Submicrovolt Resolution X-Ray Monochromators

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There are three basic methods to obtain a "monochromatic" beam from incident "white" radiation: wave length selection, frequency (or energy) selection, and (for non-zero rest mass particles) velocity selection. For X-rays only the first two methods are applicable which, for short, will be called "λ-filtering" and "ω-filtering" methods.

The radiation emitted by an ultra relativistic electron is confined to an angle $\sim \gamma^{-1}$ ($\sim 10^{-4}$ for electron energy $\sim$ 5 Gev) about the forward direction. A pulse of radiation observed in the forward direction lasts $\sim \frac{R}{c}\,\gamma^{-3}$ where R is the radius of curvature at the tangency of the observation point to the electron's orbit. Since all of this is explained in previous talks given in the conference, we confine ourselves to the observation that such a pulse has a length $\sim \lambda_c$ where λ_c is the critical wavelength, and for our purposes we consider the hard X-ray synchrotron sources where $\lambda_c \lesssim 1$Å. For definitness we shall suppose that we are attempting to extract a monochromatic beam with $\lambda \approx 1$Å and $\frac{\delta\lambda}{\lambda} \ll 1$ from this incident pulse which has $\frac{\delta\lambda}{\lambda} \approx 1$.

In the λ-filtering method one lets the pulse fall on a crystal at an angle θ to a set of Bragg planes. The pulse incident on the first plane gives rise to a small (distorted) reflected image, which is followed at a distance, 2d sin θ, by that reflected by the second plane, and so on. If the crystal has N planes then the incident pulse will result in a train of N pulses a distance 2d sin θ apart, each "imaging," the incident pulse (if N is not too large so multiple reflections are unimportant). The fundamental (spatial) period of this train is $\lambda = 2d \sin\theta$, and since it consists of only N periods, there is a spread of wavelengths given by $\frac{\delta\lambda}{\lambda} \approx \frac{1}{N}$ in the packet. We ignore the presence of the $\frac{\lambda}{2}$ and higher harmonics, they may be removed in various known ways.

Clearly the crystal monochromator works like an analog device, translating the spatial periodicity of the crystal onto the reflected field. Also, clearly, the device will select the same fundamental wavelength, $\lambda = 2d \sin \theta$, independent of the nature of the incident radiation.

This would seem to indicate that the monochromicity obtainable by λ filtering methods is limited only by the size of the perfect crystal one can obtain. That is not true, however, because for large N multiple reflections become important. The effect of these are to give a progressive decrease in the amplitude of the waves incident upon deep lying layers and consequently on the amplitudes of the waves reflected from layers which propagate to the outside of the crystal and contribute to the reflected wave. Roughly, if F is the scattering amplitude per plane then multiple reflections are unimportant only if $N F << 1$. Analysis (Darwin) shows that the individual planes reflect progressively smaller amounts so that the effective number of planes is $N_{eff} \approx \frac{\pi}{F}$

Large silicon crystals of high perfection are available. Using the Darwin expression, $F = n\lambda^2 f(2\theta)/2 \sin^2 \theta$, where n = density of atoms, $f(2\theta)$ the atomic (elastic) scattering amplitude for scattering of X-rays of wavelength λ thru the scattering angle 2θ, then we have for $\lambda = 1Å$, $\theta \approx 90°$, $n = 0.05Å^{-3}$(Si) , $f(2\theta) \approx 2.7r_0 \approx 7.5 \times 10^{-5}Å$,
$\frac{\delta\lambda}{\lambda} = \frac{F}{\pi} \approx \underline{6 \times 10^{-7}}$.

The result then is that for a perfect Si crystal monochromator the smallest energy spread it can give is $\delta E \approx 6$ mev at $E \approx 10^4$ ev. [Since F decreases rapidly, $\sim \lambda^2 f(2\theta)$, as λ decreases, δE could be decreased by selecting smaller wavelengths (assuming perfect periodicity of the Neff scattering planes).

ω-Filters

To exceed 1-10 mev resolution obtainable in the E = 10 kev range by λ-filtering we must rely on ω-filters based on nuclear resonance scattering. The nuclear resonances lying a few tens of kilovolts above the nuclear ground state necessarily have widths in the range of 10^{-6}-10^{-9}ev (and smaller). Thus, to exceed the mev resolution afforded by λ-filters, we must construct ω-filters and they will give submicrovolt resolution. These filter of course do not depend on the spatial periodicity of a set of crystal planes being translated into a periodic wave train.

If we consider a single plane, for example, then a "white" incident X-ray blip will result in a weak distorted blip being reflected from it. The reflected blip will still be "white," $\frac{\delta\lambda}{\lambda} \approx 1$. However, if the plane has atoms in it with excited nuclear states with energies in the spectral range of the incident blip, then those resonances will be shocked into excitation by the blip and will ring, continuing for a time $T_R \sim \frac{\hbar}{\Gamma}$ to send out E-M waves with frequencies $\omega_R = E_R/\hbar$, where Γ is the width of the resonance and E_R is its energy. If the resonant atoms are Fe^{57}, for example, then $E_R = 14.4$ kev ($\lambda_R = 0.87$Å) and $\Gamma = 5 \times 10^{-9}$ ev $\rightarrow T_R \approx 10^{-7}$s. In this case if a blip with a length of $\delta\lambda \approx 1$Å is incident upon the plane, then there will be a "prompt" electronically reflected blip with a length 1Å reflected from the plane (which still has a $\frac{\delta\lambda}{\lambda} \approx 1$) in a time $\sim 10^{-18}$ sec after the incident blip arrives, and a "pure tone" lasting about 10^{-7} sec $\rightarrow$ wave train of ≈ 30 m with $\lambda = 0.87$Å $\rightarrow \frac{\delta\lambda}{\lambda} \approx 3 \times 10^{-12}$, emerging after the initial excitation.

Now a single bunch of electrons in a storage ring (giving $\lambda_c \approx 1$Å) is ~ 3 cm long and in a single pass gives a light pulse at an observer lasting ~ 10^{-10} sec. When this radiation is incident upon a small sample, then the electronically scattering X-rays will emerge promptly during the pulse, but those nuclei which have been excited will send out a wave train for ~ 10^{-7} sec (Fe^{57}) after the pulse.

S. Ruby[1] in 1974 pointed out that by detecting the photons scattered from a sample containing resonant nuclei only after the prompt reflected pulse had passed (after $\approx 10^{-10}$s from the time the synchrotron pulse hit the sample) then those photons emerging in the next $\frac{\hbar}{\Gamma} \approx 10^{-7}$s ($Fe^{57}$), would have $\frac{\delta\lambda}{\lambda} \approx \Gamma/E_R \approx 10^{-12}$ (Fe^{57}).

In a demonstration experiment R. Cohen[2] showed that indeed there were these delayed photons following the prompt pulse in an experiment at Spear using an Fe^{57} crystal as scatterer. However, the slow recovery of his detectors from the massive prompt reflected pulse resulted in a large background noise making the signal detection difficult.

The Ruby scheme effects ω-filtering using the temporal separation of the "noise" and the signal following pulse excitation of a crystal containing resonant atoms. For SPEAR operating in the single bunch mode the pulse deration $\sim 10^{-10}$s and the pulse repetition time $\sim 10^{-6}$s, then for the Fe^{57} resonance with $T_R \sim 10^{-7}$s, this elegant scheme could be the basis of a practical monochromator. However if an Fe^{57} crystal is used to Bragg reflect the spectral slice $\delta E_R \approx \Gamma \approx 10^{-8}$ ev, then the average power reflected electronically in the prompt pulse is $\gtrsim 10^4$ x that of the resonant signal. In Cohen's experiment, the detectors did not recover quickly enough from the strong promptly reflected pulse to allow good discrimination of the resonant signal. For this reason, and also to allow operations in multibunch modes for which the pulse repetition times $< T_R$, it is desirable to construct ω-filters so that the average electronically reflected "noise" power is less than the nuclear reflected "signal" power.

There are several ways which have been suggested to construct such filters, and since 1975 we and coworkers[3], and independently the Russian physicists[4] and others, have been engaged in working out the theoretical details of the most promising schemes for achieving efficient nuclear resonance filters. In addition, experimental work leading to filtering is well underway in Professor Gerdau's group[5] in Hamburg, and in Russia.[6]

The filters all involve reflection devices which will give strong nuclear reflections and zero, or very weak, electronic reflections.

A conceptually simple case is that of an iron crystal consisting of alternating planes of Fe^{57} and Fe^{56}. We suppose that the incident beam has been premonochromatized by means of a crystal monochromator (plus perhaps one or more grazing incidence reflections to remove the higher orders) to a width $\Delta E \approx 1$ ev around E_R (14.4 kev). Then letting the beam fall at a Bragg angle $\theta_R = \sin^{-1}(\frac{\lambda}{4d})$ on the crystal there would be strong reflection of the resonant photons and practically no electronically reflected photons.

Without resorting to such artificial crystals a simple antiferromagnet containing Fe^{57} (or other magnetic resonant atoms), e.g., Fe_2O_3 , will give strong nuclear reflections at the antiferromagnet, X-ray forbidden, Bragg settings[7, 8] and very weak electronic scattering at those settings. Ferrimagnets, e.g., YIG[5] can similarly serve to give pure nuclear Bragg reflections.

These, and other Bragg reflection methods, will depend upon obtaining large perfect crystals to get efficient filtering. Another promising method involving grazing incidence reflections from Fe^{57} surfaces coated with suitable thin layers to eliminate, by destructive interference, the nonresonant X-ray reflections, but giving strong nuclear reflections, does not depend upon perfect crystals and should be capable of yielding efficient filters.[3] By properly tailoring, these GIAR filters can be made to have resonance responses with widths ranging from 1-100 times the natural width.

As an example of the crystal requirements for efficient Bragg reflection filters, we consider the [111] reflection in $(Fe^{57}]_2O_3$. The Fe atoms lie in layers parallel to (111) planes with the spins lying in the (111) planes and the spin directions alternating.[9] The [111] reflection is X-ray forbidden but there is a strong resonant nuclear reflection.[8] The Bragg angle (for $\lambda = 0.87$Å) for this reflection is $\theta = .094$ rad, and taking the vertical dimension of the beam ≈ 1 mm then, for the crystal cut with the surface parallel to the (111) planes, ≈ 1 cm of the crystal face is illuminated. For a thick perfect crystal the reflected power can be calculated from the Darwin-Prins like formula adapted to resonant scattering.[10] Taking the angular divergence of the incident beam to be $\Delta = 10^{-4}$ rad gives a reflected power equivalent to the incident power in an energy spread of $\approx 20\Gamma = 10^{-7}$ ev.[11] This, however, requires that the mosaic spread be less than 10^{-4} rad and the thickness of the crystal $\tilde{>} 10^{-3}$ cm. Because of the difficulty of growing such a large perfect crystal of $(Fe^{57})_2O_3$, other crystals might be preferred. Gerdau and coworkers[5] are investigating the pure nuclear reflections in YIG, for which large highly perfect crystals are available.

In conclusion, efficient submicrovolt resolution X-ray monochromators can be achieved using nuclear resonance reflections. The results of detailed calculations of the properties of these filters and their uses for inelastic scattering measurements, interferometry and X-ray holography, surface magnetism investigations, and biomolecular structure determinations, and others, will be given in papers being prepared for publication in the Physical review.