# Excitation of Semiconductor Nanosystems by Multy-Frequency Quantum Field

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**Abstract** The formation of Frenkel excitons in a 2D semiconductor nanosystem induced by frequency-multimode non-classical electromagnetic field is investigated. Strong sensitivity of excitation dynamics to the initial states of quantum field modes is found. The possibility to manage the excitation dynamics and controllably enhance a certain excitation channel by varying field frequency detuning and by choosing a proper initial state of the non-classical multimode field is demonstrated. The specific features of the excitation arising due to efficient coupling and energy exchange between initially independent field frequency modes are demonstrated. The polarization response of the considered nanosystem on the multi-frequency field impact is studied and its peculiarities arising due to the quantum features of the affecting non-classical light are found.

## 1. Introduction

The interaction of electromagnetic fields with nanostructured systems is an important direction of theoretical and experimental investigations and appears to be very promising for many practical applications. Nanostructured systems are characterized by interesting quantum features arising due to the effects of spatial quantization. At the same time, the interaction with electromagnetic fields opens up a wide range of possibilities to develop methods for controlling spatially localized excitations and current [1-10] in such structures and to use them for purposes of nanoelectronics and of quantum information algorithms. One of the relevant areas of such research is the investigation of the impact of non-classical fields since the experimental generation of different non-classical states of electromagnetic field becomes now possible [11-16]. The analysis of hybrid electron-photon systems provides the basis for the development of the full quantum photon-matter interface being very promising especially if the nanostructured systems are involved. .An important aspect in this case is the analysis of the effects arising when a set of frequency components of the quantum field are applied. In contrast to the case of a classical light [17], several quantum field modes are non-independent and can strongly influence each other via the interaction with the electronic subsystem of a nanostructure. Such possibility provides specific features of the field-induced excitation of a nanosystem and leads to new physical effects that do not take place in the classical case. The peculiarities of interaction of such multi-frequency non-classical light with different atomic and semiconductor systems remain still an open question and give rise to the development of quantum information protocols implemented with charge carriers and multiple excitations in nanostructures.

In this paper, the problem of the interaction of a semiconductor quantum well with a quantum field characterized by several frequency modes is analytically solved. In contrast to [18] different non-classical initial states of the quantum frequency modes are studied including coherent states with small mean number of photons and squeezed vacuum states and multiple excitations induced in the nanostructure are studied. Similar to [6-8] the manifold excitations of a quantum well are analyzed using the formalism of bosonic ladder operators and the time-dependent operators of creation and annihilation of excitations in electron and field subsystems are found analytically by direct solution of the Heisenberg equations. We focus mainly on the interaction of the nanostructure with two frequency modes of the quantum field with opposite detuning from resonant transition. Different regimes and time-dependent populations of different excitation channels of a quantum well are investigated and simultaneous influence of considered spectral field components are examined. The efficient energy exchange between considered field frequency modes is revealed and is shown to arise due to their interaction with electronic subsystem. The possibility to control the excitation dynamics and controllably enhance a certain excitation channel by varying field frequency detuning is demonstrated. The polarization response of the considered nanosystem on the multi-frequency field impact is studied and its peculiarities arising due to the quantum features of the affecting non-classical light are found.

## 2. Theoretical approach

The problem of excitation of a semiconductor quantum well by several frequency modes of quantum light is studied analytically with both electronic and quantum field degrees of freedom being taken into account. To provide efficient electron-field interaction the quantum well is supposed to be placed in a cavity with several eigenmodes corresponding to the frequencies of the acting field components. In such a case strong field effects can be observed even for low number of photons in a cavity. We suppose the planar dimensions of the quantum well to be infinitely large in comparison to its nanoscaled transverse size. Under such conditions the formation of excitons and multiple excitations in a nanosystem can be treated in terms of bosonic ladder operators [6-8]. For this reason the electronic subsystem as well as both field frequency modes are represented as linear harmonic oscillator with corresponded interaction terms taken in the dipole approximation. The quantum field is supposed to be characterized by two monochromatic frequency modes with opposite detuning $\pm\Delta\omega$ from the resonance.

In the frame of the rotating wave approximation [17] the total Hamiltonian of the system can be written in the form:

$$\hat{H} = \hbar\omega_0\hat{a}^+\hat{a} + \hbar(\omega_0 + \Delta\omega)\hat{b}^+\hat{b} + \hbar(\omega_0 - \Delta\omega)\hat{c}^+\hat{c} + \hbar\frac{g}{2}(\hat{a}^+\hat{b} + b^+\hat{a}) + \hbar\frac{g}{2}(\hat{a}^+\hat{c} + \hat{c}^+\hat{a}) \quad (1)$$

Here the creation operators $\hat{a}^+$, $\hat{b}^+$ and $\hat{c}^+$ stand for the electronic excitation and two frequency field modes respectively, g is the interaction strength supposed to be almost the same for both frequency modes and represented as $g = d\varepsilon_{0i}$ where $d$ is the dipole matrix element of the electronic transition and $\varepsilon_{0i}$ is a normalization constant for dimensionless electromagnetic field quadrature in each mode which depends on the cavity volume $L^3$**:** $\varepsilon_{0i} = \sqrt{\frac{4\pi\hbar\omega_i}{L^3}}$ [19,20]. We do not take into account any relaxation processes since we are interested in field-induced excitation at times shorter than characteristic damping time. We suppose the strength of atomic-field interaction which is determined by the mean photon density $< N >/ L^3$ large enough to provide the characteristic Rabi oscillations significantly faster than any decoherence processes in the system [21].

Different types of non-classical initial field states are considered including the Fock states $\Phi_k$ with different photon numbers *k*, coherent states $|\alpha >$ and squeezed states $\Psi_{sq}$ .

The coherent state of light can be expanded over different Fock states as follows [12]:

$$|\alpha > = \sum_n \exp\left(-\frac{|\alpha|^2}{2}\right)\frac{\alpha^n}{\sqrt{n!}}\Phi_n \quad (2)$$

This state is known to be characterized by the Poisson statistics of photons with mean photon number equal to $< N >= |\alpha|^2$ and photon number variance $D_n =< N >$.

At the same time the squeezed vacuum state is characterized by much more broader distribution over Fock states with extremely large photon number variance $D_n = 2\langle N\rangle^2 + 2\langle N\rangle$. The correspondent field wave function can be expanded over Fock states with only even photon numbers:

$$\Psi_{sq} = \sum_n C_{2n}\, \Phi_{2n} \tag{3}$$

with coefficients given by

$$C_{2n} = (-1)^n \sqrt{\frac{2\gamma}{1+\gamma^2}} \frac{\sqrt{(2n)!}}{2^n n!} \left(\frac{1-\gamma^2}{1+\gamma^2}\right)^n \tag{4}$$

where $\gamma$ is the squeezing parameter which determines the mean photon number as

$$\langle N\rangle = \frac{1}{4}\left(\gamma - \frac{1}{\gamma}\right)^2 \tag{5}$$

Using the Hamiltonian (1) the system of differential Heisenberg equations for the evolution of the considered exciton and photon operators is obtained and is given by:

$$\begin{aligned} &\hat{a}'(t) + \mathrm{i}\omega_0 \hat{a}(t) + \mathrm{i}\frac{g}{2}\left(\hat{b}(t) + \hat{c}(t)\right) = 0; \\ &\hat{b}'(t) + \mathrm{i}(\omega_0 + \triangle\, \omega)\hat{b}(t) + \mathrm{i}\frac{g}{2}\hat{a}(t) = 0; \\ &\hat{c}'(t) + \mathrm{i}(\omega_0 - \triangle\, \omega)\hat{c}(t) + \mathrm{i}\frac{g}{2}\hat{a}(t) = 0; \end{aligned} \tag{6}$$

The solution of these equations is found analytically in the form:

$$\hat{a}(t) = \exp[-i\omega_0 t]\,(\alpha_1(t)\hat{a}_0 + \alpha_2(t)\hat{b}_0 + \alpha_3(t)\hat{c}_0) \tag{7}$$

Here the zero-indices denote the operators in the Schrödinger representation and the time-dependent functions $\alpha_i(t)$ are given by:

$$\alpha_1(t) = \frac{1}{\frac{g^2}{2} + \Delta\omega^2}\left(\Delta\omega^2 + \frac{g^2}{2} Cos[\gamma t]\right);$$

$$\alpha_2(t) = -\alpha_3^+(t) = \frac{g}{2\left(\frac{g^2}{2} + \Delta\omega^2\right)}\left(\Delta\omega(Cos[\gamma t] - 1) - i\gamma Sin[\gamma t]\right)$$

where the parameter $\gamma = \sqrt{\Delta\omega^2 + \frac{g^2}{2}}$ is introduced. Similar expressions can be obtained for $\hat{b}(t)$ and $\hat{c}(t)$ by replacing $\alpha_i$ with another functions $\beta_i$ and $\gamma_i$ respectively (not given here due to their complexity).

The found evolution of operators allows to obtain all required characteristics of the interacting electron and field subsystems. For example, the mean energy of excitation of a nanosystem actually corresponds to the average number of excitations and can be found as follows:

$$< N_a >=< \psi_{in}|\hat{a}^+\hat{a}|\psi_{in} > \tag{8}$$

where the averaging of the time-dependent operators is performed over the initial state of the total system. Similarly, the mean number of photons in each frequency mode can be obtained. At the same time, the probability $W_{nkm}$ to find n-fold excitation of a quantum well, as well as *k* and *m* photons in the field frequency modes respectively can be obtained as follows:

$$W_{nmk} = \left|\frac{< 0|\hat{a}^n(t)\hat{b}^k(t)\hat{c}^m(t)|\psi_{in} >}{\sqrt{n!}\sqrt{m!}\sqrt{k!}}\right|^2 \tag{9}$$

Using the results of (9) the probability to have explicitly n-fold excitation of a quantum well can be obtained by averaging over field degrees of freedom and is given by:

$$W_n = \sum\nolimits_{k,m} W_{nkm} \tag{10}$$

Similarly the probability of certain number of photons in one or another quantum field mode can be calculated. Using the found probabilities the 2D distributions over different excitation channels of electronic subsystem are obtained in dependence on the field frequency detuning and time.

To obtain the time-dependent wave function of the total system the non-stationary Schrödinger equation with Hamiltonian (1)

$$i\hbar\frac{\partial\Psi}{\partial t}=\hat{H}\Psi \tag{11}$$

is solved numerically using the expansion of the solution over interaction-free eigenstates of the electronic and quantum field subsystems:

$$\psi(\vec{r},q_1,q_2,t)=\sum \mathrm{C}_{n,k,m}\,\phi_n(\vec{r})\widetilde{\phi_k}(q_1)\widetilde{\phi_m}(q_2)\exp\left[-\frac{iE_{nkm}t}{\hbar}\right] \tag{12}$$

with the interaction-free energy of the total system being equal to :

$$E_{nkm}=\hbar\omega_0\left(n+\frac{1}{2}\right)+\hbar(\omega_0+\triangle\,\omega)\left(k+\frac{1}{2}\right)+\hbar(\omega_0-\triangle\,\omega)\left(m+\frac{1}{2}\right)$$

Using the expansion (12) the system of differential equations for the probability amplitudes $\mathrm{C}_{nkm}(t)$ in (12) is obtained in the following form:

$$\begin{aligned}i\frac{d\dot{\mathrm{C}_{n,k,m}}}{dt}=(k-m)\Delta\omega C_{n,k,m}+g\sqrt{\frac{n(k+1)}{4}}\mathrm{C}_{n-1,k+1,m}+g\sqrt{\frac{k(n+1)}{4}}\mathrm{C}_{n+1,k-1,m}\\+g\sqrt{\frac{n(m+1)}{4}}\mathrm{C}_{n-1,k,m+1}+g\sqrt{\frac{m(n+1)}{4}}\mathrm{C}_{n+1,k,m-1}\end{aligned} \tag{13}$$

This system is solved numerically for different initial field states and under the assumption of the initial absence of the excitation in the electronic subsystem.

In addition, the time-dependent polarization response of the electronic subsystem $<\hat{d}(t)>$ was calculated both analytically and numerically. Taking into account that initially there is no excitation in electronic subsystem the polarization response can be calculated in terms of the time-dependent bosonic ladder operators as follows:

$$<\hat{d}(t)>=\frac{<\hat{a}^{+}(t)+\hat{a}(t)>}{\sqrt{2}}=\frac{1}{\sqrt{2}}(\alpha_2(t)<\psi_{in}|\hat{b}_0|\psi_{in}>+\alpha_3(t)<\psi_{in}|\hat{c}_0|\psi_{in}>+\text{h.c.}) \tag{14}$$

The found evolution of the mean dipole moment and its Fourier spectrum are analyzed in dependence on the properties of initial non-classical states of the quantum field modes.

## 3. Results and discussions

*3.1 Sensitivity of excitation dynamics to the initial quantum field state*

Let us first consider the influence of initial states of the quantum field frequency modes on the dynamics of the excitation. We consider 4 different cases: both modes are initially prepared in a certain Fock state, both modes are characterized by coherent states (2) with the same or different mean photon numbers, both modes are in a squeezed vacuum state (3) and finally, one mode is initially in coherent state and another in a squeezed vacuum state. Significant sensitivity of the excitation dynamics to the type of initial field states is found. Figure 1 demonstrates the time-dependent probabilities of different channel of multiple excitations induced in a nanosystem by quantum field.

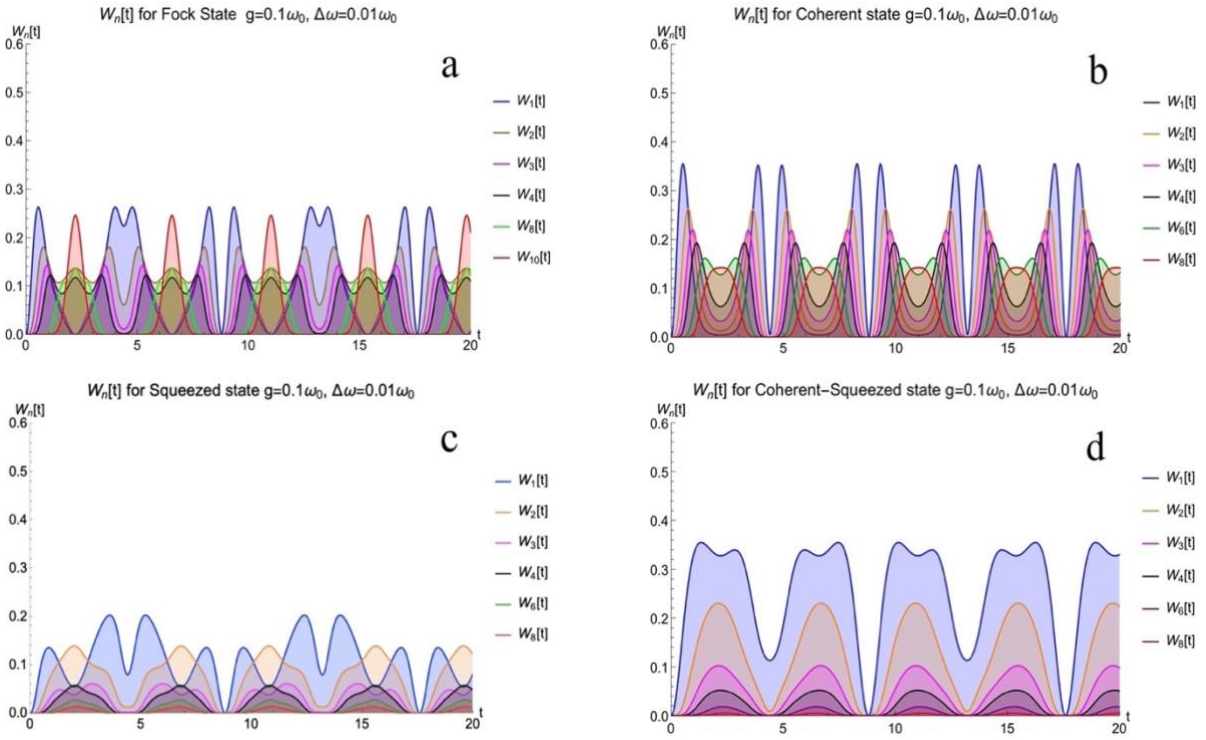


**Figure 1.** The time-dependent probabilities of different excitation channels induced in a nanosystem by different initial quantum field states: a) Fock states with equal photon numbers $n_b = 5\,, n_c = 5$, b) coherent states with equal mean photon numbers $<n_b>=2$, $<n_c>=2$, c) squeezed states with equal mean photon numbers $<n_b>=2$, $<n_c>=2$, and d) one coherent and one squeezed-vacuum state with mean equal photon numbers $<n_b>=2$, $<n_c>=2$.

It is seen that in the case of initial Fock states the dynamics of excitation differs dramatically for different excitation channels. Indeed, only in the case of Fock states (Figure 1a) the maximal probabilities of the highest and lowest excitation channels appear to be close to each other and rather high. In contrast, for two coherent initial states (Figure 1b) the excitation of channels takes place sequentially: the population of higher channels appears later in time and is significantly suppressed. As predicted, the excitation dynamics is periodic due to descrete number of photons and excitons in the field and electron subsystems respectively. Figure 1b shows the double frequency of oscillations in the case of two equal coherent states chosen initially for two field modes. Such symmetric field state gives rise to the destructive interference of excitation at a certain instant of time. As a result, a full absence of excitation is reached at time equal to half of the common period. In the case of interaction with two squeezed vacuum fields (Figure 1c) the excitation is less efficient due to large contribution of the vacuum state to the photon field and is characterized by different dynamics for odd and even excitation channels. Indeed, the local minima of odd channels coincide in time with local maxima of even channels. Such behaviour results from the specific feature of the squeezed initial state which is characterized by the population of even photon states only. Finally, in the case of one coherent and one squeezed vacuum field mode (Figure 1d) the excitation takes place simultaneously for all channels with maximal probability going down for higher channel numbers. Thus, it is possible to control the time-dependence of the excitation dynamics by varying the initial states of quantum field frequency modes.

*3.2 Control of the excitation dynamics by varying the field frequency detuning*

The time-dependent excitation probabilities discussed above are obtained for a certain value of the field frequency detuning. It is interesting to study the influence of the detuning on the excitation

process and peculiarities of its time-behaviour. In the case of only one frequency mode the dependence on the detuning is trivial: the lager is the detuning the lower is the efficiency of excitation. However for two symmetrically detuned frequency modes this is not the case since there is a mutual influence of both modes on the electronic subsytem. The 2D distributions representing the probability of the quadruple excitation in a nanosystem in dependence on time and detuning are plotted in Figure 2 for different initial field states.

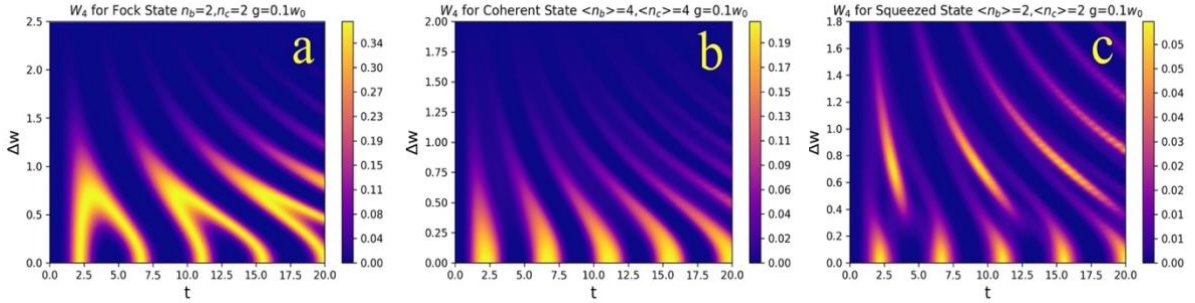


**Figure 2.** The 2D distributions representing the probability $W_4$ of the 4-th excitation channel in dependence on time and detuning for $g = 0.1\omega_0$, $\omega_0 = 10\ a.u.$ and for different initial field states: a) both field modes are in the Fock state with equal photon numbers $n_b = n_c = 2$, b) both field modes are in coherent state with equal mean photon numbers $<n_b>=<n_c>=4$, c) both field modes are in squeezed vacuum state with equal mean photon numbers $<n_b>=<n_c>=2$. Time unit corresponds to $1/10$ of the period of frequency.

The obtained results show that in the case of Fock field states (Figure 2a) the growing detuning firstly does not reduce the maximal probability of the channel but changes drammatically the time dynamics of its excitation. For sufficiently larger detuning the excitation probability goes down. In the case of two coherent initial states the behaviour is rather simple: the largest excitation probability is reached at zero detuning. A very non-trivial and interesting result is obtained for two squeezed vacuum field modes (Figure 2c). In addition to zero-value some rather large detuning values are also found to provide maximal excitation of considered channel. The found peculiarity comes from specific features of non-classical squeezed vacuum state and only even number of photons present in such field. Thus, there is a possibility to control and manage the excitation dynamics by choosing the appropriate detuning.

Nevertheless, the probability of excitation of high channels induced by squeezed modes appears to be rather low even for zero detuning. Thus, the Fock initial states of field seem to be the most promising to provide efficient excitation of high channels. Indeed, Figure 3 represents the 2 D distributions of excitation probability calculated for rather high channels of excitation induced in a nanosystem by quantum field modes with 5 and 10 initial numbers of photons.

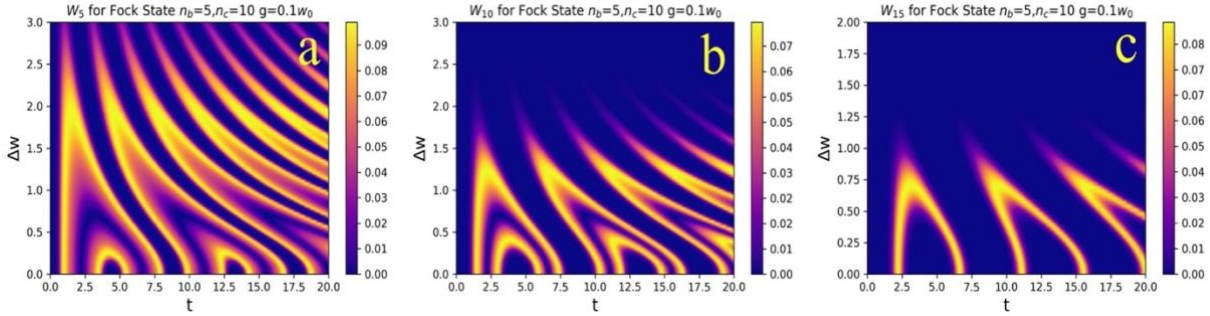


**Figure 3.** The 2 D distributions of excitation probability calculated for 5-th (a), 10-th (b) and 15-th (c) channels in dependence on time and detuning for $g = 0.1\omega_0$, $\omega_0 = 10\ a.u.$ and for field modes being initially in Fock states with 5 and 10 initial numbers of photons.

The maximal energy which can be transferred to the electronic subsystem corresponds to the excitation of the 15-th channel. The comparison of the maximal probability of excitation obtained for the 5-th, 10-th and 15-th channels shows almost the same values. Therefore the Fock initial states of field modes provide almost the same rather efficient excitation for intermediate and high channels.

*3.3 Mutual influence and energy exchange between field frequency modes*

In addition to the study of the excitation dynamics in a nanostructure, the analysis of the quantum field subsystem is of great interest. The most important feature of initially independent quantum field modes is their mutual influence and energy exchange arising due to their interaction with electronic subsystem. The distributions over different excitation channels for electronic subsystem and population of different Fock states of two frequency field modes is presented in Figure 4a-c in dependence on time for the case when only one field mode is initially populated by exactly 12 photons. Though this field mode is initially in the Fock state, the excitation of high channels in the electronic subsystem appears to besuppressed. The most part of the energy stored initially in populated field mode is efficiently transferred to the second mode via the interaction with a nanosystem. Indeed, the Figure 4c demonstrates the oscillations of the number of photons in the second field mode in time with maximal 12-th Fock state being periodically efficiently populated. Thus, the frequency modes appear to be strongly coupled to each other and exchange their energy due to the interaction with the electronic subsystem working as a transmission link.

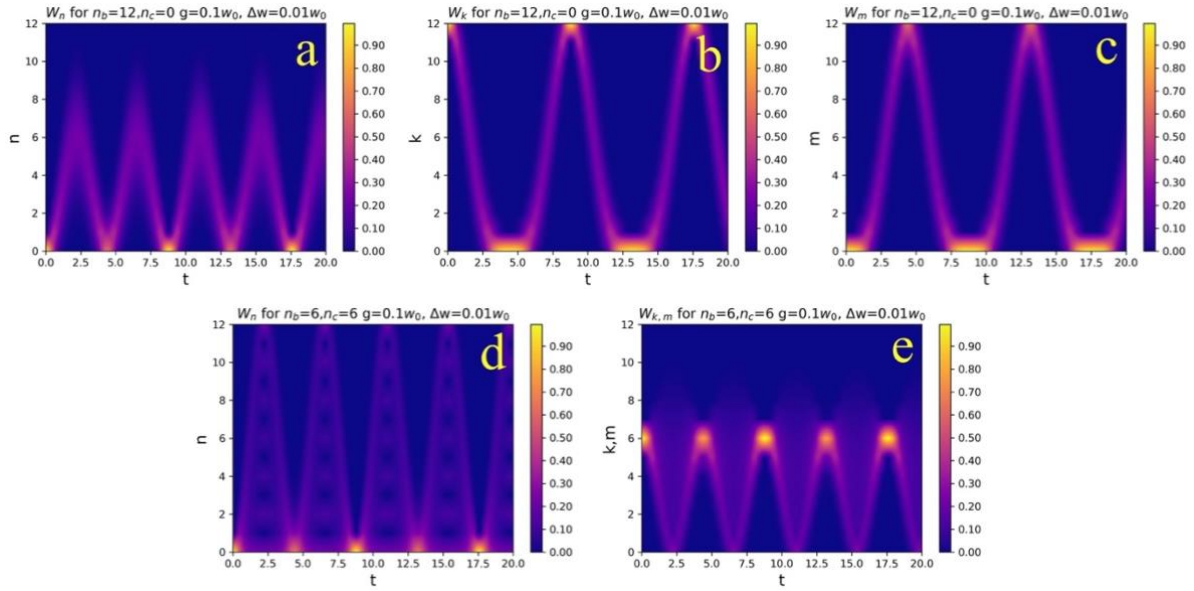


**Figure 4.** 2D- distributions over different excitation channels of the electronic subsystem (a, d) and population of different Fock states of two frequency field modes (b, c, e) in dependence on time obtained for field modes being initially in the Fock states with $n_b = 12\,, n_c = 0$ (a-c) and $n_b = 6\,, n_c = 6$ (d, e).

In the case of symmetrical initial field state when the field modes are initially in the Fock states with the same number of photons both modes contribute simultaneously to the excitation and the excitation of the highest channel is found to be enhanced (see Figure 4d). The constructive influence of two field modes consists in periodically taking place predominant population of the even excitation channels and at the same time suppression of energy transfer to the odd channels (Figure 4d). The time-dynamics of the distribution over different Fock states appears to be the same for both quantum field modes due to the symmetric initial condition. Moreover, it we have shown analytically that under such conditions the mean numbers of photons of both field modes are equal at any instant of time.

Similar distributions are obtained for the fields being initially in coherent and squeezed vacuum states. The data is presented in Figure 5 for the asymmetric condition when the second field mode is

initially empty. Transfer of energy and of the photon statistics from one field to another being initially in the ground vacuum state is clearly seen. At the same time, the distribution of electronic subsystem over different excitation channels is also characterized by the shape which is similar to the photon distribution in the acting field. Thus, it is possible to control and manage the distribution over excitation channels of a nanosystem by transfer the photon statistics to the electronic excitation.

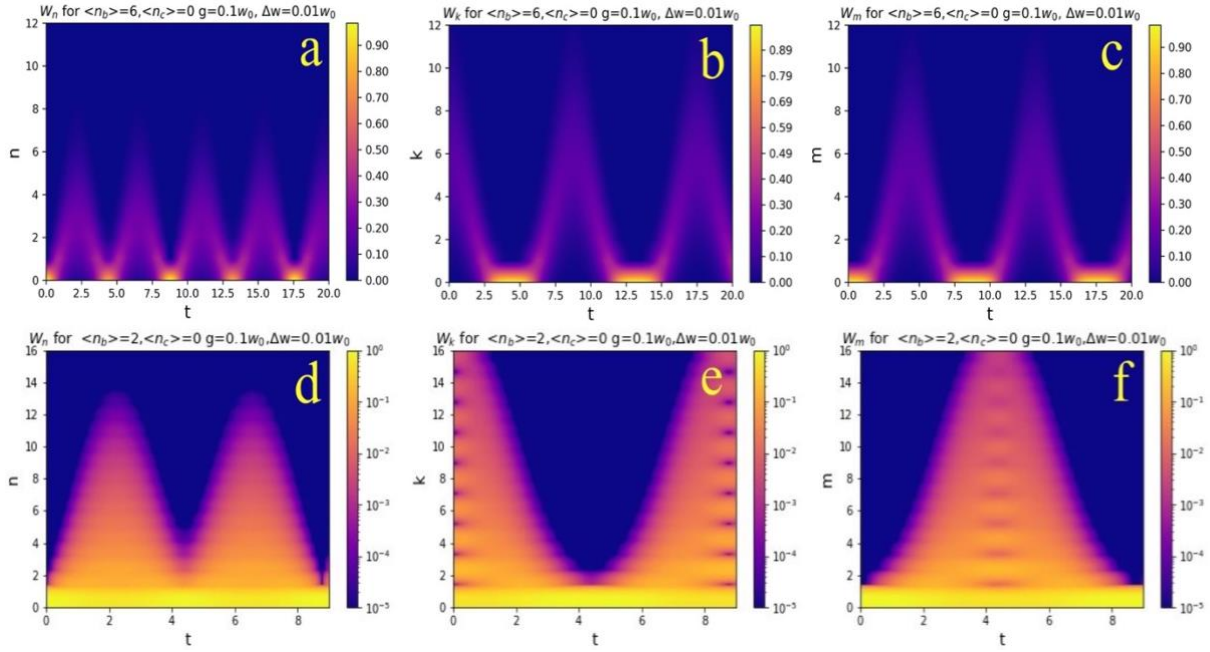


**Figure 5.** 2D-distributions over different excitation channels of the electronic subsystem (a, d) and population of different Fock states of two frequency field modes (b, c, e, f) in dependence on time obtained for asymmetrical initial condition when the second mode is empty and the first mode is in coherent state with mean photon number <$n_b$>=6 (a-c) or in squeezed vacuum state with mean photon number <$n_b$>=2 (d-f).

*3.4 Polarization response of the electronic subsystem*

One more important feature of the electronic subsystem characterizing the excitation process is the polarization response induced by quantum field modes during the interaction. The polarization response is known to be proportional to the mean dipole moment which in our case is calculated analytically in dependence on time according the Eq. (14). In the case of initial Fock states or squeezed vacuum states of field modes the dipole moment is found to be explicitly zero due to strong non-classical character of the acting fields. However, if only one or both of field modes are initially in the coherent state the polarization response is non-zero and is found to be very sensitive to the initial states and the parameters of the interaction. Figure 6 represents the time-dependent mean dipole moment of the electronic subsystem calculated analytically for two types of initial field states and different values of the interaction strength and detuning.

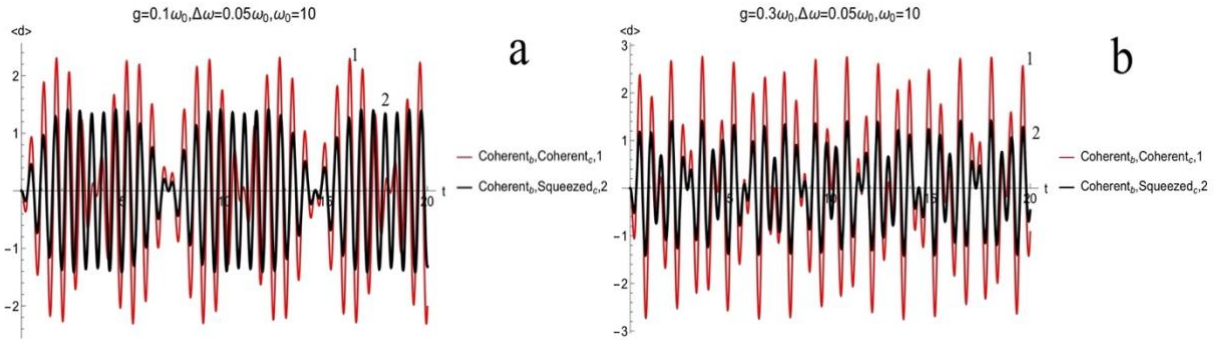


**Figure 6.** The time-dependent mean dipole moment of the electronic subsystem calculated analytically according to Eq. 14 for two coherent states (curve 1) and one coherent, one squeezed vacuum state (curve 2) of field modes with equal mean photon numbers $<n_b>=<n_c>=2$ and parameters $g = 0.1\omega_0, \Delta\omega = 0.05\omega_0$ $\omega_0 = 10$ $a.u.$ (a) and $g = 0.3\omega_0$ , the same detuning (b).

It can be seen that for relatively small interaction strength $g$ (Figure 6a) beats are observed, which occur 2 times more often for two coherent fields. At the same time, for rather strong interaction the modulation becomes less pronounced but oscillations with several frequencies contribute to the observed time-behaviour. The amplitude of oscillations is significantly smaller for the case when one of field modes in squeezed vacuum state due to large contribution of zero-Fock state in initial photon statistics.

Much more information can be obtained from the Fourier spectra of the found time-dependent polarization response which are presented in Figure 7. The results of Figure 7a, b are calculated for the data of Figure 6 for two coherent initial field states and one coherent, one squeezed vacuum state respectively.

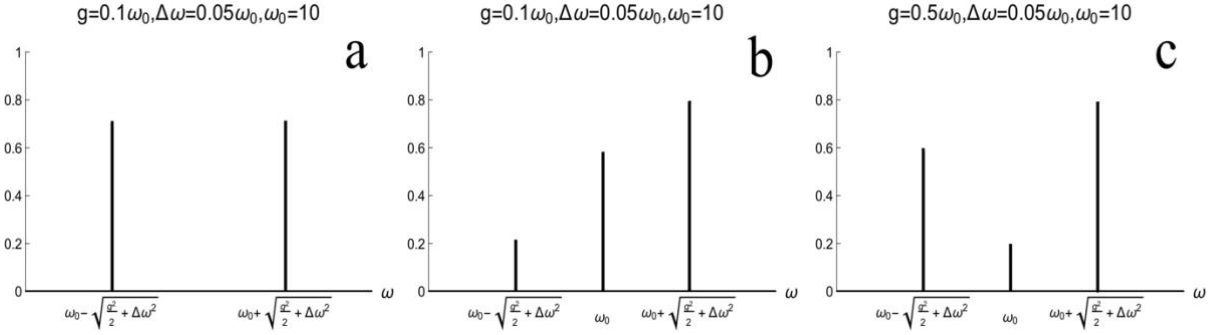


**Figure 7.** The Fourier spectra of the time-dependent polarization response <d>(t) (in arbitrary units) found in the case of two coherent initial field states (a) and one coherent, one squeezed vacuum state (b, c) for parameters of Figure 6a (a, b) and larger interaction strength (c).

It is interesting that for two coherent fields the central frequency peak is cancelled due to distructive contribution of symmetrical initial population of field modes. This found result is responsible for the doubled frequency of modulation of the mean dipole moment presented in Figure 6a. For the case of one coherent, one squeezed vacuum initial states of field modes different spectra of polarization response can be obtained. Figure 7b demonstrates a very specific spectrum with strongly asymmetrical side peaks. Such situation takes place for rather close values of the interaction strength $g$ and detuning. For more efficient interaction the spectrum takes the form mostly typical for the Rabi oscillation regime (Figure 7 c).

## 4. Conclusion

In conclusion, the excitation of a semiconductor quantum well by several frequency modes of quantum light is studied analytically and numerically with both electronic and quantum field degrees of freedom being taken into account. The time-dependent probability of excitation of different excitation channels is analyzed for different initial conditions and parameters of interaction. The

features and dynamics of excitation are found to depend dramatically on the initial states of the field frequency modes. The possibility to control the excitation dynamics and controllably enhance a certain excitation channel by varying field frequency detuning and by choosing the initial state of the non-classical multimode field is demonstrated. The specific features of the excitation arising due to mutual influence of two quantum field mods on the electron subsystem and each other are revealed. The efficient coupling and energy exchange between considered field frequency modes is found and is shown to arise due to their interaction with electronic subsystem. The polarization response of the electron subsystem induced by multi-frequency quantum field is investigated and its peculiarities arising due to the quantum features of the affecting non-classical light are found.

**Acknowledgements**

We acknowledge financial support of the Russian Science Foundation project No. 22-79-10018 and of the 'BASIS' Foundation (project No №22-2-1-69-1).

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