

# NON-ENVELOPE FORMULATION FOR FEMTOSECOND OPTICAL PULSES IN SEMICONDUCTORS

*I.V.Mel'nikov<sup>1)</sup>, D.Mihalache<sup>2)</sup>, F.Moldoveanu<sup>+</sup>, N.-C.Panoiu\**

*Institut für Festkörpertheorie und Theoretische Optik, F.Schiller - Universität Jena  
D-07743 Jena, Germany*

*<sup>+</sup>Department of Physics, University of Maryland  
MD 20781, USA*

*\*Department of Physics, New York University  
New York, NY 10003, USA*

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We analyze the response of the ensemble of 1s-excitons driven by a femtosecond optical pulse beyond traditional approach of the slowly varying amplitudes. For optical pulses of a given duration it is shown that the off-resonance optical field can evolve into a stable soliton with non-zero asymptotics.

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Over the past few years, optical pulse durations as short as 6–10 femtoseconds (fs) pulses have been demonstrated for wavelengths ranged from the blue-green to the near infrared. These have been widely exploited to generate a unipolar single-cycle electromagnetic pulse in a variety of nonlinear media [1] and has also resulted in a flurry of activity in theoretical studies in order to answer the question of whether it is possible to obtain the right knowledge relevant to the dynamics of such pulses within the traditional framework of the slowly varying envelope approximation (SVEA) operating with a quasi-monochromatic field. From this point of view, it is of a great importance the recent observation pertaining to a Kerr self-focusing that SVEA loses its justification long before the pulse duration approaches an optical cycle [2].

The problem met by the SVEA in a fs domain is that both the wide spectrum of the pulse and its intense field increase the number of harmonics have to be included into the expansion of the polarization in series of field powers and adjust phase-matching conditions for all harmonics simultaneously. This breaks down the SVEA basic assumptions of a weakly nonlinear and strongly dispersive medium, the superposition does not act what, in turn, prevents one in reducing the consideration to a finite number of interacting waves. And what is more, quantum-mechanical effects may come into action at the subwavelength scale.

The purpose of this Letter is to go beyond the SVEA to show the advantages of the self-consistent description based on the semiconductor Maxwell–Bloch equations (SMBEs) and to impose proper relationships among nonlinearity, dispersion, dissipation (or amplification), and backward-scattering effects. We derive an asymptotic analytic solution for the induced polarization of excitons at low density. It gives rise to new features in the quasiadiabatic following which are absent in the standard SVEA model; known results [3] are also recovered.

<sup>1)</sup> e-mail: ivm@gpi.ac.ru. Permanent address: General Physics Institute, Russian Academy of Sciences.

<sup>2)</sup> Department of Theoretical Physics, Institute of Atomic Physics, Bucharest, Romania.

Consider the response of semiconductor on a fs electromagnetic field. It is described by the total macroscopic polarization  $\mathcal{P}$  obtained by summing over all wavevectors  $\mathbf{k}$ ,  $\mathcal{P} = 2 \sum_{\mathbf{k}} d_{vc} P_{\mathbf{k}}$ ,  $d_{vc}$  is the interband dipole matrix element; in the summation, the factor of 2 counts for the spin degeneracy. The time dependence of  $P_{\mathbf{k}}$  is given in a Hartree-Fock limit by the semiconductor Bloch equations (SBEs) written in the notations of Ref. [4]:

$$i\hbar \partial_t P_{\mathbf{k}} = (\epsilon_{c,\mathbf{k}} - \epsilon_{v,\mathbf{k}} + \epsilon_g^0) P_{\mathbf{k}} + (n_{e,\mathbf{k}} + n_{h,\mathbf{k}} - 1) d_{cv} E - \sum_{\mathbf{q}} V_{|\mathbf{k}-\mathbf{q}|} P_{\mathbf{q}}, \quad (1)$$

$$i\hbar \partial_t n_{e,\mathbf{k}} = d_{vc} E P_{\mathbf{k}} - d_{cv} E P_{\mathbf{k}}^*, \quad (2)$$

here the subscript of  $t$  label corresponding derivative. The summation in the right-hand side of Eq.(1) is conventionally termed a Coulomb hole and is given by the departure of the screened Coulomb potential  $V_{\mathbf{k}}$  from its unscreened value. For simplicity the collision terms in Eqs.(1) and (2) are neglected; this places an upper limit of 60 fs on the pulsewidth  $\tau_p$  which can give rise to the quasiadiabatic following [5]. Since we are interested in low carrier densities, the contribution dominated by changes in the chemical potential (i.e., phase-space filling), conventionally called screened exchange, can be neglected, and the summations  $\sum_{\mathbf{q}} V_{\mathbf{k}-\mathbf{q}} P_{\mathbf{q}} n_{\mathbf{q}}$ ,  $\sum_{\mathbf{q}} V_{\mathbf{k}-\mathbf{q}} n_{\mathbf{q}} P_{\mathbf{q}}$ , and  $\sum_{\mathbf{q}} V_{\mathbf{k}-\mathbf{q}} P_{\mathbf{q}} P_{\mathbf{k}}^*$ , can be also omitted in Eqs.(1) and (2).

Following the steps used to calculate the macroscopic polarization  $\mathcal{P}$  we subsequently apply the Fourier transformation to Eqs. (1) and (2), and scale the transformed polarization  $P_{\lambda}$  and excitation density  $n_{\lambda}$  with the Wannier function  $\psi_{\lambda}$  defined in the lattice site. On this way, one can formally write out  $\tilde{P}_{\lambda}$  as

$$\tilde{P}_{\lambda}(z, t) = id_{cv} \hbar^{-1} \int_{-\infty}^t [1 - 2\tilde{n}_{\lambda}(z, \tau)] E(z, \tau) \exp[-i\omega_{\lambda}(t - \tau)] d\tau, \quad (3)$$

where  $\lambda$  labels the discrete exciton energy states. In turn, the macroscopic polarization  $\mathcal{P}$  may be written as  $\mathcal{P}(t) = 2d_{cv} \sum_{\lambda} |\psi_{\lambda}(R=0)|^2 \tilde{P}_{\lambda} + \text{c.c.}$

These equations show that if one knows a functional relation  $\tilde{P}_{\lambda} = \tilde{P}_{\lambda}(\tilde{n}_{\lambda}, E)$  among the induced polarization, excitation density, and the pulse field, then  $\tilde{n}_{\lambda}(E)$  may be determined. Furthermore, if both  $\tilde{n}_{\lambda}(E)$  and  $\tilde{P}_{\lambda}(\tilde{n}_{\lambda}, E)$  are known, then one can derive, at least formally by resolving the SBEs, the functional relation  $\mathcal{P} = \mathcal{P}(E)$ . This turns the wave equation into a nonlinear partial differential equation (PDE) for  $E(z, t)$  alone. Pursuing such a program substitute the integral (3) by

$$\int_{-\infty}^t \dots d\tau \rightarrow \frac{1}{i\omega_{\lambda}} \sum_0^m \frac{(-1)^k}{(i\omega_{\lambda})^k} \frac{\partial^k}{\partial t^k} [(1 - 2\tilde{n}_{\lambda})E], \quad (4)$$

which presumes that the dependence  $n(E)$  is a power series in  $E$  and  $\partial_t E$ . In principle, the series (4) generates an infinite hierarchy of coupled equations. Thus the best one can hope to do is to truncate this expansion, i.e. to find an expansion parameter which makes such a truncation meaningful. In general, this means long pulses, i.e.  $\omega_{\lambda} \tau_p \gg 1$ , which is essentially the approach first introduced by Crisp whose expansion parameter was  $s = 1/(\omega_g - \omega_p) \tau_p \ll 1$  [6].

From Eqs. (3) and (4) one can derive the relation

$$\mathcal{P}(z, t) = \frac{4\epsilon_{exc}r_0}{\pi\epsilon_\lambda} \left( E - \frac{\hbar^2}{\epsilon_\lambda^2} \frac{\partial^2 E}{\partial t^2} - \frac{1}{2} E \frac{E^2}{E_0^2} \right), \quad (5)$$

where  $r_0 = [1 - 2\tilde{n}_\lambda(t = -\infty)]$ ,  $E_0 = \epsilon_\lambda/2d_{cv}$ , and  $\epsilon_{exc}$  is the exciton binding energy. Note that we dropped all terms of the order higher than two in the expansion (4), and that both amplifying and absorbing semiconductors are described by Eq. (5). It is also of particular importance for the analysis below that the nonlinearity and dispersion contribute into the polarization (5) with the same sign. In the plane-wave approximation, its substitution into a classical wave equation leads to

$$\partial_{zz} E = c^{-2} \partial_{tt} [1 + 16r_0(\epsilon_{exc}/\epsilon_\lambda) (1 - (\hbar/\epsilon_\lambda)^2 \partial_{tt}) - 8r_0(\epsilon_{exc}/\epsilon_\lambda)(E/E_0)^2] E. \quad (6)$$

This can be further simplified under the assumption that  $E_t + cE_z \approx E_t$ , which physically means that we consider the fs pulse propagating in the positive direction of the  $z$ -axis, and stipulates that the backward-scattered wave is accounted on the spatial scale longer than the pulse length. Under these circumstances, Eq. (6) becomes

$$E_z + v_g^{-1} E_t + c_1 E^2 E_t + c_2 E_{ttt} = 0, \quad (7)$$

where the following set of parameters is used

$$\begin{aligned} v_g &= c[1 + 4\pi r_0 \chi_I(0)]^{-1/2}, \quad c_1 = \frac{6\pi v_g}{c^2} r_0 \chi_{nl}(0), \\ c_2 &= r_0 \frac{\pi v_g}{c^2} \left[ \frac{\partial^2 \chi_I}{\partial \omega^2} \right]_{\omega=0}, \quad \text{and} \quad \chi_{nl}(0) = \frac{1}{4\pi} \frac{\hbar \omega_{LT}}{\epsilon_\lambda} \frac{1}{E_0^2}, \end{aligned}$$

where  $\chi_I(\omega)$  is the linear susceptibility of excitons,  $\omega_{LT} = 8\pi s d_{cv}^2 \hbar^{-1}$ , and  $s$  is the Sommerfeld factor;  $\chi_{nl}(0)$  relates to the traditional cubic susceptibility of the semiconductor as follows,

$$\chi_{nl}(0) = \frac{4}{3} \chi^{(3)}(3\omega, \omega, \omega, \omega) |_{\omega=0} = 4\chi^{(3)}(\omega, \omega, \omega, -\omega) |_{\omega=0}.$$

Equation (7) is the modified Korteweg-de Vries equation (mKdV) which belongs to the class of PDEs integrable by the inverse scattering transform [7]. The general solutions of Eq. (7) are governed by the relative sign between the nonlinear and dispersion terms, the asymptotic values of the field (the boundary condition) and by the pulsewidth. Calling  $E(z = \pm\infty) = E_\infty$ , the general single soliton solution takes the form

$$E(z, t) = E_\infty \left[ 1 - \frac{4e^{-\beta}(1 - E_d/E_0)}{(1 - E_d/E_0 - 2\delta^2 e^{-\beta})^2 + 2\delta^2 e^{-2\beta}} \right]. \quad (8)$$

Here

$$\beta = (t - z/v)/\tau_p + \beta_0, \quad E_d^2 = 2E_\infty^2 + E_0^2, \quad \delta = E_\infty/E_0,$$

$$v^{-1} = v_g^{-1} [1 + 2\pi r_0 \chi_{nl}(0) (3E_\infty^2 + E_0^2)], \quad \text{and} \quad \tau_p = \sqrt{\frac{[\partial^2 \chi_I / \partial \omega^2]_{\omega=0}}{2E_0^2 \chi_{nl}(0)}},$$

where  $E_0$  labels the maximum amplitude of the bright soliton, its displacement from  $E_\infty$  is given by  $E_d$  which, in turn, is to be found for a given set of boundary conditions. It turns out that the solutions  $E(z, t)$  lie in the range  $E_\infty \leq E \leq E_\infty + 4\delta E_0^2$ . They describe either a bright soliton superimposed on a continuous wave background, i.e. a unbound soliton, or a hyperbolic-secant solitary

pulse; the behavior depends on the value of  $E_\infty$ . The transition between bound and unbound solitons is at  $E_\infty > 0$ , which may occur when the semiconductor is biased by a dc electric field. Exactly at this condition, the unbound soliton (8) is excited and will propagate through the medium of excitons.

In the limiting case  $E_\infty = 0$ , one can expect that the general solution (8) converges into the hyperbolic-secant form

$$E(z, t) = \frac{1}{\tau_p} \sqrt{\frac{[\partial^2 \chi_l / \partial \omega^2]_{\omega=0}}{\chi_{nl}(0)}} \operatorname{sech} \left( \frac{t - z/v}{\tau_p} \right), \quad (9)$$

which is determined exclusively by the given pulsewidth  $\tau_p$ ; its velocity is

$$v^{-1} = v_g^{-1} \left( 1 + \frac{\pi r_0}{\tau_p^2} \left[ \frac{\partial^2 \chi_l}{\partial \omega^2} \right]_{\omega=0} \right), \quad (10)$$

This leads to the expected result that the soliton (9) has a lower velocity than that of a low-frequency electromagnetic wave in the inverted medium ( $r_0 = 1$ ), and a greater velocity than that of a low-frequency electromagnetic wave in the absorbing one ( $r_0 = -1$ ).

In the general case of the non-zero boundary condition, the unbound soliton (8) occurs, and its behavior is a deal more complicated. In Fig.1 we plot the intensity of the single bright solitons with non-zero-boundary conditions for a variety of different ratios  $\delta$ . There is a typical spreading out of hump amplitudes for values of  $\delta \geq 1$ . Notice also the appearance of asymmetry for  $\delta > 0.5$ . This is due to the line-broadening by the dc-field  $E_\infty$  which shifts the dispersion contour, hence makes the whole pulse profile asymmetric.

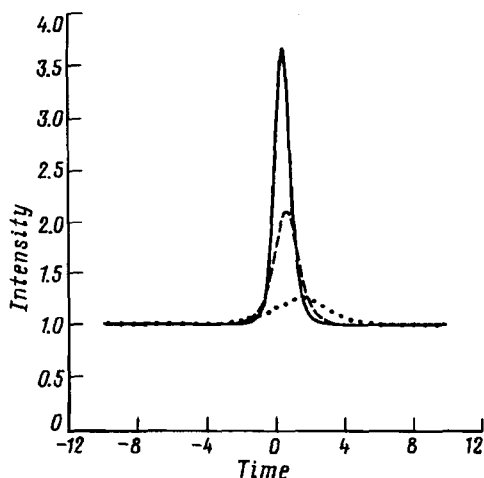


Fig.1. Normalized intensity versus normalized time for single bright solitons with non-zero boundary conditions;  $\delta = 0.75$  - solid, 1.0 - dashes, and 2.0 - dots. Note the asymmetry of the soliton acquired with the growth of  $\delta$

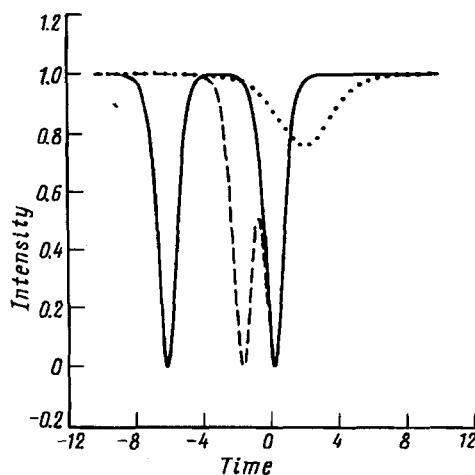


Fig.2. Dark-grey to dark-black soliton bifurcation;  $\delta = \sqrt{2}/2$  - solid, 0.72 - dashes, and 2 - dots. Note the asymmetry as in Fig.1

Let us return to Eq.(5). As we mentioned, the relative contributions to the polarization of excitons from nonlinearity and dispersion effects are of the same sign and this dictates our choice of the solution with non-zero asymptotics in the

form of (8) and rules out the following dark soliton solution

$$E(z, t) = E_{\infty} \left[ 1 - \frac{4e^{-\beta}}{(1 + 2\delta^2 e^{-\beta})^2 - 2\delta^2 e^{-2\beta}} \right], \quad (11)$$

where

$$\beta = (t - z/v)/\tau_p + \beta_0, \quad \text{and} \quad v^{-1} = v_g^{-1} [1 + 2\pi r_0 \chi_{nl}(0) (3E_{\infty}^2 - E_0^2)].$$

This solution is depicted in Fig.2 and describes the bifurcation of the dark-grey soliton state into the coupled state of two dark-black solitons of equal width, with  $\delta \rightarrow \sqrt{2}/2$  as a point of bifurcation. Although such topology of the fs field should be regarded as an illustrative one, it is worth to notice that this refutes the misconception of Hayata and Koshiba [3] that the prerequisite of its existence is the presence of a quadratic nonlinearity in the system.

It would be of interest to verify our results experimentally for, say, a GaAs/AlGaAs guiding structure in which Harten et al. observed the escape of a sub-picosecond pulse from quasiadiabatic following [8]. This was identified with carrier density oscillations in the semiconductor. It is anticipated in our study that the effects of phase-space filling and exciton screening may be quasiadiabatically ruled out, thus line broadening must be removed. On the other hand, the regime of quasiadiabatic following requires that the pulse contains several optical cycles, and hence sets a window for the pulsewidth used in experiments. In addition, one must have a structure as long as several soliton interaction lengths in order to ensure the soliton formation. The first step is thus to do shape measurements to see if the pulse reaches the steady-state shape corresponding to the given pulsewidth and the material parameters. If the soliton is observable, its shape can be changed by a seeding dc-field, and this provides a further test on the theoretical predictions. Therefore, a GaAs/AlGaAs guiding structure of a 1 cm length at room temperature may yield the predicted behavior upon a 10 GW/cm<sup>2</sup> excitation by a Ti:sapphire laser generating 20-60 fs pulses at  $\lambda = 850 - 940$  nm. Such an experiment may dramatically change the picture of quasiadiabatic following in semiconductors obtained so far within the SVEA.

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1. D.H.Auston and M.C.Nuss, IEEE J. Quantum Electron. **24**, 255 (1988); D.You, R.R.Jones, P.H.Bucksbaum, and D.R.Dykaar, Opt. Lett. **18**, 290 (1993); R.R.Jones, D.You, and P.H.Bucksbaum, Phys. Rev. Lett. **70**, 1236 (1993); C.O.Reinhold, M.Melles, H.Shao, and J.Burgdörfer, J.Phys. B **26**, L659 (1993); M.R.X. de Barros, R.S.Miranda, T.M.Jedju, and P.C.Becker, Opt. Lett. **20**, 480 (1995); A.Bonvalet, M.Joffre, J.L.Martin, and A.Migus, Appl. Phys. Lett. **67**, 2907 (1995).
  2. J.E.Rothenberg, Opt. Lett. **17**, 1340 (1992).
  3. K.Hayata and M.Koshiba, J. Opt. Soc. Am. B **11**, 2581 (1994).
  4. W.W.Chow, S.W.Koch, and M.Sargent III, *Semiconductor-Laser Physics*, Springer-Verlag, Berlin, 1994.
  5. R.Binder, D.Scott, A.Paul et al., Phys. Rev. B **45**, 1107 (1992).
  6. M.D.Crisp, Phys. Rev. A **1**, 1604 (1970).
  7. See e.g. G.L. Lamb, Jr., *Elements of Soliton Theory*, Wiley, New York, 1980, and references therein.
  8. P.A.Harten, A. Knorr, J.P. Sokoloff, et al., Phys. Rev. Lett. **69**, 852 (1992).