

Revisiting photon-statistics effects on multiphoton ionization. II. Connection to realistic systemsG. Mouloudakis^{1,*} and P. Lambropoulos^{1,2}¹*Department of Physics, University of Crete, P.O. Box 2208, 71003 Heraklion, Crete, Greece*²*Institute of Electronic Structure and Laser, FORTH, P.O. Box 1527, 71110 Heraklion, Greece*

(Received 12 March 2019; published 19 June 2019)

In this paper, we extend the results of an earlier paper [G. Mouloudakis and P. Lambropoulos, *Phys. Rev. A* **97**, 053413 (2018)] in which we had demonstrated the limitations of the notion of nonresonant multiphoton ionization, in the exploration of photon statistics effects in nonlinear processes. Through the quantitative analysis of specific realistic processes, we provide the connection to conditions of intensity and pulse duration necessary in relevant experiments, including a recent seminal experiment demonstrating the effect of superbunching found in squeezed radiation. The outlook for a future era of studies on the interplay between stochasticity and nonlinearity in transitions driven by electromagnetic radiation is also discussed.

DOI: [10.1103/PhysRevA.99.063419](https://doi.org/10.1103/PhysRevA.99.063419)**I. INTRODUCTION**

In a recent paper [1] we examined the effect of field fluctuations on near-resonant two- and three-photon ionization. Through a quantitative analysis of the role of intensity, we found that the enhancement of the process due to bunching, more often than not, will be reduced owing to the onset of Rabi oscillations, even at seemingly large detuning from resonance. This makes the notion of nonresonant ionization rather tenuous and possibly misleading in the planning of an experiment. For processes of order higher than 3, it is practically inevitable that near resonances will modify the enhancement, even at moderate intensities, simply because the level spacing decreases with increasing level excitation.

That paper came at an auspicious time as it practically coincided with an experimental breakthrough by Spasibko *et al.* [2], in which enhancement in processes of order up to 4 due to superbunched light was reported. This represents a break in the rather long period of experimental drought in this field because, despite the rather extensive relevant theoretical literature over the past 50 years [3–25], experimental results have been quite sparse [26–32]. Most significantly because photon statistics enhancement due to squeezed (superbunched) light has been observed for the first time, a development that creates an opportunity for the experimental investigation of a number of challenging open problems.

It may be useful to clarify at the outset the relevance of the term superbunched to this work, in relation to other properties of squeezed light. Following standard terminology [33–35], bunched light implies that the N th-order intensity correlation function is larger than unity. For chaotic (thermal) light the normalized intensity correlation function $g_N(0)$ is equal to $N!$, which is what is usually implied by bunched light. If a state

of light exhibits a value of the N th-order intensity correlation function larger than $N!$, it is referred to as superbunched, which is the case with squeezed light. However, squeezed light has many other features, notable among them being that it is nonclassical. Those other properties, interesting as they may be, are of no relevance to the effects discussed in this paper. Actually, any source of light, classical or otherwise, that can exhibit intensity fluctuations stronger than those of a coherent state will lead to the same effects, which result from an interplay between nonlinearity and fluctuations. That is why, in earlier work on the theory of bound-bound transitions driven by stochastic fields [20,21], the fields were modeled as random Gaussian variables, producing the same effects that quantized fields would. Further discussion of that aspect is deferred to Sec. IV.

Owing to computational demands, the numerical results in [1], illustrating the involvement of near resonances and the ensuing modification of the bunching effect with increasing intensity, were obtained with scaled intensity and atomic parameters such as Rabi frequencies and ionization cross sections. The need for scaling arose from the necessity of formulating the problem in terms of a quantized field, which then led to summations over the photon-number distributions of the various states of the field we examined. The concomitant drawback of that approach is the difficulty in translating photon numbers to the intensity of a pulsed source, which is what occurs in an experiment [2]. Our purpose in this article is to provide a bridge between the two, through a sample of realistic calculations in atomic systems.

For that purpose, we have chosen the cases of two-photon ionization in cesium in the vicinity of the intermediate $7p$ state and three-photon ionization in sodium in the vicinity of two-photon intermediate resonance with the $4d(3/2)$ and $4d(5/2)$ states. It is common knowledge that, for a nonresonant N -photon process, the ionization yield as a function of laser intensity, in a log-log plot, will be a straight line of slope N [3]. When, with increasing

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intensity, an intermediate state near resonance begins playing a role, the slope begins diminishing. The intensity at which this change of slope becomes noticeable is the intensity at which the bunching enhancement will also begin decreasing.

The most convenient formal tool for the calculation of the dependence of ion yields on intensity is the time-dependent density matrix of the atomic system, driven by a classical electromagnetic (em) field. This formulation, in addition to allowing for realistic calculations which include the source temporal pulse shape, also allows for the incorporation of the laser bandwidth, an experimentally important aspect not accessible in the formalism of [1]. Evaluating the role of the bandwidth addresses the possibility that, even with the carrier frequency tuned far from resonance, the wings of the spectral shape of the pulse may involve real excitation of a near-resonant state, thereby altering the slope and therefore the enhancement due to bunching.

A short clarification of the issue of bandwidth is needed at this point. The bandwidth of a pulsed source involves two separate contributions. One is due to the finite duration of the pulse and is referred to as the Fourier bandwidth. The second is due to stochastic phase and/or amplitude fluctuations of the field, the latter being the cause of bunching. Depending on the duration of the pulse, the stochastic bandwidth may become important and therefore would have to be included in the formulation. For the sake of completeness, we have included the effects of the stochastic bandwidth in our calculations.

For pulses of extremely short duration of the order of a few femtoseconds, it is the Fourier bandwidth that dominates. For longer pulses of hundreds of femtoseconds or longer, it is the stochastic bandwidth that dominates, which is in fact the case with sources in which bunching is significant [2]. In the examples discussed in this paper, the pulses we have chosen are of relatively long duration, so it is the stochastic bandwidth that dominates. Nevertheless, the Fourier bandwidth is inherently included in the calculation, through the solution of the time-dependent density-matrix differential equations.

In order to avoid repetition of formal aspects and derivations readily available in the literature [36], we have chosen to simply provide the basic differential equations governing the time evolution of the density matrix for the three-photon process, as a special case of which the two-photon process can be obtained. After a brief discussion and explanation of the basic equations in Sec. II, a collection of illustrative examples with discussion and conclusions are presented in Sec. III.

II. THEORY

Consider the atom to be initially in its ground state, denoted by $|1\rangle$, in the presence of an external electric field of the form $E(t) = \mathcal{E}(t)e^{i\omega t} + \text{c.c.}$, where ω is the frequency of the field and $\mathcal{E}(t)$ its complex amplitude. The field amplitude can be expressed as $\mathcal{E}(t) = |\mathcal{E}(t)|\exp[i\varphi(t)]$, where both $|\mathcal{E}(t)|$ and $\varphi(t)$ can in general be stochastic quantities, owing to fluctuations of the field. The absorption of two photons excites

the atom to the vicinity of an intermediate state $|2\rangle$, whose energy is denoted by $\omega_2(\hbar = 1)$. In addition to the coherent coupling between the ground and excited states induced by the external field, the state $|2\rangle$ can either decay spontaneously or be ionized by a single-photon absorption, with a rate denoted by $\Gamma(t)$. When state $|2\rangle$ is connected to the initial state by a two-photon transition, spontaneous decay back to $|1\rangle$ is allowed only through a cascade. For the sake of completeness, we include in our equations that rate in the sense of an effective rate denoted by γ . In any case, for the type of experiments pertaining to our problem, that rate is too small to be of relevance. The excited state $|2\rangle$, in addition to its coupling to the initial state and to the continuum, also undergoes a Stark shift through virtual transitions to all dipole-allowed bound and continuum states. It is denoted here by $S(t)$, where the time dependence is due to the fact that the shift is proportional to the instantaneous intensity. Since in this case the two-photon Rabi frequency and the ionization rate are also proportional to the intensity, the shift needs to be included in the formalism.

If ρ_{ij} , $i, j = 1, 2$, are the matrix elements of the density matrix of our effective two-level model of the three-photon process, in the rotating-wave approximation, then the differential equations governing the time evolution of the respective slowly varying matrix elements, defined by $\rho_{ii}(t) = \sigma_{ii}(t)$, $i = 1, 2$, and $\rho_{12}(t) = \sigma_{12}(t)e^{i\omega t}$, are [1]

$$\frac{\partial}{\partial t}\sigma_{11}(t) = \gamma\sigma_{22}(t) + 2\text{Im}[\Omega_{12}^*(t)\sigma_{12}(t)], \quad (1)$$

$$\frac{\partial}{\partial t}\sigma_{22}(t) = -[\gamma + \Gamma(t)]\sigma_{22}(t) - 2\text{Im}[\Omega_{12}^*(t)\sigma_{12}(t)], \quad (2)$$

$$\begin{aligned} \frac{\partial}{\partial t}\sigma_{12}(t) &= \{i[\Delta - S(t)] - \gamma_{12}(t)\}\sigma_{12}(t) \\ &+ i\Omega_{12}(t)[\sigma_{22}(t) - \sigma_{11}(t)], \end{aligned} \quad (3)$$

where $\Delta = 2\omega - (\omega_2 - \omega_1)$ is the detuning from the intermediate resonance, $\gamma_{12}(t) = \frac{1}{2}[\gamma + \Gamma(t)]$ is the off-diagonal relaxation, and $\Omega_{12}(t) = \hbar^{-2}\mu_{12}\mathcal{E}^2(t)$ is the effective two-photon Rabi frequency of the $|1\rangle \leftrightarrow |2\rangle$ transition, given by the product of the effective two-photon matrix element μ_{12} of the dipole operator and the square of the electric-field amplitude. All parameters depending on the applied em field, namely, the ionization rate, the Stark shift, and the Rabi frequency, are in general stochastic quantities, as they are subjected to the fluctuations of the field. As a result, the equations of motion of the density operator's matrix elements are also stochastic. The next step is to average the differential equations (1)–(3) over the field fluctuations, denoting such averaged quantities by angular brackets. In the process of averaging the above set of differential equations, we encounter atom-field products of the form $\langle \Gamma(t)\sigma_{ij}(t) \rangle$, $\langle \Omega_{12}(t)\sigma_{ij}(t) \rangle$, etc., $i, j = 1, 2$. The rigorous evaluation of such products requires the exact model of the stochastic properties of the field, entailing considerable mathematical complexity, which is beyond the scope of this paper. However, a detailed treatment of how this can be accomplished can be found in Refs. [20,36]. Since our purpose in this paper is to simply assess the effect of the laser bandwidth, in an approximate fashion, we adopt the decorrelation approximation, which amounts to replacing the stochastic averages of products such as those above by the

products of their averages. Assuming that the deterministic real field amplitude is constant and equal to \mathcal{E}_0 (square pulse

shape), the resulting differential equations governing the time evolution of the averaged density-matrix elements are

$$\frac{\partial}{\partial t} \langle \sigma_{11}(t) \rangle = \gamma \langle \sigma_{22}(t) \rangle + 2 \operatorname{Im} \left\{ i \bar{\Omega}_{12}^2 \int_0^t dt' e^{i[\Delta - \langle S \rangle - \langle \tilde{\gamma}_{12} \rangle](t-t')} [\langle \sigma_{22}(t') \rangle - \langle \sigma_{11}(t') \rangle] \right\}, \quad (4)$$

$$\frac{\partial}{\partial t} \langle \sigma_{22}(t) \rangle = -[\gamma + \langle \Gamma \rangle] \langle \sigma_{22}(t) \rangle - 2 \operatorname{Im} \left\{ i \bar{\Omega}_{12}^2 \int_0^t dt' e^{i[\Delta - \langle S \rangle - \langle \tilde{\gamma}_{12} \rangle](t-t')} [\langle \sigma_{22}(t') \rangle - \langle \sigma_{11}(t') \rangle] \right\}, \quad (5)$$

where $\langle \tilde{\gamma}_{12} \rangle \equiv \langle \gamma_{12} \rangle + \gamma_L = \frac{1}{2}(\gamma + \langle \Gamma \rangle + 2\gamma_L)$ and $\bar{\Omega}_{12} \equiv \hbar^{-1} \mu_{12} \mathcal{E}_0^2$ is the average Rabi frequency of the $|1\rangle \leftrightarrow |2\rangle$ transition.

Note that $\langle \Gamma(t) \rangle$ and $\langle S(t) \rangle$ have been replaced by $\langle \Gamma \rangle$ and $\langle S \rangle$, respectively, since we assumed that the deterministic field amplitude is constant. The above averaged equations contain the additional term γ_L in the off-diagonal relaxation, which represents the bandwidth of the radiation source. This result is the consequence of relations [33–35] that connect higher-order field correlation functions to the second-order field correlation function

$$\langle \mathcal{E}(t_1) \mathcal{E}^*(t_2) \rangle = \langle \mathcal{E}^2 \rangle \exp \left[-\frac{1}{2} \gamma_L |t_1 - t_2| \right], \quad (6)$$

where $\langle \mathcal{E}^2 \rangle$ is the variance of the electric field. Such field correlation functions appear in the process of averaging over the field's stochastic fluctuations and introduce the laser bandwidth in our model. The solution of the equations of motion of the averaged density-matrix elements provide the probability of ionization through the expression

$$P(t) = 1 - \langle \sigma_{11}(t) \rangle - \langle \sigma_{22}(t) \rangle, \quad (7)$$

which can be obtained either numerically or analytically with the use of Laplace transform, if the field amplitude is not pulsed but can be assumed constant.

We are particularly interested in the behavior of the ionization probability as a function of the intensity for various detunings from the intermediate resonance. In order to provide results pertaining to a realistic model, we apply our theory to the $3s \rightarrow 4d \rightarrow$ continuum process in Na, as described in the next section.

The above theoretical model, with minor modifications, reduces to the case of two-photon near-resonant ionization. The form of the density-matrix equations is the same. Since the coupling between the initial and excited states is mediated by a single-photon transition, the Rabi frequency now is proportional to the field amplitude and not the intensity. For the same reason, the relaxation constant of the off-diagonal matrix element is now given by $\tilde{\gamma}_{12} = \frac{1}{2}[\gamma + \langle \Gamma(t) \rangle + \gamma_L]$, where the laser bandwidth γ_L is not multiplied by 2, as it was in the previous three-photon case. As discussed in the next section, the model is applied to the realistic two-photon process $6s \rightarrow 7p \rightarrow$ continuum process in Cs.

III. RESULTS AND DISCUSSION

In this section we present quantitative results from the application of our theoretical model to the two realistic processes described above, namely, the $3s \rightarrow 4d \rightarrow$ continuum transition in Na and the $6s \rightarrow 7p \rightarrow$ continuum transition in Cs.

The atomic parameters [36,37] obtained through quantum-defect theory are, for the Na transition, $\omega_{21} = 4.2845$ eV, $\Gamma = 9.4I$, $\Omega = 1.46 \times 10^3 I$, and $S = 179I$, while for the Cs transition, $\omega_{21} = 2.665$ eV, $\Gamma = 11I$, and $\Omega = 0.75 \times 10^7 \sqrt{I}$, where I is the field intensity, in units of W/cm^2 , and Γ , Ω , and S are in units of Hz. Note that the Stark shift for a $(1+1)$ -photon process, such as the $6s \rightarrow 7p \rightarrow$ continuum transition, and the spontaneous decay rates for both transitions in the range of intensities considered are negligible and can be neglected. The laser bandwidth used in our calculations is $\gamma_L = 2 \times 10^{13}$ Hz.

The illustrative results are summarized in Figs. 1–4. It should first be noted that the pulse duration is an additional parameter affecting the slope of the ionization signal as a function of intensity. Obviously, for any peak intensity, if the pulse duration is sufficiently long, complete ionization will occur. In that limit, the slope will become zero, which will eventually be reached through a gradual decrease of the slope with increasing intensity, as shown in Figs. 1 and 2. A similar behavior is shown in Figs. 3 and 4, with the difference that the slope becomes zero for intensities below the complete ionization regime. This indicates that higher-order processes are more prone to depart from the nonresonant condition as we increase the intensity, compared to lower-order ones.

Aside from this difference, the feature shared by all cases is that for larger detuning (dotted as compared to solid lines) from the intermediate resonance, the unperturbed slope reflecting the overall order of the process persists for higher intensities. This behavior reflects the onset of the effect of the

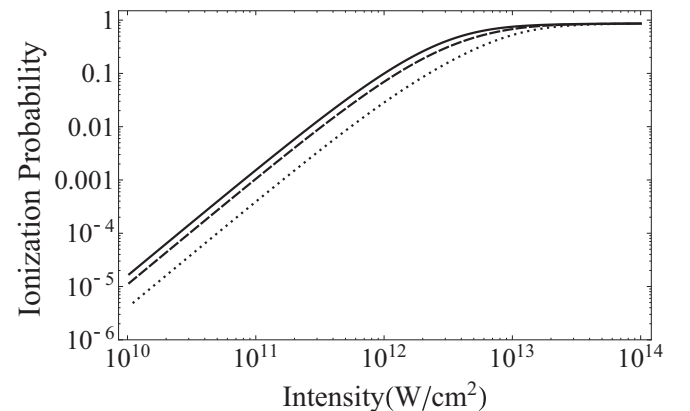


FIG. 1. Ionization probability of Cs as a function of the intensity for various detunings from the intermediate resonance and $T = 100$ fs. The solid line shows $\Delta/\omega_2 = 0.01$, the dashed line $\Delta/\omega_2 = 0.05$, and the dotted line $\Delta/\omega_2 = 0.1$.

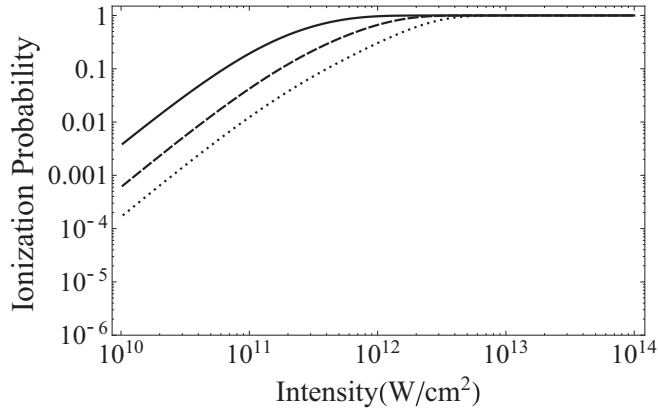


FIG. 2. Ionization probability of Cs as a function of the intensity for various detunings from the intermediate resonance and $T = 1$ ps. The solid line shows $\Delta/\omega_2 = 0.01$, the dashed line $\Delta/\omega_2 = 0.05$, and the dotted line $\Delta/\omega_2 = 0.1$.

increasing Rabi frequency, signaling the onset of departure from the notion of a nonresonant process. It is precisely the effect which in Ref. [1] was shown to herald the distortion of the photon statistics enhancement, although the process nominally is nonresonant. Therefore, the message emerging from these results is that up to 10^{10} W/cm² or so, it can be assumed that the nonresonant behavior will persist. This is the type of assessment that served as the motivation for this paper.

It could be argued that the results may be of limited usefulness as they pertain to specific atomic transitions. However, they may not be as limited as they might seem at first sight. For one thing, parameters such as matrix elements in nonlinear transitions do not differ by orders of magnitude. Cognizant of the specificity of our results, we do not claim exact limiting values of the intensities, but only a range of intensities. The validity of that range is further underscored by comparison with the results of the recent data by Spasibko *et al.* [2], who observed nonresonant slopes in harmonic generation up to

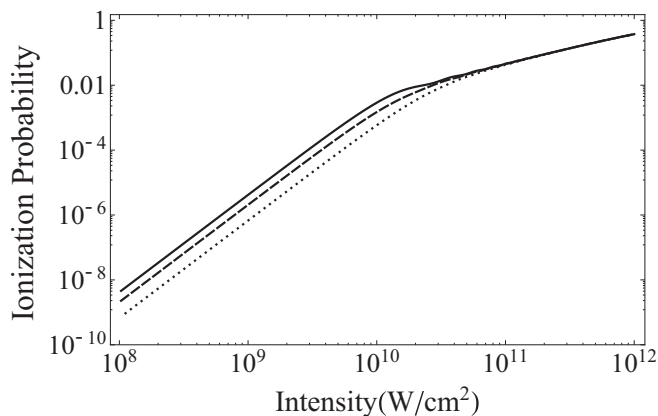


FIG. 3. Ionization probability of Na as a function of the intensity for various detunings from the intermediate resonance and $T = 100$ fs. The solid line shows $\Delta/\omega_2 = 0.01$, the dashed line $\Delta/\omega_2 = 0.05$, and the dotted line $\Delta/\omega_2 = 0.1$.

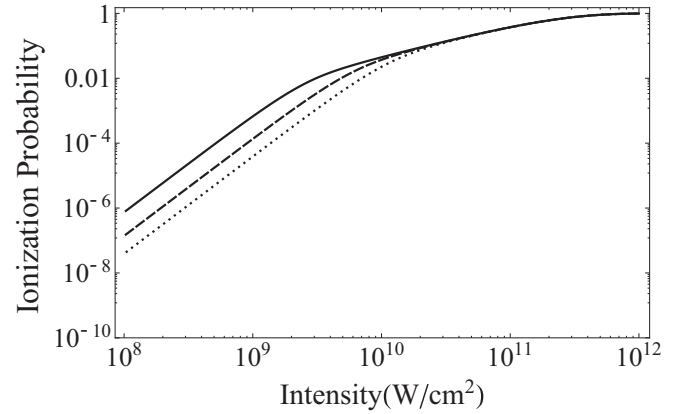


FIG. 4. Ionization probability of Na as a function of the intensity for various detunings from the intermediate resonance and $T = 1$ ps. The solid line shows $\Delta/\omega_2 = 0.01$, the dashed line $\Delta/\omega_2 = 0.05$, and the dotted line $\Delta/\omega_2 = 0.1$.

order 4, for intensities in the above range, in a completely different material.

In view of the above estimates, in combination with the results in [2], and the dramatic progress in the potential for observation of nonlinear processes induced by squeezed light, it appears that novel effects may be observed under intensities in the range of 10^9 – 10^{10} W/cm², as it is intensities in that range that are needed for the observation of such processes and are luckily becoming available even for squeezed radiation.

IV. CLOSING REMARKS AND OUTLOOK

As explained in Sec. I, the chief motivation for this paper was to provide a quantitative assessment of the intensities needed for the observation of a nonlinear process such as few-photon ionization or harmonic generation, which would enable the exploration of the interplay between stochasticity and nonlinearity. To the extent that such a process is basically nonresonant, with nearby resonances causing only modest modification of the dependence of the yield on light intensity, the yield is proportional to the intensity correlation function reflecting the order of the process. Further, in the presence of bunching due to intensity fluctuations, the yield is in that case enhanced. In contrast, the behavior of two discrete states driven by a stochastic field depends on correlation functions of all orders, which entails a much more complex and occasionally counterintuitive behavior [20].

As long as the process depends on a single intensity correlation function, it does not matter whether the source is treated classically or quantum mechanically. The experimental observation of nonlinear processes, such as those discussed in this paper, demand sufficiently high intensity. The surprising achievement of the experiment by Spasibko *et al.* [2] is that their source of squeezed light did muster the intensity necessary for the observation of even fourth harmonic. Recent developments [38–41] have led to the production of superbunched radiation, exhibiting values of intensity correlation functions larger than those of thermal light; however, the intensity available in those sources does

not seem to be sufficiently high for the driving of nonlinear transitions. It is possible that future developments may remove that barrier.

Still, at much lower intensities, the strong driving of bound-bound transitions may be feasible, provided bunched or superbunched radiation at the appropriate wavelength were available, which may be the stumbling block for the moment. Near infrared to optical is the desired range of wavelengths. As already mentioned above, a much richer variety of effects is to be expected in that context. For example, a number of hitherto unanticipated effects can be expected in a two-photon bound-bound transition driven by superbunched radiation, still an open theoretical problem. A glimpse of possible surprises can be speculated in the light of theoretical results for thermal as well as squeezed light [20,42], which have been in print for about 30 to 35 years now. The development

of new sources may finally make their observation feasible, thus presenting the opportunity for the exploration of hitherto unknown territory in stochastic processes.

Having stated above that near infrared to optical is the desirable wavelength range for the study of field stochastic effects on nonlinear photon-atom processes, we should not fail to mention another possibility, amenable through short-wavelength free-electron lasers, which are known to exhibit strong intensity fluctuations, akin to those of chaotic radiation [43–46]. Although for many applications such fluctuations may be a nuisance, still they offer a hitherto experimentally unexplored territory for the interplay between nonlinearity and stochasticity, at short wavelengths extending to x rays. A glimpse of expected effects, in the context of a strongly driven and hence nonlinear Auger transition, has been given in [46].

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