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Demonstration of harmonic lasing in a free-electron laser

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It has long been known that gain exists at the odd harmonics of a free-electron laser. Using a dispersive intracavity element to alter the cavity lengths for the fundamental and harmonic lasing, we have demonstrated harmonic lasing between 1.4 and 1.8 μ m using the Stanford Mark III infrared free-electron laser. Gain at the third harmonic was seen to agree with theory to within experimental limits. The wavelength of the third harmonic was not quite equal to one third of the fundamental due to saturation and bandwidth effects. Power output was somewhat smaller than expected. This may have been due to losses in the optical transport system.

It has been realized for some time that a free-electron laser has gain at odd harmonics of the fundamental operating frequency.^{1,2} In 1982, researchers using the ADONE storage ring at Frascati Laboratory measured the gain curves for the third-harmonic and suggested that lasing could be attained more easily at the third harmonic due to the better emittance at lower electron energy.³ Unfortunately, the gain was still too low to lase. In 1983, evidence of enhanced third-harmonic output in a $1.6-\mu m$ free-electron laser oscillator was found by Edighoffer et al.⁴ The evidence for lasing was not conclusive, however, and the third-harmonic power level never exceeded one thousandth that of the fundamental. In Edighoffer's system the fundamental and third harmonic could, in principle, lase simultaneously. Such a system is very difficult to study. In general, one gets very strong coherent emission at the third-harmonic FEL oscillators even with gain well-below threshold due to the overbunching of electrons by the laser interaction at the fundamental.⁵ It is difficult to separate the lasing signal from this coherent spontaneous signal in such a device.⁶

The optical cavity in the Stanford Mark III FEL consists of two opaque metal mirrors (uncoated amorphous silver) at each end of the laser undulator. Output coupling is accomplished by means of a calcium fluoride plate mounted a few degrees away from Brewster's angle.⁷ The plane of incidence is parallel to the plane of electron oscillations in the wiggler. Since the laser output is strongly polarized in this plane, the reflection off the plate is fairly weak. For the experiments reported here we used a 64.8° plate which yields 5.9% total output coupling at 4.8 μ m and 5.65% output coupling at 1.6 μ m. The cavity itself has losses of the order of 2% between 1 and 8 μ m.

The intracavity CaF_2 plate is dispersive and leads to a wavelength-dependent round-trip cavity time. Since the laser is driven by a radio-frequency linear accelerator, the laser gain and power are very sensitive to the cavity length. The laser can only operate within a few microns of the *synchronous length*, defined as the length at which the round-trip path length is equal to the arrival time between electron pulses. Due to laser lethargy⁸ the laser power peaks near the synchronous length, falling off very steeply for longer cavity length and somewhat less steeply for shorter lengths. At the 4.8- μ m wavelength used in these experiments, the third-harmonic radiation at 1.6 μ m should be synchronous for a cavity length 60 μ m longer than that of the fundamental. Since the power falls off very quickly on the long side of the cavity length detuning curve, there is no chance of any significant amount of coherent spontaneous radiation at 1.6 μ m. This is essential for turn-on since the initial length enhancement seen is usually smaller than the spontaneous radiation level.

Harmonic lasing was achieved as follows. The laser was started at the fundamental wavelength and tuned up for maximum power and gain. The cavity length was then changed to search for the peak in the 1.6- μ m output expected due to the third-harmonic gain. A fast Ga(Au) detector was used to measure both the fundamental and the third harmonic. This detector is very sensitive at 1.6 μ m due to its intrinsic germanium response, so most data were taken for harmonic lasing at this wavelength. A 1.6- μ m notch filter was used to filter out all radiation except the third harmonic. The cavity length was set at the peak of the length enhancement and the mirrors and electron beam were then tuned until saturation was achieved. At wavelengths other than 1.6 μ m, we used a Pellin-Broca prism to separate out the third harmonic. The rest of the procedure was the same. Saturated pulse energies were measured using a broadband pyroelectric detector.

Once saturated harmonic lasing was achieved, the laser could be switched from the third harmonic to the fundamental by changing the cavity length alone. A scan of the peak laser power versus cavity length is shown in Fig. 1. Note the asymmetric nature of both the laser curves. Different filters were used for each wavelength, so the ratio of the power in these curves is not indicative of the actual power ratio. The distance between the peaks in the curves is 58 μ m, close to the expected value of 60 μ m.

The ratio of the third-harmonic gain to the fundamental gain is given in Ref. 2. If one assumes a wiggler parameter K equal to 1.4 and an optical-mode volume proportional to the wavelength, one finds that the gain ratio is 0.97 for an ideal electron beam.

In order for the gain ratio in a realistic laser to approach unity, the effective energy spread, which includes contributions from both longitudinal and transverse momentum spreads, must be much less than 1/6N in full



FIG. 1. Laser power vs oscillator cavity length in thousandths of an inch. A larger value for the length indicates a longer cavity. The power is not to scale. Different filters were used for the third-harmonic power and fundamental power. The actual maximum powers are listed in Table II.

width, where N is the number of wiggler periods. In addition, the wiggler must be nearly ideal. The Mark III laser accelerator and wiggler⁹ satisfy this condition except for the longitudinal energy spread. The parameters of the Mark III are listed in Table I. The calculated gain reduction due to wiggler imperfections is 13%. The value of $6N(\delta\gamma/\gamma)$ is approximately unity. This reduces the gain of the third harmonic to about half that of the fundamental. The exact value of the energy spread is close to the resolution of the electron spectrometer so the gain reduction factor cannot be known to better than 20%. The measured gain, cavity losses, power, and wavelength at the fundamental and third harmonic are listed in Table II. The experimental gain ratio was close to one half, as expected from theory.

One interesting feature of harmonic lasing is that the wavelength of the third harmonic is not exactly one third of the fundamental wavelength. This is due to the asymmetry of the FEL gain curve and the distortion of the gain curve at saturation. Both of these effects are inversely proportional to the effective number of wiggler periods. In addition, the fundamental is more strongly saturated due to its higher gain and lower cavity losses. When one takes all these factors into account in laser simulations one finds a difference of 1.3% in the lasing wavelength. The measured shift was 1.6% or larger in every run in which the laser was run at the harmonic. This discrepancy may be due to three-dimensional effects which can pull the frequency to longer wavelengths for a small electron beam.¹⁰

The ratio of the power extracted from the electron beam for the two wavelengths should be 0.28. The measured ratio calculated from the known coupling efficiency and transport mirror losses is 0.12. One might think that this is because of incomplete saturation, but the spectra showed evidence of sidebands both at the fundamental and the third harmonic. These should only be present at saturation. In addition, the coherent spontaneous radiation at 530 nm for harmonic lasing was quite bright, indicating full saturation. It is possible that losses in the optical transport line are to blame. We have recently found that optical transport losses from the laser to the detector at 4.9 μ m are 25% and grow to 30% at 3.5 μ m. Since scattering by dust is strongly dependent on wavelength, it could account for the difference. In future experiments we intend to put a detector next to the laser to measure the power at the output coupler.

In order to determine whether the laser was operating in the lowest-order transverse-cavity mode, we placed an aperture in the laser far field and reduced its size until the transmitted power was reduced by half. The aperture size indicated that the laser was lasing in the TEM_{00} mode. A higher-order mode would have required a significantly larger aperture.

In conclusion we note that several free-electron laser designs rely on the option of harmonic lasing to extend the wavelength range of the laser to wavelengths not possible with the energy delivered by the accelerator system. This now seems feasible given adequate beam quality. We should also note that harmonic lasing has already been used to generate light in the range of 1.4 to 1.8 μ m for studies in materials sciences.¹¹

TABLE I. Mark III laser and accelerator parameters during harmonic lasing.

Parameter	Value	
Electron beam energy	35 MeV	
Macropulse length	3.2 µsec	
Micropulse length	2.5 ± 0.5 psec	
Beam current (peak)	20 ± 5 A	
Energy spread	$0.4 \pm 0.1\%$ full width at half maximum	
Transverse emittance $(\beta \gamma \varepsilon)$	$15 \pm \pi \mathrm{mmmrad}$	
Wiggler wavelength	2.3 cm	
Wiggler parameter K	1.39	
Number of periods in wiggler	47	
Optical cavity length	183.6 cm	
Rayleigh range	73 cm	

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	Fundamental	Third Harmonic
Net Gain	47±2%	19±1%
Total cavity losses	$7.2 \pm 0.2\%$	$8.8 \pm 0.2\%$
Output coupling	5.9%	5.65%
Ideal transport losses	14%	24%
Macropulse power	30 kW	2.4 kW
Peak wavelength	4.85 μm	1.59 μm

TABLE II. Relative performance of fundamental and third harmonic lasing.

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