Light Propagation in Field-Ionizing Media: Extreme Nonlinear Optics

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A novel equation for light propagation in underdense field-ionizing media is derived by extending concepts of nonlinear optics into the strong-field domain. The equation is first order in the propagation coordinate and is valid for arbitrarily short pulse durations. Solutions of the first-order wave equation are found to be in excellent agreement with solutions of the scalar wave equation. Furthermore, the polarization response of a field-ionizing medium is derived from semiclassical considerations. The polarization reveals, in agreement with experiments, a previously unrecognized contribution that is shown to significantly affect the absorption loss in the presence of ionization.

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Field ionization is the primary process in matter exposed to high-intensity ($I > 10^{14}$ W/cm^2) radiation triggering all strong-field processes. Whereas ionization plays a central role in strong-field atomic processes, such as high harmonic generation (HHG) [1], it did not appear to be important for high-field plasma physics so far. This is because ionization saturates at the leading edge of the previously used subpicosecond or longer pulses, so that the dominant part of the pulse interacts with a fully ionized plasma. The availability of ultrashort, few-cycle laser pulses has changed the situation, opening a novel parameter regime of plasma physics. Saturation of ionization is shifted to considerably higher intensities and ionization continues playing a role until instants close to the pulse peak. In spite of the central role of ionization in strong-field physics, there exists no simple and complete model for the evolution of laser pulses in field-ionizing media.

An exact description of strong-field atomic and plasma phenomena requires solution of Maxwells equations in combination with an analysis of the microscopic processes in the medium. This formidable task often exceeds current computer capacities and makes the use of simplified models indispensable. In this Letter we derive a novel propagation equation for the strong-field regime that simplifies and improves existing models in the following two respects.

(i) Our analysis extends concepts used in the perturbative regime of nonlinear optics into the strong-field regime. Drawing on the slowly evolving wave (SEW) approximation [2], a first-order propagation (FOP) equation for strong-field atomic and plasma physics is derived that is valid for underdense media and for arbitrarily short pulse durations. Numerical results obtained from a solution of the FOP equation and of the scalar wave equation reveal excellent agreement. *The FOP equation can be solved by more than 2 orders of magnitude faster than the scalar wave equation,* greatly facilitating the analysis of a wide range of strong-field atomic and plasma phenomena, such as high harmonic generation [3], x-ray lasing [4], and processes in relativistic nonlinear optics [5–7].

(ii) The polarization response of ionizing media [8–10] used in previous macroscopic propagation studies is incomplete [11] and is not capable of describing fundamental processes, such as the absorption loss introduced by field ionization, correctly. Our semiclassical analysis reveals an additional loss term. A comparison to absorption measurements in helium yields excellent agreement and corroborates the validity and importance of the additional term that increases the absorption loss by a factor of 3 in our experiments.

The propagation of high-intensity pulses in matter is modeled by Maxwells equations in combination with a model for the polarization response of the medium P. As long as $P(\mathbf{r},t)$ in the coordinates ($r_\perp = x, y$) transversal to the propagation direction (z) exhibits little variation over a distance comparable to the center wavelength λ_0, Maxwells equations may be substituted by the scalar wave equation. In Gaussian units and for a linearly polarized electric field the scalar wave equation is given by

$$\partial_z^2 E + \nabla_\perp^2 E - \frac{1}{c^2}\,\partial_t^2 E = \frac{4\pi}{c^2}\,\partial_t^2 P\,, \qquad (1)$$

where c is the vacuum velocity of light, $\partial_{z,t}$ refers to the partial derivatives with respect to z and t, and $\nabla_\perp^2$ is the transversal Laplace operator. The derivation of the FOP equation is performed in the spectral domain by inserting the ansatz $E(\mathbf{r},\omega) = U(\mathbf{r},\omega)\exp(i\omega z/c)$ into the Fourier transformed Eq. (1), where $E(\mathbf{r},\omega) = \int_{-\infty}^{\infty} dt\, E(\mathbf{r},t)\exp(-i\omega t)$. This yields

$$\left(\frac{2i\omega}{c}\,\partial_z + \nabla_\perp^2\right)U(\omega) = -\frac{4\pi\omega^2}{c^2}\exp\left(-\frac{i\omega z}{c}\right)\hat{F}[P]\,, \qquad (2)$$

where the operator $\hat{F}$ denotes the Fourier transform and the term $\partial_z^2 U$ was neglected. Neglect of the second space derivative has been termed SEW approximation [2] and is equivalent to elimination of backward propagating wave solutions. The SEW approximation is applicable as long as changes to the electric field induced by the polarization of the medium over a distance comparable to λ_0 are small, i.e., $|\partial_z E| \ll (\omega/c)E$. Substituting U by E followed by a

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transformation of Eq. (2) back into the time domain yields

$$\partial_\xi E(r_\perp, \xi, \tau) - \frac{c}{2} \nabla_\perp^2 \int_{-\infty}^{\tau} d\tau' \, E(r_\perp, \xi, \tau') = -\frac{2\pi}{c} \partial_\tau P[E(r_\perp, \xi, \tau)]. \quad (3)$$

Here, we have introduced the moving coordinate frame $\xi = z$ and $\tau = t - z/c$ and have utilized the fact that the Fourier transform of $U(r_\perp, z, \omega)$ is identical to the electric field in the retarded coordinate system, $E(r_\perp, \xi, \tau)$. In contrast with the perturbative regime of nonlinear optics, where the FOP equation is derived for a complex envelope, the strong-field FOP equation (3) governs evolution of the electric field. In the strong-field regime of nonlinear optics, nonlinear effects dominate linear dispersion and a decomposition of the electric field into carrier wave and complex envelope is no longer beneficial. As our derivation contains no assumptions with respect to the pulse width, Eq. (3) is valid for arbitrarily short pulse durations. The FOP equation (3) is the first major result of this paper presenting a powerful tool for the analysis of complex propagation phenomena in strong-field atomic and plasma physics.

In the remainder of this paper the FOP equation is applied to the analysis of laser pulse evolution in the presence of field ionization. Ionization is of particular importance constituting the primary event for all strong-field phenomena. In order to model evolution of a laser pulse in a field-ionizing medium, the polarization response in (3) has to be determined. Our derivation of P is restricted to moderate gas densities and moderate laser intensities, at which collisions, collective plasma processes, and relativistic effects may be neglected. For inclusion of these phenomena, see, e.g., Refs. [5,11]. Under the above assumptions the medium polarization can be written as $P = n_0 e\langle x\rangle$, where n_0 is the atomic particle density, $e\langle x\rangle = e\langle\Psi|x|\Psi\rangle$ is the single-atom dipole moment, e is the electron charge, and x represents the microscopic coordinate of the electron along the polarization of the linearly polarized laser electric field. The electron wave function Ψ is obtained by solving the time-dependent Schrödinger equation in the frame of the dipole approximation and of the single-active-electron approximation.

Although solution of the FOP equation (3) in combination with the Schrödinger equation is possible with current computer capacities, it is still a demanding problem calling for a further simplification. To this end, we derive an approximate closed-form expression for $P(\mathbf{r}, t)$ in terms of $E(\mathbf{r}, t)$ and $n(\mathbf{r}, t)$, the density of electrons set free by field ionization. The derivation draws on a simple quasistatic model [8,12], which is valid as long as the Keldysh parameter $\gamma = \sqrt{I_p/(2U_p)} < 1$ [13,14]. Here, I_p is the ionization potential of the atom, $U_p = (eE)^2/4m\omega_0^2$ is the ponderomotive potential, m is the electron mass, and ω_0 is the center frequency of the laser pulse. The quasistatic model is depicted schematically in Fig. 1. In the quasistatic limit the electron tunnels through the suppressed Coulomb barrier and the tunneling rate adiabatically follows variations of the laser field. Coupling to other bound states is negligibly small [12,14]. In this picture, the electron is "born" on the outer side of the Coulomb barrier at a distance of $x_0(t) \approx I_p/eE(t)$ from the nucleus [15] and with zero initial velocity, $v_0 \approx 0$ [12].

The classical trajectory $x(t)$ of the freed electron in the nonrelativistic regime is obtained by solving Newton's equation of motion, $m\ddot{x}(t) = eE(t)$. By using the classical trajectory the current is determined by $J = \dot{P} = e\dot{n}x + en\dot{x}$. Here, the dot/double dot refers to the first/second partial time derivative. As $\dot{n} \neq 0$ only when an electron is born, $x(t)$ is substituted by $x_0(t)$ in the first term of the current, yielding $J = e\dot{n}x_0 + en\dot{x}$. The second time derivative of the polarization is obtained from the current by $\ddot{P} = \dot{J} = e\partial_t(\dot{n}x_0) + en\ddot{x} + e\dot{n}\dot{x}$. As the free electron density is changed only when an electron is born due to ionization and as the birth velocity $v_0 = 0$, the last term in $\ddot{P}$ vanishes. After expressing x_0 and $\ddot{x}$ in terms of E, and integrating $\ddot{P}$ with respect to t, we obtain the current

$$\partial_t P(\mathbf{r}, t) = I_p\left(\frac{\partial_t n(\mathbf{r}, t)}{E(\mathbf{r}, t)}\right) + \frac{e^2}{m} \int_{-\infty}^{t} dt' \, n(\mathbf{r}, t')E(\mathbf{r}, t'), \quad (4)$$

where

$$n(\mathbf{r}, t) = n_0\left(1 - \exp\left[-\int_{-\infty}^{t} dt' \, w\{E(\mathbf{r}, t')\}\right]\right), \quad (5)$$

and $w(E)$ is the quasistatic ionization rate. Equations (4) and (5) in combination with the closed-form expressions for $w(E)$ [16] given by the ADK (Ammosov-Delone-Krainov) theory or the tabulated values of $w(E)$ [14] provide an explicit constitutive law for field-ionizing media and represent the second major result of this paper.

In comparison to previous theoretical work [8–10], our derivation reveals an additional term proportional

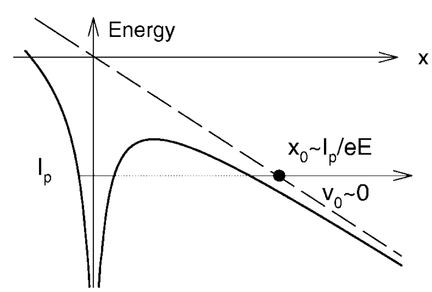

FIG. 1. Schematic of an atom in the presence of a laser pulse. The dashed and the full lines denote the electric field and the combined potential of the nucleus and of the electric field, respectively. The dot refers to the position of birth x_0.

to the ionization potential. Whereas the second term in (4) introduces predominantly phase effects, such as blueshifting and broadening of the spectrum, the first term is a pure loss term. To identify the physical effects underlying the two terms, the loss of pulse energy per volume in the presence of ionization $\Delta\rho_E = \int_{-\infty}^{\infty} dt\, EJ$ is determined. Inserting J from Eq. (4) in this expression yields

$$\Delta\rho_E = -\frac{e^2}{2mc^2}\int_{-\infty}^{\infty} A^2(t)\,[\partial_t n(t)]\,dt - n(t\to\infty)I_p\,. \tag{6}$$

Here, the first term on the right hand side has been obtained by performing partial integration twice, and the vector potential is defined by the relation $E = -(1/c)\dot{A}$ subject to the condition $A(t\to\pm\infty) = 0$. As the total energy of electrons and laser field has to be conserved, the energy gain of the electrons during ionization must be compensated by an energy loss of the laser field. During the interaction, the ionized electrons gain the potential energy I_p and a kinetic energy resulting from a drift velocity, the free electrons are left behind with after the laser field vanished. The drift velocity is given by $v_d = \lim_{t\to\infty}\dot{x}(t) = -\lim_{t\to\infty} e/(mc)[A(t) - A(t_0)] = e/(mc)A(t_0)$ [8], where t_0 is the instant of birth of the free electron and the velocity $\dot{x}$ is obtained from time integration of the Newton equation subject to the condition $\dot{x}(t_0) = v_0 = 0$. Indeed, we find that the energy gain of the ionized electrons corresponds to the energy loss of the laser pulse given by Eq. (6). The first term accounts for the energy loss due to the drift velocity and the second term on the right hand side introduces an energy loss I_p per ionized electron. The two terms are referred to as the drift loss and as the ionization loss from hereon. From Eq. (6) we find that the ratio of the ionization loss term and of the drift loss contribution scales with the square of the Keldysh parameter $\gamma^2 = I_p/2U_p$. Therefore, the influence of the ionization loss is strongest for short wavelength laser pulses and decreases with increasing field strength.

Substituting the constitutive law as given by (4) in (3) yields a novel propagation equation for the evolution of laser pulses in field-ionizing media,

$$\partial_\xi E(r_\perp,\xi,\tau) - \frac{c}{2}\nabla_\perp^2\int_{-\infty}^{\tau} d\tau'\, E(r_\perp,\xi,\tau') = -\frac{1}{2c}\int_{-\infty}^{\tau} d\tau'\,\omega_p^2(r_\perp,\xi,\tau')E(r_\perp,\xi,\tau') - \frac{2\pi I_p}{c}\,\frac{\partial_\tau n(r_\perp,\xi,\tau)}{E(r_\perp,\xi,\tau)}\,, \tag{7}$$

where $\omega_p^2(r_\perp,\xi,\tau) = 4\pi n(r_\perp,\xi,\tau)e^2/m$ is the plasma frequency squared [17]. Using the validity condition derived above we find that in the presence of ionization the FOP equation is valid for underdense plasmas, i.e., as long as $\omega_p \ll \omega_0$.

To corroborate the validity of this equation we have performed several tests. Figure 3 shows a numerical solution of the FOP equation (3), propagated by a fourth-order Runge-Kutta method, and of the scalar wave equation (1), solved by the commonly used leap-frog method, in one space dimension. The two solutions are nearly identical proving the applicability of the SEW approximation for underdense plasmas and the validity of the FOP equation. Small deviations come from a reflection at the vacuum-gas surface that cannot be accounted by the FOP equation. In the scalar wave equation, vacuum propagation of the laser pulse limits the maximum spatial step size to <0.005 μm. The major advantage of the FOP equation is that this limitation of the step size is eliminated by use of the moving coordinate frame. Consequently, for the integration of Eq. (3) a considerably larger step size (≈ 1 μm) can be used *speeding up solution of the FOP equation by more than 2 orders of magnitude.*

To demonstrate the validity of the ionization loss term revealed by our analysis we compared the experimentally measured energy loss in helium (500 Torr) with the loss obtained from a solution of the FOP equation (7) in two space dimensions assuming radial symmetry. The pulse parameters were $\lambda_0 = 800$ nm, $I_0 = 2\times10^{15}$ W/cm^2, and a FWHM pulse duration $\tau_p = 5$ fs; for a more detailed description of the laser system, see, e.g., Ref. [1]. The experimentally (full circles) and theoretically (full line) obtained energy loss is plotted versus gas interaction length in Fig. 2. The dashed line represents the numerical solution without the ionization loss term, which is the second term on the right hand side of (7). In order to eliminate possible errors originating from the limited validity of the ADK model we used the exact static

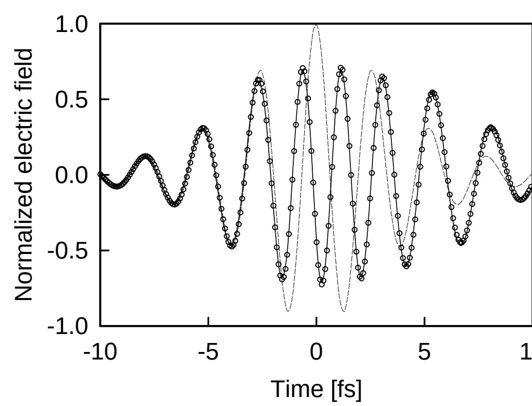

FIG. 2. Thin-dashed line: Normalized input electric field of a Ti:sapphire laser pulse. Empty circles and full line: Solution of the FOP equation (7) and of the scalar wave equation (1) with the polarization (4) in one space dimension, respectively. The pulse parameters are $I_0 = 2\times10^{15}$ W/cm^2, $\lambda_0 = 0.8$ μm, and full width at half maximum (FWHM) pulse duration $\tau_p =$ 5 fs; the remaining parameters are propagation through a 1-mm-thick He gas target at a pressure of 500 Torr corresponding to $n_0 = 1.75\times10^{19}$ cm^{-3}. At full ionization the electron density corresponds to a ratio $\omega_p/\omega_0 = 0.1$. The excellent agreement between the two solutions proves the validity of the FOP equation (7) for underdense plasmas.

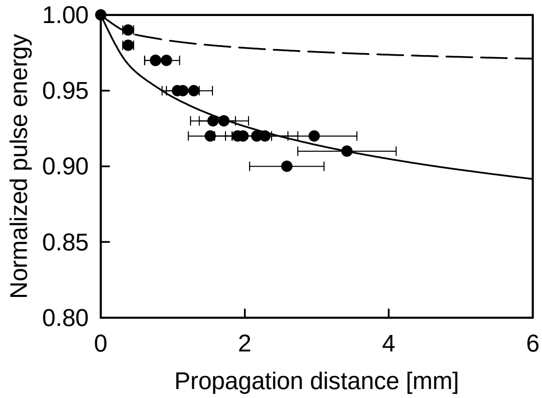

FIG. 3. Output pulse energy normalized to input pulse energy versus propagation distance for a 5 fs Ti:S laser pulse propagating in helium under conditions described in the caption of Fig. 3. The full circles denote the experimental values and the error bars refer to an estimated $\pm 20\%$ uncertainty in the measurement of the gas interaction length. The full/dashed lines denote the theoretical values calculated from a solution of the 2D wave equation (7) using cylindrical symmetry with/without the ionization loss term. Our analysis clearly reveals the necessity of the ionization loss term to account for the experimental data.

ionization rates w of helium in Eq. (5) that were derived by a full solution of the two-electron Schrödinger equation [14]. The agreement between our model's prediction and experimental data is excellent and clearly reveals the essential role of the ionization loss term in the constitutive law (4) for a proper description of the evolution of a laser pulse in a field-ionizing medium. *The ionization loss term increases the energy loss by a factor of 3 under our experimental conditions.* After 4–6 mm interaction length the pulse peak intensity has been reduced so much due to dispersion-induced pulse broadening that ionization stops, limiting the maximum loss to around 10%. We recall that the ratio of ionization loss to drift loss scales with $\gamma^2 \propto 1/\lambda_0^2$. Furthermore, the free electron induced group velocity dispersion decreases with λ_0^3, leading to longer interaction distances over which the pulse experiences loss. As a result, the ionization loss increases for shorter wavelengths. Our calculations show that a frequency doubled Ti:sapphire pulse ($\lambda_0 = 400$ nm, $\tau_p = 10$ fs, $I_0 = 3 \times 10^{15}$ W/cm^2) loses $\approx 45\%$ of its pulse energy over a propagation distance of 12 mm mainly due to the first term in (4) missing in previous theoretical models.

As more electrons are ionized at the peak of the pulse than in the pulse wings, the ionization loss leads to a stronger reduction of the pulse peak and therewith, to a lengthening of the pulse. As a result, the nonlinear interaction saturates faster and the blueshifting and spectral broadening of the pulse is reduced. The observed behavior has important implications on strong-field phenomena, such as a modification of the maximum interaction length for x-ray lasing and high harmonic generation, which will have to be analyzed in future investigations.

In conclusion, we have derived a first-order propagation equation for strong-field atomic and plasma physics which is valid for ionizing media below the critical density and for arbitrarily short pulse durations. The simplified wave equation is accurate and expedites computing by more than 2 orders of magnitude. In combination with a closed-form constitutive law for optical field ionization this first-order wave equation constitutes a powerful tool for predicting the *macroscopic* response of atomic media driven by strong laser fields.

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