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# Observation of Sub-Poisson Fluctuations in the Intensity of the Seventh Coherent Spontaneous Harmonic Emitted by a rf Linac Free-Electron Laser

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We report the observation of sub-Poisson intensity fluctuations in the coherent spontaneous harmonic radiation generated by an infrared free-electron laser in a photon counting experiment using a well-defined ensemble of electron pulses. These observations constitute the first observation of a nonclassical state of the radiation field generated by a beam of free electrons. The fluctuations observed in the experiment are smaller than those expected from semiclassical radiation theory, and larger than those expected from electron shot noise.

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Study of the radiation emitted by free charged particles has been critical to the development of electrodynamics. The predictions of classical theory for the intensity, angular, and spectral distribution of this radiation have generally been well confirmed by experiment. But less is known about the statistical fluctuations of this radiation. While these statistical fluctuations can be analyzed in the context of classical theory in terms of the statistical fluctuations in the number and coordinates of the radiating particles [1,2], and the interference of the spectral components of the radiated field at the detector [3], the results obtained are not generally consistent with the predictions of semiclassical radiation theory [4]. And the analysis of this radiation within the context of a fully quantized theory has revealed the possibility that fluctuations in the radiation emitted in these elementary processes could be reduced to values below those predicted by the semiclassical theory. Of particular note is the prediction of Becker, Scully, and Zubairy, and that of Gjaja and Bhattacharjee, that operation of a free-electron laser (FEL) could result in the generation of a squeezed state of the radiation field [5,6].

In previous research, analyses and measurements have been made of the fluctuations in the spontaneous undulator and bend-magnet radiation [7] emitted by electrons circulating in a storage ring, and the fluctuations in the self-amplified spontaneous emission emitted by a high-gain RF linac FEL [8–13].

In the research reported below, we have measured the fluctuations in the intensity of the coherent spontaneous harmonic radiation (CSHR) of an infrared (IR) FEL. In classical terms, this radiation is emitted as a consequence of the bunching of the electrons by the fundamental component of the optical field, which leads to the generation of discrete Fourier components of the electron current and enhanced radiation at the integral harmonics of the fundamental. The physics of the radiation mechanism is generally similar to that employed in previous efforts to generate enhanced harmonic radiation using an external drive laser [14].

Given the sensitivity of the higher-order Fourier components of the current to the fluctuations in the number and coordinates of the electrons, and to the amplitude of the field at the fundamental wavelength, these measurements provide a sensitive means to explore the physics responsible for the fluctuations in the radiation emitted by bunched electron beams. We found that the observed fluctuations in the number of radiated photons are substantially smaller than those predicted by semiclassical radiation theory for deterministic electron currents, and cannot be explained on the basis of electron shot noise.

In photon counting experiments of the kind reported here, it is essential to maximize the quantum efficiency (QE) and to suppress all relevant classical fluctuation mechanisms. When the quantum efficiency $\eta$ is less than unity, the measured Fano factor $F$, defined as the ratio of the variance to the photon count mean, is related to the actual Fano factor $F_i$ as $F = 1 + \eta(F_i - 1)$. It is seen that low QE would result in Poisson statistics ($F = 1$) despite the actual distribution at the source. Therefore, although it would be desirable to investigate the sub-Poisson phenomena predicted by Becker *et al.* at the lasing wavelength of the Mark III infrared FEL, the low QE of the high speed IR detectors available for these measurements has made it impossible to date to pursue this approach with currently available FELs.

Since fast photon counting detectors with good quantum efficiencies are readily available in the visible spectrum, in the experiment described below we counted the photons emitted at the seventh CSHR (382 nm) of the lasing wavelength (2.68 $\mu$m) of the Mark III FEL using a fast, high QE (25% at 382 nm) Hamamatsu R329 photomultiplier tube (PMT) in the small-signal regime as the system approached saturation. By counting the net number of photons emitted during equivalent periods of time for each member of a set of FEL macropulses, we could measure the fluctuations in the number of photons generated by a well-defined ensemble of electron bunches.

Since classical fluctuations in electron beam emittance, energy, and current can also modulate the intensity of the CSHR, it is also necessary to suppress these wholly classical fluctuations to the extent that the electron bunches responsible for radiation can be considered as the indistinguishable members of an ensemble. To produce such an ensemble of bunches, it is necessary to ensure (1) that the conditions under which the electron beam was generated and accelerated within the system's linac driver are uniform from bunch to bunch, and (2) that the conditions leading to the optical modulation of the electron beam are identical for each member of the ensemble.

The electron bunch current, energy, and emittance were stabilized in the experiment using feedback to regulate the injected beam current and the accelerating field. To check the effectiveness of these measures, the FEL small-signal gain was measured periodically during the experiment and was found to be substantially constant. Uniform optical modulation of the electron beam by the fundamental laser field was achieved in the experiment by sampling the seventh CSHR photons at a fixed infrared power level defining small-signal regime. The use of this trigger insured that the electron current present in the interaction region of the FEL, both macroscopic and microscopic, was held substantially constant over the time interval sampled by the detector [15]. The Mark III was operated for this experiment at an electron beam energy of 43.5 MeV, a peak micropulse current of 30 A, and a net small-signal gain of 46%. The general operating parameters of the Mark III are described in Ref. [16].

As seen in Fig. 1, a dichroic beam splitter was used to separate the fundamental infrared FEL and the visible CSHR. The seventh CSHR was passed first through a spatial filter to control its intensity, and then through a monochromator to suppress the other near IR and visible harmonics. An external slit in front of the monochromator was used as a spatial filter to alter the count rate without attenuating the beam. The slit, being at the focal plane of the lens shown in Fig. 1, defined the waist of the optical mode into which the seventh CSHR photons were emitted in the cavity. By altering the volume of that mode, variation of the slit width could be used to vary the number of seventh CSHR photons without attenuating those photons. At some point, the mode volume exceeded the volume of the interaction region; beyond this point, the slits served as an attenuator reducing the probability that a photon would be emitted into the selected mode.

Photons were detected by a PMT at the exit of the monochromator. The PMT was operated at −2.25 kV, providing single-photon current pulses with a peak current of 0.4 mA in the linear operating region. The FWHM of the amplified PMT pulses was 7 ns. Those pulses in excess of the discriminator threshold were registered by a dual-channel counter. The two channels on the counter were gated by a pair of 80 ns pulses. The first gate was initiated by a trigger set at 1% of the intensity reached by the Mark III FEL at saturation. By acquiring data in the small-signal regime, we eliminated the chaotic intensity fluctuations due to the sideband instability, insuring that the seventh CSHR photons were radiated at the same IR intensity level within the picosecond micropulse as required for suppression of classical noise. The second gate was delayed by 6 $\mu$s to sample the background counts between laser pulses. The counts accumulated in the second channel permitted verification of the absence of background light during the 80 ns window and/or PMT instabilities during setup of the experiment. A 1 GHz LeCroy multichannel oscilloscope was used to monitor the stability of the IR pulse during the experiment and record the PMT pulse traces at various count rates for analysis of correlation and dead-time effects.

To quantitatively determine dead time as well as to verify the reliability of the counting system, we made a series of measurements using a known Poisson light source, and compared the result with the theoretical curve for the dead-time-modified Poisson radiation (DTMPR). A red light emitting diode (LED) was chosen as the Poisson source based on the following: (1) low PMT quantum efficiency at this wavelength (less than 0.5%); (2) measured Fano factor close to unity ($F = 0.9967 \pm 0.007$) at very low count rates where dead-time effects were negligible; (3) measured differential and integral probability distribution functions closely matching the theoretical Poisson distributions; (4) absence of short- and long-term correlations; (5) the equivalence of the PMT's response to the LED and seventh CSHR as verified by the observed pulse height distributions and shapes.

The LED and seventh CSHR data are plotted together with the theoretical DTMPR curve in Fig. 2. The seventh CSHR data points in Fig. 2 include measurements taken with both the unattenuated and the attenuated CSHR photons. As expected the Fano factors for the attenuated CSHR photons converge to unity due to the low system quantum efficiency at these low count rates. In the discussion below, we consider only the data for the unattenuated

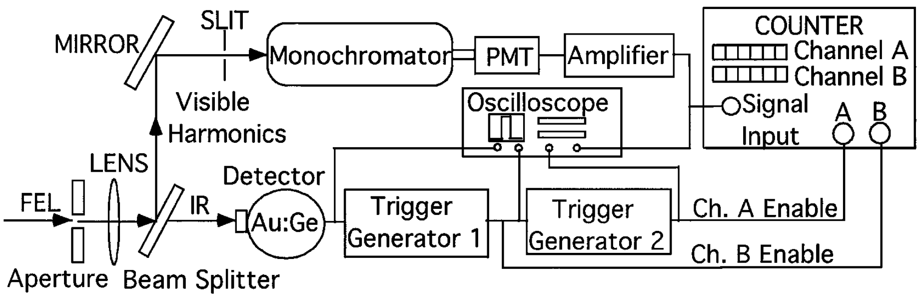

FIG. 1. Diagram of photon counting experiment on the seventh CSHR from the Mark III FEL.

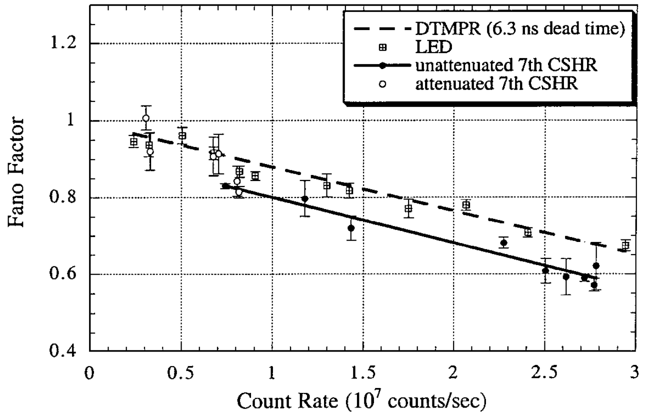

FIG. 2. Reduction of the Fano factor for seventh CSHR (solid line) compared to the theoretical DTMPR (dashed line) and an experimental Poisson source (LED). The error bars indicate the measured standard deviation of the experimental data points.

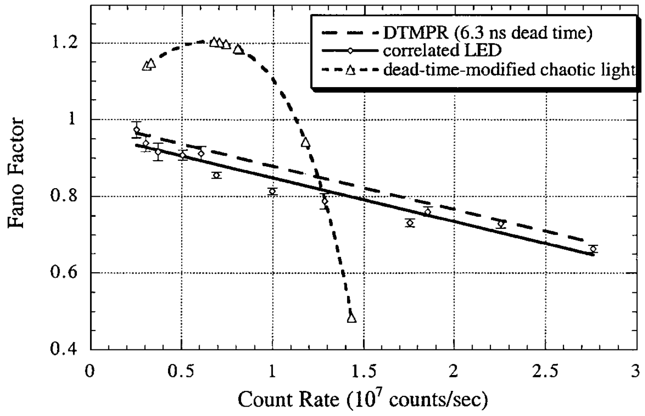

FIG. 3. Dependence of the Fano factor on the count rate for chaotic light and for light source with strong short-term intensity correlation.

case for which the net system quantum efficiency, including cavity outcoupling efficiency, was 11.76%.

The theoretical curve for the DTMPR in Fig. 2 is a reasonable fit to the LED data with a 1% $\chi^2$ confidence level (C.L.), assuming a dead time of 6.3 ns consistent with the observed PMT pulse width. Although the $\chi^2$ C.L. is only moderate, inspection of the tabulated data in Table I indicates that the difference between the best fit to the LED data and the DTMPR is at most of the order of 1 standard deviation, justifying the DTMPR curve as an acceptable fit to the LED data. Although the seventh CSHR data cannot be fit by the DTMPR curve because of an extremely low $\chi^2$ C.L. (less than $10^{-4}$), it can be fit by a straight line offset from the DTMPR curve. The $\chi^2$ C.L. for this *ad hoc* fit is quite high (25%). The offset of this straight line fit cannot be due to statistical errors in the data: as seen in Table I, the seventh CSHR data differ from the DTMPR curve by more than 3 standard deviations at a count rate of $7.5 \times 10^6$ counts/s, and more than 6 standard deviations at $2.75 \times 10^7$ counts/s.

Given that the possible effects of photon clustering can be dismissed for the reasons summarized below, the data in Table I indicate that the measured seventh CSHR Fano factor is smaller than those for DTMPR by up to 0.089 at $2.75 \times 10^7$ counts/s. Allowing for the known quantum efficiency of the detection system and the reduced effect of dead time on sub-Poisson light [17], the data in Fig. 2 and Table I indicate that the actual Fano factor for the seventh CSHR was no more than about 0.243 at the source.

To conclude that the data reported here actually represent an observation of sub-Poisson light, it must be demonstrated that the depressed Fano factors could not be due to photon clustering [18]. The possible effects of photon clustering can be analyzed in several ways. Some form of clustering can be ruled out on elementary grounds. As an example, Fig. 3 compares the functional dependence of the Fano factor on the count rate for chaotic light with the DTMPR. As seen from the figure, the functional form of dead-time-modified chaotic light is simply incompatible with the observed dependence of the seventh CSHR data shown in Fig. 2.

We can also directly evaluate the effects of short-term and long-term intensity correlations on the measured Fano factor. Short-term correlations were studied by using an LED exhibiting strong periodic intensity fluctuations (correlation exceeding 30% at the first maxima) with a period of 50 ns and a coherence time of 400 ns. The correlated LED data are compared with the DTMPR curve in Fig. 3 and Table I. While the Fano factor for the correlated LED is suppressed relative to DTMPR, the effect is small compared to the effect observed for the FEL seventh CSHR.

TABLE I. Comparison of the experimental Fano factor with the theoretical curve for dead-time-modified Poisson radiation (DTMPR). $\Delta F$ refers to the offset of the best-fit experiment curve from the DTMPR at various count rates,[a] and $\sigma_{\Delta F}$ is the standard deviation of $\Delta F$.

| Count rate | $\Delta F \pm \sigma_{\Delta F}$ | | | |
|---|---|---|---|---|
| ($10^7$ counts/s) | Reference LED | Correlated LED | Seventh CSHR | Scatter probability[b] |
| 0.0 | $-0.016 \pm 0.0135$ | $-0.032 \pm 0.009$ | $-0.063 \pm 0.030$ | 0.017 |
| 0.75 | $-0.010 \pm 0.009$ | $-0.032 \pm 0.006$ | $-0.070 \pm 0.022$ | 0.0007 |
| 1.5 | $-0.005 \pm 0.006$ | $-0.032 \pm 0.004$ | $-0.077 \pm 0.016$ | $4 \times 10^{-7}$ |
| 2.25 | $0.0005 \pm 0.008$ | $-0.032 \pm 0.005$ | $-0.084 \pm 0.013$ | Less than $3 \times 10^{-7}$ |
| 2.75 | $0.0042 \pm 0.011$ | $-0.031 \pm 0.007$ | $-0.089 \pm 0.015$ | Less than $3 \times 10^{-7}$ |

[a] Results at zero count rates are extrapolated.

[b] Probability that the difference between the straight line fit to the observed seventh CSHR data points and the theoretical curve for DTMPR could be due to the statistical uncertainties in the CSHR data.

For clustering on the time scale of seconds or minutes due to possible long-term fluctuations (caused by drifts of mirrors, slits, etc.), direct examination of the experimental data verified that the large drift in count rate occasionally observed in the experiment (as large as 40%) had no significant effect on the Fano factor. In practice, we rejected data sets with drift rates in excess of 20% in the computation of the seventh CSHR data reported in Fig. 2 and Table I.

The time-resolved PMT oscilloscope traces can also be used to check for clustering in individual and adjacent micropulses, and in the optical radiation stored in the FEL resonator cavity. The PMT data show no sign of clustering in either individual or adjacent micropulses. Had more than one photon been deposited on the PMT photocathode in a time short compared to the pulse width, the PMT pulse height distribution would have diverged from the distribution for single incident photons. But the distribution deduced from the oscilloscope traces and the dependence of the count rate on discriminator setting remained fully consistent with those expected for single incident photons. These data rule out the possibility of multiphoton emission from individual electron bunches, as well as correlated emissions from adjacent bunches. The autocorrelation function computed from the PMT traces also rules out the possibility of clustering due to stored energy in the cavity. Such clustering would result in the enhancement of the autocorrelation function at the cavity round-trip time (13.7 ns). On the contrary, the PMT autocorrelation function displayed a small local minimum at this time interval.

Photon clustering in adjacent micropulses can also be ruled out on theoretical grounds. The emission of the seventh CSHR photon from adjacent electron bunches is statistically independent because the electron bunches are uncoupled. There is therefore no basis for expecting either correlation or anticorrelation of the seventh CSHR photons radiated by successive electron bunches, ruling out clustering in adjacent photon micropulses.

Having ruled out chaotic, short-term, long-term, and periodic clustering, we may conclude that the depressed Fano factors characterizing the seventh CSHR observed in the experiment must be attributed to the sub-Poisson statistics of that light. The maximum degree of squeezing observed in the experiment exceeded 75.7% ± 12.7% at the source. This observation constitutes the first experimental demonstration of the generation of nonclassical light by a beam of free electrons.

As this experiment was conducted at a coherent harmonic of the lasing wavelength, the analysis of Becker *et al.* is not directly applicable. The observation of a nonclassical state of the electromagnetic field in this experiment characterized by sub-Poisson statistics thus neither confirms nor contradicts Becker *et al.*'s expectations for the state of radiation emitted at the lasing wavelength.

Nonetheless, this result has fundamental consequences for our understanding of the physics responsible for the FEL operation. Nonclassical states of electromagnetic field cannot be described in terms of the deterministic electric and magnetic fields assumed in the classical analyses of the FEL operation. It is also impossible to generate a nonclassical state of the field using a classically defined electron current; semiclassical theory predicts that field of a classical current would invariably display Poisson statistics [4]. Calculation of the Fano factor due to electron shot noise for the conditions of the experiment yields very small Fano factors ($F_i = 0.0273$) [19], inconsistent with both the experiment and with the general results of semiclassical theory. We conclude that neither the classical shot-noise model nor the semiclassical theory can explain the results observed in the experiment, which therefore appear to be of quantum mechanical origin.

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