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# Schemes for Time-Resolved Experiments at the TTF FEL

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## Abstract

The paper describes schemes of two-color time-resolved experiments that could be performed at the soft X-ray self-amplified spontaneous emission free electron laser (SASE FEL) at the TESLA Test Facility (TTF) at DESY and determines what additional FEL hardware and instrumentation developments will be required to bring these experiments to fruition.

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## 1 Introduction

The field of synchrotron radiation research has grown rapidly over the last 30 years due to advances in the development of the electron (positron) storage ring technology. Three successive generations of synchrotron radiation facilities have provided an increase in brilliance by more than ten orders of magnitude. However, the storage ring technology itself approaches its theoretical limits of performance with respect to average and peak brilliance, as well as to pulse duration. Recently a new era of synchrotron radiation research has begun with first user experiments on a free electron laser (FEL) based on self-amplified spontaneous emission (SASE). The results have been obtained at the Tesla Test Facility (TTF) at DESY [1,2] using radiation pulses of 80-120 nm wavelength with 30-100 fs pulse duration and a peak power in the GW range [3]. Compared to present day synchrotron radiation sources its peak brilliance is more than 100 million times higher, the radiation has full transverse coherence and the pulse duration is reduced from the 100 picoseconds down to the sub-100 femtosecond time domain. The TTF is currently extended to cover the soft X-ray spectral range down to wavelengths of 6 nm. Commissioning of this facility will start in the year 2003 [4].

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The discussion in the scientific community over the last two years has produced many ideas for novel applications using the soft X-ray SASE FEL at the TESLA Test Facility. Brilliance, coherence, and timing down to the femtosecond regime are the three properties which have the highest potential for new science to be explored with the soft X-ray FEL.

The standard technique for high-resolution time-resolved measurements is a pump-probe scheme in which a process is started by a short pulse of radiation (pump) and the evolution of the process is then observed (probed) with different delay after the start by means of a second pulse of radiation, generally at another photon energy. This paper describes schemes of two-color time-resolved experiments that could be performed at the soft X-ray SASE FEL at the TTF and determines what FEL hardware and instrumentation developments will be required to bring these experiments to fruition. It shows also which concepts are compatible with the layout of the TTF FEL, and may thus serve as a basis for the discussion of further developments at the TTF.

Four types of pump-probe techniques have been considered for the TTF FEL:

(1) independent pump and probe light sources employing a powerful optical laser system synchronized with the FEL [5],

(2) pump and probe beams that are generated by the same electron bunch from the same insertion device [6]

(3) pump and probe beams that are generated by the same electron bunch but from two insertion devices [7,8]

(4) concepts for improving the time resolution to the 10 fs level which are based on the generation of femtosecond pulses by sideband-seeded soft X-ray SASE FEL [9].

The synchronization of an optical laser with the soft X-ray FEL pulses is the most challenging task of the first type of pump-probe technique. At present the best achieved synchronization between two independent sources is of the order of 1 ps. Picosecond time-resolved work can be performed using known techniques. Sub-picosecond synchronization, on the other hand, needs further development.

The synchronization problem can be eliminated by generating the two different frequencies (colors) by the same electron bunch. The second and third concept are based on this idea. However, these approaches are limited in the level of the SASE FEL pulse duration (100 fs) that can be achieved. The 4th approach, like the first, is based on the combination of the soft X-ray SASE FEL with a femtosecond laser. However, in the proposed scheme the femtosecond soft X-ray pulse is naturally synchronized with the femtosecond optical pulse, there is no jitter between them (see section 5). It should be possible to achieve a soft X-ray pulse duration and timing accuracy close to the duration of the seed laser pulse, i.e. of the order of 10 fs. Nevertheless, many applications require even shorter soft X-ray pulses. In section 3 we propose a new type of pump-probe technique which is based on the use of the statistical properties of the third harmonic radiation from the SASE FEL. Ultra-short pulses at the third harmonic are selected by a trigger in the data acquisition system. The main advantage of this scheme is the absence of physical limitations which would prevent operation at much shorter pulse duration close to the coherence time, i.e. a few femtoseconds in the case of the TTF FEL.

The development of a versatile pump-probe facility at the TTF FEL will give us an entirely new tool offering an enormous range of research possibilities. Two-color time-resolved experiments in the range from 6 nm up to 300 000 nm with a pulse duration down to a few femtoseconds will provide unique possibilities for the study of the dynamics of electronic excitations, chemical reactions, and phase transitions of matter.

## 2 Pump-probe experiments combining an external optical laser and a soft X-ray SASE FEL

Two-color pump-probe experiments combining optical femtosecond lasers with short wavelength radiation from the FEL are very attractive for sub-picosecond resolved studies. For applications in the visible and near-visible wavelength range a pump-probe facility based on a conventional quantum laser system will be available at the TTF [5]. The scheme for two-color time-resolved experiments is shown in Fig. 1. The laser system comprises a seed pulse laser, special synchronization with the accelerator, pulse shaping, and a pump laser together with an optical parametric amplifier (OPA) (see Fig. 2). The laser will initially cover the spectral region between 750 and 900 nm and will provide a train of 100 MW-level pulses with 100 fs pulse duration. This requires the development and construction of an OPA, pumped by an unique, frequency doubled Nd:YLF laser with 10 ps pulse duration. Grating combinations will be used to adapt the duration of the seed pulses to the pulse duration of the pump laser and compress them after parametric amplification (CPA Chirped-Pulse Amplification). The laser system has been designed in such a way that it can be extended later to cover a wider spectral range and to reduce the pulse duration to the 10 fs level.

The main technical problems of this project are the development of the high power OPA and the synchronization system. The SASE FEL at the TTF can produce radiation pulses shorter than 100 fs [3], hence the synchronization should be better, possibly down to a few 10 femtoseconds. The best synchronization between two sources achieved to date is of the order of a picosecond. Even if femtosecond pulses from the laser system were synchronized to the photoinjector master clock by phase-locking, the main problem is the time jitter of the electron pulses. The latter is produced in the magnetic bunch compressors and is estimated to ±1 ps for the expected ±0.1% energy jitter of the electron bunch. One possible approach to cope with imperfect synchronization is to measure the exact time delay between pump and probe pulses by using single-pulse correlation techniques. It is in fact part of the present project to adopt such techniques to the special requirements at the TTF FEL. On the other hand, the stretched optical pulses at the output of the OPA could also be used for seeding a FEL amplifier in the (near) visible range as described below in section 4. In this case standard techniques can be used for the electronic synchronization and, in particular, the timing accuracy between the pump and probe pulses is no longer affected by the time jitter coming from the accelerator.

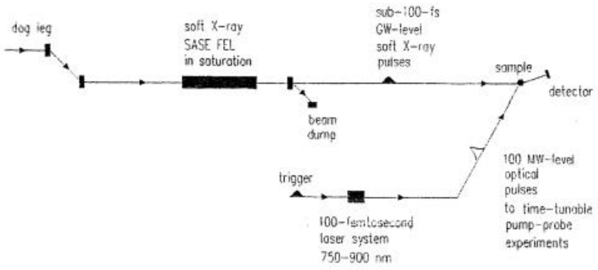

Fig. 1. Scheme for pump-probe experiments with sub-picosecond time resolution combining an external high-power optical laser and a soft X-ray SASE FEL

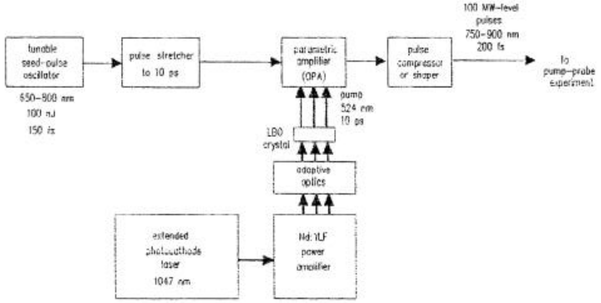

Fig. 2. Basic scheme of the external laser system for TTF-FEL pump-probe set-up

## 3 Pump-probe experiments employing soft X-ray pulses generated by a single bunch in the same insertion device

### *3.1 Single-color pump and probe beams by splitting the SASE FEL radiation*

Perfect timing of two radiation pulses for high resolution time-resolved measurements can only be achieved if the two pulses are derived from the same source using a beam splitter. Therefore, the simplest experiment would be a single-colour experiment in which a fraction of the FEL pulse excites (pumps) a system and another fraction of the same pulse probes it after a certain delay time. A typical application would be the measurement of the lifetime of an excited state: The pump pulse excites the system under study into a well-defined energy state, selected by the photon energy, and the probe pulse determines after a certain time if the state is still occupied, for example by detecting photoelectrons.

For this experiment one needs only a beam splitter and mirrors for delaying and recombining the two beams on the sample. The beam splitter is a crucial element. It has to withstand the high peak and average power of the FEL beam, must not produce diffraction e.g. by sharp edges, must not stretch the pulses, and it should be highly efficient. This is not trivial for the spectral range of the TTF FEL, approximately 6-100 nm wavelength, and needs further development, based on previous work concerned with synchrotron radiation optics and ultra-short high-order harmonic pulses [10,11]. Although they are very important, we will not dwell on these more technical aspects in this paper but rather concentrate on the general concepts for pump-probe experiments that can be done at the TTF.

### *3.2 Pump-probe experiments combining the first and the third harmonics of the SASE FEL*

SASE FELs are capable to produce powerful radiation not only at the fundamental frequency, but also at higher harmonics. When a beam is strongly bunched in the sinusoidal ponderomotive potential (formed by the undulator field and the radiation field of the fundamental frequency), the electron beam density spectrum develops rich harmonic contents. Coherent radiation at the odd harmonics can be generated in a planar undulator and significant power levels for the third harmonic can be reached before the FEL saturates [12]. It is expected that the power of the transversely coherent third-harmonic radiation can approach 1% of the fundamental power level at the TTF FEL [13,14]. It is therefore proposed to combine the GW-level SASE FEL pulses at the fundamental frequency and the 10 MW-level third-harmonic radiation pulses from the same SASE FEL for high resolution pump-probe experiments. Since the nonlinear harmonic generation occurs naturally in a planar undulator, no special synchronization nor any additional FEL hardware components are required for these experiments. It is necessary, of course, to develop optical components to select the 1st and the 3rd harmonic pulses and combine them

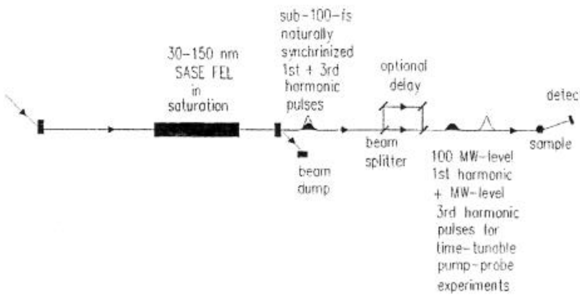

Fig. 3. Scheme for pump-probe experiments employing 1st and 3rd harmonic pulses from a SASE FEL

with a tunable delay. The experimental arrangement is schematically shown in Fig. 3.

### *3.3 Femtosecond resolution by employing the statistical properties of the third harmonic of the SASE FEL radiation*

A closer inspection of the statistical properties of the third harmonic radiation from an SASE FEL reveals that it is possible to select single, temporally coherent radiation spikes by using a simple intensity trigger. A carefully designed optical system for splitting, delaying, filtering, and recombining the radiation would then allow time-resolved measurements with a resolution down to the coherence time of the FEL, i.e. a few femtoseconds in the case of the TTF FEL. In principle, it could be possible to measure directly the liftime of atomic core levels in the soft X-ray region. For example, the lifetime of the silicon 2p level at $\simeq 100$ eV is approximatelly 30 fs, and the expected duration of a third harmonic FEL pulse at 100 eV photon energy is about 6 fs.

The selection of a single third harmonic radiation spike is based on the specific, instantaneous intensity variation within the radiation pulse. This is illustrated in Fig. 4 for a simple example. Consider the intensity function $I(t)$ of a SASE FEL pulse at the fundamental frequency shown in Fig. 4 (a). In the linear gain regime of the SASE FEL, the instantaneous intensity of the third harmonic is proportional to $[I(t)]^3$ plotted in Fig. 4 (b). Due to the nonlinear transformation the intensity variation becomes much more pronounced, leading to a distribution which is dominated by a single spike in this case. In practice this will not occur for every pulse, and for those cases where a single spike is left, we expect a large intensity fluctuation from pulse to pulse. The main question is how likely we are to observe a single bright spike in the intensity of the third harmonic radiation. Clearly, a necessary condition for this event is that the energy $E_{(3)}$ in the third-harmonic radiation pulse is much

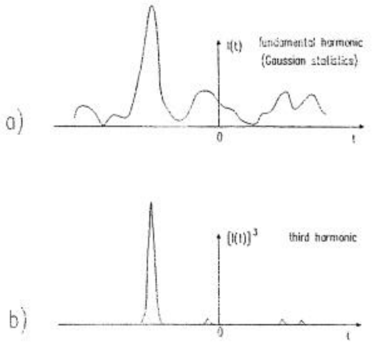

Fig. 4. Illustration of the results of a nonlinear transformation (a) sample function of fundamental harmonic instantaneous intensity for SASE FEL; (b) the nonlinear transform of Fig. (a) representing the third harmonic instantaneous intensity

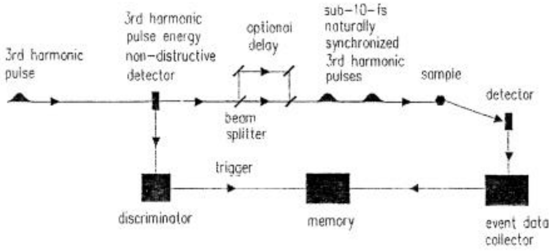

Fig. 5. Experimental setup for femtosecond time-resolved measurements. A non-destructive third-harmonic pulse-energy detector is used as a trigger rejecting events with $E_{(3)}$ smaller than 10 $\langle E_{(3)} \rangle$

larger than the average energy $\langle E_{(3)} \rangle$. The results of numerical simulations confirm these simple physical considerations. The predicted probability for the TTF FEL is about 1 %. This is completely acceptable for practical applications because the superconducting TTF accelerator can deliver up to 72 000 electron bunches per second such that a kHz-level average repetition rate of femtosecond third harmonic pulses can be obtained. The statistical properties of the SASE FEL harmonic radiation is outlined in more detail in the Appendix.

In practice single-spike third harmonic pulses can be identified by measuring their total pulse energy $E_{(3)}$ using a non-destructive technique such as gas ionization. The schematic arrangement of a pump-probe experiment employing these femtosecond pulses is shown in Fig. 5. A multilayer singles out the third harmonic radiation, a gas cell at low pressure measures the pulse energy, then the pulse is split into two parts which are delayed with respect to each other and then recombined on the sample. Only such data are selected for which the pulse energy exceeded a certain threshold, typically of the order of 10 $\langle E_{(3)} \rangle$.

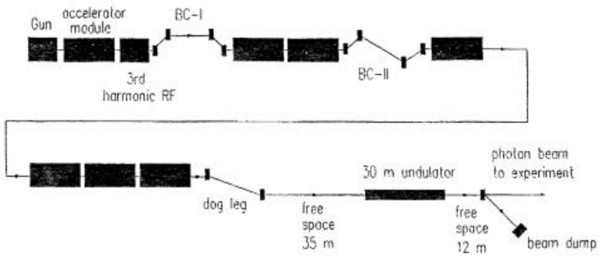

Fig. 6. Schematic layout of the soft X-ray SASE FEL facility

## 4 Pump-probe experiments employing optical and soft X-ray pulses that are generated by the same electron bunch in two different insertion devices

Sections 2 and 3 described pump-probe schemes for which no additional FEL hardware components are required. The following schemes employ additional undulators to generate the second colour radiation pulses. They require free space in front of and behind the main soft X-ray FEL undulator, and one has to make sure that the arrangement is compatible with the overal layout of the TTF FEL. Fig. 6 shows the installations of the soft X-ray FEL in the first phase, including the linear accelerator with two magnetic bunch compressors and six undulator modules at the end of the accelerator tunnel. The first goal is to reach saturation in the soft X-ray range with this configuration, using the six undulator modules in SASE mode [1]. In a second step the 35 m free space in front of the undulator will be used to build a fully coherent soft X-ray facility based on the two undulator self-seeding concept [15,16].

### *4.1 The soft X-ray SASE FEL combined with a FEL amplifier seeded by a long optical laser pulse*

The synchronization problem of a separate optical laser with the FEL (see section 2) can be avoided if the optical pulse is produced by the same electron bunch which also generates the soft X-ray FEL pulse. This can be done by using an optical laser

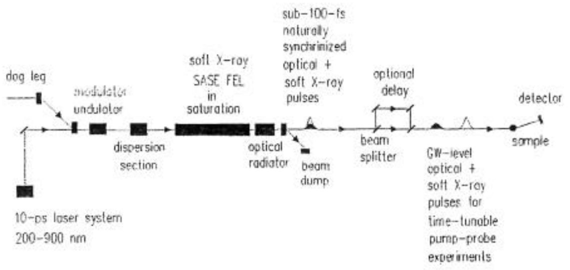

Fig. 7. Scheme for pump-probe experiments employing an optical pulse as a pump and a soft X-ray SASE pulse as a probe or vice versa. A very long laser pulse is used for modulation of the energy and density of the electrons at optical frequency. Optical photons for pump-probe experiments are generated by an additional insertion device (optical radiator) using the same electron bunch

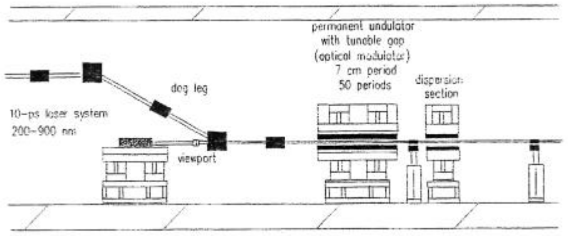

Fig. 8. Side view of the electron beam transport system, showing the location of the seed laser and the modulator

as a seed laser for a FEL amplifier tuned to the optical frequency. This concept is schematically shown in Fig. 7. We propose to separate the optical FEL into a modulator in front of the main soft X-ray SASE FEL and a radiator behind. The electron beam density can be modulated in a wide spectral range by using the synchronized optical laser described in section 2 (or a similar system) as a seed laser for an FEL amplifier which operates in the linear regime. The density modulation is only moderate and does not disturb the SASE process at short wavelengths in the main undulator. Although the electron beam leaving the soft X-ray FEL has acquired some additional energy spread, it is still a good "active medium" for an optical radiator at the end. As a result this optical source will provide intense, tunable and coherent radiation in the spectral range between approximately 200 nm and 900 nm. The pulse duration is the same as that of the soft X-ray SASE FEL, i.e. in the sub-100 fs range, the peak power is a few GW and the spectral width is transform-limited.

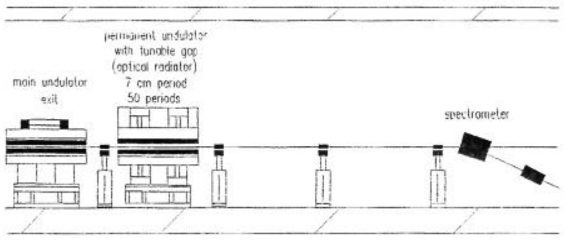

Fig. 9. Side view of the electron beam transport system, showing the location of the optical radiator

The facility consists of the seed laser, the modulator undulator, a dispersion section, the main soft X-ray SASE undulator, and the optical radiator. Figures 8 and 9 show the location of additional hardware components. The first stage is a conventional FEL amplifier seeded by the optical laser. A very long laser pulse of the order of 10 ps or more to be insensitive to any time jitter - is used for modulating the energy and density of the electron bunch at the optical frequency. The input laser power required for effective seeding is approximately 1 MW. The radiation power is exponentially amplified upon passing through the first undulator. The amplitude of the energy modulation of the electron beam at the undulator exit is about 0.4 MeV, while the beam density modulation is about 5%. The density modulation at the optical frequency is increased to the required value of about 20% when the electron beam passes through the dispersion section. The energy modulation, introduced by the modulator, is smaller than the initial energy spread of 1 MeV. As a result, the process of amplification of the radiation in the main (soft X-ray) undulator develops in the same way as in the conventional SASE FEL: fluctuations of the electron beam current density serve as input signal. The seeding optical radiation does not interact with the electron beam in the main undulator and is diffracted out of the electron beam. Optical pulses for pump-probe experiments are generated when the same modulated electron bunch passes through an additional insertion device, the optical radiator. The function of the optical radiator is to prepare very short optical pulses (100 fs) strictly synchronized with the electron bunch. The optical radiator is a conventional FEL amplifier seeded by the density modulation in the electron bunch. The amplitude of the density modulation at the optical frequency, about 10%, is much larger than the amplitude of shot noise harmonics of about 0.01%. This density modulation serves as an input signal for the optical radiator which is resonant to the seed laser frequency. The radiation is exponentially amplified along the optical radiator. The input signal is high enough to reach saturation.

An important feature of the proposed scheme is that the optical radiator uses the spent electron beam from the soft X-ray FEL. Therefore the optical radiator can operate at saturation without interfering with the soft X-ray SASE FEL. Since the optical radiator generates at least 10 times longer wavelengths compared with the main SASE FEL, the acceptable energy spread of the electron beam is rather large (1 %) and allows the effective generation of powerful radiation in the optical radiator.

### 4.2 The soft X-ray FEL combined with a far-infrared source with laser like characteristics

Another, very promising possibility is the combination of the soft X-ray FEL with an unique new source of far-infrared (FIR) radiation, giving unprecedent insight into the spectroscopy of gas-phase free radicals, cluster energy and dynamics, vibrational structure of highly excited and superexcited molecules.

The TTF is a facility producing sub-picosecond electron bunches (50 $\mu$m rms) for the generation of soft X-ray radiation in a SASE FEL. Utilizing these electron bunches can also provide a reliable and easily tunable powerful source of FIR radiation for scientific applications [8]. Intense, coherent, far-infrared (FIR) radiation can be produced from 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 radiative 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 $10^{10}$ electrons in each bunch, the radiation intensity is enhanced by this enormously large factor over the incoherent radiation.

It is important to stress that the resulting shape (spectrum) of FIR pulses can be well described analytically. For example, for the radiation within the central cone the relative spectral bandwidth is equal to $\Delta\omega/\omega \simeq 1/N_{\mathrm{w}}$, where $N_{\mathrm{w}}$ is the number of undulator periods. The energy radiated into the central cone in the case when the resonance wavelength $\lambda$ is much longer than the bunch length and the undulator parameter $K \gg 1$, is given by $\Delta E_{\mathrm{con}} \simeq \pi e^2 \omega_0 N_e^2/c$ where $e$ is the charge of the electron, $2\pi c/\omega_0 = \lambda$, $N_e$ is the number of electrons in the bunch [8]. When the electron bunch moves through the undulator, the electromagnetic wave advances the electron bunch by one wavelength after one undulator period. As a result, the FIR pulse duration is equal to $N_{\mathrm{w}}\lambda/c$. For the case of the TTF the parameters for the FIR radiation at a wavelength $\lambda = 300$ $\mu$m are 10 ps pulse duration, 10 % spectral bandwidth, 1 mJ radiation energy, and 100 MW peak power. The FIR source at the TTF can also operate at shorter wavelengths. In the range 30-100 $\mu$m the peak power would still be about 100 kW at a pulse duration of 1-3 ps which is comparable to operating IR FEL oscillators.

Figures 10 and 11 show a FIR coherent source integrated into the TTF FEL user facility. In this conceptual design we assume to use a conventional electromagnetic undulator with 10 periods, each 60 cm long. Operating the FIR source around 200 $\mu$m wavelength requires a peak value of the magnetic field of about 1.3 T. The FIR

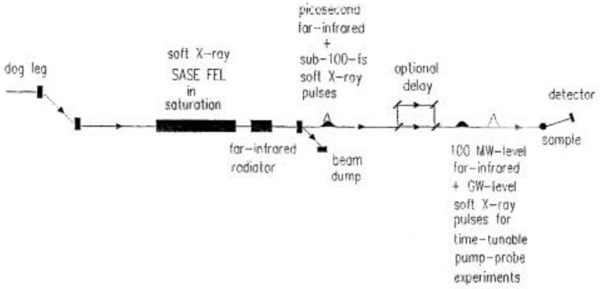

Fig. 10. Scheme for pump-probe experiments combining high-power far-infrared source with laser-like characteristics and soft X-ray SASE FEL. The far-infrared source will use spent electron beam coming from the SASE FEL. Intense, coherent far-infrared undulator radiation can be produced from electron bunches at wavelength longer or equal to the bunch length

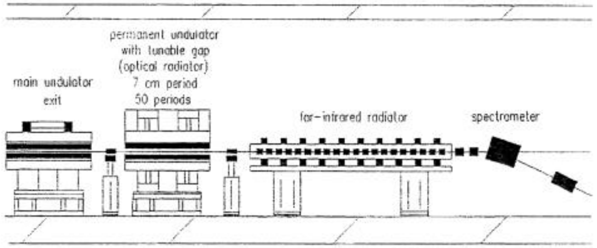

Fig. 11. Side view of the electron beam transport system, showing the location of the far-infrared radiator

source would use the spent electron beam coming from the soft X-ray SASE FEL which allows one to extend significantly the scientific potential of the TTF without interfering with the soft X-ray TTF FEL operation.

The pump-probe technique is one of the most promising methods for the application of the high-power FIR source. The TTF will allow, for the first time in the world, the integration of a FIR source and a soft X-ray beam line. The pulses of the coherent FIR radiation are naturally synchronized with the soft X-ray pulses from the main TTF FEL, enabling pump-probe techniques using either the FEL pulse as a pump and the FIR pulse as a probe, or vice versa.

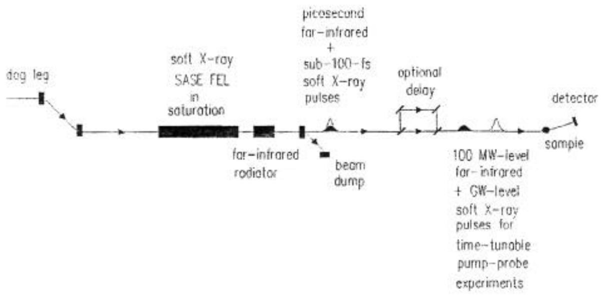

Fig. 10. Scheme for pump-probe experiments combining high-power far-infrared source with laser-like characteristics and soft X-ray SASE FEL. The far-infrared source will use spent electron beam coming from the SASE FEL. Intense, coherent far-infrared undulator radiation can be produced from electron bunches at wavelength longer or equal to the bunch length

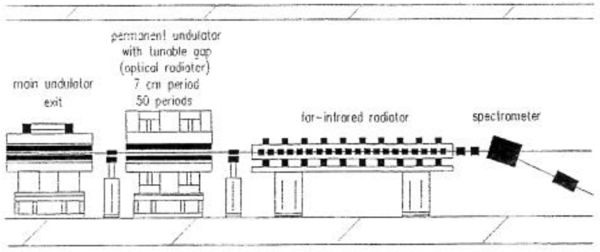

Fig. 11. Side view of the electron beam transport system, showing the location of the far-infrared radiator

source would use the spent electron beam coming from the soft X-ray SASE FEL which allows one to extend significantly the scientific potential of the TTF without interfering with the soft X-ray TTF FEL operation.

The pump-probe technique is one of the most promising methods for the application of the high-power FIR source. The TTF will allow, for the first time in the world, the integration of a FIR source and a soft X-ray beam line. The pulses of the coherent FIR radiation are naturally synchronized with the soft X-ray pulses from the main TTF FEL, enabling pump-probe techniques using either the FEL pulse as a pump and the FIR pulse as a probe, or vice versa.

## 5 Pump-probe experiments using 10 fs pulses from a sideband-seeded soft X-ray SASE FEL

For many applications the typical FEL pulse duration of 100 fs is too long. While 10 fs optical laser pulses are routinely available using CPA techniques, the FEL pulse duration is coupled with the electron bunch length which cannot easily be reduced to the 10 fs level due to space charge limitations. However, it is possible to generate soft X-ray FEL pulses shorter than the electron bunch length by using the sideband seeding technique.

This technique is based on the combination of the soft X-ray SASE FEL and a fs quantum laser [9]. The operation of the sideband seeded SASE FEL is illustrated with Figs. 12 - 14. An ultrashort laser pulse is used to modulate the electron density within a femtosecond slice of the electron bunch at a frequency $\omega_{\mathrm{opt}}$. We assume that the seed laser pulse will be timed to overlap with the central area of the electron bunch. This ultrashort laser pulse serves as a seed for a modulator which consists of an undulator and a dispersion section. The modulator/dispersion section is identical with the one described in section 4.1. The interaction of the seed laser pulse with the electron beam produces an energy modulation at $\omega_{\mathrm{opt}}$ in the central part of the electron bunch. This energy modulation is converted into a density modulation in the dispersion section. The electron beam then enters the main SASE undulator and produces soft X-ray SASE radiation at the resonant frequency $\omega_0$. The SASE process modulates the electron density at frequency $\omega_0$ leading to an amplitude modulation at the sidebands $(\omega_0 \pm \omega_{\mathrm{opt}})$ in that part of the electron bunch that overlaps with the seed laser pulse. This part is much shorter than the electron bunch. Following the SASE FEL undulator the beam enters an undulator section (radiator) which is resonant with the $\omega_0 - \omega_{\mathrm{opt}}$ radiation. Because the beam has a relatively large component of bunching at the long wavelength sideband, coherent emission at $\omega_0 - \omega_{\mathrm{opt}}$ is copiously produced within a femtosecond slice of the electron bunch. After leaving the radiator the electron beam is deflected into a beam dump, while the photon beam enters a time-compensated monochromator, which selects the femtosecond soft X-ray pulse.

We illustrate the sideband seeding for the parameters of the TTF FEL operating at a wavelength of 20 nm. The sideband seeded FEL has been optimized by using the three-dimensional time-dependent code FAST [17] taking into account all physical effects influencing the FEL amplifier operation such as diffractive effects, energy spread, emittance, slippage effect, etc. The parameters of the optical laser are: 400 nm wavelength, 6 $\mu$J energy in the laser pulse, and 25 fs FWHM pulse duration. The laser beam is focused onto the electron beam in a short (5 periods) undulator resonant at a wavelength of 400 nm. Due to the resonant interaction of the electron beam with the optical field in the undulator the electron energy is modulated. The subsequent dispersion section transforms the energy modulation into a density modulation.

The optimum length of the main SASE undulator is given by the condition of maximum spectral purity of the sideband. In the case under study the optimum

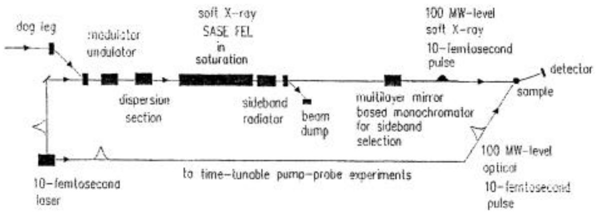

Fig. 12. Scheme for pump-probe experiments based on the generation of femtosecond pulses by a sideband-seeded soft X-ray FEL. In this scheme a femtosecond soft X-ray pulse is naturally synchronized with the femtosecond optical pulse from seed laser

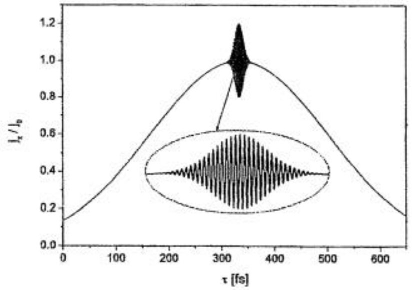

Fig. 13. Density modulation of the electron beam at the exit of the dispersion section

length of the main undulator is equal to 10 m. Figs. 15 and 16 demonstrate the high contrast of the fs radiation pulse of about 100:1. The fine structure in the central part of the pulse is due to the interference of the radiation from the main undulator with frequency $\omega_0$ and from the sideband radiator with frequency $\omega_0 - \omega_{\mathrm{opt}}$. Figure 17 shows the spectral distribution of the radiation at the exit of the sideband radiator. It is seen from Fig. 16 that after a 5 m long sideband radiator the radiation power in the fs pulse reaches a level of a few GW, and the energy in the fs pulse reaches a value of about 30 $\mu$J. Further increase of the length of the radiator does not lead to significant increase of the power, while the contrast of the fs pulse starts to reduce drastically because the sideband intensity due to noise increases in the nonlinear regime.

The 10 fs pulse at the sideband frequency is separated from the 100 fs SASE pulse at the central frequency by a monochromator. One has to take care that the short pulse duration is preserved. In principle, it is possible to design time-compensated

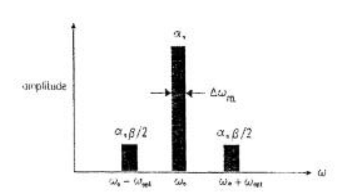

Fig. 14. The development of sidebands for an initial density modulation at $\omega_{\mathrm{opt}}$. The density modulation of the electron bunch at the exit of the SASE FEL has the form $\alpha_0(1 + \beta\cos(\omega_{\mathrm{opt}}t + c))\cos(\omega_0 t + d)$

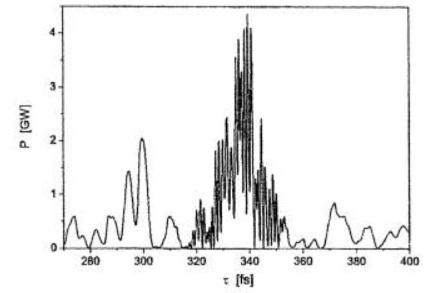

Fig. 15. Total radiation pulse at the exit of the sideband radiator. The length of the radiator undulator is equal to 5 m

spectroscopic configurations with gratings by using at least two gratings in a substrative fashion to compensate for the dispersion. This concept is well known in the study of ultrafast laser pulses, and has recently been extended to the soft X-ray region [10].

Femtosecond sideband radiation pulses can also be selected by using plane multilayer mirrors. The design of a time-compensated monochromator based on multilayers has been presented in [11]. A schematic illustration is shown in Fig. 18. The monochromator is useful in the 6-35 nm region, where multilayer mirrors have good performance. The pulse duration is not altered up to a few fs, preserving the time resolution capability. In our case, the central peak-to-sideband separation has a value of 5%. Because the SASE FEL bandwidth (about 0.5% FWHM) is much less than the separation of the sidebands from the main peak, (see Fig. 17) we obtain a clean fs pulse after the monochromator. It should be noted that the calculations of the radiation power (see Figs. 15 and 16) assumed an ideal monochromator with

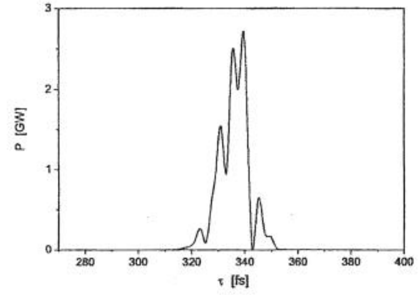

Fig. 16. The output radiation pulse of Fig 15 after spectral filtering at the sideband frequency. The calculations assumed an ideal monochromator

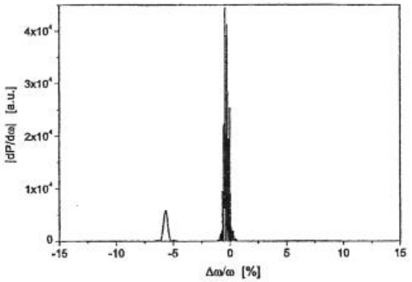

Fig. 17. Spectral distribution of the radiation behind the sideband radiator. The length of the radiator undulator is equal to 5 m

100 % transmission. The efficiency of a multilayer mirror monochromator is about 10% only, so the radiation power available for user experiments is roughly by one order of magnitude less than that shown in Figs. 15, 16.

The sideband in the SASE FEL is seeded by positioning a fs laser pulse on the electron bunch. As we discussed above, it is expected that a time jitter of the order of 1 ps will remain due to the energy jitter of the electron beam of $\simeq 0.1$ %. Due to this uncertainty not every fs optical pulse will produce a fs soft X-ray pulse. The predicted probability of positioning the interaction region on the electron bunch is about 10 % only. But although the fs soft X-ray pulses are randomly produced, they are always exactly timed relative to the fs optical seed pulse. This means that the sideband soft X-ray pulse transmitted by the monochromator can be combined with the optical seed pulse for femtosecond resolution pump-probe experiments.

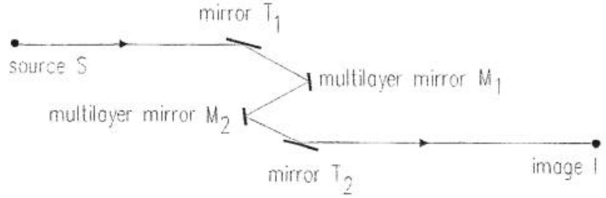

Fig. 18. Schematic view of the time-compensated monochromator for sideband selection using two anti-parallel multilayer mirrors set to the Bragg-angle of the sideband frequency. The monochromator works in parallel light. The frequency can be tuned by changing the Bragg-angle, the direction of the exit beam remains constant

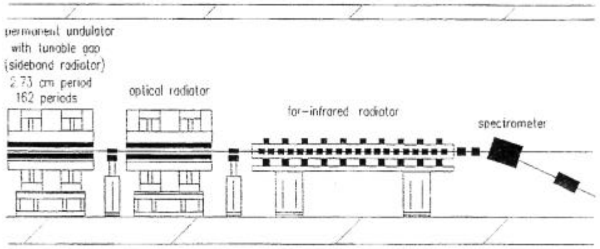

Fig. 19. Side view of the electron beam transport system, showing the location of the sideband radiator just behind the main soft X-ray FEL undulator. It is followed by the optical radiator and the FIR radiator discussed in section 4

## 6 Appendix. The statistical properties of the third harmonic of SASE FEL radiation

SASE radiation is a stochastic object and at a given time it is impossible to predict the amount of energy which flows to a detector. First we should remember that the initial modulation of the electron beam is defined by the shot noise and has a white spectrum. Second, we obtain that the high-gain FEL amplifier cuts out and amplifies only a narrow frequency band of the initial spectrum $\Delta\omega/\omega \ll 1$. In the time domain, the temporal structure of the fundamental harmonic radiation is chaotic with many random spikes, with a typical duration given by the inverse width of the spectrum envelope. Even without performing numerical simulations, we can describe some general properties of the fundamental harmonic of the radiation from the SASE FEL operating in the linear regime. Indeed, in this case we deal with Gaussian statistics. As a result, the probability distribution of the instantaneous radiation intensity $I$ should be the negative exponential probability density distribution: $p(I) = \langle I\rangle^{-1}\exp(-I/\langle I\rangle)$. Here one should realize clearly that the notion of instantaneous intensity refers to a certain moment in time, and that the analysis must be performed over an ensemble of pulses. Also the energy in the radiation pulse $E$ should fluctuate in accordance with the gamma distribution [18]:

$$p(E) = \frac{M^M}{\Gamma(M)}\left(\frac{E}{\langle E\rangle}\right)^{M-1}\frac{1}{\langle E\rangle}\exp\left(-M\frac{E}{\langle E\rangle}\right) ,$$

where $\Gamma(M)$ is the gamma function of argument $M$, and $1/M = \langle(E - \langle E\rangle)^2\rangle/\langle E\rangle^2$ is the normalized dispersion of the energy distribution. These properties are well known in statistical optics as properties of completely chaotic polarized radiation [19].

Let us start to discuss the statistical properties of the third-harmonic radiation in a SASE FEL. It should be noted that the statistics of the third-harmonic radiation from the SASE FEL changes significantly with respect to the fundamental harmonic (e.g., with respect to Gaussian statistics). It is interesting in our case to be able to determine the probability density function of the instantaneous intensity of SASE radiation after it has been subjected to the nonlinear transformation. We know the probability density function $p(I) = \langle I\rangle^{-1}\exp(-I/\langle I\rangle)$ of the fundamental intensity $I$, and $I$ is subjected to a transformation $z = (I)^3$. The problem is then to find the probability density function $p(z)$. It can be readily shown that $p(z) = (3\langle I\rangle)^{-1}z^{-2/3}\exp(-z^{1/3}/\langle I\rangle)$ Using this distribution we get the expression for the mean value: $\langle z\rangle = 6\langle I\rangle^3$. Thus the third-harmonic radiation for the SASE FEL has an intensity level roughly 6 times larger than the corresponding steady-state case, but with more shot-to-shot fluctuations compared to the fundamental [14]. This nontrivial behavior of the intensity of the third harmonic reflects the complicated nonlinear transformation of the fundamental harmonic statistics. One can see that Gaussian statistics is no longer valid.

Because of our lack of knowledge of the detailed microscopic structure of the intensity profile of the radiation pulse, it is necessary to discuss the properties of single-spike selection in statistical terms. The statistics of concern are defined over an ensemble of radiation pulses. If we define the contrast $C$ as the ratio of the number of photons in the main spike to the total number of photons in the pulse we find that $\langle C\rangle$ asymptotically approaches unity as the ratio $E_{\mathrm{th}}/\langle E_3\rangle$ increases, where $E_{\mathrm{th}}$ is the threshold level of the third-harmonic energy pulse discriminator. Clearly, the larger the threshold level of the discriminator $E_{\mathrm{th}}/\langle E_3\rangle$, the larger the number of shots per trigger pulse, $N_{\mathrm{sh}}$. Note that the number of degrees of freedom $M$ of the fundamental radiation pulse is a parameter of the functions $\langle C\rangle = F(M, E_{\mathrm{th}}/\langle E_3\rangle)$, $\langle N_{\mathrm{sh}}\rangle = f(M, E_{\mathrm{th}}/\langle E_3\rangle)$ as indeed we might have anticipated.

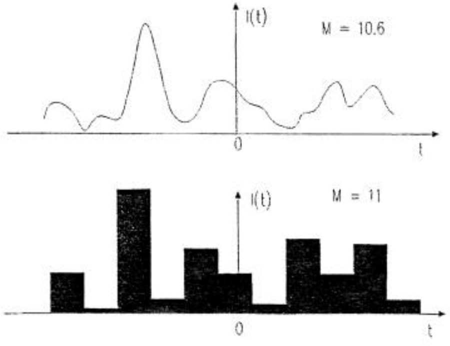

Fig. 20. Approximation of the smoothly varying instantaneous intensity by a "boxcar" function

Our goal is to find the relationship between the level of the discriminator $E_{\mathrm{th}}/\langle E_3\rangle$, contrast factor $\langle C\rangle$, and number of shots per trigger pulse $\langle N_{\mathrm{sh}}\rangle$. In addition, we wish to find effects of the number of modes $M$ on the single spike selection. A complete description of the third harmonic radiation for an SASE FEL can be performed only with a three-dimensional time-dependent numerical simulation code. Numerical calculations allow one to describe the general case of third harmonic SASE FEL operation, including the case of an arbitrary axial profile of the electron bunch, the effects of the spectral profile of the radiation, etc. Such an approach is most fundamental but is also comparatively difficult, for it requires a supercomputer. As a consequence of the obstacles associated with the rigorous approach, an alternative formalism has been chosen for this paper.

To find an approximate density function of integrated intensity we invoke a quasi-physical arguments as follows [19]. As an approximation for the fundamental harmonic, the smoothly fluctuating instantaneous intensity curve $I(t)$ may be replaced by a "boxcar" function on the interval $T$ (see Fig. 20). The time interval is divided into $M$ equal length subintervals. Within each subinterval, the approximation to $I(t)$ is constant; at the end of each subinterval, the approximate waveform jumps to a new constant value, assumed statistically independent of all preceding and following values. The probability density function of the boxcar function within any subinterval is taken to be the same as the probability density function of the instantaneous intensity at a single time instant $t$.

The integrated fundamental harmonic intensity is now approximated in terms of the area under the boxcar function as follows:

$$E = \int I(t)\,\mathrm{d}\,t \simeq \sum_{i=1}^{M} I_i\Delta t ,$$

where $\Delta t$ is the width of one subinterval of the boxcar function and $I_i$ is the value of the boxcar function in the $i$th subinterval. By hypothesis, the probability density function of each $I_i$ is taken to be the same as the density function of the instantaneous intensity. Also by hypothesis, the various $I_i$ are assumed to be statistically independent.

This particular density function of the fundamental radiation energy per pulse (integrated intensity) is known as a gamma probability density function, and accordingly the random variable $E$ is said to be (approximately) a gamma variate. Continuing with the case of the fundamental harmonic, one problem remains: the parameters of the density function must be chosen in such a way as to best match the approximate result to the true density function of $E$. The only two adjustable parameters available are $\langle I \rangle$ and variance $1/M$. The most common approach taken is to choose the parameters such that mean and variance of the approximate density function are exactly equal to the true mean and variance of the fundamental radiation energy $E$ [19].

It should be noted that, in a certain sense, our quasiphysical reasoning that led to the approximate distribution has broken down, for in general the parameter $M$ is not an integer, where as we implicitly assumed an integer number of subintervals in the boxcar function. In what follows we use the assumption: $M \gg 1$. Such assumption does not reduce the practical applicability of the result obtained. It is obvious that this single-spike scheme for time-resolved experiments has an advantage over the scheme which is described in section 3 only when the SASE radiation pulse is much longer than the single spike.

Our approximate model assumes that the smoothly fluctuating SASE intensity curve $I(t)$ can be replaced by a "boxcar" function. Such a model allows us to express the third-harmonic radiation pulse as the sum of the $M$ therms too. The integrated third-harmonic intensity is now approximated in therms of the area under the boxcar functionals follows:

$$E_3 = \int I^3(t)\,\mathrm{d}\,t \simeq \sum_{i=1}^{M} I_i^3 \Delta t \,.$$

Note, that $M$ is the average number of degrees of freedom (or modes) in the fundamental radiation pulse.

In Fig. 21 and 22 one can see the basic characteristics of the single-spike pulse selection process. The dependence of the degree of the contrast $\langle C \rangle$ on the value of energy threshold $E_{\mathrm{th}}/\langle E_3 \rangle$ is presented in Fig. 21. It is seen that the contrast increases with an increase in the value of energy threshold and it asymptotically approaches to unity. Simulations at different values of $M$ show that the degree of contrast does not differ significantly when the number of modes is within the limits $10 < M < 20$. Figure 22 shows plots of the number of shots per trigger pulse $\langle N_{\mathrm{sh}} \rangle$ on the $E_{\mathrm{th}}/\langle E_3 \rangle$ for several values of parameter $M$. From Fig. 22 it is quite clear that the dependence of $\langle N_{\mathrm{sh}} \rangle$ on the number of modes $M$ is rather strong and can not be ignored.

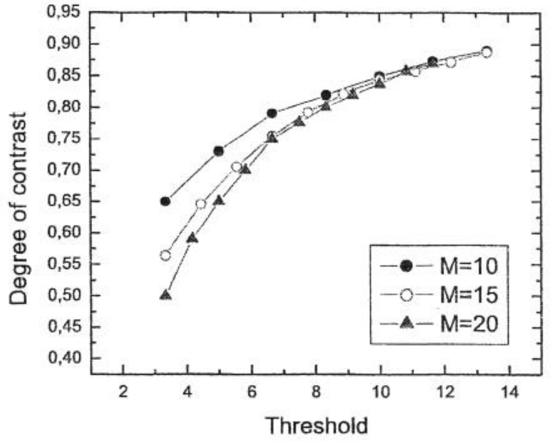

Fig. 21. Degree of contrast $\langle C \rangle$ versus energy threshold $E_{\mathrm{th}}/\langle E_3 \rangle$

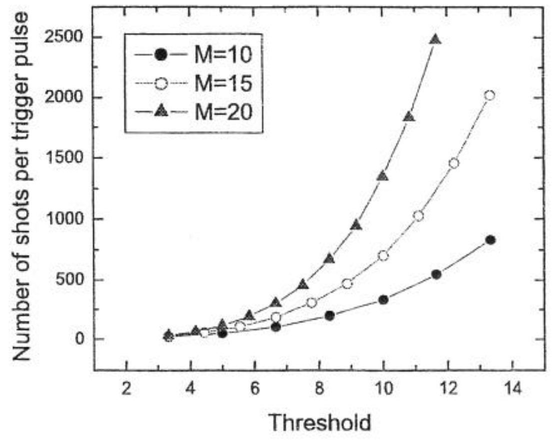

Fig. 22. Number of shots per trigger pulse $\langle N_{\mathrm{sh}} \rangle$ versus energy threshold $E_{\mathrm{th}}/\langle E_3 \rangle$

With the preceding results in hand, it should now be possible to estimate, for example, the repetition rate of the pump-probe pulse. In the TTF SASE FEL case, the number of modes in the fundamental radiation pulse at a wavelength of 30 nm is about $M \simeq 10 - 20$. Suppose that we wish to achieve a contrast of 80%. The discriminator threshold required to achieve this contrast is about $E_{\mathrm{th}}/\langle E_3\rangle \simeq 9$. If the number of modes is close to $M \simeq 15$, Fig. 22 shows that the number of shots per trigger pulse required is about 500. Hence the pump-probe pulse repetition rate is still high (about 50 single-spike pulses per second). On the other hand, if the contrast of interest is only 70% , the number of shots is about $\langle N_{\mathrm{sh}}\rangle \simeq 100$ and the pump-probe pulse repetition rate increases up to a few hundred per second.

A few additional comments are needed in closing this section. The results of numerical simulations presented above refer to the specific model which depends on one parameter $M$ only. This model has proven to be very fruitful, providing the possibility of performing fast numerical simulations of the main statistical characteristics of the high-harmonic radiation from the SASE FEL operating in the linear regime. There is no doubt that these results are useful for a quick estimate and deeper understanding of the frequency multiplication process in a SASE FEL. The defect of the method of analysis used to obtain these results arises from our neglect of effects coming from the spectral profile of the radiation and from density profile of the electron bunch. As a result, the method described above is rather crude and may need to be modified. The full analysis of single-spike generation, including both of these effects, is a very expensive to computer run.

Fortunately, in the particular case, namely, radiation originating from an electron bunch with rectangular profile, a much simplified analysis will suffice. An approach, which takes into account the spectral line shape effects, uses a well known analytical expression for the steady-state spectral Green function of the FEL amplifier [18]. We can decompose the input shot noise signal into Fourier harmonics. Since in the linear regime all the harmonics are amplified independently, we can use the results of steady-state theory for each harmonic and calculate the corresponding Fourier harmonics of the output radiation field. An expression for the electric field of the electromagnetic wave as a function of time $t$ can be obtained using the inverse Fourier transform. In the framework of this model it becomes possible to calculate the smooth sample functions $I(t)$ and $I^3(t)$.

To describe the single-spike selection, we should define the degree of contrast. A first question that arises is: what is the definition of the main spike? The question of when two closely spaced spikes are barely resolved is a complex one and lends itself to a variety of rather subjective answers. One possible definition can be made as follows. After the analysis of a smooth sample function $I^3(t)$ we find a time moment $t_{\mathrm{m}}$ when the intensity reaches its maximum value. Then we find the number of photons within the time interval $(t_{\mathrm{m}} - \tau_{\mathrm{coh}}, t_{\mathrm{m}} + \tau_{\mathrm{coh}})$, where $\tau_{\mathrm{coh}}$ is the coherence time of the third-harmonic radiation. In fact, the ability to resolve two spikes depends fundamentally on the discriminator level associated with the selected third-harmonic pulse, and for this reason for a large ratio, $E_{\mathrm{th}}/\langle E_3\rangle$, this problem does not exist at all. It can be demonstrated that all reasonable definitions for the degree of contrast are consistent in the region $E_{\mathrm{th}}/\langle E_3\rangle \gg 1$.

While adding realism, the step from a "boxcar" function to a smooth sample function drastically increases computer time. Nevertheless, one can learn much about the accuracy of the "boxcar" scheme by seeing how it reproduces "exact" results of

numerical simulations for several values of the discriminator threshold. Surprisingly, we can find that the results of the more general approach does not differ significantly from those of the far simpler analysis done previously.

## Acknowledgments

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