

Chapter 1

Introduction

This book discusses the consequence of the so-called self-focusing of a very short laser pulse in a transparent medium. Self-focusing is similar to focusing of a light/laser pulse by a lens except that this lens is a self-created one. If a laser pulse has an intensity distribution across the wave front and if the intensity is strong at the center and weak at the edge (a Gaussian distribution in intensity, for example), the high intensity central part of the wave front would see a higher index of refraction in the medium due to nonlinear increase of the index of refraction than the low intensity edge. This is similar to focusing a plane wave by a lens where the central part passes through a thicker glass medium than the edge. Because the velocity of propagation is equal to c/n where c is the velocity of light in vacuum and n is the index of refraction of the lens' material, the light wave thus propagates through the central part of the lens slower than through the edge resulting in the curvature of the wave front toward the axis of propagation. This means focusing. The same is true in self-focusing except that this is a continuous self-inflicted process throughout the propagation. It will thus come to a self-focus where ionization occurs. Different sections/slices of the pulse along the propagation direction self-focus at different positions along the propagation axis giving rise to a continuous series of self-foci which we call filament. This description will be expanded in more detail in Chapter 2.

When using/propagating femtosecond (10^{-15} s) laser pulses in any transparent optical medium, there is no way to avoid self-focusing of the pulse so long as it is sufficiently intense. When it is powerful enough, self-focusing will collapse resulting in plasma generation. In the olden days, using long laser pulses, this phenomenon was and still is a nuisance one likes to avoid inside laser systems. But with femtosecond (fs) laser pulses, the phenomenon becomes what is now popularly called filamentation with a lot of potential applications.

1.1 Mature Physics and New Development

This is a story of the revival of a so-called mature field of science. Nonlinear optics was developed following the discovery and development of lasers in the 1960s.

Phenomena such as self-focusing and self-trapping of laser beams in an optical medium (Hercher, 1964; Chiao et al., 1964), multiphoton/tunnel ionization (see for example Chin and Lambropoulos, 1984 and a recent review by Chin, 2004) and optical breakdown (see for example: Morgan, 1975; Raizer, 1991 and references therein) of materials by a laser pulse at high intensity were well known. Filamentation and spectral (red) broadening were observed by Reintjes et al. (1973), while the limitation of the laser intensity at the self-focus (which we now call “intensity clamping”) in glass was proposed already in the 1970s (Yablonovitch and Bloembergen, 1972). Probably no one would have predicted that a powerful femtosecond laser pulse could uncover a seemingly known yet totally unexpected sub-field of applied physics, namely, filamentation nonlinear optics (Chin et al., 2007). Special names, some of which became popular, were given to describe known or unexpected phenomena. Examples are filamentation, intensity clamping, background (energy) reservoir, self-transformed white light laser pulse, self-spatial filtering, self-group phase locking, self-pulse compression (down to the few cycle and even single cycle levels), clean nonlinear fluorescence, and so on. Long range propagation at high intensity which is seemingly against the law of diffraction, is probably one of the most exciting consequences of this new sub-field of nonlinear optics. Short range propagation in glass resulting in the transformation of its properties is also as fascinating. The main reason for the observation of the above mentioned new phenomena is the very short duration (femtosecond) and high power/intensity of the laser pulse. Nanosecond or even picosecond laser pulses could not have produced such results because of optical breakdown that masks the observation of the new phenomena.

Imagine the following scenario. A femtosecond laser pulse propagates and self-focuses into a small volume in air at a high intensity such that some molecules are ionized. It then keeps on propagating in this form as if it were a bullet through a long distance up to the kilometer range. A streak of plasma column is left behind by the bullet; the streaking “bullet” gives rise to the perception of a filament. As it propagates, its frequency keeps on broadening “by itself”. Soon, it turns white (very broadband) encompassing what is popularly called supercontinuum. (See for example: Alfano, 1989 and references therein.) It is called a chirped white light laser pulse. This self-transformed white light laser pulse could be considered as a white light bulb streaking in the sky whose back illumination (scattering) could be applied to molecular absorption spectroscopy, hence identifying some molecules in air at long range.

Inside the core of this filament, the high intensity can explode chemical and biological molecules/agents. Many of the neutral fragments emit fluorescence with a characteristic spectrum; i.e., each species will emit its own finger print fluorescence spectra. Because we can in principle project the filament at long distances in air, we can thus detect chemical and biological agents in air from a long distance using the laser radar (LIDAR) technique. Solid targets interacting with the strong filament can be identified at long range through what is called femtosecond laser induced breakdown spectroscopy using this remote sensing technique.

Associated with this fundamental bullet of high intensity laser light is another light bullet at the third harmonic frequency of the fundamental bullet, born through

the interaction of the fundamental bullet with air. This third harmonic bullet is dragged along by the fundamental bullet at the same speed of propagation (self-group phase locking). The third harmonic UV pulse as well as the high intensity fundamental pulse could excite fluorescence from chemical and biological molecules at long range. Sodium in air could be excited to fluoresce, hence forming an artificial “star”. The filament could in principle conduct electricity, hence providing lightning control. High spatial quality few cycle (down to single cycle) intense pulses tunable from the UV to the radiofrequency could in principle be generated inside the filament in gases by mixing it with another pulse at an appropriate frequency.

Imagine again the propagation of such pulses in glass. The filament is short. Inside the filament, the glass material is melted by the very short femtosecond pulse in an initial interaction time much shorter than the heating time. When the melted material re-solidifies, a new zone of different index of refraction is born. This leads to the so-called writing of a waveguide or generating data storage space in the material.

Many of these are previously unheard-of phenomena. The author believes that more new phenomena, deeper physical understanding and new applications are waiting to be discovered. He attempts to describe and explain the basic physics and applications of this new field of filamentation nonlinear optical science principally in air (gases), while that in condensed matters will be briefly described. The book aims at those who have an interest to enter field. The presentation of this book is at the level of graduate students in physics and researchers from other fields.

During filamentation of a femtosecond laser pulse, some fundamental nonlinear processes are involved in the propagation and interaction, namely nonlinear phase changes of the laser pulse, multiphoton and tunnel ionization, and optical breakdown. These concepts will be explained briefly in the rest of this chapter. The most popular femtosecond Ti-sapphire laser (central wavelength around 800 nm) will be used in our discussion as the laser source throughout this book.

1.2 Phase Effect of a Laser Pulse Propagating in an Optical Medium

A laser pulse is an electromagnetic pulse. Mathematically, it can be represented as a Fourier integral by the superposition of a set of plane waves propagating in the z -direction in the following form, for example

$$E(z,t) = \int_0^{\infty} E_0(\omega,t) \exp[i(\omega t - kz)] d\omega \quad (1.1)$$

$$k = \omega n/c$$

The plane wave component in the integrand contains the wave vector k which is expressed as a function of the index of refraction n . It is this index that plays a central role in the propagation, self-focusing and filamentation of a femtosecond laser pulse. This is because n is dependent on many parameters. In the linear regime, it is wavelength dependent. In a transparent material, under normal dispersion condition, longer wavelengths have a lower index. Thus, a plane wave at a longer wavelength (say red color) will travel faster than that at a shorter wavelength (blue). The initial coherent superposition of many waves at different wavelengths gives rise to a short pulse. However, once they start propagating, the red will propagate faster than the blue components and the pulse becomes longer.

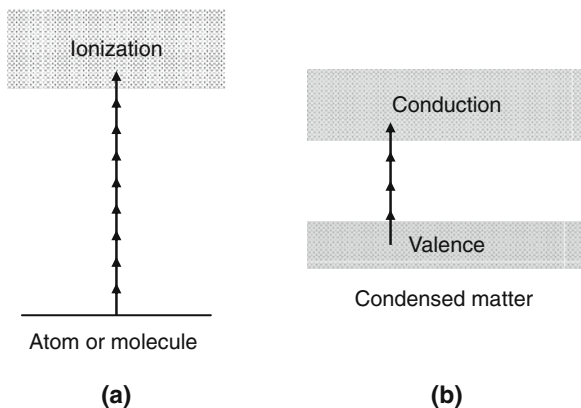
Many good books deal with the aspect of the superposition of wave and ultra short pulses. The readers are referred to the following, namely, Rulière C ed. (1998) and Diels and Rudolph (2006).

1.3 Multiphoton and Tunnel Ionization

Multiphoton ionization (Delone and Krainov, 1994; Chin and Lambropoulos, 1984) is simply an extension of the phenomenon of single photon ionization. This is a bound-free transition. In the context of the perturbation theory, an electron in a bound state in an atom or a molecule, after absorbing one (usually) energetic-photon, would have the probability of being excited into the free (electron) state or continuum. In the case of condensed matter, it is the ejection of an electron by the energetic-photon that overcomes the work function. In both these situations, the photon energy is larger than the ionization potential or the work function. This is the normal photoelectric effect in which one photon is absorbed.

What if the photon energy is less or much less than the ionization potential or the work function? Quantum mechanics, in the framework of the perturbation theory (Goeppert-Mayer, 1931), shows that if the intensity of the radiation is high enough, there will be a distinct probability that the atom or molecule or the condensed matter absorbs several photons “simultaneously” (in one cycle of the field oscillation) and excites a bound-free transition thus releasing a free electron; this is probable when the total energy of the absorbed photons is larger than the ionization potential or the work function. In Fig. 1.1a, a schematic picture of multiphoton ionization of an atom or a molecule is shown. The absorption of several photons simultaneously overcomes the ionization potential, giving rise to the probability of ejecting an electron. This picture can also represent the case of releasing a free electron from the surface of a metal (condensed matter) where now the work function is overcome resulting in the ejection of a free electron. In the case of filamentation in a condensed matter which is transparent to the incoming femtosecond laser pulse, a multiphoton transition of an electron from the valence band to the conduction band is more probable (Brodeur and Chin, 1998, 1999). This is shown in Fig. 1.1b. The electron set free into the conduction band will be further accelerated by the laser field through collisions resulting in a breakdown, but only partially in the case of femtosecond laser pulses (see below).

Fig. 1.1 Schematic diagram illustrating multiphoton transitions. (a) Multiphoton ionization of an atom or a molecule; (b) multiphoton transition from the valence to the conduction band in the case of a transparent condensed matter



However, when using femtosecond laser pulses to ionize gases, it is more appropriate to talk about tunnel ionization (Fig. 1.2). Tunnel ionization of gas atoms or molecules is similar to multiphoton ionization in the sense that both are high intensity effects. Depending on the combined condition of the laser and the ionization

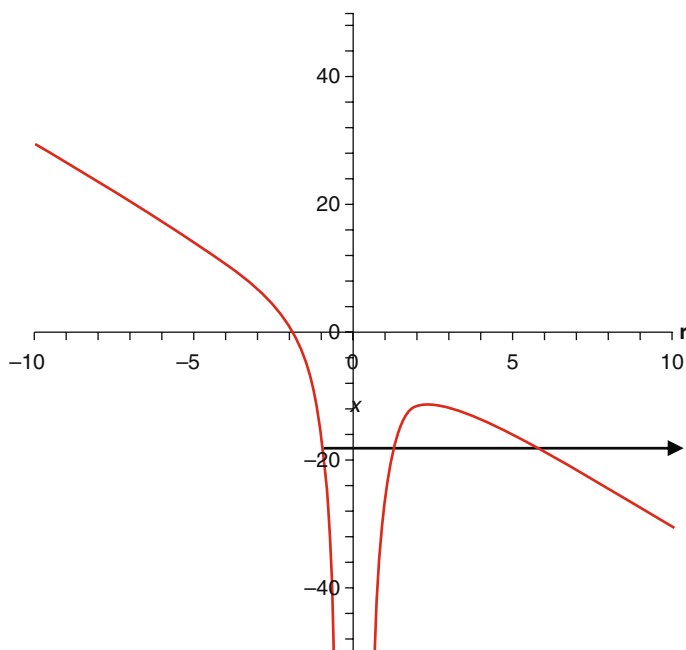


Fig. 1.2 Tunnel ionization

potential of the gas particle to be ionized, we could be in the multiphoton or the tunnelling regime. One popular parameter that distinguishes between multiphoton and tunnel ionization is the Keldysh (adiabatic) parameter γ (Keldysh, 1965). The first observation of tunnel ionization of atoms and molecules was by Chin (1984) and Chin et al. (1985), after more than 20 years of debate by those in the field. From our experimental findings, when $\gamma < 1/2$, it is in the tunnelling regime and when $\gamma > 1/2$, it is in the multiphoton regime (Chin, 2004). For a more detailed discussion of multiphoton and tunnel ionization, see Chin (2004).

1.4 Optical Breakdown

Since the invention of the Q-switched laser at the beginning of the 1960s, optical breakdown in air or condensed matter became an easily observable physical phenomenon. When focusing any intense nanosecond or picosecond laser pulse in the material, optical breakdown is bound to occur. Before filamentation was well understood, optical breakdown was a rule with no known exception.

It was started by Meyerand and Haught (1963, 1964) who observed that a focused Q-switched ruby laser pulse could generate a spark (plasma) in air. After a great deal of extensive theoretical and experimental work in many laboratories, it was finally proposed that optical breakdown of gases followed essentially a three-step process (Fig. 1.3) as follows: (1) Multiphoton ionization (MPI) of impurity molecules with low ionization potentials would easily provide a few free electrons with low initial kinetic energy in the focal volume at the front part of the pulse. (2) The free electrons in the strong laser field could absorb or emit n photons ($n = 0, 1, 2, 3, \dots$)

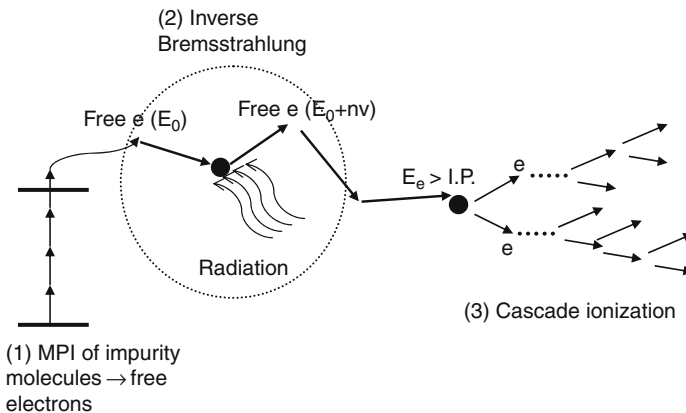


Fig. 1.3 Visualization of laser induced breakdown, a three steps process: (1) MPI of impurity molecules, (2) inverse Bremsstrahlung resulting in electron acceleration, and (3) cascade ionization

while colliding (scattering) with a much heavier particle (atom, molecule or ion). The heavy particle is to conserve momentum during the interaction. The process of absorbing n photons is called inverse Bremsstrahlung or free-free transition. (3) After one or more inverse Bremsstrahlung processes, the free electron would acquire a kinetic energy E_e higher than the ionization potential of the gas molecule/atom. Subsequent collision would give rise to the ejection of an extra electron from the molecule/atom. This would result in two low energy electrons. They would undergo the same processes as before each giving rise to two more electrons, and so on, until the gas is fully ionized. This is called cascade or avalanche ionization; i.e., breakdown (Morgan, 1975). Many experiments were performed to verify the cascade ionization process (Morgan, 1975). The fundamental process of the initiation of breakdown by the MPI of impurities was proved experimentally by Chin (1970) whereas the free-free transition which is at the center of cascade ionization was observed by Weingartshofer et al. (1977, 1979).

In condensed media transparent to the laser pulse, the interaction leading to breakdown inside the material is essentially similar to that in gases except that the first few “free” electrons are generated through multiphoton absorption by these electrons from the valence to the conduction bands. Once in the conduction band, these electrons would again undergo inverse Bremsstrahlung followed by cascade ionization resulting in the breakdown of the material (Raizer, 1991).

One major difference between breakdowns in gases and in condensed matter is that the density of atoms/molecules in a condensed medium ($\sim 10^{22} \text{ cm}^{-3}$) is about three orders of magnitude higher than that in a gas, say, at one atmosphere ($\sim 3 \times 10^{19} \text{ cm}^{-3}$). Thus it is much easier to induce breakdown in a condensed medium than in gases. In fact, using femtosecond laser pulses, practically no inverse Bremsstrahlung and cascade (avalanche) ionization could take place in gases at one atmospheric pressure at the intensity of $\sim 10^{13} \text{ W/cm}^2$. This is because the mean free time of a free electron released from an atom or a molecule through multiphoton/tunnel ionization is longer than the pulse duration. The mean free time of a free electron is given by the formula (Raizer, 1991)

$$(\Delta t)_{\text{mean}} = \frac{1}{\sigma N_a v_e} \quad (1.2)$$

where $(\Delta t)_{\text{mean}}$ is the mean free time; σ , the electron-neutral collision cross section; N_a , the atomic/molecular density; v_e , the free electron velocity in the field.

We estimate the mean free time as follows. The clamped intensity in air is $\sim 5 \times 10^{13} \text{ W/cm}^2$ (Kasparian et al., 2000a; Becker et al., 2001). According to Paulus (2007, private communication), from the observed electron spectra of the above threshold ionization (ATI) (Becker et al., 2002; Milosevic et al., 2006) of atoms at the intensity range of 10^{13} – 10^{14} W/cm^2 , the probability of an electron to become really free is more than 99%. That is to say, not more than 1% of the ionized electrons participate in the rescattering process (Corkum, 1993; Kuchiev, 1987, 1995, 1996). Thus, rescattering of the electrons could be neglected. The free electron will experience an oscillatory motion in the laser field acquiring a mean

kinetic energy (known as the ponderomotive potential) given by (units are given in the parenthesis) (Augst et al., 1991)

$$\frac{e^2 E^2}{4m\omega^2} (\text{eV}) = (9.33 \times 10^{-14}) I (\text{W/cm}^2) \lambda^2 (\mu\text{m}) \quad (1.3)$$

where e and m are the electronic charge and mass respectively; ω , the laser frequency; E , the electric field strength of the laser pulse; I , the laser intensity and λ , the wavelength. It is between 0.6 and 6 eV in the intensity range of 10^{13} – 10^{14} W/cm². Its nonrelativistic velocity is thus $v_e \sim 4.5 \times 10^7$ to 1.4×10^8 cm/s. N_a of one atmospheric air is 2.68×10^{19} cm⁻³; $\sigma \sim 10^{-15}$ cm² for both gas and condensed matter for electrons with kinetic energy ranging from 1 to 6 eV (Raizer, 1991). We obtain $(\Delta t)_{\text{mean}} \sim (3\text{--}8) \times 10^{-13}$ s or 300–800 fs. This is longer than most of the femtosecond laser pulses used in the experiments.

In the case of condensed matter, the density is 1000 times higher than that of air at one atmosphere. Thus, the mean free time is 0.3–0.8 fs. Sun et al. (2005), using 130 fs Ti-sapphire laser (800 nm) pulses, have measured that the electron collision time (mean free time) inside the filaments in fused silica is of the order of 1.7 fs. The theoretical estimation is in good qualitative agreement with the experimental result. The order of magnitude of mean free time in condensed matter could thus be roughly 1 fs. This would mean that if a whole 50 fs (at FWHM) pulse focuses into a small focal zone, there would be a few tens of cycles of collisional ionization resulting in optical breakdown. This is true when focusing the laser pulse by a very short focal length lens. However, because of slice-by-slice self-focusing (see Chapter 2), the number of collisions is much less when using a long focal length lens to focus the fs pulse into a condensed medium. We shall come back to this question in Chapter 2.

1.5 Intense Femtosecond Laser Beam Attenuation

Very often, one needs to vary the energy of the powerful femtosecond laser pulse. The classical technique is to use transmission-type attenuators. There would be some modification of the physical properties of the pulse (wave front, pulse duration, chirp, etc.) after passing through the attenuator because of nonlinear effects (self-focusing, GVD, etc.) (Tang et al., 2005a,b). To avoid this, one could use reflective attenuators. Another way is by adjusting (rotating) a half-wave-plate in front of a polarizer. The half-wave-plate is oriented such that it will rotate the polarization of the uncompressed chirped laser pulse which the fixed polarizer will transmit the selected polarization, thus attenuating the laser pulse. The half-wave-plate/polarizer pair is placed before the compressor. If the uncompressed pulse is very energetic (10 mJ or higher), it is wise to place them in the path of the seed beam before the latter is further amplified. This latter technique of using the half-wave-plate/polarizer pair ensures that the beam energy is adjusted before the compression of the pulse; it thus avoids the deterioration of the wave front of the pulse through absorption

optics due to nonlinear propagation effects. This latter technique is adopted in all the author's experiments in Laval University. However, for experiments involving a window of a vacuum system or using lenses, the same problem would arise. In the work of Tang et al. (2005a,b), this problem was discussed thoroughly. A good way is to use the half-wave-plate/polarizer pair in front of the amplifier as mentioned above and put the compressor and the beam steering optical elements and the interaction zone all inside the *same* vacuum. In the case of propagation experiment in optical media, the compressed pulse has to get out of the vacuum system and the window will always be a problem. It is a question of the tolerance of the particular experiment under study.



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