Terahertz wave generation from hyper-Raman lines in two-level quantum systems driven by two-color lasers

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Based on spatial-temporal symmetry breaking mechanism, we propose a novel scheme for terahertz (THz) wave generation from hyper-Raman lines associated with the 0th harmonic (a particular even harmonic) in a two-level quantum system driven by two-color laser fields. With the help of analysis of quasi-energy, the frequency of THz wave can be tuned by changing the field amplitude of the driving laser. By optimizing the parameters of the laser fields, we are able to obtain arbitrary frequency radiation in the THz regime with appreciable strength (as strong as the typical harmonics). Our proposal can be realized in experiment in view of the recent experimental progress of even-harmonics generation by two-color laser fields.

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Introduction

Terahertz (THz) radiation, electromagnetic radiation with typical frequency from 0.1 THz to 10 THz-lying in the spectrum gap between the infrared and microwaves, has varieties of applications in information and communication technology, biology and medical sciences, homeland security, and global environmental monitoring etc. The generation of THz wave is a challenge problem due to the lack of appropriate materials with small bandgaps in the usual optical approach. Great effort has been made to the design of THz sources1–15. THz wave can be obtained by both electronic and optical methods. The uni-travelling-carrier photodiode6 and the quantum cascade laser16,17 are two examples. The electronic approach is limited in the low frequency end of the THz regime, and the optical approach usually focuses on the cases of small energy gap. Technological innovation in photonics and nanotechnology has provided us more new ways for THz radiation generation. Recently an interesting approach based on semiconductor nanostructure driven by acoustic wave was suggested16, and electrically pumped photonic-crystal THz laser was developed17. THz wave generation by up conversion method through high-order harmonics generation(HHG) in semiconductor quantum dot(QD) was also suggested in our previous work18.

One challenge of THz wave generation in optical approach in usual system is to generate low frequency radiation from a quantum system with a large energy gap. Usually, in the emission spectrum of a quantum system, there are harmonics, as well as the associated hyper-Raman lines. Hyper-Raman lines are caused by the transitions between the dressed bound states. Nth hyper-Raman lines are defined as those associated with the nth harmonics (they appear with the corresponding harmonics simultaneously due to the same symmetry reason, and locate near n\(\omega_0\), where \(\omega_0\) is the fundamental frequency of the incident laser).19. Here is one observation that the shift of the hyper-Raman line to the associated harmonic could be small and could be tuned by the external field, even though the fundamental frequency is large. Then one may use the 0th hyper-Raman line (that associated with the 0th harmonic) to generate the low-frequency (THz) wave. There have been some studies on the harmonics and hyper-Raman lines. In many cases, there are only odd harmonics due to the particular symmetry and the associated hyper-Raman line is of high frequency and/or weak intensity. Therefore one has to solve these two problems to generate THz wave by using the hyper-Raman lines in the nonlinear optical processes.

In this article, we adopt an effective optical method to obtain THz wave in typical two-level quantum systems, which solves the two problems mentioned above. Based on the generalized spatial-temporal symmetry principle we developed recently20, we propose a novel mechanism to obtain the 0th hyper-Raman lines in THz regime by introducing additional laser with frequency 2\(\kappa\omega_0\) \((k = 1, 2, 3, \ldots)\). Using analysis of quasi-energy and optimization of laser fields, we are able to obtain considerably intense THz radiation with desired frequency. With the help of our optimization method, it is quite likely that our proposal can be realized in experiment considering the recent experimental progress of even-harmonics generation by two-color laser.

Theoretical formulism

Our two-level quantum system driven by laser fields [see the schematic diagram Fig. 1(a)] is described by the Hamiltonian

\[
H = \sum_{i=1}^{2} E_i |i\rangle \langle i| + G(t)|1\rangle \langle 2| + |2\rangle \langle 1|,
\]

where \(G(t) = F(t)e \cdot \mu_{12}\), is the Rabi frequency caused by periodic laser field \(E = F(t)e\), \(F(t) = F(t + T)\), \(e\) a unit vector. \(\mu_{12} = \langle 1|e|2\rangle\) is the dipole between state \(|1\rangle\) and state \(|2\rangle\). The energy spacing between the two states [with energies \(E_i(i = 1, 2)\)] is set as \(\Delta E = E_1 - E_2\).

The dynamics of our system is described by the equa-
tion of motion of the density matrix,
\[
\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}[H, \rho] - \Gamma \cdot \rho,
\tag{2}
\]
where $H$ is the Hamiltonian, the last term describes possible dissipative effects (such as spontaneous phonon emission) and we set $\hbar = 1$ in the following. We numerically calculate the photon emission spectra of the two-level quantum systems by solving the density matrix in Eq. (2) through Runge-Kutta method with the time step of 0.002/\(\omega_0\), total steps of 1200000 and the electron initially setting at the lower level. The average dipole can be calculated as $\mathbf{D}(t) = \sum_{ij} \mathbf{H}_{ij} \rho_{ij}(t)$. We use Fourier transformation to obtain the emission spectrum $S(\nu) = | \int dt \exp(-i\nu t) \mathbf{D}(t) |^2$. In the calculation we set $\hbar \omega_0$ (the driving field frequency) as the unit of energy. We choose $\omega_0 = 100THz$ as an example, yet in general one can also use other frequency (much larger than THz) laser as will be discussed later. The two-level quantum systems can be realized in many systems, such as atoms, molecules, and semiconductor quantum dots. Here we use the typical parameters of a quantum dot: the energy spacing $\Delta E = 100\omega_0$, the dipole moment $\mu_{12} = 0.5 e\cdot nm$, phonon emission coefficient $\Gamma_{ij} = 3.4 \times 10^{-3} \omega_0$.

We explore the emission spectra using above numerical approach and the theory on the selection rule in high-order harmonic generation based on the generalized spatial-temporal symmetry. As described in our previous work, for a quantum system described by Hamiltonian (1), if there exists one symmetric operation Q, which is the time shift $\theta : t \rightarrow t + T/2$ ($T = 2\pi/\omega_0$) combined with another operation $\Omega$ in spatial/spectral domain, i.e. $Q = \Omega : \theta$ , such that the initial condition and the Hamiltonian (up to a sign) are invariant, and the dipole operator $\hat{P}$ has a definite parity, then the emission spectrum contains no odd/even component if operator $\hat{P}$ is even/odd. For our two-level quantum system driven by a monochromatic laser, i.e. $E(t) = F_1 \cos(\omega_0 t)$, there exists only one symmetric operation $Q_1$, which is the time shift $\theta$ combined with spatial operation $\Omega_1$: $c_1 \rightarrow -c_1$ (here and in the following $c_j, j = 1, 2$, refers to the annihilation operator for state $|j\rangle$), and the Hamiltonian is invariant and the dipole operator $\hat{P}$ is odd. Then odd harmonics alone are generated because of the spatial-temporal symmetry. Other types of spatial-temporal symmetry may lead to many interesting emission patterns.

Here we give some analysis on the hyper-Raman lines and their relation to the harmonic components. In our systems driven by a periodic field, the quasienergy states has the form $|\psi_n(t)\rangle = e^{-i\omega_0 t}|\phi_n(t)\rangle$, where the Floquet state $|\phi_n(t)\rangle = |\phi_n(t + T)\rangle$ can be written in the form $|\phi_n(t)\rangle = \sum_m e^{-im\omega_0 t}|\phi_m^n\rangle$. We consider a state $\langle\psi(t)| = a_1|\psi_1\rangle + a_2|\psi_2\rangle = a_1 e^{-i\epsilon_1 t}|\phi_1\rangle + a_2 e^{-i\epsilon_2 t}|\phi_2\rangle$. Then we have
\[
\langle\psi|\hat{P}|\psi\rangle = \sum_{m,n} e^{-i(n-m)\omega_0 t}[a_1|^2\langle\phi_m^n|\hat{P}|\phi_1^n\rangle + |a_2|^2\langle\phi_m^n|\hat{P}|\phi_2^n\rangle]
\]
\[
+ \sum_{m,n} e^{-i[(\epsilon_1 - \epsilon_2) + (n-m)\omega_0] t}a_2 a_1^* \langle\phi_m^n|\hat{P}|\phi_2^n\rangle
\]
\[
+ \sum_{m,n} e^{-i[(\epsilon_1 - \epsilon_2) + (n-m)\omega_0] t}a_1 a_2^* \langle\phi_m^n|\hat{P}|\phi_1^n\rangle.
\tag{3}
\]

If the system posses a symmetry generated by the operator $\hat{Q} = \Omega \cdot \theta$ and the Floquet state $|\phi_n(t)\rangle$ has a definite parity under Q, i.e., $\hat{Q}\langle\phi_n(t)| = \pm \langle\phi_n(t)|$, then we have $\langle\phi_m^n|\hat{P}\phi_0^n\rangle = 0$ for $n-m$ even/odd number and states $\alpha, \beta$ of same/different parity. Quite often the harmonics and the associated hyper-Raman lines appear simultaneously due to the same symmetry properties of the Floquet states. In the case with a monochromatic laser, we have odd harmonics and the associated hyper-Raman lines. While in other cases with symmetry broken, more harmonics and the associated hyper-Raman lines are generated.

**Emission patterns** Let’s first look at the emission spectrum of a system driven by a monochromatic incident laser with frequency $\omega_0$. As seen in Fig.1(b), there are odd harmonics as well as the associated hyper-Raman lines due to the spatial-temporal symmetry properties of the Floquet states as analyzed above. It is natural that, for the incident laser with frequency $2\omega_0$, the emission spectrum contains components with frequencies of $2\omega_0$, $6\omega_0$ and $10\omega_0$... (odd orders of incident frequency $2\omega_0$) as shown in Fig.1(c). One notices that there is no 0th
hyper-Raman line in this case. If we use two-color lasers with frequencies $\omega_0$ and $2\omega_0$, interesting phenomena appear. As seen from Fig.1(d), even harmonics and the associated hyper-Raman lines are generated by introducing the second laser field. Especially a low frequency radiation, 0th hyper-Raman radiation is generated. We would like to point out that the emission spectrum for the case with driving field $F(t) = F_1 \cos(\omega_0 t) + F_2 \cos(2\omega_0 t)$ is not the supposition of those driven by $F_1 \cos(\omega_0 t)$ and $F_2 \cos(2\omega_0 t)$. For example, the harmonic $4\omega_0$ in Fig. 1(d) neither appears in Fig.1(b) nor in Fig.1(c). It is also not the frequency summation or difference of the harmonics of systems driven by monochromatic incident laser with frequency $\omega_0$ and $2\omega_0$. In fact, the appearance of all even components (and the associated hyper-Raman lines) in Fig. 1(d) is the consequence of symmetry breaking as discussed above.

According to our theory, the symmetry of the quantum system generated by $Q_1$ is broken by introducing the second laser with frequency $2k\omega_0$ ($k = 1, 2, 3...$), and it is not broken by introducing the second laser with frequency $(2k-1)\omega_0$ ($k = 1, 2, 3...$). These predictions are verified by our numerical results shown in Fig.1(e) (the second laser of frequency $3\omega_0$) and Fig.1(f) (the second laser of frequency $4\omega_0$). It is clear that the 0th hyper-Raman line does not appear in the cases with symmetry (see Figs.1(b) (c) (e)). Interestingly, the second laser field with frequency $4\omega_0$ leads to the appearance of 2nd harmonics (even for $n = 1$) and the associated hyper-Raman line (in particular the 0th hyper-Raman line) as predicted by our theory, since the second laser with frequency $4\omega_0$ breaks the spatial-temporal symmetry generated by $Q_1$. Using this mechanism, we can explain the experimental results of observing even harmonics in helium or plasma plumes (containing nanoparticles, carbon nanotubes, etc) driven by a two-color laser where the second-harmonic driving term, despite being very small, breaks the symmetry and thus allows additional strong even harmonic components. Our theory also naturally explains the generation of even harmonics and associated hyper-Raman lines by breaking the spatial symmetry.

As seen from equation (3), the frequency of hyper-Raman line is determined by the quasienergy, which is tunable. One may ask whether one can use the hyper-Raman line associated with 1th harmonic to generate the low frequency/THz wave? Actually, as seen from Fig. 2(a) the intensity of the hyper-Raman line associated with the 1th harmonic decreases dramatically as the frequency decreasing. To make this point clearer, we compare the low frequency components associated the 1th harmonic and 0th harmonic for the case with two-color laser shown in Fig. 2(b). One sees that in the every low frequency regime, the intensity of the 0th hyper-Raman line (associated with the 0th harmonic) is much larger than that of the 1th hyper-Raman line. Therefore one should use the 0th hyper-Raman line to generate the THz radiation effectively.

Tuning of the frequency and intensity of THz wave Now we discuss the optimization scheme of THz wave generation with driving field $F(t) = F_1 \cos(\omega_0 t) + F_2 \cos(2\omega_0 t)$ in our proposal. From equation (3), we see that the difference of the quasienergy is related to the shift of the hyper-Raman line form the corresponding harmonics, or the frequency of the 0th hyper-Raman line. This relation is verified by our numerical calculation as shown in Fig. 3. Moreover from Fig. 3, one sees that the 0th hyper-Raman line with very small frequency can be obtained in some parameter regimes, which can be used for THz wave generation. By applying this relation $\Delta = |\epsilon_1 - \epsilon_2| = h\nu (\nu$ the frequency of the 0th hyper-Raman line), any designated frequency THz wave can be obtained by tuning the magnitudes of the two external fields $F_1$ and $F_2$. For example, THz radiation of 2THz (0.02$\omega_0$) can be obtained by tuning the external field as shown in Fig. 3 [intersection points of the solid straight-line ($\nu = 0.02\omega_0$) and the line for quasienergy]. Fig. 4(a) shows the values for $F_1$ and $F_2$ under which the low frequency is $\nu = 0.02\omega_0$. There are disconnected parameter regimes of driving fields for generation of THz radiation. Furthermore, the intensity of the THz wave
Raman line with frequency $\omega$ we use the incident laser with frequency in the regime 3, but also by the frequency of the incident laser. If be tuned by the driving field intensity as seen in Fig. 4. The emitted hyper-Raman line frequency can not only can be optimized. One can find the proper parameters for the maximal intensity with desired frequency [the triangle in Fig. 4(a)]. The corresponding emission spectrum is shown in Fig. 4(b). It is seen that the intensity of THz radiation has been increased 200 times after optimization and we are able to obtain appreciable intense THz wave (as strong as typical harmonics). Here we estimate the emission power based on a system of arrays of semiconductor quantum quantum dots. The emission power of each dot is $P \sim (\mu \omega)^2 v / 3e^2 \sim 10^{-22} W$. There are about $N \sim 10^8$ dots in the regime of size of wavelength which emit wave in phase. Therefore, the total emission power from a sample of submillimeter size is around $N^2 P \sim \mu W$. 

In our approach, the typical driving field intensity is in the order of $10^{12} \sim 10^{13} W/cm^2$, which is lower than the typical driving field intensity (in the order of $10^{14} W/cm^2 \sim 10^{15} W/cm^2$) used in other methods for THz generation and/or HHG by two-color laser method. One should also use a long driving laser pulse [much longer than its period $(2\pi / \omega_0)$] as that used in Ref. [22]. The emitted hyper-Raman line frequency can not only be tuned by the driving field intensity as seen in Fig. 3, but also by the frequency of the incident laser. If we use the incident laser with frequency in the regime $\omega_0 \sim 50THz - 500THz$, we may obtain the 0th hyper-Raman line with frequency $\omega_0 \sim 1THz - 10THz$. 

In our method, there is another tunable parameter, the phase difference between two incident lasers. The main physical picture remains the same for different phases.

Unlike previous studies on non-linear driving two-level systems, the hyper-Raman lines and low frequency generation, our scheme of generating low frequency (THz) wave is based on the symmetry principle in typical two-level quantum systems of large energy spacing (in the order of $eV$). It can be easily obtained from natural systems (such as atoms, molecules, and semiconductors) and artificial structures (such as quantum dots). The large energy spacing makes it robust against thermal fluctuation and also insensitive to the initial state. Our mechanism based on the symmetry principle is different from those (based on four-wave mixing or transit current) used before. Our non-perturbative analysis and calculation show that one only needs to tune the intensity of the two-color laser for typical two-level quantum systems, which is very convenient in experiments. We may be able to obtain tunable THz radiation due to the availability of stable, tunable driving sources above the THz regime. Moreover, people have already observed even harmonics in experiments with two-color laser. Therefore it is quite likely that the 0th hyper-Raman line (in THz regime) associated with 0th harmonic can be obtained in experiment if our optimization method is used.

**Summary** A novel mechanism based on spatial-temporal symmetry breaking is proposed to obtain THz wave from 0th hyper-Raman line in two-color pumped two-level quantum system. Quasienergy is calculated to determine the parameters of the incident laser to obtain radiation of arbitrary frequency from 0.1THz to 10THz. Upon optimization of the driving fields, we are able to obtain THz wave with desired frequency and appreciable intensity (as large as that of the typical harmonics).

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