optical-frequency electric field, which can even be comparable with the internal electric field of the atom or molecule. In such a case, the nonlinear terms of the polarization expression cannot be entirely neglected and may play a vital role for various nonlinear optical effects. Based on quantum statistics, a light radiation with $\bar{n} \ll 1$ manifests the feature of a shot noise field when it interacts with matter; whereas a radiation with $\bar{n} \gg 1$ manifests the feature of a coherent wave field when it interacts with matter. That is an alternative insight to understand why so many nonlinear optical effects (especially the coherent wave mixing effects in nonlinear media) can be observed only by using laser radiation but not ordinary optical radiation.

In summary, on the one hand, the parameters of laser radiation (such as power, beam divergence, pulse duration, wavelength, spectral width, polarization status, etc.) can be well controlled or modified based on existing laser techniques. On the one hand, there is a great variety of nonlinear optical media, which can be various materials (inorganic, organic, biological, etc.), in different physical states (solid, liquid, gas, plasma, liquid crystal, etc.), and with different reaction centers (molecules, atoms, ions, atomic nuclei, electrons, color centers, phonons, excitons, plasmons, etc.). So that, it is not surprising that so many new effects and novel phenomena in nonlinear optics have been found within only three to four decades since the 1960's.[81-112] These effects and phenomena are related to the intense light-induced opto-optical, opto-electric, opto-magnetic, opto-acoustic, opto-thermal, opto-mechanical, opto-chemical, and opto-biological interactions in optical media. Generally speaking, all these kinds of interactions can be used to develop various new techniques that may provide many advantages, such as high efficiency, high resonant selectivity, high spectral resolution, high temporal resolution, high spatial resolution, and high sensitivity.

1.4 THEORETICAL FRAMEWORK OF NONLINEAR OPTICS

Basically, two major theoretical approaches can be employed in nonlinear optics as well as in laser physics.[81-112] The first is the semi-classical theory, and the second is the quantum electrodynamical theory. The most essential feature of the semi-classical theory is that the media composed of atoms or molecules are described by the theory of quantum mechanics, while the light radiation is described by the classical Maxwell's theory. The key issue of semi-classical theory in nonlinear optics is to give the expressions of macroscopic nonlinear electric polarization for optical media. For this purpose, the density matrix method, which is a special approach based on both quantum mechanics and statistical physics, is used to derive the expressions for various orders of electric susceptibilities as $\chi^{(1)}$, $\chi^{(2)}$, $\chi^{(3)}$..., and the expressions for various orders of polarization components as

$\mathbf{P}^{(1)}$, $\mathbf{P}^{(2)}$, $\mathbf{P}^{(3)}$...and so on. Substituting the appropriate nonlinear polarization components into the generalized wave equations, we are able, in principle, to predict many possible nonlinear optical responses of the medium for a given condition of the input intense optical field(s).

By contrast, the quantum theory of radiation in the regime of quantum electrodynamics treats the medium and optical field as a combined and quantized system. In other words, both the medium and the optical field should be described in the way of quantum mechanics. As a result, the wave function of the combined system is expressed as the product of the eigen function of a molecular system and the eigen function of a quantized photon field. In this case, the key issue is to determine the probability of state change of the combined system due to interaction between the photon field and the medium. Usually, the state changes of the combined system are related to the transition of molecular system from its initial eigen state to the final state and the simultaneous changes of the photon numbers among different photon modes.

It should be pointed that there is no major contradiction or inconsistency between the results and conclusions given by these two theoretical approaches. In fact, they can give the same quantitative results in many cases, such as the cross section of Raman scattering as well as the cross section of two-photon absorption. Nevertheless, these two theoretical approaches have their own usefulness and shortcomings. In this sense, these two different theoretical regimes are parallel and complementary to each other in the scope of nonlinear optics.

The most successful example of the semi-classical theory in nonlinear optics is the derivation of quantitative expressions for various orders of nonlinear susceptibilities of optical media. A semi-classical theoretical approach can be employed to explain all those nonlinear optical effects and phenomena, which are related to nonlinear electric polarization responses in the media, such as various coherent optical wave-mixing effects, and intense light-induced refractive index change effects. Nevertheless, there are some limitations inherently associated with the semi-classical theory. First, this theory cannot distinguish the difference between the stimulated and the spontaneous processes of radiation, scattering, and parametric photon emission. In order to describe the spontaneous processes, the correspondence principle has to be invoked in the semi-classical regime. For example, Einstein's coefficient relation has to be used to describe the difference between the probabilities of spontaneous emission and stimulated emission. Second, some important physical facts (such as transition relaxation and spectral linewidth) can only be considered by introducing a phenomenological damping factor into the equation of density matrix. Finally, there are a number of nonlinear optical effects (such as stimulated Raman scattering, stimulated Brillouin scattering, CARS process, two-photon absorption, third-harmonic generation, as well as induced refractive index changes), all of them can be described with a nominal third-order nonlinear susceptibility $\chi^{(3)}$. In these cases, however, the nonlinear polarization theory can not reveal the essential differences in origins and mechanisms of those entirely different effects. As a result of that failure, sometimes one may find confusion and terminological ambiguity in classification and description of some nonlinear optical processes within the regime of semi-classical theory.

In quantum electrodynamics, the quantum theory of radiation is a more rigorous theoretical approach that, in principle, can be perfectly used to explain or describe any kinds of effects and phenomena related to the interaction of radiation field with matter in both qualitative and quantitative ways. There are many well known examples that have shown the advantage of the quantum theory of radiation over the semi-classical theory. First, the relationship between stimulated emission (or scattering) and spontaneous emission (or scattering) can be naturally derived without the need of using the correspondence principle. Second, the selection rule, life-time of state, and spectral linewidth can be quantitatively determined for a given molecular system. Third, the conservation of energy and momentum between the photon field and molecular system can be logically applied to various nonlinear optical processes without the need of using the so-called Manley-Rowe relation (for conservation of energy) and the phase-matching requirement (for conservation of momentum). Finally, the most important feature of the quantum theory of radiation is that a concept of virtual energy level can be introduced, which represents an intermediate quantum state occupied by the combined system of the photon field and the medium. Based on the concept of virtual energy level or intermediate state, the principles and mechanisms of most major nonlinear optical effects can be consistently interpreted and, in many cases, clearly illustrated by an energy-level diagram involving the transitions via virtual energy levels. On the other hand, however, the mathematical derivation in the regime of quantum electrodynamics is rather lengthy and cumbersome for some specific issues. Therefore, in practice the all-quantum derivations of the related formulae are only applied in those cases where the semi-classical approach is obviously poor or failed.

Throughout this book we intend to use both of these two theoretical approaches to describe various nonlinear optical effects and phenomena in a complementary way. In many cases we will briefly discuss the principles or mechanisms of nonlinear optical processes in terms of conceptions of quantum elecrodynamics; in the meantime, we will also use the semi-classical approach of nonlinear polarization to present those formulae widely used in nonlinear optics. Only in some special cases, such as stimulated Raman scattering and stimulated Kerr scattering, the method of quantum theory of radiation is fully used to give a rigorous quantitative description.

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