^* A_2 \exp(2i(k_2 - k_1)x), \\
- \frac{i}{2} \frac{\partial A_2}{\partial x} - ik'_2 \frac{\partial A_2}{\partial t} + \frac{k''_2}{8} \frac{\partial^2 A_2}{\partial t^2} & = \frac{x_2}{F_{2k}} A_1^2 \exp(2i(k_1 - k_2)x),
\end{align*}
\]

where \( F_{1k} \) stands for \( (\partial F_1 / \partial k)_{k_1, \omega_1} \), \( F_{2k} \) for \((\partial F_2 / \partial k)_{k_2, \omega_2} \), and

\[
k_{1,2} = \frac{d^2 k_{1,2}(\omega)}{d\omega^2} |_{\omega = \omega_1}, \quad k''_{1,2} = \frac{d^2 k_{1,2}(\omega)}{d\omega^2} |_{\omega = \omega_2}.
\]

In the dimensionless variables \( \xi = x / x_0 \), and \( s = (s - k_1 x) / t_0 \), where \( x_0 = t_0 / k_1 \), \( t_0 \) is a free time parameter, the reduced equations for the renormalized fields \( a_1 = A_1 x_0 |(2x_1 \chi') / (F_{1k} F_{2k})| \), and \( a_2 = A_2 x_0 \chi' / F_{1k} \), take the form

\[
\begin{align*}
\frac{\partial a_1}{\partial \xi} - \sigma \frac{\partial^2 a_1}{\partial \xi^2} + a_1 a_2 e^{-i\gamma} & = 0, \\
\frac{\partial a_2}{\partial \xi} - i \frac{\partial a_2}{\partial s} - \gamma \frac{\partial a_2}{\partial s^2} + \Gamma a_1^2 e^{i\gamma} & = 0,
\end{align*}
\]

where \( \gamma = k''_2 / (2k''_1) \), \( \delta = 2(k_1 - k_2)x_0 / t_0 \), \( p = 2(k_1 - k_2)x_0 \), \( \sigma = \text{sign } k'_1 \), and \( \Gamma = \text{sign } (x_1 \chi' / (F_{1k} F_{2k})) \).

Equations of this kind have been studied in many papers on cascaded \( \chi^{(2)} \)-nonlinearity \([1,2,10,18,21]\) in Refs. \([10,11,18,21]\) several exact solitary wave solutions were found, and in Refs. \([20,21]\) with the use of the variational approach these equations were shown to have two-parametric families of soliton-type solutions. In Refs. \([22-24]\) it was shown that in the case of a large mismatch \( p \) these equations reduce to the nonlinear Schrödinger (NLS) equation.

For a metal–vacuum interface the linear surface waves exist for both fundamental and second harmonics when \( 2 \omega < \omega_p / \sqrt{2} \), \( \omega_p \) being the plasma frequency. In the infrared spectral region damping of linear surface waves is weak, so that the propagation length is large, about several centimeters. For the frequency \( \omega_p / c = 2 \pi \times 10^3 \text{ cm}^{-1} \) (the corresponding wavelength in vacuum is \( \lambda = 10 \mu \text{m} \)), we obtain for the parameters in Eqs. (19) and (20) the following values: \( \gamma = 2.0 \), \( \delta = -5.7 \times 10^{14} \), \( p = -3.6 \times 10^{28} \), if we set \( t_0 = 1 \) s and take \( \omega_p / c = 4.5 \times 10^5 \text{ cm}^{-1} \) for a silver surface. Therefore, Eqs. (19) and (20) reduce to the NLS equation, which in our case has the form:

\[
\begin{align*}
\frac{i}{2} \frac{\partial a_1}{\partial \xi} - \frac{\partial^2 a_1}{\partial \xi^2} + \frac{\Gamma}{p} |a_1|^2 a_1 & = 0, \\
\frac{i}{2} \frac{\partial a_2}{\partial \xi} - \frac{\partial^2 a_2}{\partial \xi^2} + \frac{\Gamma}{p} |a_1|^2 a_2 & = a_2 - \frac{\Gamma}{p} a_1^2 e^{i\gamma},
\end{align*}
\]

When \( \Gamma / p < 0 \), it has the well-known two-parametric family of soliton solutions:

\[
a_1 = \sqrt{-\sigma pK^2 / \Gamma} \exp \left[ i(\Omega \xi - i\Omega_s) \right] \cosh \left[ K (s - V \xi) \right],
\]

with two parameters \( K \) and \( \Omega \), where \( V = \sigma \Omega \), \( Q = \sigma (\Omega^2 - K^2) / 2 \).

The largest contributions to \( \psi_{1,2} \) come from the terms with nonlinear coefficients \( \mu_{12} \) and \( \mu_{12} \). The values estimated from the free-electron model \([25]\) are \( \mu_{12} = \mu_{12} = -0.8 \times 10^{-14} \text{ CGSE} \). In this case we obtain \( (\chi_0 / F_{1k}) = i \times 1.1 \times 10^{-6} \text{ CGSE}, (\chi_2 / F_{2k}) = i \times 2.2 \times 10^{-6} \text{ CGSE} \), thus, \( \Gamma = 1 \) and, therefore, the solitons can exist, indeed.

Returning to the dimensional coordinates \( x \) and \( t \), we see that due to the small value of \( c^2 k''_1 = 0.95 \times 10^7 \text{ cm} \) and \( k'_1 = 1 / c \) we can neglect \( Q \) and \( V \). Then the soliton has the shape

\[
\begin{align*}
A_1 & = \mathcal{A}_1 \exp \left[ i(\Omega / \tau_p)(x - ct) \right] \cosh \left[ \left( 1 / ct_p \right)(x - ct) \right], \\
A_2 & = \mathcal{A}_2 \exp \left[ i(\Omega / \tau_p)(x - ct) \right] \cosh \left[ \left( 1 / ct_p \right)(x - ct) \right],
\end{align*}
\]

where

\[
\mathcal{A}_1 = \sqrt{F_{1k} F_{2k}} \chi_1 \chi_2 \left( k_1 - k_2 \right), \quad \mathcal{A}_2 = \frac{F_{2k}}{\chi_1},
\]

If we take the pulse duration \( \tau_p = t_0 / K \) to be \( \tau_p = 1 \) ps, we obtain \( \mathcal{A}_1 = 0.8 \times 10^6 \text{ CGSE}, \mathcal{A}_2 = 0.9 \times 10^5 \text{ CGSE} \), while the peak power \( P_1 = 0.5 \times 10^8 \text{ W/cm} \), and \( P_2 = 0.13 \times 10^8 \text{ W/cm} \). Note that the increase of the pulse duration leads to the decrease of its energy. The peak power of the pulse decreases also in the near infrared spectral region. For example, for \( \lambda = 1.06 \mu \text{m} \) we obtain \( \mathcal{A}_1 = 1.6 \times 10^5 \text{ CGSE}, \mathcal{A}_2 = 2.4 \times 10^5 \text{ CGSE} \), while the peak power \( P_1 = 1.8 \times 10^5 \text{ W/cm} \), and \( P_2 = 0.9 \times 10^5 \text{ W/cm} \). However, the propagation length of surface waves
decreases substantially and is about tens of micrometers in this spectral region.

The situation is more favorable when the nonlinear response is associated with a layer of extended charge-transfer molecules [26] on the Al surface. In this case we obtain \( \alpha_1 = 1.7 \times 10^5 \) CGSE, \( \alpha_2 = 5.6 \) CGSE, \( P_1 = 2.0 \times 10^9 \) W/cm, \( P_2 = 2.2 \times 10^7 \) W/cm for \( \lambda = 10 \) \( \mu \)m, and \( \alpha_1 = 5.2 \times 10^5 \) CGSE, \( \alpha_2 = 7.9 \) CGSE, the peak power \( P_1 = 1.9 \times 10^5 \) W/cm, and \( P_2 = 0.11 \) W/cm for \( \lambda = 1.06 \) \( \mu \)m.

The estimates show that in the case of free silver surface the conversion into the second harmonic is weak for temporal solitons. The energy characteristics of the pulses can be substantially decreased by using the interface between a metal and a highly refractive dielectric or semiconductor.

We turn now to the consideration of spatial solitons. In the stationary picture, \( A_1, A_2 \) are independent of time, but are slowly varying functions of \( x \) and \( y \). We obtain again Eqs. (7) and (8) but with the two-dimensional wave vector \( \mathbf{k} = (k_x, k_y) \):

\[
F_1(k_x, k_y, \omega_0) A_1 = \chi_1(k_x, \omega_0) A^*_2 A_2,
\]

\[
F_2(k_x, k_y, \omega_0) A_2 = \chi_2(k_x, \omega_0) A^*_1 A_1^*,
\]

where \( F_{1,2}(k_x, k_y, \omega_0) = F_{1,2}(k^2_x + k^2_y, \omega_0) \). We consider light beams which propagate in the \( x \)-direction with the slow dependence of the amplitudes on the \( y \) coordinate and assume for simplicity that both beams are parallel to each other. This corresponds to the expansion around \( k_x = (k_x, 0) \), \( k_y = (k_0, 0) \). In the way described above we obtain the following coupled equations for the slowly varying envelope amplitudes

\[
-i \frac{\partial A_1}{\partial x} + \frac{1}{2k_1} \frac{\partial^2 A_1}{\partial y^2} = \frac{\chi_1}{F_{1k}} A_1^* A_2 \exp(2i(k_x - k_0) x),
\]

\[
-i \frac{\partial A_2}{\partial x} - \frac{1}{8k_2} \frac{\partial^2 A_2}{\partial y^2} = \frac{\chi_2}{F_{2k}} A_2^* A_1 \exp(2i(k_x - k_0) x),
\]

where the nonlinear coefficients are given by Eq. (18).

Using the dimensionless variables \( \xi = x / (k_1 y_0^2) \), \( y = y / y_0 \) and renormalized fields \( a_1 = A_1 k_1 y_0^2 / \sqrt{\chi_1 \chi_2 / (F_{1k} F_{2k})} \) and \( a_2 = A_2 k_1 y_0^2 \chi_1 / F_{1k} \), we obtain the reduced equations

\[
\frac{\partial a_1}{\partial \xi} + \frac{1}{2} \frac{\partial^2 a_1}{\partial \xi^2} + a_1 a_2 e^{-i\rho \xi} = 0,
\]

\[
\frac{\partial a_2}{\partial \xi} - \frac{\gamma}{2} \frac{\partial^2 a_2}{\partial \xi^2} + \Gamma a_1^2 e^{i\rho \xi} = 0,
\]

where \( \gamma = -k_0 / (2k_2), \rho = 2(k_x - k_1) k_1 y_0^2 \), and \( \Gamma = \text{sign}(\chi_1 \chi_2 / (F_{1k} F_{2k})) \).

The numerical estimates show that the NLS equation is not applicable in this case. The parameters suit better to the exact solution [10,11,18,21] of Eqs. (28) and (29):

\[
A_1 = \frac{\alpha_1 \exp(ik_x x)}{\cosh^2(y/l)}, \quad A_2 = \frac{\alpha_2 \exp(ik_x x)}{\cosh^2(y/l)},
\]

\[
q_1 = \frac{4k_2(k_x - k_1)}{4k_2 - k_1}, \quad q_2 = \frac{2k_1(k_x - k_1)}{4k_2 - k_1},
\]

\[
l = \frac{4k_x - k_1}{2k_1 k_2(k_x - k_1)}
\]

\[
A_1 = \frac{F_{1k}}{2\chi_1 \chi_2} \frac{3(k_x - k_1) k_1 k_2}{4k_2 - k_1},
\]

\[
A_2 = \frac{F_{1k}}{2\chi_1} \frac{3(k_x - k_1) k_1 k_1^*}{4k_2 - k_1}.
\]

For \( \lambda = 10 \) \( \mu \)m we have \( l = 111 \) \( \mu \)m, \( \alpha_1 = 0.9 \times 10^6 \) CGSE, \( \alpha_2 = 1.8 \times 10^5 \) CGSE, and for the total power \( J_1 = 1.1 \times 10^{10} \) W, \( J_2 = 8.6 \times 10^6 \) W. For the layer of the charge-transfer molecules on the Al surface we obtain \( \alpha_1 = 1.1 \times 10^5 \) CGSE, \( \alpha_2 = 0.6 \times 10^5 \) CGSE, \( J_1 = 1.8 \times 10^7 \) W, \( J_2 = 0.26 \times 10^7 \) W. The increase of the frequency leads to the decrease of the soliton width \( l \) and to the increase of the total power of the pulse.

Thus, we have shown that a new type of surface solitons can propagate along the interface between a metal and a dielectric. In spite of the absence of second order nonlinearity in the bulk media, the nonlinearity in the surface boundary conditions leads to cascaded \( \chi^{(2)} \) surface solitons. Our estimates show that in principle such solitons can be observed in experiments.

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