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of Properties and Prospectives**

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Infrared Synchrotron Radiation, Review of Properties and Prospectives.

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ABSTRACT

In this paper we review the properties of infrared sources, setting the synchrotron in perspective among lasers and thermal sources. Synchrotron radiation is ideal for spectroscopy on small samples and has enjoyed extensive utilization throughout the world with some 27 beamlines either in operation or planned. It is a broadband source, which is 1000 times brighter than standard thermal sources. It is polarized, pulsed on the nanosecond scale, highly spatially coherent and is also an absolute source making it possible to perform accurate absorption or reflection measurements. The high brightness makes it ideal for spectroscopy on samples with limited throughput and the main focus has been the realization of very high signal to noise values, not only on small samples, but in the far infrared where the 300K background is a major contributor to the noise. However, synchrotron radiation is not suitable for high power or non-linear applications. Modern free-electron lasers are up to 8 orders of magnitude brighter than synchrotron radiation at the wavelengths at which they operate.

Keywords: Synchrotron, infrared

1. INTRODUCTION

Fig. 1. Schematic of synchrotron radiation generation.

As the introductory paper, this is intended to set the stage for the meeting, by being tutorial in introducing the important concepts and terminologies relevant to light sources and their applications. Since we are concerned with accelerator produced sources of light, let us first consider some definitions and numerical examples. Light of a given wavelength can be characterized by its brightness, power, bandwidth and time structure. In many cases these definitions are evident from the units. For example, power could be expressed in watts/cm², or it could also be expressed in photons/sec/0.1% bandwidth. Brightness, also called brilliance, is defined as power/emittance where the emittance of a source is the product of its area and the angle into which it emits light. Thus brightness will be in units like watts/cm²/mm²/sr. Brightness has the unique property in being at best conserved via Liouville's theorem in an optical system. As an example we consider a 1200K thermal source of area 10mm×1mm, which emits into an angle of 2π steradians. For

this source the power could be of the order of 20 watts into a bandwidth of approximately 10,000 cm⁻¹ and the emittance is 63 mm²/sr. Thus the brightness is 20/(63×10⁴), or 30 microwatts/cm²/mm²/sr. In contrast we consider a synchrotron radiation source which emits 100 milliwatts into the same bandwidth, namely 10,000 cm⁻¹, but from a source size of 330 microns by 330 microns, and into a solid angle of 1 milliradian, giving a brightness of 0.1/(0.03²×10⁻³×10⁴), or 1 milliwatt/cm²/mm²/sr – a factor of 300 higher than the thermal source. Free electron lasers, with a total power of 100 watts per cm², emitting into a diffraction limited phase-space of λ², will, at a wavelength of 3 microns, have a brightness of 100/(3×10³)² or 10⁷ watts/cm²/mm²/sr, a factor of 10¹⁰ higher again!!

These comparisons serve to introduce the terminology and a sense of scale, but miss many important points such as temporal structure and total available bandwidth. In fact all the sources have an important role to play in scientific investigations, and it is important to match the source to the experiment. At this meeting we will hear many talks about the applications of synchrotron radiation, in which advantage is taken of its high brightness and broad bandwidth, and in some cases its temporal structure. We will also hear talks about the applications of free-electron laser sources, in which the extreme high brightness and time structure is used.

Next we will discuss how these sources operate, and also present details of the spectra.

2. CHARACTERISTICS OF SYNCHROTRON RADIATION

Synchrotron radiation^{1,2} is emitted when the velocities of relativistic electrons are modified as they enter dipole magnet fields in storage rings. Actually it is the transverse acceleration term which is responsible for synchrotron radiation emission. The velocity change gives rise to what is termed "dipole edge" radiation, which is the subject of other papers at this meeting. The passing of an electron in a dipole magnet of a storage ring results in a pulse of electric field as seen by an observer as shown in Fig. 1. The Fourier transform of this pulse yields the spectrum as indicated. For N electrons, the intensity is simply multiplied by N , since the electric fields add incoherently, but we discuss this more later. In storage rings, many electrons circulate in "bunches" whose length is governed by the radio-frequency cavity which has to restore the energy lost by synchrotron radiation. These bunches are typically 50 to 500 picoseconds long, and this time-scale is the one relevant for using the source for timing measurements.

The emitted radiation whose spectrum is shown in Fig. 1, has a characteristic emission pattern, which is shown in Fig. 2. Light polarized parallel to the orbit plane peaks in this plane while light polarized perpendicular to the orbit plane peaks slightly out of the plane.

1. Power.

The power emitted in the infrared region, which is on the left of the schematic figure at the lower right of Fig. 1, is readily calculated and in photons per second it is:

$$P(\lambda) = 4.38 \times 10^{14} \times I \times \theta \times BW \times (\rho/\lambda)^{1/3} \text{ photons/sec}$$

1

Where I the current in amperes, θ (rads) the horizontal collection angle, BW the bandwidth in %, λ the wavelength, and ρ the radius of the ring. λ and ρ are in the same units. Power in photons per second can easily be converted to watts by dividing P by $(5.04 \times 10^{18} \cdot \lambda(\text{microns}))$. The bandwidth can also easily be changed to cm^{-1} if desired.

Fig. 3. Total power of a synchrotron radiation source compared with a 10x1 mm thermal source

horizontal angle of the beamline. The corresponding vertical size is a complicated function but is something like $\rho \theta_h \theta_{\text{nat}}/8$. However, assuming that the opening angles of the beamline match the synchrotron radiation natural opening angles, and assuming that the intrinsic source size is smaller than that due to diffraction, Jim Murphy³ has shown that the brightness of synchrotron radiation is given for all rings by:

Fig. 2 Vertical distribution of synchrotron radiation power

2. Brightness.

In order to be able to calculate the brightness, we need to be able to calculate the source area and emission angle. The source size is generally close to that given by the diffraction limit, that is a Full Width Half Maximum (FWHM) of $\lambda/\theta_{\text{nat}}$ where θ_{nat} is the characteristic or "natural" opening angle given in radians by $1.66(\lambda/\rho)^{1/3}$ with λ and ρ in the same units this time. If the source size is larger than this, then it has to be added in quadrature with the size due to diffraction. Also it is important to note that if the horizontal angle is larger than the natural opening angle, the brightness is actually reduced!! This is due to the curvature of the storage ring and the large size of the source then controlled by the geometry. In this case, the horizontal size is given by $\rho \theta_h^2/8$, where θ_h is the

Where again, λ is the wavelength in microns, I the current in amperes, BW the bandwidth in %. Photons per second can easily be converted to watts as before by dividing B by $(5.04 \times 10^{18} \lambda)$. The bandwidth can also easily be changed to cm^{-1} if desired. In Figs. 3 and 4 we plot Eqs. 1 and 2, and compare the values with those for a practical beamline. For the latter we took a radius of 1.91 meters, and maximum collection angles of 90 mrad, the parameters for beamlines U4IR and U12IR at the NSLS, Brookhaven.

Fig. 4 Synchrotron radiation brightness compared to 2000K thermal source.

generally less destructive than particle probes. Infrared light is important in being able to interact with any electric dipole. Such dipoles include free or nearly free charges that may give rise to what is known as polarizability, and also those associated with atomic and molecular vibrations. Studies of the frequency dependent response of samples can provide deep insight into the physics responsible for much of the dynamics that critically determines the behavior. We see many examples of this in the studies of surface friction and in the studies of strongly correlated electron systems.

For almost a century scientists have done spectroscopy with broadband thermal infrared sources and developed an impressive range of detectors and Fourier Transform Interferometers. In general the work that can be done with such sources is limited by the brightness of the source, since usually one seeks high signal to noise, and the noise is given by the expression⁵:

$$\%N = \frac{100A^{1/2}D^*}{\phi(\nu)\Delta\nu\epsilon^{1/2}\xi} \quad 3$$

where A is the detector area, D^* the detectivity of the detector, $\phi(\nu)$ the brightness, $\Delta\nu$ the bandwidth, ϵ the experiment's throughput, t the measuring time interval and ξ the optical efficiency. The throughput, also called the etendue, is the product of the limiting area and angle of the experiment. Multiplying the brightness by the throughput yields the signal expected at the detector. For expression 3 to apply, the throughput must be less than the emittance!!

Even with the best detectors, superior signal to noise is possible if the source brightness can be increased. With the synchrotron source, the brightness - and consequently the signal-to-noise - can be 1000 times greater than for the thermal source. Thus, to attain similar quality spectra with a thermal source one would have to collect data a million times longer, i.e. a 1 second measurement would take 11 days! In practice we have found that the expected, calculated, theoretical signal to noise is not achieved due to instabilities in the electron orbits in the synchrotron which we discuss later.

Synchrotron radiation, being a broad-band source, is most naturally compared with thermal sources. For a thermal source, the power can easily be evaluated using the formula:

$$P_{bb}(\text{watts}) = \frac{2\pi\hbar c^2}{\lambda^5} \frac{d\lambda}{\frac{ch}{\lambda KT} - 1} \quad 4$$

where P_{bb} is the power emitted per unit surface area, $2\pi hc^2 = 3.7415 \times 10^{-16} \text{ W/m}^2$, and $hc/K = 1/43879 \times 10^{-2} \text{ mK}$. The brightness is easily calculated by dividing by the area $\times 2\pi$. We have added calculations based on Eq. 4 to Figs. 3 and 4 and can thus compare the brightness and power of a thermal source at 2000K, and a synchrotron source. It can be seen how the synchrotron source is superior in brightness across the entire range, with corresponding implications for noise reduction. Note also that the synchrotron source provides more power at long wavelengths, so is always superior.

Given the advantage in signal to noise for small samples, it is no surprise that the major use of synchrotron radiation is microspectroscopy, as we will hear during this workshop. Numerous other applications, such as measurements of samples at high pressures, surface vibrational spectroscopy and surface dynamics at grazing incidence, are also examples of experiments with limited throughput.

4. FUTURE PROSPECTS

It is clear from Table 1, which shows the synchrotron radiation beamlines in operation, that there has been a considerable increase in activity in this area in the past few years. Reference to Table 2 shows that this activity is likely to continue, as more and more planned beamlines come into operation.

Location	Number of IR ports	Comments
SRC, Daresbury, UK	1	Several end stations, microscopy, surface science
LURE, Paris, France	2	One on undulator for spectroscopy, one on dipole for microscopy
ALS, Berkeley	1	2 spectrometers, one for microscopy, one for surface science.
Aladdin, Wisconsin, USA	1	Edge and dipole radiation port, microscopy.
NSLS, Brookhaven	6	8 spectrometers, several end stations, microscopy, surface science, spectroscopy.
NIST, Gaithersburg, USA	1	Not operating, new ring commissioning
Max-Lab, Lund, Sweden	1	Several end stations, microscopy, gas-phase, surface science.
UVSOR, Okasaki, Japan	2	Spectroscopy, microscopy

Table 1. Synchrotron radiation facilities as of July 1999.

Location
DaΦne, Rome, Italy
SRRC, Taiwan
ANKA, Karlsruhe, Germany
NSRC, Hefei, China
BESSY-2, Berlin, Germany
SpRing-8, Japan
CAMD, Baton-Rouge, USA
CLS, Saskatoon, Canada

Table 2. Synchrotron radiation facilities in the planning or construction phases as of July 1999.

Thus at the time of writing there are no less than 15 operating synchrotron IR facilities, and 8 being planned. These IR beamlines have been found to test the stabilities of the various rings, since the brightness is not an advantage if there is source noise. Intrinsically synchrotron radiation should be a very stable source because the same bunch of electrons circulates, with very small losses. However, in practice, many of us have found that instabilities in power supplies, in radio-frequency cavities, in storage ring cooling systems, and in vibrating pumps, have led to source noise. These are of several kinds - long term drifts probably due to changes in temperature as the storage ring current decays, short term instabilities in the 10-100 Hz region due to mechanical vibrations and power supplies, and radio frequency side-bands in the kilohertz region from the accelerating cavity. Standard FTIR spectrometers and detectors are sensitive to noise in the audio range of frequencies, so reducing beam instabilities is crucial for realizing the brightness advantage of the source. We have addressed the low (<300 Hz) instabilities by installing a global feedback system⁶ on the NSLS VUV ring in which the electron orbit is measured and corrected using special additional magnets. The rf sidebands can be reduced⁷ through careful tuning of the synchrotron's rf cavity, minimizing their effects on the spectral noise. Continued improvements in reducing these noise sources will benefit all the infrared programs. This is particularly important for FTIR instruments since the

noise is multiplexed and the main advantage turns into a disadvantage. Fortunately many of these sources of electron beam instability, have been addressed aggressively by the accelerator staff at all facilities. In the future, however, we could still benefit from continuing work in this area. One way of eliminating the thermal drifts at Brookhaven was found by using top-off mode injection, in which beam was injected every few minutes to maintain an almost constant stored beam current. This maintained a baseline across the entire spectrum in absolute terms, of $\pm 0.2\%$ over a period of over an hour.

Other improvements for the future of synchrotron radiation infrared research will come with improvements in instrumentation for existing and new beamlines. Thus, we will see wider spectral ranges used, down to 1 cm^{-1} and up into the near IR to visible. In spectrometers we will see step-scan benches and faster rapid-scan benches, newer vacuum benches, more benches with higher resolution. New generations of microscopes are already on the horizon, which allow simultaneous viewing, IR spectroscopy and other capabilities. Multi-element "camera" detectors will be tested.

We will also see many additional capabilities in experimental stations such as the availability of high magnetic fields, newer cryostats, electrochemical cells, high and low pressure chambers, and protein reaction cells to name just a few.

Finally I comment on two new developments in IR brightness for synchrotron sources. The first is dipole edge radiation, which will be discussed in a paper at this meeting. It appears to have the same brightness as synchrotron radiation, although the emission characteristics are different, with smaller angular divergencies. At the time of writing, the theoretical calculations for this radiation have just been completed by several groups. The radiation itself has not been accurately measured and clearly needs to be well characterized. The second is coherent synchrotron radiation^{8,9}. If the electrons are bunched tightly longitudinally, then it is possible for the electron bunch to be shorter than the wavelength being emitted. In this case, the electric fields of the electrons add coherently, making a field N times that of a single electron, so the intensity is therefore N^2 times that of a single electron, and thus N times that of incoherent synchrotron radiation. This effect has been observed at the NSLS, Brookhaven. Since there can be up to 10^{12} electrons in a single bunch at the NSLS, even a small modulation in the bunch can cause a large enhancement in the source brightness.

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