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Generation of triphotons upon spontaneous parametric down-conversion in a resonator

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***Abstract*.** **The possibility of generating correlated three-photon states of light (triphotons) during spontaneous parametric down-conversion of light in a cubic medium in a resonator is analysed. It is shown that the number of photons per mode of the three-photon field is proportional to the square of the resonator finesse and the number of longitudinal resonator modes satisfying the triple resonance condition.**

Keywords: *spontaneous parametric down-conversion, nonclassical light, biphoton, triphoton, resonator.*

1. Introduction

The development of nonclassical light sources [1] is one of the main directions in evolution of modern quantum optics. In particular, of great interest is the generation of three-photon entangled states (triphotons), which can be used both for fundamental studies related to the verification of the fundamentals of quantum mechanics and as the main resource in many-sided quantum communication systems [2]. However, the most obvious method of generating three-photon states – the direct or cascade parametric process requires very high average pump powers or high nonlinearity coefficients. The authors of paper [3] proposed to generate three- and four-photon states due a random overlap of two pairs of photons upon two-photon parametric scattering in the case of pulse pumping. The pump pulse should be considerably shorter than the inverse width of the spectrum separated at the input to a measuring system. However, the rate of four-photon coincidences in this generation method is of the order of $10^{-2} - 10^{-1}$ s^{-1} (see experimental papers [4 – 8]), and therefore the search for analysis of other schemes for generating three-, four-photon, etc. states is still of current interest. A possible solution can be the generation of triphotons during spontaneous parametric down-conversion (SPDC) in a resonator. This process is well studied in the case of two-photon SPDC because we are dealing in fact with an optical parametric oscillator (OPO) operating considerably lower the threshold. It is known that the generation rate of biphotons in the resonator increases proportionally to the square of the resonator finesse and the width of the two-photon field spectrum can be reduced to the resonator passband width [9]. These properties can be used to develop efficient sources of two-photon [10, 11] and single-photon [12] light states with the spectral width comparable to the absorption linewidth in resonance media. The latter circumstance makes such sources especially attractive for recording and reproducing quantum states of light in quantum memory devices. However, in the case of three-photon SPDC, the generation rate of photons in a certain mode is proportional to the integrated brightness of vacuum fluctuations corresponding to all possible two-photon processes resulting in the creation of a third photon in this mode [13]. Therefore, the presence of a resonator should not only increase the rate of three-photon SPDC but also decrease it due to the narrowing of the spectrum of vacuum fluctuations producing the down conversion of pump photons. The aim of this paper is to elucidate how much the triphoton creation rate can be increased during SPDC in a resonator and to estimate the outlook for using this phenomenon for the development of nonclassical light sources.

2. Field state vector and intensity

Entangled (in polarisation) three-photon states can be obtained by using an OPO with two identical negative uniaxial crystals, which are characterised by the cubic nonlinearity $\chi^{(3)}$, are cut at the type I phase-matching angle and are oriented so that their optical axes are turned through 90° with respect to each other around the phase-matching axis (Fig. 1). The type I phase matching (eoo phase matching) means that the polarisation of the pump field in each crystal should correspond to that of the extraordinary wave, while polarisations of created photons should be the same as that of the ordinary wave. If the pump wave is polarised at 45° to optical axes, the creation of three photons in a crystal, in which the ordinary wave is, for example, polarised horizontally (H), and in the other crystal, where the ordinary wave is polarised vertically (V), occurs with the same probability amplitude. As a result, the entangled field state of the type $|\psi\rangle = \frac{1}{\sqrt{2}}(\mathrm{VVV} + \mathrm{HHH})$ appears. Such a scheme with quadratically nonlinear crystals, both without a resonator [14 – 16] and with it [10, 11], was successfully used to generate entangled two-photon states. Let us assume that the resonator is confocal, both its

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mirrors are transparent at the pump frequency and have a high reflectance at the frequencies of scattered waves. Thus, the OPO under study is a three-resonator one. We assume for simplicity that the angular divergence of the fundamental transverse resonator mode, determined by the waist size, coincides with the divergence of the SPDC radiation in each crystal in the absence of the resonator. In other words, the resonator does not produce the angular selection of modes. We also assume that a nonlinear medium occupies the entire volume of the resonator and the interaction region of the field modes is characterised by the Fresnel number, which is much greater than unity. Then, calculations can be performed in the standard approximation of an infinitely broad medium, which leads to the angular correction of photons created within one transverse resonator mode.

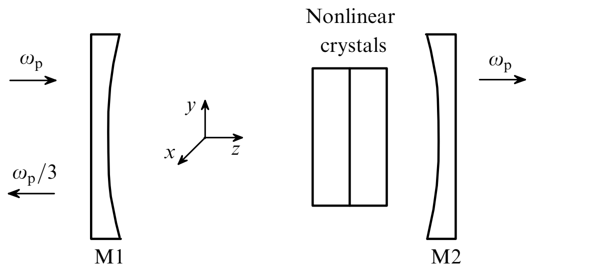

Figure 1. Scheme of an OPO: M1 and M2 are resonator mirrors; $\omega_{\rm p}$ is the pump radiation frequency. Triphotons are generated at frequencies close to $\omega_{\rm p}/3$ and come out through semitransparent mirror M1.

By considering spontaneous parametric scattering, it is reasonable to assume that the probability of the three-photon decay during the photon lifetime in the resonator is small (the pump level is considerably lower than the generation threshold). Then, the interaction Hamiltonian can be written in the form

$$\hat{H} = \varepsilon_0\chi^{(3)}\sum_{s={\rm H,V}}\int {\rm d}^3\boldsymbol{r}E_{\rm p}^{(+)}(\boldsymbol{r},t)\hat{E}_s^{(-)}(\boldsymbol{r},t) \times \hat{E}_s^{(-)}(\boldsymbol{r},t)\hat{E}_s^{(-)}(\boldsymbol{r},t) + {\rm H.c.}, \quad (1)$$

where ε_0 is the permittivity of vacuum; $E_{\rm p}^{(+)}(\boldsymbol{r},t) = E_0\exp[{\rm i}(\boldsymbol{k}_{\rm p}\boldsymbol{r} - \omega_{\rm p}t)]$ is the field of the classical pump wave with the amplitude E_0; $\omega_{\rm p}$ and $\boldsymbol{k}_{\rm p}$ are the pump frequency and wave vector;

$$\hat{E}_s^{(-)}(\boldsymbol{r},t) = -{\rm i}\int {\rm d}^3\boldsymbol{k}E(\boldsymbol{k})a_s^+(\boldsymbol{k})\exp\{-{\rm i}[\boldsymbol{kr} - \omega(\boldsymbol{k})t]\} \quad (2)$$

are the field operators corresponding to the modes of scattered light with the frequency ω and wave vector $\boldsymbol{k}$; $a_s^+(\boldsymbol{k})$ is the photon creation operator in a mode with frequency $\omega(\boldsymbol{k})$;

$$E(\boldsymbol{k}) = \frac{M(\boldsymbol{k})}{(2\pi)^{3/2}n(\boldsymbol{k})}\left[\frac{\hbar\omega(\boldsymbol{k})}{2\varepsilon_0}\right]^{1/2}; \quad |\boldsymbol{k}| = \frac{\omega(\boldsymbol{k})n(\boldsymbol{k})}{c}; \quad (3)$$

$n(\boldsymbol{k})$ is the refractive index of the medium for the ordinary wave; c is the speed of light in vacuum; the factors

$$M(\boldsymbol{k}) = \left\{\frac{c\gamma/[Ln_{\rm av}(\boldsymbol{k})]}{[\omega(\boldsymbol{k}) - \bar{\omega}(\boldsymbol{k})]^2 + \gamma^2}\right\}^{1/2} \quad (4)$$

take into account the relation between the field operators in the resonator with the field operators outside the resonator [17]; γ is the decay rate of the field amplitude in the resonator, which is assumed the same for all the modes; L is the distance between mirrors; $n_{\rm av}(\boldsymbol{k})$ is the average value of the refractive index for waves with the H and V polarisations in the presence of two crossed crystals of the same thickness; $\bar{\omega}(\boldsymbol{k})$ is the frequency of the longitudinal resonator mode, nearest to the mode frequency $\omega(\boldsymbol{k})$. Expression (3) is written in the approximation of the equal phase and group speeds of light in the medium [18], and expression (4) is valid for a resonator with the finesse much greater than unity, which is assumed below.

Taking (2) into account, Hamiltonian (1) takes the form

$$\hat{H} = {\rm i}\varepsilon_0\chi^{(3)}\sum_{s={\rm H,V}}\iiint {\rm d}^3\boldsymbol{k}'{\rm d}^3\boldsymbol{k}''{\rm d}^3\boldsymbol{k}'''E_0E(\boldsymbol{k}') \times E(\boldsymbol{k}'')E(\boldsymbol{k}''')a_s^+(\boldsymbol{k}')a_s^+(\boldsymbol{k}'')a_s^+(\boldsymbol{k}''') \times \int {\rm d}^3\boldsymbol{r}\exp[{\rm i}(\boldsymbol{k}_{\rm p} - \boldsymbol{k}' - \boldsymbol{k}'' - \boldsymbol{k}''')\boldsymbol{r}]$$
$$\times\exp\{{\rm i}[-\omega_{\rm p} + \omega(\boldsymbol{k}') + \omega(\boldsymbol{k}'') + \omega(\boldsymbol{k}''')]t\} + {\rm H.c.}, \quad (5)$$

and we obtain in the first order of the perturbation theory the field state vector

$$|\psi\rangle = |{\rm vac}\rangle - \frac{\rm i}{\hbar}\int_{-\infty}^{\infty}{\rm d}t\hat{H}|{\rm vac}\rangle = |{\rm vac}\rangle + \sum_{s={\rm H,V}}\iiint {\rm d}^3\boldsymbol{k}'{\rm d}^3\boldsymbol{k}''{\rm d}^3\boldsymbol{k}'''\mathscr{F}(\boldsymbol{k}',\boldsymbol{k}'',\boldsymbol{k}''') \times a_s^+(\boldsymbol{k}')a_s^+(\boldsymbol{k}'')a_s^+(\boldsymbol{k}''')|{\rm vac}\rangle, \quad (6)$$

where $|{\rm vac}\rangle$ is the vacuum state;

$$\mathscr{F}(\boldsymbol{k}',\boldsymbol{k}'',\boldsymbol{k}''') = \frac{\varepsilon_0\chi^{(3)}}{\hbar}E_0E(\boldsymbol{k}')E(\boldsymbol{k}'')E(\boldsymbol{k}''') \times V\varDelta(\boldsymbol{k}_{\rm p} - \boldsymbol{k}' - \boldsymbol{k}'' - \boldsymbol{k}''') \times 2\pi\delta[\omega(\boldsymbol{k}') + \omega(\boldsymbol{k}'') + \omega(\boldsymbol{k}''') - \omega_{\rm p}]; \quad (7)$$

$$\varDelta(\boldsymbol{k}) = \frac{1}{V}\int_V {\rm d}^3\boldsymbol{r}\,\exp({\rm i}\boldsymbol{kr});$$

and V is the medium volume.

Let us assume that the pump wave vector $\boldsymbol{k}_{\rm p}$ is directed along the z axis coinciding with the resonator axis and denote the longitudinal and transverse components of the wave vectors $\boldsymbol{k}'$, $\boldsymbol{k}''$ and $\boldsymbol{k}'''$ of the scattered light mode by $\boldsymbol{k}_z'$, $\boldsymbol{k}_z''$, $\boldsymbol{k}_z'''$ and $\boldsymbol{k}_\perp'$, $\boldsymbol{k}_\perp''$, $\boldsymbol{k}_\perp'''$, respectively. In the limit of an infinitely broad nonlinear medium (the Fresnel number for the mode interaction region is much greater than unity), we have

$$V\varDelta(\boldsymbol{k}) = \int_V {\rm d}^3\boldsymbol{r}\,\exp({\rm i}\boldsymbol{kr}) = L{\rm sinc}\left(\frac{k_zL}{2}\right)(2\pi)^2\delta(\boldsymbol{k}_\perp), \quad (8)$$

where ${\rm sinc}\,x = (\sin x)/x$. Thus, taking (8) into account, expression (7) contains two delta functions, one of which imposes the restriction on the frequencies: $\omega_{\rm p} = \omega(\boldsymbol{k}')+$

$\omega(\boldsymbol{k}'') + \omega(\boldsymbol{k}''')$ (frequency matching) and the other – on the transverse components of wave vectors: $\boldsymbol{k}'_\perp + \boldsymbol{k}''_\perp + \boldsymbol{k}'''_\perp = 0$ (phase matching). Taking into account the above assumptions, the expression for the state vector takes the form

$$|\psi\rangle = |\mathrm{vac}\rangle + \frac{\varepsilon_0 \chi^{(3)} (2\pi)^3 L E_0 n^{3/2}}{c^{3/2}\hbar}$$

$$\times \iint \mathrm{d}^3\boldsymbol{k}'_\perp \mathrm{d}^3\boldsymbol{k}''_\perp \iint d\omega' d\omega''\, E(\omega')E(\omega'')E(\omega_\mathrm{p} - \omega' - \omega'')$$

$$\times \operatorname{sinc}\left\{\left[k_\mathrm{p} - k'_z(\omega') - k''_z(\omega'') - k'''_z(\omega_\mathrm{p} - \omega' - \omega'')\right]\frac{L}{2}\right\}$$

$$\times \sum_{s=\mathrm{H,V}} a_s^+(\omega', \boldsymbol{k}'_\perp) a_s^+(\omega'', \boldsymbol{k}''_\perp)$$

$$\times a_s^+(\omega_\mathrm{p} - \omega' - \omega'', -\boldsymbol{k}'_\perp - \boldsymbol{k}''_\perp)|\mathrm{vac}\rangle, \qquad (9)$$

where $n = n(\omega_\mathrm{p}/3)$.

In the approximation of almost degenerate SPDC at frequencies close to $\omega_0 = \omega_\mathrm{p}/3$, it is convenient to make the change of variables $\omega' = \omega_0 + \Omega'$ and $\omega'' = \omega_0 + \Omega''$, at which $\omega''' = \omega_0 - \Omega' - \Omega''$ due to the frequency matching. Because $k_z \approx k - \frac{1}{2}k_\perp^2/k$ for small scattering angles, by expanding the modulus of the wave vectors into a series in powers of Ω, we obtain with the accuracy to the second order that

$$k_\mathrm{p} - k'_z - k''_z - k'''_z = \frac{1}{2}\frac{{k'_\perp}^2 + {k''_\perp}^2 + (\boldsymbol{k}'_\perp + \boldsymbol{k}''_\perp)^2}{k_0}$$

$$- \frac{D''}{2}[{\Omega'}^2 + {\Omega''}^2 + (\Omega' + \Omega'')^2], \qquad (10)$$

where $k_0 = k(\omega_0)$,

$$D'' = \left.\frac{\mathrm{d}^2 k'}{\mathrm{d}\omega^2}\right|_{\omega=\omega_0} = \left.\frac{\mathrm{d}^2 k''}{\mathrm{d}\omega^2}\right|_{\omega=\omega_0}$$

and it is taken into account that $k_\mathrm{p} = 3k_0$ if the pump wave propagates at the phase-matching angle to the optical axis of the crystal. As a result, the expression for the state vector takes the form

$$|\psi\rangle = |\mathrm{vac}\rangle + \frac{\varepsilon_0 \chi^{(3)} (2\pi)^3 L E_0 n^{3/2}}{c^{3/2}\hbar} \iint \mathrm{d}^3\boldsymbol{k}'_\perp \mathrm{d}^3\boldsymbol{k}''_\perp$$

$$\times \iint \mathrm{d}\Omega' \mathrm{d}\Omega'' E(\omega_0 + \Omega')E(\omega_0 + \Omega'')E(\omega_0 - \Omega' - \Omega'')$$

$$\times \operatorname{sinc}\left[\left\{\frac{1}{2}\frac{{k'_\perp}^2 + {k''_\perp}^2 + (\boldsymbol{k}'_\perp + \boldsymbol{k}''_\perp)^2}{k_0}\right.\right.$$

$$\left.\left.- \frac{D''}{2}[{\Omega'}^2 + {\Omega''}^2 + (\Omega' + \Omega'')^2]\right\}\frac{L}{2}\right]$$

$$\times \sum_{s=\mathrm{H,V}} a_s^+(\omega_0 + \Omega', \boldsymbol{k}'_\perp) a_s^+(\omega_0 + \Omega'', \boldsymbol{k}''_\perp)$$

$$\times a_s^+(\omega_0 - \Omega' - \Omega'', -\boldsymbol{k}'_\perp - \boldsymbol{k}''_\perp)|\mathrm{vac}\rangle. \qquad (11)$$

By using this expression, we find the average number of photons

$$N(\omega_0 + \Omega, \boldsymbol{k}_\perp) = \langle\psi| a_s^+(\omega_0 + \Omega, \boldsymbol{k}_\perp) a_s(\omega_0 + \Omega, \boldsymbol{k}_\perp)|\psi$$

(where a_s is the annihilation operator of a photon in a mode) generated in a field mode, which is characterised by the transverse wave vector $\boldsymbol{k}_\perp$ and the detuning frequency $\Omega \equiv \omega - \omega_0$:

$$N(\omega_0 + \Omega, \boldsymbol{k}_\perp) = \left[\frac{\varepsilon_0 \chi^{(3)} (2\pi)^3 L E_0}{c^{3/2}\hbar}\right]^2 \frac{n^{-3}}{(2\pi)^9}\int \mathrm{d}^3\boldsymbol{k}'_\perp$$

$$\times \int \mathrm{d}\Omega'\, \frac{\hbar(\omega_0 + \Omega')}{2\varepsilon_0}\frac{\hbar(\omega_0 + \Omega)}{2\varepsilon_0}\frac{\hbar(\omega_0 - \Omega' - \Omega)}{2\varepsilon_0}$$

$$\times \left\{\frac{c\gamma/[L n_\mathrm{av}(\omega_0)]}{f^2(\omega_0 + \Omega') + \gamma^2}\right\}\left\{\frac{c\gamma/[L n_\mathrm{av}(\omega_0)]}{f^2(\omega_0 + \Omega) + \gamma^2}\right\}$$

$$\times \left\{\frac{c\gamma/[L n_\mathrm{av}(\omega_0)]}{f^2(\omega_0 - \Omega' - \Omega) + \gamma^2}\right\}$$

$$\times \operatorname{sinc}^2\left[\left\{\frac{1}{2}\left[\frac{{k'_\perp}^2 + k_\perp^2 + (\boldsymbol{k}'_\perp + \boldsymbol{k}_\perp)^2}{k_0}\right]\right.\right.$$

$$\left.\left.- \frac{D''}{2}[{\Omega'}^2 + \Omega^2 + (\Omega' + \Omega)^2]\right\}\frac{L}{2}\right], \qquad (12)$$

where $f(\omega) = \omega - \bar{\omega}(\omega)$; $\bar{\omega}(\omega)$ is the frequency of the longitudinal resonator mode nearest to the ω. It is clear that only the filed modes for which the relation $|f(\omega_0 + \Omega')|$, $|f(\omega_0 + \Omega)|$, $|f(\omega_0 - \Omega' - \Omega)| < \gamma$ is fulfilled will make the contribution to the integral over frequency. Because the OPO frequency spectrum is non-equidistant due to the refractive index dispersion, the above relation is fulfilled only for some longitudinal modes forming groups (clusters) with close frequencies. As a result, the SPDC spectrum also has the cluster structure, which is typical for multiresonator OPOs (see, for example, [19]). In the case of a high finesse, which is considered here, the triple resonance is possible only within one cluster with the central frequency ω_0 (Fig. 2).

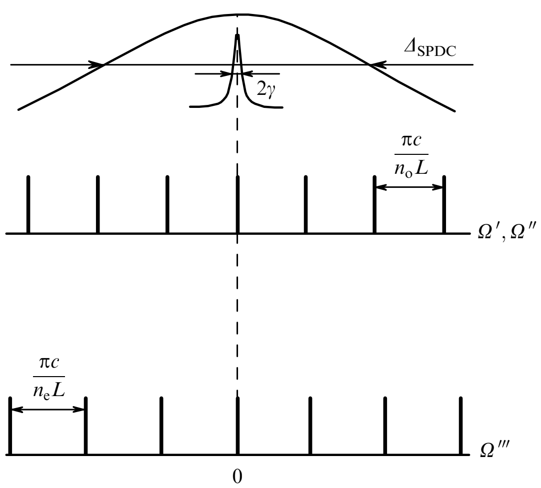

Figure 2. Typical relations between the width Δ_SPDC of the SPDC spectrum in the scheme without a resonator, the width 2γ of the resonator mode and frequencies of the longitudinal modes of a confocal resonator in the case of the frequency-degenerate matching $\Omega' = \Omega'' = \Omega''' = 0$; $\pi c/(n_{\mathrm{e,o}}L)$ are intermode intervals.

The maximum value $N(\omega_0+\Omega, \boldsymbol{k}_\perp)$ is achieved in the degenerate collinear regime, when $\Omega = 0$ and $\boldsymbol{k}_\perp = 0$. In this case, expression (12) can be written in the form

$$N(\omega_0, 0) \approx \left[\frac{\varepsilon_0 \chi^{(3)} L E_0}{\hbar}\right]^2 \left(\frac{\hbar\omega_0}{4\pi\varepsilon_0 c}\right)^3 \left(\frac{2F}{\pi n}\right)^3 \Delta\Omega S_{\boldsymbol{k}_\perp} m, \qquad (13)$$

where $F = \pi c/[\Delta\Omega L n_{\mathrm{av}}(\omega_0)]$ is the resonator finesse; $\Delta\Omega = 2\gamma$ is the half-width of the spectral line of the resonator; $S_{\boldsymbol{k}_\perp} = 0.866\pi^2 k_0/L$ is the area of the region in the plane of wave vectors perpendicular to $\boldsymbol{k}_\mathrm{p}$ within which the function sinc x considerably differs from zero; and m is the number of longitudinal resonator modes contributing to SPDC. Because $\Delta\Omega \sim F^{-1}$, we obtain that $N(\omega_0, 0) \sim F^2 m$.

Let us estimate the generation rate of triphotons upon SPDC in the resonator. By assuming that the absorption coefficient α of the crystal is 0.01 cm^{-1}, the resonator length is $L = 5$ mm, and the reflectance of resonator mirrors is 99.9 %, we obtain the resonator finesse $F = 300$. Based on the estimates made in [9], the number m of longitudinal modes for such values of F and L should be set equal to 30. Then, by using the typical values of quantities entering (13), namely, $\chi^{(3)} = 10^{-21}$ m^2 V^{-2}, $\lambda = 900$ nm, $n_{\mathrm{av}} = n = 1.5$, and the pump radiation intensity 0.1 MW cm^{-2}, we obtain $N \sim 10^{-15}$. The photon counting rate w can be found from the relation [20]

$$w = N\frac{\eta c}{\lambda^4}\Delta\Theta\Delta\lambda A, \qquad (14)$$

where η is the quantum efficiency of a detector; $\Delta\lambda$, $\Delta\Theta$ and A are the passband width, angular aperture, and area of the detector, respectively. By assuming that $\eta = 0.5$, $\Delta\lambda =$ 0.005 nm, $\Delta\Theta \approx 10^{-4}$ sr (the divergence angle of the beam in the case under study is 1°, which corresponds to the waist diameter of 60 μm), and $A = 10^{-2}$ cm^2, we obtain $w \approx 10^{-3}$ s^{-1}. Thus, the triphoton creation rate in the resonator is an order of magnitude lower than in experiments mentioned in section 1. Note, however, that in this case we consider photons within the spectral band of width ~ 2 GHz, which is three – four orders of magnitude narrower than a spectral band obtained upon usual picosecond SPDC in the scheme without a resonator.

3. Conclusions

The use of SPDC in a resonator is promising for generating both single and correlated photons within the spectral band of width of the order of the resonator spectrum width. The long lifetime of photons in the resonator (up to tens of nanoseconds) allows the use of nanosecond pump pulses instead of femtosecond pulses, which can substantially simplify experiments requiring the time synchronisation of many photons. However, unlike two-photon SPDC, the number of photons per mode in the case of three-photon SPDC proves to be proportional to the number of longitudinal resonator modes satisfying the triple (or double) resonance condition. As a result, the triphoton creation rates required to realise the corresponding experiment can be obtained only under the condition that the resonator finesse increases simultaneously with increasing the frequency interval between longitudinal modes, i.e. with decreasing the resonator length. The efficiency of three-photon SPDC in the resonator can be further increased by using a system of identical matched resonators, when the total length of the medium considerably exceeds the length of each of the resonators.

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