Frequency-pulsed regime of a spherical microcavity excitation by ultrashort chirped laser radiation

D.V. Apeksimov, Yu.E. Geints, and A.A. Zemlyanov

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk

Received August 29, 2007

Resonance excitation of the internal optical field of transparent spherical microparticles under irradiation by a train of ultrashort laser pulses has been simulated numerically. It has been determined that the incident radiation can be tuned optimally to preset high-Q resonance with a particle by varying the pulse ratio in the train with the linear frequency modulation of every pulse in the train (chirping). Analytical equations have been derived for calculation of these parameters depending on the laser pulse duration and the frequency of the resonance to be excited.

Introduction

Miniature high-sensitivity optical devices (optical biosensors,[1] polymer optical fibers,[2] nanostructure semiconductors,[3] photon crystals,[4] microlaseres[5]) are actively developed in recent years. The main element of these devices is an optical microcavity, which serves for amplification and controllable conversion of optical radiation. Such a cavity is usually represented by an optically transparent dielectric microparticles of spherical or cylinder form, whose dimensions are much larger than the wavelength of the working radiation. The operating principle is based on excitation of the so-called whispering gallery modes (WGMs).

Whispering gallery modes form a subfamility of natural electromagnetic oscillation modes of a cavity. They are characterized by a very high Q-factor ($> 10^5$), narrow spectral profile, rather long lifetimes (about nanoseconds), and the high degree of localization of the optical field near the cavity surface. Just these properties of WGMs have found the utility in optical microelectronic devices for frequency filtering and amplitude modulation of optical radiation and optical connection of elements. The also can be used in lasing at new wavelengths due to nonlinear effects of stimulated Raman scattering, stimulated fluorescence, and harmonic generation. Natural frequencies of WGMs are determined by a microparticle size and its optical properties.

As the frequency of an optical wave, incident on the particle, coincides with the frequency of one of its natural modes, there appears a resonance excitation of the internal optical field, and its spatiotemporal distribution is fully determined by the excited mode field.

A key point in the increase of the microcavity operation efficiency is achievement of the optimal regime of WGM excitation by optical radiation, which requires a rather accurate tuning to the resonance. It is not always possible and convenient to meet this requirement at a given cavity geometry by varying the frequency of the incident radiation. Application of ultrashort laser pulses with pico- and femtosecond duration allows the initially wide spectral composition of the radiation to be used. Consequently, this can facilitate the tuning of selected WGMs to the resonance.[6]

Earlier, it has been shown theoretically that the use of the frequency-pulsed excitation of a spherical microcavity by a train of ultrashort laser pulses instead of irradiation by single pulses allows the control over this process.[7] At a properly selected relative pulse duration optical fields from every pulse inside the particle are in-phasely summed, and it becomes possible to excite selectively microparticle resonance modes, whose natural frequencies fall within the spectral range of radiation.

This paper proposes a further development of this technique from the viewpoint of efficiency of resonance excitation of WGMs by varying the relative pulse duration in a train in combination with linear frequency modulation of every pulse (chirping). Variation of the chirping depth leads to redistribution of spectral energy inside the radiation profile, and, thus, it becomes possible to concentrate energy near needed frequency ranges (natural WGM frequencies).

Basic equations

The basic idea of the proposed technique of frequency-pulsed modulated optical excitation of a spherical microcavity is explained qualitatively in Fig. 1. Figure shows three spectral profiles of the power density of laser radiation incident on a microcavity $|S_\omega(\omega)|^2$, which correspond to a single Gaussian (in time) pulse and a train of ten pulses of the same duration with and without liner frequency modulation applied to every pulse.

As is seen, in contrast to the smooth Gaussian profile of a single pulse, the spectral profile of a train is a function consisting of many localized spikes (lobes), the frequency distance between which is constant and determined by the ratio of the time gap between the pulses and the relative pulse duration. Then we assume that nonresonance excitation of a microparticle is realized: $\omega_0 \neq \omega_n$, where ω_0 and ω_n are, respectively, the frequencies of the incident radiation and a selected whispering gallery mode. Then if the pulse ratio in the train is selected so that the frequency of some side lobe of the spectrum ω_j coincides with the resonance frequency of the microparticle ω_n, then the light energy is transferred optimally in this mode compared to the case of a single pulse.

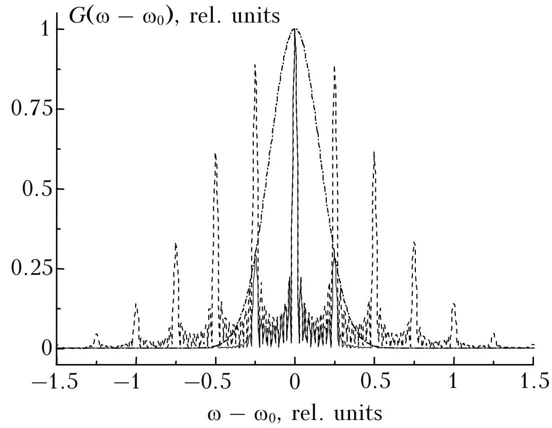

Fig. 1. Spectral profiles of a single Gaussian pulse (dot-and-dash curve) and a train of 10 pulses with (dashed curve) and without (solid curve) frequency modulation.

The efficiency of this process directly depends on the spectral energy contained in the side lobe of the spectrum of radiation exciting WGMs. For a nonmodulated train of pulses, as follows from Fig. 1, the ratio between peaks of spectral intensity at the central frequency $|S_\omega(\omega_0)|^2$ and at the frequency of, say, the first lobe $|S_\omega(\omega_1)|^2$ is rather large and characterized by the value of $\eta_1 = |S_\omega(\omega_0)|^2/|S_\omega(\omega_1)|^2 \simeq 3.8$. If we consider a train of chirped pulses with the same relative pulse duration, then this duration can be decreased significantly, and for the profiles shown in Fig. 1 it is already $\eta_1 \simeq 1.2$. Consequently, in this case, we can expect a smoother distribution of peaks of the radiation spectral energy and more efficient excitation of the microcavity.

Then we find the functional connection between the parameter η and the frequency–time characteristics of the radiation incident on the microparticle and, first of all, with the parameter of the chirping depth. For this purpose, we specify the time profile of every pulse in the train by the Gaussian profile with the duration t_p (at a level of e^{-1} of intensity maximum):

$$f_j(t) = \mathrm{e}^{-\frac{(t-t_j)^2}{2t_\text{p}^2}(1-ib)}\,\mathrm{e}^{i\omega_0 t},\ j = 1, \ldots, N_\text{p}.$$

The time dependence of the whole train of N_p equidistant pulses has the form

$$f(t) = \sum_{j=1}^{N_\text{p}} f_j(t). \qquad (1)$$

Here $t_j = t_0 + (j-1)T$, T is the pulse period; b is the depth parameter of the linear frequency modulation, and t_0 defines the position of the first pulse peak in the train on the time scale. The frequency of such radiation within every pulse varies in time by the law $\omega(t) = \omega_0 + bt/2t_\text{p}^2$.

The Fourier transform spectrum of dependence (1) describing the spectral profile of radiation is specified by the following function:

$$S_\omega(\omega) = \int_{-\infty}^{\infty} f(t)\mathrm{e}^{-i\omega t}\mathrm{d}t = S_{\omega 0}(\omega - \omega_0)\sum_{j=1}^{N_\text{p}} \mathrm{e}^{-i(\omega-\omega_0)t_j}, \qquad (2)$$

where

$$S_{\omega 0}(\omega - \omega_0) = \int_{-\infty}^{\infty} f_0(t)\mathrm{e}^{-i\omega t}\mathrm{d}t =$$

$$= \frac{(2\pi)^{3/2}\sqrt{1+ib}}{\omega_\text{p}}\exp\left[-\frac{4\pi^2(\omega-\omega_0)^2}{2\omega_\text{p}^2}\right]$$

is the envelope of the spectral profile of the whole train with the half-width $\omega_\text{p} = 2\pi\sqrt{1+b^2}/t_\text{p}$.

The equation for the spectral intensity of radiation follows from Eq. (2) and can be written as

$$\left|S_\omega(\omega)\right|^2 = \left|S_{\omega 0}(\omega-\omega_0)\right|^2 \frac{\sin^2(N_\text{p}K/2)}{\sin^2(K/2)}, \qquad (3)$$

where $K = (\omega - \omega_0)T = (\omega - \omega_0)s_\text{p}t_\text{p}$, the parameter $s_\text{p} = T/t_\text{p}$ defines the relative pulse duration. According to Eq. (3), the spectral profile of the train looks like a frequency-pulsating function and has main peaks arranged equidistantly on the frequency axis, which arise upon constructive summation of exponents in Eq. (2). The coordinates of these peaks can be found from the relation

$$\left|(\omega - \omega_0)\right| = \frac{2\pi m}{s_\text{p}t_\text{p}},\ m = 0, 1, 2, \ldots. \qquad (4)$$

Consequently, the sought ratio of the spectral intensity peaks for the first side lobe η_1 is determined as $\eta_1 = \exp\{-4\pi^2/[s_\text{p}^2(1+b^2)]\}$ and increases with the increase of the modulation depth b.

At the same time, the chirping of pulses has a negative effect as well, because at the general broadening of the spectrum the absolute value of the maximum intensity of the lobes decreases simultaneously. Actually, from Eq. (3) we obtain

$$\left|S_\omega(\omega_1)\right|^2 \sim \frac{1}{\sqrt{1+b^2}}\exp\left[-\frac{t_\text{p}^2(\omega_1-\omega_0)^2}{(1+b^2)}\right].$$

This function has a peak for the parameter $b_\text{m} = \sqrt{\left|2(\omega_1-\omega_0)^2 t_\text{p}^2 - 1\right|}$ value, which takes the following form when using condition (4) for the optimal pulse duration:

$$b_{\mathrm{m}} = \left|(2\pi/s_{\mathrm{p}}) - 1\right|^{1/2}. \qquad (5)$$

Thus, at $\omega = \omega_n$ Eq. (4) determines the rule, by which the relative pulse duration s_{p} is selected for the frequency position of a side peak of the radiation spectral intensity to coincide with the frequency of the selected resonance mode of the particle ω_n, and Eq. (5) gives the depth b of frequency modulation of the pulses, which is necessary for more efficient excitation of this mode.

Numerical example

Let us illustrate the results obtained with an example. For this purpose, we consider the laser radiation incidence in the form of a train of ten pulses ($N_{\mathrm{p}} = 10$) of picosecond duration ($t_{\mathrm{p}} = 1$ ps) with $\lambda_0 = 800$ nm on a transparent spherical water particle (complex refractive index $m_a = n_a + i\kappa_a = 1.33 - i \cdot 0$) with the radius $a_0 = 10$ μm.

The spatial intensity profile of the optical field inside such a particle $I(\mathbf{r}_b)$ can be calculated by the Mie theory taking into account the finite width of the radiation spectral profile.[6,8] It is characterized by the presence of two main peaks located near the irradiated and dark hemispheres. The efficiency of optical excitation is measured by the so-called factor of inhomogeneity of the light field $B(\mathbf{r}_b) = I(\mathbf{r}_b)/I_0$ (where I_0 is the peak intensity of the incident radiation) calculated for definiteness at the point of intensity peak in the rear hemisphere of the particle $\mathbf{r}_b$. For the selected computational conditions, this point corresponds to the spherical coordinates: $\mathbf{r}_b = (r, \theta, \varphi)|_b = (0.845;\ 0.0;\ 0.0)$.

The spectral profile of picosecond radiation is rather wide ($\omega_{\mathrm{p}} \simeq 6.3 \cdot 10^{12}$ Hz) and, in principle, it can encompass several resonance modes of the particle. This can be seen from Fig. 2, which shows the spectral response function of the particle in the form of the dependence of $B(\mathbf{r}_b)$ on the normalized frequency detuning $\Delta\bar{\omega} = (\omega - \omega_0)/\omega_0$ near the central frequency of radiation ω_0.

The letters in Figure indicate the positions of WGM resonance profiles excited in the spatial configurations of TE_n^m and TM_n^m fields, in which the numerical indices correspond to the number n and order m of a natural resonance. The higher the resonance number and the lower the resonance order, the narrower its spectral profile and the higher the resonance value of the intensity $B(\mathbf{r}_b)$ [Ref. 9].

It follows from Fig. 2 that at the chosen parameters of the numerical experiment, a single pulse excited the optical field of the particle nonresonantly, since no one natural resonance corresponds to the central frequency of the radiation ω_0 ($\Delta\bar{\omega} = 0$). At the same time, at least three resonance modes, namely, TE_{85}^3, TM_{90}^2, and TE_{81}^4, lie near the frequency center of the pulse and can be candidates for selective excitation. For definiteness, we select the tuning to the natural frequency of the TE_{85}^3 mode (shown by an arrow in Fig. 2), since it has a rather high Q-factor at a wide spectral profile, which facilitates its excitation.

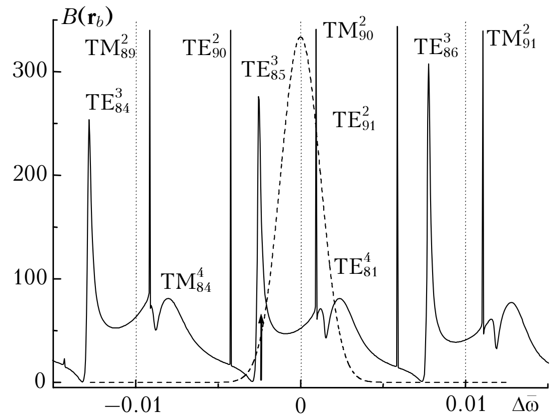

Fig. 2. Spectral response function $B(\mathbf{r}_b)$ of a water droplet with $a_0 = 10$ μm as a function of the normalized frequency detuning $\Delta\bar{\omega}$ for radiation with $\lambda_0 = 800$ nm. Positions of natural resonance modes of the particle and the spectral profile of a laser pulse (in relative units) with duration $t_{\mathrm{p}} = 1$ ps are shown.

First, following Eq. (4), we find the relative pulse duration in the train, at which the frequency of the first side peak of the radiation spectral intensity is equal to the natural frequency of the selected mode $\omega_{85,\,3}$. We obtain the following value of the pulse ratio:

$$s_{\mathrm{p}} = 2\pi \Big/ \left|(\omega_{85,3} - \omega_0)\right| t_{\mathrm{p}} \simeq 2.05.$$

Then we determine the optimal value of the chirping depth by Eq. (5) taking into account the relative pulse duration determined at the previous stage, which yields $b \simeq 4.6$. In this case, the maximal value of the factor $B(\mathbf{r}_b)$ (and, consequently, the optimal regime of excitation of selected WGM) is achieved during irradiation of the particle by a train of chirped pulses and is equal to 143, in contrast to 59.6 for a single pulse and 49.2 for continuum radiation.

Conclusions

In this paper, we have considered the problem of the most efficient excitation of resonance spatial configurations of the internal optical field (WGMs) of transparent spherical particles by laser radiation.

Our study has shown that the optimal transfer of energy of the incident radiation to a preset high-Q natural resonance of a particle can be performed with the use of frequency-pulsed irradiation in combination with the linear frequency modulation of every pulse in the train. Varying the relative pulse duration, it is possible to tune to the resonance, and varying the chirping depth, it is possible to concentrate energy in needed spectral ranges.

In micron-sized water droplets, this effect is most pronounced for radiation consisting of pulses of picosecond duration compared to radiation consisting of a train of femtosecond pulses. More than twice increase in the intensity of optical field of excited WGM has been obtained.

It is assumed that in larger particles, in which the density of natural oscillation modes per unit frequency range is high, in order to obtain the best conditions of cavity excitation, it is necessary to pass to subpicosecond frequency-modulated radiation, while the femtosecond range of pulse duration gives the best results for excitation of submicron particles.

Acknowledgements

This work was supported in part by the Combined Integration Project No. 3.13, RFBR Grant No. 06-05-64799, and the Lavrent'ev Grant of SB RAS.

References

1. F. Vollmer, D. Braun, A. Libchaber, M. Khoshima, I. Teraoka, and S. Arnold, Appl. Phys. Lett. **80**, No. 21, 4057–4059 (2002).
2. K. Etourneau and J. Marcou, Pure Appl. Opt. **6**, No. 6, 707–715 (1997).
3. V. Pokropivny, Int. J. of Nanotechnol. **1**, Nos. 1–2, 170–192 (2004).
4. J. Fu, S. Fhe, and S. Xiao, J. Phys. A **33**, No. 37, 7761–7771 (2000).
5. Yu.I. Voloshchenko, A.Sh. Dzhamalov, A.I. Koval', and A.T. Reutov, Izv. Vyssh. Uchebn. Zaved., Radioelektron. **34**, No. 7, 6–12 (1991).
6. A.A. Zemlyanov and Yu.E. Geints, Atmos. Oceanic Opt. **14**, No. 5, 316–325 (2001).
7. A.A. Zemlyanov, Yu.E. Geints, and D.V. Apeksimov, Atmos. Oceanic Opt. **16**, No. 12, 982–986 (2003).
8. K.S. Shifrin and I.G. Zolotov, Appl. Opt. **33**, No. 9, 7798–7804 (1994).
9. Yu.E. Geints, A.A. Zemlyanov, V.E. Zuev, A.M. Kabanov, and V.A. Pogodaev, *Nonlinear Optics of Atmospheric Aerosol* (SB RAS Publishing House, Novosibirsk, 1999), 260 pp.