Appearance intensities for multiply charged ions in a strong laser field

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We study multiple ionization of atoms by a strong laser field using a time-dependent approach based on the Thomas-Fermi model. The evolution of the electron density under the influence of a transient, linearly polarized radiation field is determined by numerical solution of hydrodynamic equations on a two-dimensional grid. We find a threshold "appearance intensity" for a given charge state, i.e., the laser intensity at which such a charge state first appears, which is in reasonable agreement with experimental results for high charge states.

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I. INTRODUCTION

Experiments on the interaction of atoms with short light pulses with peak intensities greater than 10^{12} W/cm^2 have led to a discovery of many new phenomena: generation of intense extreme ultraviolet and x radiation; excess-photon or above-threshold ionization; suppression of the ionization rate at high intensities ("stabilization"); and efficient production of highly charged ions. A fairly recent summary of the present state of this area of physics may be found in Ref. [1].

The main qualitative features of the first three of these phenomena—high harmonic generation, above-threshold ionization, and stabilization—can be obtained in simple models that invoke a picture of one electron under the influence of an effective potential, and they are also found in numerical experiments which integrate the time-dependent Schrödinger equation for one-electron systems under fairly realistic conditions. Thus it is reasonable to think of these as single-electron phenomena, though it may be necessary to account for many-electron effects to understand them in detail. The production of multiply charged ions by laser irradiation of neutrals is, on the other hand, of an essentially many-electron character. It has not yet been possible to perform numerical experiments with realism comparable to that which has been attained in the one-electron case, and it seems unlikely that any such treatment will be made in the near future to an atom with more than two electrons: the two-electron system in the nonperturbative regime is just beginning to be explored [2,3], though there has been extensive work on a model system of two electrons moving in one dimension [4]. However, with the increase of attainable laser intensity, experiments have moved into a regime of unprecedented high ionization: recent reports [5,6] of experiments at close to 10^{19} W/cm^2 exhibit ion charge states of more than 40+. Thus there is a need to develop a theoretical framework within which large numbers of electrons can be treated.

This paper presents an approach to this problem based on the Thomas-Fermi statistical model of the atom. It provides us with a single quantity, the electron density, whose behavior in a linearly polarized radiation field can be computed by solving a time-dependent partial differential equation in the cylindrical polar coordinates ρ and z. We find that for a given laser intensity the atom becomes ionized to a degree that is relatively independent of the initial turn on of the pulse and is stable over long integration times. Our results are in reasonable agreement with experimental data for ionic charges greater than 4 or so.

Our presentation is organized as follows. In Sec. II we review experimental results on multiple ionization in strong laser fields. Section III presents the formalism underlying the time-dependent Thomas-Fermi model, and its numerical implementation. In Sec. IV we discuss the results.

II. REVIEW OF EXPERIMENTS AND THEIR INTERPRETATIONS

The first observation of laser-induced multiple ionization of an atom, double ionization of strontium [7], showed an unexpectedly high value of the corresponding ionization cross section. Progressive increase of attainable laser intensity made it possible to strip the entire outer shells of noble gases [8–11], and there is recent evidence for substantial removal of inner-shell electrons as well [5].

The appearance of successive ionization stages may be summarized as follows. At low intensities I, the yield of an ionic species X^{q+} is proportional to $I^{n(q)}$, where $n(q)$ increases with q. For small I, only the singly charged species X^+ is observed; when X^{2+} first becomes observable as I increases, its yield can grow by several orders of magnitude with an increase of I by a factor of 2 (see, e.g., [12]). This phenomenon occurs sequentially for charge states $q > 2$ as I increases further. Thus for each q there is an "appearance intensity" I_q at which X^{q+} first becomes observable. The appearance intensity depends in principle upon the dynamic range of experimental detection, but in practice this dependence is quite weak, and there is general agreement of appearance intensities determined by independent experiments.

There are several theoretical explanations of this phenomenon and of the dependence of I_q upon atomic number Z and laser frequency ω. One of the most successful is based on Coulomb barrier suppression [13,11]. In this picture, the electric field of the laser is viewed as quasistatic, and it is superimposed upon the atom's internal field to create the typical barrier structure of the dc Stark effect. X^{q+} appears when this barrier is sufficiently low to allow q electrons to escape from the atom.

To obtain predictions from this model, one needs a description of the effective potential of a many-electron atom. In previous work [13], we utilized the Thomas-Fermi model for this purpose. As we describe below, the Thomas-Fermi model provides a framework for dealing directly with the electron density $\rho_e(\vec{r})$ of the atomic system, rather than with the coordinates of individual electrons. Thus it provides a set of basic equations for determining the electron density in the combined external and nuclear electric fields, which accounts for effects of electron-electron interaction and Fermi-Dirac statistics in an approximate way. We solved these equations to find the maximum number of electrons that could remain localized about the nucleus in such combined fields. Comparison with experiment was made by equating the static field strength to the peak electric field strength of the laser. The results are in good agreement with the experimental data for the low frequency fields, such as are provided by CO_2 laser experiments [10], and for high charge states [11]. The explanation is simple. For the static field picture to be valid, the atom should rearrange itself according to the external field on a time scale shorter than the optical period, and an electron which is above the barrier should be able to escape from the atom during a fraction of the optical period.

In actual experiments, however, we must expect that both the laser frequency and the detailed pulse shape can play significant roles in the process of ionization. Hence a time-dependent approach is needed. In the present paper we develop such an approach, and present the first results for the Thomas-Fermi atom subject to a short laser pulse of optical frequency. The equations of the model resemble a hydrodynamic equation for a droplet shaken by an external oscillating force. The droplet, however, consists of a charged electronic "fluid," and the strong internal interactions of this fluid self-consistently modify the effective potential that governs its motion.

III. A TIME-DEPENDENT THOMAS-FERMI MODEL

Even the time-independent Schrödinger equation for the multielectron atom is too complex to be solved directly. An approximate treatment of atomic structure based on a statistical model was proposed in the 1920s by Thomas and Fermi [14]. They viewed the atom as a gas of almost free electrons, whose density $\rho_e(\vec{r})$ is a continuous function. Such a coarse-grained description requires that there are many electrons in every cell in space, over which the total electrostatic potential Φ is nearly constant, and that the distribution of energies among electrons in each cell is that appropriate to a Fermi-Dirac ensemble at zero temperature. This account of Fermi-Dirac statistics is the only element of quantum theory in the model. Finally, the effects of individual electron-electron interactions are subsumed in the self-consistent electrostatic potential Φ, which satisfies a classical Poisson equation appropriate to the charge distribution $\rho_e(\vec{r})$ and with boundary conditions consistent with externally applied fields.

Over the years a number of improvements of the original Thomas-Fermi model have been proposed. Among them Dirac [15] introduced the correction describing the exchange energy of electrons while Fermi and Amaldi [16] proposed a way of subtracting a self-interaction of electrons. These two corrections improve the description of outer parts of the electronic cloud, where, in a real atom, the electron density varies rapidly. The original Thomas-Fermi model describes this region quite poorly; for a neutral atom, the density decays too slowly, and the static polarizability diverges. In our earlier treatment of the static model [13] we have investigated the effects of these corrections.

In the present paper we study the response of the Thomas-Fermi atom to a short and intense pulse of linearly polarized light. We shall describe our results in terms of the usual system of atomic units (a.u.) in which the numerical values of the electron mass m, the absolute value of electron charge e, and reduced Planck's constant $\hbar$ are equal to unity, although these constants are retained explicitly in the formulas of our derivations. In this system of units, radiation intensity of 3.5×10^{16} W cm^{-2} corresponds to a peak electric field strength of $F = 1$ a.u.

Our underlying assumption is that the oscillations of the electron cloud in a many-electron atom of nuclear charge Ze can be viewed as a motion of a fluid characterized by a mass density $\rho_e(\vec{r},t)$ and a velocity field $\vec{v}(\vec{r},t)$. The motion of this fluid is described [17] by the hydrodynamic equations

$$\frac{\partial \rho_e(\vec{r},t)}{\partial t} + \vec{\nabla}\cdot[\rho_e(\vec{r},t)\vec{v}(\vec{r},t)] = 0 \ , \tag{3.1}$$

$$\frac{\partial \vec{v}(\vec{r},t)}{\partial t} + [\vec{v}(\vec{r},t)\cdot\vec{\nabla}]\vec{v}(\vec{r},t) = -\frac{1}{\rho_e(\vec{r},t)}\vec{\nabla}P(\vec{r},t) + \frac{e}{m}\vec{\nabla}\Phi(\vec{r},t) \ .$$

The first of these is the usual continuity equation expressing the conservation of mass. The second derives from the classical equations of motion for an infinitesimal element of fluid subject to the gradients of the pressure P and the electrostatic potential Φ. The sole quantum-mechanical ingredient in this approach is the constitutive equation connecting P and ρ_e,

$$P(\vec{r},t) = \frac{1}{5}(3\pi^2)^{2/3}\frac{\hbar^2}{m}[n(\vec{r},t)]^{5/3}$$

$$[n(\vec{r},t) = \rho_e(\vec{r},t)/m] \ , \quad (3.2)$$

which derives from applying Fermi-Dirac statistics to an ensemble of noninteracting particles distributed homogeneously at a number density $n(\vec{r},t)$ per unit volume. For a radiation field polarized along the z axis and treated in the dipole approximation the electrostatic potential takes the form

$$\Phi(\vec{r},t) = \frac{Ze}{r} - \int \frac{en(\vec{r}\,',t)}{|\vec{r}-\vec{r}\,'|}d^3r' - \vec{F}(t)\cdot\vec{r} \ , \quad (3.3)$$

where $\vec{F}(t)$ is the radiative electric field. Equation (3.3) can be cast in terms of the Poisson equation:

$$\Delta\Phi(\vec{r},t) = 4\pi e n(\vec{r},t) \ , \quad (3.4)$$

which must be solved subject to appropriate boundary conditions. Solution of Φ from (3.4) rather than (3.3) is preferred from a computational standpoint.

Thus we are posed with the problem of simultaneous solution of a set of hyperbolic equations (3.1) and an elliptic equation (3.4). These equations have been solved by Ball, Wheeler, and Fireman [17] in the weak-field limit, when the fluid executes small oscillations about the Thomas-Fermi density $\rho_{\rm TF}(r)$ which describes the free atom in the absence of a radiation field. From the normal-mode spectrum of these oscillations one can determine the frequency-dependent polarizability $\alpha(\omega)$ of the atom. This approach is not adequate to describe the strong-field case, when a substantial fraction of the electron fluid is stripped from the atom. We have treated this case as follows.

Equations (3.1)–(3.4) are symmetric about the direction of polarization of the light field, which we take to be the z axis, so it is convenient to describe the system in cylindrical coordinates. We treat the system as an initial-value problem in time t, with $\vec{F}(t=0)=0$. Thus $\rho_e(\vec{r},0) = \rho_{\rm TF}(r)$; the axial symmetry of the solution at $t=0$ thus ensures that the density is a function only of the cylindrical coordinates ρ, z for all $t>0$. Since for heavy atoms a substantial amount of charge will always remain in the vicinity of the nucleus, it is convenient to deal with the difference of the electron density from its initial distribution by use of the variable

$$\rho_d(\rho,z,t) = \rho_e(\rho,z,t) - \rho_{\rm TF}(r). \quad (3.5)$$

The system of equations (3.1)–(3.4) can be written as

$$\begin{aligned}
\frac{\partial\rho_d(\rho,z,t)}{\partial t} &= -\frac{1}{\rho}\frac{\partial}{\partial\rho}\{\rho[\rho_{\rm TF}(r)+\rho_d(\rho,z,t)]v_\rho(\rho,z,t)\} \\
&\quad -\frac{\partial}{\partial z}\{[\rho_{\rm TF}(r)+\rho_d(\rho,z,t)]v_z(\rho,z,t)\} \ , \\
\frac{\partial v_\rho(\rho,z,t)}{\partial t} &= -v_\rho(\rho,z,t)\frac{\partial v_\rho(\rho,z,t)}{\partial\rho} \\
&\quad -v_z(\rho,z,t)\frac{\partial v_\rho(\rho,z,t)}{\partial z} \\
&\quad -\frac{1}{m}\frac{\partial\Phi_{\rm eff}(\rho,z,t)}{\partial\rho} \ , \\
\frac{\partial v_z(\rho,z,t)}{\partial t} &= -v_\rho(\rho,z,t)\frac{\partial v_z(\rho,z,t)}{\partial\rho} \\
&\quad -v_z(\rho,z,t)\frac{\partial v_z(\rho,z,t)}{\partial z} \\
&\quad -\frac{1}{m}\frac{\partial\Phi_{\rm eff}(\rho,z,t)}{\partial z} \ ,
\end{aligned} \quad (3.6)$$

where v_z, v_ρ are components of the velocity field in the cylindrical coordinate system, and

$$\Phi_{\rm eff}(\rho,z,t) = \frac{1}{2}(3\pi^2)^{2/3}\frac{\hbar^2}{m}[n(\rho,z,t)]^{2/3} - e\Phi_{\rm TF}(r) + \int\frac{e^2 n_d(\vec{r}\,')}{|\vec{r}-\vec{r}\,'|}d^3r' + eF(t)z \ , \quad (3.7)$$

where $\Phi_{\rm TF}$ is the electrostatic potential calculated from the static Thomas-Fermi model [in practice we use analytical approximations to $\Phi_{\rm TF}$ and $\rho_{\rm TF}(r)$ given in Ref. [18]]. The set of partial differential equations (3.6) constitutes an initial-value problem of the hyperbolic type. To solve it we used the Lax method, which is accurate to first order in the time step. Its basic idea is to introduce numerical viscosity into the equation to avoid the instability associated with the simplest forward time centered space (FTCS) difference scheme [19]. Only the first of the equations (3.6) has the so-called flux-conservative form [19]. This form is most convenient because the flux-conservation property of the differential equation is preserved in the finite-difference approximation [20]. Although the other two equations of (3.6) do not have the flux-conservative form, our method conserves the total charge on the grid (for appropriate boundary conditions). This is achieved by replacing the second and third equations by the finite-difference formulas of the Lax method, and transforming the first equation to a difference expression based on the integral form of the continuity equation.

In the applications reported here, we utilized a uniform grid in ρ and z with a step size typically equal to 0.1 a.u. to represent all dependent variables. Solution of the Poisson equation required to generate the potential (3.7) was carried out on the same grid. Spatial derivatives were approximated by central difference formulas, giving second order accuracy in the grid spacing. There are differences in the methods of solving the equations according to whether the nucleus was placed on a grid node or between nodes; the latter case was found to be better behaved in practice. We considered two different conditions imposed at the boundary of the grid region. The first,

$$\rho_d|_{\text{edge}} = 0, \quad v_\rho|_{\text{edge}} = 0, \quad v_z|_{\text{edge}} = 0, \tag{3.8}$$

does not allow fluid to flow across the boundary, and was invoked in order to check whether our integration method preserved the total charge on the grid. The second,

$$(\partial\rho_d/\partial\vec{n})_{\text{edge}} = 0, \quad (\partial v_\rho/\partial\vec{n})_{\text{edge}} = 0, \quad (\partial v_z/\partial\vec{n})_{\text{edge}} = 0, \tag{3.9}$$

corresponds to the situation of an atom in open space, in which the fluid may flow freely across the (arbitrarily chosen) boundary of the grid. Since we do not retain a representation of the fluid outside the grid region, this is effectively an absorbing boundary condition, and it provides the mechanism by which the atom is ionized in our model.

The initial conditions on our time-dependent solution are

$$\rho_d(\vec{r}, t=0) = 0, \quad v_\rho(\vec{r}, t=0) = 0, \quad v_z(\vec{r}, t=0) = 0, \quad \Phi_{eff}(\vec{r}, t=0) = 0. \tag{3.10}$$

At every time step the Poisson equation is solved to find the contribution to the effective potential (3.7) from the charge density $-en_d(\vec{r}, t)$. We used the over-relaxation method, a standard technique for elliptic equations [21]. The Poisson equation is approximated by a five-point difference equation in the cylindrical coordinate system. In every iteration the grid is scanned point by point and a new value of the potential is obtained from the difference equation. The boundary conditions at the outer edge of the grid are updated after each iteration and are calculated from the density $n_d(\vec{r})$ by a Cartesian multipole expansion. The procedure is convergent within a finite number of iterations depending on the value of the over-relaxation parameter and physical data. In propagating the solution forward in time t, we found it necessary to use several thousand time steps per optical period $T = 2\pi/\omega$ to attain numerical convergence. The laser pulse was turned on with a $\sin^2$ ramp over times of $0 - 12$ T. The pulse duration was limited to about 30 T, which is short compared to actual experiments; however, in the cases we report here, the final charge state of the ion does indeed appear to be determined fairly early in the pulse.

IV. RESULTS

We have performed an extensive study of the response of the Thomas-Fermi atom to a strong linearly polarized pulse using the method outlined in Sec. III. Here we present two sets of results: first, for the Nd:YAG (where YAG denotes yttrium aluminum garnet) laser ($\omega = 0.043$ a.u.) for which much experimental data have been obtained over the years [8,11]; second, for the strong fields produced by a KrF laser ($\omega = 0.183$ a.u.) that have been reported recently [5]. For the Nd:YAG case we are able to follow the evolution over 20 to 30 optical cycles. Reliable calculations over a realistically long pulse for lower frequencies, such as that of the CO_2 laser, are impractical for us at present due to constraints on computational time.

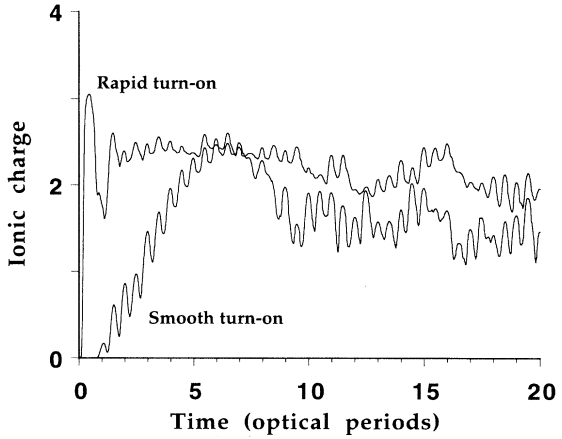

FIG. 1. Ionic charge in the "weak"-field case $F = 0.1$ a.u. for smooth and rapid turn ons of the pulse. It is not possible to determine the appearance intensity here.

All our results are for the xenon atom ($Z = 54$). Convergence of our method requires strong fields, since in weak fields very little ionization occurs and the actual amount is sensitive to the details of the pulse shape. This point is illustrated for the case $F = 0.1$ a.u. in Fig. 1. "Ionic charge" means the net electric charge of the atom, and is equal to the number of electrons that have been removed. Here it is not possible to determine an unambiguous appearance intensity. This fact is consistent with our estimates [13] that several outer electrons in the Xe atom are too slow to escape above the barrier at the Nd:YAG frequency at this field strength.

For a peak field strength $F = 0.2$ a.u., nearly four

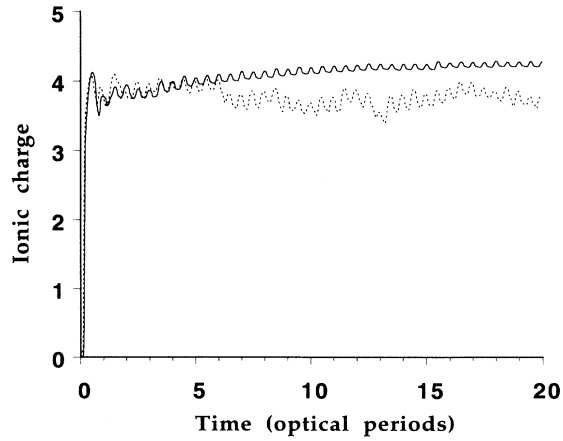

FIG. 2. Evolution of the ionic charge in a rapidly turned-on field of strength $F = 0.2$ a.u. Comparison of grids of 80×160 (dashed line) and 160×320 (solid line) points.

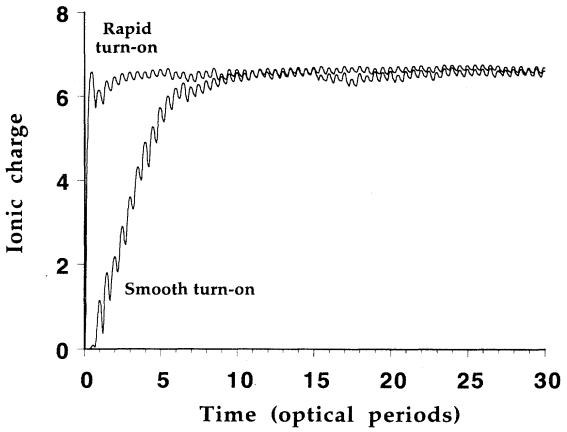

FIG. 3. Evolution of the ionic charge in a field of peak strength $F = 0.5$ a.u. Note the weak dependence upon pulse shape.

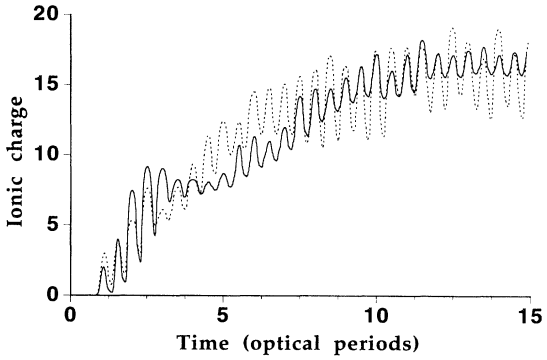

FIG. 4. Evolution of the ionic charge in a laser pulse under the conditions of the experiment of Ref. [5]: $F = 15.1$ a.u., $\omega = 0.183$ a.u. Solid line: time step of 1×10^{-5} a.u.; dashed line: time step of 2.5×10^{-5} a.u. The resulting ionic charge is much less than the corresponding value of 27 obtained from the static model.

electrons are stripped from the atom. In Fig. 2 we show the time dependence of the number of ionized electrons for two grid sizes: 80×160 points and 160×320 points (i.e., maximum values of $|x|$, $|y|$, and $|z|$ of 8 a.u. and 16 a.u., respectively). The dependence has nearly a stepwise character. The electrons are ejected during the first cycle. The subsequent evolution does not produce a noticeable electronic flux. So in this area of field strength we may define an appearance intensity that is independent of pulse shape.

For stronger fields, when electrons closer to the nucleus are affected, the droplet readjusts itself to the changing electric field almost instantaneously. This point is illustrated in Fig. 3. This shows a comparison of residual charges for a rapid turn on and for a six-cycle ramp. The charge flow is seen to follow the pulse, and the degree of ionization is simply determined by the peak intensity. There is no way one can introduce a decay rate here. The process has a binary character and the notion of the appearance intensity is very natural.

In Table I we compare the present appearance intensities with those of our previous static-approximation calculations and with the experimental data of [11]. As expected, the agreement of the present model with experiment is rather good for higher charge states.

Finally, in Fig. 4 we show the evolution of the charge for $F = 15.1$ a.u., $\omega = 0.183$ a.u., corresponding to the experimental conditions of Ref. [5] (KrF laser radiation at intensities of 10^{19} W/cm^2). This experiment observed Xe L-shell x-ray line emission, consistent with the presence of charge states in excess of 40+. An estimate of the maximum charge state that can be produced by a static field of this strength, using the methods of Ref. [13], gives 27+, well under what is required to explain the charge states observed experimentally. As seen in Fig. 4, our time-dependent calculations give an ionization of under 20, which is even further from experiment. However, these results are consistent with other estimates of the degree of atomic ionization that can be produced by direct laser-atom interaction [5,6,22]. Production of high charge states is believed to be mediated by electron-impact ionization of the initial ionic population, caused by the initial ionized electrons which are accelerated to keV quiver energies by the oscillating light field [5,6,22].

TABLE I. Appearance intensities in units of 10^{13} W cm^{-2} for charge states of Xe in a Nd:YAG laser field. Present results in comparison with static TF and TFDJ (Thomas-Fermi-Dirac-Jensen) approximations of Ref. [13] and with experiment of Ref. [11].

	Intensity			
Charge state	Present work	Static TF	Static TFDJ	Experiment
1		0	0.15	7
2		0.14	1.83	18
3		2.12	9.19	40
4	84	11.3	30.7	92
5	169	37.7	79.5	160
6	406	98.2	181.5	214
7	710	220	367	672
8	1024	442	683	802

This mechanism is expected to be more effective in dense systems, such as atomic clusters, than in isolated atoms, and we have begun work on extending the present approach to clusterlike systems.

ACKNOWLEDGMENTS

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