DEUTSCHES ELEKTRONEN-SYNCHROTRON DESY

DESY SR-79/17
July 1979

TIME RESOLVED SPECTROSCOPY WITH SYNCHROTRON RADIATION

by

N. Schwentner
Institut für Experimentalphysik Universität Kiel

U. Hahn
Deutsches Elektronen-Synchrotron DESY, Hamburg

D. Einfeld, G. Mühlhaupt
Berliner Elektronenspeicherring BESSY, Berlin

NOTKESTRASSE 85 · 2 HAMBURG 52

To be sure that your preprints are promptly included in the
HIGH ENERGY PHYSICS INDEX ,
send them to the following address (if possible by air mail) :

DESY
Bibliothek
Notkestrasse 85
2 Hamburg 52
Germany

Time Resolved Spectroscopy with Synchrotron Radiation

N. Schwentner
Institut für Experimentalphysik Universität Kiel,
Kiel, Germany

U. Hahn
Deutsches Elektronen-Synchrotron DESY, Hamburg,
Germany

D. Einfeld, G. Mühlhaupt
Berliner Elektronenspeicherring BESSY, Berlin,
Germany

Abstract

Synchrotron radiation from storage rings provides time resolution in the picosecond range for spectroscopy from the Infrared up to the X-ray region. The status of time resolved experiments at storage rings is described. Some aspects how to reduce the pulse length of storage rings and how to increase the time resolution of experimental set ups are discussed.

Synchrotron radiation is one of the most favourable light sources for time resolved spectroscopy. Due to the intense continuum, light is available from the far infrared, to the visible, vacuum ultraviolet and X-ray region[1]. In addition the radiation is highly polarized. For wavelengths below 2000 Å synchrotron radiation is superior compared to conventional light sources with respect to its time structure, its tunability, the intensity and the degree of polarisation. In the near ultraviolet and visible synchrotron radiation is an essential complement to lasers. Time resolved experiments with synchrotron radiation have been started in 1973 by Heaps et al. at Tantalus I[2]. Since that time intense work has been done at ACO which has been partly summarized bei Lopez Delgado[3]. Since 1976 time resolved experiments are carried out also at SPEAR[4] and DORIS[5]. Recent results have been presented at the V. VUV conference in Montpellier (1977)[6] and in meetings at York (1978)[7] and Stanford (1978)[8].

Pulse length and pulse separation

In electron and positron storage rings the periodic accelerating field from the r.f.-system collects the particles into circulating bunches. According to the harmonic number k of the r.f.-system, up to k bunches can be distributed over the circumference of the ring[9]. For example, in the storage ring DORIS the bunches are 3.96 cm (fwhm) long and the separation between the bunches is either 60 cm (k=480), 240 cm (k=120) or in the single bunch mode 28800 cm (k=1). Due to the strong collimation of the emitted radiation the duration of the light flash from one bunch is mainly given by the time which the bunch needs to pass the tangential point viewed by the observer. For a bunch length of 3.96 cm one gets a duration of the light flash of 130 psec. This is also true for electron synchrotrons, but storage rings offer the advantage of a high current which is stable on a time scale of hours whereas in a synchrotron the typical lifetime of the beam is in the order of msec. In table 1 we have listed the pulse length and the separation between the bunches of those storage rings which are presently used for time resolved studies together with storage rings which will become operational in the near future. The storage rings DORIS, SPEAR and NINA II provide Gaussian light pulses with fwhm of 100 - 200 psec. For the planed new storage ring BESSY the typical pulse length of 130 psec can be shortened below 20 psec by a special beam optic[10].

Due to the divergence ß of the radiation, light from different points of the electron orbit reaches the observer. Therefore an additional broadening Δt of the light pulse is caused by the difference of the straight way of the light path and the way of the electrons on the orbit between the extreme points:

$$\Delta t = \frac{\rho_0}{c} (\tan \beta - \beta)$$

where ρ_0 is the radius of curvature and c the light velocity. For a divergence of 3 mrad in the VUV (0.1 mrad for X-rays) Δt is far below 1 psec for all accelerators of table 1. A similar broadening appears when light from a larger part of the electron orbit is collected by focusing mirrors. When 60 mrad of the radiation emitted in the storage ring plane are accepted Δt will be 0.5 psec at BESSY (ρ_0 = 1.70 m) and 3 psec at DORIS (ρ_0 = 12 m).

The possibility of a large separation of the pulses is important for the investigation of long time constants. For single exponential decay processes, time constants considerabely longer than the pulse separation can be determined from the modulation depth[11], but for most applications the pulse separation (table 1) gives an upper limit for detectable long time constants. The separation between two consecutive pulses reaches 1 usec at DORIS in the single bunch mode.

Time resolution of experimental set ups:

In general obvious advantages of synchrotron radiation are the high possible repitition rates (500 MHz), the extreme small jitter in the pulse train (< 1 psec) and the availability of the trigger signals for the bunch position (for single photon counting technique) and of the modulation frequency (for fluorometry) from the ring radio frequency.

Until now time resolved experiments have been carried out in the visible and vacuum ultraviolet using luminescence spectroscopy and in one example time of flight measurements of photoelectrons[12]. A scheme of an advanced set up at DORIS[5] is shown in Fig. 1 which is typical also for set ups at ACO and SPEAR. The excitation energy can be tuned with the primary monochromator between 2 eV and 50 eV. The samples are illuminated with ≈ 10^{10} photons/sec Å. The emitted light is dispersed by a secondary monochromator working in the range from 2 eV to 20 eV. Typical counting rates are in the kHz regime. The limits are given by the acceptance of the secondary monochromator and the efficiency and bandwidth of the emission. The ratio of the excitation pulses to detected photons is of the order of 10^{-3}. The time delay between a reference pulse for the excitation time (delivered from the machine high frequency) and the luminescence pulses is converted by a time to amplitude converter and the obtained voltage is attributed by an ADC to a channel in the multichannel analyser. By collecting the events a decay curve is immediately displayed and transfered to a PDP 11 computer.

The response function combining the excitation pulse width and the time distortions due to detector and electronics can be measured exactly by analysing reflected light from the sample, which is prompt. Fig. 2 compares response functions measured with different detectors at SPEAR[13], ACO[14] and DORIS[5]. The curve for ACO represents the real excitation pulse width of ACO. The increase of the halfwidth with electron current in the ring is shown. Using a dual microchannelplate a halfwidth of 400 psec with short rise and fall times has been measured at DORIS. By a deconvolution of experimental decay curves with the response function time constants down to 50 psec have been determined and time constants of the order of 10 psec gave a significant change. By an optimized adaption of the channelplate the time resolution will be increased by a factor of two.

Shorter pulse length's and longer pulse separations:

The bunch length can be shortened by chosing an appropriate machine optic. The bunch length σ is given by[9]:

$$\sigma = (\alpha^2 c_q \ \gamma_o^2 / \ \Omega^2 J_E \rho_o)^{1/2}$$

$$\Omega^2 = \alpha e \dot{V}_{\psi_o} / T_o E_o$$

In the special case of small energy oscillations

$$\Omega^2 = \alpha e V_o \omega_{HF} \cos \psi_o / T_o E_o$$

c_q, γ_o, e, ρ_o, ω_{HF}, T_o and E_o are constants for a given storage ring and also the partition number $J_E \approx 2$ depends only little upon the machine parameters. The bunch length is determined by the momentum compaction factor α and the angular frequency Ω of the energy oscillations. Ω depends on the peak voltage V_o which also detrmines the synchronous phase ψ_o. ($\sin\psi_o = U_o/eV_o$, U_o: radiation energy emitted by one electron during one revolution around the ring).

For stability reasons Ω has to be kept much smaller than $1/T_o$. Therefore, in order to get a small σ it is more desirable to reduce α instead of increasing Ω with a larger peak voltage V_o. α is given by integrating the off-momentum function $\eta(s)$ around the circumference L of the ring.

$$\alpha = \frac{1}{L} \oint \frac{\eta(s)}{\rho_o} ds$$

For the lattice of BESSY with three rectangular bending magnets (B_1, B_2, B_3) and a pair of quadrupol magnets between two of them (B_1 B_2; B_2 B_3) it is possible to obtain different signs of $\eta(s)$ at different magnets and the contributions to the integral are partly canceled. The result of an optimal design[10] for the beam optics of BESSY is a momentum compaction factor α as small as $\alpha = 0.00016$. With an appropriate peak voltage V_o extreme small bunch length's corresponding to a pulse width's of 5 psec to 15 psec are expected. In this consideration electromagnetic interaction of the electrons in the bunch has been neglected. For high beam currents an additional broadening of the bunches has to be taken into account. Further due to the smaller phase stability the number of electrons contained in the bunch will be reduced for smaller α. As a rough estimate it can be expected that the current per picosecond can be kept constant. For a bunch length of 10 psec 20 mA of current seem to be realistic.

The separation between the pulses can be increased up to msec by a mechanical chopper in the light path which opens only for each n^th light flash[15]. For dedicated sources like BESSY an aperture in the light path can be used to shadow the light flashes except for each n^th pulse by sweeping the beam position vertically across the aperture with the machine magnets.

Improvements in time resolution and intensity of experimental set ups:

Using timing techniques specially adopted to the pulse structure of the synchrotron radiation improvements are possible. In the phase fluorometric method the phase shift Θ between a modulated excitation source and the consequently modulated luminescence output yields the decay time τ from

$$\tan \Theta = \omega\tau \cdot$$

The determination of short decay times requires high modulation frequencies ω due to the limitations of the accuracy of phase shift measurements to 0.1 to 1 degree. The timing function F(t) of the synchrotron radiation consist of a train of Gaussian pulses $\sigma^{-1}\ (2\pi)^{-1/2} \exp (-t^2/2\sigma^2)$ with a separation of the pulses of t_o (table 1):

$$F(t) = \sigma^{-1}\ (2\pi)^{-1/2} \sum_{n=-\infty}^{\infty} \exp\ (-(t-nt_o)^2/2\sigma^2)$$

The harmonic content is the Fourier transform $G(\omega)$ in the frequency space

$$G(\omega) = \omega_o \exp\ (-\sigma^2\omega^2/2) \sum_{n=-\infty}^{\infty} \delta(\omega - n\,\omega_o)$$

with the fundamental frequency $\omega_o = 2\pi/t_o$. The spectrum $G(\omega)$ shows a train of delta functions spaced by ω_o. It extends to very high overtones[8)] of ω_o due to the small pulse width σ. For the pulse width of ≈ 130 psec of DORIS or SPEAR the overtones at 5 GHz still have 20 % of the amplitude of the fundamental.

Sabersky and Munro[16)] measured the power spectrum of Spear in the single bunch mode with t_o = 780 nsec and obtained at the 280 overtone, which corresponds to the storage ring r.f. of 358 MHz, a signal to noise ratio of 20 dB using a conventional RCA 8850 phototube. A decay time

experiment has been simulated by measuring the phase shift to the storage ring reference singnal when the photomultiplier was moved. The result of 4.7 ± 0.5 degrees per cm of motion, corresponding to 36 ± 4 psec per cm, suggests a time resolution of 8 psec. The intensity of each overtone in $G(\omega)$ increases with ω_o for larger fundamentals ω_o. When DORIS is operated in the multibunch mode at 500 MHz than the intensity in all overtones will be increased by a factor of 500 compared to the single bunch mode (1 MHz). Also the spacing of the overtones is increased from 1 MHz to 500 MHz.

For the very short pulse length of BESSY the amplitude at 10 GHz will be ≈ 90 % of the fundamental. When the higher intensity in the multibunch mode and the higher frequencies of overtones in the GHz regime are combined with the very fast microchannel plate detectors with transit times of 50 psec a time resolution with the phase shift technique in the picosecond and subpicosecond regime will be obtained. Finally other techniques for picosecond spectroscopy like single photon counting streak cameras and techniques developed in combination with picosecond laser spectroscopy are available.

Intensity will become a problem in all fast timing experiments. At DORIS a new beam line[17)] has been designed accepting 50 mrad of the emitted radiation with a premirror focussing in the horizontal plane. The available intensity at the sample will be considerabely increased reaching a mean photon flux at the sample of 4×10^{12} photons/Å sec or 1×10^{15} photons/Å sec cm^2. During the bunch the flux will be 7×10^{17} photons/Å sec cm^2. A resolution of the primary monochromator up to 0.07 Å will be available.

The authors are grateful to Profs. R. Haensel and G. Zimmerer for the support of the project at DORIS.

References

1) For example "Synchrotron Radiation" ed. C. Kunz in Topics in Current Physics Vol. 10, Springer, Berlin 1979

2) Wm. S. Heaps, D.S. Hamilton, and W.M. Yen opt. commun. 9 (1973) 304

3) R. Lopez-Delgado, "Proc. of the course on synchrotron rad. res." Int. Coll. on Appl. Phys., eds A.N. Mancini and I.F. Quercia, Alghero, Italy 1976, Vol.I, p.63

4) K.M. Monahan and V. Rehn Nucl. Instr. and Meth.152 (1978) 255

5) U. Hahn and N. Schwentner Nucl. Instr. and Meth. 152 (1978) 201

6) Vth Int. Conf. on VUV Physics, Supplément au Journal des Physique Fasc. 7 C-4 1978

7) 4. EPS General Conference "Trends in Physics", York, UK, Sept. 1978

8) E. Gratton and R. Lopez-Delgado in Stanford Report SSRL 78/09 (1978)

9) M. Sands, "The Physics of Electron Storage Rings. An Introduction", Stanford, SLAC Report 121, Nov. 1970

10) D. Einfeld and G. Mühlhaupt, Technical Report BESSY TB 8/78

11) O. Benoist d' Azy, R. Lopez-Delgado, and A. Tramer Chem. Phys. 9 (1975) 327

12) R.Z. Bachrach, M. Skibowski, and F.C. Brown Phys. Rev. Lett. 37 (1976) 40

13) J. Munro, I. Pecht, and L. Stryer to be published in Proc. Nat. Acad. Sci (USA) 1978

14) R. Lopez-Delgado, J.A. Miehe, B. Sipp, and H. Zyngier Nucl. Instr. and Meth. 133 (1976) 231

15) Proposal for DORIS by D. Haaks, University of Wuppertal, Germany

16) A.P. Sabersky and I.H. Munro in "Picosecond Phenomena" of Chemical Physics 4, eds. C.V. Shank, E.P. Ippen, S.L. Shapiro, p. 85-89, Springer Berlin 1978

17) Proposal for DORIS by N. Schwentner and R. Haensel University of Kiel, Germany

Figure captions

1) Top: scheme of the experimental arrangement
MP: primary monochromator; FS: focusing mirror;
P: sample holder; L: luminescence light;
MS: secondary monochromator; PM: photomultiplier;
VK: cryostat; GE: gas inlet tube.
Bottom: scheme of the time resolving electronics
DORIS: storage ring; BT: bunch trigger;
DL: delay line; CD: constant fraction discriminator;
TAC: time to amplitude converter; ADC: analog to digital converter; MC: multichannel analyser; PL: plotter

2) Time course of the synchrotron light pulses at SPEAR[8), ACO[9), and DORIS[7) as detected by a single photon counting system (Fig.1) with photomultipliers as indicated. The times for full width half maximum are also shown.

Table 1

storage ring	location	pulse width fwhm (2.355σ) in (nsec)	pulse separation t_0 in (nsec)		
			multiple bunch	single bunch	
DORIS	Hamburg	0.13	2; 8;	960	timing experiments in operation
ACO	Paris	0.8 -1.4		73	
SPEAR	Stanford	0.08-0.4		780	
TANTALUS I	Stoughton	1 -4		31	
BESSY	Berlin	0.01-0.13	2;	200	planned for 1981
NINA II	Daresbury	0.17		320	planned for 1980

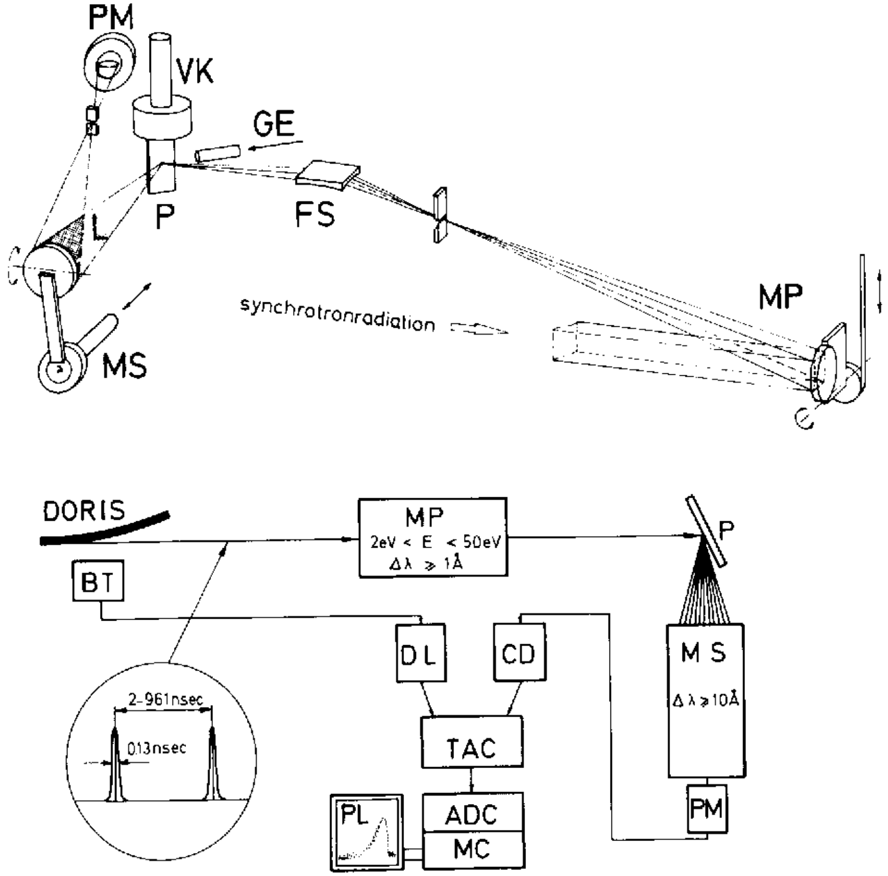
PM
VK
GE
P
FS
L
MS
MP
synchrotronradiation
DORIS
MP
2eV < E < 50eV
Δλ ≥ 1Å
P
BT
DL
CD
M S
Δλ ≥ 10Å
TAC
ADC
MC
PL
PM
2-961nsec
0.13nsec

Fig. 1

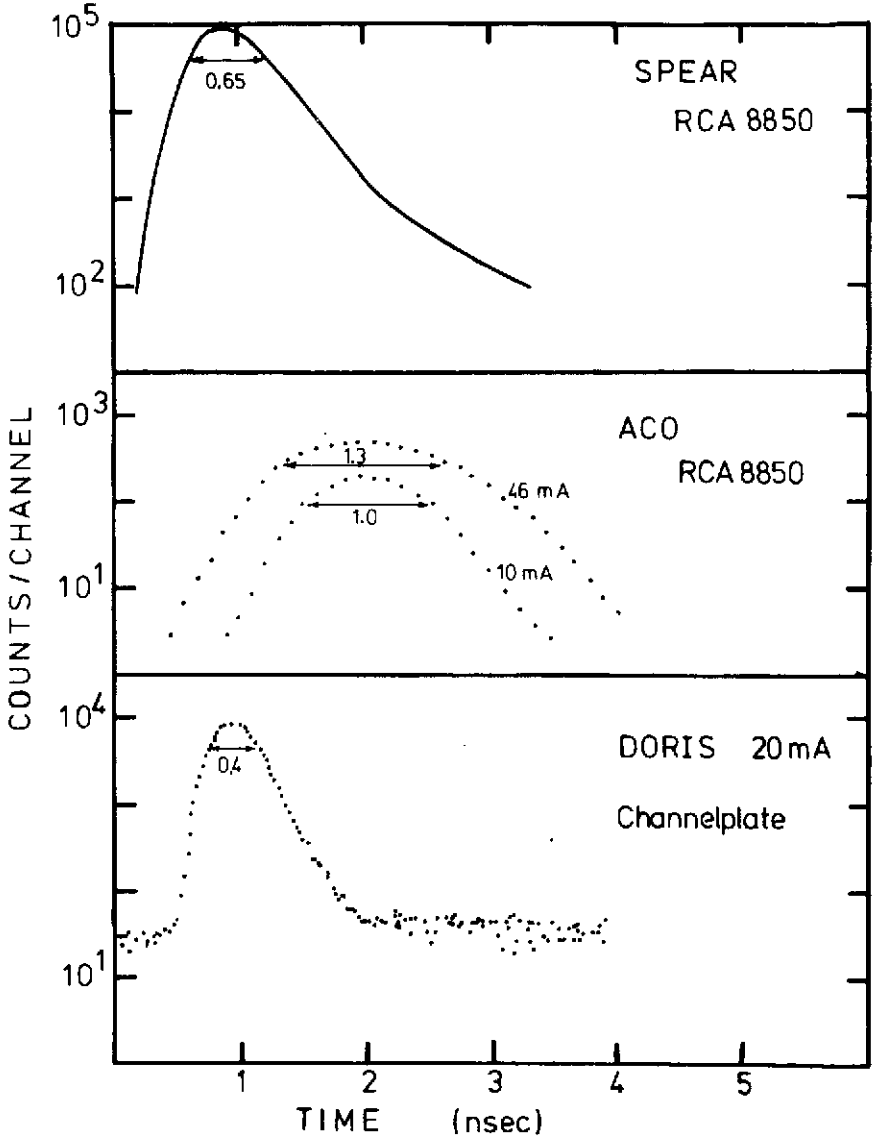
10^5
10^2
0.65
SPEAR
RCA 8850
10^3
10^1
1.3
1.0
46 mA
10 mA
ACO
RCA 8850
10^4
10^1
0.4
DORIS 20 mA
Channelplate
COUNTS / CHANNEL
1
2
3
4
5
TIME (nsec)

Fig. 2