

# Quantum lithography on bound-free transitions

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**Abstract.** A new protocol for quantum lithography is presented. A formula which describes the single-quantum bound-free transition to the center of the continuous spectral zone under the action of two monochromatic photon beams is obtained. The derivation is based on the Markov approximation and takes into account all orders of the interaction parameter. The probabilities of bound-free transition for several initial field states are represented:  $N$ -photon, entangled  $N$ -photon and coherent states cases. The possibility of obtaining thin geometric structures on the surface of photoresist is discussed.

## 1 Introduction

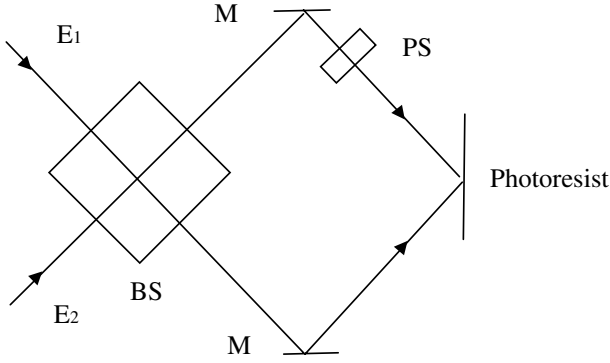
Optical lithography is an important tool of the semiconductor industry which makes it possible to create fine structures on the surface of substrates. New methods which improve the resolution of lithography and overcome the Rayleigh diffraction limit are presented. The different version of semi-classical optical lithography (classical description of light and quantum mechanical description the substrate) is presented in references [1–5]. In references [1–4] it is shown that the resolution of lithography processes can be increased by the use of substrates with a nonlinear (multiphoton) coefficient of absorption ( $N$ -photon substrates). So, references [1,2] suggest irradiating a two-photon substrate with three-frequency light whose frequencies satisfy a two-photon resonance condition. It has been shown that the distance between the stripes of an interference pattern can be reduced to  $\lambda/4$ . In references [3,4] the alternative method which uses interference between short pulses on a two-photon substrate was presented. Resolution of  $\lambda/4$  may be obtained using lithographic material with a narrow spectral absorption bandwidth and pulses with durations shorter than the lifetime of atomic excitation. In reference [5] has been shown that the high-frequency Rabi oscillations, caused by short driving pulses of light, can form a contrasting interference pattern on the surface of substrate with  $N$ -photon absorption.

The quantum interference of photon states is found to be a new direction of quantum optics, which was called quantum lithography. A recent review devoted to this field can be found in reference [6]. In references [7–10] it is shown that the quantum entangled  $N$  photon states of light can be used to increase the resolution of classical

photolithography. It is possible to increase the resolution of optical instruments up to a value of  $\frac{\lambda}{2N}$  using an entangled  $N00N$  state, with  $|N00N\rangle = \frac{|N,0\rangle + |0,N\rangle}{\sqrt{2}}$ . Here  $\lambda$  is the emission wavelength and  $N$  is the number of photons in the considered entangled state. Reference [10] presents the results of an experiment with a two-photon  $N00N$  state. In this work it is shown that resolution higher than the Rayleigh diffraction limit may be obtained. One difficult implementation of  $N$ -photon lithography is the development of photoresists which allow  $N$ -photon sensitivity. In optical lithography, different types of photoresists are used [11]. Photoresists are chemical substances whose chemical and physical properties are changed by the action of light. For example, photoresists formed from ortho-naphthoquinone or diazide- novolak resins act by photochemically dissociating to carbene and molecular nitrogen. Photochemical dissociation starts the reaction of photopolymerization. Atomic photoionization and molecular photodissociation are consequences of transitions from a bound state to a continuum spectrum state (bound-free transition) [12]. Tunable laser radiation can selectively excite any quantum state of the atoms and molecules of a certain type. Also, it is possible to selectively transfer significant energy to the atom or molecule by multistage excitation, causing their selective photoionization, photodissociation and other phototransformations [13].

Many aspects of quantum interference phenomena with participating continuum spectrum states have been investigated. For instance, the so-called Fano resonances [14] are well documented in the literature; a recent review can be found in reference [15]. Fano's resonance is revealed in the form of a sharp maximum in the absorption spectrum of inert gases, and also by the phenomenon of antiresonance, when the ionization probability at a specific frequency is equal to zero. In references [16–19] the

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**Fig. 1.** Diagram of quantum lithography experiment. Two field modes of  $E_1$  and  $E_2$  interfere on the photoresist, causing the necessary photo transformations. BS – beamsplitter, PS – phase plate, M – mirror.

methods for calculating the probabilities of bound-free transitions for a quantum system from discrete states to the continuum spectrum states are developed. Here the following methods of calculations were used: a perturbation theory, Markov approach, theory  $S$ -matrix and other methods.

In this paper, the interference pattern of two quantized field modes is studied, causing a single-photon transition of a quantum system into a state of a zone of continuous spectrum (bound-free transition). Nonlinear formulas of the one-photon transition probability were obtained in the Markov [20] approximation for the different photon quantum states. Graphs of the probability against the phase difference of the interfering fields are presented. The plausibility of inducing thin geometric structures on the substrate through interference patterns is discussed.

## 2 The probability of bound-free transitions in the Markov approximation

Let us assume that two field modes (Fig. 1) interact with the film. Both modes have identical polarizations which are parallel to this film.

Mode wave vectors are denoted by  $\mathbf{k}_1, \mathbf{k}_2$ ,  $|\mathbf{k}_1| = |\mathbf{k}_2| = k$ . Under the influence of photons an electron jumps from a bonding orbital to a continuum spectrum state (antibonding orbital). This passage leads to the breaking of an intramolecular bond and to the starting of a photochemical reaction in the film of photoresist. We simplify the description of this complex photochemical process by modeling the problem of the electron transition from one discrete level with the vector  $|0\rangle$  in a zone of the continuous spectrum. The continuum spectrum states are designated by  $|\varepsilon\nu\rangle$ . The problem in this formulation is considered in various contexts, for example, for describing the spectrum of autoionization [14], for the description of photodetection process [16], and for the analysis of ionization probabilities of weakly bound electrons [17]. In references [18,19] formulas for probabilities of bound-free transitions are obtained for the single-photon ionization,

nonlinearly depending on an exciting field. In contrast, the electromagnetic field in our work is assumed to be quantized. The Hamiltonian of electrons, modes, and their interaction takes the form

$$H = \sum_{\mu=1,2} \omega \hat{a}_{\mathbf{k}_\mu}^\dagger \hat{a}_{\mathbf{k}_\mu} + \sum_{\nu} \sum_{\varepsilon=-\Delta E/2}^{\Delta E/2} \times \left[ \left( \varepsilon + E_0 + \frac{\Delta E}{2} \right) |\varepsilon\nu\rangle \langle \varepsilon\nu| + \sum_{\mu=1,2} \left( f_{\mu;\varepsilon\nu}(\mathbf{r}) |\varepsilon\nu\rangle \langle 0| \hat{a}_{\mathbf{k}_\mu} + \{H.C\} \right) \right]. \quad (1)$$

Here  $|\varepsilon\nu\rangle$  is the continuum spectrum state with the energy  $E = \varepsilon + E_0 + \frac{\Delta E}{2}$ ,  $\nu$  represents additional quantum numbers of the state in the continuous spectrum,  $|0\rangle$  is the vector of the ground state, orthogonal to the states of continuous spectrum,  $E_0$  is lower boundary of the continuous spectrum and  $\Delta E$  is the width of the energy band. Also,  $f_{\mu;\varepsilon\nu}(\mathbf{r}) = d_{\mu;\varepsilon\nu} \exp i((\mathbf{k}_\mu, \mathbf{r}) + \varphi_\mu)$ ,  $d_{\mu;\varepsilon\nu}$  is the parameter of interaction between modes and an electron which transfer from the ground state to a continuum spectrum state with the energy  $E$ ,  $\mathbf{r}$  is the coordinate of the atomic nucleus, and  $\varphi_\mu$ , ( $\mu = 1, 2$ ) are additional phases in the beam. We will find the time evolution operator of the atom-field system:

$$i \frac{\partial}{\partial t} W(t) = H W(t), \quad W(0) = I.$$

The time evolution operator in the resonance representation is:

$$U(t) = \exp \left\{ i t \sum_{\nu} \sum_{\varepsilon=-\Delta E/2}^{\Delta E/2} \omega |\varepsilon\nu\rangle \langle \varepsilon\nu| \right\} \times \exp \left\{ i t \sum_{\mu=1,2} \omega \hat{a}_{\mathbf{k}_\mu}^\dagger \hat{a}_{\mathbf{k}_\mu} \right\} W(t).$$

The Hamiltonian in the resonance representation is:

$$H_R = \sum_{\nu} \sum_{\varepsilon=-\Delta E/2}^{\Delta E/2} \left[ \varepsilon |\varepsilon\nu\rangle \langle \varepsilon\nu| + \sum_{\mu=1,2} \left( f_{\mu;\varepsilon\nu}(\mathbf{r}) |\varepsilon\nu\rangle \langle 0| \hat{a}_{\mathbf{k}_\mu} + \{H.C\} \right) \right].$$

The field frequency coincides with the transition frequency to the center of the energy band:

$$\omega = E_0 + \frac{\Delta E}{2}.$$

We will decompose the time evolution operator in the atomic basis  $|0\rangle$ ,  $|\varepsilon\nu\rangle$

$$U(t) = |0\rangle\langle 0|U_{0,0} + \sum_{\nu} \sum_{\varepsilon=-\Delta E/2}^{\Delta E/2} (U_{\varepsilon\nu;0}|\varepsilon\nu\rangle\langle 0| + U_{0;\varepsilon\nu}|0\rangle\langle \varepsilon\nu|) + \sum_{\nu,\nu'} \sum_{\varepsilon,\varepsilon'=-\Delta E/2}^{\Delta E/2} U_{\varepsilon\nu;\varepsilon'\nu'}|\varepsilon\nu\rangle\langle \varepsilon'\nu'|.$$

The system of equations for the matrix elements of the time evolution operator is:

$$i\frac{d}{dt}U_{0,0} = \sum_{\nu} \sum_{\varepsilon=-\Delta E/2}^{\Delta E/2} \sum_{\mu=1,2} f_{\mu;\varepsilon\nu}^*(\mathbf{r}) \hat{a}_{\mathbf{k}\mu}^{\dagger} U_{\varepsilon\nu;0},$$

$$i\frac{d}{dt}U_{\varepsilon\nu;0} = \varepsilon U_{\varepsilon\nu;0} + \sum_{\mu=1,2} f_{\mu;\varepsilon\nu}(\mathbf{r}) \hat{a}_{\mathbf{k}\mu} U_{0,0}.$$

The initial conditions are:

$$U_{0,0}|_{t=0} = I, \quad U_{\varepsilon\nu;0}|_{t=0} = 0.$$

The integro-differential equation for  $U_{0,0}$  has the following form

$$\frac{d}{dt}U_{0,0} = -2\pi \sum_{\mu,\mu'=1,2} \hat{a}_{\mathbf{k}\mu}^{\dagger} \hat{a}_{\mathbf{k}\mu'} \int_0^t \frac{1}{2\pi} \sum_{\varepsilon=-\Delta E/2}^{\Delta E/2} \times \sum_{\nu} f_{\mu;\varepsilon\nu}^*(\mathbf{r}) f_{\mu';\varepsilon\nu}(\mathbf{r}) \exp\{i\varepsilon(t'-t)\} U_{0,0} dt'.$$

The Markov approximation [20] is utilized to solve this equation:

1.  $f_{\mu;\varepsilon\nu}^*(\mathbf{r}) f_{\mu';\varepsilon\nu}(\mathbf{r}) \approx f_{\mu;0\nu}^*(\mathbf{r}) f_{\mu';0\nu}(\mathbf{r})$ ,
2.  $\Delta E$  sufficiently large,
3.  $g(\varepsilon) \approx g(0)$ .

Here  $g(\varepsilon)$  is the density of states with the energy  $\varepsilon$ ,  $g(0)$  is the density of states in the center of band. We make the transition from summation to integration in the energy spectrum and obtain as a result

$$\frac{d}{dt}U_{0,0} \approx -2\pi\hat{\Gamma} \int_0^t \delta(t-t') U_{0,0} dt',$$

$$\hat{\Gamma} = \sum_{\mu,\mu'=1,2} \hat{a}_{\mathbf{k}\mu}^{\dagger} \hat{a}_{\mathbf{k}\mu'} \sum_{\nu} f_{\mu;0\nu}^*(\mathbf{r}) f_{\mu';0\nu}(\mathbf{r}) g(0), \quad (2)$$

and

$$\frac{d}{dt}U_{0,0} \approx -\pi\hat{\Gamma}U_{0,0},$$

and then

$$U_{0,0} = U_{0,0}(t) = \exp\{-\pi\hat{\Gamma}t\}.$$

The probability of a bound-free transition is determined by the formula

$$P_{BF}(t) = 1 - Sp_f \left( \rho_f(0) \exp(-2\pi\hat{\Gamma}t) \right). \quad (3)$$

Here  $\rho_f(0)$  is the initial density matrix of field modes. Supposed, that an atom at the initial moment is found in the ground state. Let us study the properties of the operator  $\hat{\Gamma}$  (2), which we write as:

$$\hat{\Gamma} = \gamma \left( \hat{N} + \hat{a}_1^{\dagger} \hat{a}_2 e^{i\varphi} + \hat{a}_2^{\dagger} \hat{a}_1 e^{-i\varphi} \right).$$

Here

$$\gamma = \left| \sum_{\nu} f_{\mu;0\nu}^*(\mathbf{r}) f_{\mu';0\nu}(\mathbf{r}) g(0) \right|,$$

$\varphi = (\mathbf{k}_2 - \mathbf{k}_1, \mathbf{r}) + \varphi_2 - \varphi_1$  is the phase difference between beams and  $\hat{N} = \hat{a}_1^{\dagger} \hat{a}_1 + \hat{a}_2^{\dagger} \hat{a}_2$  is the photon number operator. We introduce operators corresponding to the su(2) Lie algebra ( $S$  is the index of representation)

$$S_- = \hat{a}_2^{\dagger} \hat{a}_1, \quad S_+ = \hat{a}_1^{\dagger} \hat{a}_2,$$

$$S_0 = \frac{\hat{a}_1^{\dagger} \hat{a}_1 - \hat{a}_2^{\dagger} \hat{a}_2}{2}, \quad S = \frac{N}{2}, \quad S^2 = S(S+1).$$

Operator  $\hat{\Gamma}$  takes the form

$$\hat{\Gamma} = \gamma \left( \hat{N} + S_+ e^{i\varphi} + S_- e^{-i\varphi} \right).$$

We introduce the notation of the eigenvectors of the operator  $S_0$

$$S_0 |S, m\rangle = m |S, m\rangle, \quad \hat{N} |S, m\rangle = 2S |S, m\rangle = N |S, m\rangle, \\ -S \leq m \leq S.$$

The Fock and su(2) basis are related as follows:

$$|N-n, n\rangle_F = |S, m\rangle = \left| \frac{N}{2}, \frac{N}{2} - n \right\rangle, \quad n = 0, 1, \dots, N.$$

$\hat{\Gamma}$  is then diagonalised

$$\hat{\Gamma}_d = V^{\dagger} \hat{\Gamma} V = \gamma \left( \hat{N} + 2S_0 \right),$$

by a unitary transformation (transfer to the coherent representation of the su(2) algebra [21])

$$V = \exp(i\varphi S_0) \exp\left(-\frac{\pi}{4}(S_+ - S_-)\right).$$

The spectrum and the eigenvectors of the operator  $\hat{\Gamma}$  are

$$\hat{\Gamma} \exp(i\varphi S_0) |I_{S,m}\rangle = 2\gamma(S+m) \exp(i\varphi S_0) |I_{S,m}\rangle, \\ |I_{S,m}\rangle = \exp\left(-\frac{\pi}{4}(S_+ - S_-)\right) |S, m\rangle.$$

Here  $|\Gamma_{S,m}\rangle$  are the coherent states of  $\text{su}(2)$ . Equation (3) for the probability can be written in the basis  $|\Gamma_{S,m}\rangle$ .

$$P_{BF}(t) = 1 - \sum_{S=0}^{\infty} \sum_{k=-S}^S \exp(-4\pi(S+k)\gamma t) \langle \Gamma_{S,k} | \exp(-i\varphi S_0) \rho_f(0) \exp(i\varphi S_0) | \Gamma_{S,k} \rangle. \quad (4)$$

Vector  $|\Gamma_{S,m}\rangle$  can be expanded in terms of the basis  $|S, m\rangle$

$$\begin{aligned} |\Gamma_{S,m}\rangle &= \exp\left(-\frac{\pi}{4}(S_+ - S_-)\right) |S, m\rangle \\ &= \sum_{q=-S}^S \langle S, q | \exp\left(-\frac{\pi}{4}(S_+ - S_-)\right) |S, m\rangle |S, q\rangle. \end{aligned}$$

The matrix elements of the operator  $\exp(-\frac{\pi}{4}(S_+ - S_-))$  take the form [21]

$$\begin{aligned} \langle S, q | \exp\left(-\frac{\pi}{4}(S_+ - S_-)\right) |S, m\rangle &= 2^m (-1)^{q-m} \sqrt{\frac{(S+q)!(S+m)!}{(S-q)!(S-m)!}} \\ &\times \sum_{p=\max\{0, m-q\}}^{S+m} \left(-\frac{1}{2}\right)^p \frac{(S-m+p)!}{p!(S+m-p)!(q+p-m)!}. \end{aligned}$$

The vector  $|\Gamma_{S,-S}\rangle$ , which corresponds to zero eigenvalue, takes the following form in the Fock basis

$$\begin{aligned} |\Gamma_{S,-S}\rangle &= \left(\frac{1}{\sqrt{2}}\right)^N \sum_{n=0}^N (-1)^n \sqrt{\frac{N!}{(N-n)!n!}} |n, N-n\rangle_F \\ &= \frac{1}{\sqrt{N!}} \left(\frac{-\hat{a}_1^\dagger + \hat{a}_2^\dagger}{\sqrt{2}}\right)^N |0\rangle_F. \end{aligned}$$

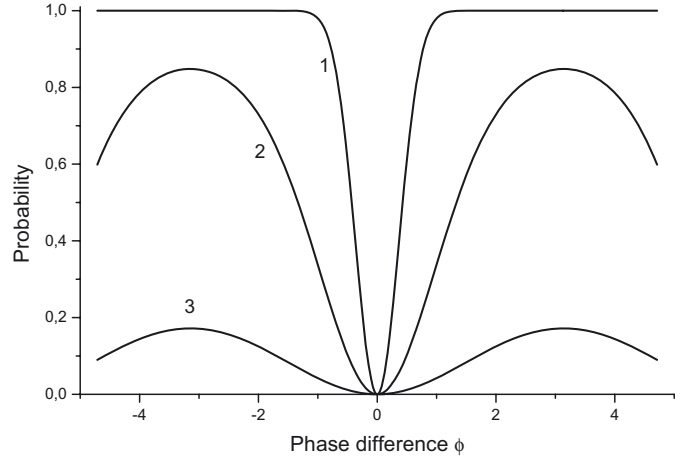
### 3 Nonlinear antiresonance on bound-free transitions

We calculate and graph the probability  $P_{BF}(t)$  (4) for several photon states. We choose the initial pure  $N$ -photon state of two modes  $|N\rangle = \exp(i\varphi_0 S_0) |\Gamma_{S,-S}\rangle$ . The initial density matrix takes the form

$$\rho_f(0) = |N\rangle \langle N|. \quad (5)$$

It is easy to obtain this state, after passing the  $N$ -photon state of the first mode  $|N, 0\rangle_F$  through the beam splitter BS and the phase plate PS with the transfer matrix  $PS \times BS$  (Fig. 1)

$$\begin{aligned} \begin{pmatrix} \hat{a}_1^\dagger \\ \hat{a}_2^\dagger \end{pmatrix}_{in} &= PS \times BS \begin{pmatrix} \hat{a}_1^\dagger \\ \hat{a}_2^\dagger \end{pmatrix} \\ &= \begin{pmatrix} \exp(i\varphi_0/2) & 0 \\ 0 & \exp(-i\varphi_0/2) \end{pmatrix} \begin{pmatrix} -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{pmatrix} \begin{pmatrix} \hat{a}_1^\dagger \\ \hat{a}_2^\dagger \end{pmatrix}. \end{aligned}$$



**Fig. 2.** Probability  $P_{BF}(t)$  as a function of the phase difference  $\varphi$  of the interfering beams. Initial state of the two modes:  $|N\rangle = \frac{1}{\sqrt{N!}} \left(\frac{-\hat{a}_1^\dagger + \hat{a}_2^\dagger}{\sqrt{2}}\right)^N |0\rangle_F$ . Number photons:  $N = 15$ . Initial phase:  $\varphi_0 = 0$ . Graph 1 –  $\gamma t \rightarrow \infty$ , graph 2 –  $\gamma t = 0.01$ , graph 3 –  $\gamma t = 0.001$ .

Then the probability  $P_{BF}(t)$  will depend on the phase difference  $\varphi - \varphi_0$

$$\begin{aligned} P_{BF}(t) &= 1 - \sum_{k=-S}^S \exp(-4\pi(S+k)\gamma t) \\ &\times |\langle \Gamma_{S,k} | \exp(-i(\varphi - \varphi_0)S_0) | \Gamma_{S,-S} \rangle|^2, \quad S = \frac{N}{2}. \end{aligned} \quad (6)$$

In the asymptotic limit  $4\pi\gamma t \gg 1$  and the  $k = -S$  term dominates the sum, giving

$$\begin{aligned} P_{BF}(t) &= 1 - |\langle \Gamma_{S,-S} | \exp(-i(\varphi - \varphi_0)S_0) | \Gamma_{S,-S} \rangle|^2 \\ &= 1 - \left(\cos \frac{\varphi - \varphi_0}{2}\right)^{2N}. \end{aligned} \quad (7)$$

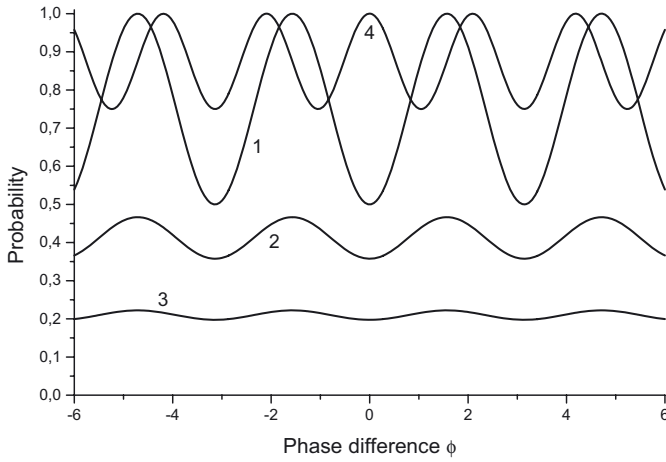
Equation (7) describes narrow interference hole (antiresonance) of probability when  $\varphi \approx \varphi_0$ . The half-width at half-height of the hole  $\Delta\varphi(\xi)$  depends on  $N$

$$\Delta\varphi \approx \sqrt{\frac{2}{N}}.$$

The graphs dependence of  $P_{BF}(t)$  on a phase difference  $\varphi$  for several values of the exposure time are represented in Figure 2. The graphs are calculated by equations (6) and (7) for the initial density matrix (5).

Now we select the initial pure entangled  $N$ -photon state of two modes ( $N00N$  state)

$$|N\rangle = |N00N\rangle = \frac{|N, 0\rangle_F + |0, N\rangle_F}{\sqrt{2}} = \frac{|S, -S\rangle + |S, S\rangle}{\sqrt{2}}. \quad (8)$$



**Fig. 3.** Probability  $P_{BF}(t)$  as a function of the phase difference  $\varphi$  of the interfering beams. Initial state of the two modes:  $|N00N\rangle = \frac{|N,0\rangle_F + |0,N\rangle_F}{\sqrt{2}}$ . Initial phase:  $\varphi_0 = 0$ . Graph 1 –  $\gamma t \rightarrow \infty$ ,  $N = 2$ , graph 2 –  $\gamma t = 0.05$ ,  $N = 2$ , graph 3 –  $\gamma t = 0.02$ ,  $N = 2$ , graph 4 –  $\gamma t \rightarrow \infty$ ,  $N = 3$ .

We substitute (8) into (4) and obtain

$$P_{BF}(t) = 1 - \left(\frac{1}{2}\right)^N \left( (1 + e^{-4\pi\gamma t})^N + (-1)^N (1 - e^{-4\pi\gamma t})^N \cos(\varphi N) \right). \quad (9)$$

In the asymptotic limit  $4\pi\gamma t \gg 1$  we have

$$P_{BF}(t) = 1 - \left(\frac{1}{2}\right)^N \left( 1 + (-1)^N \cos(\varphi N) \right). \quad (10)$$

With an increase in the number of photons the fringe width  $\frac{\lambda}{2N}$  decreases and the visibility of the interference pattern  $\frac{2^{-N}}{1-2^{-N}}$  at the same time substantially decreases. The graphs dependence of  $P_{BF}(t)$  on the phase difference  $\varphi$  for several values of the exposure time are represented in Figure 3. The graphs are calculated from equations (9) and (10) for the initial density matrix (8).

We examine the initial coherent state of two modes with amplitudes  $\xi_1$  and  $\xi_2$

$$\rho_f(0) = |\xi_1, \xi_2\rangle\langle\xi_1, \xi_2|, \quad |\xi_1, \xi_2\rangle = e^{-\frac{\xi_1^2 + \xi_2^2}{2}} \times \sum_{S=0}^{\infty} \sum_{m=-S}^S \frac{(\xi_1 \xi_2)^S}{\sqrt{(S+m)!(S-m)!}} |S, m\rangle. \quad (11)$$

Then the ionization probability is:

$$\begin{aligned} P_{BF}(t) &= 1 - \sum_{S=0}^{\infty} \sum_{k=-S}^S \exp(-4\pi(S+k)\gamma t) |\langle F_{S,k} | \\ &\quad \times \exp(-i\varphi S_0) |\xi_1, \xi_2\rangle|^2 \\ &= 1 - \exp \left\{ -\frac{1 - \exp(-4\pi\gamma t)}{2} (|\xi_1|^2 + |\xi_2|^2 \right. \\ &\quad \left. + \xi_1^* \xi_2 e^{i\varphi} + \xi_2^* \xi_1 e^{-i\varphi}) \right\}. \end{aligned} \quad (12)$$

For short times  $t$  we obtain Fermi's golden rule, and also Mandel's formula for the photodetection rate [22]

$$\frac{P_{BF}(t)}{t} \approx 2\pi\gamma (|\xi_1|^2 + |\xi_2|^2 + \xi_1^* \xi_2 e^{i\varphi} + \xi_2^* \xi_1 e^{-i\varphi}).$$

For the case  $|\xi_1| = |\xi_2| = |\xi|$ , we obtain the known formula for the interference pattern

$$\frac{P_{BF}(t)}{t} \approx 4\pi\gamma |\xi|^2 (1 + \cos(\varphi - \delta)). \quad (13)$$

Here  $\delta$  is the phase difference of the coherent amplitudes  $\xi_1$  and  $\xi_2$ . In equation (13) the bandwidth of the interference fringes does not depend on the light intensity. In the asymptotic limit  $4\pi\gamma t \gg 1$  we have

$$P_{BF}(t) \xrightarrow{t \rightarrow \infty} 1 - \exp(-|\xi|^2 (1 + \cos(\varphi - \delta))). \quad (14)$$

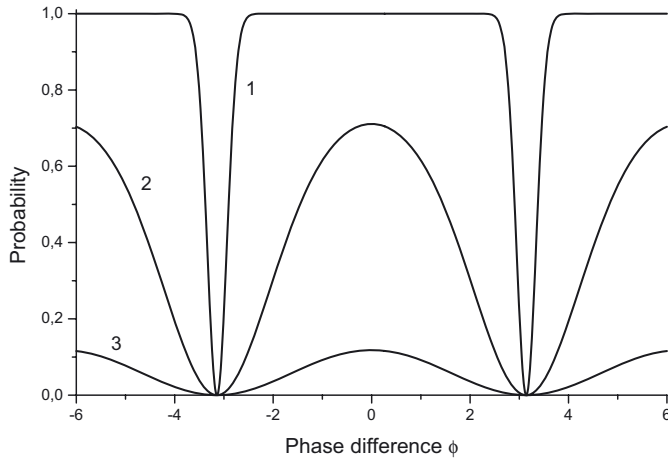
In contrast to (13), equation (14) describes a narrow interference hole (antiresonance) in the probability around  $\varphi - \delta \approx \pi$ . The half-width at half-height of the hole  $\Delta\varphi(\xi)$  depends on the average number of photons in the mode

$$\Delta\varphi(\xi) \approx \frac{\sqrt{\ln 4}}{|\xi|}.$$

The graphs dependence of  $P_{BF}(t)$  on the phase difference  $\varphi$  for several values of the exposure time are represented in Figure 4. Graphs are calculated from equations (12) and (14) for the initial density matrix (11).

## 4 Conclusion

The purpose of an optical lithography is to create a thin geometric structure on the surface of the substrate. Diffraction of light diffuses the structure, the edges of stripes have low contrast, and the distances between the stripes are limited by the diffraction limit. In references [1–10] several methods of overcoming the diffraction limit are proposed. In our work the new protocol of quantum lithography is formulated. The interference of two quantized modes, which interact with the substrate, and which cause the single-quantum transition of molecules into the continuum spectrum states (bound-free transition) is investigated. We obtained equation (3), which describes a single-quantum bound-free transition to the center



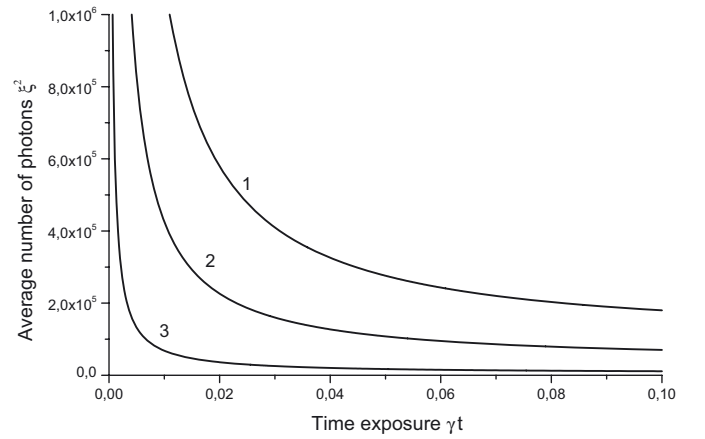
**Fig. 4.** Probability  $P_{BF}(t)$  as a function of the phase difference  $\varphi$  of the interfering beams. Initial state of the two modes:  $|\xi, \xi\rangle = e^{-\xi^2} \sum_{S=0}^{\infty} \sum_{m=-S}^S \frac{(\xi)^{2S}}{\sqrt{(S+m)!(S-m)!}} |S, m\rangle$ . The average number of photons in the coherent state:  $\xi^2 = 25$ . Initial phase:  $\delta = 0$ . Graph 1 –  $\gamma t \rightarrow \infty$ , graph 2 –  $\gamma t = 0.002$ , graph 3 –  $\gamma t = 0.0002$ .

of the continuous spectrum zone under the action of two monochromatic beams of photons. The derivation is based on the Markov approximation and takes into account all orders of the interaction parameter. An image on the photoresist is formed during irradiation with ultraviolet light at a wavelength of 250 nm [11]. Characteristic radiation doses were around  $5 \times 10^{-2}$  J/cm<sup>2</sup>. With the aid of equation (12) it is possible to obtain the dependence of the average number of photons in the mode  $n(t) = \xi^2$  on the time of exposure with the fixed values  $P_{BF}(t) = P_{cr}$  and  $\Delta\varphi(\xi, t) = \Delta\varphi_{cr}$

$$n(t) = \frac{-\ln(1 - P_{cr})}{2 \left( \sin\left(\frac{\Delta\varphi_{cr}}{2}\right) \right)^2 (1 - \exp(-4\pi\gamma t))}. \quad (15)$$

Figure 5 shows dependence  $n(t)$  on a time exposure  $\gamma t$  with  $P_{cr} = 0.8$  and three values of the half-width of antiresonance  $\Delta\varphi_{cr} = 0.005, 0.008, 0.02$ .

Equation (15) gives the possibility to select the exposure time in the dependence on the parameter  $\gamma$  and the photon numbers in the mode so as to obtain a narrow stripe in the photoresist on the substrate where the polymerization reaction does not passed. The described protocol can be used in two ways. For the negative photoresists this protocol makes it possible to create on the substrate a stripe with a width of  $\frac{\lambda}{2}$  and sharp contrast edges. For a positive photoresists, due to the nonlinearity, thin stripes can be formed on the substrate with a width that is smaller than wavelength, but the distance between these stripes is equal to  $\frac{\lambda}{2}$ . With the use of  $N00N$  photon states it is possible to obtain surface structures with high resolution  $\frac{\lambda}{2N}$ , but with a visibility  $\frac{1}{2N-1}$ . The proposed protocol complements and extends the capabilities of the protocols proposed in the above-mentioned works. The effect presented



**Fig. 5.** Average number of photons  $n(t) = \xi^2$  in the mode at the exposure time  $\gamma t$  with fixed values  $P_{cr} = 0.8$  and  $\Delta\varphi_{cr} = 0.005, 0.008, 0.02$ . The initial state of the two modes are coherent  $|\xi, \xi\rangle = e^{-\xi^2} \sum_{S=0}^{\infty} \sum_{m=-S}^S \frac{(\xi)^{2S}}{\sqrt{(S+m)!(S-m)!}} |S, m\rangle$ . Graph 1 –  $\Delta\varphi_{cr} = 0.005$ , graph 2 –  $\Delta\varphi_{cr} = 0.008$ , graph 3 –  $\Delta\varphi_{cr} = 0.02$ .

here can be used to increase the resolution of the photolithography process.

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