Turning Light into a Liquid via Atomic Coherence

Humberto Michinel and María J. Paz-Alonso

Área de Óptica, Facultade de Ciencias de Ourense, Universidade de Vigo, As Lagoas s/n, Ourense, 32004 Spain

Víctor M. Pérez-García

Departamento de Matemáticas, E.T.S.I. Industriales, Universidad de Castilla-La Mancha, Ciudad Real, 13071 Spain

We study a four-level atomic system with electromagnetically induced transparency with giant $\chi^{(3)}$ and $\chi^{(5)}$ susceptibilities of opposite signs. This system will allow us to obtain multidimensional solitons and light condensates with surface tension properties analogous to those of usual liquids.

PACS numbers: 42.65.Tg, 42.50.Gy

The applications of nonlinear optical media mostly rely on the adequate dependence of their refractive indices on the amplitude of light fields. It is well known that the figure of merit of a suitable material for practical devices includes a fast and strong response to the light field as well as low losses [1], which has motivated an active search for optical materials with the appropriate properties [2].

On the other hand, a significant breakthrough in quantum optics has been the realization of giant optical nonlinearities in gases by means of atomic coherence and interference [3]. A technique that has attracted much attention is electromagnetically induced transparency (EIT) [4–6], in which an opaque medium becomes transparent to a probe laser beam by the addition of an appropriate coupling laser beam. The adequate choice of an atomic level scheme and driving fields can yield to controllable nonlinearities with very interesting applications in the design of nonlinear optical devices. This has been the basis for many studies on the resonant enhancement of nonlinear optical phenomena via EIT [7–13]. However, only a few of these works have investigated the formation of transverse solitons [12] and mostly considered the role played by the giant Kerr nonlinearity.

In this Letter we study the optical properties of a system where atomic coherence can be used to control the dependence of the refractive index on the amplitude of the light field. Many novel nonlinear optical phenomena beyond the giant Kerr effect are described, the most interesting being the obtention of the so-called *liquid light condensates* [14], i.e., robust solitonic distributions of light with analogies to ordinary fluid droplets.

We consider the propagation of a weak probe light field of frequency ω_p in a medium composed of four-level atoms and a coupling light field of frequency ω_c (see, e.g., [12]). A scheme of our system is shown in the inset of Fig. 1. In this kind of system, a coupling field of frequency ω_c changes the level structure [15] and induces transparency for a probe beam of frequency ω_p. A second effect is the enhancement of the optical Kerr nonlinearity. γ_2, γ_3, γ_4 denote the decay rates of the atomic states and

Δ_{13}, Δ_{23}, Δ_{24} are light detunings. Direct electric-dipole transitions between the two ground states $|1\rangle$ and $|2\rangle$ are forbidden.

Paraxial propagation along z of a probe laser beam through an optical medium, is given by

$$2ik_p \frac{\partial E_p}{\partial z} + \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) E_p = -k_p^2 \chi E_p. \qquad (1)$$

$k_p = 2\pi/\lambda_p$ and E_p are the wave number and amplitude of the beam. The optical susceptibility χ in the rotating wave and adiabatic approximations for EIT in the presence of a coupling beam E_c take the form [12]

$$\chi(E_p, E_c) = -\frac{\eta|\mu|^2}{\epsilon_0 \hbar \Gamma_3} + \frac{\eta|\mu|^2}{\epsilon_0 \hbar \Gamma_3 A} \left(\frac{|\Omega_c|}{\Gamma_3} + \frac{|\Omega_c|^2 |\Omega_p|^2}{\Gamma_3^2 B} \right)$$
$$- \frac{\eta|\mu|^2 |\Omega_c|^2 |\Omega_p|^2}{\epsilon_0 \hbar |\Gamma_3|^2 \Gamma_4 |A|^2} \left| 1 + \frac{|\Omega_p|^2}{\Gamma_3 B} \right|^2, \qquad (2)$$
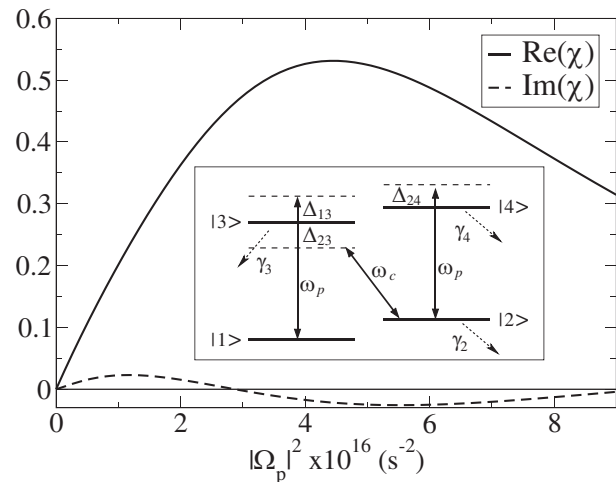

FIG. 1. Real (solid curve) and imaginary (dashed curve) parts of the susceptibility χ given by Eq. (2) for $\gamma = 30$ MHz, $\gamma_2 = 10^{-8}\gamma$, $\gamma_3 = \gamma_4 = 0.8\gamma$, $\Delta_{13} = \Delta_{23} = 1.5\gamma$, $\Delta_{24} = -5\gamma$, $\Omega_c = \gamma/2$, as a function of $|\Omega_p|^2 = |\mu|^2 |E_p|^2/4\hbar^2$. Inset: Schematic plot of the energy levels and optical couplings of the four-level atomic system.

where $|\Omega_{p,c}|^2 = |\mu|^2|E_{p,c}|^2/4\hbar^2$ are squared Rabi frequencies and $\Gamma_2 = \Delta_{23} - \Delta_{13} - i\gamma_2$, $\Gamma_3 = \Delta_{13} + i\gamma_3$, $\Gamma_4 = \Delta_{24} + \Delta_{13} - \Delta_{23} + i\gamma_4$, and $A = B + |\Omega_c|^2|\Omega_p|^2/(\Gamma_3^2 B)$, with $B = \Gamma_2 + |\Omega_p|^2/\Gamma_4 + |\Omega_c|^2/\Gamma_3$, being γ_2, γ_3, and γ_4 the decay rates of the atomic states and Δ_{13}, Δ_{23}, and Δ_{24} the light detunings. For our numerical examples to be presented later we choose an atomic density $\eta = 10^{14}$ cm^{-3}, an electric-dipole moment $\mu = 3 \times 10^{-29}$ Cm (for alkali atoms such as Rb or Ce, assuming for simplicity $\mu_{13} = \mu_{23} = \mu_{24} = \mu$), $\gamma_2 = 10^{-8}\gamma$, $\gamma_3 = \gamma_4 = 0.8\gamma$ (with $\gamma = 30$ MHz), $\Delta_{13} = \Delta_{23} = 1.5\gamma$, $\Delta_{24} = -5\gamma$, $\Omega_c = \gamma/2$, and $\lambda_p = 800$ nm.

From Eq. (1) we get the coefficients $\chi^{(j)}$ of the Taylor expansion of the susceptibility $\chi = \sum_{j=0}^{\infty} \chi^{(2j+1)}|E_p|^{2j}$:

$$\chi^{(1)} = -\eta|\mu|^2\Gamma_2/(\epsilon_0\hbar C), \tag{3a}$$

$$\chi^{(3)} = \frac{\eta|\mu|^4|\Omega_c|^2}{4\epsilon_0\hbar^3}\left[\frac{\frac{1}{\Gamma_3} - \frac{1}{\Gamma_4}}{C^2} - \frac{|\Omega_c|^2}{\Gamma_3 C^3} - \frac{1}{\Gamma_4|C|^2}\right], \tag{3b}$$

$$\chi^{(5)} = \frac{\eta|\mu|^6|\Omega_c|^2}{16\epsilon_0\hbar^5\Gamma_4}\left[\frac{D}{C^3} + \frac{|\Omega_c|^2(3 - \Gamma_4/\Gamma_3)}{C^4} + \frac{|\Omega_c|^4\Gamma_4}{C^5\Gamma_3}\right.$$
$$\left. + \frac{D + \frac{|\Omega_c|^2}{C}}{C|C|^2} + \frac{D^* + \frac{|\Omega_c|^2}{C^*}}{C^*|C|^2}\right], \tag{3c}$$

$C = \Gamma_2\Gamma_3 + |\Omega_c|^2$, $D = \Gamma_3/\Gamma_4 - 1$.

In Fig. 1 we show the real and imaginary parts of the susceptibility χ as a function of the squared Rabi frequency of the probe light $|\Omega_p|^2 = |\mu|^2|E_p|^2/4\hbar^2$, for our parameter choice. As it can be appreciated from Re$[\chi(\Omega_p)]$ (see Fig. 1), the real part of the susceptibility of the medium grows linearly with $|E_p|^2$ for low powers (due to the effect of a positive $\chi_R^{(3)}$) and decreases for high powers (due to a negative $\chi_R^{(5)}$), while the losses are comparatively small in this range. Thus, we have a balance of diffraction plus self-focusing for low field amplitudes and self-defocusing for larger amplitudes. This type of competition is also found in media with the so-called nonlinearity of cubic-quintic type, i.e., those with a refractive index of the form $n = n_0 + n_2|E|^2 + n_4|E|^4$. These nonlinearities have attracted a lot of theoretical attention recently [14,16–20] because of their predicted ability, when $n_4 < 0$, to prevent collapse of laser beams for sufficiently large powers, thus yielding to different stable two-dimensional light distributions [16]. The robustness of these *light bullets* has been recently connected with the formation of a liquid light condensate with surface tension properties similar to those of usual liquids [14,21]. It has been shown recently [21] that the surface tension of the light condensates grows linearly with the radius, as in the case of usual liquid droplets. These media are able to support stable vortex beams [17–19,22] and display interesting nonlinear phenomena [20].

For our choice of parameters, using Eqs. (3) and the relation $n(E_p) \simeq n_0 + (\chi_R^{(3)}/2n_0)|E_p|^2 + \frac{1}{2n_0}[\chi_R^{(5)} -$

$(\chi_R^{(3)}/2n_0)^2]|E_p|^4 + \cdots$, we obtain $n_2^R = 2.2713 \times 10^{-7}$ m^2/V^2, $n_4^R = -3.4390 \times 10^{-13}$ m^4/V^4, which are, respectively, $\sim 10^{13}$ and $\sim 10^{22}$ larger than those measured for usual nonlinear optical materials [23]. These facts provide some analogies between our system and cubic-quintic (CQ) media. However, the contribution of higher order and dissipative terms will be relevant for us.

First we will construct stationary transverse self-trapped solutions of Eq. (1) of the form: $E_p(r, z) = \psi_l(r)e^{i\beta z}e^{il\theta}$, where β is the propagation constant. For $l \neq 0$ the beam host a vortex of topological charge l. To this end we set $\chi_I = 0$ and solve numerically the problem

$$\left[\frac{d^2}{dr^2} + \frac{1}{r}\frac{d}{dr} - \frac{l^2}{r^2} + k_p^2\chi_R(\psi_\ell) - 2k_p\beta\right]\psi_l = 0, \tag{4}$$

with boundary conditions $\psi_l'(0) = 0$ and $\psi_l(\infty) = 0$.

This gives us stationary beam shapes corresponding to different powers as a function of β. Let us first consider beams with $l = 0$. In Fig. 2 we show the results for $\beta = 0.2$, 1.4, and 1.7 μm^{-1}. Low values of β yield to light distributions with quasi-Gaussian profiles. As β is incremented, the spatial shapes become narrower, but still keep a Gaussian shape. For larger values of β, the beam flux grows rapidly and the peak intensity of the light distribution saturates due to the effect of a negative n_4, yielding to light distributions with almost super-Gaussian profiles. Because of the giant nonlinear response, these powers can be experimentally achieved by using mW continuous lasers provided the sources are highly stabilized in frequency (typically 1 MHz bandwidth). A warning is in order: for our parameter combination, the probe beam has a power smaller but close to that of the coupling beam, thus a fully quantitative treatment should consider a vector extension of Eq. (1) including the effect of the probe beam on the coupling beam. Since this extension makes the analysis even more complex, in this Letter we restrict ourselves to the scalar model and the full vector model will be the subject of future research.
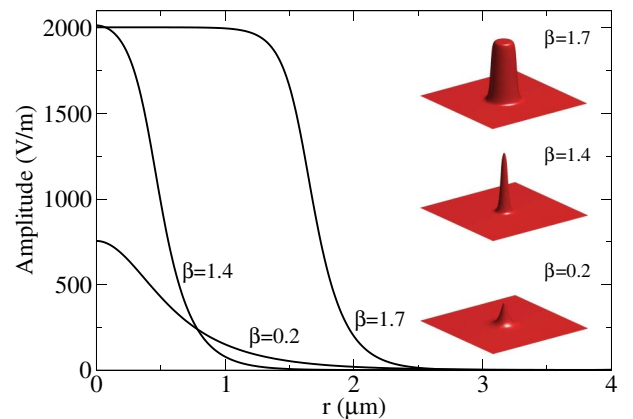

FIG. 2 (color online). Solutions of Eq. (4) for $\beta = 0.2$, 1.4, and 1.7 μm^{-1} for our parameter set (values given in the text).
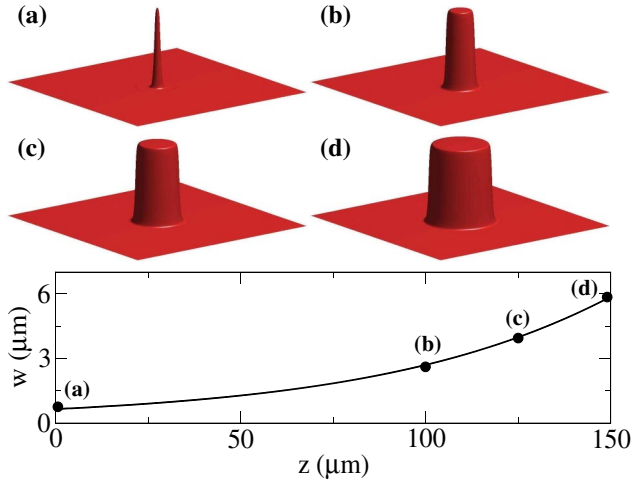
FIG. 3 (color online). Propagation of an eigenstate of Eq. (4) with $\beta = 1.5$ μm^{-1} in as medium with the full complex susceptibility given by Eq. (2). (a)–(d) 3D plots of $|E_p(x, y, z)|^2$ for $z = 0, 100, 125,$ and 150 μm. (e) Beam width $w(z)$.

We have used the eigenstates of the nondissipative case as input conditions for propagation in a medium with the full complex susceptibility of Eq. (2) (i.e., including the imaginary part of χ). We observe that for $\beta = 1.1$ μm^{-1} the eigenstate keeps its shape while propagating in such a medium. In this situation the amount of energy pumped into the soliton and taken out by the linear and nonlinear gain and dissipation, respectively, achieves an equilibrium [24]. Eigenstates with $\beta < 1.1$ μm^{-1} tend to spread during propagation since they do not achieve the critical power for the formation of a soliton, while those with $\beta > 1.1$ μm^{-1} keep their peak amplitude and increase their width during propagation as it can be seen in Fig. 3. In this situation the energy available in the medium is responsible for the broadening of the liquid light droplet in a similar way to the process of growth of a fluid droplet in a super-
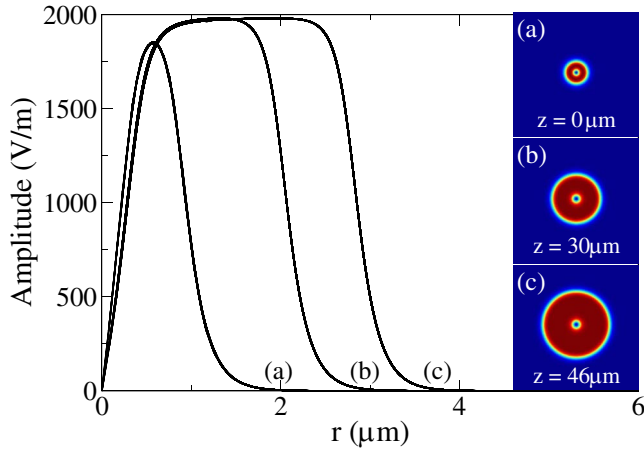

FIG. 4 (color online). Evolution of an eigenstate with $l = 1$ and $\beta = 1.5$ μm^{-1} for several propagation distances. The gain enlarges the radial size of the beam preserving the vortex.
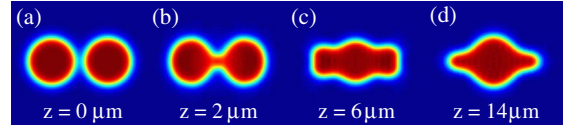

FIG. 5 (color online). Coalescence of two eigenstates with $\beta = 1.65$ μm^{-1} launched in parallel with the same phases.

saturated atmosphere. This means that although small, nonconservative effects play an important role in the propagation of wave packets for this set of parameters. We have found numerically with a very high accuracy that the radius of the light droplet varies as $R(z) \sim z^3$, which is faster than typical fluid droplet growth phenomena [25] or models similar to CQ ones such as the Ginzburg-Landau equations [26]. In both cases the growth takes the form z^q with $q < 1$. Even pure diffractive propagation leads an exponent $q = 1$, which is smaller than the one observed in our system.

Next, we have constructed eigenstates of Eq. (4) with $l = 1$. Eigenstates with $\beta \leq 1.07$ μm^{-1} tend to spread during propagation. However, for $\beta > 1.07$ μm^{-1} the beams reach a critical value of the energy [17] so that the liquid light condensate is formed and the surface tension is able to sustain the vortex within the beam. Thus, the effect of gain is to enlarge the radial size of the beam without destroying the vortex, as can be seen in Fig. 4. As the beam propagates the width of the beam surrounding the vortex increases while keeping the peak density constant, which again resembles the growth of an incompressible fluid. Note that the radial profiles from Fig. 4 are very similar to those found in Ref. [22] in the study of the stability of multiply charged two-dimensional vortices in cubic-quintic media. It is remarkable that the maximum densities in Figs. 2 and 4 are very similar.

To study the robustness of these solitons we have made a series of numerical experiments with collisions of different beams (we show results for $l = 0$). First we have launched initially parallel beams in phase corresponding to eigenstates with $\beta = 1.65$ μm^{-1}. Their mutual effective interaction, as it happens with solitons of the 1D nonlinear Schrödinger equation, is attractive and leads to their fusion and subsequent transverse oscillations of the new bound state as it can be seen in Fig. 5. In a different series of numerical experiments we have studied the collisions of the same beams launched initially with opposite phases as
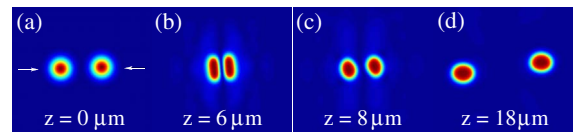

FIG. 6 (color online). Collision of two hyper-Gaussian beams launched with opposite phases, amplitudes $A = 2000$ V/m and width $w = 4$ μm, separated by 3 μm and one of them slightly displaced in the y axis. The initial angle is 0.25 rad.
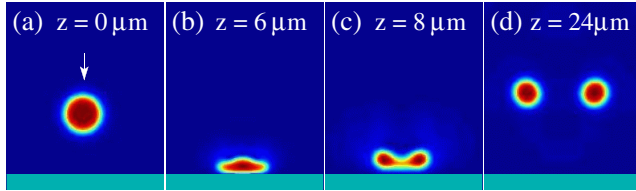
FIG. 7 (color online). Total reflection at a nonlinear-linear interface of an eigenstate with $\beta = 1.6$ μm^{-1} and an incidence angle of 0.01 rad, at $z = 0, 6, 8,$ and 24 μm.

shown in Fig. 6. As it can be seen, both beams survive the collision behaving as light droplets, although slight transverse oscillations of the beams are observed after the collision by excitation of surface modes. Both phenomena arise in fluid droplets collisions [27].

Finally, we have launched an eigenstate with $\beta = 1.6$ μm^{-1} with an incidence angle of 0.01 rad against the frontier between a nonlinear and a linear material. The results are shown in Fig. 7, where the breakup of the beam into smaller droplets is observed analogously to splashing fluid droplets [28].

In conclusion, we have shown that the adequate choice of the parameters of a specific EIT scheme leads to a giant response for both n_2 and n_4 (but different signs) and could allow us to obtain stable two-dimensional liquid light condensates with surface tension properties similar to those of usual liquids. Our theoretical and computational results could be the basis for real experiments in nonlinear optics with continuous mW lasers showing this phase transition and nice liquidlike properties of light.

[1] S. R. Marder *et al.*, Science **276**, 1233 (1997).
[2] V. Degiorgio and C. Flytzanis, *Nonlinear Optical Materials: Principles and Applications* (IOS, Amsterdam, 1995).
[3] J. E. Field, K. H. Hahn, and S. E. Harris, Phys. Rev. Lett. **67**, 3062 (1991); A. Kasapi *et al.*, Phys. Rev. Lett. **74**, 2447 (1995); H. Schmidt and A. Imamoglu, Opt. Lett. **21**, 1936 (1996); A. S. Zibrov *et al.*, Phys. Rev. A **65**, 043817 (2002).
[4] S. E. Harris, Phys. Today **36**, No. 3, 36 (1997).
[5] U. Rathe *et al.*, Phys. Rev. A **47**, 4994 (1993).
[6] C. Szymanowski and C. H. Keitel, J. Phys. B **27**, 5795 (1994).
[7] A. Andre *et al.*, Phys. Rev. Lett. **94**, 063902 (2005).
[8] H. Schmidt and A. Imamoglu, Opt. Lett. **21**, 1936 (1996); A. Imamoglu, H. Schmidt, G. Woods, and M. Deutsch, Phys. Rev. Lett. **79**, 1467 (1997).
[9] S. E. Harris and Y. Yamamoto, Phys. Rev. Lett. **81**, 3611 (1998); S. E. Harris and L. V. Hau, Phys. Rev. Lett. **82**, 4611 (1999).
[10] D. Petrosyan and G. Kurizki, Phys. Rev. A **65**, 033833 (2002).
[11] M. Masalas and M. Fleischhauer, Phys. Rev. A **69**, 061801(R) (2004).
[12] T. Hong, Phys. Rev. Lett. **90**, 183901 (2003).
[13] R. S. Bennink, R. W. Boyd, C. R. Stroud, and V. Wong, Phys. Rev. A **63**, 033804 (2001); X.-J. Liu, H. Jing, and M.-L. Ge, Phys. Rev. A **70**, 055802 (2004).
[14] H. Michinel *et al.*, Phys. Rev. E **65**, 066604 (2002).
[15] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).
[16] A. H. Piekara, J. S. Moore, and M. S. Feld, Phys. Rev. A **9**, 1403 (1974); C. Josserand and S. Rica, Phys. Rev. Lett. **78**, 1215 (1997); K. Dimitrievski *et al.*, Phys. Lett. A **248**, 369 (1998); M. Quiroga-Teixeiro, A. Berntson, and H. Michinel, J. Opt. Soc. Am. B **16**, 1697 (1999); A. Desyatnikov, A. Maimistov, and B. Malomed, Phys. Rev. E **61**, 3107 (2000).
[17] H. Michinel and M. Quiroga-Teixeiro, J. Opt. Soc. Am. B **14**, 2004 (1997).
[18] B. A. Malomed, L. C. Crasovan, and D. Mihalache, Physica D (Amsterdam) **161**, 187 (2002).
[19] M. J. Paz-Alonso *et al.*, Phys. Rev. E **69**, 056601 (2004); H. Michinel, J. R. Salgueiro, and M. J. Paz-Alonso, Phys. Rev. E **70**, 066605 (2004).
[20] D. Mihalache *et al.*, Phys. Rev. Lett. **88**, 073902 (2002); D. Mihalache *et al.*, Phys. Rev. E **68**, 046612 (2003).
[21] M. J. Paz-Alonso and H. Michinel, Phys. Rev. Lett. **94**, 093901 (2005).
[22] T. A. Davydova and A. I. Yakimenko, J. Opt. A Pure Appl. Opt. **6**, S197 (2004).
[23] S. Saltiel, S. Tanev, and A. D. Boardman, Opt. Lett. **22**, 148 (1997).
[24] J. M. Soto-Crespo, N. Akhmediev, and A. Ankiewicz, Phys. Rev. Lett. **85**, 2937 (2000); K. Staliunas, Phys. Rev. Lett. **91**, 053901 (2003); E. A. Ultanir *et al.*, Phys. Rev. Lett. **90**, 253903 (2003); N. N. Rozanov *et al.*, Opt. Spectrosc. (USSR) **97**, 88 (2004); N. N. Rozanov, Opt. Spectrosc. (USSR) **96**, 569 (2004).
[25] J. S. Langer and A. J. Schwartz, Phys. Rev. A **21**, 948 (1980).
[26] D. Gomila *et al.*, Phys. Rev. Lett. **87**, 194101 (2001).
[27] M. Orme, Prog. Energy Combust. Sci. **23**, 65 (1997).
[28] See, e.g., R. L. Vander Wal *et al.*, in *Proceedings of the Fifth Microgravity Fluid Physics and Transport Phenomena Conference, Cleveland* (2000), CP-2000-210470, p. 1395, available at www.ncmr.org/research/examples/splashing drop.pdf.