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## Applications of Short Laser Pulses

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### 1. Introduction

Even though the phrase “short pulse” has been in widespread use for more than two decades, there has been no indication of serious attempts to provide a clear limit on pulse duration beyond which the pulse is no more considered to be short. Unless this ambiguity is resolved devising applications peculiar to the short pulses will be prohibitively difficult as it has been the case over past years despite significant advances in untangling the phenomena arising from the interaction of laser pulses with matter. The parameter space determining the outcome of such interactions, which include wavelength, pulse duration, peak intensity, polarization state, density of the matter, and the energy levels of the molecular constituents of the matter, is too enormous to allow an efficient design appropriate for providing performances levels superior to what can be achieved using traditional techniques. Moreover, the parameter space becomes more complex when two or more temporally overlapping pulses of different wavelengths are desired to be applied for providing more flexibility. Thus, we are well justified to devote a whole section for the definition of “short pulse” in the context of the potential applications that are expected to emerge in coming years. Then, two application areas, which we consider to be promising despite being scarcely discussed in academic literature, will be discussed in some details. These include laser assisted high specific mass spectroscopy and flat panel display dicing.

### 2. Relevancy of applications and definition of “short pulse”

In the introduction we used the phrase “relevant application” without clearly specifying why the adjective relevant needed to be used in the first place. Unfortunately, there are many claims in the literature on the applications of short laser pulses while few, if any, of these claims live up to markets’ expectations and soon sink into oblivion. Therefore, we believe that the relevant applications are much fewer than the large list one can compile by gathering those suggested by the researchers in universities. We support our assertion by providing as an example related to one of our recent experiences. Someone approached us for advice on a flat transparent thin sheet of inert polymer having mono-dispersed pores for application in health science to remove bacteria from biological fluids. The pores were desired to have diameters of  $0.4\ \mu\text{m}$  with a pore density of about  $10^8$  pores per  $\text{cm}^2$ . Obviously, such pores cannot be drilled using commercially available high repetition rate lasers since the pore diameter is smaller than their wavelength. However, recalling the published work on drilling to sub-wavelength diameters we suggested using the high repetition 100 fs Ti:Sapphire laser pulses as a suitable and feasible method. To our dismay, few days later, we learnt of commercially available

polycarbonate membrane filters with well characterized characteristics having list prices as low as 0.2 \$/cm<sup>2</sup>. Unfortunately, achieving similar levels of perfection and cost does not appear to be practical with short laser pulses; the number of the pores is too large to have a wide area filter to be manufactured in a relatively short time. Thus, as usual the merits of chemistry based fabrication techniques are easily manifested and the economical realities will dictate their selection.

We are aware that cases similar to the aforementioned example abound in technology transfer offices of the universities. The announcement of a new application may generate excitement in research circles and provide a new hot topic for the researchers looking for satisfaction by addressing real world problems and justifying large funds that are spend for conducting application oriented researches. Then, at the first glimpse of seemingly new observation patents are files with the hope that soon they will be licensed by some high tech firm. This approach suffers from the lack of locating the invention on a detailed development landscape. Such a map, often, is prepared at the lunch of development phase at the established industries by evaluating the survival potential of the invention encounter with the harsh realities of the markets.

In table 1 we provide a list of topics which were considered as breakthroughs for a while. For each item the most plausible reason for its final demise is briefly mentioned. The list is

The suggested application of short pulses	The reason for failing the relevancy criterion
Using the short laser pulses as a replacement to the mechanical drills in dentistry to avoid the transfer of AIDS virus from one patient to the other.	The early overwhelming AIDS scare subsided when the clinical reports did not support the prevalence of viral infections through dentistry tools. In addition, a user friendly laser system was not introduced into the market, perhaps, due to the difficulties in justifying the development costs.
Writing FBG's	The dotcom crash of late nineties nearly dried up the market for the FBGs. Remaining small market is very well served by the installations utilizing UV lasers equipped with phase masks, which enable the manufacturing of high performance FBGs.
Pattern carving on wooden structure without leaving behind burn traces.	The short pulses are not able to efficiently carve out deep enough as required by the industry. It is not clear if there is any demand for shallow and colourless patterns.
Using filaments generated by the short pulses for remote sensing of target molecules in atmosphere.	Efficient and selective detection has not been validated by data collected in the field.
Using filaments generated for lightening control.	Reviewing the published results does not support the claims that the clouds can indeed be discharged by filaments.
Continuum free spectroscopy of gases	Despite providing clean spectra with very low continuum noise, the method lacked enough specifity required for discrimination of molecules having similar structures.

Table 1. A short list of suggested applications for short pulses that could not live up to their initial promises.

very short and only presents few items that the author had been personally involved. In order to do justice to those who initiated these topics, we should add that applying the relevancy criterion, i.e. examining the proposed application with scrutiny, is not easy at the time of announcements due to the lack of required feedbacks from the intended end users. Returning to the challenge of defining “short pulse”, we select the multiphoton/tunnelling ionization of Xe interacting with 200 fs Ti:Sapphire laser pulses reported by Larochelle et al (1998). They measured the ion yield versus peak intensity curve, the laser was focused using f#100 optics into an UHV chamber having a background pressure of  $2 \times 10^{-9}$  Torr. Ion species were separated with a time-of-flight mass spectrometer. Ion curve was produced by combining a series of intensity scans, each having a different fill pressure in the interaction chamber. The experimentally measured ion yield versus intensity curves were compared with that predicted by the PPT model. The results are presented in figure 1.

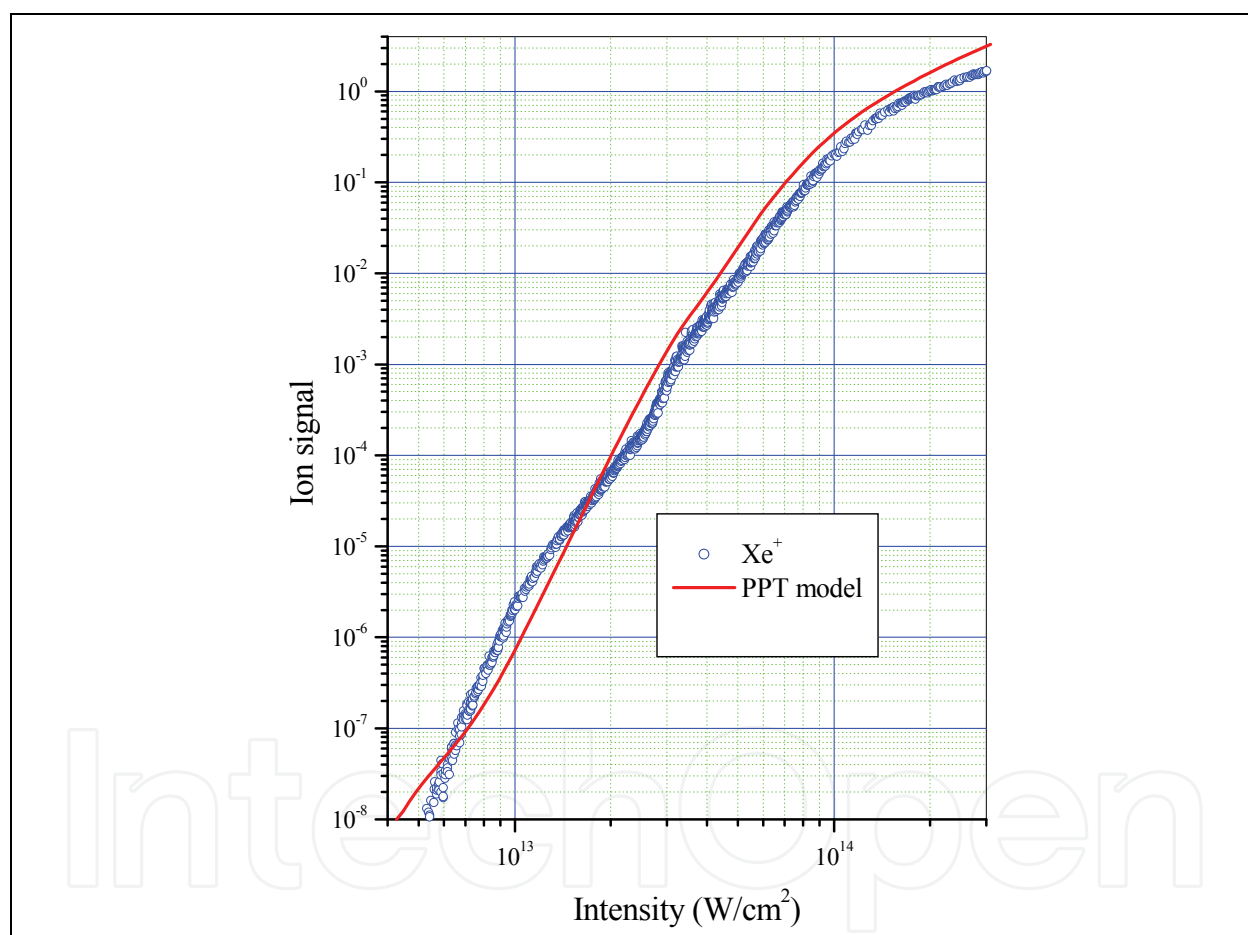


Fig. 1. Ion versus peak intensity plot for Xe interacting with linearly polarized laser pulses from a Ti:Sapphire laser (800 nm). Each datum corresponds to a three point average. The theoretical ion yield is calculated using PPT model.

The derivation of the PPT model proceeds as following (Talebpour 1998a). By finding the exact solution of the time dependent Schrodinger equation for a potential  $V = \sqrt{2E_i} \delta(\mathbf{r})$  ( $E_i$  is the magnitude of the ionization potential of the atom), the probability current  $\mathbf{J}(\mathbf{r}, t)$  is calculated. Then, the total ionization rate from short range potential for linear polarization,  $W$ , is found from

$$W = \left( \frac{\omega}{\pi} \right) \lim_{x \rightarrow \infty} \int_0^{2\pi/\omega} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} J_x(r, t) dy dz dt$$

where  $\omega$  is the frequency of the laser, which is assumed to be polarized in the direction of the x axis. The effect of the ionic potential, which behaves like  $Z/r$  ( $Z$  is the charge of atomic core) at a long distance from the nucleus, is calculated through first order correction on the semi-classical action. Accordingly, the PPT model simply disregards the excited energy levels of the atom. The overall fit between the model and the measured ion signal is amazingly good. However, the measured signal exhibits unexpected slope changes at intensities less than  $3 \times 10^{13}$  W/cm<sup>2</sup>. These are believed to indicate the involvement of multiphoton resonances of some excited energy levels with the ground state followed by Lambda type trapping. The mechanism of the lambda type trapping is schematically presented in figure 2. At the rising part of the pulse (a) the excited state (with two neighboring levels 1 and 2 which are degenerate within the laser bandwidth) are not in multiphoton resonance with the ground state. The electron is ionized through multiphoton coupling with the continuum. As the intensity of the pulse is increased the excited state and the continuum are shifted in energy due to the Stark shift. At the peak of the pulse (b) the excited states go into multiphoton resonance with the ground state and most of the population is transferred from the ground state to the excited states 1. Coupling starts instantaneously and continues on through the rest of the pulse. As the intensity starts to decrease (c), the two states are coupled through continuum and the population is trapped in a coherent superposition of the two states.

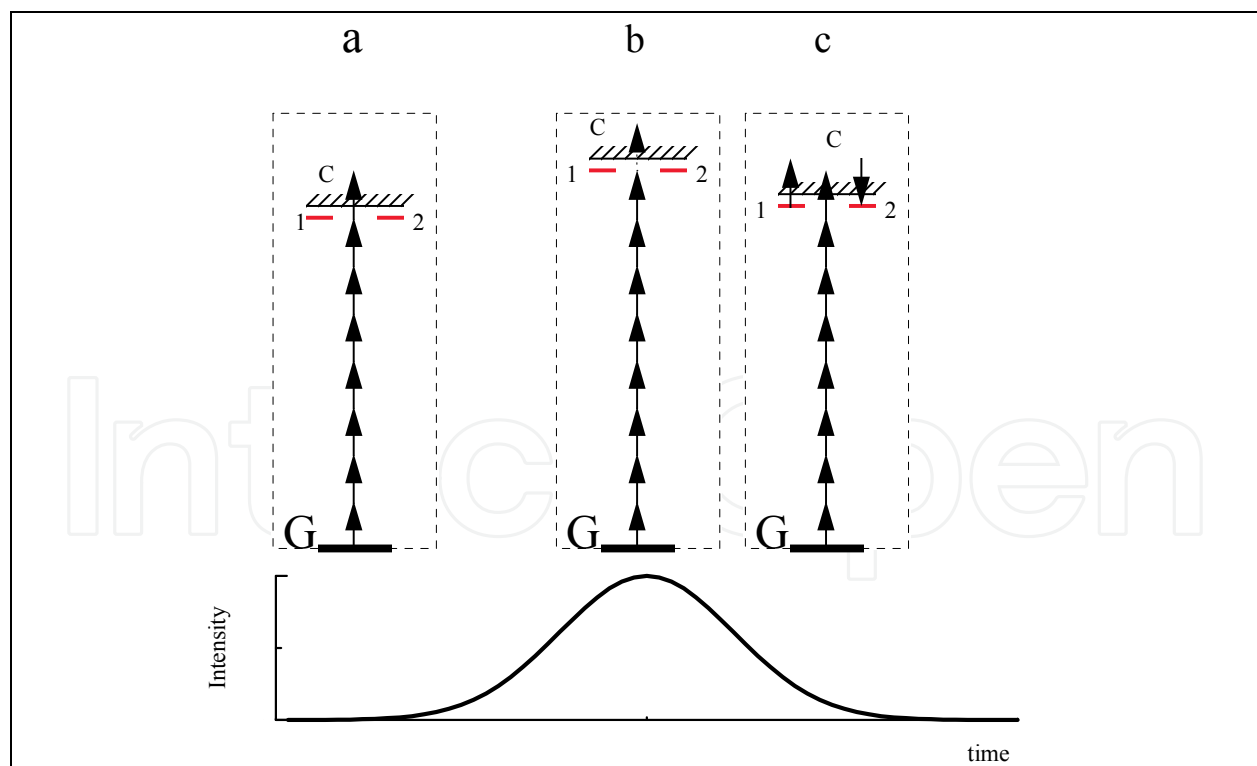


Fig. 2. The schematic presentation of lambda type population trapping. G is the ground state of the atom. 1 and 2 are two degenerate excited states. After the population is transferred to the states due to multiphoton resonance, these states are coupled through continuum c and the population is trapped in the superposition of these states.

If the multiphoton resonance had not been occurred at the peak of the pulse, the population transfer to the excited state would not be appreciable. Within the dressed atom picture, the ground state dressed by  $m$  photons and the resonant state undergo an avoided crossing at the resonance intensity  $I_r$ . The minimum distance,  $V_m$ , at the avoided crossing is proportional to the generalized Rabi frequency  $\Omega(t) = \Omega_m I(t)^{m/2}$  coupling the two states. According to Story *et al* (1994), the probability of remaining in the ground state,  $P_g$ , is given by  $P_g = \exp\left(-\frac{2\pi V_m^2}{dW/dt}\right)$ , where  $W$  is the time-dependent energy difference between the two dressed states. In interaction with a short pulse, if the dynamic resonance is reached in the rising or the falling part of the pulse, the population practically remains in the ground state and the effect of multiphoton resonances may be neglected. However, if the states go onto resonance at the peak of the pulse, where  $dW/dt$  is zero, then the excited state is populated. After being populated, since the ionization potential of the excited state is small, it is expected that the electron will be instantly ionized or subsequent interaction with field stabilize the atom against ionization.

Now we offer our definition of short pulse for the cases where the density of the interacting matter is very low.

*A pulse interacting with a dilute gas is considered to be short if the rule of excited atomic or molecular levels in the outcome of the interaction phenomenon under consideration can be ignored.*

According to our definition, in the case of the multiphoton ionization of Xe (corresponding to the data of figure 1) the pulses are in the brink of being considered short. The data of Larochelle *et al* (1998) indicate that ion versus peak intensity curves of rare gas atoms with high ionization potentials, such as Ar, do not have noticeable slope changes. Therefore, for the multiphoton ionization of Ar interacting with Ti:Sapphire laser pulses 200 fs pulse duration is short.

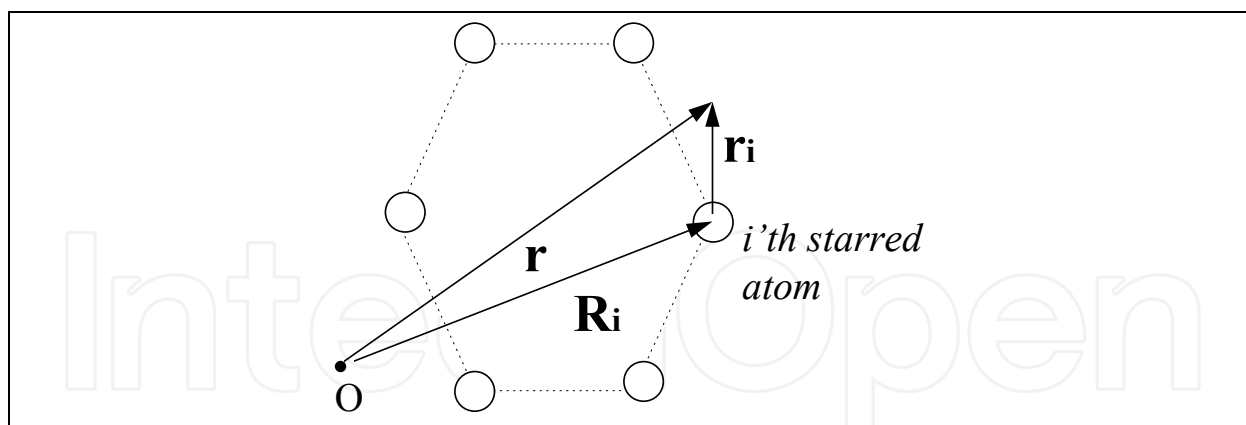


Fig. 3. The schematic diagram of an unsaturated hydrocarbon. The starred atoms are presented by circles. Note that the molecule contains other atoms like H which in Huckel's approximation are neglected. O is the origin of coordinate system.

The hydrocarbon molecules, whose interactions with lasers may be more important, have ionization potentials that are less than the ionization potential of Xe. However, a pulse with duration of 200 fs can be safely taken as short for the following reason. We consider an unsaturated hydrocarbon, such as benzene, which is schematically presented in figure 3. As it is well known from organic chemistry, using Huckel's theory, the normalized wavefunction of the active electron in the molecule can be approximately represented as



$$\Psi(r) = \sum_i c_i \psi_i(r_i)$$

where  $c_i \psi_i(\mathbf{r}_i)$  is the contribution of the  $p_z$  electron wavefunction of the  $i$ 'th carbon atom (starred atom in the nomenclature of organic chemistry) situated at point  $\mathbf{R}_i$  and  $c_i$ 's are constants ( $\sum |c_i|^2 = 1$ ), determined by variationally minimizing the total energy of the molecule. The variable  $\mathbf{r}_i$  is the position vector in a coordinate system with origin at  $\mathbf{R}_i$ . A schematic definition of different variable are presented in fig. 3. Talebpour *et al* (1998b) have argued that the ionization amplitude can be thought as being a linear superposition of the ionization amplitudes of individual starred atoms. The MPI process in an unsaturated hydrocarbon might be considered as the emission of electron waves from multiple centers. At any observation point in space these waves interfere and, depending on their relative phase, give rise to constructive or destructive interference. The result is that the ionization of the unsaturated hydrocarbons are noticeably suppressed and the ionization process instead of occurring the pure multiphoton channel, by absorbing multiple photons, proceed through tunnel ionization for which the intermediate resonances play no rule in the electron detachment process. In order to verify our claim, we have presented the ion signal versus peak intensity of benzene in figure 4. Clearly there are no observable kinks, reminiscent of the involvement of the excited energy levels in the process, on the ion curve. Therefore, the pulses with wavelength of 800 nm and duration of 200 fs that interact with unsaturated hydrocarbons are short based on our definition.

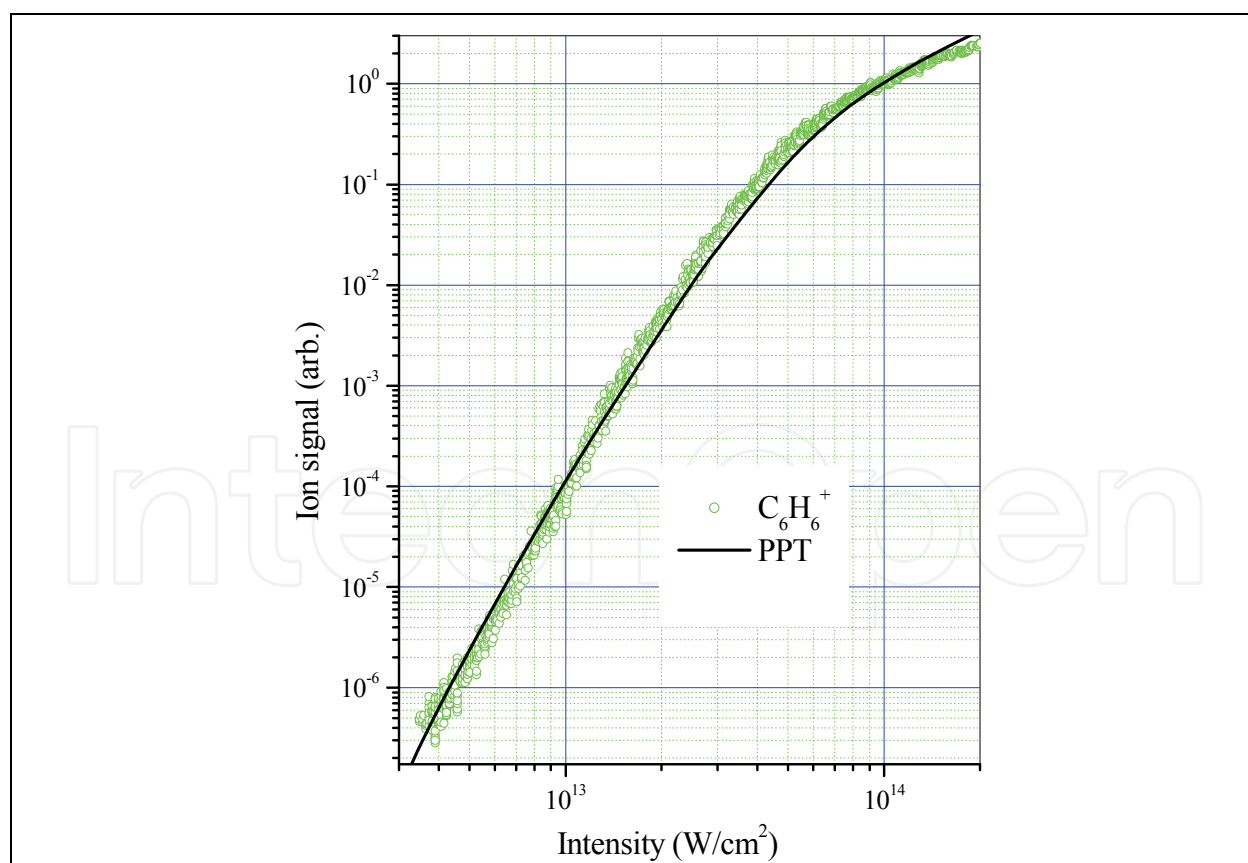


Fig. 4. Ion versus peak intensity plot for benzene interacting with linearly polarized laser pulses from a Ti:Sapphire laser (800 nm). Each datum corresponds to a three point average. The theoretical ion yield is calculated using PPT model.

In the case of interaction with dense media another definition for short pulse may appear to be necessary. Following ionization the electron is being accelerated by inverse Bremsstrahlung and can collide with the nearby molecules and generate new electrons through collisions. If the pulse duration is long, the newly ionized electrons can be accelerated and eventually avalanche or cascade ionization follows. Once the density of the electrons reached a critical value breakdown occurs and high density plasma is created which has no memory of the laser pulse. So, the criterion for the shortness of a pulse in dense media is as the following:

*A pulse interacting with a dense matter is considered to be short if during the interaction the threshold for the avalanche ionization is not reached.*

At the first glance this definition may appear to be too limiting. Fortunately, due to the delicately balanced behavior of the pulses in dense media the threshold does not reached easily. The phenomenon responsible for the balance is the well known filamentation process during the propagation of strong laser pulses in dense media. The vast amount of studies have resulted in the emergence of a qualitative understanding, which describes the phenomenon as being the manifestation of a self-tuning between two competing processes; the nonlinear self focusing of the pulses and the multiphoton ionization. The electrons are exclusively created through the MPI in the leading edge of the pulse. As time passes, the increase in the energy of the pulse cannot lead to an appreciable increase in the density of the electrons, since the defocusing effects caused by the plasma and by the high order nonlinearities decrease the local intensity and thus stop the generation of more electrons. Talebpour *et al* (2000a) have estimated that, in the case of propagation in atmospheric pressure D<sub>2</sub> gas the plasma density does not reach above 10<sup>-5</sup> times the gas density. This is the reason why the strength of the light contributed by the continuum of radiation is so low in the visible spectrum of air monitored from the side of filament. Moreover the characteristic atomic lines resulting from the breakdown induced fragmentation of air molecules are missing. These statements can be easily verified by referring to figure 5, which presents the spectrum of the atmospheric pressure air interacting with 200 fs, 800 nm laser pulses.

### 3. Examples of potential applications for the short laser pulses

In this section we will discuss two areas that short pulses may provide relevant applications. These are highly specific mass spectroscopy of molecules and flat panel display dicing. In the first case the basic physics has been very well developed by the contributions from multitude of research groups all over the world. What remain are intensive development efforts preferably supported by industry to resolve the issues related to robust instrumentations. We are aware that the second case is already being introduced in electronic industry particularly in south Korea.

In what follows the scientific background of the fragmentation of molecules, as related to the mass spectroscopy will be reviewed with sufficient detail. The second application will be briefly described by referring to a recent PCT patent application by Kamata *et al* (2008). Due to the lack of information on marketing front, we are not able to construct a solid case to unambiguously demonstrate the relevance of the suggested applications areas. The future events will prove whether or not our speculations are sound.



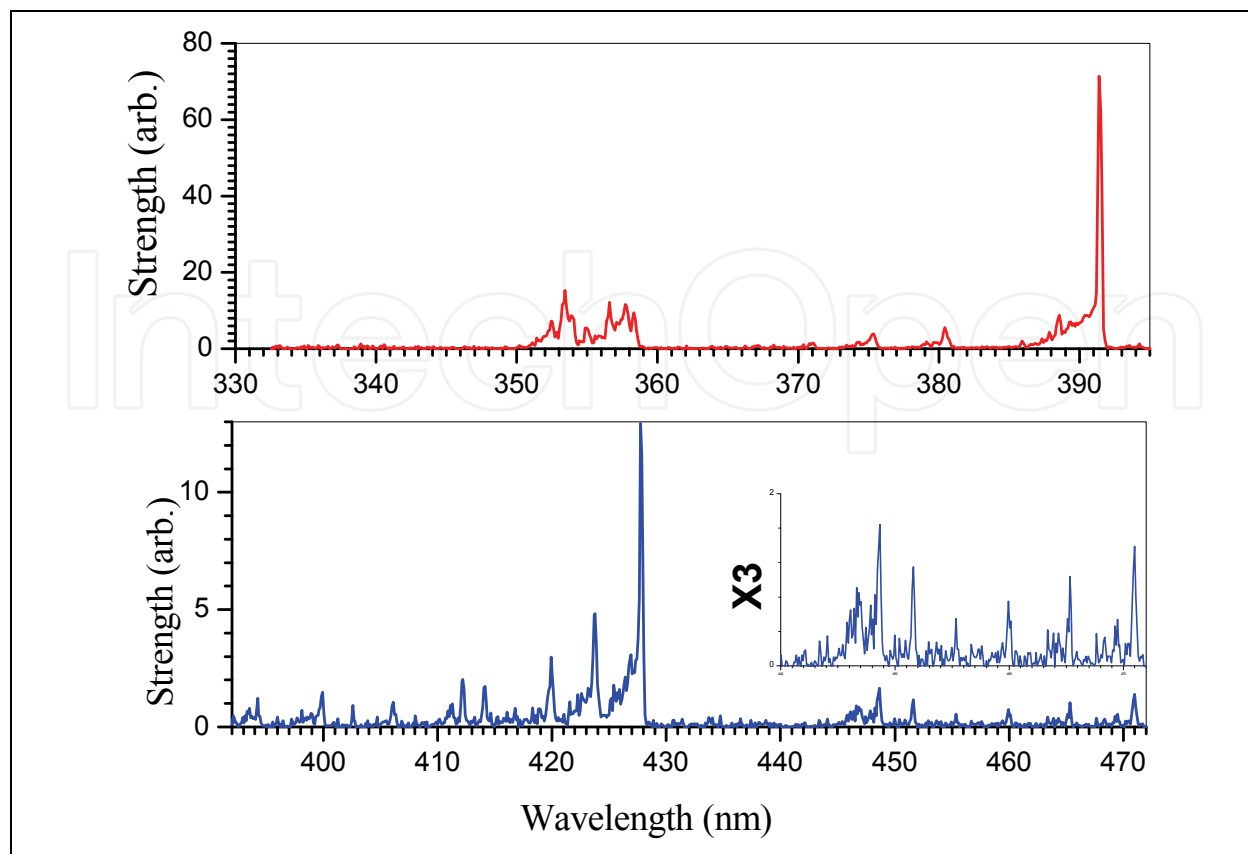


Fig. 5. The visible spectrum of air interacting with linearly polarized Ti:Sapphire pulses having a duration of 200 fs.

### 3.1 Potential application of short laser pulses as an ion source for high performance mass spectroscopy

#### 3.1.1 Single pulse

The discussions of the previous section indicated that according to the criteria put forward by our definitions, pulses with durations of less than 200 fs are considered to be short. We do not set any restriction on the laser wavelength as the wavelengths of the commercially available lasers and their second harmonics are more or less in the 400-1200 nm range. It is true that all multiphoton process are wavelength dependent but such dependencies are not expected to result in failing the two aforementioned “shortness” characteristics of the pulses. Therefore, in the present subsection, except one case, the discussions will be restricted to the case of 200 fs pulses delivered by a Ti:Sapphire laser that has a wavelength of 800 nm.

We start by reviewing the mass spectra of Cyclohexane that have been prepared at some selected laser intensities (see figure 6). The experiment employed a commercial laser system that consists of a Ti:Sapphire oscillator followed by a regenerative and two multi-pass Ti:Sapphire amplifiers that can deliver pulses with energies of up to 100 mJ. The pulse duration is 200 fs (FWHM) and the central wavelength  $\lambda$  is 800 nm. The laser pulses were focused using  $f/100$  optics (1 m focal length lens and a beam diameter of 1 cm) into an ultra-high vacuum chamber having a background pressure of  $2 \times 10^{-9}$  Torr. The liquid was introduced into the vacuum chamber through a needle valve and the ions resulting from the dissociative ionization of the target molecule are extracted into a time-of-flight (TOF) mass

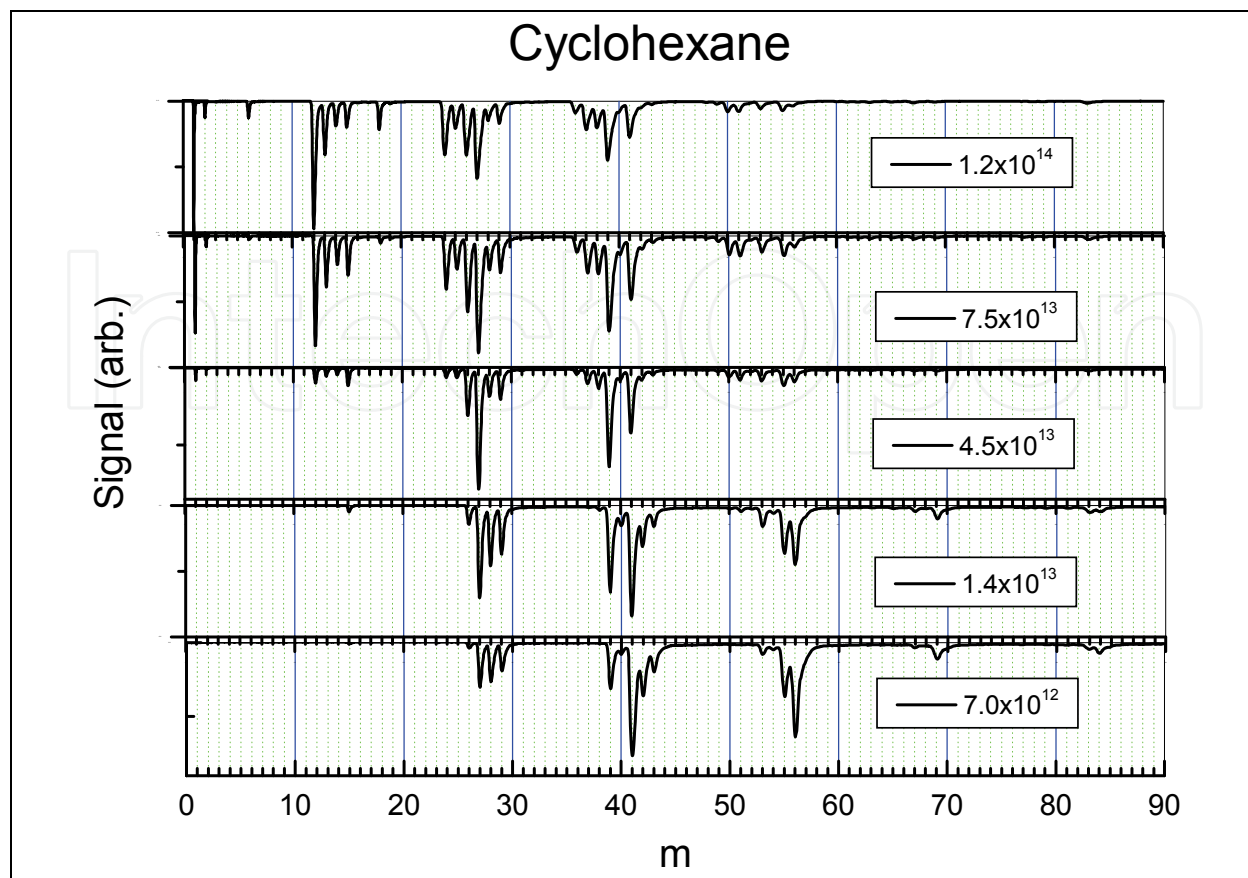


Fig. 6. The mass spectra of Cyclohexane interacting with linearly polarized Ti :Sapphire pulses having a duration of 200 fs.

spectrometer having a 60 cm long drift tube and detected with an electron multiplier in conjunction with a digital storage oscilloscope. The large  $f$ -number focusing lens was used to maximize the focal volume of the interaction region so as to obtain ion signals from very low ionization rates. The gas pressure in the interaction chamber was controlled by a precision and set at  $10^{-7}$  Torr. The laser intensity was determined by measuring the laser energy, the pulse width and the focal spot size. The energy measurement was taken from a 4% reflection off a wedged beam-splitter situated after the interaction region and was detected with a pyroelectric energy meter. The pulse width was measured by a second-order autocorrelator. To measure the spot size, the beam was attenuated by successive reflections from high-quality glass plates. The beam was then focused by the 100 cm focal length lens and the image of the focused area was analyzed by a microscopic objective with an associated CCD camera. The beam had a nice Gaussian profile and a roundness of 0.95. The focal spot diameter (at  $1/e^2$ ) was found to be  $100\ \mu\text{m}$ . Using this information the peak laser intensity was calculated and recorded on the corresponding spectrum.

When the molecule was exposed to 1 ns pulses from a  $\text{CO}_2$  laser the mass spectrum only possessed  $\text{H}^+$  and  $\text{C}^+$  peaks. Thus, the main characteristic peculiar to the mass spectra obtained using short pulses is the richness of the spectrum, as any peak expected from fragmentation of the molecule through known dissociation channels can be observed by varying the laser intensity in a  $10\times$  intensity interval. In this respect the short pulse ion source is superior to the well established electron beam based ion sources, a fact that can be easily verified by referring to figure 7.

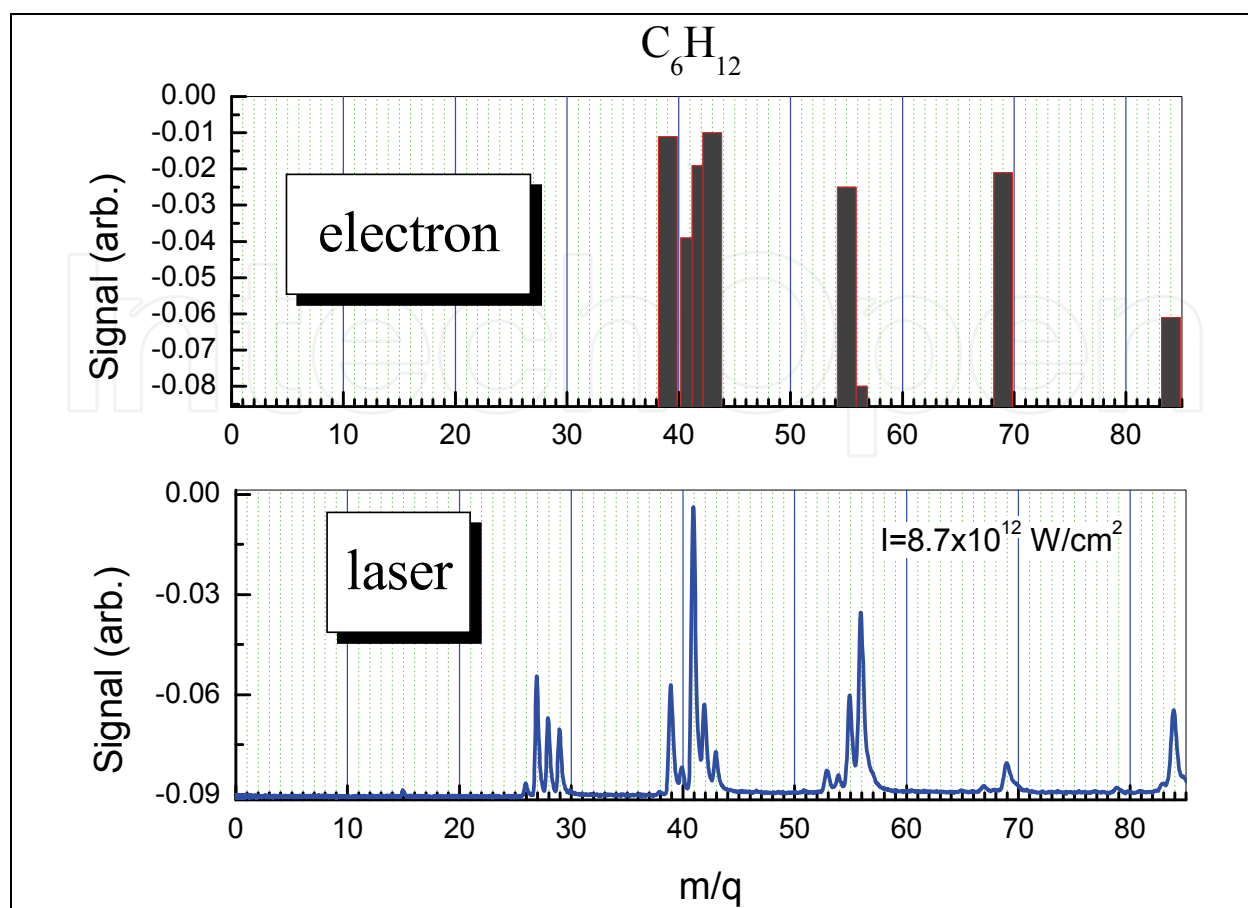


Fig. 7. The mass spectra of Cyclohexane interacting with linearly polarized Ti:Sapphire pulses having a duration of 200 fs.

The mechanism responsible for the fragmentation of polyatomic molecules interacting with short laser pulses was developed by Talebpour *et al* (2000b). Their model describes the short pulse induced fragmentation of molecules as happening in three steps:

- An electron ionizes from either the outer or one of the inner orbitals of the molecule this generates a molecular ion. If the ionized electron had originated from an inner orbital the ion will be left in ro-vibrational levels of an excited electronic state;
- The excited ion makes a rapid radiationless transition to the high-lying ro-vibrational levels of a lower electronic state; and
- Then the ion dissociates to different fragments through various fragmentation channels. 100  $\mu\text{m}$ .

The importance of the short duration of the pulse is that due to very high intensity the ionization steps occur rapidly without involving the intermediate excited states. Then, the pulse ceases before appreciably perturbing the geometry of the excited ion. So, the task of the pulse is rapidly ionizing the molecule by coupling the ground state of an electron to the continuum and then leaving the resulting ion alone to follow the path to fragmentation. A long duration pulse stays too long and heavily involves the intermediate states in the ionization process. In addition, there is the possibility that the molecule undergoes dissociation before being ionized. Moreover, the pulse survives so long that the fragments interact with it and undergo their own fragmentation route. This explains why in the case of using 1 ns  $\text{CO}_2$  laser pulses the molecule dissociates down to its constituent elements.

Now we are in a position to address the main aspect of the short pulse induced molecular fragmentation, i.e. the higher specificity compared to the other ion sources, through experimental studies. The intention is to show that the selectivity provided by a short pulse based source is superior to that expected when using the conventional electron ionization based sources. The distinction becomes important when the simultaneous analysis of mixtures containing multiple isomers is required. Let us take, as an example, the mass spectra of butane isomers studied by Bennett and Cook (200). Visual inspection of the reported results indicates that the differences between the mass spectra of different isomers are not sufficient for accurately quantifying the partial concentrations of the isomers in a mixture. Any source capable of increasing the difference would improve the accuracy by which the concentration of each isomer is determined. In order to verify if the short pulse based ion source provides enhanced performance Sharifi et al (2008) studied the fragmentation of 1-butene and cis-2-butene, whose structures are presented in figure 8) interacting with femtosecond Ti:Sapphire laser pulses. The resulting fragmentation patterns were compared for the two isomers and it was shown that their difference, as quantified by the spectral similarity index (SI), was more pronounced than the case of using 100 eV electrons as ionization/dissociation source. The experimental setup and conditions were similar to what was mentioned above in connection to the mass spectra of Cyclohexane.

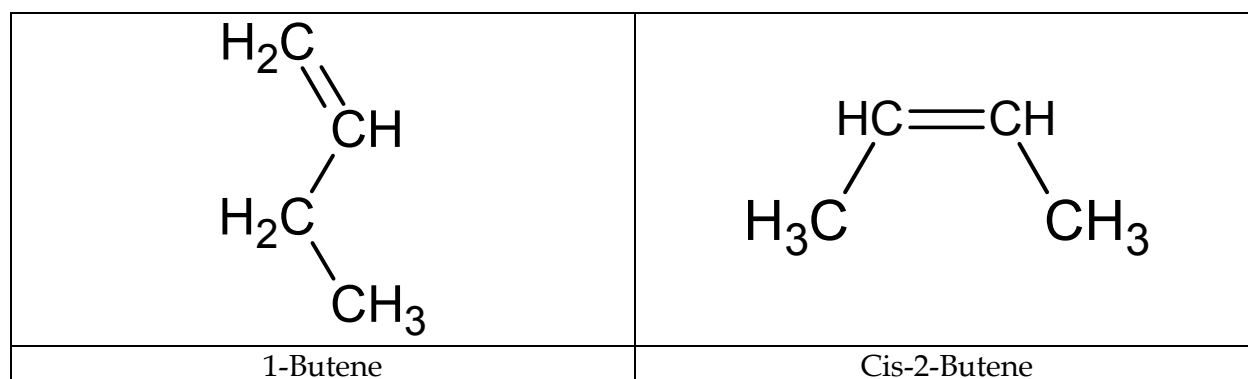


Fig. 8. The structure of 1-Butene and Cis-2-Butene.

The mass spectra of 1-butene and cis-2-butene were recorded at two different peak laser intensities and presented in figures 9 and 10. In both figures the signal corresponding to singly charged ion of cis-2-butene, at the mass to charge ratio of  $m/q=56$ , has been scaled to an arbitrary value of 100 and the peaks corresponding to other ions have been accordingly scaled. As it is observed at both peak intensities the difference in the spectra of the isomers are more pronounced than the cases of using electron impact ionization of reference.

In order to quantify the performance of the new ion source in terms of determining the composition of a gas containing the two isomers, we calculate the similarity index (SI), which is defined as,

$$SI \equiv \left( \sum_{\frac{m}{q}} \sqrt{I_x I_y} \right)^2 / \left( \sum_{\frac{m}{q}} I_x \sum_{\frac{m}{q}} I_y \right)$$

where x and y denote the two isomers, m and q are the mass and charges of a given ion, and  $I_x$  and  $I_y$  are the relative abundances of peaks for equal value of  $m/q$  on 1-butene or cis-2-

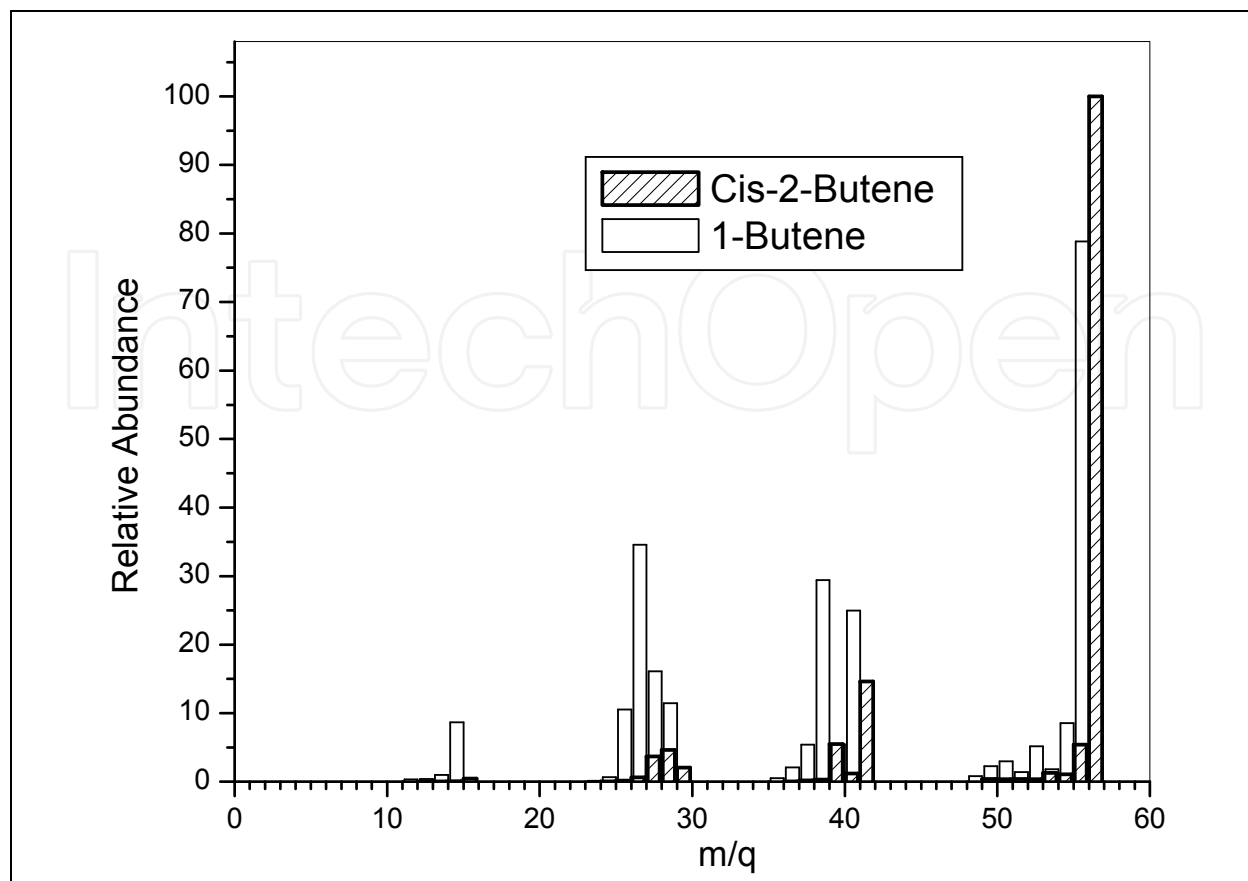


Fig. 9. Comparison of mass spectra for Cis-2-Butene and 1-Butene prepared by exposing pure gases to linearly polarized short laser pulses focused to peak intensity of  $2 \times 10^{13} \text{ W/cm}^2$  (SI=0.793).

butene mass spectra. The SI values were calculated to be SI=0.793 and SI=0.918 for the spectra presented in figures 2 and 3, respectively. The corresponding value reported in reference, in the case of using electron source for ionization, is SI=0.9957. This observation readily indicates that the dissociative ionization by ultra-fast laser pulses can potentially provide a high performance ion source for applications in mass spectroscopy. In addition, the dependence of the SI on the laser peak intensity could provide a practical optimization tool. We elaborate on these observations in the following section.

The peak laser intensity is the main parameter that can be used to optimize the dissociative multiphoton ionization as a source for high performance mass spectroscopy. This is due to the action of two well established phenomena; increase in ionization potential resulting from AC Stark shift and the field induced barrier suppression. Let us denote the appearance potential of a particular fragment by AP, which is defined as the minimum energy needed to form the fragment from a neutral molecule in the ground state. AP can be expressed as  $AP = E_i + \Delta E$ , where  $E_i$  is one of the ionization potentials (from the inner valence orbital) and  $\Delta E$  is the minimum energy above that ionization potential needed to form the ion. At the peak laser intensity,  $I_0$ ,  $E_i$  increases by the so-called ponderomotive potential  $Up_{(ev)} = 9.33 \times 10^{-14} I_{0(W/cm^2)} \lambda^2_{(\mu m)}$ , where  $\lambda$  is the wavelength of laser. The ponderomotive potential is the average energy of the oscillatory motion of a free electron in an electromagnetic field and explicitly appears in analytical formula for the multiphoton

ionization of atoms with applying the strong field approximation. In contrast to the ionization potential,  $\Delta E$  decreases by an amount  $\delta E$  due to field induced barrier suppression. The magnitude of  $\delta E$  is estimated to be given by  $\delta E \approx 0.5FR$ , where  $F$  is the strength of the laser field and  $R$  is the size of the parent ion. For example at a peak intensity of  $2 \times 10^{13} \text{ W/cm}^2$  and for an ion with  $R=2$  atomic unit,  $\delta E$  is calculated to be 0.62 eV.

The aforementioned arguments can be applied to qualitatively explain the dependence of SI on the peak laser intensity. For this purpose we define partial SI by excluding the signal of the parent ion from the summations in equation 1. This quantity is calculated to be  $SI=0.868$  and  $SI=0.967$  at low and high intensities, respectively. The relatively elevated value of the partial SI indicates that at higher intensities the fragmentation patterns of the two isomers are more similar. This can be explained by noting that the quantity  $\delta E$  at higher intensity ( $6.5 \times 10^{13} \text{ W/cm}^2$ ) is higher than its value at the low intensity ( $2 \times 10^{13} \text{ W/cm}^2$ ) by a factor of 1.8. Consequently, most of the parent ions resulting from the multiphoton/tunnel ionization of inner valence electrons will indiscriminately dissociate at higher intensities resulting in a mass spectrum that is independent of the differences in the geometries of the isomers. One might conclude that lower intensities are preferable for giving rise to smaller partial SI. However, we should remember that the ion signal decrease rapidly with lowering the intensity and the potential gain due to reduced SI might be compromised by the detection limit of the mass spectrometer. Therefore, a balance between (partial) SI and the magnitude of ion signal would lead to optimum ion source.

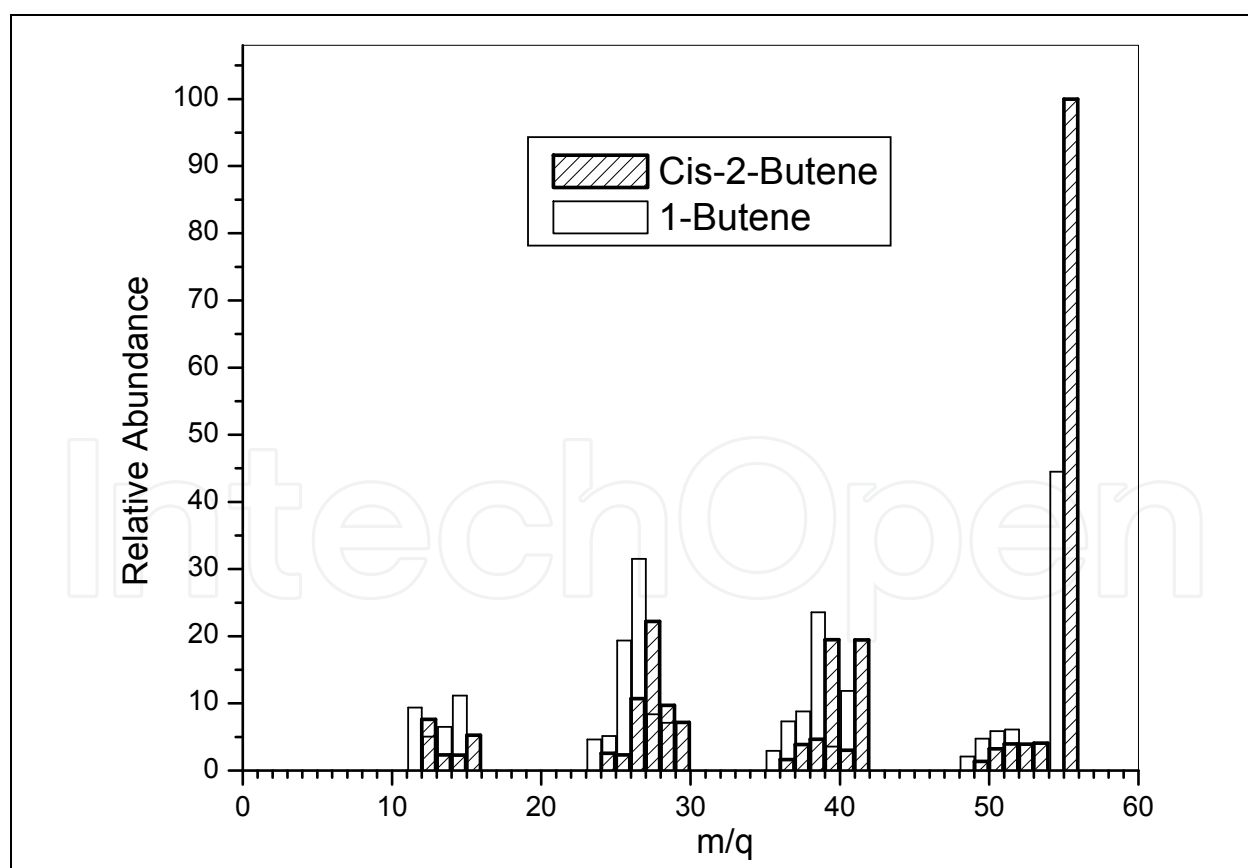


Fig. 10. Comparison of mass spectra for Cis-2-Butene and 1-Butene prepared by exposing pure gases to linearly polarized short laser pulses focused to peak intensity of  $6.5 \times 10^{13} \text{ W/cm}^2$  ( $SI=0.793$ ).



Another parameter that can be used to optimize the short pulse based ion source is the laser wavelength. We have noticed that the fragmentation pattern is wavelength dependent therefore, in theory, by changing the wavelength we may enhance the SI of two isomers. Unfortunately, tuning wavelength is not easy and requires expensive setup, which often is bulky and is not able to supply stable output. This difficulty can be overcome by using our dual wavelength setup that by varying the angle between the polarization vectors of  $\omega$  and  $2\omega$  fields results in an interaction outcome equivalent to what was expected from using a single frequency,  $\omega'$ , in the range  $\omega < \omega' < 2\omega$ . This finding will be discussed in the next section.

3.1.2 Dichroic pulses

In a recent publication (Sharifi *et al* (2010)) we studied the interaction of a two-color field by taking the angle between the polarization vectors of the two fields as the control parameter. The experimental setup is presented in figure 11. A Ti:Sapphire laser system provides a ten hertz train of 800-nm pulses having a full-width-at-half-maximum time duration of 150 fs. A KDP crystal of thickness 2 mm, and phase-matched for the second-harmonic generation,

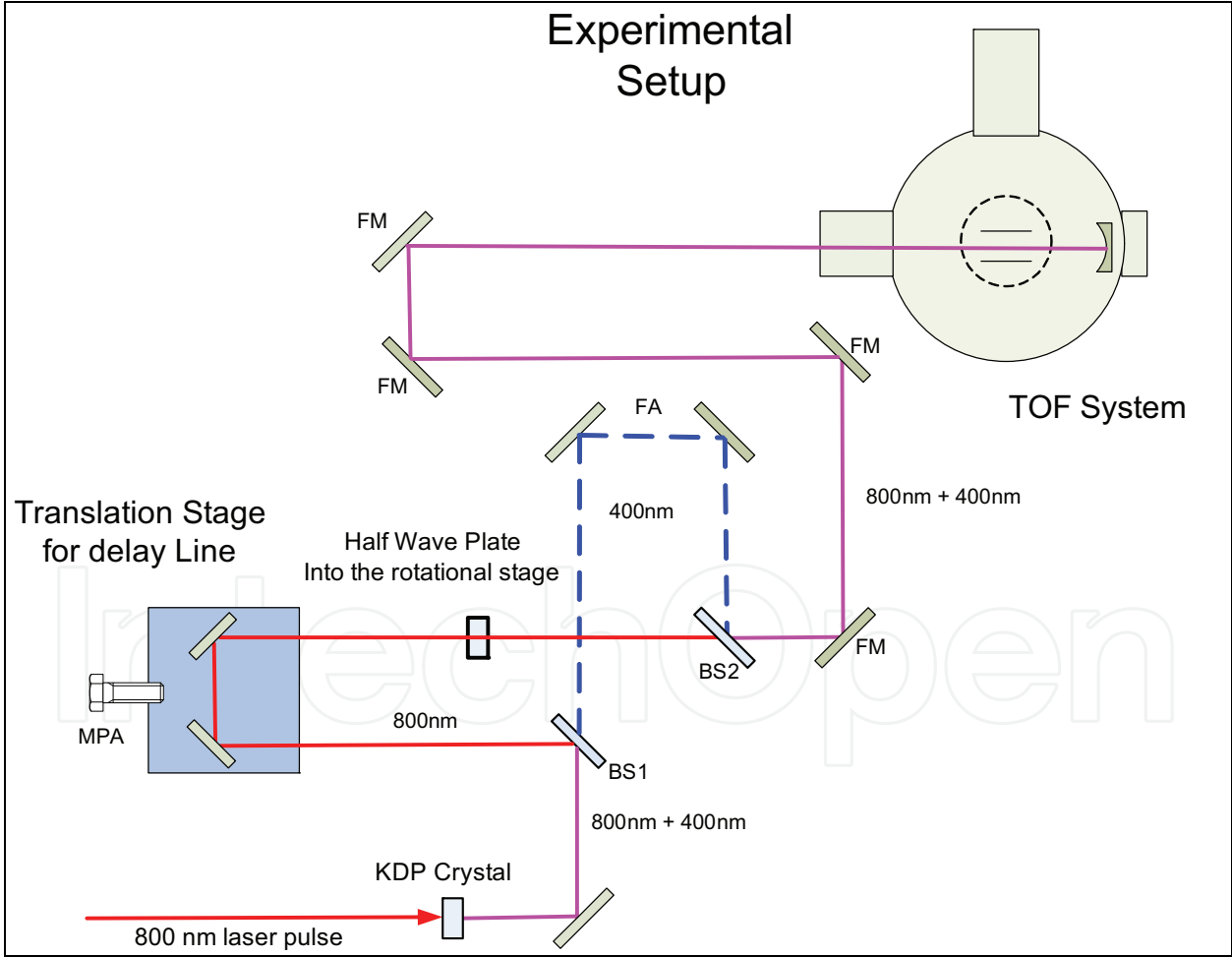


Fig. 11. Comparison Figure 1- Schematic diagram of the experimental setup for two-color multiphoton process study where, BS1: Harmonic separator HR@800 nm, AR/HT@400 nm; BS2: Harmonic separator, HR@400 nm, AR/HT@800 nm; MPA: Micrometer piezoelectric actuator; FA: Fixed arm ~400-nm arm; FM: Folding mirror.

was used to produce the 400-nm pulses whose characteristics will be explained later in this section. The fundamental and second-harmonic beams emerging from the doubling crystal are passed into a Michelson interferometer with a dichroic beam splitter. The IR arm of the interferometer includes a wave-plate oriented such that the angle between the polarization vectors of the two beams,  $\theta$ , can be varied in the range of 0 to  $\pi/2$ . The laser pulses from both the arms of the set-up were collinearly focused using a 10-cm focal length concave mirror in the vacuum chamber.

The total field causing the interaction is given by  $E(t) = E_\omega \cos(\omega t) + E_{2\omega} \cos(\theta) \cos(2\omega t)$ . By defining two variables  $\alpha = (1 + E_\omega / E_{2\omega} \cos(\theta))^{-1}$  and  $F = E_\omega + E_{2\omega} \cos(\theta)$  the field can be expressed as;  $E(t) = F((1 - \alpha) \cos(\omega t) + \alpha \cos(2\omega t))$ . We have shown that, under the condition  $I_{2\omega} \ll I_\omega$ , the rate of electron tunneling in this field can be calculated by applying the following transformations,

$$I_\omega \rightarrow I_\omega + I_{2\omega} \cos^2(\theta) + 2\sqrt{I_\omega I_{2\omega}} \cos(\theta) \quad \omega \rightarrow \omega\sqrt{1 + 3\alpha}$$

Thus, the two color field is equivalent to a single laser whose wavelength can be adjusted by varying the angle between the polarization vectors of the two fields. This effect can be very useful for the mass spectroscopy application as we mentioned in the previous subsection.

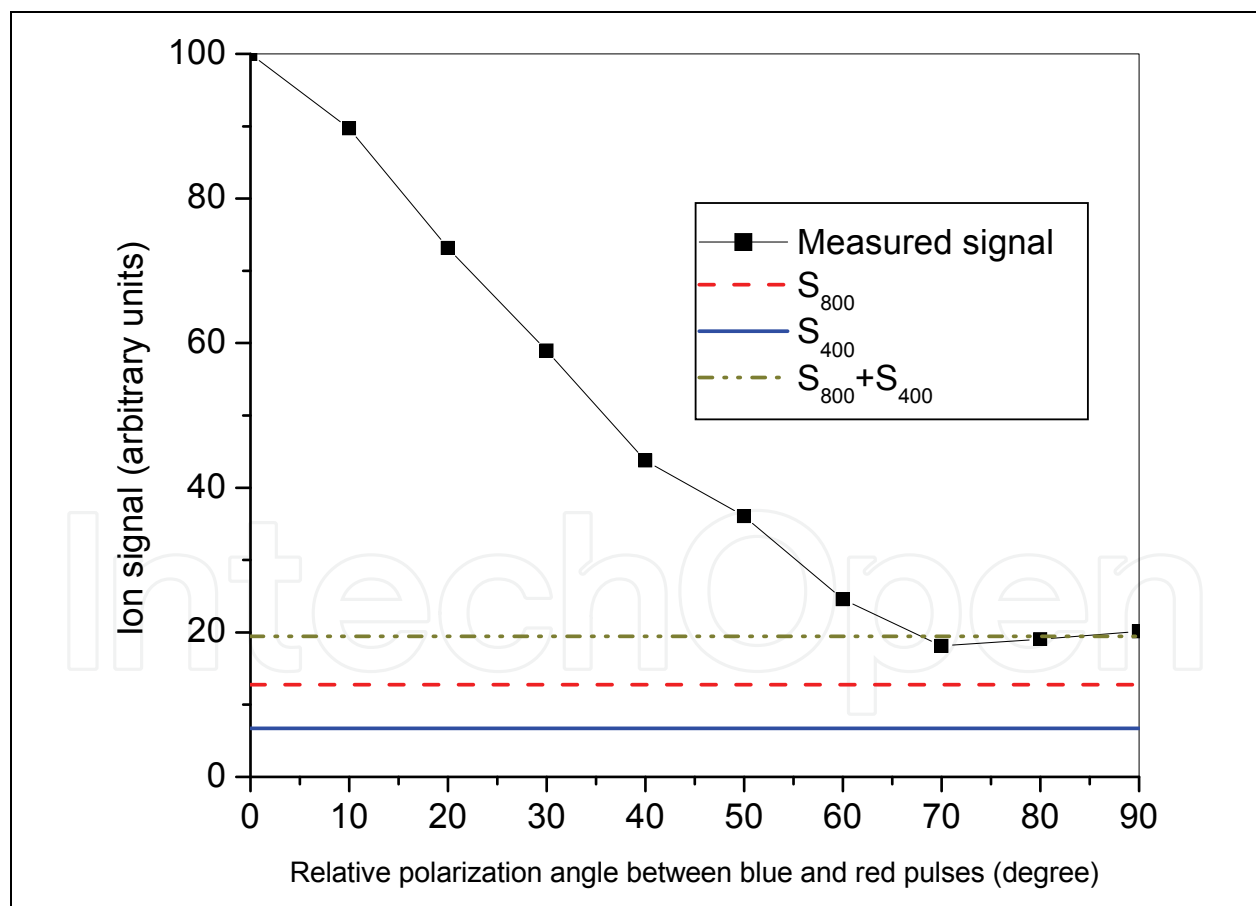


Fig. 12. The dependence of  $\text{Xe}^{++}$  signal on  $\theta$  which is the angle between the polarization vectors of  $\omega$  and  $2\omega$  pulses. The “measured signal” plot corresponds to the case when both pulses are present. The dashed and solid horizontal lines indicate the ion signals when either of the  $\omega$  or  $2\omega$  pulses is present alone.

The key finding from the tests using the setup of figure 11, as related to the potential use for generating permanent index changes in transparent solids, is furnished by the following experimental result. the ionization of Xe was studied by setting the intensities of  $\omega$  and  $2\omega$  pulses such that  $I_{\omega} = 7 \times 10^{13}$  W/cm<sup>2</sup> and  $I_{2\omega} = 7 \times 10^{12}$  W/cm<sup>2</sup>, respectively. In figure 12, the signal of doubly charged ion is presented as a function of the angle between the polarizations of the two fields,  $\theta$ . In a purely classical picture, it is expected that the rescattered electron (ionized and rescattered by the action of  $\omega$  field) to be deflected by the action of  $2\omega$  field. Accordingly, the electron-ion collision probability and the resulting double ionization rate depends on the angle  $\theta$ , because the double ionization process at our working intensity is dominated by the non-sequential channel.

3.2 Flat panel display dicing

Flat panel dicing using short laser pulses is, in our opinion, is a relevant application due to the fact that as the glass substrates become thinner it becomes more difficult to improve the process yield using conventional diamond blade dicing techniques. Using short pulses dicing speeds of over 400 mm/s has been successfully demonstrated on non-alkali glass and borosilicate glass, using a 50 kHz, 5W high-power femtosecond laser. The working principal of the technique is presented in figure 13, which has been adapted from Kamata *et al* (2008).

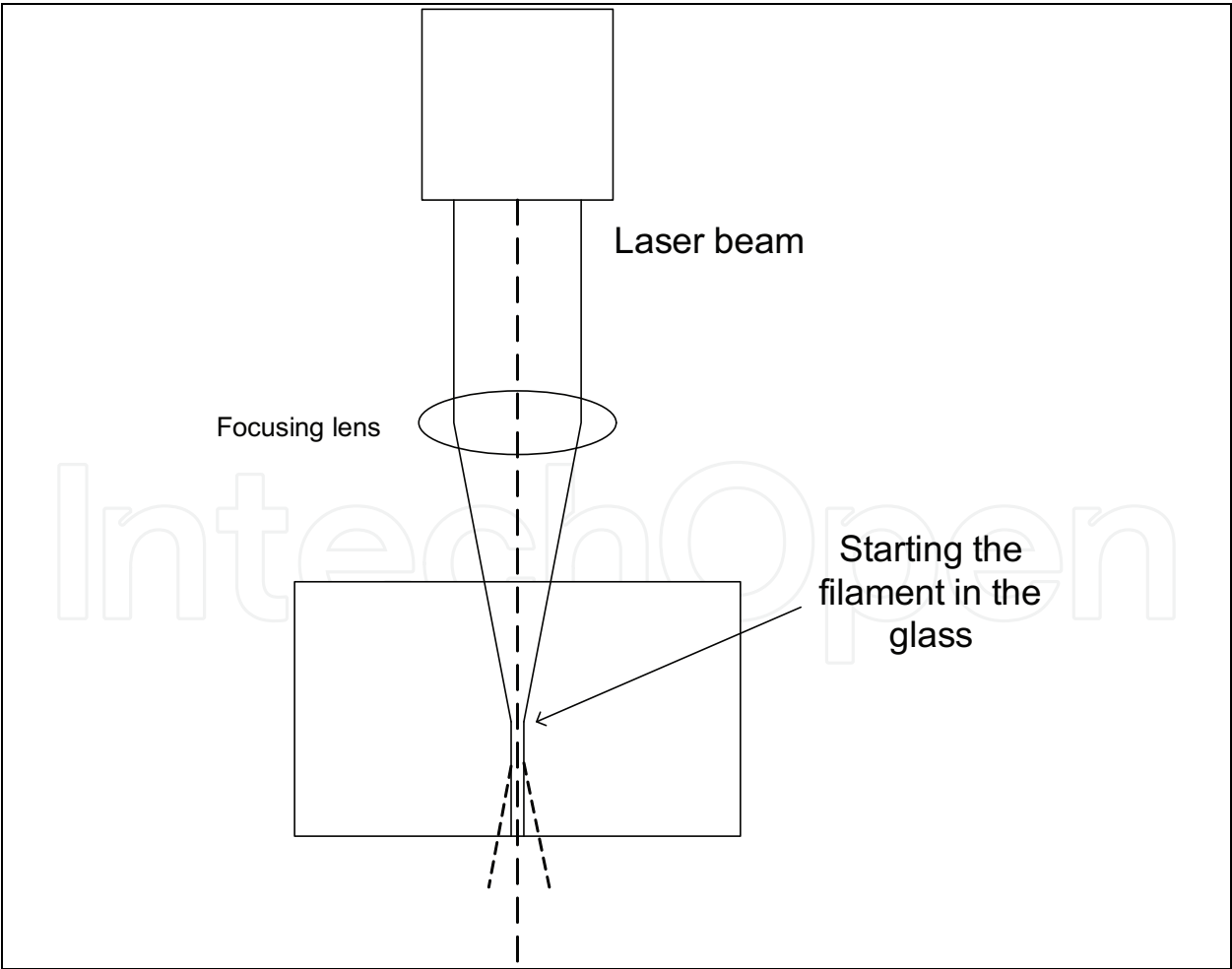


Fig. 13. Schematic of short laser pulse dicing technique.

The short pulse laser beam having a wavelength to which the work is transparent is directed to the front surface of the work toward the back surface and focused. A filament in the light beam traveling direction from the beam waist is formed by the auto-focusing action due to the laser beam propagation in the work is formed. The substance in the filament is decomposed by the laser beam and can be discharged from the back surface, and a cavity is formed in the channel. While forming the cavity, the laser beam is scanned, a machined surface is formed, and thereafter the work can be cut with a weak bending stress.

#### 4. Conclusion

Over past two decades, robust lasers capable of providing pulses with sub-picosecond durations have been developed. This has resulted in extensive research activities attempting to find applications based on light-matter interactions involving, predominantly, electronic couplings. Such interactions minimize the, potentially undesirable, effects of transitions between the ro-vibrational states of the molecules, which may result in heating and uncontrolled disintegration of molecules. Though, even a single wide-spread application has not materialized yet, but nuclei of the potential applications have already formed. We discussed the fact that not all the suggested applications are relevant, in terms of addressing the market needs in cost effective manner, and will be adopted by the high technology companies. However, there are unexplored areas with the potential of offering viable solutions to the real life problems. We identified and discussed two areas with some details. These include the use of ultrafast pulses as an ion source for mass spectroscopy and flat panel display dicing.

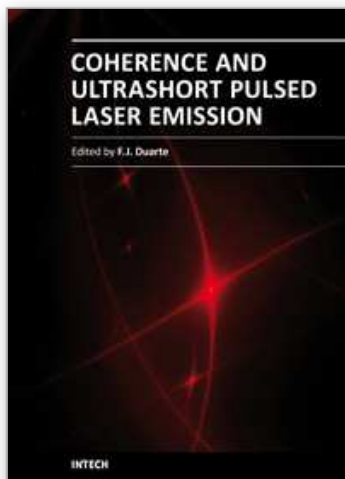
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In this volume, recent contributions on coherence provide a useful perspective on the diversity of various coherent sources of emission and coherent related phenomena of current interest. These papers provide a preamble for a larger collection of contributions on ultrashort pulse laser generation and ultrashort pulse laser phenomena. Papers on ultrashort pulse phenomena include works on few cycle pulses, high-power generation, propagation in various media, to various applications of current interest. Undoubtedly, Coherence and Ultrashort Pulse Emission offers a rich and practical perspective on this rapidly evolving field.

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