

Instrumentation for the Accurate Measurement

and Control of Optical Phase

Final Technical Report

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Instrumentation for the Accurate Measurement and Control of Optical Phase

John M. J. Madey, FEL Laboratory, Duke University

SYNOPSIS

This grant was sponsored by the FY95 Defense University Research Instrumentation Program (DURIP). Free electron lasers hold a unique place in laser physics as high power, broadly tunable sources of optical radiation with unprecedented spectral resolution. These sources are also capable of generating spectrally pure isolated optical pulses and pulse trains which are particularly well suited to spectroscopic applications conveying the capability for detection and analysis at extreme levels of sensitivity. These sources can also uniquely generate high power amplitude and/or phase squeezed state radiation. The objective of this grant was to acquire and upgrade instrumentation for the accurate measurement and control of optical phase of the infrared and ultraviolet free electron laser (FEL) systems at Duke University, thereby enhancing the breadth of capabilities of these sources for supporting the DoD-sponsored research programs of the Laboratory and of our collaborating institutions. The instrumentation and equipment acquired with funds from this grant include a Fox-Smith Laser Resonator, infrared optical and data acquisition instrumentation, and VUV/XUV optics and instrumentation. The technical content of the research funded by this grant is summarized in the attached paper, "High-Power, Fox-Smith Resonator for Tunable, Phase-Locked Operation of an Infrared Free-Electron Laser", by Eric B. Szarmes.

BACKGROUND

This report documents the results of funding received by Duke University from the FY95 Defense University Research Instrumentation Program (DURIP) for the acquisition of *Instrumentation for the Accurate Measurement and Control of Optical Phase*. This grant, amounting to \$544,000, was administered by the Air Force Office of Scientific Research (AFOSR); Dr. Alan E. Craig served as AFOSR's Program Manager.

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In response to the Department of Defense (DoD) areas of interest specified in Section VII of BAA 95-2, specifically for the accurate measurement and control of optical phase, the instrumentation proposed for this grant was intended to provide:

- Production and control of a phase locked, GHz-rate infrared pulse trains using the Mark III FEL;
- Stabilization of the phase of the optical radiation generated by the OK-4 FEL system;
- High resolution, doppler-free, nonlinear and self-calibrating spectroscopy;
- Coherent spectroscopy and control of optical phase, with application to atmospheric studies;
- Ultra-low level dark-detection of trace impurities, with application to atmospheric studies; and
- Interferometric and geophysical applications involving squeezed states of light.

PROGRESS

Instrumentation was acquired and fabricated for application of the existing and evolving Duke University Free Electron Laser Laboratory (FEL) light sources to the research goals specified above for accurate measurement and control of optical phase. The list of equipment includes: an interferometer-stabilized (Fox-Smith) infrared resonator system; high resolution (Fabry-Perot) infrared spectrometer and crossed-beam 2nd-harmonic detector instrumentation; real-time infrared data acquisition instrumentation; a UV resonator with multiple ports for UV extraction; and optical instrumentation for the UV, VUV, and XUV light sources. Most of proposed equipment has been assembled and tested. A few items of customized equipment are still in fabrication should be delivered for assembly and installation by spring of 1998.

CONCLUSION

The most essential parts of the instrumentation for the accurate measurement and control of optical phase of the Duke FEL light sources have been built and tested. A few remaining pieces of customized equipment are due for delivery very shortly. Thanks to an extension of the Grant period, we were able to use the available budget for most important and essential elements of the system, avoiding duplication with equipment supported by other grants and contracts.

ATTACHMENT

Technical paper: "High-Power, Fox-Smith Resonator for Tunable, Phase-Locked Operation of an Infrared Free-Electron Laser", by Eric B. Szarmes; presented at the *International Conference on Lasers '97* on December 16, 1997, in New Orleans, LA, and accepted February 3, 1998 for publication in the Conference Proceedings.

HIGH POWER, FOX-SMITH RESONATOR FOR TUNABLE, PHASE-LOCKED OPERATION OF AN INFRARED FREE-ELECTRON LASER

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Abstract

The Mark III free-electron laser (FEL) is a continuously tunable, ps-pulse laser delivering a 2.86-GHz train of transform-limited, MW-power optical pulses between the wavelengths of 1-9 μ m. The GHz repetition rate and kW average power of this laser have important applications in high-resolution nonlinear spectroscopy, and considerable effort has been devoted in recent years to develop tunable, high-power intracavity optics for establishing and controlling the interpulse phase coherence required in such applications. Present methods successfully employ an intracavity Michelson interferometer to optically couple the 39 circulating pulses in the laser and induce a high degree of interpulse phase coherence in the output pulse train, but broad tunability and single-mode phase coherence cannot be achieved simultaneously. This paper describes the design and operation of a continuously tunable, high-power Fox-Smith resonator for phase-locked operation of the Mark III FEL which, in contrast to the Michelson interferometer, is capable of delivering single-mode radiation across the entire laser spectrum, with at least 20 dB sidemode attenuation in pulse-train durations of several microseconds. The interferometer is part of a complete upgrade of the Mark III optical resonator, presently under construction, which will extend the flexibility and performance of the laser.

I. Introduction

The broad and continuous tunability of rf-linac free-electron lasers (FELs), coupled with their GHz-rate, picosecond time structure and kilowatt average power, have made important contributions to fundamental and applied research in materials science, chemistry, and medicine. While such a combination of physical attributes is unique to these devices, the fabrication of suitable resonators and optics for such lasers have historically invoked considerable engineering challenges due to the constraints imposed by the nature of rf-linac and undulator technologies. In particular, realization of the full utility of FELs has in all cases required special attention to optics and hardware that can survive exposure, in spatially restricted high-vacuum environments, to ultra-high intracavity peak- and average-powers and to extreme levels of ionizing radiation, while at once offering a broadband optical tuning capability. In addition to the above research, the further possibility to perform high resolution spectroscopy with FELs, requiring a high level of interpulse phase coherence and spectral stability in the output pulse train, has motivated development of the 'phaselocked FEL'1-4 for which the associated advances in FEL resonator technology have again been confronted by the above challenges. The notable progress-to-date in these latter efforts have resulted in the proof-of-principle demonstration of single-mode laser outputs with sub-MHz spectral resolution and tens of watts of optical power in the near and far infrared, ^{5,6} and work has been continuing to develop a robust system matching the intrinsic breadth and flexibility of the FEL. The present paper describes the evolution and design of such a resonator for the Mark III infrared FEL at Duke University. This resonator, which is now under construction, has successfully evolved from a program of engineering development and is expected to firmly establish the efficacy of the phase-locked FEL as a tool for applied spectroscopy.

The Mark III FEL^{7,8} is a continuously tunable, picosecond-pulse laser operating between $1 - 9 \mu m$ that delivers a 2.86-GHz train of transform-limited optical pulses with peak powers of several megawatts and energies of several microjoules. Broad band operation of the laser is achieved using all-metal cavity mirrors and Brewster plate output coupling. The laser is synchron-ously pumped and harmonically mode locked by relativistic, picosecond electron bunches from a pulsed-rf linac in pulse-trains of microsecond duration that repeat at 1 - 30 Hz, and due to harmonic mode-locking of the 2-meter cavity by the GHz electron beam, the laser contains 39 circulating pulses that build up independently from noise with random relative optical phases. In the absence of an interpulse coupling mechanism, the corresponding optical spectrum contains laser lines separated by the relatively narrow 73 MHz round-trip frequency of the resonator (instead of the GHz repetition rate of the pulse train), which is an otherwise severe limitation for applications in high resolution spectroscopy.

Interpulse phase coherence is established in the Mark III FEL by optically coupling adjacent pulses in the cavity. Present methods employ an intracavity Michelson interferometer as a straightforward extension of the Brewster plate output coupling geometry, arranged so that the secondary arm of the interferometer incorporates an extra round-trip delay of one rf period. The circula-

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ting pulses are split by the intracavity beamsplitter but are recombined with their adjacent neighbors, thereby communicating their phase information along the entire pulse train and forcing the circulating optical pulses to evolve from pass to pass with a fixed relative phase as they turn on from noise. The temporal and spectral features of the individual picosecond pulses are preserved, but the interpulse coherence of the pulse train is characterized by an axial mode separation equal to the GHz rf frequency of the linac instead of the MHz round-trip frequency of the resonator. In this paper, for brevity, we refer to this mode reduction as *single-mode operation* or *absolute phase locking*. Of course, the pulse train retains its multimode structure, but each rf band of the spectrum contains only a *single* mode (instead of 39); one of these 2.86-GHz rf bands in the output spectrum can then be filtered externally from the phase locked pulse train to extract the true, single-mode field. The degree of mode reduction can be analyzed for a given resonator design in a straightforward and systematic way by calculating the frequency-dependent axial modes losses in the resonator, ³ which analysis has proven to be an indispensable guide in the design of the present system.

The insertion of a beamsplitter into the harsh intracavity FEL environment poses a severe engineering challenge. The high average power and broad tunability of the free-electron laser discourage intracavity interferometers of any kind, because the circulating intensities or fluences in these lasers can easily exceed the damage thresholds of typical dielectric coatings used to fabricate the intracavity beamsplitter, especially if the coating is extremely broadband. Two options are available, both of which were developed on the Mark III FEL. The first is to simply use the uncoated, Brewster plate output coupler as the beamsplitter. The optic is broad band and damage resistant, but the small reflectance of only a few percent is insufficient to produce single-mode operation. Nevertheless, this technique yields substantial mode reduction to bandwidths of several hundred MHz, and has been used in Doppler-limited spectral scans in acetylene.⁹ Moreover, even a reflectance of 1% induces sufficient interpulse phase coherence to nearly eliminate the two unwanted outcoupled reflections at the beamsplitter surface, and the technique is now routinely used in the Mark III FEL to double the Brewster plate outcoupling efficiency.

The second option, which is the basis of the present upgrade to the Mark III resonator, employs an extremely broadband, birefringent beamsplitter that provides sufficient reflectance without the use of any coatings.¹⁰ The optic is simply a sapphire plate, inclined near the Brewster angle, that uses S-polarized (TE) reflection at one of the surfaces to provide the coupling in the interferometer; for practical angles of incidence (greater than the Brewster angle), reflectances up to 45% can be obtained. Prohibitive cavity losses at the second surface are eliminated by exploiting the birefringence to yield P- (TM-) polarization at that surface, resulting in low-loss reflections that are suitable for output coupling. This beamsplitter has been used in the Michelson resonator (Fig. 1a) to produce single-mode coherence in the Mark III FEL at $3 \,\mu m$.⁶ but is tunable in the Michelson configuration only over



Figure 1. a) Michelson resonator with birefringent beamsplitter, and b) Fox-Smith resonator with birefringent beamsplitter, for a horizontally polarized gain interaction. OC is the output coupling surface, BS is the beamsplitter surface, and M1, M2, M3 are the resonator mirrors. Intracavity beams are indicated by bold lines. δ_{OC} , δ_L , and δ_R are output coupling, beamsplitter leakage, and spurious reflection losses respectively. The angle of incidence θ_{inc} is typically greater than the Brewster angle.

a few isolated and narrow ranges in wavelength. On the other hand, if the beamsplitter is used in the Fox-Smith interferometer (Fig. 1b), it can be tuned over the entire wavelength range of the laser, and also provides much greater side-mode attenuation for single-mode operation. The analyses of phase locking in these configurations have been developed in a previous publication.¹⁰

The present paper is organized as follows. Section II describes the scope of the Mark III resonator upgrade, outlines the main features and functions of the various resonator options, and presents the results of the analysis predicting the phase-locking performance and capabilities for high resolution spectroscopy. Section III briefly discusses some of the engineering problems encountered in the course of the design and describes particular solutions. Finally, Section IV presents a summary and conclusions, and outlines several ideas for future research using this resonator.

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II. Resonator Design and Optical Analysis

1. Scope of Upgrade

The primary objective of the Mark III resonator upgrade, which will replace completely the present resonator vacuum system, is to commission an actively stabilized, tunable Fox-Smith interferometer for experiments in high resolution spectroscopy. The new resonator will include four major sub-systems: 1) an upstream vacuum chamber housing the Fox-Smith interferometer, the upstream cavity mirrors (including an option for hole-coupling), and Brewster plate output couplers; 2) the wiggler vacuum chamber, which will include a taller vacuum bore allowing operation at wavelengths up to 15 μ m, plus a displaced secondary bore for an independent cavity stabilization laser beam; 3) the cavity stabilization laser, hardware, and optics providing continuous tunability with sub-MHz stability, and 4) the downstream vacuum chamber housing the downstream cavity mirror and piezo-driven mirror mounts for cavity stabilization. The basic configuration of the upstream chamber is shown in Figure 2.



Figure 2. Layout of the upstream vacuum chamber showing the resonator optics, interferometers, and Brewster plate output couplers.

The geometry of the vacuum chamber includes four cavity configurations. The first two, retained from the present resonator design, are a two-mirror cavity with Brewster plate output coupling using solid metal mirrors, and the Michelson extension of this geometry in which the beamsplitter is a 71° CaF₂ Brewster plate providing tunable, Doppler-limited (200 MHz) resolution, with the interferometer mirror located outside of the vacuum chamber through a BaF₂ Brewster window. The third resonator, which is new, provides an option for hole-output coupling using an independent mirror accessed by a 45° reflector on the output coupler carousel, and the fourth is the Fox-Smith resonator using either a sapphire beamsplitter at 71° incidence to provide single-mode spectra at wavelengths between 1-4 μ m, or alternately, an uncoated, birefringent *cadmium selenide* beamsplitter at 71° incidence, replaced in the same optic mount, to provide single-mode operation from 4-9 μ m. The beamsplitter assembly and interferometer mirror for the Fox-Smith are affixed independently of the upstream cavity mirror on a separate, movable stage that locks into position, but which can be retracted when the output coupler carousel is being used. This output coupler carousel is oriented vertically and has twelve lockable positions, allowing six separate Brewster plates of various materials or angles of incidence to be inserted into the beam, plus six intermediate positions to remove the Brewster plates from the beamline when the Fox-Smith is in use.

The cavity stabilization system employs a second optical resonator, displaced by 1/2" from the FEL cavity but connected rigidly to the respective mounting plates for the main mirrors, which is actively frequency-locked to a commercial, stabilized red He-Ne laser using the Pound-Drever-Hall method.¹¹ Single-mode tunability at any given FEL wavelength is then achieved using an independent piezoelectric drive on the downstream FEL mirror. The stability of the He-Ne laser is better than 1 MHz over 8 hours, which means that the corresponding stability in the infrared laser at 3 μ m, which has the same cavity length as the stabilization resonator, should be on the order of 200 kHz. This stability coincides with the optimum spectral resolution of the FEL axial modes as determined by the microsecond duration of the pulse-trains.

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2. Analysis and Performance

The hypermode description of harmonically mode-locked lasers provides a powerful theoretical basis for the straightforward analysis of phase-locking,^{3,4} and previous detailed analyses of the tunability and axial mode decay in the birefringent Michelson and Fox-Smith resonators¹⁰ have completely encompassed every single detail in the design of the present system.

In the Fox-Smith resonator, broad tunability is realized by rotating the birefringent beamsplitter in the plane of the crystal without adversely compromising the alignment of the interferometer. For both the sapphire and CdSe crystals in present design, the c-axis is inclined from the plane of incidence at an angle (29° and 22° respectively) determined by the 71° angle of incidence, and the projection of the c-axis onto that plane is perpendicular to the surface of the crystals. At this incidence, the S-polarized beamsplitter reflectance is 39% in sapphire and 56% in CdSe. The corresponding P-polarized reflectance in sapphire is 4% and provides the output coupling from the second surface of that crystal; thus, we have ground a vertical wedge of 7 arc-min into the sapphire crystal to separate the beamsplitter leakage from the outcoupled beam in the far field. In CdSe, the 71° incidence is too close to the Brewster angle for the 0.5% P-polarized reflectance to be useful for output coupling, and so that crystal is plane parallel, and outcoupling is achieved using a second Brewster plate on the carousel. Both the sapphire and CdSe beamsplitters are designed with a thickness of 1830 μ m (yielding nominal 5 π phase retardation at ~3 μ m for sapphire, and 3 π retardation at ~7 μ m for CdSe) so they can otherwise be mounted into the same optical assembly.

The minimum-loss wavelengths λ at order m of phase retardation in a crystal with c-axis inclination angle β and thickness t_c, as a function of rotation angle ρ about the surface normal, are given by¹⁰

$$\lambda(\rho) = 2\Delta n \left(\frac{t_c}{2m+1}\right) \left[\frac{1 - (\cos\phi\cos\beta - \sin\phi\sin\beta\sin\rho)^2}{\cos\phi}\right]$$
(1)

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where $\Delta n \equiv n_0 - n_e$ is the birefringence and φ is the internal angle of refraction. The associated cavity losses δ for the kth hypermode (i.e. the kth decaying sideband mode) in a cavity containing N circulating optical pulses are then given by

$$\delta = 2 \left(1 - \cos \xi \right) \left[\left(\frac{1}{T_s^2} + \frac{1}{T_p^2} \right) - \left(\frac{s^2}{T_s} + \frac{c^2}{T_p} \right) - \left(\frac{c^2}{T_s} + \frac{s^2}{T_p} \right)^2 \right]$$
(2)

where $\xi \equiv 2\pi k/N$, $T_{s,p}$ are the S- and P-polarized transmittances of the beamsplitter surface, and 'c' and 's' are shorthand notations for $\cos(2\psi)$ and $\sin(2\psi)$, where the angle ψ determines the decomposition of the incident polarization into ordinary and extraordinary components and is given as a function of rotation angle ρ by

$$\Psi(\rho) = \arccos \frac{\cos \rho}{\sqrt{\cos^2 \rho + (\sin \phi \cot \beta + \cos \phi \sin \rho)^2}}$$
(3)

Figure 3 shows the first order hypermode losses at the various $[2m+1]\pi$ phase retardations for both the sapphire and CdSe beamsplitters over their respective tuning ranges, as accessed by rotation of the beamsplitter through an angle of $\pm 22^{\circ}$. Note that the *entire lasing range* can be accessed within these angular limits, with some overlap, by choosing the appropriate order of phase retardation. From 1-4 µm the relative hypermode losses are greater than 2.5% per pass for the sapphire plate, and greater than 6.5% per pass for CdSe from 4-9µm. Thus, after 400 passes in the cavity (5.5 µs in the Mark III FEL) the relative side-mode intensities are, respectively, $(1-0.025)^{400} = 4(10^{-5})$ or 44 dB for sapphire, and $(1-0.065)^{400} = 2(10^{-12})$ or 116 dB for CdSe!



Figure 3. Round-trip, sideband mode losses for the 9π , 7π , 5π , and 3π phase retardations in sapphire, and for the 3π phase retardation in CdSe, across their respective tuning ranges for beamsplitter rotation angles of $\pm 22^{\circ}$.

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Of course, the FEL produces broadband, picosecond optical pulses, and so the question of the bandwidth of the birefringent optics must be addressed. In fact, Jones Matrix calculations of the spectral passbands for the sapphire beamsplitter show the cavity losses for the surviving hypermode, which supports the propagation of the phase locked optical pulses in the cavity, are less than 10% in a bandwidth of $\Delta\lambda/\lambda \sim 2\%$ for all rotation angles of the beamsplitter (with a minimum loss of 8% due to the P-polarized double-pass through the outcoupling surface). These cavity losses are quite moderate for Mark III operation, and the 2% bandwidth is three times greater than typical laser bandwidths corresponding to picosecond pulse durations. We also find, again for all rotation angles of the beamsplitter, that the relative 1st order hypermode losses are more than 1% greater than the fundamental losses over an even larger bandwidth of $\Delta\lambda/\lambda \sim 3\%$. Therefore, while the losses remain low enough to support the propagation of the picosecond pulses, the axial mode decay rates remain high enough to ensure a high degree of phase locking across the pulse spectrum. The corresponding bandwidths for transmission and phase-locking are even larger for the CdSe beamsplitter, due to both the small 3π phase retardation and the higher surface reflectance.

In the phase locked FEL, discrete tuning of the axial modes is accomplished by scanning the interferometer mirror on the scale of an optical wavelength.^{2,3} Perfect phase coherence, with zero beamsplitter leakage, is obtained when the relative optical displacement between adjacent coupled pulses in the interferometer is an integral multiple k of λ /N (corresponding to a phase shift of $\xi = 2\pi k/N$). If the mirror displacement yields intermediate values of the phase shift, then the leakage losses increase, and phase locking takes longer to occur due to the fact that the relative losses of two of the competing hypermodes become comparable. If the phase shift is an exact half-integral multiple of λ /N, then the two competing modes both survive and yield the maximum leakage losses due to destructive interference. Therefore, the stability of the interferometer mirror must be good enough to ensure a high degree of phase locking and side-band mode reduction for a given surviving mode.

The relative intensities of the two competing hypermodes (say, the $k = 0^{th}$ and $k = 1^{st}$) can be calculated from the ratio of the relative growth rates $\gamma \equiv 1 - \delta$ in which the previous expression for the phase shift ξ following Eq. 2 is replaced by $\xi = 2\pi k/N - \phi$, where ϕ is the round-trip phase difference between coupled pulses imposed by the interferometer mirror. For the Fox-Smith resonator, the ratio of the intensities after p passes simplifies (from Eq. 2 with $\psi = \pi/4$) to

$$\left[\frac{\gamma_{k=1}}{\gamma_{k=0}}\right]^{p} = \exp\left\{-p\frac{2(1-T_{s})}{T_{s}^{2}}\left[1-\cos\left(\frac{2\pi}{N}\right)-\phi\sin\left(\frac{2\pi}{N}\right)\right]\right\}$$
(4)

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This result was used to calculate the data in Table 1 below, which summarizes the absolute mirror stability required to enforce a high degree of mode reduction in the Fox-Smith resonator at wavelengths of 2 μ m, 3 μ m, and 4 μ m using the sapphire beamsplitter. As indicated above, mode hops occur when the interferometer mirror is displaced by $(1/2)\lambda/N$, for example, 38 nm at $\lambda = 3 \mu$ m, where N = 39 in the Mark III FEL. From Table 1, we see that we can maintain at least 20 dB mode reduction at *all* wavelengths for mirror displacements as great as 50% of this mode-hopping range! Greater stability is required for the shorter wavelengths, but in all cases the required stability can be achieved with modern commercial piezoelectronics coupled with careful optical design.

	useful mode reduction	using the sapphire beamsplit	ter.	
Relative Phase	Fraction of tuning range	Absolute mirror stability	Hypermode attenuation	
Offset	$[= 2\phi/(2\pi/39)]$	(nm) [= Frac x (λ/2)/39]	(400 passes; 5.5 μs)	
Wavelength = 2 μm				
0			40.dB	
0.040	50 %	13 nm	20 dB	
0.050	62 %	16 nm	15 dB	
Wavelength = 3 μm				
0			43 dB	
0.043	54 %	21 nm	20 dB	
0.052	65 %	25 nm	15 dB	
Wavelength = 4 µm				
0			41 dB	
0.042	. 52 %	27 nm	20 dB	
0.051	63 %	33 nm	15 dB	

Table 1. Absolute mirror stability required in the Fox-Smith interferometer for

When scanning the interferometer mirror for discrete mode selection, the leakage transmission induced in the cross-over regions of strong mode competition compares significantly with Brewster plate output coupling. For the sapphire beamsplitter this leakage is as high as 0.6%, for which the peak power of the micropulses can approach 1 MW! This fact was one of the reasons for separating the leakage from the outcoupled beam using a wedge. Also, the direct detection of the leakage power provides an independent monitor of phase locking quality, and yields a redundant and reliable check for counting the mode hops during computerized spectral scans.

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III. Resonator Hardware and Vacuum Mechatronics

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Several physical constraints have governed the present design of the resonator and vacuum chamber, mainly relating to the restricted space for locating the optical hardware in the electron beamline for the all of resonator configurations described in Section II.1. The available space between the steering magnets in Figure 2 measures 9.5 inches along the beamline. In addition, provisions must be engineered for remote control and adjustment of all subsystems in a vacuum environment maintaining 10⁻⁷ Torr, and both active and passive interferometric stabilization must be incorporated. Finally, the location of the optics must satisfy the mode locking condition, by which the distances between the cavity mirrors and the arms of the interferometers are constrained by the 2.86 GHz frequency of the electron and optical pulse trains. Several relevant optical parameters for the resonator are listed in Table 2.

To alleviate the effects of secular variations in atmospheric pressure, which unavoidably causes the vacuum chamber walls to bow, the upstream mirror mount, Fox-Smith interferometer, and outcoupler carousel are mounted independently on a base plate that is internally elevated from the floor of the vacuum chamber by a three-point contact at the edges. The variations of atmospheric pressure would otherwise impose uncontrolled variations on the order of microns in the interferometer path lengths between any optics mounted directly to the floor. By similar considerations, the steering feedthroughs for the upstream mirror mount are affixed near the edge of the upstream wall where the shift due to bowing is minimal, and the mirror mount is constructed in independent 'frames' that mechanically decouple the angular adjustments from the longitudinal position of the mirror. An invar bracing system extends externally between the upstream and downstream vacuum chambers, over a length of 2 meters, and provides sufficient thermal stability for active stabilization of the cavity length to be accomplished using piezoelectric displacements of a few microns.

The fabrication of the Fox-Smith interferometer, in the restricted space between the upstream mirror mount and the output coupler carousel, provided the greatest engineering challenge. Due to the mode locking condition, a total path length of only 5.25 cm between the interferometer mirrors is available for the insertion of the beamsplitter, which must be inclined at 71° and rotatable by $\pm 22^{\circ}$. The assembly must also include a steering adjustment mechanism for the crystal, a steering capability on the interferometer (independently of the upstream and downstream cavity mirrors), a fine piezoelectric tuning capability on the interferometer mirror, and finally, allow for the entire Fox-Smith assembly to be removed from the beamline when not in use. The detailed layout of the hardware which was engineered to accommodate all of these features is shown in Figure 4.





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Table 2. Optical parameters for the Mark III resonator		
Pulse separation	10.49 cm	
Cavity length (nominal)	2.046 m (39 pulses round-trip)	
Cavity length (hole-coupled)	1.994 m (38 pulses round-trip)	
Rayleigh range (symmetric resonator)	0.5232 m	
Length of wiggler vacuum chamber	1.32 m	
Height (= 1/2 width) of wiggler vacuum bore	6.50 mm	
Optical beam radius at mirrors (for $\lambda = 4 \mu m$)	1.8 mm	
Tuning range (extended)	1 μm - 15 μm	

Several specific features of the design of this assembly should be noted. The 7 arc-minute wedge in the sapphire beamsplitter, by which the leakage beam is separated from the outcoupled reflection, yields substantial beam steering in the interferometer when the crystal is rotated by $\pm 22^{\circ}$. The mirrors can in principle, of course, be steered to compensate, but this solution requires more space for the steering mechanics than would be available in the vicinity of the interferometer mirror. To eliminate the beam steering entirely, we have included a 21 arc-min, *counter-wedged* BaF₂ Brewster plate in front of the sapphire beamsplitter which rotates with the beamsplitter on the same assembly. The angular wander of the beams in the interferometer is thereby reduced to $\pm 100 \,\mu$ rad for the entire rotation range of $\pm 22^{\circ}$, with a slight residual steering due to the different spectral dependences of the refractive indices. The interferometer mirror can now be aligned to this beam using small piezoelectric drivers that possess both the range and resolution to yield optimum, stable alignment of the Fox-Smith interferometer. The BaF₂ plate does not eclipse the outcoupled laser beam that is reflected from the sapphire beamsplitter, but due to the single pass of this outcoupled beam through BaF₂ plate, the outcoupled beam wanders by $\pm 0.25^{\circ}$ as the beamsplitter is rotated by $\pm 22^{\circ}$. However, *this wander* is easily compensated by an external mirror that directs the outcoupled beam to the user laboratory.

The rotating beamsplitter assembly is oriented as shown in Figure 4, out of the way of both the electron beam and the outcoupled laser beam. The rotating part that holds the optics is mounted on a spring-loaded bearing assembly capable of rotation by $\pm 22^{\circ}$, and rides on three precision, sapphire balls azimuthally positioned 120° apart between ground steel ways. The rotator is connected by a shaft through the back of the mount, which is coupled in turn to a steel bellows that redirects the axis of rotation, via a 19° bend, to a feedthrough assembly at the wall of the vacuum chamber. The entire assembly sits on a lockable translation stage, providing precise motion in two perpendicular directions, that can be removed from the optical axis without affecting the rotational alignment. Manual differential screws in the beamsplitter mount provide a one-time alignment of the beamsplitter assembly to the axis of rotation to within 50 µrad resolution.

IV. Summary and Conclusions

We have designed a robust optical resonator for broadband, and continuously tunable, phase-locked operation of the Mark III FEL. The resonator incorporates a high-power birefringent beamsplitter in an intracavity Fox-Smith interferometer, for which the choice of sapphire or cadmium selenide selectively covers the entire wavelength range of the laser from 1-9 µm. Previous experiments on the Mark III FEL using the sapphire beamsplitter in the Michelson interferometer have confirmed both its damage resistance and its theoretical operation. The new design is part of a general upgrade of the Mark III resonator to provide actively stabilized, single-mode radiation with sub-MHz resolution and stability for applications in high resolution infrared spectroscopy. The design also incorporates several flexible output coupling configurations, including Brewster plate and hole-output coupling, as well as an intracavity Michelson resonator for phase-locked operation of the laser to provide Doppler-limited spectral resolution and enhanced Brewster-plate output coupling efficiency.

The new resonator will be the driver in a comprehensive program of high resolution spectroscopy using the Mark III FEL. The single-mode powers of several hundred watts delivered by this laser are expected have unprecedented applications in nonlinear spectroscopy and ultra-sensitive linear absorption spectroscopy. One of the primary research goals is to exploit Doppler-free, multiphoton spectroscopy as a probe for studying extremely narrowband, nonlinear optical processes, for characterizing molecular energy transfer mechanisms in long-lived excited states, for performing highly resolved and efficient spectral hole burning, and for studying the efficiency of nonlinear frequency-mixing interactions. The high power and narrow spectral resolution should also facilitate efficient chemical reaction control for isotope separation. Finally, the phase locked FEL will provide the basis for a new technique in ultra-sensitive linear absorption spectroscopy, based on dark-detection of absorbed single-mode radiation from the phase locked pulse train. The method, which was recently demonstrated for the first time on the Mark III FEL,¹² is reminiscent of FM laser spectroscopy but offers at least four orders of magnitude greater sensitivity in the measurement of weak absorption features than the quantum-limited regime of FM spectroscopy. Research goals include implementing, testing, and optimizing the dark-signal detection method on the Mark III FEL, identifying and studying the relevant spectral features in a wide range of trace impurities. and ultimately developing the technique for environmental monitoring, process control diagnostics, and selective gas sensing.

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