



## Development of a Pump-Probe Facility Combining a Far-Infrared Source with Laser-Like Characteristics and a VUV Free Electron Laser

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### Abstract

This paper presents the analysis of a possibility to construct a high power far-infrared coherent source at the TESLA Test Facility at DESY (TTF). The TTF is a facility producing sub-picosecond electron pulses for the generation of VUV or soft X-ray radiation in a free electron laser. The same relativistic, sub-picosecond electron pulses would also allow the direct production of high-power coherent radiation by passing the electron beam through an undulator. Intense, coherent far-infrared undulator radiation can be produced from sub-picosecond electron bunches at wavelengths longer than or equal to the bunch length. The coherent radiation energy is proportional to the square rather than linear proportional to the number of radiating electrons. Since there are  $6 \times 10^9$  electrons in each bunch, the radiation intensity is enhanced by this large factor over incoherent radiation. The source described in this paper provides, in the wavelength range 100–300  $\mu\text{m}$ , a train of 3–10 ps radiation pulses, with 0.3–1 mJ of optical energy per pulse radiated into the central cone. The average output power can exceed 30–100 W. In this conceptual design we assume to use a conventional electromagnetic undulator with a 60 cm period length and a maximum field of 1.5 T. The far-infrared source will use the spent electron beam coming from the VUV FEL which allows one to extend significantly the scientific potential of the TTF without interfering with the main option of the TTF FEL operation. This far-infrared source combines high peak power (30–100 MW), high repetition rate, wide tunability of the radiation in the far-infrared wavelength range and short-pulse operation, and is transform-limited in terms of bandwidth. In addition, the pulses of the coherent far-infrared radiation are naturally synchronized with the VUV pulses from the main TTF FEL, enabling pump-probe techniques using either the FEL pulse as a pump and the far-infrared pulse as a probe, or vice versa.

## 1 Introduction

The far-infrared (FIR) range of the electromagnetic spectrum is not well covered by intense sources except of a few operating FELs. User facilities are available in the Center for FEL Studies at the University of California, Santa Barbara, USA, (UCSB FELs), and at the FOM-Institute for Plasma Physics, Rijnhuizen, in the Netherlands (FELIX). The wavelength range of the UCSB FELs is from 60 to 1000  $\mu\text{m}$ . The UCSB FEL is driven by an electrostatic-beam recirculating accelerator. Unlike rf accelerator driven FELs, the normal output pulse structure from the UCSB FEL is quasi-cw with an adjustable pulse duration of 25  $\mu\text{s}$  and a repetition rate of one Hz. The peak power is typically a few kilowatts [1]. FELIX covers the 5–200  $\mu\text{m}$  wavelength range. It operates at a repetition rate of 5 Hz and produces a 10  $\mu\text{s}$  long train of 1–10 ps long radiation pulses, with a few  $\mu\text{J}$  of optical energy per pulse. The peak and average radiation power in the far-infrared range are about 100 kW and 50 mW, respectively [2]. Another source is the FIREFLY at Stanford University, USA. The FIR beam from this FEL is tunable from 20 to 100  $\mu\text{m}$ . The pulse train is 5 ms long with a repetition rate of 20 Hz and the pulses are separated by 85 ns. The pulse duration can be varied from 2 to 10 ps. The peak and average power in the far-infrared range are about 100 kW and 1 W, respectively [3].

The analysis of the parameters of existing FIR FELs shows that practical sources of broadly tunable, powerful coherent FIR radiation remain essentially unavailable at wavelengths beyond 200  $\mu\text{m}$ . This situation, however, might change soon. The development of magnetic bunch compression systems, together with advances in superconducting accelerator technology and design, now offers the new possibility of laser-like sources in the far-infrared wavelength range. The generation of relativistic, sub-picosecond electron pulses allows the direct production of high-power, coherent, narrow-band, FIR radiation by passing the electron beam through an undulator. This provides a reliable and easily tunable powerful source of FIR radiation for scientific applications [4, 5].

The TTF is a facility producing sub-picosecond electron pulses (50  $\mu\text{m}$  rms) for the generation of VUV or soft X-ray radiation. Utilizing these sub-picosecond electron bunches can also provide broadband FIR source. Intense, coherent FIR radiation can be produced from sub-picosecond electron bunches at wavelengths longer than or equal to the bunch length. The total radiation from an electron bunch is the summation of the electric fields emitted by each individual electron and the total radiated energy is then equal to the square of the total electric field. The coherent radiation energy is proportional to the square rather than linear proportional to the number of radiating electrons. Since there are  $6 \times 10^9$  electrons in each bunch, the radiation intensity is enhanced by this large factor over the incoherent radiation. This paper describes such a coherent source, proposed as part of the TTF FEL user facility. The FIR source addresses the needs of the science

community for a high-brightness, tunable source covering a broad region of the far-infrared spectrum – from 100 to 300  $\mu\text{m}$ . The FIR radiator described in this paper provides a train of 3–10 ps long pulses with 0.3–1 mJ of optical energy per pulse into the central cone. The average output power can exceed 30–100 W.

## 2 Scientific opportunities

The far-infrared radiation band overlaps the principal excitations in condensed matter systems, the transition energies in Rydberg atoms, and is a region of great interest in surface science. A tunable, picosecond scale duration, far-infrared source will permit time-resolved studies of low frequency molecular vibrations. Collective motions of large molecules, including many biologically important systems, can be directly excited and probed. With Rydberg atoms short pulses of far-infrared light can be used to excite multiple transitions, observe above threshold ionization, and study high harmonic generation.

For surface science this source enables the next generation of surface vibrational spectroscopy experiments, probing modes inaccessible with conventional lasers. Sensitive vibrational spectra of adsorbate can be measured by FIR-visible Sum Frequency Generation (SFG) [6]. In the far-infrared, photodetectors are not sufficiently sensitive to detect the few photons that are produced by the molecular monolayer. In SFG the far-infrared spectroscopic information is converted to the visible, thereby greatly improving the detection efficiency. A (fixed wavelength) pulsed YAG laser is mixed with a (tunable) FIR source: if the substrate is centrosymmetric only the surface contributes to the process, which becomes resonant if the FIR wavelength corresponds to an absorption arising from the surface. Therefore the nonlinearity of the process, requiring high peak power, is efficient in discriminating the surface from the bulk. The coherent undulator radiation source is particularly suited for this application since it is widely tunable and of high power (100 MW). The SFG allows one to characterize the molecular structure and coupling of energy from the molecule to the substrate, surface or interface. The additional advantages of using a coherent undulator radiation source coupled SFG spectrometer are the high average power available (100 W) and the possibility of generating Fourier transform limited short pulses of picosecond duration which is particularly attractive for both spectroscopic and time resolved studies of surface or interface vibrational properties.

Nonlinear optical properties of semiconductor quantum well systems have been investigated extensively in recent years due to their technological importance in optoelectronics. These nonlinear optical properties have almost exclusively been studied in the 10-100  $\mu\text{m}$

range [7]. Coherent FIR undulator radiation from sub-picosecond electron bunches suits ideally for this type of experiments.

The pump-probe technique is one of the most promising methods for the application of a high power FIR source [8]. It is the aim of the present project to develop a user facility for pump-probe experiments in the picosecond regime, combining FIR and short-wavelength FEL radiation. The TESLA Test Facility will allow, for the first time, the integration of a far-infrared coherent radiation source and a VUV beamline. One type of experiments will use the VUV FEL beam as a pump and the far-infrared photon beam as a probe; in this mode, researchers will be able to study the vibrational structure of highly excited and superexcited molecules. The other mode - far-infrared beam pump and VUV beam probe - can be used to study cluster energetics and dynamics. The FIR radiation can be used to excite the clusters, which can subsequently be dissociated or ionized by the VUV radiation. Spectroscopic and structural information can thus be extracted. Spectroscopy of gas-phase free radicals will also benefit from the FIR beam pump and VUV beam probe experiments. In these experiments, a cold molecular beam containing a small concentration of radicals would be excited by the intense FIR beam, tunable across the absorption spectrum. Since the density of radicals in the beam is not high enough to allow the direct measurement of absorption, a VUV beam from the TTF FEL would be used to detect the infrared-excited states of molecules by selectively ionizing the vibrationally excited radicals.

### 3 Temporal coherent undulator radiation

The electron beam current is made up of moving electrons randomly arriving at the entrance to the undulator:

$$I(t) = (-e) \sum_{k=1}^N \delta(t - t_k) ,$$

where  $\delta(\dots)$  is the delta function,  $(-e)$  is the charge of the electron,  $N$  is the number of electrons in a bunch, and  $t_k$  is the random arrival time of the electron at the undulator entrance. The electron bunch profile is described by the profile function  $F(t)$ . The beam current averaged over an ensemble of bunches can be written in the form:

$$\langle I(t) \rangle = (-e)NF(t) . \tag{1}$$

For instance, for an electron beam with Gaussian distribution of the current along the beam, the profile function  $F(t)$  is

$$F(t) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{t^2}{2\sigma^2}\right) .$$

The probability of arrival of an electron during the time interval  $t, t + dt$  is equal to  $F(t)dt$ .

The electron beam current,  $I(t)$ , and its Fourier transform,  $\bar{I}(\omega)$ , are connected by

$$\bar{I}(\omega) = \int_{-\infty}^{\infty} e^{i\omega t} I(t) dt = (-e) \sum_{k=1}^N e^{i\omega t_k} . \quad (2)$$

Let us calculate  $\langle |\bar{I}(\omega)|^2 \rangle$ :

$$\langle |\bar{I}(\omega)|^2 \rangle = e^2 N + e^2 \sum_{k \neq n} \langle \exp(i\omega t_k) \rangle \langle \exp(-i\omega t_n) \rangle .$$

Taking into account (1) and (2), we find that  $\langle \exp(i\omega t_k) \rangle$  is equal to the Fourier transform of the bunch profile function  $F(t)$ :

$$\langle \exp(i\omega t_k) \rangle = \int_{-\infty}^{\infty} F(t_k) e^{i\omega t_k} dt_k = \bar{F}(\omega) .$$

Thus, we can write:

$$\langle |\bar{I}(\omega)|^2 \rangle = e^2 N + e^2 N(N-1) |\bar{F}(\omega)|^2 .$$

Electron beams from a linear accelerator have often a more compressed electron distribution of a form between a Gaussian and a rectangular distribution. For the specific cases of a Gaussian and rectangular profile of the electron bunch, the Fourier transforms of the profile functions have the form:

Gaussian profile:

$$\bar{F}(\omega) = \exp\left(-\frac{\omega^2 \sigma^2}{2}\right) ,$$

Rectangular profile with pulse duration T:

$$\bar{F}(\omega) = \left[\frac{\omega T}{2}\right]^{-1} \sin\left[\frac{\omega T}{2}\right] .$$

The radiation field at the frequency  $\omega$  from a single electron is

$$\bar{E}_k(\omega) \propto e^{i\omega t_k} .$$

The radiation power is proportional to the square of the radiation field and summing over all electrons we get

$$P(\omega) \propto \sum_{k,n} \bar{E}_k(\omega) \bar{E}_n^*(\omega) \propto \sum_{k,n} e^{i\omega(t_k - t_n)} .$$

The radiation power, averaged over an ensemble, is given by the expression:

$$\langle P(\omega) \rangle = p(\omega) [N + N(N-1) |\bar{F}(\omega)|^2] ,$$

where  $p(\omega)$  is the radiation power from one electron. For wavelengths shorter than the bunch length the form factor reduces to zero and approaches unity for longer wavelengths.

To optimally meet the needs of basic research with FIR coherent radiation, it is desirable to provide specific radiation characteristics. To generate this characteristics, radiation is produced from undulator installed along the electron beam path. The undulator equation

$$\omega = 2ck_w\gamma^2 \left[ 1 + \frac{K^2}{2} + \gamma^2\theta^2 \right]^{-1}$$

tells us the frequency of radiation as a function of undulator period  $\lambda_w = 2\pi/k_w$ , undulator parameter  $K$ , electron energy  $\gamma$ , and polar angle of observation  $\theta$ . Note that for radiation within the cone of half angle

$$\theta_{\text{con}} = \frac{\sqrt{1 + K^2/2}}{\gamma\sqrt{N_w}}$$

the relative spectral bandwidth  $\Delta\omega/\omega \simeq 1/N_w$ , where  $N_w$  is the number of undulator periods. The spectral and angular density of the radiation energy emitted by a single electron during the undulator pass is given by the expression (at zero angle):

$$\frac{d^2\mathcal{E}}{d\omega d\Omega} = \frac{e^2 N_w^2 \gamma^2 A_{JJ}^2 K^2 \sin^2[\pi N_w(\omega - \omega_0)/\omega_0]}{2c(1 + K^2/2)^2 [\pi N_w(\omega - \omega_0)/\omega_0]^2}. \quad (3)$$

Here  $\omega_0 = 2\gamma^2 k_w / (1 + K^2/2)$  is the resonance frequency,  $A_{JJ} = [J_0(Q) - J_1(Q)]$ , where  $J_n$  is the Bessel function of  $n$ th order,  $Q = K^2/(4 + 2K^2)$ . In the small-angle approximation the solid angle is equal to  $d\Omega = \theta d\theta d\varphi$ . Integration of (3) over  $\omega$  and over  $\varphi$  gives us factors  $\omega_0/N_w$  and  $2\pi$ , respectively. We also have to integrate over  $\theta$  from 0 to  $\theta_{\text{con}}$ . Thus the energy radiated into the central cone, for a single electron, is given by

$$\Delta\mathcal{E}_{\text{con}} \simeq \pi e^2 A_{JJ}^2 \omega_0 K^2 / [c(1 + K^2/2)]$$

The coherent radiation enhances the energy radiated into the central cone by a factor of  $N|\bar{F}(\omega_0)|^2$ .

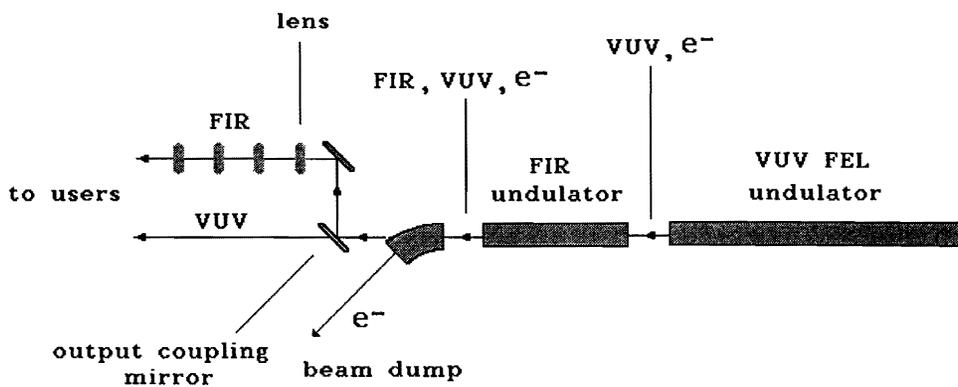
#### 4 Facility Description

In the far-infrared, beyond 100  $\mu\text{m}$ , a source based on coherent undulator radiation has unique capabilities. In this paper we propose to integrate such a source into the TESLA Test Facility at DESY [9, 10]. This source will be able to deliver up to 800  $\mu\text{s}$  long trains of FIR pulses at a separation of 111 ns with 3–10 ps duration<sup>1</sup>, 0.3–1 mJ energy radiated into the central cone, and 100–300  $\mu\text{m}$  wavelength. The superconducting linac will operate at 1 % duty factor, and the average output power of coherent FIR radiation can exceed 100 W.

<sup>1</sup> When the electron bunch moves along the undulator, the electromagnetic wave advances the electron beam by one wavelength at one undulator period. So, the duration of the optical pulse emitted by a short electron bunch passing  $N_w$  undulator periods is equal to  $\lambda N_w/c$ .

**Table 1.** Parameters of the FIR coherent radiation source

<u>Electron beam</u>	
Energy	1000 MeV
Number of electrons per bunch	$6 \times 10^9$
rms bunch length	$50 \mu\text{m}$
rms normalized emittance	$2 \pi \text{ mm mrad}$
rms (VUV FEL induced) energy spread	2.5 MeV
Number of bunches per train	7200
Repetition rate	10 Hz
<u>Undulator</u>	
Type	Planar (electromagnetic)
Period	60 cm
Peak magnetic field	1-1.5 T
Number of periods	10
<u>Output radiation (into central cone)</u>	
Wavelength	100-300 $\mu\text{m}$
Bandwidth	Transform-limited
Peak power	30-100 MW
Average power	30-100 W
Micropulse duration (rectangular profile)	3-10 ps
Micropulse energy	0.3-1 mJ
Bunch spacing	111 ns



**Fig. 1.** Schematic layout of the FIR-VUV pump-probe facility

We propose to install an additional undulator after the VUV SASE FEL. Because the FIR source uses the electron beam coming from the VUV FEL, the proposed source operates in a "parasitic" mode not interfering with the main mode of the VUV FEL operation (see Fig. 1). Starting point of the design are the project parameters of the electron beam after the VUV FEL at the TTF (see Table 1). The planar undulator is an inexpensive electromagnetic device with 10 periods, each 60 cm long. At the operation wavelength of the FIR source around  $300 \mu\text{m}$  the peak value of the magnetic field is about 1.5 T.

Many practical applications require to control the shape and time characteristics of the FIR pulse. For instance, closely spaced picosecond FIR pulses with controllable phase relations are needed for coherent multi-photon excitation and selective excitation of, for example, certain molecular vibrations. The proposed FIR source provides wide possibilities

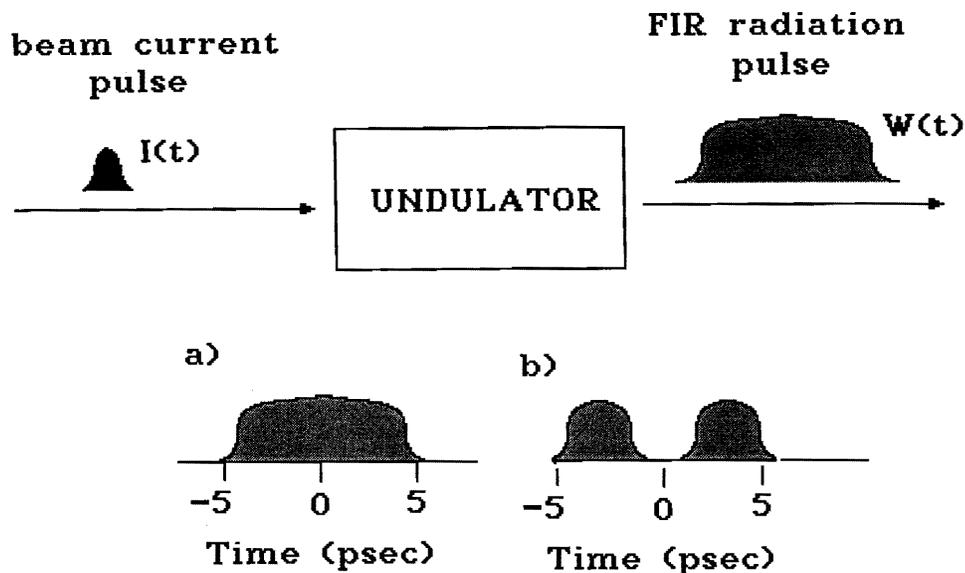


Fig. 2. FIR pulse profiles generated by the FIR undulator in the central cone ( $300 \mu\text{m}$  wavelength). Plot (a) illustrates the pulse produced by a uniform undulator (rectangular pulse of 10 ps duration). Plot (b) illustrates the case when four central poles are switched off (two pulses of 3 ps duration each delayed by 3 ps)

to control and modify in a well-defined manner the shape of the radiation pulse on a picosecond time scale (see Fig. 2). The electron beam passing uniform undulator produces FIR pulse with rectangular profile. FIR optical pulses can be shaped in a complicated manner by means of individual tuning of the magnetic field in each period. It is important to stress that resulting shape (spectrum) of FIR pulses can be well described analytically for this source.

The main goal of the present study has been to design a FIR source which is compatible with the layout of the TTF and the VUV FEL under construction at DESY, and which can be realized with minimal additional effort. The undulator and outcoupling optical system proposed can be installed in the unoccupied straight vacuum line used to transfer the electron beam, and in the VUV beam line behind the dipole magnet separating the electron beam from the VUV beam (see Fig. 1).

In order to make use of the FIR radiation an additional mirror is needed to couple out the major fraction of the optical power in the central cone and to direct it to the experimental area. The distance between the mirror and the exit of the second undulator is 10 m, the distance between the mirror and the exit of the FEL undulator is about 16 m. The minimum size of the hole in the outcoupling mirror is defined by the condition that VUV radiation losses due to hole aperture limitation should be avoided. The fraction of the output FEL power passing the mirror hole is calculated from the angular distribution of the VUV radiation ( $20 \mu\text{rad}$  rms). For a hole diameter of 2 mm the fraction of VUV power directed into the experimental area is close to 100% (see Fig. 3).

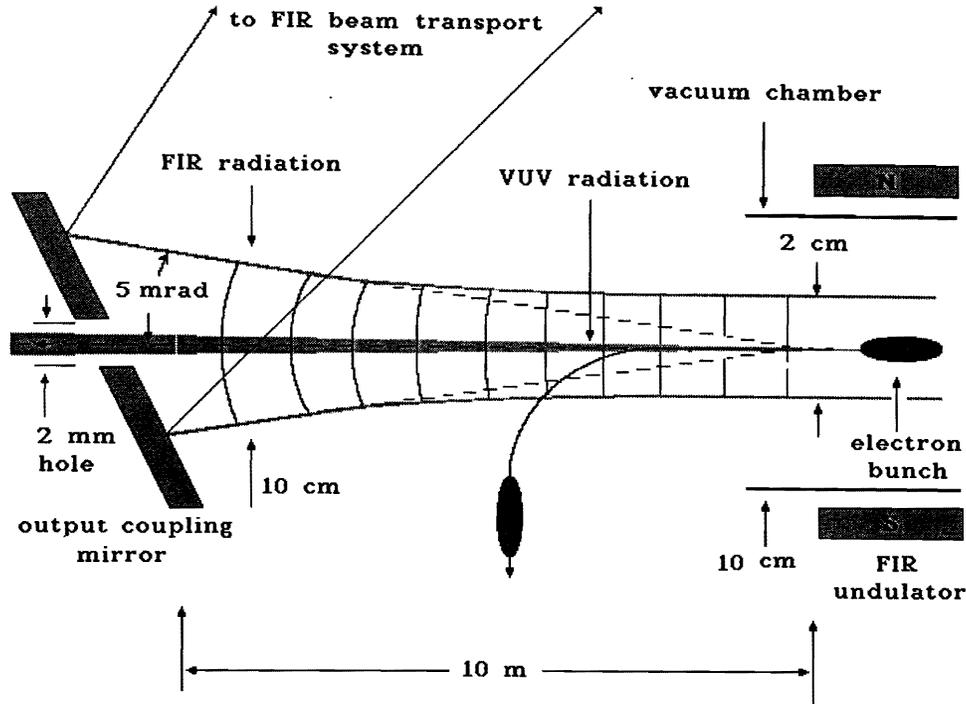


Fig. 3. Schematic of a mirror-coupling arrangement

Due to the angle-frequency correlation of the coherent undulator radiation the required frequency bandwidth of radiation could be provided by angular selection. The average solid angle accepted by a coupling mirror of radius  $a$  located at distance  $D$  from the downstream end of the second undulator of length  $L$  is

$$\langle d\Omega \rangle = \frac{\pi a^2}{D(D+L)} .$$

To provide a natural selection of coherent radiation in the central cone ( $\theta_{\text{con}} \simeq 5 \text{ mrad}$ ), the radius of the mirror should be equal to 6 cm at a distance of about 10 m between the exit of the second undulator and the mirror (see Fig. 3).

The operation of the proposed FIR source is insensitive to the emittance of the electron beam, since the condition of optimal electron beam transverse size is:

$$\sigma^2 \ll Lc/\omega \simeq 1 \text{ cm}^2 .$$

The analysis of the parameters of the FIR source has shown that it will operate reliably even for an emittance exceeding the project value of  $2\pi \text{ mm mrad}$  by two orders of magnitude.

An undulator is a sequence of bending magnets where particles with different energies have different path length. As a result, the energy spread in the beam leads to the bunch lengthening in the undulator. When an electron bunch passes the FIR undulator,

radiation interaction induces additional energy spread in the electron beam which also can lead to bunch lengthening. Recently, the problem connected with radiative interaction of the particles in a line-charge microbunch moving in an undulator has been investigated analytically [11]. In the case of a Gaussian bunch profile the induced energy spread  $\Delta\mathcal{E}_f$  is given by:

$$\frac{\Delta\mathcal{E}_f}{\mathcal{E}} = \frac{r_e N K^2 L}{\sqrt{2\pi}\sigma_z^2 \gamma^3} G(p, K), \quad (4)$$

where,  $r_e$  is the classical electron radius,  $p = k_w \sigma_z \gamma^2 / [(1 + K^2/2)]$  is the bunch length parameter, and  $\sigma_z$  is the rms bunch length. Parameter of the FIR source project are:  $N = 6 \cdot 10^9$ ,  $\sigma_z = 50 \mu\text{m}$ ,  $K = 50$  (for  $100 \mu\text{m}$  wavelength),  $L = 6 \text{ m}$ , and  $\gamma = 2 \cdot 10^3$ . The value of  $G$  is  $G \simeq 0.5$ . Substituting these values into (4), we obtain an induced correlated energy spread at the exit of undulator  $\Delta\mathcal{E}_f/\mathcal{E} \simeq 1\%$ . This leads to an increase of the bunch length:

$$\Delta l_b \simeq \frac{L(1 + K^2/2)}{2\gamma^2} \frac{\Delta\mathcal{E}_f}{\mathcal{E}} \simeq 10 \mu\text{m}.$$

Since this value is much less than the radiation wavelength  $\lambda \sim 100 \mu\text{m}$ , we can conclude that bunch decompression in the undulator due to induced energy spread should not be a serious limitation in our case.

## 5 Optical beam transport

The FIR radiation must propagate at least 30 meters to reach the experimental hall; then, depending on the location of the experiment, it must travel an additional 10–20 meters within the hall. When the far-infrared optical beam propagates in vacuum, it

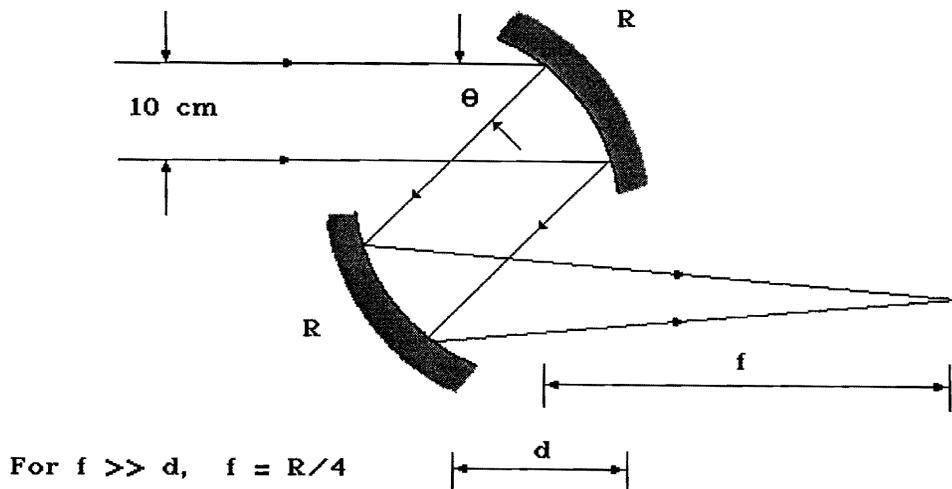


Fig. 4. Arrangement for the FIR focusing module with reflective optics

diverges due to diffraction effects. A suitable beam transport system must be provided to guide the beam over a distance up to 50 meters, at the same time maintaining a reasonable transverse beam size. Application of reflective optics is an optimal solution for the optical transmission line for the wavelength range of 100–300  $\mu\text{m}$  [8]. For maximum flexibility, all transport optics are high-reflectivity, overcoated copper mirrors. FIR transport system can be designed using focusing modules presented schematically in Fig. 4. Each module consists of a pair of matched copper mirrors, separated such that the angle  $\theta$  is less than  $10^\circ$  to minimize aberrations. In this arrangement, the effective focal length of the system is given by  $f \simeq R/4$ , where  $R$  is the radius of mirror curvature. The entire beam path will be evacuated to  $10^{-6}$  Torr to minimize absorption losses. A Brewster-angle exit window of an appropriate material (polyethelene) separates the transport system vacuum from the experimental devices.

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