γ -ray-pulse formation in a vibrating recoilless resonant absorber

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We study propagation of γ radiation from a Mössbauer radioactive source through a vibrating recoilless resonant absorber and find the optimal conditions to produce a periodic train of γ -ray pulses with maximum peak intensity, several times higher than the intensity from the source, and minimum duration, much shorter than the lifetime of the emitting nuclear state of the source. The shape, duration, and repetition rate of the pulses are tunable in a wide range. We propose modifications of the recently reported experiment [F. Vagizov *et al.*, Nature (London) **508**, 80 (2014)] to produce pulses with higher peak intensity and shorter duration using absorbers enriched by the resonant nuclei and discuss possible applications of the generated pulses for the time-domain Mössbauer spectroscopy.

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I. INTRODUCTION

The generation of the ultrashort hard-x-ray and γ -ray pulses draws persistent attention due to the important diagnostic applications in material science as well as in tracing and steering of the ultrafast chemical and biological processes. The extremely short wavelength of the x rays and γ radiation potentially enables one to investigate the material structure on atomic and subatomic scales. The time-resolved x-ray– Mössbauer spectroscopy and dynamical x-ray diffraction based on the resonant scattering of x-ray and γ -ray pulses allow extracting rich information about the properties of the nuclear levels, local fields, phonon spectra, and transient structures [1–4].

Picosecond x-ray pulses of synchrotron radiation or femtosecond pulses of sliced synchrotron radiation and of x-ray free-electron lasers (XFELs) are typically used in such studies, which limits this research to a few existing facilities in the world [5–7]. Besides, these sources produce extremely broadband radiation and the x-ray pulses are far from being transform limited, which restricts their spectroscopic applications. The sources of coherent transform-limited ultrashort pulses, such as self-seeded XFELs [8] and high-order-harmonicgeneration sources [9], in the (1–10)-keV range have just started to emerge.

The formation of the ultrashort pulses of high-energy radiation plays an important role in the extension of the concepts of coherent optics into the x-ray and γ -ray frequency ranges. The active search for methods to control the interactions of the γ radiation with nuclear transitions has a rather long history (see Refs. [1–4,10–15]), including some important experimental results such as the observation of the ac Zeeman effect [16], the so-called γ -echo effect [17,18], and magnetic switching of nuclear forward scattering [19–23]. This field has been rapidly developing recently. It includes pioneering experimental demonstrations of the collective Lamb shift [24], cavity electromagnetically induced transparency [25], and vacuum-assisted generation of coherences [26] as well as a number of interesting theoretical ideas related to the field of γ -ray quantum optics such as the possibility of single-photon entanglement via magnetic switching [27], coherent control of branching ratios [28], modulation-induced transparency [29], coherent control of inhomogeneous broadening of nuclear transitions [30], single-photon coherent storage [31], and stimulated Raman adiabatic passage [32]. All these and many other recent developments (see the recent review in [33] and references therein) clearly indicate that γ -ray quantum optics is a fast-growing area of research.

There have been several attempts to produce γ -ray pulses via compression of frequency-modulated radiation from a Mössbauer radioactive source due to the resonant or nonresonant interaction with a Mössbauer absorber [10,17,18,20–22,34–39]. In Refs. [10,17,18,34,35,37–39] the resonant interaction of δ -function-like frequency-modulated (or, equivalently, stepwise phase-modulated) incident radiation with a Mössbauer absorber was considered. The δ -functionlike frequency modulation can be theoretically achieved via rapid acceleration and deceleration of the radiating Mössbauer source (which is sequentially kicked and stopped). However, in an experiment, a kicked source fixed on a piezoelectric plate tends to oscillate, so the δ -function-like frequency modulation is extremely hard to achieve. According to the estimate presented in [10], the time of motion of the Mössbauer absorber should not exceed $1/100\Gamma$, where $1/\Gamma$ is the lifetime of the excited nuclear state of the source. Consequently, this approach is applicable to the Mössbauer sources with especially long lifetimes, such as ⁶⁷Ga [34,10], and rather limited for other sources, such as ⁵⁷Co [17,18,35,37,39]. Another approach was proposed in [36], where the possibility of producing a train of Mössbauer γ -ray pulses much shorter than the lifetime of the excited state of radiating nucleus was shown theoretically via compression of Mössbauer radiation with harmonic frequency modulation, emitted by an oscillating source, in a far-off-resonance Mössbauer absorber. However, because of strong nonresonant losses, typical for Mössbauer

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absorbers, the intensity of the output radiation appears to be much smaller than the incident radiation intensity since most of the γ quanta are lost in the medium. The problem of γ -ray pulse formation was also considered in Refs. [20–22], which are devoted to the generation of γ -ray pulses on the basis of an abrupt change of energy of nuclear excitation due to magnetization reversal of a ⁵⁷FeBO₃ absorbing crystal. This method has experimentally allowed for the formation of isolated pulses of approximately 10-ns duration from polarized 14.4-keV Mössbauer γ radiation of modest intensity, limited by considerable nonresonant losses in an optically thick absorber and polarizer.

Recently, transformation of 14.4-keV photons spontaneously emitted by a radioactive ⁵⁷Co source into a periodic sequence of pulses of duration shorter than the lifetime of the excited state of the nuclei was realized [40] via radiation passage through a harmonically vibrating foil containing resonantly absorbing nuclei of ⁵⁷Fe. The ability to control the shape, duration, and repetition period of the produced pulses was shown both experimentally and theoretically. A uniquely large ratio of the resonance energy to the resonance bandwidth of the Mössbauer transitions (3×10^{12}) for the 14.4-keV transition of ⁵⁷Fe) makes effective use of the Doppler effect in the photon-nuclei interaction for generation of the wellseparated phase-locked γ -ray sidebands and formation of the γ -pulse train. The technique proposed in [40] is experimentally feasible due to the combination of the advantages of the methods in Refs. [34,36], namely, (i) the resonant interaction of γ radiation with a Mössbauer absorber and (ii) the harmonic motion of the absorber, respectively. The same technique in combination with a time-delayed coincidence measurement has allowed us to demonstrate the possibility of controlling the waveforms of single γ photons [40], which is an important contribution to the fast developing quantum γ optics and its applications in quantum information processing [24-26,31-33].

In the present paper we investigate the ultimate possibility for γ -ray pulse formation from radiation of a Mössbauer radioactive source in a single vibrating resonant absorber under readily available experimental conditions. We find the optimal conditions to produce a periodic train of pulses with maximum peak intensity, exceeding several times the intensity of the incident radiation, and the maximum ratio of the peak to the average output γ -ray intensity. We show that the total efficiency, defined as the ratio between the average intensity of the incident Mössbauer radiation (the average number of γ quanta detected per unit time) and the average intensity of the output radiation, i.e., a pulse train, under optimal conditions can be as high as 70%–80%for the realistic parameters of a Mössbauer absorber. We discuss the possibility of controlling the shape, duration, and peak intensity of the pulses via variation of parameters of the vibrating absorber and propose modifications of the experiment [40] to produce pulses with higher peak intensity, shorter duration, and better shape using absorbers with larger optical thickness (achieved via increasing concentration of the resonant nuclei). In general, the possibility of producing pulses with higher intensity and shorter duration originates from interference of the incident and the coherently scattered waves and intensification of the coherently scattered wave with increasing optical thickness of the resonant absorber (see [38]

and references therein). As is well known, in the absence of vibrations the interference between the incident and coherently scattered fields leads to a speedup effect and dynamical beats in nuclear forward scattering [12,13,41]. Oscillation of the absorber alters the coherently scattered field via generation of the vibrational sidebands and dramatically changes the character of interference. Under some optimal conditions (corresponding to the phase matching of all the generated spectral components) it provides constructive interference of the resonantly scattered and incident radiation fields within the short intervals of the vibration cycle and thus allows pulses to be produced with a peak intensity exceeding the intensity of the incident field. At the same time, during the rest of the vibration cycle the interference is destructive, leading to a reduction of the time-averaged output intensity with respect to the incident one [42]. The character of interference between the incident and coherently scattered fields can be altered not only by vibration but also by other techniques, such as the instantaneous shift of the emitter relative to the resonant absorber or vice versa [17,18,35,37-39], as well as by magnetization reversal [20–23], also resulting in γ -ray pulse formation under some optimal conditions.

An increase in the optical thickness of the absorber leads to an increase in amplitude of the resonantly scattered radiation and hence both an increasing peak intensity of the produced pulses and a decreasing time-averaged intensity of the output radiation. In this paper we show that the use of the incident radiation, detuned properly from the absorber resonance, allows pulses to be produced with higher peak intensity without considerable resonant attenuation in optically thick Mössbauer absorbers.

The paper is organized as follows. In the next section we present the theoretical model. In Sec. III we give the numerical optimization results, accompanied by estimates of the experimental parameter values. We conclude in Sec. IV with a discussion of the capabilities and limitations of the method and its possible applications in the area of timeresolved x-ray Mössbauer spectroscopy.

II. THEORETICAL MODEL

The transformation of Mössbauer γ radiation in a vibrating resonant recoilless absorber was studied in Refs. [40,43–50]. Except for [40], previous works focused on the spectral rather than temporal properties of the transmitted γ radiation. A similar problem was considered in Refs. [10,34,35,51,52], which were devoted to the experimental and theoretical study of the time dependence of the intensity of Mössbauer radiation, emitted by a vibrating source and propagated through the resonant absorber at rest. It was observed that the output γ radiation acquires an amplitude modulation that is highly sensitive to small energy shifts and can be used for their measurement. A quite general theory applicable to absorbers of arbitrary optical depth was derived in [52]. However, the authors analyzed only the case of a thin resonant absorber. As a result, they predicted and observed only an amplitude modulation of a small depth and did not discuss the possibility of γ -ray pulse formation. Our model is closer to that presented in Refs. [45]. However, we focus on modifications of the temporal properties of γ radiation, propagated through an

optically thick vibrating absorber, and consider the more general case, assuming that the linewidth of the resonant transition of the absorber can differ from the transition linewidth of the radioactive source.

Radiation of the source is considered as a flow of photons emitted at random times. The electric field of a single photon within the semiclassical approach can be represented as a quasimonochromatic wave propagating along the coordinate z_l in the laboratory frame of reference [12,13]

$$E_{\rm inc}(z_l,t) = E_0 \theta((t-t_0) - (z_l - z_0)/c) \\ \times e^{-(i\omega_s + \Gamma_s/2)[(t-t_0) - (z_l - z_0)/c] + i\phi_0} + {\rm c.c.}, \quad (1)$$

where we assume that the Mössbauer source has an unsplit transition line with the angular frequency ω_s and the excitedstate lifetime Γ_s^{-1} ; t_0 is the instant of formation of the excited state of the radiating nucleus, z_0 is the coordinate of the radiating nucleus, c is the speed of light in vacuum, φ_0 is the random phase, $\theta(x)$ is the unit step function, and c.c. stands for the complex conjugate.

The resonant absorber oscillates as a whole along the direction of propagation of γ photons with amplitude *R*, angular frequency Ω , and the initial phase of oscillation ϑ_0 . The coordinate in the laboratory reference frame z_l is related to the coordinate in the reference frame of the oscillating absorber z_a as

$$z_l = z_a + R\sin(\Omega t + \vartheta_0). \tag{2}$$

This equation assumes that the absorber thickness h is much smaller than the wavelength of sound

$$h \ll 2\pi V_s / \Omega, \tag{3}$$

where V_s is the speed of sound in the absorber. The latter condition is typically satisfied in the experiments dealing with oscillating Mössbauer sources or absorbers.

Let us consider propagation of the γ photon (1) in the reference frame associated with the vibrating absorber. Transformation from the laboratory to the absorber's frame of reference is accomplished via Eq. (2). Since the speed of vibration is much slower than the speed of light $R\Omega/c \ll 1$, time in the reference frame of the oscillating absorber t_a is the same as in the laboratory reference frame t_l , $t_a = t_l = t$.

In the reference frame of the oscillating absorber, the electric-field strength of the incident γ photon takes the form

$$E_{\rm inc}(z_a,t) = E_0 \theta((t-t_0) - [z_a + R\sin(\Omega t + \vartheta_0) - z_0]/c) \\ \times \exp(-(i\omega_s + \Gamma_s/2)\{(t-t_0) - [z_a + R\sin(\Omega t + \vartheta_0) - z_0]/c\} + i\phi_0) + {\rm c.c.}$$
(4)

Neglecting modulation of the decay as compared to modulation of the carrier frequency (which is justified for Mössbauer radiation since $\omega_s \gg \Gamma_s/2$) and taking into account that $\exp[ikR\sin(\Omega t + \vartheta_0)] =$ $\sum_{n=-\infty}^{+\infty} J_n(kR) \exp[in(\Omega t + \vartheta_0)]$, where $J_n(kR)$ is Bessel function of the first kind of order *n*, one obtains

$$E_{\rm inc}(z_a,t) = E_0\theta((t-t_0) - (z_a - z_0)/c) \times e^{-(i\omega_s + \Gamma_s/2)[(t-t_0) - (z_a - z_0)/c] + i\phi_0} \times \sum_{n=-\infty}^{+\infty} J_n(kR) \exp[in(\Omega t + \vartheta_0)] + {\rm c.c.}, \quad (5)$$

where $k = \omega_s/c$ is the wave number of the γ photon in vacuum. At the front edge of the absorber the incident radiation has the form

$$E_{\rm inc}(z_a = 0, t) = \exp\{-i\omega_s((t - t_0) + z_0/c) + i\phi_0\}$$
$$\times \sum_{n = -\infty}^{+\infty} \exp\{in(\Omega t + \vartheta_0)\}$$
$$\times \int_{-\infty}^{+\infty} E_n^{\rm inc}(\omega) \exp(-i\omega t)d\omega + {\rm c.c.}, \qquad (6)$$

where

$$E_n^{\rm inc}(\omega) = \frac{E_0}{2\pi} J_n(kR) \frac{\exp[i\omega(t_0 - z_0/c)]}{\Gamma_s/2 - i\omega}.$$
 (7)

Propagation of γ radiation through the Mössbauer absorber is described by the wave equation

$$\frac{\partial^2 E}{\partial z_a^2} - \frac{1}{c'^2} \frac{\partial^2 E}{\partial t^2} = \frac{2\delta_e}{c'} \frac{\partial E}{\partial t} + \frac{4\pi}{\varepsilon c'^2} \frac{\partial^2 P}{\partial t^2},\tag{8}$$

where *P* is the resonant polarization, which arises due to interaction with the nuclei, $c' = c/\sqrt{\varepsilon}$, and $\varepsilon \approx 1$ is the nonresonant dielectric permitivity of the absorber for γ radiation. The nonresonant absorption, primarily caused by interaction with electrons, is taken into acount via the linear photoelectric absorption coefficient $2\delta_e$.

The resonant polarization of the Mössbauer absorber with an unsplit transition line, induced by the incident radiation, has the form

$$\vec{P} = f_a N \cdot \vec{d}_{12} \rho_{21} + \text{c.c.},$$
 (9)

where f_a is the recoilless absorption probability (the Lamb-Mössbauer factor), N is concentration of the resonant nuclei, \vec{d}_{21} is the dipole moment of the resonant transition, and ρ_{21} is the nuclear coherence (the nondiagonal density-matrix element of the nuclei). The nuclear response is adequately described by Bloch-like equations (see, e.g., [10]), which for the coherence ρ_{21} take the form

$$\frac{d\rho_{21}}{dt} + i(\omega_a - i\gamma_a)\rho_{21} = \frac{i}{\hbar}n_{12}(\vec{d}_{21}\cdot\vec{E}), \qquad (10)$$

where ω_a is the angular frequency of the absorber transition, γ_a is the decay rate of the resonant coherence, and $n_{12} = \rho_{11} - \rho_{22}$ is the population difference between the resonant energy levels. We will further consider the isotropic medium, where $(\vec{d}_{21} \cdot \vec{E})$ can be replaced by $d_{21}E$, d_{21} being the root mean square of the dipole moment matrix element. Since the intensity of Mössbauer radiation is quite small compared to the saturation value, the population difference n_{12} between the lower and upper levels of the resonant transition remains unperturbed: $n_{12} \cong n_{12}^0 \cong 1$, where n_{12}^0 is the population difference in equilibrium.

Since reflection of γ radiation from the edges of Mössbauer absorber is negligible, we seek the solution of Eqs. (8)–(10)

inside the medium $0 \leq z_a \leq h$ in the form

$$E(z_a, t) = e^{-i\omega_s[(t-t_0) - (z_a - z_0)/\epsilon'] + i\phi_0} \sum_{n = -\infty}^{+\infty} \exp[in(\Omega t + \vartheta_0)]$$
$$\times \int_{-\infty}^{+\infty} E_n(z_a, \omega) \exp(-i\omega t) d\omega + \text{c.c.}, \qquad (11)$$

$$P(z_a,t) = e^{-i\omega_s[(t-t_0)-(z_a-z_0)/c']+i\phi_0} \sum_{n=-\infty}^{+\infty} \exp[in(\Omega t + \vartheta_0)]$$

$$\times \int_{-\infty}^{+\infty} P_n(z_a,\omega) \exp(-i\omega t) d\omega + \text{c.c.}$$
(12)

In the rotating-wave approximation $|\omega - \omega_a| \ll \omega + \omega_a$, from Eqs. (9) and (10) one finds

$$P_n(z_a,\omega) = \frac{n_{12}^0 f_a N |d_{12}|^2}{\hbar(\omega_a - \omega_s + n\Omega - \omega - i\gamma_a)} E_n(z_a,\omega).$$
(13)

In the additional approximation of slowly varying envelopes, which implies (i) $N_{\text{max}}\Omega \ll \omega_s$, where N_{max} is the number of spectral components of the radiation (11) with the appreciably nonzero amplitudes [for the incident radiation (5) and (6), $N_{\text{max}} \approx 2kR + 1$], and (ii) $|\partial E_n(z_a,\omega)/\partial z_a| \ll k|E_n(z_a,\omega)|$, the wave equation (8) takes the form

$$\frac{\partial E_n(z_a,\omega)}{\partial z_a} + \left(\delta_e + \frac{2\pi n_{12}^0 f_a N |d_{12}|^2}{\sqrt{\varepsilon} \hbar [\gamma_a + i(\omega_a - \omega_s + n\Omega - \omega)]} \frac{\omega}{c}\right) \times E_n(z_a,\omega) = 0.$$
(14)

Equation (14) takes into account that the absorber thickness is much smaller than both the decay length of the photon $h \ll c/\Gamma_s$ and the smallest beat length between its spectral components $h \ll c/N_{\text{max}}\Omega$. The last inequality is satisfied if the condition (3) is fulfilled.

The solution of Eq. (14), which satisfies the boundary condition $E_n(z_a = 0, \omega) = E_n^{\text{inc}}(\omega)$, is

$$E_n(z_a,\omega) = E_n^{\text{inc}}(\omega) \exp(-\delta_e z_a) \exp[-g_n(\omega) z_a], \quad (15)$$

where $g_n(\omega)$ are the complex coefficients of resonant absorption

$$g_n(\omega) = \frac{2\pi n_{12}^0 f_a N |d_{12}|^2}{\sqrt{\varepsilon} \hbar [\gamma_a + i(\omega_a - \omega_s + n\Omega - \omega)]} \frac{\omega}{c}.$$
 (16)

Thus, in the reference frame of the oscillating absorber, γ radiation inside the absorber has the form

$$E(z_a,t) = e^{-i\omega_s[(t-t_0)-(z_a-z_0)/c']+i\phi_0} \sum_{n=-\infty}^{+\infty} \exp[in(\Omega t + \vartheta_0)]$$
$$\times e^{-\delta_c z_a} \int_{-\infty}^{+\infty} E_n^{\rm inc}(\omega) \exp[-g_n(\omega) z_a]$$
$$\times \exp(-i\omega t)d\omega + \rm c.c.$$
(17)

Consequently, the output radiation in the frame of reference of the oscillating absorber satisfying the boundary condition $E_n^{\text{out}}(\omega) = E_n(z_a = h, \omega)$ has the form

$$E_{\text{out}}(z_a, t) = e^{-i\omega_s[(t-t_0)-(z_a-z_0)/c]+i\phi_0'} \sum_{n=-\infty}^{+\infty} \exp[in(\Omega t + \vartheta_0)] \times \int_{-\infty}^{+\infty} E_n^{\text{out}}(\omega) \exp(-i\omega t)d\omega + \text{c.c.}, \quad (18)$$

$$E_n^{\text{out}}(\omega) = \frac{E_0}{2\pi} \exp(-T_e/2)$$

$$\times J_n(kR) \exp\left(-\frac{\gamma_a T_a/2}{\gamma_a + i(\omega_a - \omega_s + n\Omega - \omega)}\right)$$

$$\times \frac{\exp[i\omega(t_0 - z_0/c)]}{\Gamma_s/2 - i\omega},$$
(19)

where $\phi'_0 = \phi_0 + \omega_s(\sqrt{\varepsilon} - 1)h/c$ is the modified radiation phase, $T_a = \frac{4\pi n_{12}^2 f_a N |d_{21}|^2}{\sqrt{\varepsilon} \hbar y_a} \frac{\omega_a}{c}h$ is the resonant optical (Mössbauer) thickness of the absorber, and $T_e = 2\delta_e h$ is the exponent of photoelectric absorption.

The backward transformation from the reference frame of the oscillating absorber to the laboratory reference frame is accomplished via Eq. (2), $z_a = z_l - R \sin(\Omega t + \vartheta_0)$, and simply leads to multiplication of the field (18) by the factor $\exp[-ikR\sin(\Omega t + \vartheta_0)]$. One should note that the different spectral components of radiation possess different wave numbers and while it is possible to neglect this difference inside the absorber, where the thickness is limited by the inequality $h \ll c/N_{\text{max}}\Omega$, one cannot do it for an arbitrarily long propagation length in vacuum. Thus, in the laboratory reference frame one obtains

$$E_{\text{out}}(z_l,t) = e^{-i\omega_s(\xi-\xi_0)+i\phi_0'} \sum_{m=-\infty}^{+\infty} \sum_{n=-\infty}^{+\infty} J_m(kR) \exp[i(n-m)(\Omega\xi+\vartheta_0)] \int_{-\infty}^{+\infty} E_n^{\text{out}}(\omega,\xi_0) \exp(-i\omega\xi)d\omega + \text{c.c.}, \quad (20)$$

where $\xi = t - z_l/c$ is the local time and $\xi_0 = t_0 - z_0/c$. The intensity of the output single-photon radiation $I = \frac{c}{4\pi} \overline{E^2}^{2\pi/\omega}$ [where $\overline{(\cdots)}^{2\pi/\omega}$ stands for averaging over the radiation cycle] or, equivalently, the probability of γ -quantum detection per unit time, is given by

$$I_{\text{out}}(\xi) = I_0 \exp(-T_e) \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} J_n(kR) J_m(kR) \exp[i(n-m)\vartheta_0] \exp[i(n-m)\Omega\xi] \\ \times \frac{1}{4\pi^2} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \frac{\exp[-i(\omega_1 - \omega_2)(\xi - \xi_0)]}{(\Gamma_s/2 - i\omega_1)(\Gamma_s/2 + i\omega_2)} \exp\left(-\frac{\gamma_a T_a/2}{\gamma_a + i(\Delta\omega_n - \omega_1)} - \frac{\gamma_a T_a/2}{\gamma_a - i(\Delta\omega_m - \omega_2)}\right) d\omega_1 d\omega_2, \quad (21)$$

where $I_0 = \frac{c}{2\pi} E_0^2$ is intensity of the incident Mössbauer radiation $\Delta \omega_n \equiv \omega_a - \omega_s + n\Omega$.

γ -RAY-PULSE FORMATION IN A VIBRATING . . .

Up to now we have considered the transformation of Mössbauer radiation of a radioactive source at rest in a vibrating resonant absorber. Alternatively, one may consider a vibrating source and the resonant absorber at rest. Motion of the absorber relative to the source is equivalent to the motion of the source relative to the absorber. Thus, if the source vibrates along the axis, pointing from the source to the absorber,

$$z_0(t) = z_0 - R\sin(\Omega t + \vartheta_0), \tag{22}$$

at the absorber's entrance the incident single-photon field is given by

$$E_{\rm inc}(z_l,t) = E_0\theta((t-t_0) - [z_l + R\sin(\Omega t + \vartheta_0) - z_0]/c)\exp(-(i\omega_s + \Gamma_s/2)\{(t-t_0) - [z_l + R\sin(\Omega t + \vartheta_0) - z_0]/c\} + i\phi_0) + c.c.$$
(23)

Equations (4) and (23) coincide if z_a is replaced by z_l so that the field of the vibrating source in the laboratory reference frame coincides with the field of the source at rest in the reference frame of the vibrating absorber. Therefore, the solution obtained previously for transformation of a γ photon in the vibrating absorber also describes transformation of the photon emitted by the vibrating source in the resonant absorber at rest. In the latter case the output radiation (in the laboratory reference frame) is given by

$$E_{\text{out}}(z_l,t) = e^{-i\omega_s(\xi-\xi_0)+i\phi_0'} \sum_{n=-\infty}^{+\infty} \exp[in(\Omega\xi+\vartheta_0)] \int_{-\infty}^{+\infty} E_n^{\text{out}}(\omega,\xi_0) \exp(-i\omega\xi) d\omega + \text{c.c.},$$
(24)

where $E_n^{\text{out}}(\omega)$ are defined by Eq. (19). The corresponding intensity is expressed by Eq. (21) since

$$\left|\exp\left[-ikR\sin(\Omega\xi+\vartheta_0)\right]\right|^2 = \left|\sum_{m=-\infty}^{+\infty} J_m(kR)\exp\left[-im(\Omega\xi+\vartheta_0)\right]\right|^2 = 1.$$

To conclude this section let us calculate the intensity of the overall stochastic flow of γ photons propagating through the vibrating resonant absorber. By the choice of a zero moment of time one can set $\vartheta_0 \equiv 0$, however the instant of formation of the excited state of the radiating nucleus t_0 is absolutely undefined, so ξ_0 in Eq. (21) is a random value spreading from 0 to 2π with equal probability. Thus, the experimentally observed detection probability of γ quanta corresponds to that averaged over ξ_0 . Summing up the intensities (21) of individual γ photons, radiated in different instants of time, one obtains

$$I_{\Sigma}(\xi) = \frac{M}{\Gamma_s} I_0 \exp(-T_e) \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} J_n(kR) J_m(kR) B_{nm} \exp[i(n-m)\Omega\xi], \qquad (25)$$

where M is the average number of γ quanta detected per unit time and

$$B_{nm} = \frac{\Gamma_s}{2\pi} \int_{-\infty}^{+\infty} \frac{1}{\omega^2 + \Gamma_s^2/4} \exp\left(-\frac{\gamma_a T_a/2}{\gamma_a + i(\Delta\omega_n - \omega)}\right) \exp\left(-\frac{\gamma_a T_a/2}{\gamma_a - i(\Delta\omega_m - \omega)}\right) d\omega.$$
(26)

In dimensionless variables Eqs. (25) and (26) take the form

$$I_{\Sigma}(x) = \frac{M}{\Gamma_s} I_0 \exp(-T_e) \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} J_n(kR) J_m(kR) B_{nm} \exp\left(i(n-m)\frac{\eta x}{2\kappa}\right),\tag{27}$$

$$B_{nm} = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{1}{z^2 + 1} \exp\left(-\frac{T_a/2}{1 + i(\Delta_n - \kappa z)}\right) \exp\left(-\frac{T_a/2}{1 - i(\Delta_m - \kappa z)}\right) dz,$$
(28)

where $x = \Gamma_s \xi$, $v = \frac{\omega_a - \omega_s}{\gamma_a}$, $\eta = \frac{\Omega}{\gamma_a}$, $\Delta_n = v + \eta n$, $\kappa = \frac{\Gamma_s}{2\gamma_a}$, and $z = \frac{2\omega}{\Gamma_s}$. It is worth noting that in the limit of a very small linewidth of the Mössbauer source as compared to either the linewidth of the absorber or the detuning of each spectral component from the resonance $\kappa \ll \Delta_n, \Delta_m$ and the relatively small Mössbauer thickness of the absorber $\frac{T_a/2}{1+\Delta_n^2}, \frac{T_a/2}{1+\Delta_m^2} \leq 1$, the coefficients B_{nm} take the form $B_{nm} =$ $\exp(-\frac{T_a/2}{1+i\Delta_n})\exp(-\frac{T_a/2}{1-i\Delta_m})$. This result can be obtained if one assumes that radiation of the Mössbauer source is monochromatic. Therefore, in this limiting case the output γ -radiation intensity can be adequately calculated within the approximation of monochromatic incident radiation.

According to (25) and (27), the time dependence of the intensity of the output γ radiation is determined by interference of the spectral components, separated from each other by the frequency of vibration Ω . If (i) the output bandwidth substantively exceeds the vibration frequency, (ii) the amplitudes of a considerable number of output spectral components are of the same order of magnitude, and (iii) the difference between the initial phases of every pair of neighboring spectral components is approximately the same, the time dependence of the output intensity corresponds to a train of pronounced pulses. As we will show in the next section, under optimal conditions the pulse duration can be much smaller than the period of vibration, while the peak pulse intensity can be several times higher than the intensity of the incident radiation.

III. RESULTS OF OPTIMIZATION AND POSSIBLE EXPERIMENTS

The time dependence of intensity of the output radiation (27) and (28) is determined by six dimensionless parameters

$$P_{\omega} = kR, \quad T_{a} = \frac{4\pi n_{12}^{0} f_{a} N |d_{21}|^{2}}{\sqrt{\varepsilon} \hbar \gamma_{a}} \frac{\omega_{a}}{c} h, \quad T_{e} = 2\delta_{e} h,$$
$$\nu = \frac{\omega_{a} - \omega_{s}}{\gamma_{a}}, \quad \eta = \frac{\Omega}{\gamma_{a}}, \quad \kappa = \frac{\Gamma_{s}}{2\gamma_{a}}, \tag{29}$$

fully characterizing the interaction between the incident γ radiation and the resonant absorber. Here P_{ω} is the modulation index; T_a is the Mössbauer thickness of the absorber; T_e is the exponent of photoelectric absorption; ν is the detuning of the absorber resonant frequency from the central frequency of the source, normalized to the halfwidth of the absorber spectral line; η is the frequency of vibration, normalized to the halfwidth of the absorber spectral line; and κ is the linewidth of the Mössbauer source, normalized to the absorber linewidth.

An arbitrary set of parameters (29) corresponds to the time dependence of the output intensity (21) given by a superposition of beats of different spectral components of the output radiation. However, as shown in [40], under optimal conditions the output radiation acquires the form of a train of ultrashort pulses. Since the time dependences of the output intensity in the laboratory reference frame and in the absorber reference frame coincide, the pulse formation can be viewed as a conversion of the frequency-modulated incident γ radiation (in the absorber's reference frame in the case of an oscillating absorber and in the laboratory reference frame in the case of an oscillating source) into an amplitude-modulated output radiation. This conversion occurs due to selective attenuation and phase incursion acquired by the spectral components of γ radiation during its propagation through the absorber due to the resonant absorption and dispersion, respectively (for more details see [40]). Alternatively, one may consider pulse formation as a consequence of interference of the incident and the resonantly scattered radiation in the time domain [17,18,20–23,37–39]. However, in the case of harmonic motion of either the source or absorber, the spectral representation is generally more convenient.

In this section we study the optimal conditions of pulse formation numerically using the two criteria of optimality. First, we seek the solution with the maximum pulse peak intensity max{ I_{max}/I_0 }, where I_{max} is the peak intensity of the produced pulses and I_0 is the intensity of the incident Mössbauer radiation. Then we seek solutions with the maximum ratio between the pulse peak intensity and the time-averaged intensity of the output Mössbauer radiation max{ I_{max}/I_{av} }, where I_{av} is the time-averaged output intensity. We present the results of global optimization of intensity (27) and (28) versus the parameters P_{ω} , T_a , η , and ν for the fixed values of $\kappa = 1$ and $T_e = T_a/180$ in the experimentally feasible regions $1 \leq P_{\omega} \leq 7$, $1 \leq T_a \leq 20$, $1 \leq \eta \leq 45$, and at arbitrary ν . A discussion of the expected results outside this



FIG. 1. (Color online) Generic scheme of the proposed experimental setup: γ -ray photons from the ⁵⁷Co Mössbauer radioactive source propagate through the resonant ⁵⁷Fe absorber, mounted on a piezoelectric plate that oscillates along the direction of γ -ray propagation with the frequency Ω , the amplitude *R*, and the initial phase ϑ_0 under the action of harmonic voltage, produced by a radiofrequency (rf) generator. The rf generator also produces start signals, synchronized to some fixed phase of the absorber vibration. These signals start the clock in the processing unit. The clock stops upon receiving the stop signals from detector of the resonant 14.4-keV γ -ray photons, placed behind the absorber. Such a technique allows measuring the probability of γ -photon detection vs the phase of the absorber's vibration, which corresponds to the time dependence of the intensity of the output γ radiation, averaged over the time of formation of the excited state of radiating nuclei.

region and for different values of the parameters κ and T_e is given after the results of optimization.

The discussed approach to formation of the ultrashort γ -ray pulses can be experimentally realized using various Mössbauer sources and absorbers. Let us consider the most popular ⁵⁷Co radioactive source, which is characterized by a photon energy of 14.4 keV, corresponding to a radiation wavelength of 0.861 Å, a transition linewidth of $\Gamma_s/2\pi = 1.13$ MHz (corresponding to a lifetime of the radiating nucleus of 141 ns), and the probability of recoilless emission at room temperature $f_s =$ 0.77. In particular, this kind of source was used in our recent experiment [40]. A sketch of the proposed experimental setup is drawn in Fig. 1 and is described in the caption to this figure (a more detailed description is given in [40].) As a resonant absorber let us consider the stainless-steel foil 100% enriched by ⁵⁷Fe isotope, possessing an unsplit resonant transition line with the natural linewidth $2\gamma_a = \Gamma_s$ (corresponding to $\kappa = 1$). This differs from the experiment in [40], where the stainless-steel absorber had a natural abundance, $\sim 2\%$, of ⁵⁷Fe. Since γ radiation resonantly interacts only with ⁵⁷Fe nuclei while it is nonresonantly absorbed because of photoionization from inner electronic shells of all the atoms of a compound, an increase in concentration of ⁵⁷Fe allows one to achieve the same value of Mössbauer thickness of the absorber at a proportionally reduced physical length and consequently to reduce the undesired nonresonant absorption and increase the conversion efficiency of the incident Mössbauer radiation into a pulse train. In the case of stainless steel 100% enriched by ⁵⁷Fe, the coefficient of resonant absorption equals $2g(\omega_a) =$ $8.8 \,\mu m^{-1}$, while the decrement of nonresonant absorption is approximately $2\delta_e = 0.05 \,\mu \text{m}^{-1}$ [12], corresponding to $T_e = T_a/180$. Accordingly, even for the maximal considered



FIG. 2. (Color online) Time dependence of the intensity of γ radiation transformed in a vibrating resonant absorber under the conditions optimal for formation of pulses with the highest peak intensity (see the text). The output intensity I is normalized to the intensity of the incident radiation I_0 . Time is normalized to the cycle of the absorber's vibration $2\pi / \Omega$. The pulse peak intensity is $I_{\text{max}} = 2.77I_0$ and the time-averaged output intensity is $I_{\text{av}} = 0.79I_0$. The duration of the main pulse is $\tau_{\text{pulse}} = \pi / 6\Omega \approx 0.09 / \Gamma_s$. In the case of the ⁵⁷Co radioactive source and ⁵⁷Fe resonant absorber $\tau_{\text{pulse}} = 12.5$ ns and the pulse repetition period equals $2\pi / \Omega = 147$ ns.

Mössbauer thickness of the absorber $T_a = 20$, the nonresonant losses $1 - \exp(-T_e)$ do not exceed 11%.

Let us first consider the results of optimization according to the criterion $I_{\text{max}}/I_0 \rightarrow \text{max}$, which corresponds to the formation of pulses with the highest peak intensity. The time dependence of the output radiation intensity, corresponding to the optimal parameter values $P_{\omega} = 6.75$, $T_a = 20$, $\eta = 11.96$, and v = -65.78, is plotted in Fig. 2. The peak intensity of the produced pulses is nearly three times higher than the intensity of the incident γ radiation $I_{\text{max}} = 2.77 I_0$, the time-averaged output intensity is $I_{\rm av} = 0.79I_0$, and the duration of pulses is $\tau_{\rm pulse} \simeq \pi/6\Omega \simeq 0.09/\Gamma_s$. In the case of a ⁵⁷Co radioactive source and a stainless-steel absorber 100% enriched by ⁵⁷Fe, the above listed values of dimensionless parameters correspond to the frequency of the absorber vibration $\Omega/2\pi =$ 6.8 MHz, the amplitude of vibration R = 0.93 Å, the absorber thickness $h = 2.3 \,\mu\text{m}$, and the constant velocity of the source towards the absorber $V_s = 3.22 \text{ mm/s}$. The pulse duration is 12.5 ns, which is an order of magnitude smaller than the lifetime of the excited state of the radiating nucleus; the pulse repetition period equals 147 ns. However, the major pulses are accompanied by trains of minor peaks, which indicates that the phases of the output spectral components are not fully aligned (there remains an uncompensated frequency chirp). This fact can be most easily explained in the reference frame of the resonant absorber (recollecting once again that the time dependences of the intensity of the output γ radiation in the laboratory reference frame and in the reference frame associated with the absorber coincide). In the absorber's reference frame the complex amplitude of the *n*th incident spectral component (7) [note that according to Eq. (6), the frequency of the *n*th component is smaller than the central frequency of the incident radiation by $n\Omega$] is proportional to Bessel function of the corresponding order $J_n(P_{\omega})$. Since the sign of the Bessel function is unperiodically altered



FIG. 3. (Color online) Spectra of the incident (dashed blue line) and output (solid blue line) γ radiation, resonant absorption (bold solid red line), and dispersion (bold dashed black line), corresponding to the time dependence of the intensity, shown in Fig. 2. All the spectra are plotted in the reference frame associated with the resonant absorber.

with a change of its order, the dependence of phases of the incident spectral components on their frequencies is nonlinear. With an increase of the modulation index P_{ω} the number of substantially nonzero spectral components of the incident radiation increases $\sim 2P_{\omega} + 1$ and the spectral dependence of their phases becomes more complicated. Since the possibility of transforming the incident spectrum (7) into the output spectrum (19) is limited to attenuation of the spectral components via the resonant absorption and incursion of their phases via the resonant dispersion, for large values of the modulation index it turns out to be impossible to fully align phases of all the appreciably nonzero output components. A more detailed discussion of this issue will be given in connection with Figs. 4 and 5. This mismatching of phases of the output spectral components (and the oscillating tails behind the major pulses) can be reduced via a reduction of the modulation index at the cost of a decreasing pulse peak intensity and increasing pulse duration. It is noteworthy that a reduction of the modulation index makes the produced pulses closer to the bandwidth-limited ones. Accordingly, the pulse peak intensity only slightly decreases with a reduction of the output radiation bandwidth.

The spectra of the incident and the output γ radiation corresponding to the time dependence of intensity are shown in Fig. 2 and the curves of resonant absorption and dispersion are plotted in Fig. 3. The spectra are drawn in the absorber's reference frame (which oscillates in the case of the oscillating absorber and coincides with the laboratory reference frame in the case of the oscillating source). As can be seen from these spectra, the radiation bandwidth greatly exceeds the linewidth of the absorber's transition and none of the spectral components is in resonance. Thus, the resonant absorption plays a minor role in the transformation of the incident radiation, although it cannot be completely neglected. The major role is played by the resonant dispersion, which leads to positive phase incursion for the spectral components above resonance and negative phase incursion for the spectral components below resonance, thus enabling us to adjust their phases inside the absorber. We would like to note the two advantages of the resonant dispersion versus the nonresonant one for



FIG. 4. (Color online) Dependence of the maximum achievable peak intensity of the pulses I_{max} normalized to the intensity of the incident Mössbauer radiation I_0 on the modulation index $P_{\omega} = kR$ and the dimensionless vibration frequency $\eta = \Omega/\gamma_a$. The results are presented for the optimal (and generally different for each point of the surface) values of Mössbauer thickness of the absorber T_a and dimensionless detuning of the resonance frequency of the absorber from the central frequency of the source $v = (\omega_a - \omega_s)/\gamma_a$.

compensation of the harmonic frequency modulation [36]. First, it enables us to transform the incident Mössbauer radiation into a pulse train in a relatively thin absorber $T_a \leq 20$, where the nonresonant electronic absorption is not substantial (for the stainless-steel absorber, 100% enriched by ⁵⁷Fe, it does not exceed 11%), while conversion of Mössbauer radiation via nonresonant dispersion requires an increase of the absorber thickness by orders of magnitude, resulting in drastic nonresonant damping of the output radiation. Second, in the limit $T_a \gg 1$, $\Omega/\gamma_a \gg 1$, $T_a\gamma_a/\Omega = \text{const}$, the resonant dispersion allows better compensation of the harmonic frequency modulation than the nonresonant one [36], even in the hypothetic case of zero electronic absorption.

Let us now examine the dependence of the peak pulse intensity on the values of the parameters in (29) and analyze the optimal conditions of pulse formation. The dependence of the peak pulse intensity I_{max} , normalized to the intensity of the incident radiation I_0 , on modulation index $P_{\omega} = kR$, the dimensionless vibration frequency $\eta = \Omega/\gamma_a$ for the optimal (and generally different for each point of the surface) values of the Mössbauer thickness T_a , and dimensionless detuning of the resonance frequency of the absorber from the central frequency of the source $v = (\omega_a - \omega_s)/\gamma_a$ is plotted in Fig. 4. Generally, the resonant absorber either works as a nonlinear dispersive element leading to different phase incursions of different spectral components of the incident radiation in the absorber's reference frame (6) and (7), performs selective attenuation of the most intensive antiphase (to the majority) spectral component, or combines phase incursions with selective attenuation. As shown in Fig. 4, there are two major parameter regions where the peak pulse intensity approaches its maximum value. The first region corresponds to vibration frequencies exceeding by 6-7 times the absorber's linewidth $\Omega = 10\gamma_a - 15\gamma_a$ and modulation indeces $P_{\omega} \ge 3.5$. In this region the pulse formation relies on the resonant dispersion, which provides partial compensation



FIG. 5. (Color online) Dependence of the maximum achievable peak intensity of the pulses I_{max} normalized to the intensity of the incident Mössbauer radiation I_0 on the modulation index $P_{\omega} = kR$ and the Mössbauer thickness of the absorber T_a for the optimal (and generally different for each point of the surface) values of the dimensionless vibration frequency $\eta = \Omega/\gamma_a$ and dimensionless detuning of the absorber's resonance frequency from the central frequency of the source $\nu = (\omega_a - \omega_s)/\gamma_a$.

for the harmonic frequency modulation of the incident γ radiation. The pulses with a better shape, similar to those reported in [40] but with a bit smaller peak intensity, can be produced in the second optimal region $1.5 < P_{\omega} < 2.5, \Omega =$ $20\gamma_a - 40\gamma_a$, where the resonant absorption, rather than the resonant dispersion, plays a crucial role. The main effect of the absorber on the incident radiation in such a case is suppresion of the strongest antiphased component of its spectrum in the absorber's reference frame [40]. In between these regions $2.5 < P_{\omega} < 3.5$, $\Omega = 15\gamma_a - 25\gamma_a$, intense pulses can be produced due to theh joint action of the resonant absorption and dispertion on γ radiation. In Fig. 5 we show the dependence of the normalized peak intensity of the pulses I_{max}/I_0 on the modulation index $P_{\omega} = kR$ and Mössbauer thickness T_a for the optimal (and generally different) values of dimensionless vibration frequency $\eta = \Omega/\gamma_a$ and dimensionless detuning of frequency of the absorber from the frequency of the source $v = (\omega_a - \omega_s)/\gamma_a$. As shown in Fig. 5, the pulse peak intensity rises monotonically in the region $1 \leq T_a \leq 20$ with increasing Mössbauer thickness of the absorber T_a . This rise originates either from the possibility of increasing the frequency of vibration Ω , preserving phase incursions of the spectral components, and thus decreasing absorption of nonresonant components in the regime of dominating resonant dispersion or via the ability to reduce further the antiphased resonant component in the regime of dominating resonant absorption or due to both of these mechanisms. The dependence of the pulse peak intensity on the modulation index P_{ω} is quasiperiodic with a quasiperiod, reflecting alternations of sign of the Bessel functions $J_n(P_{\omega})$ of successive orders *n*. This periodicity is caused by modification of the phase distribution of the incident spectral components (6) and (7), which occurs with an alternation of sign of Bessel functions $J_n(P_{\omega})$, and changes the optimal conditions of the radiation-matter interaction. As shown in Fig. 5, the pulse peak intensity just slightly grows with an increase of the modulation index P_{ω} from 2 up to 7. Such behavior originates from the impossibility of



FIG. 6. (Color online) Time dependence of the intensity of the γ radiation transformed in a vibrating resonant absorber under the conditions optimal for the generation of pulses with the highest ratio between the peak and the time-averaged intensity (see the text). The output intensity I is normalized to the intensity of the incident Mössbauer radiation I_0 . Time is normalized to the cycle of absorber's vibration $2\pi / \Omega$. The pulse peak intensity is $I_{\text{max}} = 2.52I_0$, the time-averaged output intensity equals $I_{\text{av}} = 0.70I_0$, and the ratio between the peak and the time-averaged intensity is $I_{\text{max}}/I_{\text{av}} = 3.61$. The pulse duration is $\tau_{\text{pulse}} = \frac{\pi}{2.75\Omega} \approx 0.13/\Gamma_s$. In the case of the ⁵⁷Co radioactive source and ⁵⁷Fe resonant absorber $\tau_{\text{pulse}} = 18$ ns, the pulse repetition period equals $2\pi / \Omega = 99$ ns.

full compensation of the harmonic frequency modulation via interaction with a single resonant absorber for $P_{\omega} \ge 2$. As a result, with an increase of the modulation index above $P_{\omega} = 2$ the oscillatory tails grow behind the pulses, although the peak pulse intensity slightly increases. In the opposite case $P_{\omega} < 2$ there is only one negative Bessel function of substantially nonzero amplitude, that is, $J_{-1}(P_{\omega})$. In such a case, the pulse formation implies either suppression of the corresponding -1spectral component via tuning it to the absorber resonance [40] or inversion of its sign via π phase incursion on the tail of the resonance. Whatever approach is chosen, in the case $P_{\omega} < 2$ the produced pulses are nearly bandwidth limited. Further reduction of the modulation index leads to narrowing of both the incident and the output radiation spectra, a decrease of the pulse peak intensity, and an increase of the pulse duration. Finally, as shown in both Figs. 4 and 5, there are minimal values of the parameters $P_{\rm min} \sim 1$, $T_{a \rm min} \sim 5$, and $\eta_{\rm min} \sim 1$ below which the pulse train cannot be formed.

Let us now turn to the results of optimization according to the criterion of highest ratio between the peak and the time-averaged intensity of the produced pulse train $I_{\rm max}/I_{\rm av} \rightarrow$ max. The highest value of this ratio inside the considered parameter region equals $I_{\text{max}}/I_{\text{av}} = 3.61$ and is achieved for $P_{\omega} = 2.0$, $T_a = 20$, $\eta = 17.94$, and $\nu = -22.425$. The time dependence of the intensity of the output radiation corresponding to these parameter values is plotted in Fig. 6. The peak pulse intensity $I_{\text{max}} = 2.52I_0$, the average intensity of the output radiation $I_{av} = 0.70I_0$, and the pulse duration $\tau_{\text{pulse}} = \pi / 2.75 \,\Omega$, that is $\tau_{\text{pulse}} \approx 0.13 / \Gamma_s$ for $\eta = 17.94$. The pulse train shown in Fig. 6 can be produced using a stainlesssteel absorber 100% enriched by ⁵⁷Fe, vibrating at a frequency of $\Omega/2\pi = 10.1$ MHz with an amplitude of R = 0.27 Å and a ⁵⁷Co radioactive source, moving towards the absorber with a constant velocity of $V_s = 1.09 \text{ mm/s}$. The required



FIG. 7. (Color online) Spectra of the incident (dashed blue line) and output (solid blue line) γ radiation, resonant absorption (bold solid red line), and dispersion (bold dashed black line), corresponding to the time dependence of the intensity, shown in Fig. 6. All the spectra are plotted in the reference frame of the resonant absorber.

absorber thickness is $h = 2.3 \,\mu$ m. The duration of pulses is $\tau_{\rm pulse} = 18 \, \rm ns$ and the pulse repetition period equals $2\pi / \Omega =$ 99 ns. The corresponding spectra in the absorber's reference frame are shown in Fig. 7. The –1 and –3 spectral components of the incident γ radiation (6) and (7) are in antiphase to the other components, but the amplitude of the -3 component is rather small to affect the time dependence of the output radiation, so the role of the resonant absorber is to adjust the -1 component; this is done by means of both the resonant attenuation and the resonant phase incursion. The later is close to π , providing constructive interference of the -1 spectral component with the other components outside the absorber. It is worth noting that in the case of a thinner Mössbauer absorber, the optimal regime will be based on the full resonant absorption of the -1 spectral component, while if one admits thicker absorbers, the optimal regime will be based solely on the resonant phase incursion.

In Figs. 8 and 9 we show the dependences of the ratio between the peak pulse intensity I_{max} and the time-averaged intensity of the output Mössbauer radiation I_{av} on the modulation index P_{ω} and either the dimensionless vibration frequency $\eta = \Omega / \gamma_a$ or the Mössbauer thickness of the absorber T_a . The graphs are plotted for the optimal (and generally different for each point of the surfaces) values of dimensionless detuning of the resonance frequency of the absorber from the central frequency of the source $v = (\omega_a - \omega_s)/\gamma_a$ and either the Mössbauer thickness T_a or the dimensionless vibration frequency η , respectively. Figures 8 and 9 resemble the dependences of the absolute value of the pulse peak intensity on the same parameters shown in Figs. 4 and 5. However, the maximum value of the ratio $I_{\text{max}}/I_{\text{av}}$ is achieved at smaller modulation indices $P_{\omega} = 1.75 - 2.0$ and corresponds to pulses of better shape as compared to those that maximize the peak intensity.

At this point we would like to discuss what should be expected outside the considered parameter region $1 \le P_{\omega} \le 7$, $1 \le T_a \le 20$, $1 \le \eta \le 45$ and for values of the parameters κ and T_e that are different from $\kappa = 1$ and $T_e = T_a/180$. An increase of the modulation index P_{ω} (the oscillation amplitude) leads to a proportionally increase of the output radiation bandwidth and under optimal conditions to a decrease of the pulse duration. However, because of unperiodically alternating



FIG. 8. (Color online) Dependence of the ratio between the peak intensity of the pulses I_{max} and the time-averaged intensity of the output radiation I_{av} on the modulation index $P_{\omega} = kR$ and dimensionless vibration frequency $\eta = \Omega/\gamma_a$. The results are presented for the optimal (and generally different for each point of the surface) values of the Mössbauer thickness of the absorber T_a and the dimensionless detuning of the resonance frequency of the absorber from the central frequency of the source $v = (\omega_a - \omega_s)/\gamma_a$.

signs of Bessel functions $J_n(P_{\omega})$ of different orders n, the phases of both the incident and the output spectral components (20) become more and more disordered with increasing P_{ω} . Therefore, a sequence of minor pulses grows behind the major pulse with increasing P_{ω} . As discussed in connection to Fig. 4, an increase of the Mössbauer thickness of the absorber T_a allows an increase in both the peak pulse intensity I_{max} and the ratio between the peak and the time-averaged output radiation intensity $I_{\text{max}}/I_{\text{av}}$. However, an increase of the Mössbauer thickness T_a is accompanied by an increase of the exponent of photoelectric absorption inside the absorber T_e , leading to essential attenuation of the output radiation. Therefore, for a fixed ratio T_a/T_e (which is determined by



FIG. 9. (Color online) Dependence of ratio between the peak intensity of the pulses I_{max} and the time-averaged intensity of the output radiation I_{av} on the modulation index $P_{\omega} = kR$ and the Mössbauer thickness of the absorber T_a for the optimal (and generally different for each point of the surface) values of the dimensionless vibration frequency $\eta = \Omega/\gamma_a$ and dimensionless detuning of the absorber's resonance frequency from the central frequency of the source $\nu = (\omega_a - \omega_s)/\gamma_a$.

the concentration of ⁵⁷Fe nuclei in the absorber) there is an optimal Mossbauer thickness T_a (generally different for each value of P_{ω}) at which the largest values of I_{max} or $I_{\text{max}}/I_{\text{av}}$ can be achieved. An increase of the dimensionless oscillation frequency η enables an increase of both I_{max} and $I_{\text{max}}/I_{\text{av}}$ if the Mössbauer thickness T_a can be simultaneously increased. With increasing oscillation frequency both the pulse duration and the pulse repetition period are proportionally reduced and can take values much smaller than those considered above. A decrease of the source's linewidth relative to the absorber's linewidth κ for the fixed values of all the other parameters enhances both I_{max} and $I_{\text{max}}/I_{\text{av}}$. The best results would be obtained for monochromatic incident radiation $\kappa = 0$. However, the source's linewidth cannot be smaller than the natural limit, thus a decrease of the parameter κ below $\kappa = 1$ can be achieved only via broadening of the absorber's transition line, which leads either to a decrease of the Mössbauer thickness T_a for a fixed T_e (and fixed absorber thickness h) or to an increase of the exponent of the photoelectric absorption T_e for a fixed T_a (and increased absorber thickness h). Both a decrease of T_a and an increase of T_e affect pulse formation negatively. Thus, although a slight decrease of the parameter κ below $\kappa = 1$ might lead to certain enhancement of I_{max} and $I_{\text{max}}/I_{\text{av}}$, a substantial decrease of κ via an increase of the absorber's linewidth diminishes the effect.

We would like to stress the remarkably high efficiency of conversion of the incident radiation into the γ -pulse train, defined as the ratio between the intensity of the incident Mössbauer radiation (the average number of γ quanta detected per unit time) and the time-averaged intensity of the output radiation, i.e., a pulse train under optimal conditions. This total efficiency equals 79% for the pulses, shown in Fig. 1, and 70% for the pulses, shown in Fig. 6.

IV. CONCLUSION

In this paper we investigated the ultimate possibilities of the recently proposed method for γ -ray pulse formation [40] based on the resonant interaction of γ quanta with a vibrating absorber or the resonant interaction of γ quanta radiated by a vibrating source with an absorber at rest. We discussed the possibility of experimental realization using a 57Co Mössbauer radioactive source of 14.4-keV γ photons and the fully enriched by 57Fe stainless steel Mössbauer absorber with natural transition linewidths, corresponding to the excited-state lifetime of 141 ns. We have shown the experimental possibility of generating (10-20)-ns pulses with a peak intensity up to 2.8 times higher than the intensity of the incident radiation. The efficiency of conversion into the pulse train can be as high as 70%–80% under the experimentally feasible conditions. A reduction of the pulse duration to 2-5 ns can be achieved via an increase of the vibration frequency. In such a case, if the pulse formation relies on the resonant dispersion, the increase of vibration frequency should be accompanied by the proportional increase of the Mössbauer thickness of the absorber. However, if the resonant absorption plays a major role, the optimal value of the Mössbauer thickness remains unchanged. A further reduction of the pulse duration below 1 ns might be possible via broadening of the output radiation

spectrum through an increase of the vibration amplitude and phase matching of the generated spectral components either in a set of vibrating resonant absorbers [53] or in the Mössbauer absorber(s), moving with constant velocity [54]. Use of the piezoelectric transducers vibrating at gigahertz frequencies [55,56] would allow a proportional reduction of the pulse duration (by two and more orders of magnitude as compared to the above case of vibration frequencies of tens of megahertz). Since the discussed method of γ -ray pulse formation is based on the resonant interaction of γ radiation with the nonstationary absorber, it is inherently related to our recent works on generation of extremely short (attosecond and femtosecond) few-cycle optical pulses [57-61]. As compared to the previously proposed techniques for γ -raypulse formation from radiation of a Mössbauer radioactive source [10,17,18,20–22,34–39] for a ⁵⁷Co source and ⁵⁷Fe absorber pair, the discussed approach promises the formation of shorter pulses than that generated via abrupt displacement of the source or absorber and more intense than the pulses produced via magnetization reversal. The discussed method is especially fruitful for the purpose of pulse train formation with a high repetition rate, considerably exceeding the relaxation rate of the resonant nuclear transition. This is possible since the pulse formation in a vibrating absorber does not require the resonantly scattered radiation to approach the steady-state

- P. Gütlich, E. Bill, and A. X. Trautwein, *Mössbauer Spectroscopy and Transition Metal Chemistry* (Springer, Berlin, 2010).
- [2] R. Röhlsberger, Nuclear Condensed Matter Physics Using Synchrotron Radiation, Springer Tracts Modern Physics Vol. 208 (Springer, Berlin, 2005).
- [3] Y. V. Shvyd'ko, X-Ray Optics: High Energy-Resolution Applications (Springer, Berlin, 2004).
- [4] *Nonlinear Optics, Quantum Optics, and Ultrafast phenomena with X-Rays*, edited by B. Adams (Kluwer, Dordrecht, 2003).
- [5] European Synchrotron Radiation Facility, http://www.esrf.eu/.
- [6] Linac Coherent Light Source, https://portal.slac.stanford.edu/ sites/lcls_public/Pages/Default.aspx.
- [7] European XFEL, http://www.xfel.eu/.
- [8] J. Amann *et al.*, Demonstration of self-seeding in a hard-x-ray free-electron laser, Nat. Photon. 6, 693 (2012).
- [9] T. Popmintchev, M.-C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Ališauskas, G. Andriukaitis, T. Balčiunas, O. D. Mücke, A. Pugzlys, A. Baltuška, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernández-García, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, Bright coherent ultrahigh harmonics in the keV x-ray regime from mid-infrared femtosecond lasers, Science 336, 1287 (2012).
- [10] E. Ikonen, P. Helistö, T. Katila, and K. Riski, Coherent transient effects due to phase modulation of recoilless γ radiation, Phys. Rev. A **32**, 2298 (1985).
- [11] G. V. Smirnov, Nuclear resonant scattering of synchrotron radiation, Hyperfine Interact. 97-98, 551 (1996).
- [12] G. V. Smirnov, General properties of nuclear resonant scattering, Hyperfine Interact. 123-124, 31 (1999).
- [13] Y. Kagan, Theory of coherent phenomena and fundamentals in nuclear resonant scattering, Hyperfine Interact. 123-124, 83 (1999).

value before the formation of each pulse, unlike the methods based on instantaneous displacement of the source or absorber or magnetization reversal. On the other hand, the combination of the discussed approach with the technique of resonant interruption of γ radiation [19,21,23] or optimized unperiodic motion of the source or absorber might be useful for generation of isolated ultrashort γ -ray pulses of large amplitude. The proposed approach is promising for the development of a compact source of nearly bandwidth-limited γ -ray pulses and realization of the precise time-resolved x-ray–Mössbauer experiments in a laboratory.

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- [14] U. van Bürck, Coherent pulse propagation through resonant media, Hyperfine Interact. 123-124, 483 (1999).
- [15] Y. Rostovtsev and O. Kocharovskaya, Modification of Mössbauer spectra under the action of electromagnetic fields, Hyperfine Interact. 135, 233 (2001).
- [16] F. Vagizov, The splitting of hyperfine lines of ⁵⁷Fe nuclei in RF magnetic field, Hyperfine Interact. **61**, 1359 (1990).
- [17] P. Helistö, I. Tittonen, M. Lippmaa, and T. Katila, Gamma echo, Phys. Rev. Lett. 66, 2037 (1991).
- [18] I. Tittonen, M. Lippmaa, P. Helisto, and T. Katila, Stepwise phase modulation of recoilless gamma radiation in a coincidence experiment: Gamma echo, Phys. Rev. B 47, 7840 (1993).
- [19] Y. V. Shvyd'ko, G. V. Smirnov, S. L. Popov, and T. Hertrich, Observation of intensified forward γ-ray emission in spontaneous nuclear decay, JETP Lett. 53, 69 (1991).
- [20] Y. V. Shvyd'ko, S. L. Popov, and G. V. Smirnov, Resonant forward nuclear scattering of γ rays after a stepped change in the energy of an excited nuclear state, JETP Lett. **53**, 231 (1991).
- [21] G. V. Smirnov, Enhancement of the radiative channel in nuclear forward scattering, Hyperfine Interact. 72, 63 (1992).
- [22] Y. V. Shvyd'ko, S. L. Popov, and G. V. Smirnov, Coherent re-emission of γ -quanta in the forward direction after a stepwise change of the energy of nuclear excitation, J. Phys.: Condens. Matter **5**, 1557 (1993).
- [23] Y. V. Shvyd'ko, T. Hertrich, U. van Bürck, E. Gerdau, O. Leupold, J. Metge, H. D. Rüter, S. Schwendy, G. V. Smirnov, W. Potzel, and P. Schindelmann, Storage of nuclear excitation energy through magnetic switching, Phys. Rev. Lett. 77, 3232 (1996).
- [24] R. Röhlsberger, K. Schlage, B. Sahoo, S. Couet, and R. Rüffer, Collective Lamb shift in single-photon superradiance, Science 328, 1248 (2010).

- [25] R. Röhlsberger, H.-C. Wille, K. Schlage, and B. Sahoo, Electromagnetically induced transparency with resonant nuclei in a cavity, Nature (London) 482, 199 (2012).
- [26] K. P. Heeg, H.-C. Wille, K. Schlage, T. Guryeva, D. Schumacher, I. Uschmann, K. S. Schulze, B. Marx, T. Kämpfer, G. G. Paulus, R. Röhlsberger, and J. Evers, Vacuum-assisted generation and control of atomic coherences at x-ray energies, Phys. Rev. Lett. 111, 073601 (2013).
- [27] A. Pálffy, C. H. Keitel, and J. Evers, Single-photon entanglement in the keV regime via coherent control of nuclear forward scattering, Phys. Rev. Lett. **103**, 017401 (2009).
- [28] A. Pálfy, C. H. Keitel, and J. Evers, Coherent control of the cooperative branching ratio for nuclear x-ray pumping, Phys. Rev. B 83, 155103 (2011).
- [29] Y. V. Radionychev, M. D. Tokman, A. G. Litvak, and O. Kocharovskaya, Acoustically induced transparency in optically dense resonance medium, Phys. Rev. Lett. 96, 093602 (2006).
- [30] P. Anisimov, Y. Rostovtsev, and O. Kocharovskaya, Concept of spinning magnetic field at magic-angle condition for line narrowing in Mössbauer spectroscopy, Phys. Rev. B 76, 094422 (2007).
- [31] W.-T. Liao, A. Pálffy, and C. H. Keitel, Coherent storage and phase modulation of single hard-x-ray photons using nuclear excitons, Phys. Rev. Lett. **109**, 197403 (2012).
- [32] W.-T. Liao, A. Pálffy, and C. H. Keitel, Nuclear coherent population transfer with x-ray laser pulses, Phys. Lett. B 705, 134 (2011).
- [33] B. W. Adams, C. Buth, S. M. Cavaletto, J. Evers, Z. Harman, C. H. Keitel, A. Pálffy, A. Picón, R. Röhlsberger, Y. Rostovtsev, and K. Tamasaku, X-ray quantum optics, J. Mod. Opt. 60, 2 (2013).
- [34] P. Helistö, E. Ikonen, T. Katila, and K. Riski, Coherent transient effects in Mössbauer spectroscopy, Phys. Rev. Lett. 49, 1209 (1982).
- [35] P. Helistö, E. Ikonen, and T. Katila, Enhanced transient effects due to saturated absorption of recoilless γ-radiation, Phys. Rev. B 34, 3458 (1986).
- [36] E. Kuznetsova, R. Kolesov, and O. Kocharovskaya, Compression of γ -ray photons into ultrashort pulses, Phys. Rev. A **68**, 043825 (2003).
- [37] R. N. Shakhmuratov, F. Vagizov, and O. Kocharovskaya, Radiation burst from a single γ -photon field, Phys. Rev. A **84**, 043820 (2011).
- [38] R. N. Shakhmuratov, Reversible absorption of weak fields revealed in coherent transients, Phys. Rev. A 85, 023827 (2012).
- [39] R. N. Shakhmuratov, F. Vagizov, and O. Kocharovskaya, Single gamma-photon revival from sandwich absorbers, Phys. Rev. A 87, 013807 (2013).
- [40] F. Vagizov, V. Antonov, Y. V. Radeonychev, R. N. Shakhmuratov, and O. Kocharovskaya, Coherent control of the waveforms of recoilless γ-ray photons, Nature (London) 508, 80 (2014).
- [41] Y. M. Kagan, A. M. Afanas'ev, and V. G. Kohn, Excitation of isomeric nuclear states by synchrotron radiation, J. Phys. C 12, 615 (1979).
- [42] R. P. Feynman, R. B. Leighton, and M. Sands, *The Feynman Lectures on Physics* (Addison-Wesley, Reading, 1963), Vol. III, Chap. 31.
- [43] T. E. Cranshaw and P. Reivari, A Mössbauer study of the hyperfine spectrum of ⁵⁷Fe, using ultrasonic calibration, Proc. Phys. Soc. (London) **90**, 1059 (1967).

- [44] J. Asher, T. E. Cranshaw, and D. A. O'Connor, The observation of sidebands produced when monochromatic radiation passes through a vibrated resonant medium, J. Phys. A 7, 410 (1974).
- [45] L. T. Tsankov, The spectrum of Mossbauer radiation passed through a vibrating resonant medium, J. Phys. A 13, 2959 (1980).
- [46] L. T. Tsankov, Experimental observations on the resonant amplitude modulation of Mossbauer gamma rays, J. Phys. A 14, 275 (1981).
- [47] A. Y. Dzyublik, Effect of forced vibrations on the scattering of x-rays and Mössbauer radiation by a crystal (I), Phys. Status Solidi B 123, 53 (1984).
- [48] A. Y. Dzyublik, Effect of forced vibrations on the scattering of x-rays and Mössbauer radiation by a crystal. II. Dynamical effects, Phys. Status Solidi B 134, 503 (1986).
- [49] S. L. Popov, G. V. Smirnov, and Y. V. Shvyd'ko, Observed strengthening of radiative mechanism for a nuclear reaction in the interaction of γ radiation with nuclei in a vibrating absorber, JEPT Lett. **49**, 747 (1989).
- [50] Y. V. Shvyd'ko and G. V. Smirnov, Enhanced yield into the radiative channel in Raman nuclear resonant forward scattering, J. Phys.: Condens. Matter 4, 2663 (1992).
- [51] G. J. Perlow, Quantum beats of recoil-free γ radiation, Phys. Rev. Lett. **40**, 896 (1978).
- [52] J. E. Monahan and G. J. Perlow, Theoretical description of quantum beats of recoil-free γ radiation, Phys. Rev. A **20**, 1499 (1979).
- [53] Y. V. Radeonychev, V. A. Antonov, F. G. Vagizov, R. N. Shakhmuratov, and O. Kocharovskaya, Conversion of recoilless gamma-radiation into a periodic sequence of ultrashort intense pulses in a set of several sequentially placed resonant absorbers (unpublished).
- [54] V. A. Antonov, Y. V. Radeonychev, F. G. Vagizov, R. N. Shakhmuratov, and O. Kocharovskaya (unpublished).
- [55] A. Ambrosy and K. Holdik, Piezoelectric PVDF films as ultrasonic transducers, J. Phys. E 17, 856 (1984).
- [56] B. P. Sorokin, G. M. Kvashnin, A. P. Volkov, V. S. Bormashov, V. V. Aksenenkov, M. S. Kuznetsov, G. I. Gordeev, and A. V. Telichko, AlN/single crystalline diamond piezoelectric structure as a high overtone bulk acoustic resonator, Appl. Phys. Lett. 102, 113507 (2013).
- [57] Y. V. Radeonychev, V. A. Polovinkin, and Olga Kocharovskaya, Extremely short pulses via Stark modulation of the atomic transition frequencies, Phys. Rev. Lett. **105**, 183902 (2010).
- [58] V. A. Polovinkin, Y. V. Radeonychev, and O. Kocharovskaya, Few-cycle attosecond pulses via periodic resonance interaction with hydrogenlike atoms, Opt. Lett. 36, 2296 (2011).
- [59] V. A. Antonov, Y. V. Radeonychev, and O. Kocharovskaya, Formation of a single attosecond pulse via interaction of resonant radiation with a strongly perturbed atomic transition, Phys. Rev. Lett. **110**, 213903 (2013).
- [60] V. A. Antonov, Y. V. Radeonychev, and O. Kocharovskaya, Formation of ultrashort pulses via quantum interference between Stark-split atomic transitions in a hydrogenlike medium, Phys. Rev. A 88, 053849 (2013).
- [61] V. A. Antonov, T. R. Akhmedzhanov, Y. V. Radeonychev, and O. Kocharovskaya, Attosecond pulse formation via switching of resonant interaction by tunnel ionization, Phys. Rev. A 91, 023830 (2015).