Nonlinear Nanofocusing in Tapered Plasmonic Waveguides

Arthur R. Davoyan,[1] Ilya V. Shadrivov,[1] Alexander A. Zharov,[2] Dmitri K. Gramotnev,[3] and Yuri S. Kivshar[1]

[1]*Nonlinear Physics Center, Research School of Physics and Engineering, Australian National University, Canberra ACT 0200, Australia*

[2]*Institute for Physics of Microstructures, Nizhny Novgorod 603950, Russia*

[3]*Nanophotonics, GPO Box 786, Brisbane, Queensland 4035, Australia*

(Received 10 June 2010; revised manuscript received 29 July 2010; published 9 September 2010)

We suggest using tapered waveguides for compensating losses of surface plasmon-polaritons in order to enhance nonlinear effects at the nanoscale. We study nonlinear plasmon self-focusing in tapered metal-dielectric-metal slot waveguides and demonstrate that, by an appropriate choice of the taper angle, we can effectively suppress the mode attenuation achieving stable propagation of *a spatial plasmon soliton*. For larger tapering angles we observe plasmon-beam *nanofocusing* in both spatial dimensions.

DOI: 10.1103/PhysRevLett.105.116804 PACS numbers: 73.20.Mf, 42.65.Jx, 42.65.Wi

Recent advances in the study of light localization in metal-dielectric structures demonstrate a strong potential of surface plasmon polaritons for nanoscale light manipulation, with further perspectives for subwavelength all-optical devices (see, e.g., Refs. [1–3]). Light coupling to the electron plasma in metals leads to a tight confinement of the electromagnetic energy that can be employed in many applications including medicine [4], biological and chemical sensing [5], and nanoscale lasers [6]. Among various methods of light focusing in plasmonic structures, the tapering of plasmonic waveguides was suggested as one of the most efficient and promising approaches [7], which allows creating light spots much smaller than the operating wavelength.

Although focusing of plasmons with tapered nanorods and metal film waveguides was suggested theoretically more than a decade ago [8,9], it was shown later that in lossless tapered metallic nanorods, surface plasmons slow down towards a thinner part of the rod and stop asymptotically near the tip [7]. Such behavior of surface plasmons is associated with a dramatic growth of the field amplitude towards the taper tip. More detailed studies of the plasmon nanofocusing in two-dimensional tapered waveguides [10,11] suggest that even in lossy systems the field enhancement is still possible. In Ref. [12], explicit analytical results were obtained for the plasmon nanofocusing in V-shaped grooves with linearly decreasing groove angles. The theoretical results were confirmed by several experiments [13–17] where the extreme focusing of the electromagnetic energy was observed.

The ability to dynamically tune and control the properties of surface plasmons is of a key importance for applications of plasmonic devices. Tuning plasmons by utilizing nonlinear properties of materials seems to be an attractive approach for controlling light propagation at the nanoscale [18]. Typically, to access nonlinear phenomena, it is required to use high power light beams. This is where tapered plasmonic waveguides have an edge over the traditional dielectric optics: due to extremely strong field nanofocusing and enhancement [7,10] nonlinear effects can be achieved at moderate power levels. It was already shown that it is possible to observe the nonlinear photoluminescence of erbium ions near the taper tip [19].

Spatial control of light can be achieved by nonlinear self-action that manifests itself in the formation of spatial optical solitons. Spatial optical solitons are generated when the effect of diffraction is compensated by a nonlinearity-induced change of the refractive index. In plasmonic systems, however, the presence of strong losses suppresses dramatically the nonlinear self-action [20].

In this Letter we suggest tapered plasmonic waveguides for mitigating the effect of losses in nonlinear processes. As an example, we study nonlinear light localization and self-focusing in tapered two-dimensional metal-dielectric-metal slot waveguides filled with the Kerr-type nonlinear dielectric. We show that the plasmon nanofocusing allows manipulating light trapping and guiding, thus controlling the strength of nonlinear effects. We demonstrate that in contrast to the light focusing in straight waveguides [20], an appropriate choice of the taper angle allows an effective compensation of attenuation with the formation of spatial plasmon-soliton. For larger tapering angles, we observe significant soliton narrowing leading to three-dimensional spatial light nanofocusing.

Figure 1 shows the geometry of our problem, where a tapered nonlinear dielectric waveguide is sandwiched between two metals. In the absence of tapering, such a slot waveguide is known to support symmetric and antisymmetric modes, classified with respect to the magnetic field profile [21–23]. An antisymmetric mode has a cutoff for smaller slots and, as a result, it is not suitable for light focusing with tapers. As an example, at the wavelength $\lambda = 1.55\ \mu$m the antisymmetric mode becomes evanescent with the slot below 100 nm. Thus, we focus our study on the symmetric mode, shown in the inset of Fig. 1.

In an adiabatically tapered waveguide, the plasmon mode is continuously transforming along the propagation direction. We assume small taper angles, $\partial h/\partial z \ll 1$,

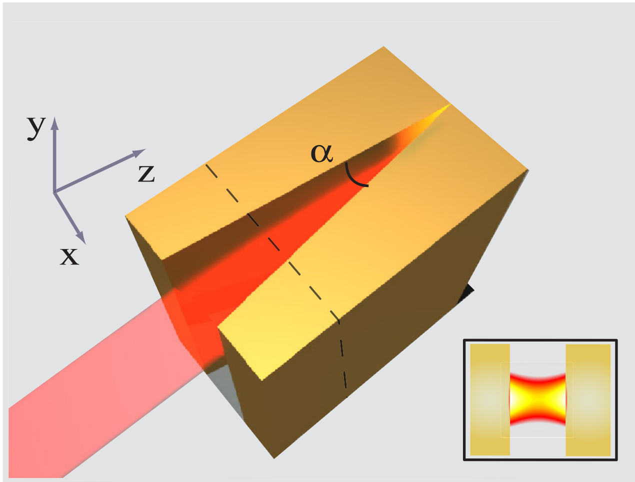

FIG. 1 (color online). Schematic of the three-dimensional plasmon focusing in a tapered slot waveguide. Focusing occurs in the horizontal plane, due to a taper, and in the vertical plane, due to the nonlinear self-focusing effect. Inset shows the waveguide cross section with the magnetic field distribution of a symmetric plasmon mode.

where h is the slot width, and a weak change of the mode dispersion on the scale of the plasmon wavelength, $|\partial\beta/\partial z| \ll \beta^2$. Here, we assume that the coordinates and plasmon wavelength are normalized to c/ω, and $\beta(h(z))$ is the plasmon guide index that depends on the slot width. Using this approximation, we present the electromagnetic field in the taper as a slowly varying eigenmode of a planar slot waveguide [10], and look for solutions in the form

$$\mathbf{E} = [A(y,z)\mathbf{E}_0(x,\beta) + \delta\mathbf{E}]\exp(i\int_0^z \beta d\xi) + \text{c.c.}, \quad (1)$$

where $A(y,z)$ is the slowly varying mode amplitude, $|\beta(\partial A/\partial z)| \gg |\partial^2 A/\partial z^2|$, $\mathbf{E_0}$ is the electric field of the symmetric eigenmode of the planar slot waveguide without losses, and $\delta\mathbf{E}$ is a small correction to the eigenmode profile. We note that in this approximation we neglect a change of the transverse mode structure, $\mathbf{E_0}(\mathbf{x},\beta)$, due to nonlinear effects. The waveguide mode is TM polarized, i.e., $\mathbf{E}_0 = (E_{x0}, 0, jE_{z0})$ and $\mathbf{H}_0 = (0, H_{y0}, 0)$, and it can be readily found from Maxwell's equations for the planar lossless slot waveguide [21,23]

$$\hat{L}_{\text{lin}}\mathbf{E}_0 \equiv \begin{pmatrix} -\beta^2 + \varepsilon' & -i\beta\frac{\partial}{\partial x} \\ -i\beta\frac{\partial}{\partial x} & \frac{\partial^2}{\partial x^2} + \varepsilon' \end{pmatrix}\begin{pmatrix} E_{x0} \\ jE_{z0} \end{pmatrix} = 0, \quad (2)$$

where $\varepsilon' \equiv \text{Re}[\varepsilon(x)]$ is the real part of the dielectric permittivity. Below in this Letter, we consider the metal to be silver, and we use the Drude-Lorentz model for describing analytically its dielectric permittivity [2]. Note that field components E_{x0} and E_z0 are real values.

Substituting the expression for the total field (1) into the wave equation, $\nabla\times\nabla\times\mathbf{E} + [\varepsilon(x) + \nu|\mathbf{E}|^2]\mathbf{E} = 0$, we obtain the equation for the field perturbation $\delta\mathbf{E}$ in the form, $(\hat{L}_{\text{lin}} + \delta\hat{L})\delta\mathbf{E} = \hat{L}\mathbf{E}_0$, where operator $\delta\hat{L}$ describes the action of nonlinearity and losses on the field correction $\delta\mathbf{E}$. The operator $\hat{L}$ is

$$\hat{L} \equiv \begin{pmatrix} \frac{\partial^2 A}{\partial y^2} + 2i\beta\frac{\partial A\cdot}{\partial z} + \eta A & -iA\frac{\partial^2\cdot}{\partial x\partial z} \\ iA\frac{\partial^2\cdot}{\partial z\partial x} & \frac{\partial^2 A}{\partial y^2} + \eta A \end{pmatrix}, \quad (3)$$

where $\eta(x) \equiv [i\frac{\partial\beta}{\partial z} + \varepsilon''(x) + \nu(x)|\mathbf{E}|^2]$. Here $\varepsilon''(x) \equiv \text{Im}[\varepsilon(x)]$ is the imaginary part of dielectric permittivity and $\nu(x)$ is the nonlinear coefficient, being nonzero only in the metal and dielectric layers, respectively.

We assume that, in accord with an asymptotic perturbation theory, the field $\delta\mathbf{E}$ and the operator $\delta\hat{L}$ are of the same order of magnitude, and higher-order terms can be neglected. Keeping the terms of the same order, we obtain the equation in the first-order approximation

$$\hat{L}_{\text{lin}}\delta\mathbf{E}_0 = \hat{L}\mathbf{E}_0. \quad (4)$$

According to the Fredholm theorem of alternative [24], the solutions $\delta\mathbf{E}_0$ of Eq. (4) are nondiverging only when the eigenmodes of the homogeneous equation $\hat{L}_{\text{lin}}\delta\mathbf{E}_0 = 0$ are orthogonal to the perturbation $\hat{L}\mathbf{E}_0$. The solution of the homogeneous equation is the plasmonic mode itself. Thus, we can write the orthogonality condition for the right-hand side of Eq. (4) with the plasmon eigenmode $\mathbf{E}_0$ and integrate it over the transverse coordinate x:

$$\int_{-\infty}^{+\infty} \mathbf{E}_0^*\hat{L}\mathbf{E}_0 dx = 0. \quad (5)$$

From this condition we obtain the required equation for the slowly varying plasmon amplitude $A(y,z)$,

$$2i\sigma\frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial y^2}I + iA\left(\frac{\partial\sigma}{\partial h}\frac{dh}{dz} + \Gamma\right) + N_{\text{nln}}|A|^2 A = 0, \quad (6)$$

where the $I = \langle E_{x0}^2 + E_{z0}^2\rangle_x$ is the effective beam intensity, $\sigma = \langle E_{x0}H_{y0}\rangle_x$ is proportional to the overall energy flow in the propagation direction per unit length, $\Gamma = \langle\varepsilon''(E_{x0}^2 + E_{z0}^2)\rangle_x$ is the effective dissipation in the system defined with the mode structure, and $N_{\text{nln}} = \langle\nu(E_{x0}^2 + E_{z0}^2)^2\rangle_x$ is the effective nonlinear coefficient, where $\langle\cdot\rangle_x$ corresponds to the integration over the transverse coordinate x.

Equation (6) derived above is the familiar nonlinear Schrödinger equation with the coefficients varying along the propagation direction. We remind here that the total magnetic field is a product of the mode structure function and the slow modal amplitude, $H = A(y,z)H_{y0}(x,z)e^{i\beta z} +$ c.c.. Similar equations were derived in the theory of pulse propagation in tapered optical fibers [25–27].

It follows from Eq. (6) that the effective losses or attenuation of plasmons is described by the term $(\frac{\partial\sigma}{\partial z} + \Gamma)$. The quantity σ decreases with the slot width. Hence, the derivative $\partial\sigma/\partial z$ is negative for plasmons propagating towards the tip of the taper. It is easy to show that the condition $\partial\sigma/\partial z + \Gamma = 0$ describes the taper, where the amplitude A does not decay due to losses in the system (however, the total energy flow still decreases). Our analysis shows that it is possible to find *an optimal tapering angle* for a straight taper so that the latter condition is fulfilled with good accuracy. Ideal compensation of the loss-induced attenuation is possible in a slightly curved taper. Moreover, when the taper angle is larger than the optimal value, a growth of the wave amplitude towards the taper tip is observed. In two-dimensional waveguides, this regime was identified as *focusing* of waves which are uniform in the third spatial dimension [10]. However, in the three-dimensional problem considered here, the decay of amplitude is additionally caused by spatial beam diffraction in the lateral direction, making plasmon nanofocusing impossible in the linear regime. However, as we show below, the effect of diffraction can be compensated by nonlinear self-focusing, usually associated with the physics of spatial optical solitons [28].

To study the evolution of plasmon waves in our waveguide, we solve Eq. (6) numerically using the beam propagation method. For our numerical simulations we consider dielectric as chalcogenite glass [29], sandwiched between two silver plates. The dielectric constant of chalcogenite glass is $\varepsilon_{\rm chal} = 4.84$, and the nonlinear coefficient is $\chi^{(3)} = 1.4 \times 10^{-19}\ {\rm m}^2/{\rm V}^2$. We assume that the frequency of light corresponds to the free-space wavelength of 1550 nm, and at the $z = 0$ the initial beam profile is taken to be of a fundamental solitonlike shape with the peak intensity $|\mathbf{E}|^2 \simeq 2 \times 10^6\ {\rm V}^2/\mu{\rm m}^2$, found for the flat slot waveguide [30]. The taper has the width of 600 nm at $z = 0$ and it is decreasing towards the tip.

Next we study the amplitude evolution in three different regimes corresponding to different taper angles. For small angles such as $\alpha = 0.8°$, when the tapering angle is below the optimal value, the losses are not compensated in the waveguide. Hence the amplitude decays, as shown by the curve (1) in Fig. 2(a). The analysis of the field distribution at the metal-dielectric interface [see Fig. 3(a)] suggests that the amplitude decay weakens the nonlinear self-action with plasmon propagation. Consequently, the beam broadening due to diffraction is observed, see the curve (1) in Fig. 2(b). The similar dynamics has been observed in planar nonlinear plasmonic waveguides [20,31].

When the tapering angle is close to the optimal value, $\alpha \approx 1.2°$, the effective losses are practically compensated, so that $\partial\sigma/\partial z + \Gamma \simeq 0$. Thus, the amplitude A remains almost unchanged in the presence of losses, as shown in Fig. 2(a), curve (2). Consequently, the diffraction is dynamically compensated by nonlinear self-action along the plasmon propagation in the taper. Thus, both the beam width and amplitude A remain unchanged, see the magnetic field distribution shown in Fig. 3(b). The similar effect has been observed in tapered optical fibers [25,26]. We notice that the taper with the optimal angle does not allow the complete effective compensation of losses, so that both the beam amplitude and its width still vary

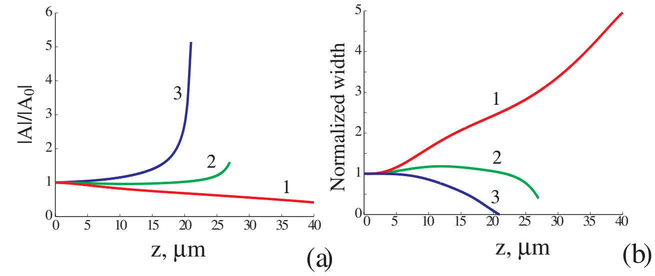

FIG. 2 (color online). Variation of (a) normalized amplitude at the beam center and (b) normalized beam width (along y axis) for three different values of the tapering angle α: (1) 0.8°, (2) 1.2°, and (3) 1.6°, respectively.

slightly with propagation. Close to the taper tip beam amplitude is increasing and the width is decreasing [curve 2 in Fig. 2], which is caused by the domination of nonlinear self-action over the diffraction, reduced due to plasmon wavelength decrease with propagation.

Finally, when the tapering angle is larger than the optimal value, we observe an effective growth of the mode amplitude associated with the nanofocusing effect [see curve 3 in Fig. 2(a)]. In the linear regime, nanofocusing was discussed for two-dimensional tapers in Ref. [10]. For the nonlinear taper discussed here, three-dimensional nanofocusing occurs when nonlinearity dominates over diffraction, and the corresponding mode experiences spatial self-focusing. In this case, the beam width narrows with propagation, and the energy becomes focused in the lateral dimension y as well, causing the amplitude A to increase even more than expected in the two-dimensional case. The magnetic field distribution at the metal-dielectric interface, shown in Fig. 4(a), manifests the spatial beam narrowing and nonlinear amplitude growth. The transfor-

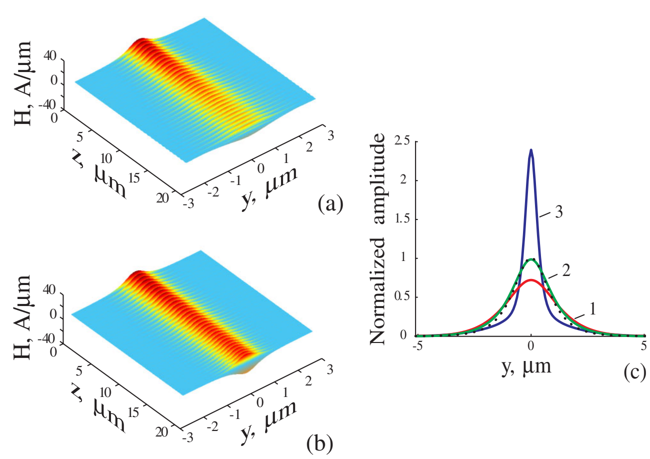

FIG. 3 (color online). Variation of the magnetic field in the nonlinear slot waveguide for two values of the tapering angle: (a) $\alpha = 0.8°$ and (b) $\alpha = 1.2°$. (c) Magnetic field profiles at the input and after 20 μm of propagation. Curves 1–3 correspond to the same tapering angles as discussed in Fig. 2. Dotted curve corresponds to the input soliton profile.

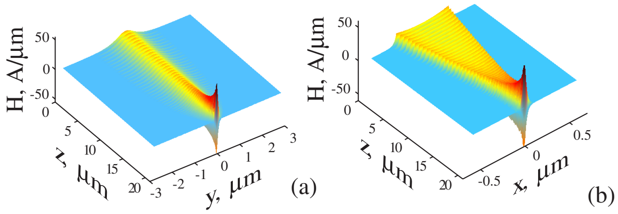

FIG. 4 (color online). (a,b) Variation of the magnetic field in the slot plasmonic waveguide for the tapering angle $\alpha = 1.6°$, shown for two cross sections. The mode tapering with the beam focusing in the lateral dimension y are clearly observed.

mation of the plasmon mode, presented in Fig. 4(b), reveals the effect of the plasmonic superfocusing. Therefore, two effects—energy focusing due to tapering and spatial beam narrowing with nonlinear amplitude concentration—are observed simultaneously. The energy concentration is very high near the tip, thus the cubic approximation for the nonlinear response may become not valid, and the nonlinearity saturation should be taken into account.

Finally, we have studied the effect of the amplitude profile variation for all three tapering angles. As follows from Fig. 3(c), after the propagation for 20 μm the amplitude profile remains unchanged in the case of the optimal taper, whereas both spatial beam narrowing and nonlinear focusing are observed for larger tapering angles.

We also notice that in all three cases studied above, the plasmon wavelength decreases with propagation. Despite this, close to the tip (e.g. within $\sim$1 μm), a variation of the plasmonic eigenmode may become so rapid that the slowly varying approximation used above may be inapplicable.

In conclusion, we have demonstrated that in using tapered waveguides, one can effectively compensate for the effect of losses, thus enhancing the nonlinear interactions. We have studied nonlinear propagation of surface plasmon polaritons in tapered slot waveguides with an intensity-dependent dielectric core. We have revealed three regimes of the wave propagation and focusing defined by different tapering angles. For the tapering angles below the optimal value, we have observed that the amplitude of the guided mode decays and the mode broadens. For the taper angles near the optimal value, the beam diffraction is compensated by nonlinear focusing, and we observe the solitonlike propagation. Finally, for the tapers with the angles larger than the optimal value, we have observed the regime of nonlinear nanofocusing, comprising of a growth of a plasmon-mode amplitude due to simultaneous action of nonlinear self-action and plasmonic nanofocusing due to a taper. We expect that our approach can be used for enhancing various nonlinear effects in plasmonics, including plasmon parametric amplification, second harmonic generation, and switching.

The authors acknowledge the support of the Australian Research Council and the Russian Fund for Basic Research (Grant No. 08-02-00379), and thank S. I. Bozhevolnyi and A. E. Miroshnichenko for useful comments.

[1] *Surface Plasmon Nanophotonics*, edited by M. L. Brongersma and P. G. Kik (Springer-Verlag, New York, 2007).
[2] S. A. Maier, *Plasmonics: Fundamentals and Applications* (Springer-Verlag, Berlin, 2007).
[3] *Plasmonic Nanoguides and Circuits*, edited by S. I. Bozhevolnyi (Pan Stanford, Singapore, 2009).
[4] A. M. Gobin *et al.*, Nano Lett. **7**, 1929 (2007).
[5] S. Vedantam, H. Lee, J. Tang, J. Conway, M. Staffaroni, and E. Yablonovitch, Nano Lett. **9**, 3447 (2009).
[6] D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003).
[7] M. I. Stockman, Phys. Rev. Lett. **93**, 137404 (2004).
[8] Kh. V. Nerkararyan, Phys. Lett. A **237**, 103 (1997).
[9] A. J. Babadjanyan, N. L. Margaryan, and Kh. V. Nerkararyan, J. Appl. Phys. **87**, 3785 (2000).
[10] D. K. Gramotnev, J. Appl. Phys. **98**, 104302 (2005).
[11] D. K. Gramotnev, D. F. P. Pile, M. W. Vogel, and X. Zhang, Phys. Rev. B **75**, 035431 (2007).
[12] S. I. Bozhevolnyi and K. V. Nerkararyan, Opt. Lett. **35**, 541 (2010).
[13] F. Keilmann, J. Microsc. **194**, 567 (1999).
[14] H. G. Frey, S. Witt, K. Felderer, and R. Guckenberger, Phys. Rev. Lett. **93**, 200801 (2004).
[15] C. Ropers, C. C. Neacsu, T. Elsaesser, M. Albrecht, M. B. Raschke, and C. Lienau, Nano Lett. **7**, 2784 (2007).
[16] E. Verhagen, A. Polman, and L. K. Kuipers, Opt. Express **16**, 45 (2008).
[17] V. S. Volkov *et al.*, Nano Lett. **9**, 1278 (2009).
[18] M. Durach, A. Rusina, M. I. Stockman, and K. Nelson, Nano Lett. **7**, 3145 (2007).
[19] E. Verhagen, L. Kuipers, and A. Polman, Nano Lett. **7**, 334 (2007).
[20] A. R. Davoyan, I. V. Shadrivov, and Yu. S. Kivshar, Opt. Express **17**, 21732 (2009).
[21] B. Prade, J. Y. Vinet, and A. Mysyrowicz, Phys. Rev. B **44**, 13556 (1991).
[22] A. R. Davoyan, I. V. Shadrivov, and Yu. S. Kivshar, Opt. Express **16**, 21209 (2008).
[23] A. R. Davoyan, I. V. Shadrivov, S. I. Bozhevolnyi, and Yu. S. Kivshar, J. Nanophoton. **4**, 043509 (2010).
[24] G. A. Korn and T. M. Korn, *Matematical Handbook for Scientists and Engineers* (McGraw-Hill, London, 1961).
[25] K. Tajima, Opt. Lett. **12**, 54 (1987).
[26] H. H. Juehel, J. Opt. Soc. Am. B **5**, 709 (1988).
[27] E. Ryder and D. F. Parker, IMA J. Appl. Math. **49**, 293 (1992).
[28] Yu. S. Kivshar and G. P. Agrawal, *Optical Solitons: From Fibers to Photonic Crystals* (Elsevier, New York, 2003).
[29] M. Asobe, T. Kanamori, and K. Kubodera, IEEE J. Quantum Electron. **29**, 2325 (1993).
[30] E. Feigenbaum and M. Orenstein, Opt. Lett. **32**, 674 (2007).
[31] A. Marini and D. V. Skryabin, Phys. Rev. A **81**, 033850 (2010).