Module 1 : Introduction and Background Material

Lecture 1 : Introduction

Objectives

In this lecture we will look at

- Linear versus nonlinear response of a system.
- A perspective on light interaction with matter.
- Non-linearity in optics.
- Brief history of nonlinear optics and why it is important?
- General references.

1 Linear versus nonlinear response

A common method of exploring properties of a physical system is to study its response to a probe or stimulus. There exists a functional relationship between the effect E and stimulus S. Dependence of E on S is the system's property. *Depending upon whether this relationship linear or nonlinear, a system called linear or nonlinear*.

1.1 Some Illustrations:

We start with a familiar example of a mechanical system - a simple pendulum shown in figure 1, which, when displaced (*stimulus*) from equilibrium, executes oscillatory motion with variable acceleration (*effect*) - under the gravitational force component directed towards the equilibrium position. The equation of motion is

$$\frac{d^2\theta}{dt^2} = -(g/l)\sin\theta \qquad (1.1)$$

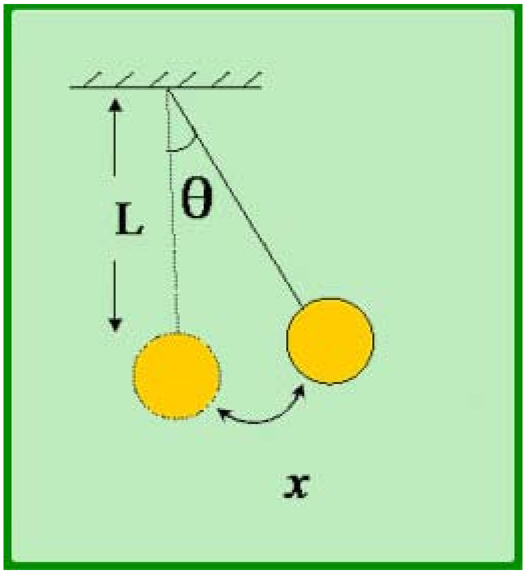

Figure1.1: motion of a simple pendulum

To obtain a description of the motion of the pendulum we need to solve this equation with the initial condition that at *t=0*, the pendulum starts with zero speed and a given amplitude θ_0 . Although, the exact solution of this problem is known, simplification can be obtained for small amplitude. For $\theta << 1$, we can write $\sin\theta \simeq \theta$ .Then this equation of motion is linear with the well known harmonic solution.

$$\theta = \theta_0 \cos(\sqrt{(g/l)}t) \qquad (1.2)$$

For large displacements equation (1.1) is no more linear. In this simple case the exact solutions of eq (1.1) are well known in terms of Jacobi elliptic functions. For larger values of the initial amplitude θ_0 the motion remains periodic but not a simple cosine function and the period depends on the initial amplitude. If we resolve this motion into a number of sinusoidal functions we find that the motion contains many harmonic frequencies. It is important to note that there is no threshold in the problem i.e. there is no value of initial amplitude θ_0 below which these corrections are zero, but of course they are *small*. Now, we know that if two pendula are close to each other, oscillations can transfer from one to another.

What happens if the frequency (as obtained in the linear approximation) of one is thrice that of another?

Our description above suggests that a pendulum with resonant frequency ω_0 can excite one at frequency $3\omega_0$ and this transfer will depend on the amplitude of the first pendulum. Thus we see that such a simple system actually turns out to be nonlinear and in the process of linearization we miss out on many interesting phenomena e.g., the possibility of exciting this system resonantly at a harmonic frequency, the nonsinusoidal nature of oscillations and the dependence of the frequency on the initial amplitude.

Yet another example of a nonlinear system is the p-n junction diode, the simplest semiconductor device. Its current voltage characteristic defined through

$$i_d = i_0\left[\exp\left(\frac{eV_d}{k_B T}\right)-1\right] \tag{1.3}$$

is essentially nonlinear as shown in figure 2(a). Even more interesting is the Esaki(tunnel) diode (characteristic shown in figure 2(b)) which exhibits bistable behavior. Naturally all other more complex semiconductor devices such as transistors, thyristors and varistors which are made of multiple junctions are expected to be and indeed are nonlinear.

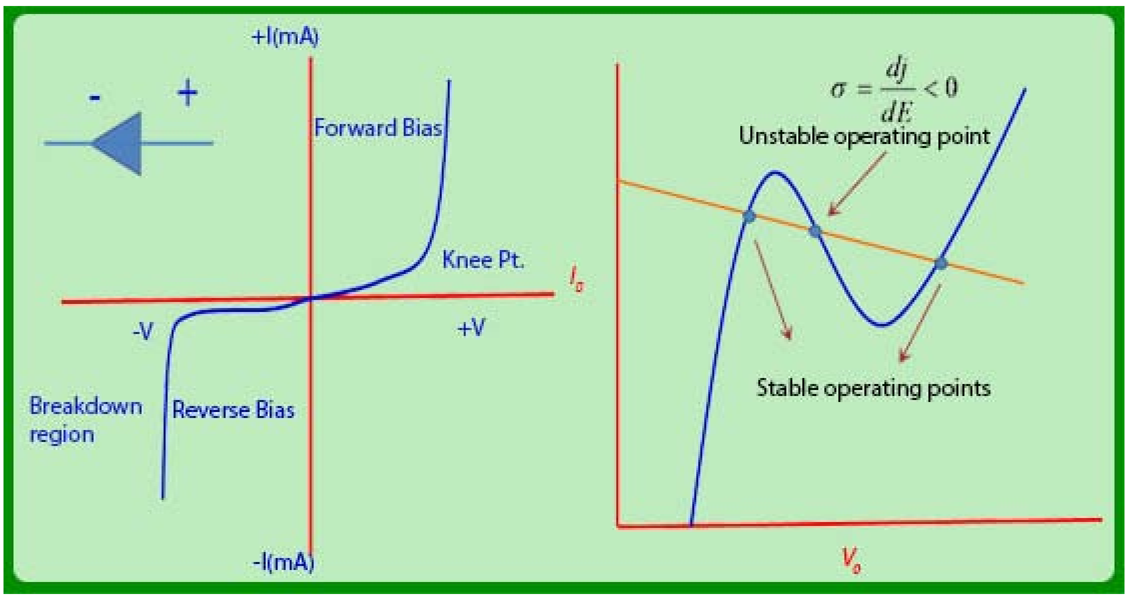

Figure 1.2 I-V Characteristics of (a) p-n junction diode (b) Tunnel diode

In fact, nonlinear systems are abundant in nature, be it the chemical reactor involving activator-inhibitor reaction kinetics or morphogenesis in biology.

It thus is not an exaggeration to say that almost every system in nature is nonlinear and optics is no exception

1.2 Optical response of matter

In an elementary optics course, we learn that light is electromagnetic in nature. The main cause of its interaction with matter has to lie in the interaction between the electric and magnetic fields and the charged particles in matter. For transparent dielectrics, a useful model is that the oscillating electric field of light sets up electric dipole oscillations in the molecules or atoms of the medium, and they in turn radiate electromagnetic waves. In the first approximation, we expect that the induced moments would be proportional to the incident fields and would therefore *oscillate at the same frequency*. This field radiated by the dipoles together with the incident field produces the resultant optical waves. When the medium is homogeneous and the interface smooth then these fields form the reflected and the refracted waves. When there are fluctuations either in the medium or at the interface, there is scattering. If the optical wave has a frequency such that the corresponding photon energy matches with a possible transition energy in the medium, we learn that the medium can absorb energy from the optical beam- which may be partly re-emitted by the medium (luminescence) and party converted into other excitations of the medium which in a very broad way is called heating of the medium.

All these phenomena, are covered by what we will now call ***linear optics***, since the amplitude of the induced dipole is taken as proportional to the incident field it is linearly dependent on the incident field. Yet another way of saying the same thing is that the optical response of the medium is linear where we envisage that the incident light is a probe of the medium, and the resultant phenomena reflection, refraction, scattering, absorption and luminescence are all response of the medium. In the simple picture, all these responses are describe by the polarization $\vec{P}(\vec{x},t)$ which is the induced dipole moment per unit volume at $\vec{x}$ at time t.

Thus, linear optics is the description of optical phenomena with the assumption that the induced dipole amplitudes are proportional to the electric field that induced them. If the field oscillates at an angular frequency ω, so do induce dipoles (see, Jackson). The total field in the medium-the incident field and the generated field- oscillates at frequency ω. In such a case no new frequencies can be generated and waves at different frequencies do not interact. The spectrum of incident light can of course be modified by selective absorption of a part of the spectrum of incident light but no frequency that is absent in the incident light is generated.

2. Nonlinear Optics:

Nonlinear optics deals with the study of the light- matter interaction in nonlinear regime which leads to myriads of phenomena hitherto unknown in the realm of the linear regime.

In the following sections, we will discuss the brief history of nonlinear optics and origin of optical nonlinearity in terms of simple classical picture.

2.1 Brief History of Nonlinear Optics:

Probably, mirage is the oldest natural phenomenon experienced by human beings that is akin to a nonlinear optical phenomenon. It is a manifestation of the light fluence dependent refractive index (thermal nonlinearity) as you will learn in these web lectures later. However, the prelude to the story of the scientific quest in nonlinear optics can be marked with the discovery of the Kerr effect [1] in 1875. Kerr in his experiments (see figure 5) with liquid and glasses demonstrated the rotation of the plane of polarization of the light propagating through a dielectric medium subjected to large electric field. This is caused by the anisotropic field dependent variation of its refractive index. In 1886, Bruhl [2] observed the Kerr effect to be stronger in conjugated organic molecule- recognized as an important class of nonlinear optical materials now. The first ever, truly nonlinear optical phenomenon of the saturation of absorption or state-filling effect was observed in 1926 by Vavilov and Levshin in Uranium glass using intense light from a spark [3]. Further growth of the exciting field of nonlinear optics was hampered due to unavailability of intense light sources and prospered only with the advent of lasers.

Within a year after the operation of the first laser, P. Franken demonstrated the generation of the second harmonic of Ruby laser radiation in 1961 [4]. Soon after that Kaiser and co-workers [5] observed the nonlinear optical phenomenon of two-photon absorption which was theoretically predicted by Goppert Mayer [6] in 1931. With the availability of the more powerful lasers a multitude of nonlinear optical phenomena such as sum and difference frequency generation [7], parametric generation and amplification of light [8.], optical rectification [9], stimulated Raman scattering [10], self focusing and filamentation [11-13], self and cross phase modulation and self steepening phenomena [14] to name a few were discovered in the 1960's. Fundamental theoretical contributions of Bloembergen [15], Akhmanov and Khokhlov [16] established nonlinear optics as an independent branch of optical sciences by mid sixties. In 1971, Stepanov demonstrated optical phase conjugation in degenerate four-wave-mixing scheme [17]. Z'eldovich used the stimulated Brillouin Scattering process in 1972 to achieve the same [18]. Time reversed wavefront generation using optical phase conjugation became work horse for the variety of applications including aberration correction and self targeting [19,20]. Observation of optical bistability in 1976 by Gibbs and coworkers [21], which was predicted by Szoke et-al in 1969 [22], sparked intense research activity in nonlinear optics and its applications to all optical computing. Investigations on the effects of cubic nonlinearity on the propagation of the laser pulses in the optical media led to the realization of optical solitons in 1983 which opened new vistas for all optical communications [23]. A variety of very versatile and ultra high resolution- both time and frequency- spectroscopic techniques based on degenerate and nondegenerate four-wave-mixing processes such as coherent raman scattering, pump-probe, laser excited grating(LEGs), photon echo, saturation and multiphoton spectroscopies, to name a few, were developed in the due course of time which generated wealth of new insights into the physics and chemistry of materials [24]. These discoveries sparked intense research activity in area of nonlinear optics during the 1980s. Bloembergen was awarded Nobel prize for his outstanding contributions to the field in 1981. With the introduction of chirped-pulse-amplification [25], intense laser sources took new leap to a regime where the electric field strengths started to exceed the intra-atomic coulomb field and added a new chapter of intense field-matter interaction[26]. Discovery of newer phenomenasuch as higher harmonic generation(HHG) [27-29], Tunneling ionization [30,31] and the barrier suppression ionization [32], above threshold ionization(ATI) [33] followed. Lasers approaching the fundamental intensity limit-Schwinger's limit [34]- are being developed to study relativistic nonlinear optical phenomena.

3. Physical picture and important scales in laser- matter interaction :

Perturbed by an oscillating electric field $E(\omega)e^{-j\omega t}$ a neutral atom acquires an oscillating dipole. In a homogenous medium, this is best described in terms of Polarization $\vec{P}$ Which represents the dipole moment per unit volume.

First order perturbation gives

$$\vec{P}^{(1)} - \vec{E} \qquad (1.4)$$

Next higher order terms in the perturbation series give terms that are:

$$\vec{P}^{(2)} - \vec{E}\vec{E} \qquad (1.5)$$

$$\vec{P}^{(3)} - \vec{E}\vec{E}\vec{E} \qquad (1.6)$$

Higher order terms are important because even though they are very small in magnitude they make qualitatively new phenomena possible. An analogy with spices in food is apt just as spices- even in very small proportions – make a qualitative difference to the taste of food, nonlinearities make qualitative changes to the behavior of optical waves. Smallness parameter for perturbation series is E_{laser} / E_{atom}, where E_{laser} is the electric field of the electromagnetic field representing the laser beam and E_{atom} is the electric field that is seen by an electron in an atom.

Exercise: Verify that the electric field in a laser beam with intensity ***Ilaser =1GW/cm²*** is much smaller than that seen by an electron in the first Bohr orbit. Hint Recall, that

$$E_{laser}(V/cm) = \sqrt{2\mu_0 c I_{laser}} \simeq \sqrt{27 I_{laser}(W/m^2)}$$

$$E_{atom} \sim \frac{e^2}{4\pi a} \simeq 10^9 \qquad (V/cm) \qquad (1.7)$$

For the perturbation series to converge we require that

$$E_{laser} \ll E_{atom} \qquad (1.8)$$

However, even for small fields, higher order terms may become important when resonant enhancement of susceptibilities occurs. A different, non-perturbative theory is then required. The other case in which greater sophistication of the theoretical treatment is necessary is when the laser field exceeds the atomic field. This is covered under *extreme nonlinear optics*.

Other approximation made in the above description is the *electric- dipole approximation* which amounts to retaining only the electric dipole term in the multipole expansion of the field. In general, the field $Ee^{-i(\omega t - \vec{q}.\vec{r})}$ can also excite higher multipoles which are neglected. The smallness parameter in the multipole expansion is *atomic size/ wavelength of light*. This too may need modification near a surface where the fields vary rapidly.

It is important in nonlinear optics specially to keep track of all approximations that are made because we are dealing with qualitative new effects of ***small*** terms in polarization. Often, we may find that it becomes necessary to go beyond the simplest description. And yet, we will also see that the perturbative nonlinear optics itself describes a great variety of phenomena which in turn yield a large number of devices and methods.

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Recap:

In this lecture we have learned about the

- Differences in the response of a linear and a nonlinear system.
- A perspective on light-matter interaction.
- Nonlinearity in optics.
- Brief history of nonlinear optics and why it is important?
- General references.