## Measurement of the Coherent Harmonic Emission from a Free-Electron Laser Oscillator

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We have measured the emitted energy and the spectral and temporal distributions of the first seven harmonics which are self-generated by the Mark III free-electron laser oscillator at Stanford University. The bandwidth scaling shows that current theory will have to be revised. We present our results and discuss the implications for using free-electron lasers as a source of tunable coherent extreme ultraviolet photon beams.

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A free-electron laser (FEL) operating near saturation naturally generates coherent light at all harmonics of the fundamental. The relativistic equations of motion for the electrons produce an amplitude for emission at the harmonics by each electron even in a monochromatic undulator field. Coherent harmonic emission is generated when the laser fundamental bunches the electron beam, producing Fourier components at the harmonics. Because the light is propagating in a vacuum, all harmonics are automatically phase matched.<sup>1</sup>

Our main interest in this work is to identify the characteristics of the coherent harmonic radiation produced by FEL oscillators. For a laser operating in the visible region of the spectrum, these harmonic beams lie in the vacuum ultraviolet (vuv) and extreme ultraviolet (xuv) wavelengths at which high-intensity tunable coherent radiation is not readily available from other sources. As was the case for synchrotron radiation, a rather simple modification of an FEL facility to allow the harmonics to emerge will permit their use in applications as diverse as chemical, biomedical, physical, and materials sciences.<sup>2</sup> An additional reason for interest in the harmonic emission is the damage it can do to the oscillator mirrors of the FEL.<sup>3-5</sup> Mirror damage can shorten the laser operating lifetime and may limit the short wavelength FEL operating range.

One can have little hope of controlling the intensity or bandwidth of the harmonics if the process of harmonic generation is not understood. The energy and the spectral, temporal, and spatial characteristics are all important parameters. In this work we have taken the first steps towards understanding the relationships among these parameters. The coherent harmonics are also sensitive probes of the electron phase motion in FEL's, and can be useful tools for studying the basic physics of the interaction. Important issues are the importance of wiggler-field errors, electron-beam emittance, electronbeam energy spread, and the coherence of the fundamental.

Although spontaneous harmonic power was observed from the time of the initial storage ring FEL experiments in Novosibirsk,<sup>6</sup> and in Orsay,<sup>7</sup> the first observation of harmonic generation during laser operation was not made until later at Stanford.<sup>8</sup> In this helical undulator FEL, a small signal was obtained at the second harmonic, but no higher harmonics could be seen. The following year, a linear undulator experiment on the same accelerator<sup>9</sup> showed strong emission at the third harmonic which was interpreted as simultaneous thirdharmonic lasing. Shortly thereafter, experiments at Los Alamos<sup>10</sup> and Orsay<sup>11</sup> demonstrated photon multiplication in an FEL amplifier. In the Mark III oscillator coherent harmonic beams are visible to the naked eye.<sup>12</sup> Third-harmonic lasing has recently been demonstrated on this system.<sup>13</sup> In this work, we have investigated the energies, the spectral properties, and the temporal dependence of the coherent harmonic emission. The basic theory of harmonic emission has been described elsewhere<sup>14-16</sup>; we will compare our results with detailed numerical calculations<sup>17</sup> based on this theory.

We have given typical operating parameters for the Mark III FEL and a detailed description of the experimental procedure elsewhere.<sup>18</sup> Briefly, the Mark III produces a 43-MeV electron beam with a 20-A peak current,  $20\pi$  and  $25\pi$  mm-mrad horizontal and vertical normalized emittances, and less than 0.3% rms energy spread. This beam interacts with the magnetic field produced by a linear wiggler (47 periods of wavelength 2.3 cm). The FEL resonator uses silver-plated mirrors for broadband operation, and the  $3-\mu m$  laser light is extracted with an intracavity CaF<sub>2</sub> plate. We disperse the radiation from the FEL and isolate the harmonics with a Pellin-Broca prism system. The harmonic of interest is directed onto a detector for time-resolved absolute power measurements, or into a monochromator for time-resolved spectral measurements.

Typical temporal profiles are presented in Fig. 1. The rise time decreases for the higher harmonics, and saturation occurs earlier. Because of the progressively stronger saturation, the higher harmonics develop a flat top, and their energy fluctuates less than the fundamental (by factors of 2 to 4). These temporal profiles are integrated to yield absolute energies, which are shown in Fig. 2 for three different settings of the wiggler magnetic field.



FIG. 1. Typical single-macropulse temporal profiles of harmonic power for the first three harmonics. The curves have been multiplied by the scale factors on the right-hand side of the figure. Each harmonic has zero intensity at time zero, but has been vertically displaced for clarity. The micropulse time structure is not resolved.

(The wiggler parameter K is  $eB\lambda_W/2\pi m_0c^2$ , where B is the peak magnetic field and  $\lambda_W$  is the wiggler period.) As expected, the energies drop rapidly with increasing n. The estimated accuracy of the absolute energy measurements, based on uncertainties in the calibration factors involved (including detector calibration and optical losses between output coupler and detector), is  $\pm 30\%$ . The experimental precision, given by pulse-to-pulse fluctuations, depends on the harmonic, <sup>18</sup> but is typically less than 5% rms.

Shown for comparison are numerical calculations of the coherent harmonic energies by Schmitt.<sup>17</sup> These calculations assume that the harmonic radiation is generated during a single pass through an ideal wiggler field with perfect electron injection and with the laser operating at saturation. [The fundamental intracavity power is set at either 200 MW (K=1.4) or 40 MW (K=1.07), close to the peak circulating powers which is calculated from the measured peak power, output coupling fraction =2.9%, and micropulse length.] Larger emittance has a small but significant effect on the calculated harmonic intensities; for example, doubling the emittance causes the harmonics to be reduced by factors of 2 to 4.<sup>17</sup> This can be taken as the theoretical error since our emittance is uncertain within about a factor of 2.

Both experiment and theory indicate in Fig. 2 that the harmonic energy increases as the wiggler field increases. This is expected because larger magnetic fields increase the nonlinear transverse electron amplitude. For K = 1.07, the experimental energies are in reasonably good (within a factor of 3) agreement with the calculations of Schmitt<sup>17</sup> for the first four harmonics. For the higher harmonics the experimental values are considerably lower than the predicted values.

The experimental data lack the even-odd intensity al-



FIG. 2. Output energy per macropulse as a function of harmonic number for three different settings of the wiggler parameter K. Calculated energies from the HELEX simulation (Ref. 17) are shown for comparison.

ternation predicted by the theory. This discrepancy, and the surprisingly low energies of the higher harmonics, might be explained by the presence of magnetic field errors or off-axis electron injection, both of which make the even harmonics relatively stronger.<sup>17,19</sup> The errors<sup>20</sup> for the Mark III wiggler measured in 1984 are not large enough to cause significant decreases<sup>21</sup> in the harmonic intensities, and the Mark III gain has apparently not been degraded since then. However, other linac-based undulators have shown serious degradation, and an effect too small to change the gain might have a serious impact on the high harmonics. Because the electron beam steering was adjusted to optimize the fundamental energy, errors should be small. The odd and even harmonics are reduced by similar amounts when larger emittance is used in the calculations.<sup>17</sup>

The linewidth of the fundamental radiation varies as a function of cavity mirror spacing because of short-pulse effects.<sup>22</sup> The spacing which minimizes the linewidth does not maximize the fundamental macropulse energy.<sup>18</sup> In Fig. 3 the instantaneous linewidth (FWHM) of the radiation (averaged over a 140-ns-long interval centered near the peak of the third-harmonic temporal profile) as a function of harmonic number is shown at the minimum-linewidth cavity mirror spacing.

The linewidths of the harmonics (Fig. 3) do not behave as expected. We anticipated that the coherence length of the harmonics would equal that of the fundamental. This leads to the prediction that the harmonic linewidth decreases linearly with the harmonic number. In the Orsay frequency multiplication experiment<sup>11</sup> the third-harmonic linewidth was measured to be less than one-half of the fundamental, in agreement with this prediction. However, we observe a rising or roughly constant linewidth, or a coherence length which decreases roughly linearly with n.

As is evident from our measurements of the macropulse temporal profiles, <sup>18</sup> the harmonic intensities satu-



FIG. 3. Average instantaneous FWHM linewidth  $(\Delta\lambda/\lambda)$  as a function of harmonic number with the laser cavity length adjusted to minimize the fundamental linewidth. Error bars show 1 sample standard deviation.

rate early. This implies that the length of the micropulses is slightly longer for the harmonics than the fundamental. The observed reduction in the coherence length is therefore not explained by a physical reduction in pulse length. Several factors discussed above which affect the harmonic intensities, including field errors, beam steering errors, and electron-beam emittance, affect the harmonic linewidths only in second order, and are negligible. We believe a new physical effect is present in this experiment which reduces the coherence length of the harmonics but which is not included in the present models of the harmonic-generation process.

A theoretical effort<sup>23</sup> to explain our linewidth results has identified saturation effects as the likely cause. This treatment will not yield quantitative predictions for our experiment until short-pulse effects have been included; however, it shows that the instantaneous harmonic intensity within a micropulse is an oscillatory function of the instantaneous fundamental intensity, with a period inversely proportional to the harmonic number. A smoothly varying temporal micropulse in the fundamental will therefore yield multiple peaks in the harmonic time structure, producing harmonic coherence lengths which are reduced by a factor proportional to the harmonic number. This result could be very significant to the applications of FEL's since it identifies a way to reduce the harmonic bandwidths and stabilizes the fundamental intensity in the micropulse.

Coherent FEL harmonic emission in the xuv could be a useful tool for scientific research. Although frequency summation using visible dye lasers can be used to generate reasonable amounts of tunable coherent radiation throughout the vuv spectral region,<sup>24</sup> similar schemes have more limited efficiency and tunability in the xuv.<sup>25,26</sup> The FEL envisioned<sup>27</sup> for the storage ring now under construction at Duke University would be capable of oscillating throughout the visible and ultraviolet regions of the spectrum, with the potential of vuv and xuv lasing in the more distant future. When operating in the uv this laser is expected to produce 100-500-ps pulses with up to 10-MW output power. The conversion efficiency for third-harmonic generation should be close to the value of about  $10^{-3}$  observed in the present experiment; this number is 3 orders of magnitude larger than the efficiency obtained using frequency tripling of uv dye-laser radiation in a molecular beam.<sup>25</sup> We can thus expect to obtain kilowatt levels of tunable xuv power with a full octave tuning range  $(\Delta\lambda\lambda = 2)$ , as compared with fractions of a watt available now over a  $\Delta\lambda\lambda \approx 0.1$  tuning range.<sup>25</sup> Kilowatt xuv power levels might also be attainable using frequency multiplication of an external laser in an optical klystron.<sup>28</sup>

Our results show that coherent harmonic radiation self-generated by an FEL oscillator can be useful source of tunable xuv light for scientific research if an effort is made to couple it out of the resonator. The need to understand the factors which determine the coherence length of this radiation should stimulate further theoretical and experimental research.

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 $^{l}$ At short wavelengths, the index of refraction of the electron beam produces a negligible effect for any reasonable undulator length.

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