Postionization medium evolution in a laser filament: A uniquely nonplasma response

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Theoretical consideration of the optical response of nascent free electrons in the process of laser filamentation reveals that the initial microscopically inhomogeneous charge distribution causes a transient electromagnetic response of the medium that differs drastically from that of a homogeneous plasma with the same degree of ionization. An analytical model, describing the forced oscillations of virtually isolated and expanding electron clouds, predicts considerable enhancement of these oscillations caused by transient resonance with the laser field. The transient resonance processes should play a role in the currently accepted picture of filament formation dynamics.

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I. INTRODUCTION

Laser filamentation in air or other gaseous media has been an area of intense research interest due both to the unique, coupled light-matter structure created in the filament and to the new technical capabilities that filaments enable [1,2]. The process is initiated through the intensity-dependent index of refraction that accompanies the passage of radiation through a medium. If the laser beam power exceeds the medium-specific critical value, the nonlinear refraction index inducing Kerr lensing overcomes diffraction and causes the laser beam to self-focus. The self-focusing is accompanied by broadening of the spectral distribution to the blue side of the pump frequency through the process of pulse shortening known as self-steepening. The processes arresting the ultimate collapse of the beam, leading to filament stabilization, are the subject of some controversy [3–7]. The conventional mechanism involves a negative contribution to the refractive index due to the presence of the freed electrons, pictured as an underdense plasma in the framework of Drude theory [1,2]. Recently, however, this notion has been challenged, on both experimental and theoretical grounds, by invoking a negative contribution to the refraction index by higher-order nonlinearities [3,8,9]. The issue of ionization, or the absence thereof, in laser filamentation remains a topic of some debate [4,5,10]. Moreover, the dynamics of laser-induced ionization in atmospheric-pressure gases has recently come to the fore in two other areas of active current research: laser wakefield acceleration [11–13] and indirect-drive inertial confinement ignition efforts [14–17]. In both of these experiments, the ionization is also a crucial step for unfolding multiple nonlinear processes, and in both cases it is not well understood, requiring *ad hoc* fitting parameters to properly describe the situation. In this paper, we challenge the assumption that the ionization process results immediately in locally homogeneous plasma, using laser filamentation in air as the primary example.

II. THE MODEL

In this section, we delineate and then present a model for a fundamental difference between the nascent state of ionization in the filament-bearing medium and a low-density plasma. The characteristic distance between atoms or molecules in a gas at atmospheric pressure is about $\sim$30 Å. At the same time, at the typical ion density in a filament [2,18], $n \sim 10^{15}-10^{16}$ cm^{-3}, the average distance between ions is $\sim$500$-$1000 Å. At the velocity of $\sim 10^{17}$ cm/s, the released electron covers only $\sim$20 Å during an $\sim$20 fs laser pulse. This means that during filamentary propagation, the ionization process does not produce a uniform plasma by the end of the laser pulse (at $t \approx 20$ fs). Rather, the isolated and well-separated ions are each surrounded by an expanding electron cloud. This cloud is initially formed by the wave packet emerging from the ionization process, and the spatial extent of the wave packet is estimated as $\hbar/\Delta p \sim \hbar/\sqrt{2m\Delta E} \sim 2.5$ Å, where ΔE is the uncertainty in the freed electron energy for an eight-photon ionization using a laser with 60 meV of bandwidth. [At the focused laser intensity of $\sim 10^{13}$ W/cm^2 [1,2], the electron kinetic-energy distribution is strongly peaked at low kinetic energies; as the ponderomotive potential is $<$1 eV (with 800 nm laser wavelength), the exponentially decreasing tail of high-kinetic-energy above-threshold-ionization electrons can be safely neglected.] The expansion and thinning of this initial electron cloud, that is, the spatial evolution of the wave packet in the presence of the ion-core potential, is a quantum process that is still largely unaccounted for, except for a few limiting cases [19]. However, the essence of the medium response effects considered here does not depend on the details of the expansion process, thus allowing for very general (and simplifying) assumptions.

Accordingly, we consider the initial electron wave packet of a general form having spherically symmetric and asymmetric components, which we will model as a classical electron cloud having the initial charge density $\rho(r,0) = (1-\mu)\rho_0(r,0) + \mu(3/2)\rho_1(r,0)\cos^2\theta$ where r is the distance to the ion core and θ is the polar angle in the corresponding spherical coordinate system, the parameter $\mu \leqslant 1$ quantifies the relative contribution of the asymmetric component, and the distribution functions $\rho_0(r,0)$ and $\rho_1(r,0)$ are normalized

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to the fundamental charge e. Note that $\rho_0(r,0)$ is nonzero at the center, while $\rho_1(r,0) \propto r^2$ (at least) when $r \to 0$. As a simple assumption, we model this expansion as a self-similar process, where the distribution functions $\rho_0(r,0)$ and $\rho_1(r,0)$ maintain their form while the characteristic scale grows with t at a constant rate α: $\rho_{0,1}(r,t) = C(t)\tilde{\rho}_{0,1}[r/(r_0+\alpha t)]$, thus causing the electron cloud to spread, decreasing the charge density $\rho_0(0,t)$ in the middle of the cloud and increasing the effective distance between the two lobes of the asymmetric distribution $\rho_1(r,0)$. In this expression, $\tilde{\rho}_{0,1}$ are function of a dimensionless argument, referring to the initial charge distributions: $\tilde{\rho}_{0,1}(r/r_0) = \rho_{0,1}(r,0)$. The time-dependent coefficient $C(t)$ is obtained from the normalization condition $\int dV \rho_{0,1}(r,t) = e$, which gives $C(t) = r_0^3/(r_0+\alpha t)^3$, so that $\rho_{0,1}(r,t) = [r_0^3/(r_0+\alpha t)^3]\tilde{\rho}_{0,1}[r/(r_0+\alpha t)]$.

Consider the system of the ion core and the electron charge distribution in the oscillating electric field of the laser pulse, $E(t) = E_0\cos(\omega t)$; neglecting the slow motion of the heavy ion, this electric field just shifts the electron cloud as a whole with respect to the ion charge. If the oscillating displacement distance z is much smaller than the characteristic scale of the charge distribution, $|z| \ll r_0 + \alpha t$, the restoring force $F(z,t)$ exerted on the shifted cloud by the ion charge can be obtained by expanding the slowly evolving potential energy $U(z,t) = e\int d^3r\,\rho(r,t)/\sqrt{r^2+z^2-2rz\cos\theta}$, to find

$$F(z,t) \approx -z\frac{4\pi}{3}\frac{eQr_0^3}{(r_0+\alpha t)^3},$$
$$Q = (1-\mu)\tilde{\rho}_0(0) + \frac{12}{5}\mu\int_0^\infty dr'\frac{\tilde{\rho}_1(r')}{r'}, \tag{1}$$

where the integral in the second term in Q converges due to the mentioned asymptotic properties of $\rho_1(r,0)$. One can note the physically distinct nature of the two terms in the expression for Q. The first term arises from the shift of spherically symmetric charge distribution and corresponds to effective attraction of the charged sphere of radius z as follows from the Gauss theorem. The second term corresponds to the shift of the positive charge from the middle position between two equal lobes of the negative charge distribution. The equation of motion for the newly released electron can be recast as an equation for the effective oscillating dipole $d = ez$:

$$\frac{\partial^2}{\partial\tau^2}\tilde{d}(\tau) = -\frac{\gamma}{(1+\beta\tau)^3}\tilde{d}(\tau) + \cos(\tau+\tau_0), \tag{2}$$

with the initial conditions $\tilde{d}(0) = 0, (\partial\tilde{d}/\partial\tau)|_{\tau=0} = 0$ and where τ_0 signifies the ionization moment. In Eq. (2), we use the ponderomotive radius $a = eE_0/(m_e\omega^2)$ as the length scale, so that $d = ea\tilde{d}$, and we choose $\tau = \omega t$ to effect a dimensionless form. The contribution of the first term in the right-hand side is determined by two dimensionless coefficients $\beta = \alpha/(\omega r_0) \ll 1$ and $\gamma = (4\pi/3)(Qr_0/E_0)(a/r_0) \geqslant 1$. For example, if we take $\alpha \sim 5\times10^6$ cm/s and $r_0 \sim 3$ Å, then $\beta \sim 10^{-2}$. Then, setting $\mu \approx 1/2$ in Eq. (1), we obtain $\gamma \sim 3$. As seen in Eq. (2), the physical meaning of γ is the initial squared frequency of the free dipole oscillations of the nascent electron cloud as related to the laser carrier frequency, while β signifies the relatively slow rate at which this frequency decreases. It is noteworthy that in a realistic situation, $\gamma > 1$, that is, the initial free-oscillation frequency is greater than the carrier frequency.

As the electron cloud expands, the value of γ inevitably passes through unity [at $\tau = (\gamma^{1/3}-1)/\beta \gg 1$], and this point of transient resonance is a major factor determining the dynamics of the medium response to the laser pulse.

III. ANALYSIS OF FORCED OSCILLATIONS

Equation (2) is formally an equation for a forced harmonic oscillator with slowly varying natural frequency $\Omega(\beta\tau) = \gamma^{1/2}/(1+\beta\tau)^{3/2}$. Assuming $\beta \ll 1$, we apply the two-scale approach that has been developed for such cases [20]. We consider $\tilde{d}(\tau)$ as a function of two temporal variables, slow ς and fast ξ: $\tilde{d}(\tau) = \tilde{d}(\xi,\varsigma)$. Specifically, we choose

$$\varsigma = \beta\tau;$$
$$\xi = \frac{1}{\beta}\int_0^{\beta\tau} dx\,\Omega(x) = \frac{2\gamma\tau}{(1+\beta\tau)^{1/2}[(1+\beta\tau)^{1/2}+1]}. \tag{3}$$

Then, Eq. (2) becomes

$$\frac{\partial^2\tilde{d}}{\partial\xi^2} + \frac{2\beta}{\Omega(\varsigma)}\frac{\partial^2\tilde{d}}{\partial\xi\partial\varsigma} + \frac{\beta^2}{\Omega^2(\varsigma)}\frac{\partial^2\tilde{d}}{\partial\varsigma^2} + \frac{\beta}{\Omega^2(\varsigma)}\frac{\partial\Omega(\varsigma)}{\partial\varsigma}\frac{\partial\tilde{d}}{\partial\xi}$$
$$= -\tilde{d} + \frac{1}{\Omega^2(\varsigma)}\cos[\tau(\xi)+\tau_0]. \tag{4}$$

We look for the solution in the form $\tilde{d}(\xi,\varsigma) = \tilde{d}_0(\xi,\varsigma) + \beta\tilde{d}_1(\xi,\varsigma) + \beta^2\tilde{d}_2(\xi,\varsigma) + \cdots$, collect the terms of the same power of β, and impose cancellation of resonance terms in higher-order contributions that would destroy the hierarchy. Thus, we obtain in the main approximation:

$$\tilde{d}_0(\xi,\varsigma) = \frac{a_0}{\sqrt{\Omega(\varsigma)}}\cos(\xi+\theta_0) + \frac{1}{\Omega^2(\varsigma)-1}\cos[\tau(\xi)+\tau_0]. \tag{5}$$

Here, the constants a_0 and θ_0 are determined by the initial conditions to Eq. (2) as

$$a_0 = -\frac{\sqrt{\Omega(0)}}{\Omega^2(0)-1}\cos(\tau_0)\sqrt{1+\frac{\tan^2(\tau_0)}{\Omega^2(0)}},$$
$$\tan(\theta_0) = \frac{\tan(\tau_0)}{\Omega(0)}. \tag{6}$$

The most interesting feature of the solution is that as the "slow time" ς progresses, the denominator in the second term becomes zero at the point $\varsigma = \varsigma_R = \gamma^{1/3} - 1$. Physically, this means that the natural oscillation frequency of the expanding electron cloud becomes resonant with the laser field oscillations. The two-scale approximation (5) does not apply in the vicinity of the resonance and as a result, the constants a_0' and θ_0' in Eq. (5) after the resonance will differ from a_0 and θ_0 determined in Eq. (6).

In the vicinity of the resonance, we use an approach [21] where the reference point to ς_R is shifted and the slow variable is scaled as $\tilde{\varsigma} = \gamma^{-1/3}(\varsigma - \varsigma_R) = \varepsilon\tilde{\tau}$, where $\tilde{\tau} = \tau - \varsigma_R/\beta$, $\tilde{\tau}_0 = \tau_0 + \varsigma_R/\beta$, and $\varepsilon = \gamma^{-1/3}\beta \ll 1$, so that and $\Omega(\tilde{\varsigma}) = (1+\tilde{\varsigma})^{-3/2}$. Then the solution to Eq. (4) is written as $\tilde{d}(\tilde{\tau}) = a(\varepsilon\tilde{\tau})\cos[\tilde{\tau} + \varphi(\varepsilon\tilde{\tau})] = \mathrm{Re}[z(\varepsilon\tilde{\tau})\exp(i\tilde{\tau})]$, where $z(\varepsilon\tilde{\tau}) = a(\varepsilon\tilde{\tau})\exp[i\varphi(\varepsilon\tilde{\tau})]$. Upon substituting this $\tilde{d}(\tilde{\tau})$

in Eq. (4), the approximate expression for $z(\tilde{\varsigma})$ is obtained as

$$z(\tilde{\varsigma}) = e^{ig(\tilde{\varsigma})}\left(-\frac{i}{2\varepsilon}e^{i\tilde{\tau}_0}\int_0^{\tilde{\varsigma}} d\tilde{\varsigma}' e^{-ig(\tilde{\varsigma}')} + z_0\right),$$
$$g(\tilde{\varsigma}) = \frac{1}{2\varepsilon}\int_0^{\tilde{\varsigma}} \tilde{\varsigma}[\Omega^2(\tilde{\varsigma}') - 1]. \quad (7)$$

Here, the constant z_0 sets the amplitude of the oscillations at the point of the transient resonance, $\tilde{\varsigma} = 0$, as it is eventually determined by the initial conditions. Thus, to find the value of z_0, we need to match $\tilde{d}(\tilde{\tau})$ with that given by the expression of Eq. (5) at some sufficient distance from the resonance, where $\tilde{\varsigma}$ is still small while the values of $g(\tilde{\varsigma})$ in Eq. (7) are already large enough to allow asymptotic expansions. In this range, for negative values of $\tilde{\varsigma}$ (that is, in the region before the resonance),

$$\tilde{d}(\tilde{\tau}) = \mathrm{Re}\left[e^{i(\tilde{\tau}+g(\varepsilon\tilde{\tau}))}\left(z_0 + \frac{i}{2\varepsilon}A_- e^{i\tilde{\tau}_0}\right)\right] + \frac{\cos(\tilde{\tau}+\tilde{\tau}_0)}{\Omega^2(\tilde{\varsigma}) - 1}, \quad (8)$$

where $A_\pm = \pm\int_0^{\pm\infty} d\varsigma e^{-ig(\tilde{\varsigma})} \approx (\varepsilon\pi/6)^{1/2}(1+i)$. By comparing Eqs. (8) and (5), we obtain $z_0 \approx a_0 \exp[i(\xi_R + \theta_0)] + \exp(i\tilde{\tau}_0)(1/2)(\pi/6\varepsilon)^{1/2}(1-i)$, where, according to Eq. (3), $\xi_R = 2\gamma^{5/6}(\gamma^{1/6} - 1)/\beta$. Performing an expansion similar to that of Eq. (8) in the region of positive $\tilde{\varsigma}$, we relate z_0 to the constants a_0' and θ_0', and then obtain the relation between (a_0', θ_0') and (a_0, θ_0):

$$\begin{aligned} a_0' \exp(i\theta') &= a_0 \exp(i\theta_0) - (i/2\varepsilon)\exp[i(\tilde{\tau}_0 - \xi_R)](A_- + A_+) \\ &\approx a_0 \exp(i\theta_0) + \exp[i(\tilde{\tau}_0 - \xi_R)](\pi/6\varepsilon)^{1/2}(1-i). \end{aligned}$$

IV. RESULTS AND DISCUSSION

The considerations outlined in the previous section sum up to the following analytic expressions for the oscillating dipole. For the values of τ outside the resonance zone,

$$\begin{aligned} \tilde{d}(\tau) = &-\frac{(1+\beta\tau)^{3/4}}{\gamma - 1}\left(\cos(\tau_0)\cos[\xi(\tau)] - \frac{\sin(\tau_0)}{\gamma^{1/2}}\sin[\xi(\tau)]\right) \\ &+ \frac{(1+\beta\tau)^{3/4}}{\gamma^{1/12}}\Theta(\tau - \tau_R)\left(\frac{\pi}{3\beta}\right)^{1/2} \\ &\times \cos\left(\xi(\tau) + \tau_0 + \tau_R - \xi_R - \frac{\pi}{4}\right) \\ &+ \frac{(1+\beta\tau)^3}{\gamma - (1+\beta\tau)^3}\cos(\tau + \tau_0), \end{aligned} \quad (9)$$

where $\xi(\tau)$ is given by the expression (3). In the immediate vicinity of the resonance,

$$\begin{aligned} \tilde{d}(\tau) = &\left(\frac{\pi}{6\varepsilon}\right)^{1/2}\cos[f(\tau - \tau_R) + \tau_0 + \tau_R]\left[S\left((\tau - \tau_R)\sqrt{\frac{3\varepsilon}{2\pi}}\right) + \frac{1}{2}\right] + \sin[f(\tau - \tau_R) + \tau_0 + \tau_R]\left[C\left((\tau - \tau_R)\sqrt{\frac{3\varepsilon}{2\pi}}\right) + \frac{1}{2}\right] \\ &- \frac{(1+\gamma^{1/3}\varepsilon\tau)^{3/4}}{\gamma - 1}\left(\cos(\tau_0)\cos[f(\tau - \tau_R) + \xi_R] - \frac{1}{\sqrt{\gamma}}\sin(\tau_0)\sin[f(\tau - \tau_R) + \xi_R]\right), \end{aligned} \quad (10)$$

where $f(x) = x - 3\varepsilon x^2/4$, while $S(x)$ and $C(x)$ are the Fresnel integrals. A typical evolution of the oscillating dipole is shown in Fig. 1. Note that the dipole oscillation undergoes a significant nonlinear enhancement as it passes through the region of the inevitable transient resonance with the laser carrier frequency. Moreover, in contrast to the typical situation with mechanical systems, the amplitude gains are retained as the system evolves away from the resonance, because there are no damping terms in the master equation (2), in accordance with the physical picture of a virtually isolated ion-electron pair whose interaction with the surroundings is at this stage negligibly weak. Note also that the transient resonance effect does not require a specific form of the function $\Omega(\beta\tau)$ and will take place under any temporal dependency of the electron cloud expansion. As seen in Fig. 2, the amplitude-enhancement interval is positioned roughly between the eighth and twelfth laser cycles. Given the accepted value of the parameter α, this corresponds to ~10 Å radius of the electron cloud. The corresponding effective charge density is $\sim 10^{21}$ $e\,\mathrm{cm}^{-3}$ which compares well with the critical plasma density for 800 nm laser light, 1.7×10^{21} cm^{-3}.

As the ion-electron pairs continue to emerge in the medium during the laser pulse (summing up to a total of about 10^3 per cubic wavelength), local polarization in the medium will result from the compounded action of all the "preplasma" dipoles that

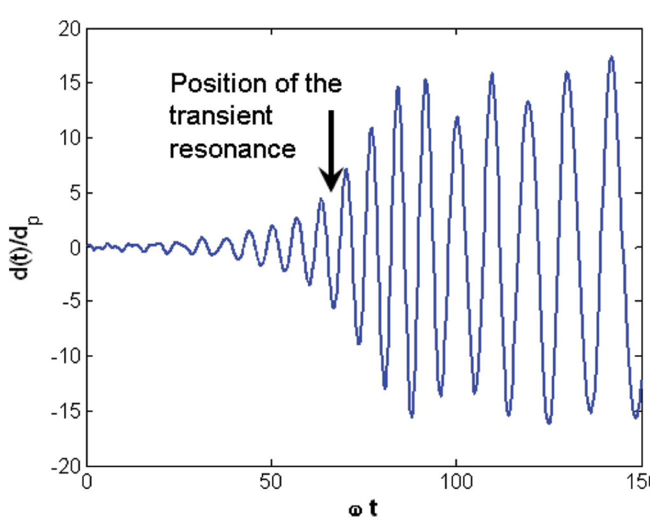

FIG. 1. (Color online) Postionization evolution of the effective dipole moment of an individual parent ion–expanding electron cloud system.

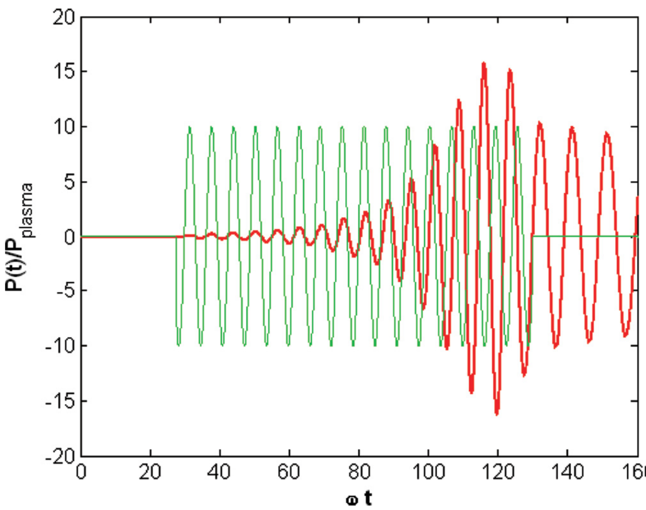

FIG. 2. (Color online) The cumulative polarization response of a medium that is being tenuously ionized by a laser pulse with rectangular envelope. (The laser electric field oscillations are shown for comparison, not to scale.)

have been initiated prior to the moment of observation:

$$P(t') = 4\pi \sum_{t_0<t} n(t_0)d(t',t_0) \to 4\pi \int_{-\infty}^{t'} dt_0 n(t_0)d(t',t_0)$$
$$= 4\pi d_p \int_{-\infty}^{t'} dt_0 n(t_0)\tilde{d}(\omega(t'-t_0),\omega t_0), \quad (11)$$

where $n(t_0)$ is the number-density rate of ionization, which depends (very nonlinearly) on $E(t_0)$. This nonlinear and nonsimultaneous polarization differs drastically from what one would expect from a homogeneous low-density plasma. As a simple illustration of the cumulative polarization effect, we calculate the transitional polarization response of the medium to a rectangular laser pulse $E(t_0) = E_0\Theta(t_0)$, where $\Theta(t_0)$ is the Heaviside step function. In this case, the ionization rate in Eq. (11) is reduced to a constant, $n(t_0) = n_0$, so that the integrand maintains its transpositional property under $t_0 \to t_0 + 2\pi/\omega$. The results of simulation are shown in Fig. 2. The time-dependent polarization retains the main features of the individual dipole response, viz., a sharp rise associated with the transient resonance and sustained large (negative) values, more than an order of magnitude greater than those of a homogeneous plasma having the same degree of ionization.

V. CONCLUSIONS

The proposed model is open to possible refinements to address more subtle effects and to account for atomic or molecular specifics. Note, however, that any such refinements will not change the two conceptual features: the localized, unplasmalike nature of the postionization response and the transient resonance enhancement of the electron oscillations. The physics revealed by the model suggests that the negative contributions to the index of refraction by the ionized electrons will be much more significant than those suggested by the currently accepted Drude model of homogeneous plasma. The differences will be especially pronounced for shorter laser pulses and thus may be expected to play a considerable role at the peak intensity developed during the filamentation process.

Finally, we comment briefly on the other two relevant areas mentioned in the introduction: wakefield acceleration and indirect-drive inertial confinement ignition. Although in these cases the peak laser intensities and the resulting eventual plasma densities are much larger than for laser filamentation, reaching the range of 10^{19} cm^{-3}, the distances between the nascent electron-ion pairs are still ∼40 Å, thus allowing for the spread of the initial electron cloud. Then the predicted effects will take place in these situations, provided that the ponderomotive wiggles of the electron cloud are smaller than the cloud size, as assumed in the model presented. For the typical parameters of the laser pulse in the gas-filled *Hohlraum* ionization case (multiple beams of 351 nm carrier wavelength, ∼10 ns duration, and 10^{16} W/cm^2 total focused intensity [14]), the ponderomotive radius $a = eE_0/(m_e\omega^2)$ is roughly estimated as $a \sim 10$ Å. In this case, the model may well serve as a qualitative guidance, but a nonlinear version is required for quantitative predictions. For the typical wakefield acceleration situation (a 30 fs pulse of 800 nm carrier wavelength with focused intensity of 10^{19} W/cm^2 [12]), the estimates give $a \sim 100$ Å in the peak region. However, even in this unfavorable case the model will be applicable to the leading ramp of the pulse and to the side areas of the laser beam where the critical instabilities are expected to emerge.

In conclusion, we considered the evolution of a gaseous system immediately following a multiphoton ionization event caused by an intense laser pulse. At a realistically low degree of ionization this initial evolution concerns individual electron-ion pairs rather than a low-density homogeneous plasma. The electromagnetic response of this pairwise system in the laser field differs significantly from the plasma picture. The individual dipole contributions undergo considerable modifications as their oscillations evolve through transient resonance. The results call for reevaluation of current laser filamentation models in the most critical issue of the filament stabilization.

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