THE SUPER-ACO FREE ELECTRON LASER SOURCE

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Abstract

A Free Electron Laser (FEL) source has been implemented on the storage ring Super-ACO in Orsay (France), dedicated for synchrotron radiation, since 1988. Dynamics studies were performed in the longitudinal direction and the transverse plane, leading to an analysis of the FEL intrinsic evolution and the interaction between the FEL and the positron beam. As a longitudinal feedback has been implemented, source stability issues are discussed. In addition, the optical FEL performances are clearly related to the characteristics of the employed mirrors, and the results of systematic studies lead to operation of the FEL at shorter wavelengths. Finally, a user program has been initiated in 1993, using one color with only the FEL or two colors, employing FEL and synchrotron radiation, for pump-probe experiments in biology, surface science and physical-chemistry.

1 HISTORY

Super-ACO is a 800 MeV storage ring dedicated for applications of synchrotron radiation that has been operated in Orsay since March 1987. In 1989, one year after the FEL optical klystron's installation, the first FEL oscillation was observed in the visible at a wavelength of 633 nm [1].

Table 1: Characteristics of the Super-ACO FEL

Beam energy	800 MeV
Spectral range	vis. – 300 nm
Spectral tunability	10 nm
$\Delta\lambda/\lambda$	10^{-4}
SR temporal width	30 – 120 ps (RMS)
FEL temporal width	20 ps (RMS)
Inter-pulse period	120 ns
Average power	0.3 W (at 350 nm)
Photon rate	few 10^{17} s^{-1}
Laser gain	2-3 %

Over the past years a continuous development, consisting of FEL beam studies, the installation of feedback systems [2] [3], optics improvements and the implementation of a 500 MHz harmonic cavity [4] has made the Super-ACO FEL a powerful coherent UV-source that provides sufficient stability and output power for user applications. The latest milestone was the achievement of laser oscillation at 300 nm with an output power of 10 mW in 1999.

2 DYNAMICS STUDIES

2.1 FEL Evolution

The macro temporal structure of the FEL is governed by the revolution frequency of the positron bunches stored in the ring. It is therefore naturally synchronized with the synchrotron radiation, a fact that is exploited especially for user experiments. Nevertheless, the FEL's behavior will depend strongly on the exact tuning between the optical resonator and the main RF cavity of the storage ring.

A typical detuning curve for the Super-ACO FEL, measuring the laser's intensity as a function of the main cavity frequency's detuning from the perfect synchronization between positrons and photons ($\Delta f_{RF}=0$), exhibits five distinct zones that are arranged symmetrically around perfect tuning. At $\Delta f_{RF}=0$, the extracted laser power is at its maximum, temporal and spectral width of the FEL are minimized and the beam is cw. Nonetheless, micro pulses can lead to a temporal jitter, which in turn results in fluctuations of intensity and a spectral drift. In the two neighboring zones ($\Delta f_{RF}\approx\pm10Hz$) the FEL is pulsed on a ms timescale and the extracted power decreases. If the detuning is again increased ($\Delta f_{RF}\approx\pm50Hz$), the laser returns to cw operation at further reduced output power, but with increased stability of intensity and position.

2.2 Interaction between FEL and Positron Beam

The effect of the stochastic interaction between the laser pulse and the positron bunch has been studied by means of the analysis of synchrotron radiation emitted by the bunch in a bending magnet. The transverse radiation profile was detected by means of a CCD array that allowed the determination of the beam's energy dispersion. Measurements performed on the beam as a

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function of beam current and in the two distinct situations FEL on – FEL off clearly showed that the interaction with the laser increases the beam's energy dispersion. This increase can be explained by the so called Renieri limit, which can for this purpose be written as

$$P_{s,FEL} \propto \Delta \left(\frac{\sigma_\gamma}{\gamma} \right)^2 ,$$

where $P_{s,FEL}$ denotes the saturation power that can be extracted from the FEL and $\Delta(\sigma_\gamma/\gamma)^2$ equals the squared variation of the energy dispersion due to the laser saturation. As the bunch length is proportional to the energy dispersion, it increases as well.

2.3 Longitudinal Feedback System

One desired feature of the FEL is the stability of its beam intensity and laser pulses as well as of the transversal maximum of the laser's intensity in space. This becomes even more important when the laser is used in applications that rely significantly on these properties. A beam being stable in this sense during a time duration ranging from several minutes up to several hours is necessary in those cases.

The temporal stability of the laser is affected by instabilities of the RF system (frequency, amplitude, phase), but also by a 50 Hz modulation of the general power supply affecting components of the storage ring. The resulting beam jitter deteriorates the laser's performance and needs thus to be compensated for.
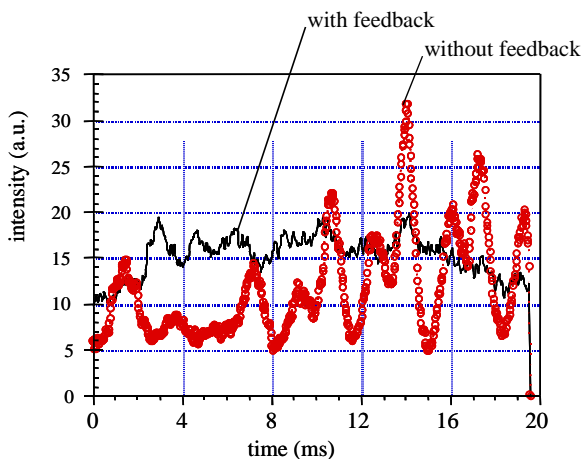

Figure 1: Temporal profiles of the FEL with and without longitudinal feedback measured with a streak camera.

A longitudinal feedback system has been developed and installed to fulfill this task [2]. It measures the temporal position of the laser pulse relative to a reference position by means of a dissector. The time difference is converted into a voltage signal, which is proportional to the measured duration. The voltage signal is in turn used to drive a modulation of the main RF cavity's frequency. The latest improvements of the feedback system's electronics allowed to reach a sampling rate of the system of up to 5 kHz.

The efficiency of the longitudinal feedback has been demonstrated to both compensate the temporal jitter of the FEL as well as to stabilize its intensity and is being used routinely in FEL user operation shifts. Figure 1 illustrates the improved stability of the laser's intensity.

3 UV OPTICS

In order to enable operation of the Super-ACO FEL in the UV, mirrors with specific properties were needed.

The most important feature of the mirrors employed is their reflectivity at the laser's wavelength. As the FEL gain available in Super-ACO does typically not exceed 2% in the UV, the roundtrip reflectivity of the optical resonator needs to be better than 98%. The means of choice are dielectric multilayer mirrors, made of a stack of high and low refractive index materials on a substrate of either fused silica or sapphire. The choice of the substrate material depends on whether the mirror is to be used facing the incident undulator radiation and the corresponding power distribution or on the other end of the optical cavity. As the thermal conductivity of sapphire is higher than of fused silica, sapphire mirrors are used for facing the undulator beam for allowing a better heat dissipation by conduction. Especially for user applications the maximum power that can be extracted from the FEL plays an important role, another strong reason for keeping the total losses of the mirrors as low as possible.

The other important requirement concerning the FEL mirrors is their ability to withstand the higher harmonics of the undulator radiation and the high intra-cavity laser power. Both phenomena lead to a degradation of the mirror's reflectivity during FEL operation.

The total losses of each mirror are given by the sum of its absorption, transmission and scattering. All quantities can be measured independently before and after utilization of a mirror in the FEL. Currently an absorption mapping experiment is being installed that will allow to measure spatially resolved the absorption of multilayer mirrors. This is especially interesting to be done with mirrors that have been exposed to only the undulator radiation during degradation tests, as a correlation between the spatial and spectral characteristics of the incident undulator beam should be possible.

The total losses of mirrors that have been degraded can recover by means of an oxygen plasma treatment followed by annealing at 200°C during several hours. It was found that the initial losses of the mirrors can be restored almost perfectly. Figure 2 shows an example of the total losses and absorption values of a mirror that has been used in the FEL and was then treated as described above.
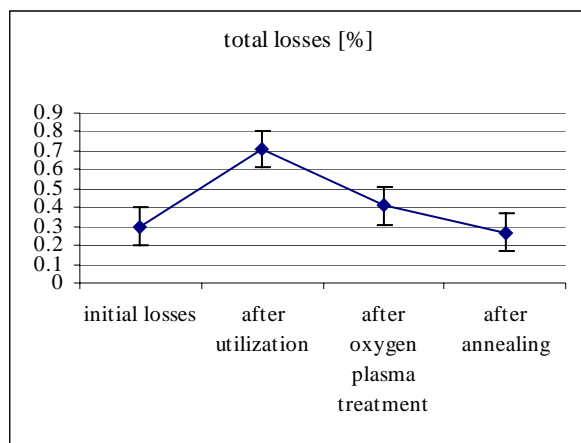
Figure 2: Recovery of used FEL mirrors.

Lasing of the Super-ACO FEL at 300 nm was achieved with mirrors made of a Ta_2O_5/SiO_2 stack on a sapphire substrate and a ZrO_2/SiO_2 stack on fused silica.

4 USER EXPERIMENTS

A program for user applications of the Super-ACO FEL has been launched in 1993. The first studies, which investigated the fluorescence decays and rotational dynamics of the NADH coenzyme [6] demonstrated the feasibility of the FEL's user application. Starting in 1994, the surface photo-voltage effect was studied on a number of different semi-conductor interfaces [7] (GaAs/Ag (1994), Si (1995), SiO2/Au (1998-99)), followed by two-photon surface photoemission since 1999.

The two-color pump-probe technique is currently being used in transient absorption (TA) spectroscopy [8].

For this purpose the synchrotron radiation emitted by a bending magnet, providing an intense white light source from IR wavelengths down to X-rays, is combined with the FEL that serves as a coherent and intense UV source. The two sources are naturally synchronized in a one to one shot-ratio at a repetition rate of the order of MHz. The availability of UV photons makes TA a tool particularly suited for photochemistry and photobiology, where among others fast chemical reactions, excited-state electron transfer can be studied. The experimental set-up at Super-ACO covers here the sub-nanosecond and nanosecond timescale.

For a first demonstration of feasibility the TA technique has been used on a dye molecule (POPOP) in aqueous solution, which appeared to be well matched with the capabilities of the experiment itself in terms of dynamics and spectroscopic range. Following that, TA is to be used on molecules of biological interest such as antitumoral drugs.

The basic principle of the TA procedure can be described as follows: The FEL beam is used as a pump pulse, which excites a fraction of the molecules in question. After a given time, which can be adjusted by means of an optical delay line, the excited state is probed by the white synchrotron radiation.

In terms of a simplified electronic energy level diagram, the FEL pump pulse excites a certain number of molecules from the ground state S_0 to the first (S_1) electronic singlet state. The decrease of transmitted probe light intensity induced by a pre-excitation of the sample is the TA spectrum of interest.

Recently, a delay time dependent effect of the TA signal has been observed when the POPOP molecule is pumped at 350 nm and probed around 450 nm. The declining population of the S_1 excited state was clearly demonstrated on a ns time scale (see figure 3).
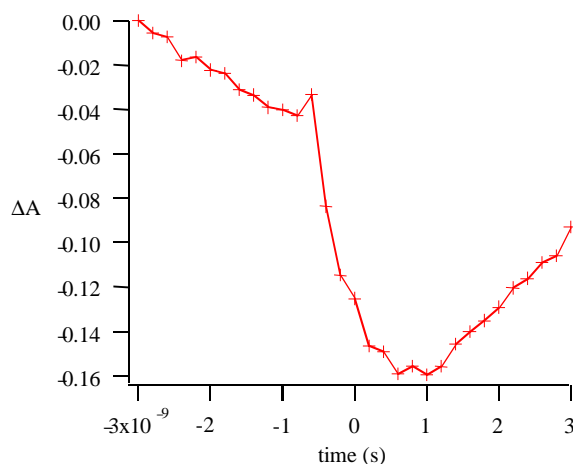

Figure 3: Temporal dependence of the TA signal obtained for a two-color experiment on POPOP. The molecule is pumped at 350 nm and after a time delay as indicated on the x-axis probed around 450 nm.

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