Superradiance of an ensemble of nuclei excited by a free electron laser

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In 1954 Dicke predicted the accelerated *initial decay* of multiple atomic excitations [1], laying the foundation for the concept of superradiance. Further studies [2-4] suggested that emission of the *total energy* was similarly accelerated, provided that the system reaches the inversion threshold. Whereas, superradiant emission of the total energy has been confirmed by numerous studies [4-12], the acceleration of the *initial decay* has not yet been experimentally demonstrated. Here we use resonant diffraction of x rays from the Mössbauer transition [13] of ^{57}Fe nuclei to investigate superradiant decay, *photon by photon*, along the entire chain of the de-excitation cascade of up to 68 simultaneous coherent nuclear excitations created by a pulse of an x-ray free-electron laser. We find agreement with Dicke's theory [1] for the accelerated initial decay as the number of excitations is increased. We also find that our results are in agreement with a simple statistical model, providing a necessary baseline for discussing further properties of superradiance, within and beyond the low-excitation regime.

Dicke's model introduces superradiance as an accelerated *initial decay* of multiple atomic excitations, and provides exact predictions for the ensemble behavior as a function of the number of atoms and number of excitations in the system [1]. It considers superradiant states emerging from incoherent excitation of a system of a size much smaller than the radiation wavelength, or created by coherent excitation of an extended system, and predicts for these cases an identical initial decay [1]. More specifically, Dicke gave an explicit closed form expression for the accelerated decay of the *first photon* of the de-excitation cascade of a multiply excited system (see eqn (24) and figure 1 of [1])

The predicted behavior of the initial decay differs qualitatively from the emission of the *total energy*, which exhibits a threshold behavior. In the low-excitation regime, the emission of the total energy follows a conventional single-photon decay; approaching inversion conditions allows for an

amplified spontaneous emission; and inverted systems can have an accelerated superradiant decay [2-9].

In contrast, the accelerated initial decay appears without threshold, i.e., it is expected for even small numbers of excitations. The application of Dicke's formulation for "maximally radiative" systems with cooperation number $r=n_a/2$ (see [1]), gives the initial decay rate I_N of a system containing N photons and n_a resonant "atoms" should be accelerated, relative to the single photon (N=1) response I_1, by a factor [1]

$$I_N/I_1 = N\,(1- N/n_a +1/n_a)\,. \qquad (1)$$

Even when $N << n_a$, this is a very large, N-fold, acceleration. Therefore, the acceleration of the initial decay predicted by Dicke can be studied even in the low-excitation regime.

We studied the accelerated initial decay of multiple coherent nuclear excitations created by an x-ray pulse of the SPring-8 Angstrom Compact free electron LAser (SACLA) [14], the only source that can presently provide temporally and spatially coherent pulses of many photons within the bandwidth of the convenient 14.4 keV nuclear transition of ^{57}Fe. For x rays, the small-system limit is fundamentally excluded because the wavelength is similar to interatomic distances. However, one can create a phased excitation of an extended system. The ideal "*x-ray lattice*" [10] is offered by atomic periodicity, and the "*seeded coherence*" [12] is provided in nuclear resonance diffraction conditions [15]. Similar to the "*collective dipole*" of atoms coupled to the light field in an infrared optical cavity [10-12], the "*compound*" excited state [16, 17] of an ensemble of nuclei under diffraction [15] or forward scattering [18, 19] conditions leads to enhancement of emission and strong "*speedup*" of the collective response [15-23]. In contrast to the optical regime, the theory of the quantum-mechanical ensemble of nuclei and x rays [20-23] is relatively tractable, since for short wavelength the system is in the limit of dynamical diffraction [24]. This brings the spatial aspect of theory to the utmost clarity and allows for excellent agreement of closed-form theory with nearly all observations [15, 18, 19, 23, 25].

The high energy of x rays and the long lifetime of nuclear levels allows us, using a fast detector, to follow, *photon by photon*, the decay of multiple excitations along the entire chain of the de-excitation cascade. Thus, this first application of an x-ray free electron laser (XFEL) to nuclear resonance scattering provided direct experimental access to the process matching Dicke's original calculation.

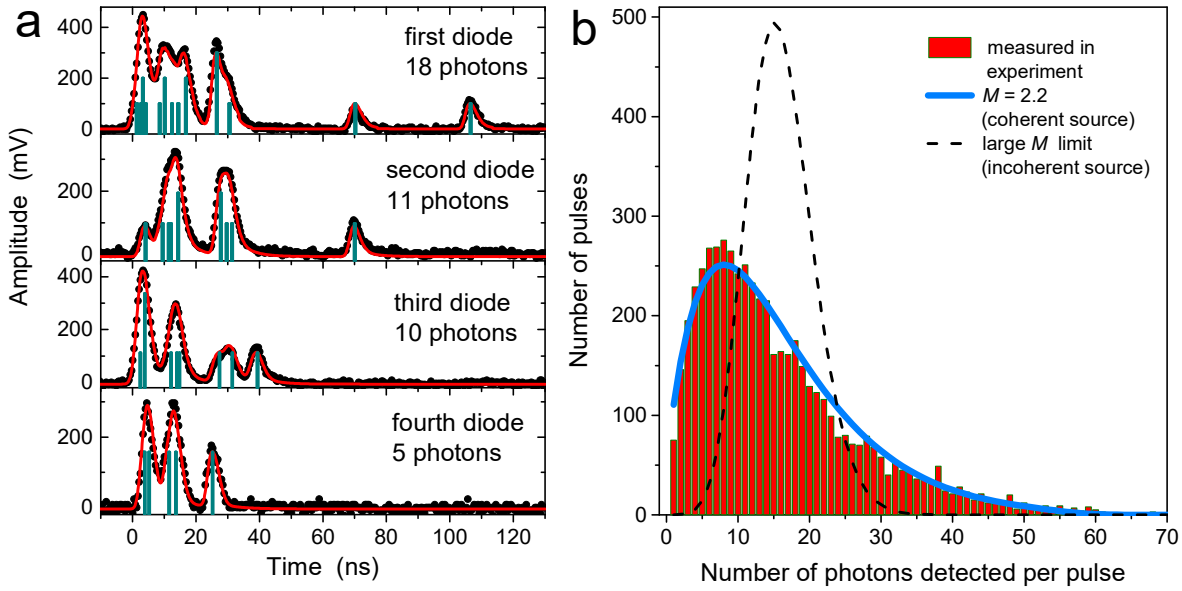

Figure 1: Measuring the multi-photon response. (a) Scope traces from the APD detectors after one pulse of 44 photons and the fits used to analyze the distribution. (**b**) Distribution of multi-photon events measured in the APD detectors, as compared to a model incorporating a coherent source with few modes (M=2.2) and an incoherent source (large M limit). See also discussion in the text and the supplemental materials.

The experimental scheme (see *Methods*) used monochromatized pulses from the SACLA x-ray free electron laser incident on a single crystal of $^{57}FeBO_3$ at the Bragg condition for the (111) pure nuclear reflection [25]. The diffracted photons were measured by 4 semi-transparent avalanche photo-diode (APD) detectors [26] placed in series. Scope traces from each detector were recorded and fit with a sum of single photon responses allowing us to determine the time of arrival of each photon after the exciting pulse in each detector (**Fig. 1a**). For the most intense pulses, the data allow for a single shot hyperfine spectroscopy, giving an access to Mössbauer spectroscopy with femtosecond time resolution (see the *Supplementary Material* for the details). **Figure 1b** shows the measured distribution of the detected multi-photon events summed over all APDs. The good agreement of the data with a distribution assuming a small number of modes (M=2.2) serves to confirm the coherence of the XFEL radiation (see also the *Supplementary Material*).

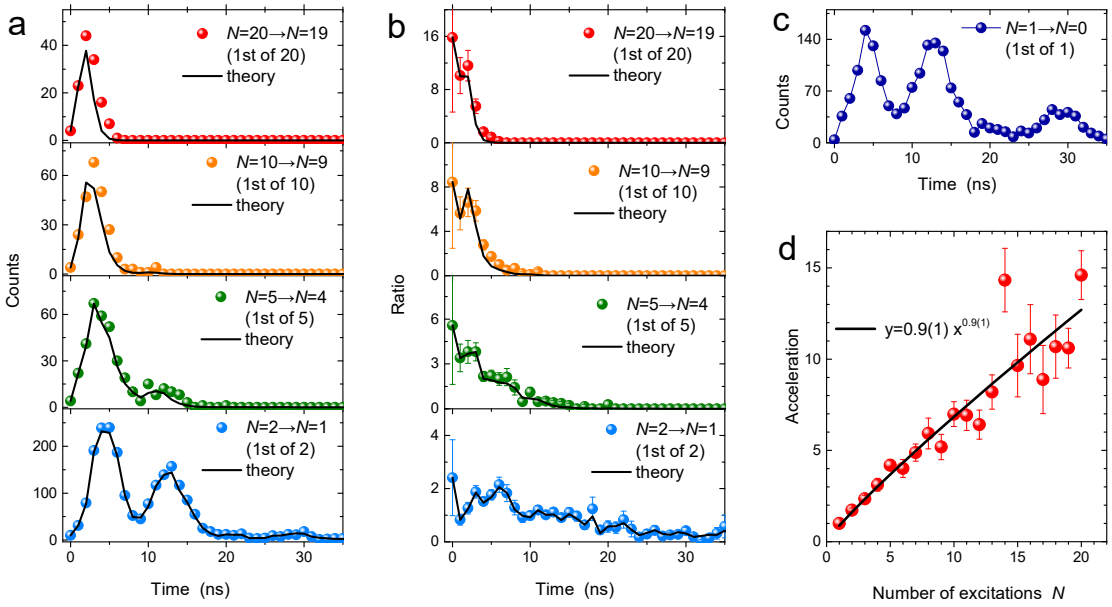

Figure 2: Initial decay rate. The increase of the initial decay rate for the transitions from N to N-1 excited states revealed **(a)** by the accelerated decay of the first out of N detected photon, $P_N^1(t)$, **(b)** by the ratios $P_N^1(t)/P_1^1(t)$ of these data to the single-photon decay $P_1^1(t)$ (shown in **(c)**), and **(d)** by the estimated (*Supplementary Materials*) acceleration rates $(P_N^1/P_1^1)|_{t\to 0}$. The solid lines in **(a, b)** are the calculations according to eq.(2). The solid line in **(d)** is the power fit. The error bars show the standard deviations related to the numbers of counts per channel in the raw data (**b**) and to the uncertainty of the linear fit to the ratios $P_N^1(t)/P_1^1(t)$ (*Supplementary Materials*).

Figure 2 presents the main experimental results of our investigation. The left panel, **(a)** shows the distribution of the times of arrival of the first photon for various total numbers N of photons detected after the pulse. The complex modulations of the decay are quantum beats from the magnetic hyperfine splitting of the nuclear levels [27] (see also *Supplementary Materials*), as has also been observed in superradiant lasing on multiple optical transitions [12]. Faster decay of the first photon for events with larger numbers of photons is immediately evident in the raw data **(a)**, and is more clearly seen after the division **(b)** by the single-photon decay **(c)**. The estimated (*Supplementary Materials*) y-intercept of the data in **(b)** gives the acceleration rate $(P_N^1/P_1^1)|_{t\to 0}$ shown in (d). Within statistics, this is quantitatively in agreement with Dicke's [1] N-fold acceleration of the transitions from the N to N-1 excited states, as described by eq. (1).

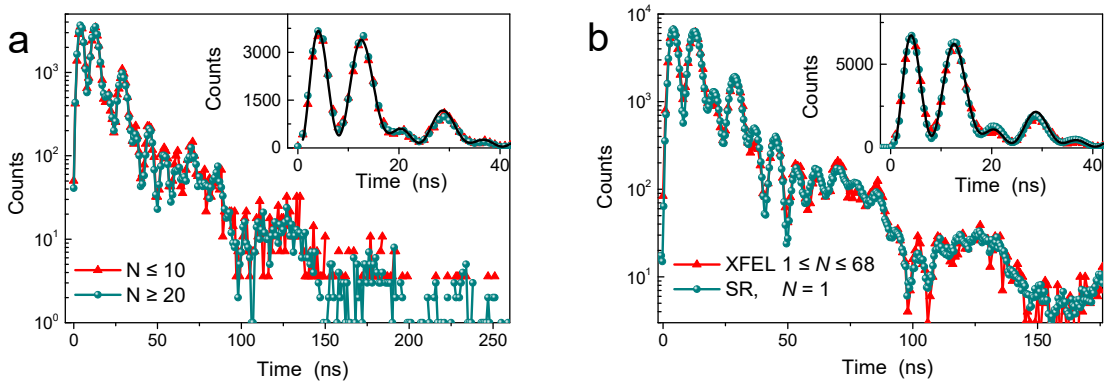

Figure 3: Emission of the total energy. (a) The decay including all photons for the events where either N≤10 or N≥20 photons were detected. These are the same within statistics. **(b)** The decay for over all photons measured at the XFEL with multi-photon events is the same as what is measured at a synchrotron radiation beamline with single photon events. The solid line in inserts in **(a, b)** is theory based on [22] showing that decay of the nuclear ensemble can be calculated with great accuracy for single-photon excitations.

Figure 3, **(a)** and **(b)**, confirm that, as expected for the low-excitation regime [2, 28], the decay of the total energy in the system remains un-affected by the number of photons. Panel **(a)** compares events with low or high number of detected photons at the XFEL, while **(b)** compares a single-photon pulse of incoherent synchrotron source with the multi-photon pulse of coherent XFEL. The decay, in all cases, remains essentially identical to the 1-photon distribution. Thus, we observe a strongly accelerated *initial decay*, as expected from Dicke's work [1], and no change in the shape of the decay of the *total energy*, as expected for a system far from inversion [2, 28].

We are able to interpret these results, the accelerated decay of the first photon and the unchanged decay of the total energy, by considering statistical properties of independent events. We assume the time response of our system after excitation by a pulse containing a single photon is given by a function *f(t)* so that *f(t)dt* is the probability of observing the photon to be re-emitted in an interval *dt* about time *t* after excitation. Then, assuming the system is excited with *N* photons, the probability of finding the *K*-th photon at a time *t* is given by the product of the probability of observing any single photon at time *t*, the probability that there are *K*-1 photons observed before time *t*, and the probability that there are *N*-*K* photons observed after time *t*. This is seen to be (see *Supplementary Materials*)

$$P_N^K(t)/f(t) = K \cdot C_N^K \cdot \left[\int_0^t f(t')dt'\right]^{K-1} \cdot \left[\int_t^\infty f(t')dt'\right]^{N-K}, \qquad (2)$$

where C_N^K is the binomial coefficient.

The solid lines in **Figure 2a** and **Figure 2b** show the calculated $P_N^1(t)$ and $P_N^1(t)/f(t)$, respectively, for several values of *N*, and one finds that the agreement of the data with this simple approach is quite good. The calculations are performed without any adjustable parameter. The probability $f(t)$ is given by the measured single-photon decay (**Fig. 2c**). The vertical scaling is given by the total number of the observed excitations with a given *N*. The model also correctly describes

the behavior of each intermediate transition in the entire cascade (*Supplementary Materials*). For an exponential shape of the single-photon decay $f(t) = exp(-t)$, eq.(2) gives an exact *N*-fold acceleration

$$P_N^1(t) = N \exp(-Nt) ,$$

which agrees with the Dicke's result [1].

Further, the equation is consistent, both considering the derivation (assumption of independent events) and by explicit evaluation (see *Supplementary Materials*), with the fact that the time distribution of the total power is the same as that of the single photon response, namely,

$$\sum_{K=1}^{N} P_N^K(t) \; = \; N \;\; f(t). \tag{3}$$

The agreement of the data with the model suggests that the multi-photon emission is straight-forwardly related to the single-photon response, independent of its possible complexity, if the number of photons in the system is much less than the total number of atoms. The response is the product of a potentially complicated single photon response, *f(t)*, and the simple statistics given by eq.(2). As the number of photons becomes comparable to the number of atoms or nuclei, we expect this approximation may fail. But, for small numbers of photons, this provides an accurate approach to understand the system behavior. This regime will be a typical case for pump-probe nuclear resonance experiments with ~femto-second time resolution (see *Supplementary Materials*) and, possibly, for emerging superradiant laser with ultra-narrow line widths [10-12].

Conceptually, our results show that the statistical aspect of multiple excitations provides an important baseline for discussing further properties of superradiance. For our system, it properly describes all experimental data, i.e., each intermediate transition in the entire cascade. For the case considered by Dicke [1], exponential single-photon decay, it also properly gives the *N*-fold acceleration. Within the limits of this study, these statistical properties are indistinguishable from the superradiant response of systems. One may need to move beyond this simple approach when one reaches regimes of higher excitation, as may occur at future facilities. In this sense, this study shows that the appearance of the higher-excitation effects has to be traced not in the *N*-fold acceleration of the initial decay, but rather in deviations from this trend. Thus, in order to correctly identify when new concepts may be needed to understand the results, it is essential that the system's response should also be compared to a statistical model.

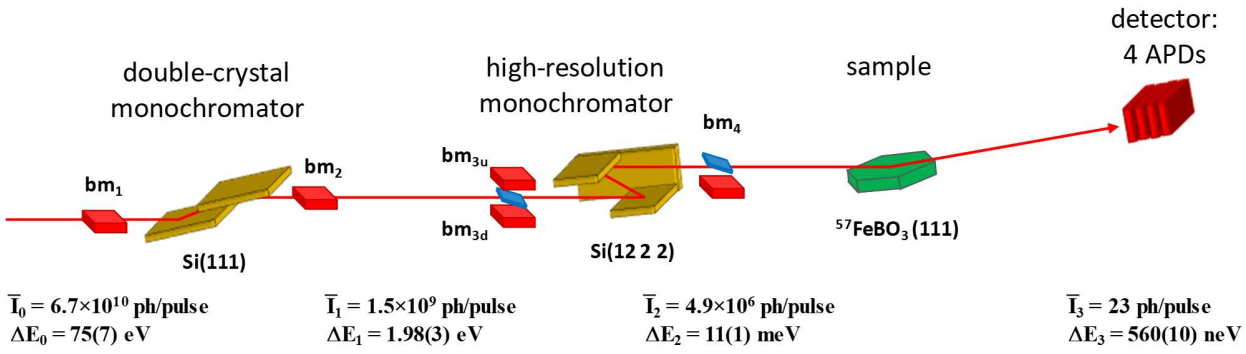

Figure 4: The experimental setup and the parameters of the x-ray beam. The double-crystal monochromator consists of two separate silicon crystals set in symmetric Si(111) reflections. The high-resolution monochromator is a silicon channel-cut crystal using two asymmetric Si(12 2 2) reflections. The sample is an iron borate $^{57}FeBO_3$ single crystal (green) set in a pure nuclear (111) symmetric reflection. The beam monitors bm_1 and bm_2 are the quadrant photo-diodes detecting scattering of the x-ray beam passing the nano-diamond foil. The beam monitors bm_{3u}, bm_{3d}, and bm_4 are the single photo-diodes detecting scattering of the x-ray beam passing through Kapton foils (blue in the figure). The detector is composed of four semi-transparent avalanche photo diodes (APDs) placed in series. $\bar{I}_0 - \bar{I}_3$ and ΔE_0 - ΔE_3 are the experimentally determined mean intensities and energy bandwidths (FWHM), respectively, of the x-ray beam in four points of the optical scheme.

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Methods.

The SPring-8 Angstrom Compact free electron LAser provides hard x-ray pulses in the energy range of 4–20 keV. For this experiment, the photon energy of SACLA was tuned to the 14.412 keV energy of the nuclear transition in ^{57}Fe isotope. The laser was operated with the repetition rate of 30 Hz. The mean pulse energy of SACLA was 155 μJ/pulse, giving an average of 6.7×10^{10} photons per pulse in a 75 eV bandwidth. The fluctuations in intensity and flux within 30 shots were about 13% (standard deviation from the mean).

The experimental setup and the experimentally determined intensities and energy bandwidths of the beam in various points of the optical scheme are shown in **Figure 4**. All reflections were set up in the vertical geometry of scattering given the undulator provides horizontally polarized light. The double crystal monochromator and beam monitors bm_1 and bm_2 (standard SACLA components) are located in transport channel, the rest of the equipment was set up in the experimental hutch EH2 (see *Supplementary Materials*). The detailed description of the SACLA equipment may be found in Ref. [29].

The double-crystal monochromator with an energy bandwidth of ~2 eV delivered an average of 1.5×10^9 photons per pulse with a time duration of about 5 fs. If such a beam would incident directly on the sample, the sample experienced a recoil force of about 30 mN. In order to avoid this mechanical disturbance and to decrease the heat load on the sample, the x-ray beam after the double-crystal monochromator was further monochromatized down to the bandwidth of 11 meV by a high-resolution monochromator consisting of a channel-cut asymmetric Si (12 2 2) reflection.

The sample is a high-quality iron borate $^{57}FeBO_3$ single crystal enriched in resonant ^{57}Fe isotope by 95%. The crystal has the shape of a platelet with the size of ~3×5 mm^2 and thickness of ~35 μm. The surface of the platelet is parallel to (111) atomic planes. The crystal was kept at the temperature of the SACLA experimental hall (28°C).

Iron borate crystal is a canted antiferromagnet. The hyperfine magnetic fields on two iron atoms of the unit cell are aligned in almost opposite directions. The magnetic fields on both atoms and a small residual ferromagnetic moment lie in (111) plane [30]. In order to obtain a single magnetic domain state of the crystal with a well-defined orientation of the hyperfine magnetic field, the crystal was magnetized by an external magnetic field of ~120 Oe perpendicular to the scattering plane. Under these conditions, the hyperfine magnetic fields on iron atoms is aligned almost in the scattering plane.

The detector consists of four EG&G Optoelectronics reach-through avalanche photo diodes [26] each with an area of 5×5 mm^2 and thickness of ~110 μm. The efficiency of a single diode is ~25%, the total efficiency of four diodes in series is ~69%. The time resolution (FWHM) of the diodes is about 1 ns. The analogue outputs of all four diodes were digitized in parallel using four channels of Tektronix DPO3054 oscilloscope with a bandwidth of 500 MHz and sampling rate of 2.5 GSPS. The trigger for the oscilloscope was delivered by the SACLA bunch-timing system. Traces were stored manually.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding authors on reasonable request.

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Author contributions

The experiment was conceived by AIC in discussion with TI and AQRB and organized by AIC. Specific equipment was prepared by GVS, AIC, AQRB, CS, OL, YI, KT, TK and MY. Experiments were done by AIC, AQRB, IS, CS, OL, YS, RR, YI, KT, and MY. Data analysis was done by AIC, CS, IS and AQRB, with IS proving eq. (2). All authors discussed the results. The paper was written by AQRB and AIC with input from all authors.

Additional information

Supplementary information is available in the online version of the paper. Correspondence and requests for materials should be addressed to AIC or AQRB.

Competing financial interests

The authors declare they have no competing financial interests.