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Retarded Electromagnetic Interaction and Symmetry Violation of Time Reversal in High Order Stimulated Radiation and Absorption Processes of Lights as Well as Nonlinear Optics – Influence on Fundamental Theory of Laser and Non-Equilibrium Statistical Physics

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1. Introduction

Based on the perturbation method of quantum mechanics and retarded electromagnetic interaction, it is proved that the transition probabilities of light's high order stimulated radiation and absorption are not the same [1]. It indicates that the processes of light's stimulated radiation and absorption as well as nonlinear optics violate time reversal symmetry actually, although the motion equation of quantum mechanics and the interaction Hamiltonian are still invariable. This result can be used to solve the famous irreversibility paradox in the evolution processes of macro-systems which has puzzled physics community for a long time.

Einstein put forward the theory of light's stimulated radiation and absorption in 1917 in order to explain the Planck blackbody radiation formula based on equilibrium theory. According to the Einstein's theory, the parameters of stimulated radiation and absorption are equal to each other with $B_{ml} = B_{lm}$. The same result can also be obtained by means of the calculation of quantum mechanics for the first order process under dipole approximation without considering the retarded interaction (or multiple moment effect) of radiation fields [1]. Because light's stimulated radiation process can be regarded as the time reversal of stimulated absorption process, the result means that light's stimulated radiation and absorption processes have time reversal symmetry.

Nonlinear optics was developed in the 1960's since laser was invented. Also by the dipole approximation without considering the retarded interaction of radiation fields, nonlinear susceptibilities in nonlinear optics are still invariable under time reversal [2]. So the processes of light's radiation and absorption as well as nonlinear optics are considered to be time reversal symmetry at present. In fact, it is a common and wide accepted idea at present that all micro-processes controlled by electromagnetic interaction are symmetrical under time reversal, for the motion equations of quantum mechanics and the Hamiltonian of electromagnetic interaction are unchanged under time reversal.

However, most processes related to laser and nonlinear optics are actually high non-equilibrium ones. As we known that time reversal symmetry will generally be violated in non-equilibrium processes. It is proved in this paper that after the retarded effect of radiation fields is taken into account, the time reversal symmetry will be violated in light's high order stimulated radiation and absorption processes with $B_{ml} \neq B_{lm}$, although the Hamiltonian of electromagnetic interaction is still unchanged under time reversal.

The transition probability of third order process is calculated and the revised formula of nonlinear optics polarizability is deduced in this paper. Many phenomena of time reversal symmetry violation in non-linear optics just as sum frequency, double frequency, different frequencies, double stable states, self-focusing and self-defocusing, echo phenomena, as well as optical self-transparence and self absorptions and so on are analyzed.

The reason to cause symmetry violation is that some filial or partial transition processes of bounding state atoms are forbidden or can't be achieved due to the law of energy conservation. These restrictions can cause the symmetry violation of time reversal of other partial transition processes which can be actualized really. These realizable filial or partial processes which violate time reversal symmetry generally are just the practically observed physical processes. The symmetry violation is also relative to the initial state's asymmetries of bounding atoms before and after time reversal. For the electromagnetic interaction between non-bounding atoms and radiation fields, there is no this kind of symmetry violation of time reversal. For example, in the experiments of particle physics in accelerators, we can not observe the symmetry violation of time reversal.

At last, the influences of symmetry violation of time reversal on the foundation theory of laser and non-equilibrium statistical physics are discussed. The phenomena of producing laser without the reversion of particle population and transition without radiation can be well explained. The result indicates that the irreversibility of evolution processes of macro-systems originates from the irreversibility of micro-processes. The irreversibility paradox can be eliminated thoroughly. By introducing retarded electromagnetic interaction, the forces between classical changed particles are not conservative ones. Based on them, we can establish the revised Liouville equation which is irreversible under time reversal. In this way, we can lay a really rational dynamic foundation for classical non-equilibrium statistical mechanics.

2. The transition probability of the first order process

For simplification, we consider an atom with an electron in its external layer. Electron's mass is μ , charge is q . When there is no external interaction, the Hamiltonian and the wave function of electron are individually

$$\hat{H}_0 = -\frac{\hbar^2}{2m}\nabla^2 + \hat{U}(r) \qquad |\psi_0\rangle = \sum_n e^{-\frac{1}{\hbar}E_n t}|n\rangle \tag{1}$$

After external electromagnetic field is introduced, the interaction Hamiltonian is

$$\hat{H}' = -\frac{q}{c\mu}\hat{A}\cdot\hat{p} + \frac{q^2}{2c^2\mu}\hat{A}^2 + q\phi \tag{2}$$

Because the charge and current densities of radiation field are zero, we can take the gauge condition $\nabla\cdot\hat{A} = 0$ and $\phi = 0$ and write $\hat{H}' = \hat{H}_1' + \hat{H}_2'$ with

$$\hat{H}_1' = -\frac{q}{c\mu}\hat{A}\cdot\hat{p} \qquad \hat{H}_2' = \frac{q^2}{2c^2\mu}\hat{A}^2 \tag{3}$$

Where $\hat{H}_1'$ has the order v/c and $\hat{H}_2'$ has the order v^2/c^2. In the current discussion for light's stimulated radiation and absorption theory, $\hat{H}_2'$ is neglected generally. Because $\hat{H}_2'$ has the same order of magnitude as the second order effects of nonlinear optics, it is remained in the paper. Suppose that electromagnetic wave propagates along $\vec{k}$ direction. Electric field strength is $\vec{E} = \vec{E}_0\sin(\omega t - \vec{k}\cdot\vec{R})$. Here $\vec{R}$ is a direction vector pointing from wave source to observation point. Both $\hat{H}_1'$ and $\hat{H}_2'$ can also be written as

$$\hat{H}_1' = -\frac{q\vec{E}_0}{2\omega\mu}\cdot\left[e^{i\left(\omega t-\vec{k}\cdot\vec{R}\right)} + e^{-i\left(\omega t-\vec{k}\cdot\vec{R}\right)}\right]\hat{p}$$

$$\hat{H}_2' = \frac{q^2\vec{E}_0^2}{2c^2k^2\mu}\left[e^{i\left(\omega t-\vec{k}\cdot\vec{R}\right)} + e^{-i\left(\omega t-\vec{k}\cdot\vec{R}\right)}\right]^2 = \frac{q^2\vec{E}_0^2}{2\omega^2\mu}\left[e^{i2\left(\omega t-\vec{k}\cdot\vec{R}\right)} + e^{-i2\left(\omega t-\vec{k}\cdot\vec{R}\right)} + 2\right] \tag{4}$$

we can write

$$\hat{H}_1' = \hat{F}_1 e^{i\omega t} + \hat{F}_1^+ e^{-i\omega t}$$

in which

$$\hat{F}_1 = -\frac{q\vec{E}_0}{2\omega\mu}\cdot e^{-i\vec{k}\cdot\vec{R}}\hat{p} \qquad \hat{F}_1^+ = -\frac{q\vec{E}_0}{2\omega\mu}\cdot e^{i\vec{k}\cdot\vec{R}}\hat{p} \tag{5}$$

Because we always have $\vec{k}\perp\vec{E}_0$ for electromagnetic wave, we have the commutation relation

$$\left[\vec{k}\cdot\vec{R},\ \vec{E}_0\cdot\hat{p}\right] = i\hbar\vec{k}\cdot\vec{E}_0 = 0 \tag{6}$$

So it can be proved that $\vec{E}_0\cdot\hat{p}$ and $\exp(i\vec{k}\cdot\vec{R})$ are also commutative. In this way, (6) can also be written as

$$\hat{F}_1 = -\frac{q\vec{E}_0}{2\omega\mu}\cdot\hat{p}e^{-i\vec{k}\cdot\vec{R}} \quad \hat{F}_1^+ = -\frac{q\vec{E}_0}{2\omega\mu}\cdot\hat{p}e^{i\vec{k}\cdot\vec{R}} \tag{7}$$

By using perturbation method in quantum mechanics to regard $\hat{H}'$ as perturbation, we write the motion equation and wave function of system as

$$i\hbar\frac{\partial}{\partial t}|\psi\rangle=\left(\hat{H}_0+\hat{H}'\right)|\psi\rangle \qquad |\psi\rangle=\sum_m a_m(t)e^{-\frac{i}{\hbar}E_m t}|n\rangle \tag{8}$$

Let

$$a_m(t)=a_m^{(0)}(t)+a_m^{(1)}(t)+a_m^{(2)}(t)+\cdots,\ E_m-E_n=\hbar\omega_{mn},$$

Substituting them in the motion equation, we can get

$$i\hbar\frac{d}{dt}\left[a_m^{(0)}(t)+a_m^{(1)}(t)+a_m^{(2)}(t)+\cdots\right]=\sum_n\left(\hat{H}'_{1mn}+\hat{H}'_{2mn}\right)\left[a_n^{(0)}(t)+a_n^{(1)}(t)+a_n^{(2)}(t)+\cdots\right]e^{i\omega_{mn}t} \tag{9}$$

The items with same order on the two sides of the equation are taken to be equal to each other. The first four equations are

$$i\hbar\frac{d}{dt}a_m^{(0)}(t)=0 \qquad i\hbar\frac{d}{dt}a_m^{(1)}(t)=\sum_n\hat{H}'_{1mn}a_n^{(0)}(t)e^{i\omega_{mn}t} \tag{10}$$

$$i\hbar\frac{d}{dt}a_m^{(2)}(t)=\sum_n\hat{H}'_{2mn}a_n^{(0)}(t)e^{i\omega_{mn}t}+\sum_n\hat{H}'_{1mn}a_n^{(1)}(t)e^{i\omega_{mn}t} \tag{11}$$

$$i\hbar\frac{d}{dt}a_m^{(3)}(t)=\sum_n\hat{H}'_{2mn}a_n^{(1)}(t)e^{i\omega_{mn}t}+\sum_n\hat{H}'_{1mn}a_n^{(2)}(t)e^{i\omega_{mn}t} \tag{12}$$

Suppose that an electron is in the initial state $|l\rangle$ with energy E_l at time $t=0$, then the electron transits into the final state $|m\rangle$ with energy E_m at time t, we have $a_m^{(0)}(t)=\delta_{ml}$ from the first formula of (10). Put it into the second formula, the probability amplitude of first order process is

$$a_m^{(1)}(t)=\frac{1}{i\hbar}\int_0^t\hat{H}'_{1ml}e^{i\omega_{ml}t}dt=-\frac{\hat{F}_{1ml}\left[e^{i(\omega+\omega_{ml})t}-1\right]}{\hbar(\omega+\omega_{ml})}+\frac{\hat{F}^{+}_{1ml}\left[e^{-i(\omega-\omega_{ml})t}-1\right]}{\hbar(\omega-\omega_{ml})} \tag{13}$$

Where

$$\hat{F}_{1ml}=\langle m|\hat{F}_1|l\rangle,\ \hat{F}^{+}_{1ml}=\langle m|\hat{F}^{+}_1|l\rangle.$$

The formula represents the probability amplitude of an electron transiting from the initial state $|l\rangle$ into the final state $|m\rangle$. In current theory, the so-called rotation wave approximation is used, i.e., only the first item is considered when $\omega=-\omega_{ml}$ and the second one is considered when $\omega=\omega_{ml}$ in (13). But up to now we have not decided which one is the state of high-energy level and which one is the state of low-energy level. In fact, electron can either transit into higher-energy state $|m\rangle$ from low-energy state $|l\rangle$ by absorbing a photon, or transit into low-energy state $|m\rangle$ from high-energy state $|l\rangle$ by emitting a photon. Because photon's energy is always positive, by the law of energy conservation, it is proper for us to think that the condition $\omega = \omega_{ml}$ corresponds to the situation with $E_m > E_l$, in which an electron transits into the high-energy final state $|m\rangle$ from the low-energy initial state $|l\rangle$ by absorbing a photon with energy $\hbar\omega_{ml} = E_m - E_l > 0$. This is just the simulated absorption process with the transition probability in unit time

$$W^{(1)}_{\omega=\omega_{ml}} = \frac{2\pi}{\hbar^2}\left|\hat{F}^{+}_{1ml}\right|^2 \delta(\omega - \omega_{ml}) \tag{14}$$

Therefore, the condition $\omega = -\omega_{ml}$ corresponds to the situation with $E_m < E_l$, in which an electron transits from the high-energy initial state $|l\rangle$ into the low-energy final state $|m\rangle$ by emitting a photon with energy $-\hbar\omega_{ml} = \hbar\omega_{lm} = E_l - E_m > 0$. This is just the simulated radiation process with the transition probability in unit time

$$W^{(1)}_{\omega=-\omega_{ml}} = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2 \delta(\omega + \omega_{lm}) \tag{15}$$

So $W^{(1)}_{\omega=\omega_{ml}}$ and $W^{(1)}_{\omega=-\omega_{ml}}$ represent the different physical processes. It is necessary for us to distinguish the physical meanings of $W^{(1)}_{\omega=\omega_{ml}}$ and $W^{(1)}_{\omega=-\omega_{ml}}$ clearly for the following discussion. As shown in Fig.1, we image a system with three energy levels: medium energy level E_l, high energy $E_m(up)$ and low energy $E_m(down)$. The difference of energy levels between E_l and $E_m(up)$ is the same as that between $E_m(down)$ and E_l. Suppose that the electron is in medium energy level at beginning. Stimulated by radiation field, the electron can either transit up into high-energy level or down into low energy level. In this case, $W^{(1)}_{\omega=\omega_{ml}}$ represents the probability the electron transits up into high-energy level and $W^{(1)}_{\omega=-\omega_{ml}}$ represents the probability the electron transits down into low-energy level.

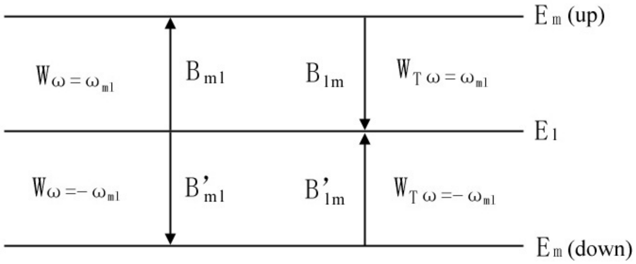

Fig. 1. Electron's transitions among three energy levels.

For visible light with wavelength $\lambda \sim 10^{-7}m$ and common atoms with radius $R \sim 10^{-10}m$, we have $\vec{k}\cdot\vec{R} \sim 10^{-3} << 1$. So in the current theory, dipolar approximation $\exp i\vec{k}\cdot\vec{R} \sim 1$ is used. However, it should be noted that the ratio of magnitude between the first order processes and the second order processes in nonlinear optics is just about 10^{-3}. Meanwhile, for the interaction between external fields and electrons in atoms, such as the situations of laser and nonlinear optics, we have $R \approx 0.1 \sim 1m$ so that $\vec{k}\cdot\vec{R} = 10^6 \sim 10^7$ with the macro-order of magnitude. In fact, factor $\vec{k}\cdot\vec{R}$ represents the retarded interaction of electromagnetic field. It can't be neglected in general in the problems of laser and nonlinear optics. It will be seen below that it is just this factor which would play an important role in the symmetry violation of time reversal in light's absorption and radiation processes

Let $\vec{R}_0$ represents the distance vector pointing from radiation source to atomic mass center, $\vec{r}$ represents the distance vector pointing from atomic mass center to electron, we have $\vec{R} = \vec{R}_0 + \vec{r}$. For the interaction process between external electromagnetic field and atom in medium, we have $R_0 = 0.1 \sim 1m$, $\vec{k} \cdot \vec{R}_0 = 10^6 \sim 10^7 >> 1$ and $\vec{k} \cdot \vec{r} << 1$. If radiation fields come from atomic internal, we have $\vec{R}_0 \approx 0$, $\vec{k} \cdot \vec{r} << 1$. In the following discussion, we approximately take:

$$e^{-i\vec{k}\cdot\vec{R}} \approx e^{-i\vec{k}\cdot\vec{R}_0}\left[1 - i\vec{k}\cdot\vec{r} - \left(\vec{k}\cdot\vec{r}\right)^2/2\right] \tag{16}$$

Here $\vec{k} = \omega\vec{\tau} / c$, $\vec{\tau}$ is unit direction vector. By considering relations $\omega_{lm} = -\omega_{ml}$, $\hat{p} = -i\hbar\nabla$ and

$$\langle m|\hat{p}|l\rangle = \mu\langle m|d\hat{r} / dt|l\rangle = \frac{\mu}{i\hbar}\langle m|\left[\hat{r},\ \hat{H}_0\right]\ |l\rangle = i\mu\omega_{ml}\langle m|\hat{r}|l\rangle \tag{17}$$

as well as (7), we can get

$$\hat{F}_{1ml} = \left[-\frac{iq\omega_{ml}}{2\omega}\vec{E}_0\cdot\langle m|\vec{r}|l\rangle + \frac{q\hbar}{2c\mu}\vec{E}_0\cdot\langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle - \frac{iq\hbar\omega}{4c^2\mu}\vec{E}_0\cdot\langle m|\left(\vec{\tau}\cdot\vec{r}\right)^2\nabla|l\rangle\right]e^{-i\vec{k}\cdot\vec{R}_0} \tag{18}$$

$$\hat{F}^{+}_{1ml} = \left[-\frac{iq\omega_{ml}}{2\omega}\vec{E}_0\cdot\langle m|\vec{r}|l\rangle - \frac{q\hbar}{2c\mu}\vec{E}_0\cdot\langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle - \frac{iq\hbar\omega}{4c^2\mu}\vec{E}_0\cdot\langle m|\left(\vec{\tau}\cdot\vec{r}\right)^2\nabla|l\rangle\right]e^{i\vec{k}\cdot\vec{R}_0} \tag{19}$$

The first item is the result of dipolar moment interaction. The second item is the result of quadrupolar moment interaction and the third item is the result of octupolar moment interaction. The wave functions of stationary states $|m\rangle$ and $|l\rangle$ have fixed parities. The parities of operator $\vec{r}$ and $\left(\vec{\tau}\cdot\vec{r}\right)^2\nabla$ are odd and the parity of operator $\vec{\tau}\cdot\vec{r}\nabla$ is even. So by the consideration of symmetry, if matrix element $\langle m|\vec{r}|l\rangle \neq 0$, we would have $\langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle = 0$ and $\langle m|\left(\vec{\tau}\cdot\vec{r}\right)^2\nabla|l\rangle \neq 0$. Conversely, if $\langle m|\vec{r}|l\rangle = 0$, we would have $\langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle \neq 0$ and $\langle m|\left(\vec{\tau}\cdot\vec{r}\right)^2\nabla|l\rangle = 0$. Suppose $\langle m|\vec{r}|l\rangle \neq 0$, $\langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle = 0$ and $\langle m|\left(\vec{\tau}\cdot\vec{r}\right)^2\nabla|l\rangle \neq 0$, we have $\hat{F}_{1ml} \neq \hat{F}^{+}_{1ml}$ but $\left|\hat{F}_{1ml}\right|^2 = \left|\hat{F}^{+}_{1ml}\right|^2$. So after retarded interaction is taken into account for the first order processes, we still have $W^{(1)}_{\omega=\omega_{ml}} = W^{(1)}_{\omega=-\omega_{ml}}$, i.e., the transition probabilities of stimulated radiation and stimulated absorption are still the same.

3. The time reversal of the first order process

Let's discuss the time reversal of the first order process below. According to the standard theory of quantum electrodynamics, the time reversal of electromagnetic potential is $\vec{A}\left(\vec{x},t\right) \to -\vec{A}\left(\vec{x},-t\right)$. Meanwhile, we have $\hat{p} \to -\hat{p}$ when $t \to -t$. The propagation direction of electromagnetic wave should be changed from $\vec{k}$ to $-\vec{k}$ under time reversal (Otherwise retarded wave would become advanced wave so that the law of causality would be violated.). Let subscript T represent time reversal, from (3), (5) and (6), we have $\hat{H}'_{1T}(\vec{x},t)=\hat{H}'_1(\vec{x},t)$ and $\hat{H}'_{2T}(\vec{x},t)=\hat{H}'_2(\vec{x},t)$. The interaction Hamiltonian is unchanged under time reversal. On the other hand, when $t\to -t$, (8) becomes

$$-i\hbar\frac{\partial}{\partial t}|\psi\rangle_T=\left(\hat{H}_{0T}+\hat{H}'_T\right)|\psi\rangle_T \qquad |\psi\rangle_T=\sum_n a_n(-t)e^{\frac{i}{\hbar}E_n t}|n\rangle \tag{20}$$

Let $a_m(-t)=a_m^{(0)}(-t)+a_m^{(1)}(-t)+a_m^{(2)}(-t)+\cdots$ and put it into the formula above, the motion equation becomes

$$-i\hbar\frac{d}{dt}\left[a_m^{(0)}(-t)+a_m^{(1)}(-t)+a_m^{(2)}(-t)+\cdots\right]$$

$$=\sum_n\left(H'_{1Tmn}+H'_{2Tmn}\right)\left[a_n^{(0)}(-t)+a_n^{(1)}(-t)+a_n^{(2)}(-t)+\cdots\right]e^{-i\omega_{mn}t} \tag{21}$$

Take index replacements $m\to l$ and $n\to k$ in the formula, then let the items with same order to be equal to each other, we get

$$-i\hbar\frac{d}{dt}a_l^{(0)}(-t)=0 \qquad -i\hbar\frac{d}{dt}a_l^{(1)}(-t)=\sum_k\hat{H}'_{1Tlk}a_k^{(0)}(-t)e^{-i\omega_{lk}t} \tag{22}$$

$$-i\hbar\frac{d}{dt}a_l^{(2)}(-t)=\sum_k\hat{H}'_{2Tlk}a_k^{(0)}(-t)e^{-i\omega_{lk}t}+\sum_k\hat{H}'_{1Tlk}a_k^{(1)}(-t)e^{-i\omega_{lk}t} \tag{23}$$

$$-i\hbar\frac{d}{dt}a_l^{(3)}(-t)=\sum_k\hat{H}'_{2Tlk}a_k^{(1)}(-t)e^{-i\omega_{lk}t}+\sum_k\hat{H}'_{1Tlk}a_k^{(2)}(-t)e^{-i\omega_{lk}t} \tag{24}$$

On the other hand, under time reversal, the initial state becomes $|m\rangle$ with $a_k^{(0)}(-t)=\delta_{km}$. Put it into the second formula of (22), we get

$$a_l^{(1)}(-t)=-\frac{1}{i\hbar}\int_0^t\hat{H}'_{1Tlm}e^{-i\omega_{lm}t}dt=-\frac{1}{i\hbar}\int_0^t\hat{H}'_{1lm}e^{-i\omega_{lm}t}dt \tag{25}$$

Because $\hat{H}'_1$ is the Hermitian operator with

$$\hat{H}'_{1lm}=\langle l|\hat{H}'_1|m\rangle=\langle m|\hat{H}'_1|l\rangle^*=\hat{H}'^*_{1ml},$$

we can write

$$\hat{H}'^*_{1ml}=\hat{F}'_{1ml}e^{i\omega t}+\hat{F}'^+_{1ml}e^{-i\omega t}$$

and get

$$\hat{F}'_{1ml}=\left(\hat{F}^+_{1ml}\right)^*=\left[\frac{iq\omega_{ml}}{2\omega}\vec{E}_0\cdot\langle m|\vec{r}|l\rangle^*-\frac{q\hbar}{2c\mu}\vec{E}_0\cdot\langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle^*+\frac{iq\hbar\omega}{4c^2\mu}\vec{E}_0\cdot\langle m|(\vec{\tau}\cdot\vec{r})^2\nabla|l\rangle^*\right]e^{-i\vec{k}\cdot\vec{R}_0} \tag{26}$$

$$\hat{F}'^{+}_{1ml} = \left(\hat{F}_{1ml}\right)^* = \left[\frac{iq\omega_{ml}}{2\omega}\vec{E}_0 \cdot \langle m|\vec{r}|l\rangle^* + \frac{q\hbar}{2c\mu}\vec{E}_0 \cdot \langle m|\vec{\tau}\cdot\vec{r}\nabla|l\rangle^* + \frac{iq\hbar\omega}{4c^2\mu}\vec{E}_0 \cdot \langle m|(\vec{\tau}\cdot\vec{r})^2\nabla|l\rangle^*\right]e^{i\vec{k}\cdot\vec{R}_0} \quad (27)$$

Let $a_{mT}(t)$ represent the time reversal of amplitude $a_m(t)$. Because the original final state becomes $|l\rangle$ and the original initial state becomes $|m\rangle$ under time reversal, we have $a^{(1)}_{mT}(t) = a^{(1)}_l(-t)$. So according to (25), after time reversal, the transition amplitude becomes

$$\begin{aligned} a^{(1)}_{mT}(t) &= -\frac{1}{i\hbar}\int_0^t \hat{H}'^{*}_{1ml} e^{i\omega_{ml}t}dt = \frac{\hat{F}'_{1ml}\left[e^{i(\omega+\omega_{ml})t}-1\right]}{\hbar(\omega+\omega_{ml})} - \frac{\hat{F}'^{+}_{1ml}\left[e^{-i(\omega-\omega_{ml})t}-1\right]}{\hbar(\omega-\omega_{ml})} = \\ &= \frac{\hat{F}'_{1ml}\left[e^{i(\omega-\omega_{lm})t}-1\right]}{\hbar(\omega-\omega_{lm})} - \frac{\hat{F}'^{+}_{1ml}\left[e^{-i(\omega+\omega_{lm})t}-1\right]}{\hbar(\omega+\omega_{lm})} \end{aligned} \quad (28)$$

So the condition $\omega = -\omega_{lm} = \omega_{ml}$ corresponds to the situation with $E_m > E_l$, indicating that an electron emits a photon with energy $-\hbar\omega_{lm} = E_m - E_l > 0$ and transits from the initial high-energy state $|m\rangle$ into the final low-energy state $|l\rangle$. This process is the time reversal of stimulated absorption process described by (14). By considering (27), the transition probability in unite time is

$$W^{(1)}_{T\omega=\omega_{ml}} = \frac{2\pi}{\hbar^2}\left|\hat{F}'^{+}_{1ml}\right|^2 \delta(\omega+\omega_{lm}) = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2 \delta(\omega-\omega_{ml}) \quad (29)$$

Comparing with (14) and considering the result $\left|\hat{F}_{1ml}\right|^2 = \left|\hat{F}^{+}_{1ml}\right|^2$, we still have $W^{(1)}_{T\omega=\omega_{ml}} = W^{(1)}_{\omega=\omega_{ml}}$. Because we define the time reversal of stimulated absorption process as the stimulated radiation process, the result shows that the transition probability of stimulated absorption is equal to that of stimulated radiation after time reversal for the first order process when retarded interaction is considered. The process is unchanged under time reversal.

Similarly, the condition $\omega = \omega_{lm} = -\omega_{ml}$ corresponds to the situation with $E_m < E_l$, indicating that an electron emits a photon with energy $\hbar\omega_{lm} = E_l - E_m > 0$ and transits from the initial low-energy state $|m\rangle$ into the final high-energy state $|l\rangle$. This process is the time reversal of stimulated radiation process described by (15). By considering (26), the transition probability in unite time is

$$W^{(1)}_{T\omega=-\omega_{ml}} = \frac{2\pi}{\hbar^2}\left|\hat{F}'_{1ml}\right|^2 \delta(\omega-\omega_{lm}) = \frac{2\pi}{\hbar^2}\left|\hat{F}^{+}_{1ml}\right|^2 \delta(\omega+\omega_{ml}) \quad (30)$$

After retarded effect is considered, we also have $W^{(1)}_{T\omega=-\omega_{ml}} = W^{(1)}_{\omega=-\omega_{ml}}$ for the first order process. The process is unchanged under time reversal.

The transition relations can be seen clearly in Fig.1. There are two stimulated absorption parameters B_{ml}, B'_{lm} and two stimulated radiation parameters B_{lm}, B'_{ml}. The initial and final states of B_{ml} and B_{lm} are opposite. So they are time reversal states. The initial and final states of B'_{ml} and B'_{lm} are also opposite, so they are also time reversal states. Therefore, if B_{ml} is defined as stimulated absorption parameter, B_{lm} should be defined as stimulated radiation parameter for their initial final states are just opposite. Similarly, if B'_{ml} is defined as stimulated absorption parameter, B'_{lm} should be defined as stimulated radiation parameter. Meanwhile, if B_{ml} (or B'_{lm}) is defined as stimulated absorption parameter, B'_{ml} (or B_{lm}) should not be defined as stimulated radiation parameter, for they have same initial and final states and do not describe corresponding stimulated radiation and absorption processes. For the first order process, we have $B_{ml} = B_{lm} = B'_{lm} = B'_{ml}$. But as shown below that in the high order processes, this relation can't hold.

4. The transition probability of the second order process

The second order processes are discussed below. We write $\hat{H}'_2 = \hat{F}_2 e^{2i\omega t} + \hat{F}_2^+ e^{-2i\omega t} + \hat{F}_0$, in which

$$\hat{F}_2 = \frac{q^2 E_0^2}{2\omega^2 \mu} e^{-i2\vec{k}\cdot\vec{R}} \qquad \hat{F}_2^+ = \frac{q^2 E_0^2}{2\omega^2 \mu} e^{i2\vec{k}\cdot\vec{R}} \qquad \hat{F}_0 = \frac{q^2 E_0^2}{\omega^2 \mu} \tag{31}$$

When $m \neq l$ we have $\langle m | l \rangle = 0$. Suppose $\langle m | \vec{\tau}\cdot\vec{r} | l \rangle \neq 0$, we have $\langle m | (\vec{\tau}\cdot\vec{r})^2 | l \rangle = 0$. According to (16), we have

$$\hat{F}_{2ml} = -\frac{iq^2 E_0^2}{c\omega\mu} e^{-i2\vec{k}\cdot\vec{R}_0} \langle m | \vec{\tau}\cdot\vec{r} | l \rangle \quad \hat{F}_{2ml}^+ = \frac{iq^2 E_0^2}{c\omega\mu} e^{i2\vec{k}\cdot\vec{R}_0} \langle m | \vec{\tau}\cdot\vec{r} | l \rangle \quad \hat{F}_{0ml} = 0 \tag{32}$$

Similarly, we suppose the initial condition is $a_n^{(0)} = \delta_{nl}$. Substituting (13) into (13) and taking the integral, we can obtain the transition probability amplitude of the second order process

$$\begin{aligned}
a_m^{(2)}(t) &= \frac{1}{i\hbar}\int_0^t \hat{H}'_{2ml} e^{i\omega_{ml}t} dt + \frac{1}{i\hbar}\sum_n \int_0^t \hat{H}'_{1mn} a_n^{(1)}(t) e^{i\omega_{mn}t} dt \\
&= -\frac{\hat{F}_{2ml}\left[e^{i(2\omega+\omega_{ml})t} - 1\right]}{\hbar(2\omega+\omega_{ml})} + \frac{\hat{F}_{2ml}^+\left[e^{-i(2\omega-\omega_{ml})t} - 1\right]}{\hbar(2\omega-\omega_{ml})} \\
&+ \sum_n \frac{\hat{F}_{1mn}\hat{F}_{1nl}}{\hbar^2(\omega+\omega_{nl})}\left\{\frac{e^{i(2\omega+\omega_{nl}+\omega_{mn})\,t} - 1}{2\omega+\omega_{nl}+\omega_{mn}} - \frac{e^{i(\omega+\omega_{mn})\,t} - 1}{\omega+\omega_{mn}}\right\} \\
&- \sum_n \frac{\hat{F}_{1mn}\hat{F}_{1nl}^+}{\hbar^2(\omega-\omega_{nl})}\left\{\frac{e^{i(\omega_{nl}+\omega_{mn})\,t} - 1}{\omega_{nl}+\omega_{mn}} - \frac{e^{i(\omega+\omega_{mn})\,t} - 1}{\omega+\omega_{mn}}\right\} \\
&+ \sum_n \frac{\hat{F}_{1mn}^+\hat{F}_{1nl}}{\hbar^2(\omega+\omega_{nl})}\left\{\frac{e^{i(\omega_{nl}+\omega_{mn})\,t} - 1}{\omega_{nl}+\omega_{mn}} + \frac{e^{-i(\omega-\omega_{mn})\,t} - 1}{\omega-\omega_{mn}}\right\} \\
&+ \sum_n \frac{\hat{F}_{1mn}^+\hat{F}_{1nl}^+}{\hbar^2(\omega-\omega_{nl})}\left\{\frac{e^{-i(2\omega-\omega_{nl}-\omega_{mn})\,t} - 1}{2\omega-\omega_{nl}-\omega_{mn}} - \frac{e^{-i(\omega-\omega_{mn})\,t} - 1}{\omega-\omega_{mn}}\right\}
\end{aligned} \tag{33}$$

The formula contains the transition processes of single photon's absorption and radiation with $\omega = \pm\omega_{ml}$ as well as the processes of double photon's absorption and radiation with $2\omega = \pm\omega_{ml}$. We only discuss the absorption process of a single photon here. By rotation wave approximation, only the items containing factor $\left(e^{-i(\omega-\omega_{ml})t} - 1\right)/(\omega - \omega_{ml})$ are remained. Let $n = l$ in the fifth and sixth items of the formula, the transition probability amplitude is

$$a_m^{(2)}(t)_{\omega=\omega_{ml}} = \frac{\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll} - \hat{F}_{1ll}^{+}\right)\left[e^{-i(\omega-\omega_{ml})t} - 1\right]}{\hbar^2\omega(\omega - \omega_{ml})} = \frac{\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll} - \hat{F}_{1ll}^{+}\right)\left[e^{-i(\omega-\omega_{ml})t} - 1\right]}{\hbar^2\omega_{ml}(\omega - \omega_{ml})} \tag{34}$$

Therefore, after the second order process is considered, the total transition probability amplitude is

$$a_m(t)_{\omega=\omega_{ml}} = a_m^{(1)}(t)_{\omega=\omega_{ml}} + a_m^{(2)}(t)_{\omega=\omega_{ml}} = \frac{\hat{F}_{1ml}^{+}\left[e^{-i(\omega-\omega_{ml})t} - 1\right]}{\hbar(\omega - \omega_{ml})}\left(1 + \frac{\hat{F}_{1ll} - \hat{F}_{1ll}^{+}}{\hbar\omega_{ml}}\right) \tag{35}$$

Because we have $\omega_{ll} = 0$ and $\langle l|\vec{r}|l\rangle = 0$, when $m = l$, the first item in (18) and (19) are equal to zero, but the second items are not equal to zero in general. By the consideration of symmetry, the third item are also zero, or can be neglected by comparing with the second item. So it is enough for us only to consider quadrupole moment interaction in this case. We have

$$\hat{F}_{1ll} - \hat{F}_{1ll}^{+} = \frac{q\hbar}{c\mu}\vec{E}_0 \cdot \langle l|\vec{\tau} \cdot \vec{r}\nabla|l\rangle \cos\vec{k} \cdot \vec{R}_0 = B_{1l} + iB_{2l} \tag{36}$$

Let

$$A_l^2 = B_{1l}^2 + 2\hbar\omega_{ml}B_{1l} + B_{2l}^2 \tag{37}$$

When $\omega = \omega_{ml}$ we get the transition probability of the second order stimulated absorption process

$$W_{\omega=\omega_{ml}}^{(2)} = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2\left\{1 + \frac{A_l^2}{\hbar^2\omega_{ml}^2}\right\}\delta(\omega - \omega_{ml}) \tag{38}$$

The magnitude order of the revised value of the second order process is estimated below. The wave function of bounding state's atoms can be developed into series with form $|l\rangle \sim \sum b_n(\theta,\phi)r^n$ in general. If $\langle l|\vec{\tau} \cdot \vec{r}\nabla|l\rangle \neq 0$, we have $\vec{\tau} \cdot \vec{r}\nabla|l\rangle \sim r\partial/(\partial r)|l\rangle \sim |l\rangle$ approximately, or $\langle l|\vec{\tau} \cdot \vec{r}\nabla|l\rangle \sim \langle l|l\rangle \sim 1$. By taking $\omega_{ml} = 10^{16}$, we have

$$\frac{A_l^2}{\hbar\omega_{ml}^2} \sim 4q^2E_0^2\left|\langle l|\vec{\tau} \cdot \vec{r}\nabla|l\rangle\right|^2/(c\mu\omega_{ml})^2 \sim 1.4 \times 10^{-26}E_0^2 \tag{39}$$

In weak electromagnetic fields with $E_0 << 10^{13} V/m$, the revised values of the second order processes can be neglected. When the fields are strong enough with $E_0 \approx 10^{12} \sim 10^{13} V/m$, the revised value is big enough to be observed. The revised factor A_l of the second order processes is only related to initial state, having nothing to do with final states. On the other hand, if the retarded effect of radiation fields is neglected with $\vec{k}\cdot\vec{r} \sim \vec{\tau}\cdot\vec{r} = 0$, we have $\langle l|\vec{\tau}\cdot\vec{r}\nabla|l\rangle = 0$, the revised value of the second order process vanishes.

5. The time reversal of the second order process

The time reversal of the second order process is discussed now. Under time reversal, the initial state becomes $a_k^{(0)}(-t) = \delta_{km}$. By means of relations $\hat{H}'_{1Tlm} = \hat{H}'^{*}_{1ml}$ and $\hat{H}'_{2Tlm} = \hat{H}'^{*}_{2ml}$, we get the time reversal of transition amplitude for the second order processes according to(11)

$$a_l^{(2)}(-t) = -\frac{1}{i\hbar}\int_0^t \hat{H}'_{2Tlm} e^{-i\omega_{lm} t} dt - \frac{1}{i\hbar}\sum_k \int_0^t \hat{H}'_{1Tlk} a_k^{(1)}(-t) e^{-i\omega_{lk} t} dt$$

$$= -\frac{1}{i\hbar}\int_0^t \hat{H}'^{*}_{2ml} e^{i\omega_{ml} t} dt - \frac{1}{i\hbar}\sum_k \int_0^t \hat{H}'^{*}_{1kl} a_k^{(1)}(-t) e^{i\omega_{kl} t} dt \tag{40}$$

Let

$$\hat{H}'^{*}_{2ml} = \hat{F}'_{2ml} e^{i2\omega t} + \hat{F}'^{+}_{2ml} e^{-i2\omega t} + \hat{F}'_{0ml},$$

when $m \neq l$, we have $\langle m|l\rangle = 0$ and

$$\hat{F}'_{2ml} = -\frac{iq^2 E_0^2}{c\omega\mu} e^{-i2\vec{k}\cdot\vec{R}_0} \langle m|\vec{\tau}\cdot\vec{r}|l\rangle^* \quad \hat{F}'^{+}_{2ml} = \frac{iq^2 E_0^2}{c\omega\mu} e^{i2\vec{k}\cdot\vec{R}_0} \langle m|\vec{\tau}\cdot\vec{r}|l\rangle^* \quad \hat{F}'_{0ml} = 0 \tag{41}$$

So the time reversal of transition probability amplitude of the second process is

$$a_{Tm}^{(2)}(t) = a_l^{(2)}(-t) = \frac{\hat{F}'_{2ml}\left[e^{i(2\omega+\omega_{ml})t} - 1\right]}{\hbar(2\omega+\omega_{ml})} - \frac{\hat{F}'^{+}_{2ml}\left[e^{-i(2\omega-\omega_{ml})t} - 1\right]}{\hbar(2\omega-\omega_{ml})}$$

$$+\sum_k \frac{\hat{F}'^{+}_{1mk}\hat{F}'^{+}_{1kl}}{\hbar^2(\omega-\omega_{mk})}\left\{\frac{e^{-i(2\omega-\omega_{mk}-\omega_{kl})\,t} - 1}{2\omega-\omega_{mk}-\omega_{kl}} - \frac{e^{-i(\omega-\omega_{kl})\,t} - 1}{\omega-\omega_{kl}}\right\}$$

$$-\sum_k \frac{\hat{F}'^{+}_{1mk}\hat{F}'_{1kl}}{\hbar^2(\omega-\omega_{mk})}\left\{\frac{e^{i(\omega_{mk}+\omega_{kl})\,t} - 1}{\omega_{mk}+\omega_{kl}} - \frac{e^{i(\omega+\omega_{kl})\,t} - 1}{\omega+\omega_{kl}}\right\}$$

$$+\sum_k \frac{\hat{F}'_{1mk}\hat{F}'^{+}_{1kl}}{\hbar^2(\omega+\omega_{mk})}\left\{\frac{e^{i(\omega_{mk}+\omega_{kl})\,t} - 1}{\omega_{mk}+\omega_{kl}} + \frac{e^{-i(\omega-\omega_{kl})\,t} - 1}{\omega-\omega_{kl}}\right\}$$

$$+\sum_k \frac{\hat{F}'_{1mk}\hat{F}'_{1kl}}{\hbar^2(\omega+\omega_{mk})}\left\{\frac{e^{i(2\omega+\omega_{mk}+\omega_{kl})\,t} - 1}{2\omega+\omega_{mk}+\omega_{kl}} - \frac{e^{i(\omega+\omega_{kl})\,t} - 1}{\omega+\omega_{kl}}\right\} \tag{42}$$

When $\omega = \omega_{ml}$, we take $k = m$ in the third and fifth items of the formula. Also by rotation wave approximation, the time reversal of probability amplitude is

$$a_{mT}^{(2)}(t)_{\omega=\omega_{ml}} = \frac{\hat{F}_{1ml}'^{+}\left(F_{1mm}' - F_{1mm}'^{+}\right)\left[e^{-i(\omega-\omega_{ml})t} - 1\right]}{\hbar^2\omega(\omega - \omega_{ml})} = \frac{\hat{F}_{1ml}'^{+}\left(F_{1mm}' - F_{1mm}'^{+}\right)\left[e^{-i(\omega-\omega_{ml})t} - 1\right]}{\hbar^2\omega_{ml}(\omega - \omega_{ml})} \tag{43}$$

The time reversal of the total stimulated absolution process is

$$a_{Tm}(t)_{\omega=\omega_{ml}} = a_{Tm}^{(1)}(t)_{\omega=\omega_{ml}} + a_{Tm}^{(2)}(t)_{\omega=\omega_{ml}} = \frac{\hat{F}_{1ml}'^{+}\left[e^{-i(\omega-\omega_{ml})t} - 1\right]}{\hbar(\omega - \omega_{ml})}\left(1 + \frac{\hat{F}_{1mm}' - \hat{F}_{1mm}'^{+}}{\hbar\omega_{ml}}\right) \tag{44}$$

Comparing with (34), because of $\hat{F}_{mm}' \neq \hat{F}_{ll}$, we have

$$a_{mT}(t)_{\omega=\omega_{ml}} \neq a_m(t)_{\omega=\omega_{ml}},$$

the transition probability amplitude can not keep unchanged. Similarly, by considering $\omega_{mm} = 0$ and from (26) and (27), we have

$$\hat{F}_{1mm}' - \hat{F}_{1mm}'^{+} = -\frac{q\hbar}{c\mu}\vec{E}_0 \cdot \langle m|\vec{\tau}\cdot\vec{r}\nabla|m\rangle^* \cos\vec{k}\cdot\vec{R}_0 = -B_{1m} + iB_{2m} \tag{45}$$

Let

$$A_m'^2 = B_{1m}^2 - 2\hbar\omega_{ml}B_{1m} + B_{2m}^2 \tag{46}$$

When $\omega = \omega_{ml}$, the time reversal of stimulated absorption probability of second process is

$$W_{T\omega=\omega_{ml}}^{(2)} = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}'^{+}\right|^2\left\{1 + \frac{A_m'^2}{\hbar^2\omega_{ml}^2}\right\}\delta(\omega - \omega_{ml}) = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2\left\{1 + \frac{A_m'^2}{\hbar^2\omega_{ml}^2}\right\}\delta(\omega - \omega_{ml}) \tag{47}$$

The revised factor A_m is also only relative to initial state. Because $A_m' \neq A_l$, we have

$$W_{\omega=\omega_{ml}}^{(2)}\left(\hat{F}_{1ml}, A_l\right) \neq W_{T\omega=\omega_{ml}}^{(2)}\left(\hat{F}_{1ml}, A_m'\right) \tag{48}$$

The second process of stimulated absolution violates time reversal symmetry. The parameter of symmetry violation of the second order process can be is defined as

$$\beta = \frac{W_{T\omega=\omega_{ml}}^{(2)} - W_{\omega=\omega_{ml}}^{(2)}}{W_{\omega=\omega_{ml}}^{(2)}} \sim \frac{\left(A_m^2 - A_l^2\right)}{\hbar^2\omega_{ml}^2} \sim 10^{-26}E_0^2 \tag{49}$$

When the radiation fields are strong enough with $E_0 \approx 10^{12} \sim 10^{13} V/m$, the time reversal symmetry violation of the second order process would be great.

Meanwhile, by means of (33) and (42), for the second order process with $\omega = -\omega_{ml}$, the transition amplitude and probability can be obtained with

$$a_m^{(2)}(t)_{\omega=-\omega_{ml}} = \frac{\hat{F}_{1ml}\left(\hat{F}_{1ll} - \hat{F}_{1ll}^{+}\right)\left[e^{i(\omega+\omega_{ml})t} - 1\right]}{\hbar^2 \omega_{ml}(\omega + \omega_{ml})} \tag{50}$$

$$W_{\omega=-\omega_{ml}}^{(2)} = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2 \left\{1 + \frac{A_l^2}{\hbar^2 \omega_{ml}^2}\right\}\delta(\omega + \omega_{ml}) \tag{51}$$

Their time reversals are

$$a_m^{(2)}(t)_{T\omega=-\omega_{ml}} = \frac{\hat{F}'_{1ml}\left(\hat{F}'_{1mm} - \hat{F}'^{+}_{1mm}\right)\left[e^{i(\omega+\omega_{ml})t} - 1\right]}{\hbar^2 \omega_{ml}(\omega + \omega_{ml})} \tag{52}$$

$$W_{T\omega=-\omega_{ml}}^{(2)} = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2 \left\{1 + \frac{A_m''^2}{\hbar^2 \omega_{ml}^2}\right\}\delta(\omega + \omega_{ml}) \tag{53}$$

Also, the process violates time reversal symmetry. It is easy to prove that for the second order processes of double photon absorptions with $2\omega = \pm\omega_{ml}$, the transition probabilities are unchanged under time reversal. The symmetry violation appears in the third order processes.

6. Accumulate solution of double energy level system and its time reversal

What has been discussed above is that the radiation fields are polarized and monochromatic light. It is easy to prove that when the radiation fields are non-polarized and non-monochromatic light, time reversal symmetry is still violated after the retarded effect of radiation field is taken into account in the high order processes. But we do not discuss this problem more here. The approximation method of perturbation is used in the discussion above. In order to prove that symmetry violation of time reversal is not introduced by the approximate method, we discuss double energy level system below. The wave function of double energy lever system can be written as

$$|\psi\rangle = a(t)e^{-\frac{i}{\hbar}E_1 t}|1\rangle + b(t)e^{-\frac{i}{\hbar}E_2 t}|2\rangle \tag{54}$$

Thus the motion equations of quantum mechanics are

$$i\hbar\dot{a}(t) = \hat{H}'_{11}a(t) + \hat{H}'_{12}e^{-i\omega_{21}t}b(t) \qquad i\hbar\dot{b}(t) = \hat{H}'_{21}e^{i\omega_{21}t}a(t) + \hat{H}'_{22}b(t) \tag{55}$$

For simplicity, we only consider the first item of the Hamiltonian (2) to take $\hat{H}'_1 \neq 0$, $\hat{H}'_2 = 0$ and $\hat{H}' = \hat{H}'_1$. By taking dipolar approximation with $\vec{k}\cdot\vec{R} = 0$, we have $\langle 1|\hat{H}'|1\rangle = \langle 2|\hat{H}'|2\rangle = 0$. By the rotation wave approximation again, the motion equations becomes

$$\ddot{a}(t) - i(\omega - \omega_{21})\dot{a}(t) + A^2 a(t) = 0 \qquad \ddot{b}(t) + i(\omega - \omega_{21})\dot{b}(t) + A^2 b(t) = 0 \tag{56}$$

Here $A=\left|\hat{F}_{21}\right|^2/\hbar^2$. These two equations have accurate solutions, i.e., the so-called Rabi solutions. Suppose that atom is in the state $|1\rangle$ at beginning with $a(t=0)=1$ and $b(t=0)=0$, we can get [1]

$$\left|b(t)\right|^2=\frac{4A^2\sin^2\sqrt{(\omega-\omega_{21})^2+4A^2}\,t/2}{(\omega-\omega_{21})^2+4A^2} \tag{57}$$

If the atom is in the state $|2\rangle$ at beginning with $b(t=0)=1$ and $a(t=0)=0$, we have

$$\left|a(t)\right|^2=\frac{4A^2\sin^2\sqrt{(\omega-\omega_{21})^2+4A^2}\,t/2}{(\omega-\omega_{21})^2+4A^2} \tag{58}$$

So for the Rabi process, the probability that atom transits from $|1\rangle$ into $|2\rangle$ is the same as that atom transits from $|2\rangle$ into $|1\rangle$. The processes are symmetrical under time reversal. In fact, let $t\to -t$ in (56), we get

$$\ddot{a}(-t)+i(\omega-\omega_{21})\dot{a}(-t)+V^2a(-t)=0 \quad \ddot{b}(-t)-i(\omega-\omega_{21})\dot{b}(-t)+V^2b(-t)=0 \tag{59}$$

Comparing these two formulas with (56), we know that as long as let $a(-t)=b(t)$ and $b(-t)=a(t)$, the motion equations are the same under time reversal.

However, if retarded effect is considered with $\vec{k}\cdot\vec{R}\neq 0$, we have $\langle 1|\hat{H}'|1\rangle\neq 0$ and $\langle 2|\hat{H}'|2\rangle\neq 0$. By taking $\hat{H}_1'\neq 0$, $\hat{H}_2'=0$ similarly, we have

$$\ddot{a}(t)-i\left\{\frac{1}{\hat{H}_{12}'}\left[(\omega-\omega_{21})\hat{F}_{12}e^{i\omega t}-(\omega+\omega_{21})\hat{F}_{12}e^{-i\omega t}\right]-\frac{1}{\hbar}\left(\hat{H}_{11}'+\hat{H}_{22}'\right)\right\}\dot{a}(t)$$

$$-\left\{\frac{\hat{H}_{11}'}{\hbar\hat{H}_{12}'}\left[(\omega-\omega_{21})\hat{F}_{12}e^{i\omega t}-\frac{1}{\hbar}(\omega+\omega_{21})\hat{F}_{12}e^{-i\omega t}\right]\right.$$

$$\left.+\frac{\omega}{\hbar}\left(\hat{F}_{12}e^{i\omega t}-\hat{F}_{12}^{+}e^{-i\omega t}\right)+\frac{1}{\hbar^2}\left(\hat{H}_{11}'\hat{H}_{22}'-\hat{H}_{12}'\hat{H}_{21}'\right)\right\}a(t)=0 \tag{60}$$

$$\ddot{b}(t)+i\left\{\frac{1}{\hat{H}_{12}'}\left[(\omega-\omega_{21})\hat{F}_{12}^{+}e^{-i\omega t}-(\omega+\omega_{21})\hat{F}_{12}e^{i\omega t}\right]-\left(\hat{H}_{11}'+\hat{H}_{22}'\right)\right\}\dot{b}(t)$$

$$-\left\{\frac{\hat{H}_{22}'}{\hbar\hat{H}_{12}'}\left[(\omega-\omega_{21})\hat{F}_{12}^{+}e^{-i\omega t}-\frac{1}{\hbar}(\omega+\omega_{21})\hat{F}_{12}e^{i\omega t}\right]\right.$$

$$\left.+\frac{\omega}{\hbar}\left(\hat{F}_{12}e^{i\omega t}-\hat{F}_{12}^{+}e^{-i\omega t}\right)+\frac{1}{\hbar^2}\left(\hat{H}_{11}'\hat{H}_{22}'-\hat{H}_{12}'\hat{H}_{21}'\right)\right\}b(t)=0 \tag{61}$$

The equations have no accurate solutions in this case. Under time reversal, we have $t \to -t$, $\hat{H}_{T12} = \hat{H}_{21}^*$, $\hat{F}_{T12} = \hat{F}'_{21}$, $\hat{F}_{T12}^+ = \hat{F}'^+_{21}$, the formulas above become

$$\ddot{a}(-t) + i\left\{\frac{1}{\hat{H}'^*_{21}}\left[(\omega-\omega_{21})\hat{F}'_{21}e^{-i\omega t} - (\omega+\omega_{21})\hat{F}'_{21}e^{i\omega t}\right] - \frac{1}{\hbar}\left(\hat{H}'^*_{11} + \hat{H}'^*_{22}\right)\right\}\dot{a}(-t)$$

$$-\left\{\frac{\hat{H}'^*_{11}}{\hbar\hat{H}'^*_{21}}\left[(\omega-\omega_{21})\hat{F}'_{21}e^{-i\omega t} - \frac{1}{\hbar}(\omega+\omega_{21})\hat{F}^*_{21}e^{i\omega t}\right]\right.$$

$$\left.+\frac{\omega}{\hbar}\left(\hat{F}'_{21}e^{-i\omega t} - \hat{F}'^+_{21}e^{i\omega t}\right) + \frac{1}{\hbar^2}\left(\hat{H}'^*_{11}\hat{H}'^*_{22} - \hat{H}'^*_{21}\hat{H}'^*_{21}\right)\right\}a(-t) = 0 \quad (62)$$

$$\ddot{b}(-t) - i\left\{\frac{1}{\hat{H}'^*_{21}}\left[(\omega-\omega_{21})\hat{F}'^+_{21}e^{i\omega t} - (\omega+\omega_{21})\hat{F}'_{21}e^{-i\omega t}\right] - \left(\hat{H}'^*_{11} + \hat{H}'^*_{22}\right)\right\}\dot{b}(-t)$$

$$-\left\{\frac{\hat{H}'^*_{22}}{\hbar\hat{H}'^*_{21}}\left[(\omega-\omega_{21})\hat{F}'^+_{21}e^{i\omega t} - \frac{1}{\hbar}(\omega+\omega_{21})\hat{F}'_{21}e^{-i\omega t}\right]\right.$$

$$\left.+\frac{\omega}{\hbar}\left(\hat{F}'_{21}e^{-i\omega t} - \hat{F}'^+_{21}e^{i\omega t}\right) + \frac{1}{\hbar^2}\left(\hat{H}'^*_{11}\hat{H}'^*_{22} - \hat{H}'^*_{21}\hat{H}'^*_{12}\right)\right\}b(-t) = 0 \quad (63)$$

Because of $\hat{H}'_{11} \neq \hat{H}'^*_{22}$, $\hat{F}'_{21} \neq \hat{F}'^+_{21}$ and $\hat{F}'_{21} \neq \hat{F}'^*_{21}$, even by taking $a(-t) \to b(t)$, $b(-t) \to a(t)$, the motion equations can't yet keep unchanged under time reversal. So after retarded effect of radiation field is considered, the double energy level system can't keep unchanged under time reversal. It means that symmetry violation of time reversal is an inherent character of systems, not originates from the approximate method of perturbation.

7. The time reversal of the third order process of double photons

According to (33), by considering rotation wave approximation, when $2\omega = \omega_{ml}$ we have the transition probability amplitude of the second order process of double photon stimulated absorption

$$a_m^{(2)}(t)_{2\omega=\omega_{ml}} = \frac{\left[e^{-i(2\omega-\omega_{ml})t} - 1\right]}{\hbar(2\omega-\omega_{ml})}\left[\hat{F}^+_{2ml} + \frac{2\hat{F}^+_{1ml}\left(\hat{F}^+_{1ll} - \hat{F}^+_{1mm}\right)}{\hbar\omega_{ml}}\right] \quad (64)$$

So the total transition probability of stimulated absorption of double photons in the first and second processes in unit time is

$$W^{(2)}_{2\omega=\omega_{ml}} = \frac{2\pi}{\hbar^2}\left\{\left|\hat{F}^+_{2ml}\right|^2 + \frac{4}{\hbar^2\omega^2_{ml}}\left|\hat{F}^+_{1ml}\right|^2\left|\hat{F}^+_{1ll} - \hat{F}^+_{1mm}\right|^2\right.$$

$$+\frac{4}{\hbar\omega_{ml}}\text{Re}\left[\left(\hat{F}_{2ml}^{+}\right)^{*}\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll}^{+}-\hat{F}_{1mm}^{+}\right)\right]\Bigg\}\delta\left(2\omega-\omega_{ml}\right) \tag{65}$$

Here " Re " represents the real part of the function. Similarly, we can obtain the time reversal of transition probability of stimulated absorption of double photons in the second process

$$a_{Tm}^{(2)}(t)_{2\omega=\omega_{ml}}=-\frac{\left[e^{-i(2\omega-\omega_{ml})t}-1\right]}{\hbar\left(2\omega-\omega_{ml}\right)}\left[\hat{F}_{2ml}^{\prime+}+\frac{2\hat{F}_{1ml}^{\prime+}\left(\hat{F}_{1ll}^{\prime+}-\hat{F}_{1mm}^{\prime+}\right)}{\hbar\omega_{ml}}\right] \tag{66}$$

We have relations

$$\left|\hat{F}_{2ml}^{\prime+}\right|^{2}=\left|\hat{F}_{2ml}\right|^{2}\qquad\left|\hat{F}_{1ml}^{\prime+}\right|^{2}=\left|\hat{F}_{1ml}\right|^{2}\qquad\hat{F}_{1ml}^{\prime}=\left(\hat{F}_{1ml}^{+}\right)^{*}$$

$$\hat{F}_{1ml}^{\prime+}=\hat{F}_{1ml}^{*}\qquad\hat{F}_{2ml}^{\prime}=\left(\hat{F}_{2ml}^{+}\right)^{*}\qquad\hat{F}_{2ml}^{\prime+}=\hat{F}_{2ml}^{*} \tag{67}$$

The time reversal of total transition probability of stimulated absorption of double photons in the first and second processes is

$$W_{T2\omega=\omega_{ml}}^{(2)}=\frac{2\pi}{\hbar^{2}}\Bigg\{\left|\hat{F}_{2ml}\right|^{2}+\frac{4}{\hbar^{2}\omega_{ml}^{2}}\left|\hat{F}_{1ml}\right|^{2}\left|\hat{F}_{1ll}-\hat{F}_{1mm}\right|^{2}$$

$$+\frac{4}{\hbar\omega_{ml}}\text{Re}\left[\hat{F}_{2ml}^{*}\hat{F}_{1ml}\left(\hat{F}_{1ll}-\hat{F}_{1mm}\right)\right]\Bigg\}\delta\left(2\omega-\omega_{ml}\right) \tag{68}$$

According to the formulas (18), (19) and (32), we have

$$\left(\hat{F}_{2ml}^{+}\right)^{*}\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll}^{+}-\hat{F}_{1mm}^{+}\right)=\hat{F}_{2ml}^{*}\hat{F}_{1ml}\left(\hat{F}_{1ll}-\hat{F}_{1mm}\right) \tag{69}$$

as well as $\left|\hat{F}_{2ml}\right|^{2}=\left|\hat{F}_{2ml}^{+}\right|^{2}$, $\left|\hat{F}_{1ml}\right|^{2}=\left|\hat{F}_{1ml}^{+}\right|^{2}$, $\left|\hat{F}_{1ll}^{+}-\hat{F}_{1mm}^{+}\right|^{2}=\left|\hat{F}_{1ll}-\hat{F}_{1mm}\right|^{2}$, we have $W_{T2\omega=\omega_{ml}}^{(2)}=W_{2\omega=\omega_{ml}}^{(2)}$.So there is no symmetry violation of time reversal in the first and second order processes of double photons. We should consider the third order process. According to (12), the transition probability amplitude of the third order process is

$$a_{m}^{(3)}(t)=\frac{1}{i\hbar}\sum_{n}\int_{0}^{t}\hat{H}_{2mn}^{\prime}a_{n}^{(1)}(t)e^{i\omega_{mn}t}dt+\frac{1}{i\hbar}\sum_{n}\int_{0}^{t}\hat{H}_{1mn}^{\prime}a_{n}^{(2)}(t)e^{i\omega_{mn}t}dt \tag{70}$$

By taking the integral of the formula, we can obtain the probability amplitude of the third order processes. The result is shown in appendix. For the double photon absorption processes with $2\omega=\omega_{ml}$, by rotation wave approximation, the probability amplitude and transition probability in unit time are individually

$$a_m^{(3)}(t)_{2\omega=\omega_{ml}} = \left[\frac{e^{-i(2\omega-\omega_{ml})t}-1}{\hbar(2\omega-\omega_{ml})}\right]\frac{4\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll}^{+}-\hat{F}_{1mm}^{+}\right)\left(\hat{F}_{1ll}-\hat{F}_{1ll}^{+}\right)}{\hbar^2\omega_{ml}^2} \tag{71}$$

$$W_{2\omega=\omega_{ml}}^{(3)} = \frac{2\pi}{\hbar^2}\left\{\left|\hat{F}_{2ml}^{+}\right|^2 + \frac{4\left(1+4A_l^2\right)}{\hbar^2\omega_{ml}^2}\left|\hat{F}_{1ml}^{+}\right|^2\left|\hat{F}_{1ll}^{+}-\hat{F}_{1mm}^{+}\right|^2\right.$$

$$\left. + \frac{4}{\hbar\omega_{ml}}\mathrm{Re}\left[\left(1+i2A_l\right)\left(\hat{F}_{2ml}^{+}\right)^{*}\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll}^{+}-\hat{F}_{1mm}^{+}\right)\right]\right\}\delta(2\omega-\omega_{ml}) \tag{72}$$

Here $iA_l = \hat{F}_{1ll} - \hat{F}_{1ll}^{+}$. On the other hand, by taking $k \to j$ in (24), we can get the time reversal of (70)

$$a_{Tm}^{(3)}(t) = a_l^{(3)}(-t) = -\frac{1}{i\hbar}\sum_j\int_0^t \hat{H}'_{2Tlj}a_j^{(1)}(-t)e^{i\omega_{jl}t}dt - \frac{1}{i\hbar}\sum_j\int_0^t \hat{H}'_{1Tlj}a_j^{(2)}(-t)e^{i\omega_{jl}t} \tag{73}$$

By considering relations $\hat{H}'_{2Tlj} = \hat{H}'^{*}_{2jl}$, $\hat{H}'_{1Tlj} = \hat{H}'^{*}_{1jl}$, $\hat{F}'_{1ml} = \left(\hat{F}_{1ml}^{+}\right)^{*}$, $\hat{F}'^{+}_{1ml} = \hat{F}^{*}_{1ml}$, $\hat{F}'_{2ml} = \left(\hat{F}_{2ml}^{+}\right)^{*}$ and $\hat{F}'^{+}_{2ml} = \hat{F}^{*}_{2ml}$ as well as by the same method to do integral and take rotation wave approximation,, we get the time reversals of probability amplitude and transition probability in the third order process of double photon absorption individually

$$a_{Tm}^{(3)}(t)_{2\omega=\omega_{ml}} = \left[\frac{e^{-i(2\omega-\omega_{ml})t}-1}{\hbar(2\omega-\omega_{ml})}\right]\frac{4\hat{F}'^{+}_{1ml}\left(\hat{F}'^{+}_{1ll}-\hat{F}'^{+}_{1mm}\right)\left(\hat{F}'_{1mm}-\hat{F}'^{+}_{1mm}\right)}{\hbar^2\omega_{ml}^2}$$

$$= -\left[\frac{e^{-i(2\omega-\omega_{ml})t}-1}{\hbar(2\omega-\omega_{ml})}\right]\frac{4\hat{F}^{*}_{1ml}\left(\hat{F}^{*}_{1ll}-\hat{F}^{*}_{1mm}\right)\left(\hat{F}_{1mm}-\hat{F}^{+}_{1mm}\right)^{*}}{\hbar^2\omega_{ml}^2} \tag{74}$$

$$W_{T2\omega=\omega_{ml}}^{(3)} = \frac{2\pi}{\hbar^2}\left\{\left|\hat{F}_{2ml}\right|^2 + \frac{4\left(1+4A_m^2\right)}{\hbar^2\omega_{ml}^2}\left|\hat{F}_{1ml}\right|^2\left|\hat{F}_{1ll}-\hat{F}_{1mm}\right|^2\right.$$

$$\left. + \frac{4}{\hbar\omega_{ml}}\mathrm{Re}\left[(1+i2A_m)\hat{F}_{2ml}\hat{F}^{*}_{1ml}\left(\hat{F}^{*}_{1ll}-\hat{F}^{*}_{1mm}\right)\right]\right\}\delta(2\omega-\omega_{ml}) \tag{75}$$

Here $iA_m = \hat{F}_{1mm} - \hat{F}_{1mm}^{+}$. Comparing with (72), we know that the difference $A_l \neq A_m$ leads to time reversal symmetry violation. But if the high order multiple moment effects are omitted, symmetry violations of time reversals in the third order processes would not exist.

8. Sum frequency process and its time reversal

The process of sum frequency process in non-linear optics is that an electron translates into higher energy lever $|m\rangle$ from low energy level $|l\rangle$ by absorbing two photons with frequencies ω_1 and ω_2 individually, then emits out a photon with frequency $\omega_3 = \omega_1 + \omega_2$ and translates from higher energy level $|m\rangle$ into low energy level $|l\rangle$ again. Suppose that incident light is parallel one containing frequencies ω_1, ω_2 and $\omega_3 = \omega_1 + \omega_2$, and the strength of electrical field is $\vec{E}_0$, the interaction Hamiltonians between electron and radiation field are

$$\hat{H}_1' = \sum_{\lambda=1}^{3}\left(\hat{F}_{1\lambda}e^{i\omega_\lambda t} + \hat{F}_{1\lambda}^{+}e^{-i\omega_\lambda t}\right) \quad \hat{H}_2' = \sum_{\lambda=1}^{3}\left(\hat{F}_{2\lambda}e^{i\omega_\lambda t} + \hat{F}_{1\lambda}^{+}e^{-i\omega_\lambda t} + \hat{F}_{0\lambda}\right) \tag{76}$$

Here

$$\hat{F}_{1\lambda} = -\frac{q\vec{E}_0}{2\omega_\lambda \mu}\cdot e^{-i\vec{k}_\lambda\cdot\vec{R}}\hat{p} \qquad \hat{F}_{1\lambda}^{+} = -\frac{q\vec{E}_0}{2\omega_\lambda \mu}\cdot \hat{p}^{+}e^{i\vec{k}_\lambda\cdot\vec{R}} \tag{77}$$

$$\hat{F}_{2\lambda} = \frac{q^2E_0^2}{2\omega_\lambda^2\mu}e^{-i2\vec{k}_\lambda\cdot\vec{R}} \qquad \hat{F}_{2\lambda}^{+} = \frac{q^2E_0^2}{2\omega_\lambda^2\mu}e^{i2\vec{k}_\lambda\cdot\vec{R}} \qquad \hat{F}_{0\lambda} = \frac{q^2E_0^2}{\omega_\lambda^2\mu} \tag{78}$$

When we calculate probability amplitude, the result corresponds to let $\omega \to \omega_\lambda$ and take sum over index λ in (33) and the formula in the appendix of this paper. For the process an electron absorbs two photons with frequencies ω_1 and ω_2, transits from low energy level $|l\rangle$ into high energy level $|m\rangle$, transition probability corresponds to let $2\omega = \omega_1 + \omega_2 = \omega_{ml}$ in (33) of double photon absorption process. When the electron transits back from high energy level $|m\rangle$ into low energy level $|l\rangle$ by emitting a photon with frequency ω_3, after the retarded effect and high order processes are considered, according to (47), the transition probability is

$$W_{\omega_3=\omega_{ml}}^{(2)} = \frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2\left\{1 + \frac{A_m'^2}{\hbar^2\omega_{ml}^2}\right\}\delta\left(\omega_3 - \omega_{ml}\right) \tag{79}$$

Therefore, for the sum frequency process that an electron translates into state $|m\rangle$ from state $|l\rangle$ by absorbing two photons with frequencies ω_1 and ω_2, then translates back into original state $|l\rangle$ from state $|m\rangle$ by emitting out a photon with frequencies $\omega_3 = \omega_1 + \omega_2$, the total transition probability is

$$W_{\omega_1+\omega_2=\omega_{ml}}^{(2)} + W_{\omega_3=\omega_{ml}}^{(2)} = \frac{2\pi}{\hbar^2}\left\{\left|\hat{F}_{2ml}^{+}\right|^2 + \frac{4\left(1+4A_l^2\right)}{\hbar^2\omega_{ml}^2}\left|\hat{F}_{1ml}^{+}\right|^2\left|\hat{F}_{1ll}^{+} - \hat{F}_{1mm}^{+}\right|^2\right.$$

$$\left. + \frac{4}{\hbar\omega_{ml}}\mathrm{Re}\left[\left(1+i2A_l\right)\left(\hat{F}_{2ml}^{+}\right)^{*}\hat{F}_{1ml}^{+}\left(\hat{F}_{1ll}^{+} - \hat{F}_{1mm}^{+}\right)\right]\right\}\delta\left(2\omega - \omega_{ml}\right)$$

$$+\frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2\left\{1+\frac{A_m'^2}{\hbar^2\omega_{ml}^2}\right\}\delta\left(\omega_3-\omega_{ml}\right) \tag{80}$$

Meanwhile, according the calculation in (38), after the retarded effects and high order processes are considered, the transition probability that an electron transits from state $|l\rangle$ into state $|m\rangle$ by absorbing a photon with frequency ω_3 is

$$W^{(2)}_{T\omega_3=\omega_{ml}}=\frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}\right|^2\left\{1+\frac{A_l^2}{\hbar^2\omega_{ml}^2}\right\}\delta\left(\omega_3-\omega_{ml}\right) \tag{81}$$

So for the time reversal of sum frequency process that an electron absorbs a photon with frequency ω_3 and transits from state $|l\rangle$ into state $|m\rangle$, then emits two photons with frequencies ω_1 and ω_2, and transits back into original state $|l\rangle$, the total transition probability is

$$W^{(2)}_{T\omega_1+\omega_2=\omega_{ml}}+W^{(2)}_{T\omega_3=\omega_{ml}}=\frac{2\pi}{\hbar^2}\left\{\left|\hat{F}_{2ml}\right|^2+\frac{4\left(1+4A_m'^2\right)}{\hbar^2\omega_{ml}^2}\left|\hat{F}_{1ml}\right|^2\left|\hat{F}_{1ll}-\hat{F}_{1mm}\right|^2\right.$$

$$\left.+\frac{4}{\hbar\omega_{ml}}\mathrm{Re}\left[\left(1+i2A_m'\right)\hat{F}_{2ml}\hat{F}_{1ml}^*\left(\hat{F}_{1ll}^*-\hat{F}_{1mm}^*\right)\right]\right\}\delta\left(2\omega-\omega_{ml}\right)$$

$$+\frac{2\pi}{\hbar^2}\left|\hat{F}_{1ml}^+\right|^2\left\{1+\frac{A_l^2}{\hbar^2\omega_{ml}^2}\right\}\delta\left(\omega_3-\omega_{ml}\right) \tag{82}$$

Similarly, because of $A_l \neq A_m'$, sum frequency process violates time reversal symmetry.

By the same method, we can prove that the other processes of non-linear optics just as double frequency, difference frequency, parametric amplification, Stimulated Raman scattering, Stimulated Brillouin scattering and so on are also asymmetric under time reversal. The reason is the same that the light's high order stimulated radiation and absorption processes are asymmetric under time reversal after retarded effect of radiation fields are taken into account.

9. Non-linear polarizations and symmetry violation of time reversal

What is discussed above is based on quantum mechanics. But in non-linear optics, we often calculate practical problems based on classical equations of electromagnetic fields. So we need to discuss the revised non-linear polarizations when the retarded effect of radiation field is taken into account. According to the current theory of nonlinear optics, polarizations are unchanged under time reversal. This does not coincident with practical situations. The reason is that current theory only considers the dipolar approximation without considering the high order processes and the retarded effects of radiation fields. We now discuss the revision of non-linear polarizations after the retarded effect of radiation fields and the high order perturbation processes are taken into account.

Let B_{ml} represent the stimulated absorption probability of an electron (unit radiation density and unit time) transiting from initial low-energy state $|l\rangle$ to final high-energy state $|m\rangle$, B_{lm} represent the probability of stimulated radiation of an electron (unit radiation density and unit time) transiting from the initial high-energy state $|m\rangle$ into the final low-energy state to $|l\rangle$, we have

$$B_{ml} = \frac{4\pi^2}{3\hbar^2}\left|\vec{D}_{ml}\right|^2 (1+\lambda_{ml}) \qquad B_{lm} = (B_{ml})_T = \frac{4\pi^2}{3\hbar^2}\left|\vec{D}_{ml}\right|^2 (1+\lambda'_{ml}) \tag{83}$$

Here $\vec{D}_{ml}$ is the dipolar moment of an electron. We have $\lambda_{ml} \neq \lambda'_{ml}$ and $B_{ml} \neq B_{lm}$ in general, i.e., the parameters of light's stimulated radiation and absolution are not the same. It also means that the nonlinear optical processes would violate time reversal symmetry in general. Let $\vec{D}'_{ml} = \sqrt{1+\lambda_{ml}}\vec{D}_{ml}$ representing the revised dipolar moment after retarded effect is considered, $\vec{D}'_{lm} = \sqrt{1+\lambda_{lm}}\vec{D}_{lm}$ representing the time reversal of $\vec{D}'_{ml}$, we have $\lambda_{ml} \neq \lambda_{lm}$ and $\vec{D}'_{ml} \neq \vec{D}'_{lm}$ in general. Therefore, as long as we let $\vec{D}_{ml} \rightarrow \vec{D}'_{ml}$ in the current formula $\chi^{(n)}_{ij\ldots k}$ of non-linear polarizations, we obtain the revised formula after the retarded effect of radiation fields and the high order perturbation processes are taken into account. Correspondingly, we let $\vec{D}_{lm} \rightarrow \vec{D}'_{lm}$ and obtain the time reversal formula $\chi^{(n)}_{Tij\ldots k}$. It is obvious that non-linear polarizations can not keep unchanged under time reversal. For example, for the non-linear polarizations of the second order processes, we have

$$\chi^{(2)}_{ijk} = (-\omega_1 - \omega_2, \omega_1, \omega_2) = \frac{N}{4\varepsilon_0\hbar^2}\sum_{nn'}\sum_{p}\frac{(D'_i)_{\beta n'}(D'_j)_{n'n}(D'_k)_{n'\beta}}{(\omega_{n'\beta}+\omega_2)(\omega_{n\beta}+\omega_1+\omega_2)} \tag{84}$$

Its time reversal is

$$\chi^{(2)}_{Tijk} = (-\omega_1 - \omega_2, \omega_1, \omega_2) = \frac{N}{4\varepsilon_0\hbar^2}\sum_{nn'}\sum_{p}\frac{(D'_i)_{n'\beta}(D'_j)_{n\,n'}(D'_k)_{\beta\,n'}}{(\omega_{n'\beta}+\omega_2)(\omega_{n\beta}+\omega_1+\omega_2)} \tag{85}$$

In general, we have $\vec{D}'_{ml} \neq \vec{D}'_{lm}$ and $\chi^{(2)}_{ijk} \neq \chi^{(2)}_{Tijk}$, so we have $\chi^{(n)}_{ij\ldots k} \neq \chi^{(n)}_{Tij\ldots k}$. Therefore, the polarization formula of electrical medium and its time reversal are

$$\vec{P} = \varepsilon_0\left(\chi^{(1)}_i E_i + \chi^{(2)}_{ij} E_i E_j + \chi^{(3)}_{ijk} E_i E_j E_k \cdots\right) \tag{86}$$

$$\vec{P}_T = \varepsilon_0\left(\chi^{(1)}_{Ti} E_i + \chi^{(2)}_{Tij} E_i E_j + \chi^{(3)}_{Tijk} E_i E_j E_k \cdots\right) \tag{87}$$

We have $P_T \neq P$ in general. So the motion equation of classical electrical field and its time reversal are also asymmetrical in general with forms

$$\nabla^2\vec{E} - \mu_0\varepsilon_0\frac{\partial^2}{\partial t^2}\vec{E} = \mu_0\frac{\partial^2}{\partial t^2}\vec{P} \qquad \nabla^2\vec{E} - \mu_0\varepsilon_0\frac{\partial^2}{\partial t^2}\vec{E} = \mu_0\frac{\partial^2}{\partial t^2}\vec{P}_T \tag{88}$$

Because (86) ~ (88) are the basic equations of nonlinear optics, we can see that the general nonlinear optical processes violate time reversal symmetry.

In fact, by analyzing nonlinear optics phenomena without complex calculations, we can know the irreversibility of nonlinear optical processes. Although the irreversibility concept is not completely the same as that of the asymmetry of time reversal, they coincide in essence. So let's analyze some practical examples to exposure the irreversibility of nonlinear optics processes below.

10. Irreversibility of nonlinear optics processes

As we know that the processes of linear optics such as light's propagations, reflection, refraction, polarization and so on in uniform mediums are reversible. For example, light's focus through a common convex mirror shown in Fig.6. When a beam of parallel light is projected into a convex mirror, it is focused at point O. If we put a same convex mirror at point B and O is also the focus of convex mirror B, light emitted from point O will become a beam of parallel light again when it passes the convex mirror B. The process that light moving from $O \rightarrow B$ can be regarded as the time reversal process of light moving from $A \rightarrow O$. It is obvious that the process is reversible. The second example is that a beam of white sunlight can be decomposed to a spectrum with different colors by a prism. When these lights with difference colors are reflected back into prism along same paths, white sunlight will be formed again. The third example is that a beam of light can become two different polarization lights with different propagation directions when the light is projected into a double refraction crystal. If these two polarization lights are reflected back into the crystal along same path again, the original light is formed. All of these processes are reversible. But in the processes of non-linear optics, reversibility does not exist. Some examples are shown below.

10.1 Light's multiple frequency, difference frequency and parameter amplification

As shown in Fig.2, a beam of laser with frequency ω is projected into a proper medium and proper phase matching technology is used. The light with double frequency 2ω is found in out going light besides original light with frequency ω. If the lights with frequencies ω and 2ω are reflected back into the same medium, as shown in Fig.2, they can't be completely synthesized into the original light with a single frequency ω. Some light with frequency ω will become light with multiple frequency again by multiple frequency process and some light with frequency 2ω will become the light with frequency ω by difference frequency process. Meanwhile, some light with frequencies ω and 2ω will penetrate medium without being changed as shown in Fig. 3. So the original input light can' be recovered and the reversibility of process is broken. The situations are the same for sum frequency, difference frequency and parameter amplification processes and so on.

10.2 Bistability of optics [3]

As shown in Fig. 4 and 5, the processes of optical bistability are similar to the polarization and magnetization processes of ferroelectrics and ferromagnetic. In the processes the hysteretic loops are formed between incident and outgoing electrical field strengths. In the polarization and magnetization processes of ferroelectrics and ferromagnetic, electromagnetic fields changing along positive directions can be regarded as the time reversal of fields changing along negative directions. There exists electric and magnetic hysteresis. The hysteretic loops are similar to heat engine cycling loops. After a cycling, heat dissipation is produced and the reversibility of process is violated.

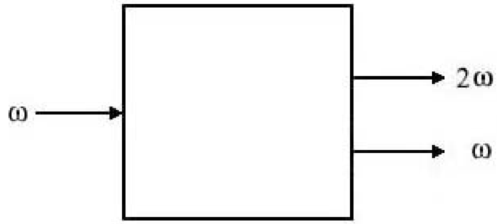

Fig. 2. Process of light's multiple frequency

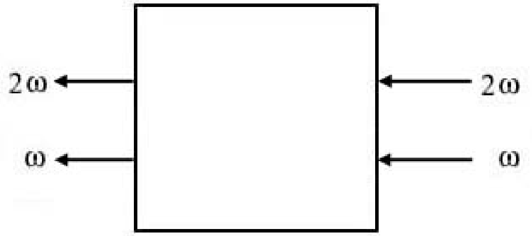

Fig. 3. Time Reversal process of Light's multiple frequency

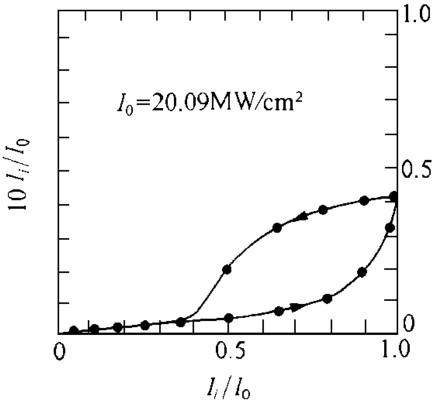

Fig. 4. Optical bistability of nitrobenzene

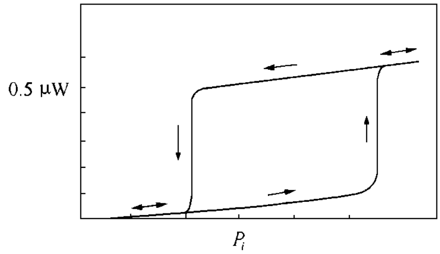

Fig. 5. Optical bistability of mixing type

10.3 Self-focusing and self-defocusing processes of light [4]

Medium's refractive index will change nonlinearly when a beam of laser with uneven distribution on its cross section, for example the Gauss distribution, is projected into a proper medium. The result is that medium seems becoming a convex or concave mirror so that parallel light is focused or defocused. This is just the processes of self-focusing and self-defocusing of lights. The stationary self-focusing process is shown in Fig.7. Parallel light is focused at point O. Then it becomes a thin beam of light projecting out medium. We compare it with common focusing process shown in Fig. 6. If the self-focusing process is reversible, the light focused at pint O would become parallel light again when it projecting out the medium as shown in dotted lines in Fig.7. But it dose not do actually. So the self-focusing process is irreversible. And so do for the self-defocusing process of light.

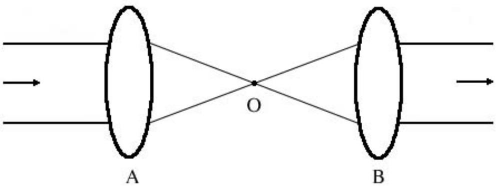

Fig. 6. Focusing process of light.

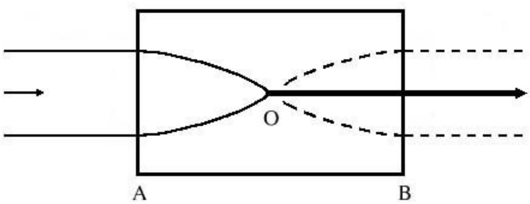

Fig. 7. Self-focusing process of light.

10.4 Double and multi-photon absorption [5]

In double absorption process of photons, an electron in low-energy level will absorb two photons with frequencies ω_1 and ω_2, then transits to high-energy level. But if the electron at high-energy level transits back to low-energy level, it either gives out only a photon at frequency $\omega_3 = \omega_1 + \omega_2$, or two photons at frequencies $\omega_1' \neq \omega_1$, $\omega_2' \neq \omega_2$ in general. It will not produce two photons with original frequencies ω_1 and ω_2. Double photon absorption process is irreversible. And so is for multi-photon absorption.

10.5 Photon echo phenomena [6]

Under certain temperature and magnetic field condition, a beam of laser can be split into two lights with a time difference by using a time regulator of optics. Then two lights are emitted into a proper crystal. Thus three light signals can be observed when they pass through the crystal. The last signal is photon echo. This is a kind of instant coherent phenomena of light. If these three lights signals are imported into same medium again, they can't return into origin two lights. Either three signals are observed (no new echo is produced) or more signals are observed (new signals are produced). In fact, besides photon echo, there are electron spin echo, ferromagnetic echo and plasma echo and so on. All of them are irreversible and violate time reversal symmetry.

10.6 Light's spontaneous radiation processes

As we known that there exist two kinds of different processes for light's radiations, i.e., spontaneous radiation and stimulated radiation. However, there exists only one kind of absorption process, i.e., stimulated radiation without spontaneous absorption in nature. An electron can only transform from high energy level into low energy level by emitting a photon spontaneously, but it can not transform from low energy level into high energy level by absorbing a photon spontaneously. So the processes of light's absorptions themselves are obviously asymmetrical under time reversal.

11. Influence on the fundamental theory of laser

The influence of higher order revision on the fundamental theory of laser is discussed below. Let us first discuss the double energy level system. Let N_1 represent the number of electrons on lower energy level and N_2 represent the number of electrons on higher energy level. With higher order revision, we have $B_{12} \neq B_{21}$. Under the circumstance of having no electron population revision $N_2 < N_1$, as long as B_{21} is larger enough than B_{12}, we still have $B_{21}N_2 > B_{12}N_1$. This means that without the reversion of electron population, laser can still be produced. At present, many experiments have verified this result [7]. In fact, the number of electrons on an energy level can not be determined directly at present. What can be determined by experiments is the number of photons emitted by atoms. And the numbers of photons are calculated by utilizing $\rho B_{21}N_2$, $\rho B_{12}N_1$ and $A_{21}N_2$. By considering the high order retarded effect of radiation fields, the condition of stimulated amplification to produce laser should be changed from $N_2 > N_1$ to $B_{21}N_2 > B_{12}N_1$.

Secondly, according to the current theory, we must have at least three energy levels to produce laser. For the systems with two energy levels, there is a so-called fine balance with $B_{12}N_1\rho(\nu) = A_{21}N_2 + B_{21}N_2\rho(\nu)$. If $B_{12} = B_{21}$ and $A_{21} = k_{21}B_{21}$, we have

$$\frac{N_2}{N_1} = \frac{\rho}{(\rho + \kappa_{21})} < 1 \tag{89}$$

That is $N_2 < N_1$. In this case, we do not have population reversion, therefore no laser is produced. According this paper, suppose we still have $A_{21} = k_{21}B_{21}$, when the balance is reached, we still have

$$\frac{N_2}{N_1} = \frac{\rho B_{12}}{(\rho + \kappa_{21})B_{21}} \tag{90}$$

Because of $B_{12} \neq B_{21}$, as long as relation $\rho B_{12} > (\rho + \kappa_{21})B_{21}$ is satisfied, we still have $N_2 > N_1$ so that population reversion can still be caused. But in this case, we have $B_{21}N_2 < B_{12}N_1$. That is to say, for the steady system of double energy levels, even under the condition of population reversion, laser can still not being produced. For the non-steady system of double energy levels, we have two cases

$$\frac{dN_2}{dt} = B_{12}N_1\rho - A_{21}N_2 - B_{21}N_2\rho > 0 \tag{91}$$

$$\frac{dN_2}{dt} = B_{12}N_1\rho - A_{21}N_2 - B_{21}N_2\rho < 0 \tag{92}$$

When $dN_2 / dt > 0$, we have $B_{12}N_1\rho > A_{21}N_2 + B_{21}N_2\rho$, so $B_{12}N_1\rho > B_{21}N_2\rho$, no laser is produced. When $dN_2 / dt < 0$, we have $B_{12}N_1\rho < A_{21}N_2 + B_{21}N_2\rho$. In this case, if $B_{12}N_1\rho < B_{21}N_2\rho$, laser can be produced. If $B_{12}N_1\rho > B_{21}N_2\rho$, laser can be produced.

Next, we discuss the influence on the system of three energy levels. The standard stimulated radiation and absorption process in the system of three energy levels is shown in Fig.8. In the current theory, however, the processes to produce laser is actually simplified as shown in Fig 9. By analyzing the difference between them, we know the significance of this paper's revision. According to Fig.8, when particles which are located on ground state E_1 at the beginning are pumped into E_3 energy level, they can transit into E_2 energy level through both radiation transition and non-radiation transition. The population reversion can be achieved between E_1 and E_2, so that the laser with frequency ω_{21} can be produced. Comparing with Fig.8, the process shown in Fig.9 omits the spontaneous radiation and stimulated radiation transitions, as well as particle's transition from E_2 level into E_3 level. According to the Einstein's theory, we have $B_{13} = B_{31}$. The possibility is the same for a particle transiting from ground state into E_3 energy level and transiting from E_3 energy level back into ground state. Suppose that the number of particles transiting in unit time from ground state into E_3 energy level is $\rho(v_{13})B_{13}N_1$. There will be $\rho B_{31}N_3$ particles transiting back to ground state from E_3 energy level by stimulated radiation, and

$A_{31}N_3 = \kappa_{31}B_{31}N_3$ particles transiting back to ground state from E_3 energy level by spontaneous radiation. Therefore, most particles which have transited to E_3 energy level will come back to original ground state by emitting photons with frequency ω_{31}, so that population reversion between E_1 and E_2 energy levels will be affected greatly. Meanwhile, because of $B_{23} = B_{32}$, some particles on E_2 energy level coming from E_3 energy level will transit back into E_3 energy level by stimulated absorption, so that population reversion between E_1 and E_2 will also be reduced. These results indicate that the Einstein's theory is only suitable for equilibrium processes, rather than the non-equilibrium process of laser production.

The current theory of laser uses a fussy method to avoid theses problems. The probability a particle transits back to ground state from E_3 energy level is not considered directly. In stead, we use a pumping speed R replaces $\rho_{13}B_{31}N_1 - (\rho_{13} + \kappa_{31})B_{31}N_3$. On the other hand, the non-radiation transition is used to replace $(\rho_{23} + \kappa_{32})B_{32}N_3 - \rho_{23}B_{32}N_2$. In this way, the complexity of process is simplified.

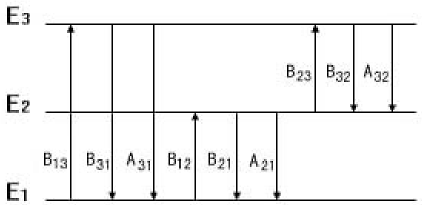

Fig. 8. Transition among three energy levels.

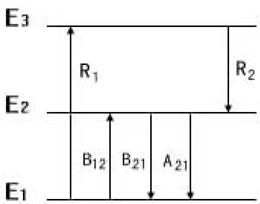

Fig. 9. Simplification of Fig.8.

According to the revision in this paper, we have $B_{ml} \neq B_{lm}$. We can provide a simpler and rational picture for the production of laser in the system of three energy levels. In this case, we can have $B_{31} << B_{13}$ and $(\rho_{13} + \kappa_{31})B_{31}N_3 << \rho_{13}B_{31}N_1$, so that only a few particles can transit back to ground state from E_3 energy level by stimulated radiation and spontaneous radiation after they transit from ground state into E_3 energy level. Most of particles on E_3 energy level will transit into E_2 energy level. On the other hand, because of $B_{23} << B_{32}$, we have $\rho_{23}B_{23}N_2 << (\rho_{23} + \kappa_{32})B_{32}N_3$. Most of particles can not transit back into E_3 energy level after they transit to E_2 energy from E_3 energy level. Meanwhile, because of $B_{12} << B_{21}$, it is difficult for particles on ground state to transit into E_2 energy level from ground state by stimulated absorption, but is easy to transit to ground state from E_2 energy level by stimulated radiation. Therefore, a high effective and ideal laser system of three energy levels should satisfy the conditions $B_{21} << B_{12}$, $B_{31} << B_{13}$ and $B_{23} << B_{32}$. It is obvious that as long as $B_{ml} \neq B_{lm}$, we can simply and rationally explain the production of laser of the system of three energy levels.

In this way, we can also well explain the phenomenon of optical self-transparence and self absorptions [8]. Experiments show that in strong electric fields, some medium can have the saturated absorption of light, so that the medium will become transparent for light. The current explanation of saturated absorption is that the number N_1 of particles located on low energy level becomes smaller and the absorption of light is proportional to the number of particles located on low energy level, therefore the stimulated absorption becomes smaller. Meanwhile, the transmission light increases due to the stimulated radiation of particles located on high energy level, i.e., the self-transparence phenomena of saturated absorption appears. The problem of this explanation is that if the number N_1 of particles located on low energy level decreases and the number N_2 of particles located on high energy level increase, the spontaneous radiation will also increase. When stationary states are reached, we always have $A_{21}N_2$ photons emitted in the form of spontaneous radiation in unit time. Because spontaneous radiation is in all directions of space, it is difficult for medium to achieve real transparence.

According to the revised theory of this paper, the revised factor is $\alpha_{ml} \sim E_0^2$. If $\alpha_{ml} < 0$, in strong field, we may have $\alpha_{ml} \sim -1$ for some mediums, so that the stimulated absorption parameter B_{ml} is very small even becomes $B_{ml} \sim 0$. In this case, even though a great number of particles are still located on low energy level, the saturated absorption of light is still possible so that the medium become transparence. However, according to current theory, we have $B_{ml} \sim E_0^2$. When E_0 increases, the stimulated absorption parameter will increase so that it is impossible for us to have $B_{ml} \sim 0$. Conversely, if $\alpha_{ml} > 0$ with $\alpha_{ml} \sim E_0^2$, light's absorption for some mediums will increase greatly in strong field. This is just the phenomena of self absorption. In the current non-linear optics, we explain the phenomena of self absorption with the absorptions of double photons or multi-photons, as well as stimulated scattering. Based on this paper, besides the absorptions of double photons or multi-photons, the process of single photon can also cause trans-normal absorption. It is obvious that the revised theory can explain theses phenomena more rationally.

12. Discussion on the reasons of symmetry violation of time reversal

We need to discuss the reason of the symmetry violation of time reversal In the paper, semi-classical method is used, i.e., quantum mechanics is used to describe charged particles and classical electromagnetic theory is used to describe radiation fields. The limitation of this method is that spontaneous radiation can not be deduced automatically from the theory. The spontaneous radiation formula has to be obtained indirectly by means of the Einstein's theory of light's radiation and absorption. Strictly, we should discuss the problems using complete quantum theory, from which we can deduce spontaneous radiation probability automatically.

However, as we known, except the spontaneous radiation, the results are exactly the same by using both the semi-classical method and the complete quantum method to calculate light's stimulated radiation and absorption probabilities. It also means that if we use complete quantum mechanics to discuss light's stimulated radiation and absorption, time reversal symmetry will also be violated after the retarded effects of radiation fields are taken into account. It is just the spontaneous radiation which indicates the asymmetry of time reversal in the processes of interaction between light and charged particles, for there exits only light's spontaneous radiation without light's spontaneous absolution in nature. This result is completely asymmetrical.

In fact, in complete quantum mechanics, we use photon's creation and annihilation operator $\hat{a}^{+}$ and $\hat{a}$ to replace the factor $-q\vec{E}_0 / 2\omega\mu$ in semi-classical theory. This kind of correspondence does not change the results of time reversal symmetry violation in calculation processes. The problem is that if photon's creation or annihilation operators are used, some complexity and problem will be caused in high order processes so that it may be too difficult to calculate. So in the problems of light's stimulated radiation and absorption and nonlinear optics, we use actually semi-classical or even complete classical theory and methods and always obtain satisfied results at present.

Because the interaction Hamiltonian and the motion equation of quantum mechanics keep the same under time reversal, what causes the symmetry violation of time reversal? The method of rotation wave approximation is used in the paper. Does this approximation method cause the symmetry violation of time reversal? Let's discuss this issue in the following.

Suppose that micro-states are described by $|\psi\rangle$ and $|\phi\rangle$. Their time reversal are $|\psi_T\rangle = T|\psi\rangle$ and $|\phi_T\rangle = T|\phi\rangle$. Suppose that the interaction Hamiltonian remains the same under time reversal, according to quantum mechanics, we have the so-called detail balance formula

$$\langle\psi|\hat{H}|\phi\rangle = \langle\phi_T|\hat{H}|\psi_T\rangle \tag{93}$$

It indicates that the probability amplitude keeps the same under time reversal in the quantum transition process. For the problem of light's stimulated radiation and absolution, if the radiation field is only one with a single frequency, the interaction Hamiltonian is

$$\hat{H} = \hat{F}_0 + \hat{F}_1 e^{i\omega t} + \hat{F}_1^{+} e^{-i\omega t} + \hat{F}_2 e^{i2\omega t} + \hat{F}_1^{+} e^{-i2\omega t} \tag{94}$$

Meanwhile, if only a single particle state is considered, we have

$$|\psi\rangle = |\phi\rangle = \sum_m a_m(t) e^{-\frac{i}{h}E_m t}|m\rangle \qquad |\psi_T\rangle = |\phi_T\rangle = \sum_m a_m(-t) e^{\frac{i}{h}E_m t}|m\rangle \tag{95}$$

Substitute (94) and (95) into (93), we obtain

$$\sum_{m,l} e^{\frac{i}{h}(E_m - E_l)\ t} a_m(t) a_l(t) \langle m|\hat{F}_0 + \hat{F}_1 e^{i\omega t} + \hat{F}_1^{+} e^{-i\omega t} + \hat{F}_2 e^{i2\omega t} + \hat{F}_1^{+} e^{-i2\omega t}|l\rangle$$

$$= \sum_{m,l} e^{-\frac{i}{h}(E_m - E_l)\ t} a_m(-t) a_l(-t) \langle m|\hat{F}_0 + \hat{F}_1 e^{i\omega t} + \hat{F}_1^{+} e^{-i\omega t} + \hat{F}_2 e^{i2\omega t} + \hat{F}_1^{+} e^{-i2\omega t}|l\rangle \tag{96}$$

The formula is the sum of multinomial. It means that the total probability amplitude is unchanged under time reversal. However, by the constraint of energy conservation law, in the formula above, only a few items which satisfy the condition $E_m - E_l = \pm n\hbar\omega$ can be realized really. Those items which do not satisfy the condition are forbidden actually. Keeping the items which satisfy the condition of energy conservation and giving up the items which do not, the procedure is just the so-called rotation wave approximation. It is obvious that the two sides of equation (96) will not equal to each other after going through the procedure, i.e., the symmetry of time reversal will be violated.

The paper calculates the transition and time reversal problems of partial items corresponding to the operators $\hat{F}_1 e^{-i\omega t}$ and $\hat{F}_1^+ e^{i\omega t}$ under the situation $n=1$. Because (68) can not be accurately calculated, we use approximation method and let $a_m(t) = a_m^{(0)}(t) + a_m^{(1)}(t) + a_m^{(2)}(t) + \cdots$. For the first order approximation, we have $a_m(t) = a_m^{(0)}(t) + a_m^{(1)}(t)$. Suppose that an atom transits from state $l\rangle$ into state $m\rangle$, we get transition probability (14) and its time reversal (29). It indicates that the first process is unchanged under time reversal after retarded effect of radiation field is considered. For the second processes, let $a_m(t) = a_m^{(0)}(t) + a_m^{(1)}(t) + a_m^{(2)}(t)$ and assume in the same way that an atom transits from state $l\rangle$ into state $m\rangle$, we get (38) and its time reversal (47). The result violates the symmetry of time reversal and the symmetry violation is relative to the asymmetry of initial states of bounding state atoms before and after time reversal. The uniform values of the Hamiltonian operator for the initial states of an atom before and after time reversal are not equal to each other. Therefore, one reason that causes the symmetry violation of time reversal is that the condition of energy conservation forbids some transition processes between bounding state atoms, so that realizable processes violates time reversal symmetry with

$$W_{\omega=\pm\omega_{ml}}\left(\hat{F}_{1ml}\right) + W_{2\omega=\pm\omega_{ml}}\left(\hat{F}_{1ml}, A_l\right) + W_{3\omega=\pm\omega_{ml}}\left(\hat{F}_{1ml}, A_l\right) + \cdots$$

$$\neq W_{T\omega=\pm\omega_{ml}}\left(\hat{F}_{1ml}\right) + W_{T2\omega=\pm\omega_{ml}}\left(\hat{F}_{1ml}, A'_m\right) + W_{T3\omega=\pm\omega_{ml}}\left(\hat{F}_{1ml}, A'_m\right) + \cdots \quad (97)$$

Meanwhile, for concrete atoms, the other restriction conditions just like the wave function's symmetries should be also considered. So only a few and specific transitions can be achieved actually. Most processes in (96) can not be completed. These realizable processes are just what we can observe and measure. They are irreversible in general. Therefore, the symmetry violation of time reversal in the filial or partial processes of light's stimulated radiation and absolution do not contradict with the fine balance formula (93) actually.

On the other hand, the symmetry violation of time reversal is also related to the asymmetry of initial states of bounding state's atoms before and after time reversal. For the interaction between radiation fields and the non-bounding state's atoms with continuous energy levels, there exists no symmetry violation of time reversal. In this case, there is no the asymmetry problem of the initial states before and after time reversal. This is why we can not find symmetry violation of time reversal in the particle collision experiments for changed particles created by accelerators are non-bounding ones.

Meanwhile, there is a difference of negative sign between A_l shown in (37) and A'_m shown in (46). This difference is caused by the interference of amplitudes between the first order and the second order processes before and after time reversal. But if the retarded effect of radiation field is neglected, the processes of light's stimulated radiation and absolution will be symmetrical under time reversal. So the reasons of symmetry violation of time reversal are caused by multi-factors and are quite complex.

13. Influence on non-equilibrium statistical mechanics

As well-known that although classical equilibrium state statistical physics has been a very mature one, the foundation of non-equilibrium state statistical physics has not be established up to now day. The key is that the evolution processes of macro-systems controlled by the second law of thermodynamics are irreversible under time reversal, but the processes of micro-physics are considered reversible. Because macro-systems are composed of micro-particles, there exists a sharp contradiction here. This is so-called reversibility paradox which has puzzled physics community for a long time [9]. Though many theories have been proposed trying to resolve this problem, for example, the theories of coarseness and mixing current and so on [10], none is satisfied.

The significance of this paper is to provide us a method to solve this problem. We known that macro-systems are composed of atoms and molecules, and atoms and molecules are composed of charged particles. By the photon's radiations and absorptions, charged particles of bounding states and radiation fields interact. According to the discussion in the paper, after the retarded effects of radiation fields are considered, the time reversal symmetry of light's stimulated radiation and absolution is violated, even though the interaction Hamiltonian is unchanged under time reversal. Only when the system reaches macro-equilibrium states, or the probabilities of micro-particles radiating and absorbing photons are the same from the point of view of statistical average, the processes are reversible under time reversal. Therefore, it can be said that irreversibility of macro-processes originates from the irreversibility of micro-processes actually.

By introducing retarded electromagnetic interaction, the forces between charged particles will become non-conservative ones. Based on it, we can establish the revised Liouville equation which is irreversible under time reversal. In this way, we can lay a really rational dynamic foundation for classical non-equilibrium statistical mechanics. The united description can be reached for classical equilibrium and non-equilibrium statistical mechanics. The detail will be provided later.

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15. Appendix

15.1 The transition probability amplitude of the third order process for light's stimulated radiation and absorption

$$
\begin{aligned}
a_m^{(3)}(t) = \frac{1}{\hbar^2}\sum_n \Bigg\{ & \frac{\hat{F}_{2mn}\hat{F}_{1nl}}{\omega+\omega_{nl}}\left[\frac{e^{i(3\omega+\omega_{nl}+\omega_{mn})\,t}-1}{3\omega+\omega_{nl}+\omega_{mn}} - \frac{e^{i(2\omega+\omega_{mn})\,t}-1}{2\omega+\omega_{mn}}\right] \\
& -\frac{\hat{F}_{2mn}\hat{F}^{+}_{1nl}}{\omega-\omega_{nl}}\left[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}} - \frac{e^{i(2\omega+\omega_{mn})\,t}-1}{2\omega+\omega_{mn}}\right] - \frac{\hat{F}^{+}_{2mn}\hat{F}_{1nl}}{\omega-\omega_{nl}}\left[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}} - \frac{e^{-i(2\omega-\omega_{mn})\,t}-1}{2\omega-\omega_{mn}}\right] \\
& +\frac{\hat{F}^{+}_{2mn}\hat{F}^{+}_{1nl}}{\omega-\omega_{nl}}\left[\frac{e^{-i(3\omega-\omega_{nl}-\omega_{mn})\,t}-1}{3\omega-\omega_{nl}-\omega_{mn}} - \frac{e^{-i(2\omega-\omega_{mn})\,t}-1}{2\omega-\omega_{mn}}\right] + \frac{\hat{F}_{0mn}\hat{F}_{1nl}}{\omega+\omega_{nl}}\left[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}} - \frac{e^{i\omega_{mn}\,t}-1}{\omega_{mn}}\right] \\
& +\frac{\hat{F}_{0mn}\hat{F}^{+}_{1nl}}{\omega-\omega_{nl}}\left[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}} + \frac{e^{i\omega_{mn}\,t}-1}{\omega_{mn}}\right] + \frac{\hat{F}_{1mn}\hat{F}_{2nl}}{2\omega+\omega_{nl}}\left[\frac{e^{i(3\omega+\omega_{nl}+\omega_{mn})\,t}-1}{3\omega+\omega_{nl}+\omega_{mn}} - \frac{e^{i(\omega+\omega_{mn})\,t}-1}{\omega+\omega_{mn}}\right] \\
& +\frac{\hat{F}_{1mn}\hat{F}^{+}_{2nl}}{2\omega-\omega_{nl}}\left[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}} + \frac{e^{i(\omega+\omega_{mn})\,t}-1}{\omega+\omega_{mn}}\right] + \frac{\hat{F}_{0nl}\hat{F}_{1mn}}{\omega_{nl}}\left[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}} - \frac{e^{i(\omega+\omega_{mn})\,t}-1}{\omega+\omega_{mn}}\right] \\
& +\frac{\hat{F}^{+}_{1mn}\hat{F}_{2nl}}{2\omega+\omega_{nl}}\left[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}} + \frac{e^{-i(\omega-\omega_{mn})\,t}-1}{\omega-\omega_{mn}}\right] - \frac{\hat{F}^{+}_{1mn}\hat{F}^{+}_{2nl}}{2\omega-\omega_{nl}}\left[\frac{e^{-i(3\omega-\omega_{nl}-\omega_{mn})\,t}-1}{3\omega-\omega_{nl}-\omega_{mn}} - \frac{e^{-i(\omega-\omega_{mn})\,t}-1}{\omega-\omega_{mn}}\right] \\
& -\frac{\hat{F}_{0nl}\hat{F}^{+}_{1mn}}{\omega_{nl}}\left[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}} - \frac{e^{-i(\omega-\omega_{mn})\,t}-1}{\omega-\omega_{mn}}\right] \\
+\frac{1}{\hbar^3}\sum_{n,k}\Bigg\{ & -\frac{\hat{F}_{1mn}\hat{F}_{1nk}\hat{F}_{1kl}}{\omega+\omega_{kl}}\left[\frac{e^{i(3\omega+\omega_{nl}+\omega_{mn})\,t}-1}{(2\omega+\omega_{kl}+\omega_{nk})(3\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})}\right. \\
& -\frac{e^{i(\omega+\omega_{mn})\,t}-1}{(2\omega+\omega_{kl}+\omega_{nk})(\omega+\omega_{mn})} - \frac{e^{i(2\omega+\omega_{nk}+\omega_{mn})\,t}-1}{(2\omega+\omega_{nk}+\omega_{mn})(\omega+\omega_{nk})} \\
& \left.+\frac{e^{i(\omega+\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(\omega+\omega_{mn})}\right] + \frac{\hat{F}_{1mn}\hat{F}_{1nk}\hat{F}^{+}_{1kl}}{\omega-\omega_{kl}}\left[\frac{e^{i(\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})}\right. \\
& \left.-\frac{e^{i(\omega+\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega+\omega_{mn})} - \frac{e^{i(2\omega+\omega_{nk}+\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(2\omega+\omega_{nk}+\omega_{mn})} + \frac{e^{i(\omega+\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(\omega+\omega_{mn})}\right] \\
& -\frac{\hat{F}_{1mn}\hat{F}^{+}_{1nk}\hat{F}^{+}_{1kl}}{\omega+\omega_{kl}}\left[\frac{e^{i(\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{mn})(\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})}\right. \\
& \left.-\frac{e^{i(\omega+\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega+\omega_{mn})} + \frac{e^{i(\omega_{nk}+\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(\omega_{nk}+\omega_{mn})} - \frac{e^{i(\omega+\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(\omega+\omega_{mn})}\right] \\
& +\frac{\hat{F}_{1mn}\hat{F}^{+}_{1nk}\hat{F}^{+}_{1kl}}{\omega-\omega_{kl}}\left[\frac{e^{-i(\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})\,t}-1}{(2\omega-\omega_{kl}-\omega_{nk})(\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})}\right. \\
& \left.+\frac{e^{i(\omega+\omega_{mn})\,t}-1}{(2\omega-\omega_{kl}-\omega_{nk})(\omega+\omega_{mn})} + \frac{e^{i(\omega_{nk}+\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(\omega_{nk}+\omega_{mn})} - \frac{e^{i(\omega+\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(\omega+\omega_{mn})}\right]
\end{aligned}
$$

$$\begin{aligned}
&-\frac{\hat{F}_{1mn}^{+}\hat{F}_{1nk}\hat{F}_{1kl}}{\omega+\omega_{kl}}\Bigg[\frac{e^{i(\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega+\omega_{kl}+\omega_{nk}+\omega_{mn})}\\
&+\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(2\omega+\omega_{kl}+\omega_{nk})(\omega-\omega_{mn})}-\frac{e^{i(\omega_{nk}+\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(\omega_{nk}+\omega_{mn})}-\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(\omega-\omega_{mn})}\Bigg]\\
&-\frac{\hat{F}_{1mn}^{+}\hat{F}_{1nk}\hat{F}_{1kl}^{+}}{\omega-\omega_{kl}}\Bigg[\frac{e^{i(\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})}\\
&-\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega-\omega_{mn})}+\frac{e^{i(\omega_{nk}+\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(\omega_{nk}+\omega_{mn})}+\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(\omega+\omega_{nk})(\omega-\omega_{mn})}\Bigg]\\
&+\frac{\hat{F}_{1mn}^{+}\hat{F}_{1nk}^{+}\hat{F}_{1kl}}{\omega+\omega_{kl}}\Bigg[\frac{e^{-i(\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})}\\
&-\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(\omega_{kl}+\omega_{nk})(\omega-\omega_{mn})}+\frac{e^{-i(2\omega-\omega_{nk}-\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(2\omega-\omega_{nk}-\omega_{mn})}-\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(\omega-\omega_{mn})}\Bigg]\\
&+\frac{\hat{F}_{1mn}^{+}\hat{F}_{1nk}^{+}\hat{F}_{1kl}^{+}}{\omega-\omega_{kl}}\Bigg[\frac{e^{-i(3\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})\,t}-1}{(2\omega-\omega_{kl}-\omega_{nk})(3\omega-\omega_{kl}-\omega_{nk}-\omega_{mn})}\\
&-\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(2\omega-\omega_{kl}-\omega_{nk})(\omega-\omega_{mn})}-\frac{e^{-i(2\omega-\omega_{nk}-\omega_{mn})\,t}-1}{(2\omega-\omega_{nk}-\omega_{mn})(\omega-\omega_{nk})}+\frac{e^{-i(\omega-\omega_{mn})\,t}-1}{(\omega-\omega_{nk})(\omega-\omega_{mn})}\Bigg]
\end{aligned}$$

15.2 The time reversal of transition probability amplitude of third order process of light's stimulated radiation and absorption

$$\begin{aligned}
a_{Tm}^{(3)}(t)=\frac{1}{\hbar^{2}}\sum_{n}\Bigg\{&\frac{\hat{F}_{2nl}'\hat{F}_{1mn}'}{\omega+\omega_{mn}}\Bigg[\frac{e^{i(3\omega+\omega_{nl}+\omega_{mn})\,t}-1}{3\omega+\omega_{nl}+\omega_{mn}}-\frac{e^{i(2\omega+\omega_{nl})\,t}-1}{2\omega+\omega_{nl}}\Bigg]\\
&-\frac{\hat{F}_{2nl}'\hat{F}_{1mn}'^{+}}{\omega-\omega_{mn}}\Bigg[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}}-\frac{e^{i(2\omega+\omega_{nl})\,t}-1}{2\omega+\omega_{nl}}\Bigg]-\frac{\hat{F}_{2nl}'^{+}\hat{F}_{1mn}'}{\omega+\omega_{mn}}\Bigg[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}}-\frac{e^{-i(2\omega-\omega_{nl})\,t}-1}{2\omega-\omega_{nl}}\Bigg]\\
&+\frac{\hat{F}_{2nl}'^{+}\hat{F}_{1mn}'^{+}}{\omega-\omega_{mn}}\Bigg[\frac{e^{-i(3\omega-\omega_{nl}-\omega_{mn})\,t}-1}{3\omega-\omega_{nl}-\omega_{mn}}-\frac{e^{-i(2\omega-\omega_{nl})\,t}-1}{2\omega-\omega_{nl}}\Bigg]+\frac{\hat{F}_{0nl}'\hat{F}_{1mn}'}{\omega+\omega_{mn}}\Bigg[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}}-\frac{e^{i\omega_{nl}\,t}-1}{\omega_{nl}}\Bigg]\\
&-\frac{\hat{F}_{0nl}'\hat{F}_{1mn}'^{+}}{\omega-\omega_{mn}}\Bigg[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}}-\frac{e^{i\omega_{nl}\,t}-1}{\omega_{nl}}\Bigg]+\frac{\hat{F}_{1nl}'\hat{F}_{2mn}'}{2\omega+\omega_{mn}}\Bigg[\frac{e^{i(3\omega+\omega_{nl}+\omega_{mn})\,t}-1}{3\omega+\omega_{nl}+\omega_{mn}}-\frac{e^{i(\omega+\omega_{nl})\,t}-1}{\omega+\omega_{nl}}\Bigg]\\
&+\frac{\hat{F}_{1nl}'\hat{F}_{2mn}'^{+}}{2\omega-\omega_{mn}}\Bigg[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}}+\frac{e^{i(\omega+\omega_{nl})\,t}-1}{\omega+\omega_{nl}}\Bigg]+\frac{\hat{F}_{0mn}'\hat{F}_{1nl}'}{\omega_{mn}}\Bigg[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}}-\frac{e^{i(\omega+\omega_{nl})\,t}-1}{\omega+\omega_{nl}}\Bigg]\\
&+\frac{\hat{F}_{1nl}'^{+}\hat{F}_{2mn}'}{2\omega+\omega_{mn}}\Bigg[\frac{e^{i(\omega+\omega_{nl}+\omega_{mn})\,t}-1}{\omega+\omega_{nl}+\omega_{mn}}+\frac{e^{-i(\omega-\omega_{nl})\,t}-1}{\omega-\omega_{nl}}\Bigg]+\frac{\hat{F}_{1nl}'^{+}\hat{F}_{2mn}'^{+}}{2\omega-\omega_{mn}}\Bigg[\frac{e^{-i(3\omega-\omega_{nl}-\omega_{mn})\,t}-1}{3\omega-\omega_{nl}-\omega_{mn}}-\frac{e^{-i(\omega-\omega_{nl})\,t}-1}{\omega-\omega_{nl}}\Bigg]\\
&-\frac{\hat{F}_{0mn}'\hat{F}_{1nl}'^{+}}{\omega_{mn}}\Bigg[\frac{e^{-i(\omega-\omega_{nl}-\omega_{mn})\,t}-1}{\omega-\omega_{nl}-\omega_{mn}}-\frac{e^{-i(\omega-\omega_{nl})\,t}-1}{\omega-\omega_{nl}}\Bigg]\Bigg\}
\end{aligned}$$

$$+\frac{1}{\hbar^3}\sum_{n,k}\left\{-\frac{\hat{F}'_{1nl}\hat{F}'_{1mk}\hat{F}'_{1kn}}{\omega+\omega_{mk}}\left[\frac{e^{i(3\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})\ t}-1}{(2\omega+\omega_{mk}+\omega_{kn})(3\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})}\right.\right.$$

$$\left.-\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(2\omega+\omega_{mk}+\omega_{kn})(\omega+\omega_{nl})}-\frac{e^{i(2\omega+\omega_{kn}+\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(2\omega+\omega_{kn}+\omega_{nl})}+\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(\omega+\omega_{nl})}\right]$$

$$-\frac{\left(\hat{F}'^{+}_{1nl}\right)^*\hat{F}'^{+}_{1mk}\left(\hat{F}'^{+}_{1kn}\right)^*}{\omega-\omega_{mk}}\left[\frac{e^{i(\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})}\right.$$

$$\left.-\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega+\omega_{nl})}-\frac{e^{i(2\omega+\omega_{kn}+\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(2\omega+\omega_{kn}+\omega_{nl})}+\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(\omega+\omega_{nl})}\right]$$

$$+\frac{\hat{F}'_{1nl}\hat{F}'_{1mk}\hat{F}'^{+}_{1kn}}{\omega+\omega_{mk}}\left[\frac{e^{i(\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})}\right.$$

$$\left.-\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega+\omega_{nl})}+\frac{e^{i(\omega_{kn}+\omega_{nl})\ t}-1}{(\omega-\omega_{kn})(\omega_{kn}+\omega_{nl})}-\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(\omega-\omega_{kn})(\omega+\omega_{nl})}\right]$$

$$-\frac{\hat{F}'_{1nl}\hat{F}'^{+}_{1mk}\hat{F}'^{+}_{1kn}}{\omega-\omega_{mk}}\left[\frac{e^{-i(\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})\ t}-1}{(2\omega-\omega_{mk}-\omega_{kn})(\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})}\right.$$

$$\left.+\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(2\omega-\omega_{mk}-\omega_{kn})(\omega+\omega_{nl})}+\frac{e^{i(\omega_{nk}+\omega_{nl})\ t}-1}{(\omega-\omega_{kn})(\omega_{kn}+\omega_{nl})}-\frac{e^{i(\omega+\omega_{nl})\ t}-1}{(\omega-\omega_{kn})(\omega+\omega_{nl})}\right]$$

$$-\frac{\hat{F}'^{+}_{1nl}\left(\hat{F}'^{+}_{1mk}\right)^*\hat{F}'^{+}_{1kn}}{\omega+\omega_{mk}}\left[\frac{e^{-i(\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})}\right.$$

$$\left.-\frac{e^{-i(\omega-\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega-\omega_{nl})}+\frac{e^{-i(2\omega-\omega_{kn}-\omega_{nl})\ t}-1}{(\omega-\omega_{kn})(2\omega-\omega_{kn}-\omega_{nl})}-\frac{e^{-i(\omega-\omega_{nl})\ t}-1}{(\omega-\omega_{kn})(\omega-\omega_{nl})}\right]$$

$$+\frac{\hat{F}'^{+}_{1nl}\left(\hat{F}'^{+}_{1mk}\right)^*\left(\hat{F}'^{+}_{1kn}\right)^*}{\omega+\omega_{mk}}\left[\frac{e^{i(\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})\ t}-1}{(2\omega+\omega_{mk}+\omega_{kn})(\omega+\omega_{mk}+\omega_{kn}+\omega_{nl})}\right.$$

$$\left.+\frac{e^{-i(\omega-\omega_{nl})\ t}-1}{(2\omega+\omega_{mk}+\omega_{kn})(\omega-\omega_{nl})}-\frac{e^{i(\omega_{kn}+\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(\omega_{kn}+\omega_{nl})}-\frac{e^{-i(\omega-\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(\omega-\omega_{nl})}\right]$$

$$+\frac{\hat{F}'^{+}_{1nl}\hat{F}'^{+}_{1mk}\hat{F}'_{1kn}}{\omega-\omega_{mk}}\left[\frac{e^{-i(\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})}\right.$$

$$\left.-\frac{e^{-i(\omega-\omega_{nl})\ t}-1}{(\omega_{mk}+\omega_{kn})(\omega-\omega_{nl})}+\frac{e^{i(\omega_{kn}+\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(\omega_{kn}+\omega_{nl})}+\frac{e^{-i(\omega-\omega_{nl})\ t}-1}{(\omega+\omega_{kn})(\omega-\omega_{nl})}\right]$$

$$-\frac{\hat{F}'^{+}_{1nl}\hat{F}'^{+}_{1mk}\hat{F}'^{+}_{1kn}}{\omega-\omega_{mk}}\left[\frac{e^{-i(3\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})\ t}-1}{(2\omega-\omega_{mk}-\omega_{kn})(3\omega-\omega_{mk}-\omega_{kn}-\omega_{nl})}\right.$$

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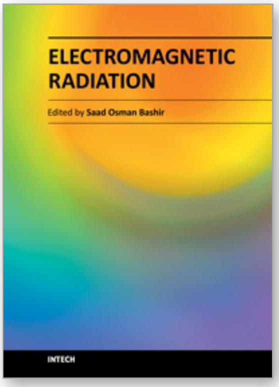

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