Boosting Photoabsorption by Attosecond Control of Electron Correlation

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(Received 24 May 2013; published 20 September 2013)

Electron correlation plays an essential role in a wide range of fundamentally important many-body phenomena in modern physics and chemistry. An example is the importance of electron-electron correlation in multiple ionization of multielectron atoms and molecules exposed to intense laser pulses. Manipulating the dynamic electron correlation in such photoinduced processes is a crucial step toward the coherent control of chemical reactions and photobiological processes. The generation of an attosecond extreme ultraviolet (EUV) pulse may enable such controls. Here, we show for the first time, from full-dimensional *ab initio* calculations of double ionization of helium in intense laser pulses ($\lambda = 780$ nm), that the electron-electron interactions can be instantaneously tuned using a time-delayed attosecond EUV pulse. Consequently, the probability of producing energetic electrons from excessive photoabsorption can be enhanced by an order of magnitude, by the attosecond control of electron-electron correlation.

DOI: 10.1103/PhysRevLett.111.123003 PACS numbers: 32.80.Rm, 32.80.Fb, 42.65.Re

Electron correlation has been of extreme importance to account for a variety of many-body phenomena in modern physics [1,2] and chemistry [3], ranging from superconductivity, molecular structure formation, and chemical reaction to high-order harmonic generations [4]. For example, when multielectron atoms and molecules are exposed to an intense laser pulse, multiple electrons can be ionized even if the laser field is not strong enough to strip them one by one *sequentially*. The dynamical electron correlation is believed to be responsible for such *nonsequential* multiple ionization [5–7] in strong fields. Namely, upon its return, a laser-driven electron can either directly knock out other electrons [8] or excite them into high-lying states that can subsequently be ionized by the strong laser field [9,10]. Both pathways essentially depend on the dynamical electron-electron correlation under the oscillating electromagnetic field.

The pioneering investigations on electron correlations in strong laser fields started 30 years ago when the nonsequential double ionization (NSDI) process was first observed [11–13]. It was shown that a surprising orders-of-magnitude enhancement of double ionization yield was measured for atoms exposed to intense laser pulses. Since then many experimental [14–18] and theoretical efforts [19–25] have been put forth to understand the underlying mechanism. Great progress has been made by these kinematically complete measurements [14–18], especially with the use of a single-cycle pulse in a recent NSDI experiment [26] in which only a single recollision was allowed. On the theory side, full-dimensional *ab initio* calculations of NSDI are still lacking, particularly in the infrared wavelength regime ($\lambda \approx 800$ nm), where most experiments were performed.

Over the past decades, the recollision mechanism [27,28] has become an important framework for conceptually understanding the nonsequential double ionization. This ''step-by-step'' approach interprets the occurrence of NSDI through either the inelastic $(e, 2e)$ process [8] or the recollision-induced excitation with subsequent ionization [9,10] for certain laser intensities, above which the returning electron can be energetic enough to either directly kick out the second electron or promote it to the lowest excited state. Apparently, in this picture these inelastic processes were made possible solely by the returned electron itself. It was therefore thought that there should be certain intensity thresholds for them to occur, which has been puzzling for the NSDI observations below such intensity thresholds [13,17].

In principle, quantum mechanics allows any additional photons to be absorbed during the recollision process; however, the more photons involved, there is less probability it may occur for a perturbative absorption process. But if the external field is strong enough to compare with the internal electron-ion or electron-electron interactions, the nonperturbative picture predicts the nonvanishing probabilities even for high-order nonlinear processes. This is exactly the mechanism that explained the extended energy plateau of above-threshold ionization up to $\sim 10\,U_p$ (U_p is the ponderomotive potential), observed in atomic single ionization in strong laser fields [29]. For a multielectron system, the electron-electron correlation plays a crucial role in determining the exact wave functions of the initial, intermediate, or final states involved in photoabsorption or photoemission processes. Therefore, the final cross section of photoabsorption or photoemission for multielectron systems is essentially sensitive to the inherent e-e correlations, which have been illustrated by many calculations and experiments [30,31]. One important example is the enhanced ionization of atoms through the e-e correlation-induced autoionization resonance [32]. In general, if the electron-electron correlation in multielectron systems can be manipulated to be comparable with any external laser-electron interaction, nonperturbative nonlinear

photoabsorption could be possible in a multiple-ionization process. The generation of attosecond pulses [33–35] may enable such manipulation of electron correlation in NSDI. Here, we present full-dimensional *ab-initio* studies on the NSDI control of atomic helium by tuning the electron-electron interaction with a time-delayed attosecond extreme ultraviolet (EUV) pulse.

The numerical details have been described in Ref. [36]. Briefly, we consider a fully correlated, two-active-electron helium atom under the combined irradiation of an intense, linearly polarized attosecond EUV pulse and a few-cycle pulse (FCP) [37] of intensity 2×10^{14} W/cm^2 at $\lambda=780$ nm. The field strength of $T=5$ fs (total pulse length) FCP with a carrier-envelope phase $\phi_0=0$ is plotted by the dashed line in Fig. 1(a). The FCP field is equal to the time derivative of its vector potential, i.e., $E(t)=-\partial A(t)/\partial t$ with $A(t)=-(E/\omega)\sin^2[\pi(t+T/2)/T]\sin[\omega(t+T/2)+\phi_0]$ and $-T/2<t<T/2$. Under the interaction of external fields, the fully correlated two-electron motion is governed by the six-dimensional (6D), time-dependent Schrödinger equation (TDSE), having the following form (atomic units are used throughout):

$$i\frac{\partial}{\partial t}\Phi(\mathbf{r}_1,\mathbf{r}_2|t)=\left[-\frac{1}{2}(\Delta_{\mathbf{r}_1}+\Delta_{\mathbf{r}_2})-\frac{2}{\mathbf{r}_1}-\frac{2}{\mathbf{r}_2}+\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}+\mathbf{E}_{\rm tot}(t)\cdot(\mathbf{r}_1+\mathbf{r}_2)\right]\Phi(\mathbf{r}_1,\mathbf{r}_2|t), \quad (1)$$

where $\mathbf{r}_1$ and $\mathbf{r}_2$ are the position vectors of each electron, with respect to the nucleus. The infrared (IR) + EUV pulse interaction is expressed by the last term in the above equation. Following the time-dependent close-coupling method [38–40], we can expand the 6D unknown wave function $\Phi(\mathbf{r}_1,\mathbf{r}_2|t)$ by using the orthonormal basis functions of bipolar-spherical harmonics $y^{L,M}_{l_1l_2}(\Omega_1,\Omega_2)$,

$$\Phi(\mathbf{r}_1,\mathbf{r}_2|t)=\sum_{L,M}\sum_{l_1l_2}\frac{\Psi^{(L,M)}_{l_1l_2}(r_1,r_2|t)}{r_1r_2}y^{L,M}_{l_1l_2}(\Omega_1,\Omega_2),$$

for a specific symmetry (L, M). We can also expand the Coulomb repulsion term $1/|\mathbf{r}_1-\mathbf{r}_2|$ and the field interaction

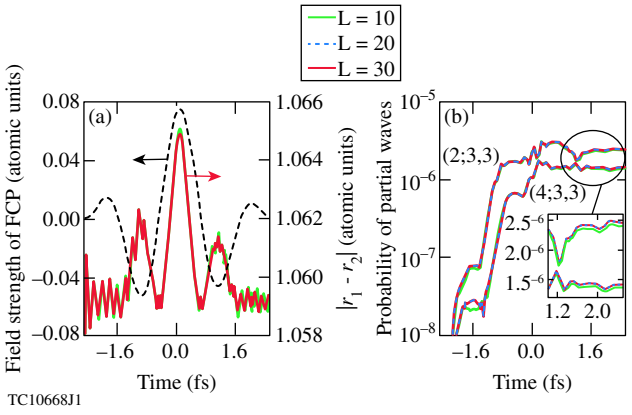

FIG. 1 (color online). (a) The pulse shape of a few-cycle pulse (FCP) and the expected value of interelectron distance $|r_1-r_2|$ versus time; (b) the probability of most-populated partial waves as a function of time, for different partial-wave expansions.

$\mathbf{E}_{\rm tot}(t)\cdot(\mathbf{r}_1+\mathbf{r}_2)$ in terms of spherical harmonics. Substituting these expansions into the above Schrödinger equation (1), multiplying the conjugate of the basis functions, and integrating over the angles Ω_1 and Ω_2, we end up with a set of coupled partial differential equations for a series of partial waves (with only two radial variables r_1 and r_2 remaining):

$$i\frac{\partial}{\partial t}\Psi(r_1,r_2|t)=[\hat{T}_1+\hat{T}_2+\hat{V}_{\rm C}]\Psi_j(r_1,r_2|t)+\sum_k\hat{V}^I_{j,k}(r_1,r_2|t)\Psi_k(r_1,r_2|t), \quad (2)$$

where the partial wave index j runs from 1 to the total number of partial waves used for expansion. In such a referencing scheme, each index j corresponds to a specific momentum combination (L, M, l_1, l_2). In Eq. (2), the diagonal operators $\hat{T}_1$, $\hat{T}_2$, and $\hat{V}_{\rm C}$ give the kinetic energies and the Coulomb attractions between each electron and the nucleus, while the off-diagonal potential term $\hat{V}^I_{j,k}(r_1,r_2|t)$ consists of the Coulomb repulsion between two electrons and their interactions with external fields. Since the photons of linearly polarized IR and EUV pulses have zero magnetic quantum number ($m=0$), the photoabsorption would not increase the total magnetic quantum number of the system. Therefore, for the singlet ground state He ($M=0$) being chosen as the initial state, the magnetic quantum number of the system is constantly kept at zero.

Combining the real-space-product algorithm [41] with the fine-gridding scheme of the finite-element discrete variable representation (FEDVR) [42], we have numerically solved the coupled Eq. (2) on a two-dimensional grid. This method has been successfully applied to three-body Coulomb collisions [43–45] as well as few-photon ionization of atoms and molecules with EUV pulses in the past [46–48]. It is very challenging, however, to perform such *ab initio* calculations in the Ti:sapphire wavelength regime ($\lambda\sim800$ nm) because a larger number of partial waves are needed for convergence. In addition, the two radial dimensions must be large enough to accommodate the large electron excursion during the long IR pulse. For the calculations presented here, we have used 240 finite elements with five-point DVR per FE, which gives a maximum radius of ~305 bohr. Before the field interactions, we relaxed a trial wave packet with 11 partial waves ($l_1=l_2=0$ to 10 and $L=0$) in imaginary time and obtained a ground state energy of -2.9001 atomic units (a.u.), in good agreement with the experimental value of -2.9037 a.u. for He. To the best of our knowledge, the *ab initio* studies presented here are the first full-dimensional TDSE calculations of NSDI in the Ti:sapphire wavelength regime, in contrast to previous efforts in the wavelength regime of 390 nm [49]. This is made possible through the optimization of our finite-element discrete variable representation-time-dependent close-coupling code [36], which speeds up the calculations on supercomputers by an order of magnitude for extremely large partial waves involved. The convergence has been tested for different numbers of partial waves: $L=10$ (201 partial waves), $L=20$ (261 partial waves), and $L=30$ (295 partial waves). The testing results are

shown in Figs. 1(a) and 1(b). The colored lines in Fig. 1(a) represent the expectation values of the interelectron distance $|r_1-r_2|$ for the three cases (FCP only). The evolution of two partial waves [with angular momentum combinations of $(L; l_1, l_2) = (2; 3, 3)$ and $(4; 3, 3)$], having the most populations, is plotted in Fig. 1(b), where the details are enlarged in the inset. One sees that the convergence is indeed reached at the $L = 20$ and $L = 30$ cases. All our results presented below have used the converged expansion of total angular momentum up to $L = 30$ (295 partial waves).

The plot of $|r_1-r_2|$ as a function of time in Fig. 1(a) indicates the basic tunneling and recollision behavior of a single electron. The two minima of interelectron distance at around $t = -0.4$ and $t = +0.8$ fs manifest the two recollision instants during the IR pulse interaction. Since the ionization probability of He is very small for the IR intensity used, the variation of $\langle|r_1-r_2|\rangle$ is only on the level of 10^{-3}. It is noted that for the IR-FCP parameters considered, the maximum returning energy of the tunneled electron ($\sim 3.2\,U_p \simeq 36$ eV) is below the first excitation of He^+ [$\Delta E(1s$–$2p) \simeq 40.8$ eV)]. One can imagine that the ground-state $1s$ electron, sitting in the deep potential well of He^+, may experience only a perturbation from the recolliding electron wave packet. Depending on the composition of the returning electron wave packet, high-angular momentum waves may approach only to certain distance from the $1s$ electron because of the repulsive centrifugal barrier of $+l(l+1)/2r^2$. However, if the ground-state $1s$ electron is timingly promoted to the $2p$ state of He^+ by an attosecond EUV pulse at the recollision instants, it can undergo a strong, nonperturbative interaction with the returning electron. The nonpertubative e-e recollision in the external laser field may lead to strongly nonlinear photoabsorption. For attosecond excitation before the recollision, the IR field ionization could remove the "target" $2p$ electron. While, if the excitation occurs after the recollision, the e-e correlation may become weak because of the increasing distance between them, so that they may behave independently in the external IR field.

To test this idea of attosecond control of the e-e correlation, we carried out *ab initio* calculations for NSDI of He in the IR-FCP together with a time-delayed attosecond EUV pulse. The attosecond EUV pulse had a pulse duration of 500 asec and an intensity of 2×10^{14} W/cm^2, with $\hbar\omega \simeq 41$ eV matching the single-photon $1s$-$2p$ transition of He^+ [without excitation of other states due to the narrow full-width-half-maximum (FWHM) bandwidth of $\Delta\omega \simeq 12$ eV]. We controlled the time delay of the attosecond EUV pulse relative to the IR-FCP, from $t_d = -2.0$ fs to $t_d = +2.0$ fs with a step of 200 as. The time delay t_d is defined as the temporal gap between the IR laser peak and the peak of the attosecond pulse. At the end of the overall interaction ($t = 5$ fs), we plotted the electron wave packet probability density in the plane spanned by r_1 and r_2. Some examples are shown in Fig. 2 for different time delays. Figure 2(c) shows the large double-ionization probability (in the region at larger r_1 and r_2) for $t_d = 0.8$ fs, in contrast

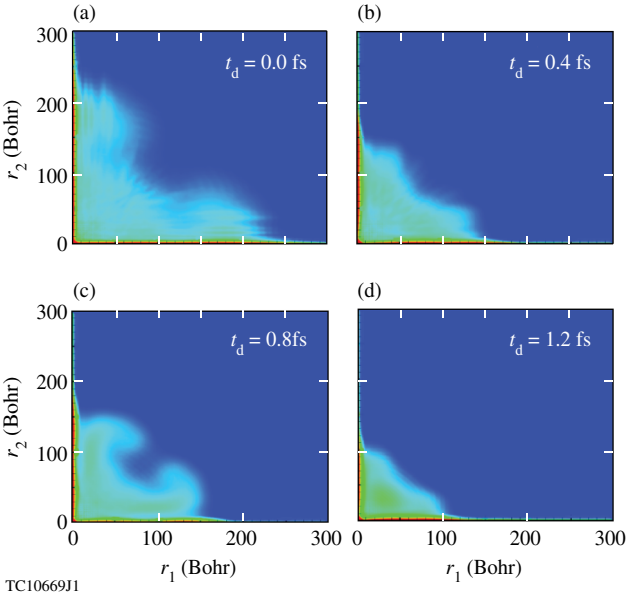

FIG. 2 (color online). The final electron probability distribution on the plane spanned by the radial coordinates r_1 and r_2 for different time delays between the attosecond extreme ultraviolet (XUV) pulse and the FCP: (a) $t_d = 0.0$ fs, (b) $t_d = 0.4$ fs, (c) $t_d = 0.8$ fs, and (d) $t_d = 1.2$ fs.

to other panels at different time delays. To compute the probability of double ionization, we projected the final wave packets onto the two-electron continuum (a product of two single-electron continua). The corresponding NSDI probability distributions in momentum space are displayed in Fig. 3 for the cases shown in Fig. 2. One sees from Fig. 3(c) that at $t_d = +0.8$ fs, the returning electron undergoes a strong e-e interaction with the attosecond EUV-pulse-excited $2p$ electron of He^+, thereby leading

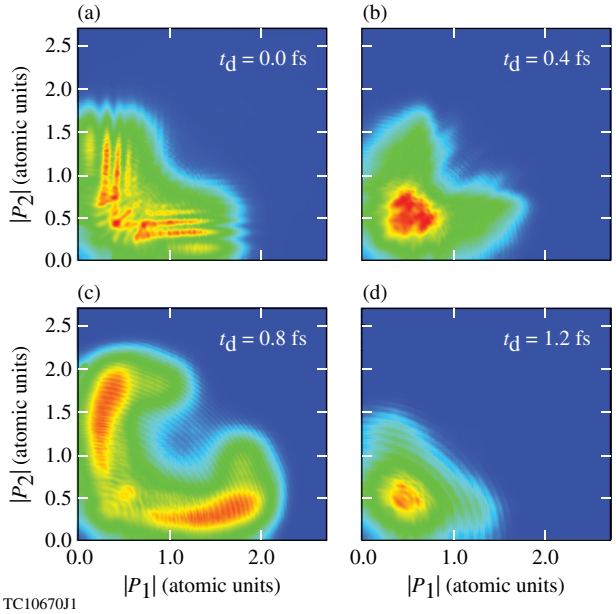

FIG. 3 (color online). The double-ionization momentum distribution for the cases shown in Fig. 2.

to the generation of energetic electrons. The fast-moving electron has energies over ~54 eV ($p \geq 2$ a.u.), which is even higher than the maximum energy ($3.2\,U_p \simeq 36$ eV) of the returning electron. If the returning electron knocks out only the $2p$ electron ($I_p \simeq -13.6$ eV) *without* any additional photoabsorption from the IR field, the freed two electrons may share only the excess energy of ~22 eV. Taking into account the additional "momentum kick" by the dressing IR field at the moment when the two electrons are freed, for example at $t_d = 0.8$ fs [note that the instantaneous IR-laser fields become smaller than its peak value shown in Fig. 1(a)], the classical energy gain for each electron is about ~8 eV at maximum. Counting all of these, the total energy of the two electrons can reach a value of only ~38 eV, which is still significantly less than the observed ~60–70 eV plateau. Therefore, the energetic electron generation indicates the excessive absorption of IR photons during the recollision as a result of the controlled, strong e-e correlation. Next we assume that each electron equally absorbs a maximum photoenergy of ~18 eV from the IR field during the recollision (including the classical momentum kick discussed above); the excited $2p$ electron will then end up with an outgoing energy of +4.4 eV (= −13.6 + 18 eV), while the recolliding electron will be boosted to a maximum energy of +54 eV (= $3.2\,U_p$ + 18 eV). This corresponds exactly to the case shown in Fig. 3(c), and the same amount of energy gain from the IR field by the two electrons indicates the strong electron-electron correlation. For other time delays shown in Fig. 3, the excitation of the $2p$ electron is not occurring at the instant of recollision. The correlation between the tunneled electron and the "target" $2p$ electron of He^+ is not very strong, causing fewer photons to be involved; therefore, energetic electrons have not been seen for these time delays.

To further explore the strong-correlation-induced excessive photoabsorption, we have plotted the total energy spectra of the attosecond-controlled NSDI in Fig. 4 by scanning the time delays. It is clearly seen that when the strong e-e correlations are turned on at $t_d = -0.4$ fs [Fig. 4(c)] and $t_d = +0.8$ fs [Fig. 4(g)], a plateau appears in the NSDI sprectra in which the almost-constant probability extends up to high energies of $E_1 + E_2 \simeq 70$ eV. Such a plateau vanishes and reappears in between the two time delays, indicated by Figs. 4(c)–4(g). The plateau feature has been seen in the highly nonlinear, nonperturbative single ionization of atoms in strong fields. Here, the observation of a NSDI plateau, from a true two-active-electron dynamics, manifests the nonperturbative nature of the attosecond-controlled electron-electron interaction, exposed to the external IR laser field.

Figure 5 plots the double-ionization probability near the plateau's cutoff region, e.g., at the total electron energy of $E_1 + E_2 = 60$ eV, as a function of the time delay. The probability increase by an order of magnitude has been seen when t_d is coincident with the two recollision instants

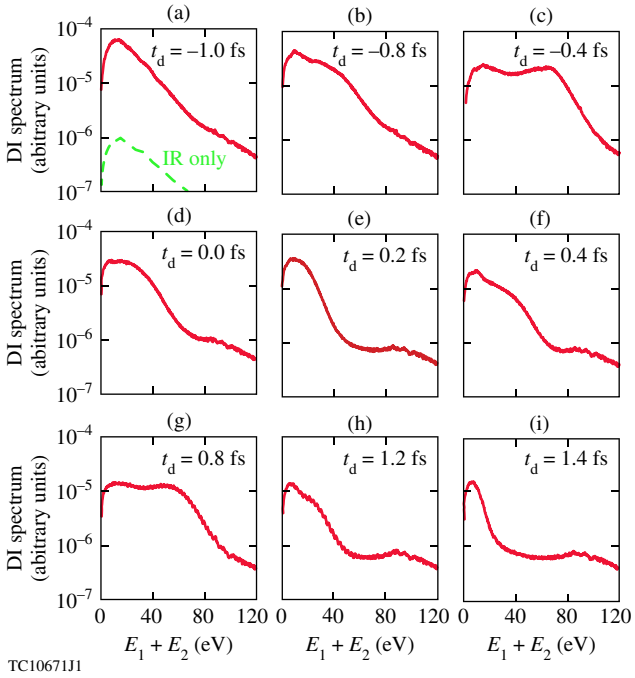

FIG. 4 (color online). The energy spectrum of double-ionized electrons for different time delays: (a) $t_d = -1.0$ fs, (b) $t_d = -0.8$ fs, (c) $t_d = -0.4$ fs, (d) $t_d = 0.0$ fs, (e) $t_d = 0.2$ fs, (f) $t_d = 0.4$ fs, (g) $t_d = 0.8$ fs, (h) $t_d = 1.2$ fs, and (i) $t_d = 1.4$ fs. The appearance of the "plateau" manifests the highly nonlinear and nonperturbative nature of the attosecond controlled nonsequential double ionization (NSDI). The green dashed line in (a) represents the IR-only case.

of −0.4 and +0.8 fs. The control of strong electron-electron correlation can significantly boost photo-absorption in the NSDI process. The two enhancements are separated exactly by half an cycle of the IR field as expected. Such a subcycle dynamics has also been observed in attosecond transient absorption measurements of IR-dressed He [50].

In summary, we have performed full-dimensional, *ab initio* investigations on the attosecond-controlled

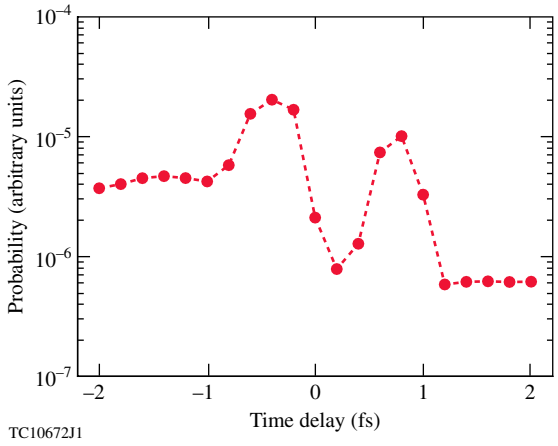

FIG. 5 (color online). The probability of double ionization with energy at $E_1 + E_2 = 60$ eV is plotted as a function of the relative time delay. The measurable double peaks indicate the exact time recollision occurs.

NSDI of He, exposed to both an IR-FCP ($\lambda = 780$ nm) and a time-delayed attosecond EUV pulse. It has been shown that a strong e-e correlation, turned on by the properly timed attosecond pulse, can significantly enhance the photoabsorption from the dressing IR field during the recollisions. A plateau feature appears in the total energy spectra for such highly nonlinear and nonperturbative NSDI process. These results shed light on understanding the essential role that a strong e-e correlation plays in the excessive photoabsorption observed in multielectron systems exposed to external intense fields.

This material is based upon work supported by the Department of Energy National Nuclear Security Administration under Award No. DE-NA0001944, the University of Rochester, and the New York State Energy Research and Development Authority. The support of DOE does not constitute an endorsement by DOE of the views expressed in this article. The author gratefully acknowledges the support by the National Science Foundation (NSF) under the NSF-TeraGrid Grant No. PHY110009. This work was conducted utilizing the NICS' Kraken Supercomputer with more than 4000 cores per run.

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