New Ideas for Future Sources

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What aspect of X-ray for future?

Short pulse (100 fsec or less) for pump prove experiment.

Whip, strong pin-pon-pan

Coherent Wave for X-ray imaging, and coherent X-ray detection.

Mountain horn, Tunable

Where should we go?

- *Major Science in 21th Century will be Biology, and it related.*
- *Optical microscope, Electron Microscope, X-ray crystalography and X-ray imaging will be used in combination.*

- *Biological material is soft, low Z (low X-ray interaction, low contrast). Specially water H*, and H_2O plays important role.*

  X-ray Scattering power is small: $\sim Z^2$
- *Biological sample is weak against X-ray damage. (binding energy is low, remember a boiling egg changes..)*
- *Need improvement in X-ray detection.*
- *Scientists want have X-ray instrument in their laboratory to quickly study living cells.*
- *Need compact and low cost machine.*

After the Brilliance

- *Brilliance*

$$B = \frac{N}{\Delta x \Delta x' \Delta y \Delta y' (\Delta\lambda / \lambda)}$$

- *History of SR*
  - *started from target X-ray source*
  - *to SR-source : larger N and lower Δy, Δy'*
  - *to undulator : narrow Δλ/λ, lower Δx, Δx'*

- *what's next?*

Using Coherent Radiation Process

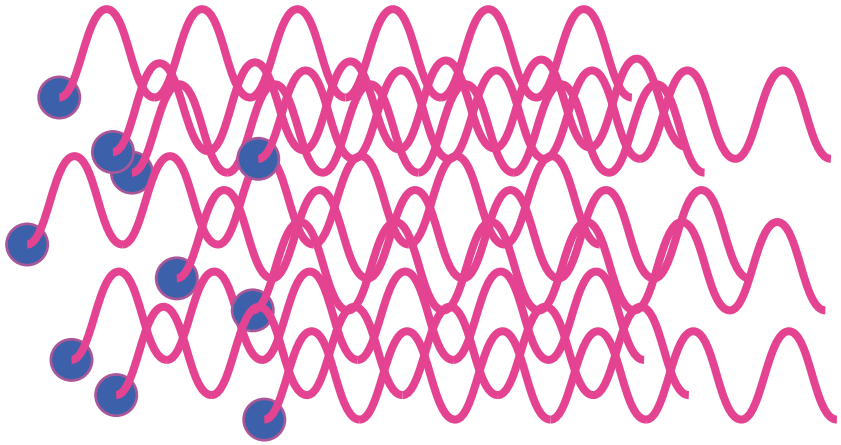
- *Not for higher peak power, like SASE-FEL.*
- *But for higher energy conversion efficiency (we can reduce the e-beam current)*

- *We can not always make good crystal from all kind of proteins. Need to analyze imperfect crystal. MEM will help. Minimum Enthalpy Method, which needs coherent light.*
- *Diffraction imaging for transparent imaging of living cells and nano-technology.*

From SR to FEL

SR or ERL

Spontaneous Radiation



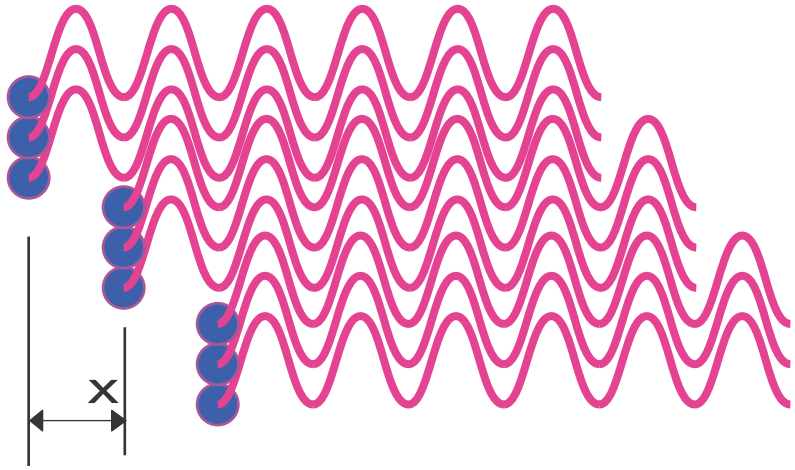
***N*-electrons**
random distribution

$E_{spt} \sim \sqrt{N}\, E_1$

$P_{spt} \sim N\, P_1$

FEL: Free Electron Laser

Coherent Radiation



***N*-electrons**
micro-bunched

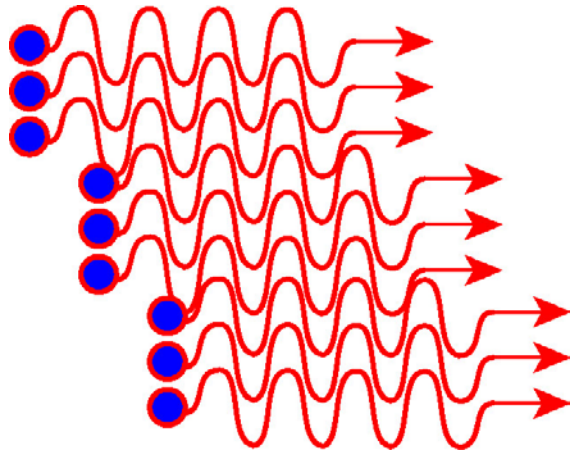
$E_{coherent} \sim N\, E_1$

$P_{coherent} \sim N^2 P_1$

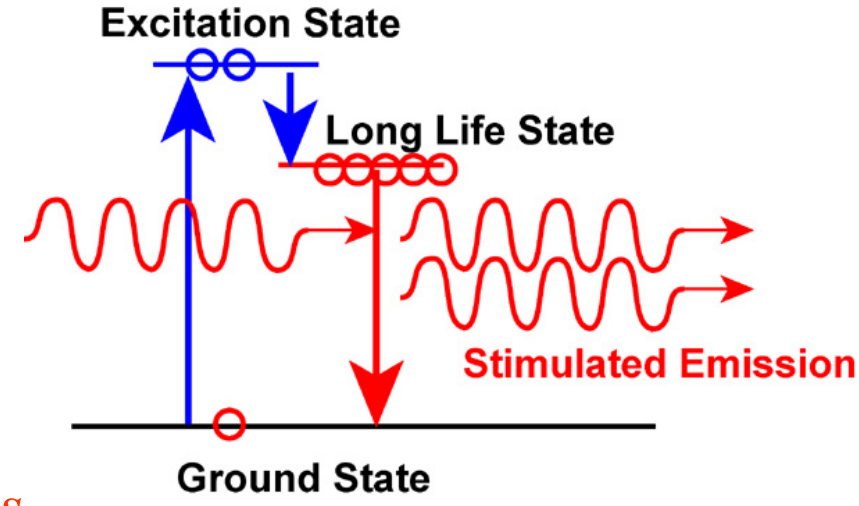
x 10^6...10^8 times higher

Super-Radiation v.s. Stimulated Emission

Super Radiation Mode in FELs



Stimulated Emission in Laser



- *Each electron emit spontaneous emission.*
- *Because, electrons are aligned in wavelength pitch, emissions are overlap each others in phase.*
- *Polarization is defined by undulator.*

- *Because of stimulated process, all lights are matched in phase and polarization.*
- *Emitting atoms are randomly distributed.*

Talk at 24th ICFA FLS Workshop, May 2002, SPring8
Presented at Wrokshop March 1999, KEK Japan

Wavelength Compression

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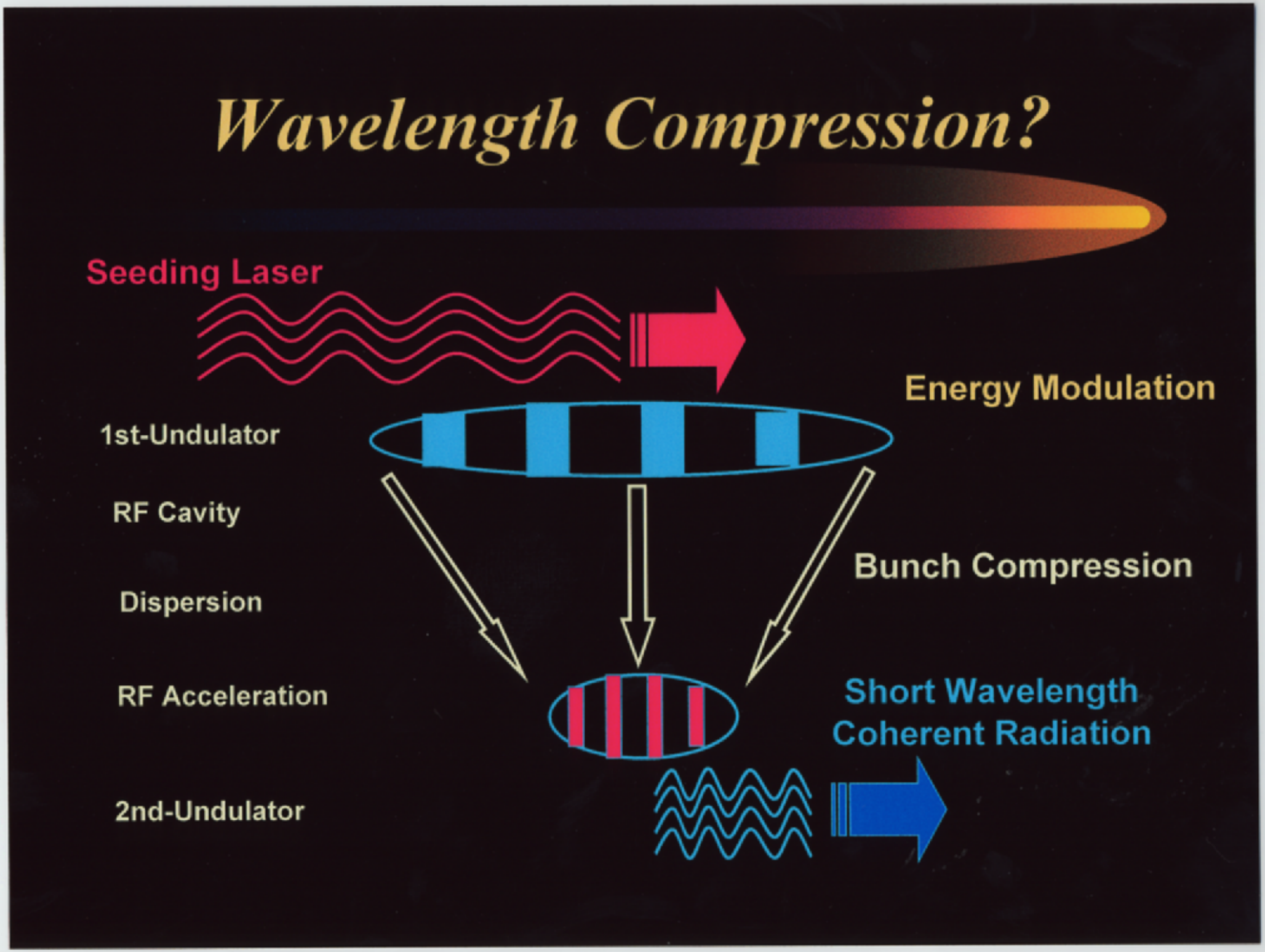
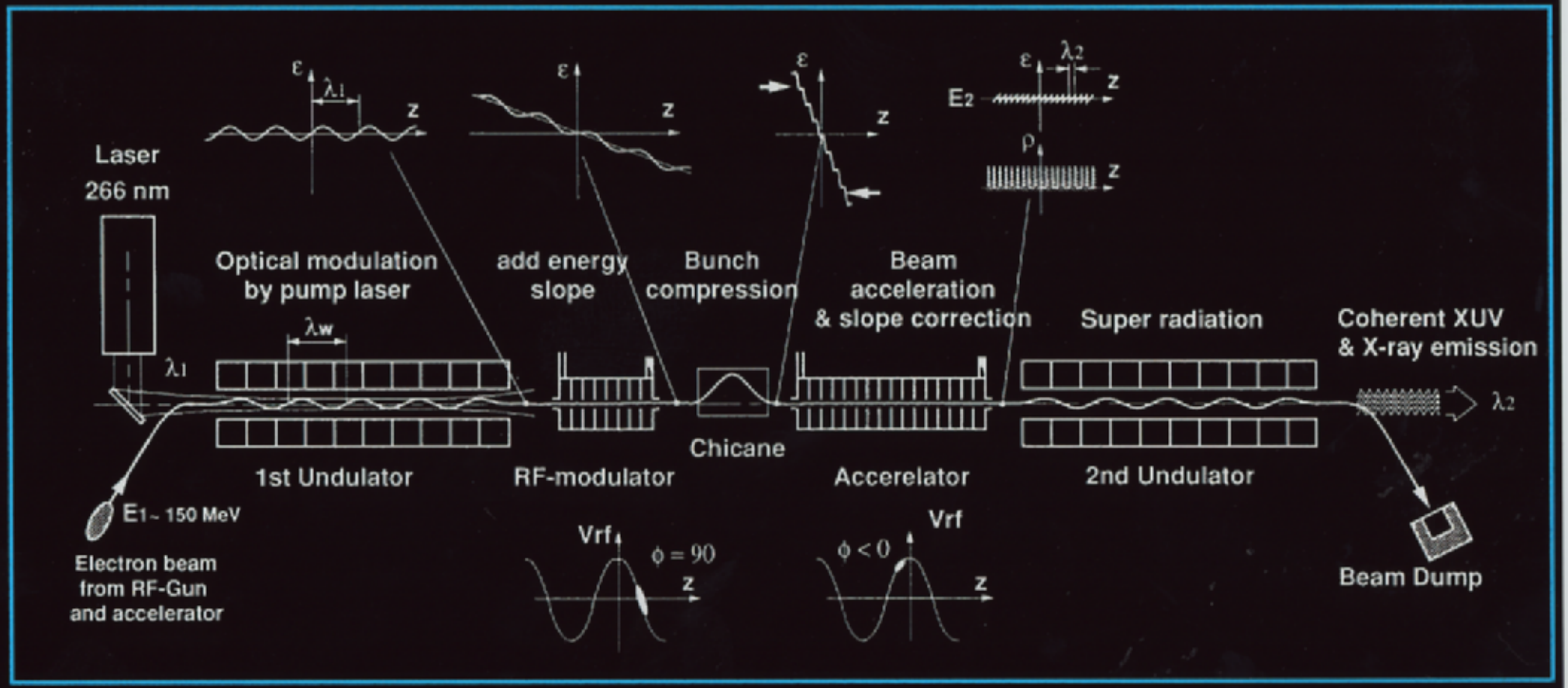
- Optical Klystron with Bunch Compressor
- Frequency up-converter of non integer up-conversion.
- CSR Effect amplifies bunching.
- Fully coherent in longitudinal direction, narrow line width, stable pulse (Fourier limited pulsed).
- For VUV, soft-X-ray.

Wavelength Compression?

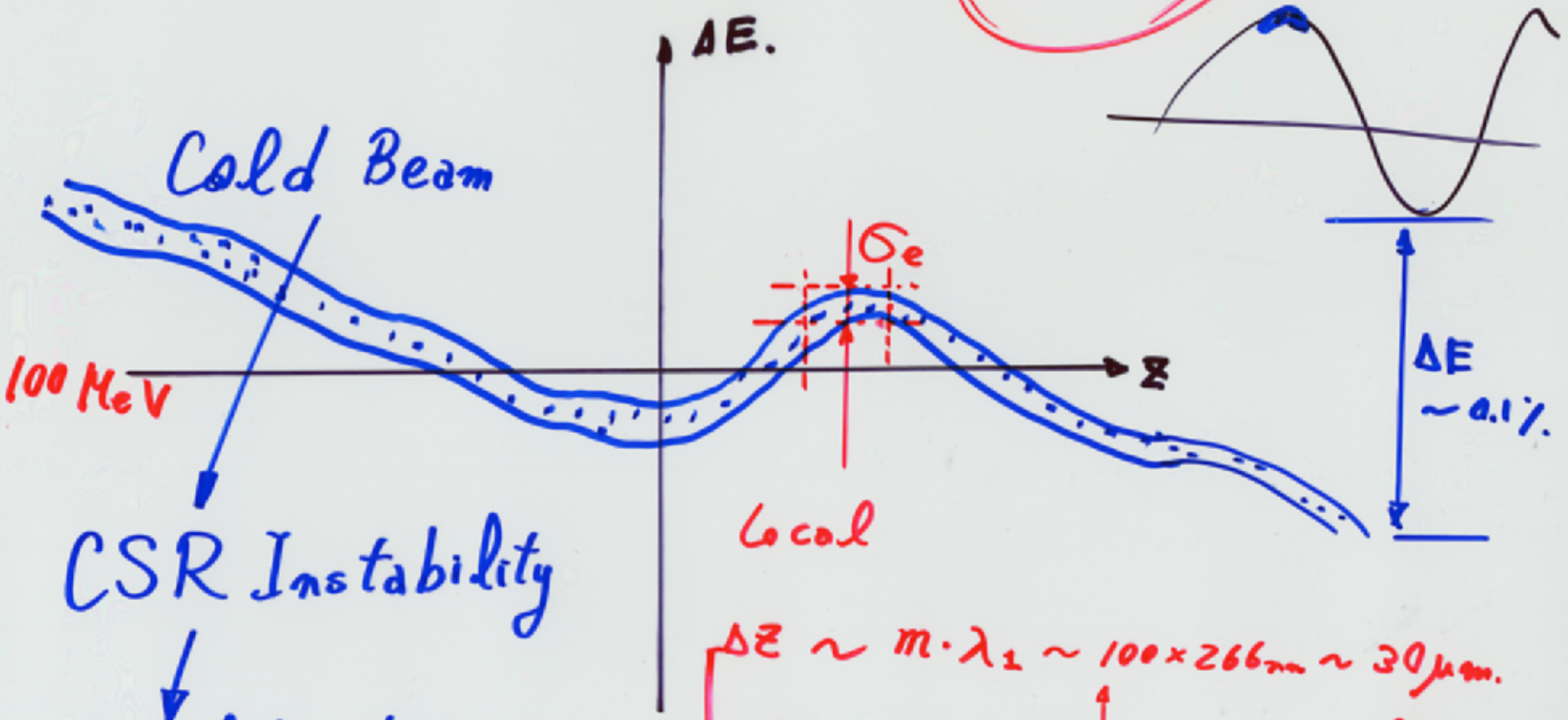


System Diagram



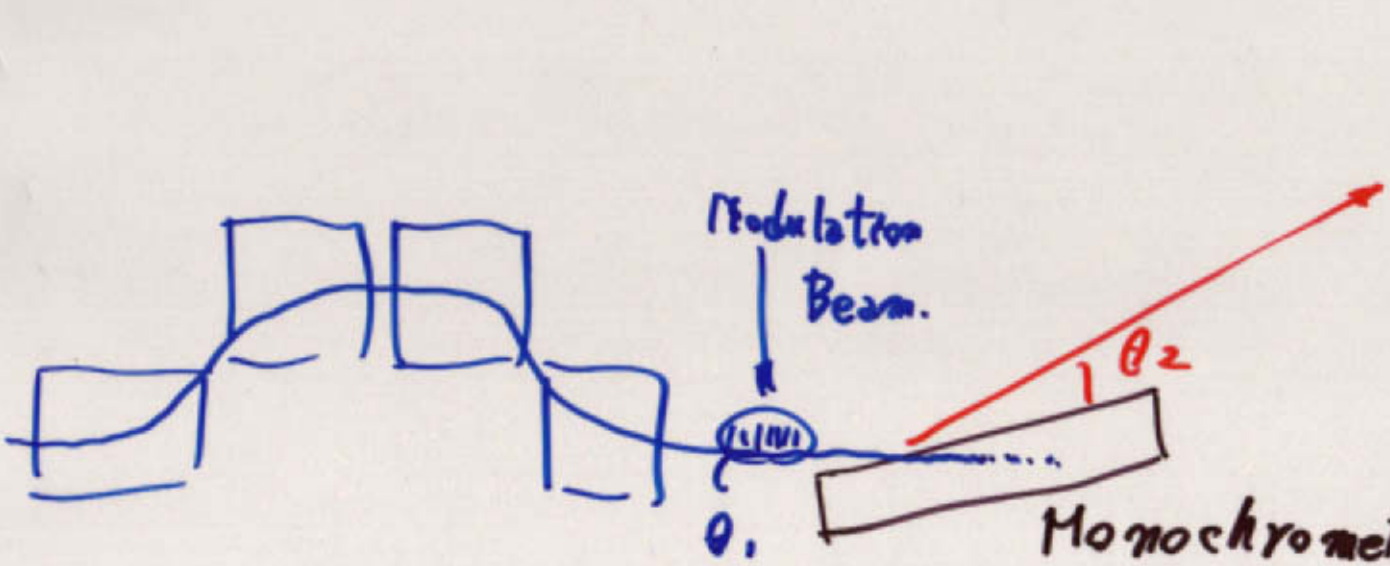
Micro-Energy Spread Slice

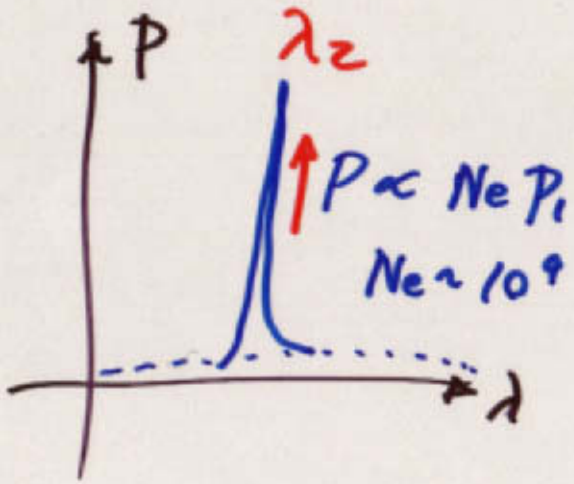
$\sigma_e/E \sim 10^{-7}$! or $\sigma_e \sim 10\,eV$?



c. i Coherent Bremsstrahlung. FEL !?

2002/01/17.
T. Shintake.





$P = N_e \cdot P_1$

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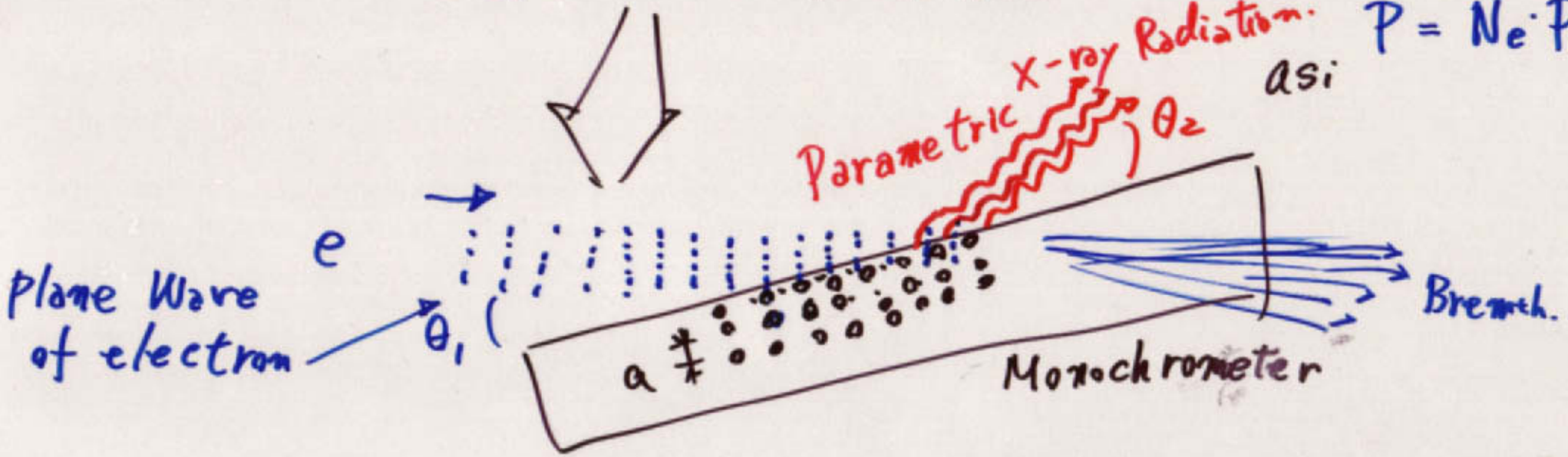


Spontaneous Parametric X-ray Radiation (PXR)

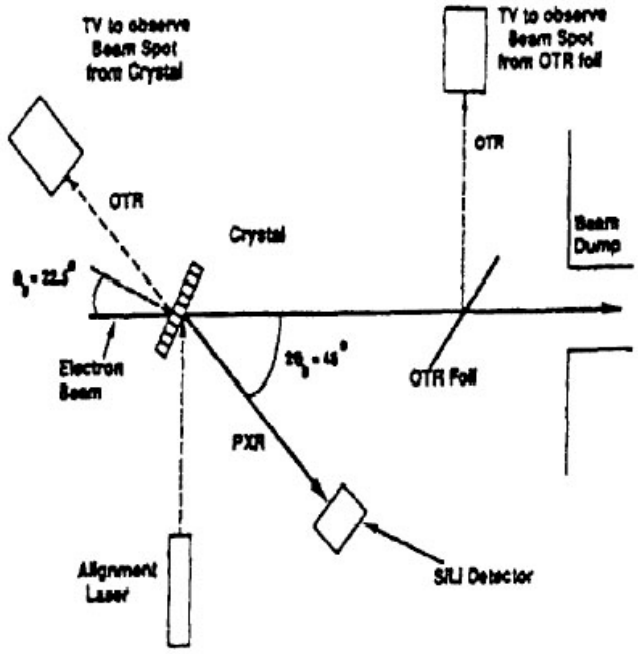


Figure 2. Experimental setup for observing parametric x-ray in the Laue geometry with $\theta_B = 22.5°$. In the Bragg geometry, the photons exit from the same side as the incoming electron beam.

order n	Energy (keV)	PXR Yield (N/e) 10^{-9}	Theory Yield (N/e) 10^{-9}	Data / Theory
1	4.88	1670	5230	0.3
2	9.53	1720	990	1.7
3	14.29	850	240	3.6
4	19.08	420	80	5.3
5	23.88	230	34	6.9
6	28.68	130	16	8.3
7	33.56	68	8	8.8
8	38.44	34	4	8.8

Table 1. Measured and Theoretical PXR yields in graphite.

X.K. Maruyama, et al, Naval Postgraduate School. Monterey CA, "A Compact Tunable X-ray Source Based on Parameteric X-ray Generation by Moderate Energy Linacs",PAC 1993

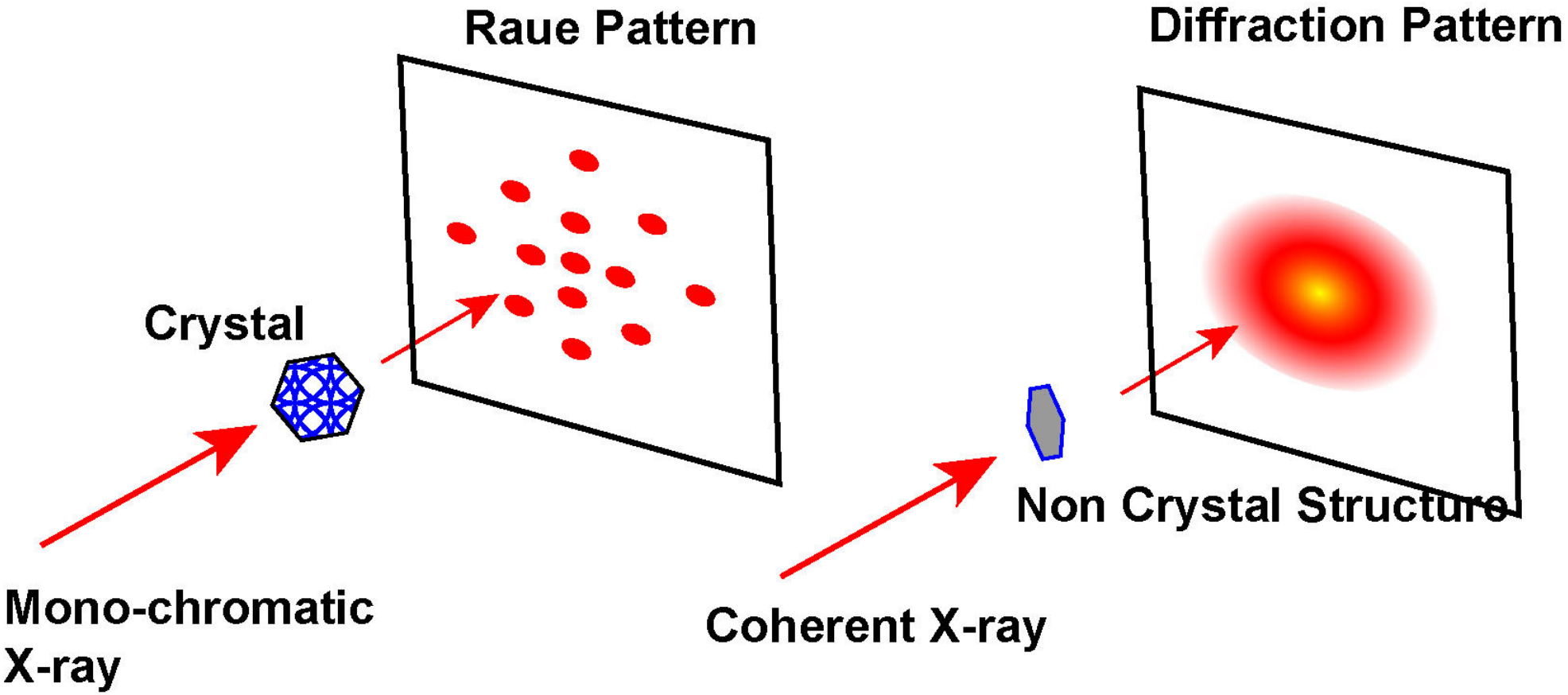
Coherent Parametric X-ray Radiation

- *Spontaneous PXR Yield is 10^{-6} photons/e.*
  Very low!
- *Bunch with Modulation Ne = 10^{6} (0.001 nC)*
  (1% density modulation in 0.1 nC bunch)
- *Intensity of coherent PXR X-ray will be*
  1 photon/electron

This is almost same value in conventional undulator.

- *Radiation is coherent!*

X-ray Diffraction Imaging



Resolve object by inverse Fourier process with phase retrieval.

High Resolution 3D X-Ray Diffraction Microscopy

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We have imaged a 2D buried Ni nanostructure at 8 nm resolution using coherent x-ray diffraction and the oversampling phasing method. By employing a 3D imaging reconstruction algorithm, for the first time we have experimentally determined the 3D structure of a noncrystalline nanostructured material at 50 nm resolution. The 2D and 3D imaging resolution is currently limited by the exposure time and the computing power, while the ultimate resolution is limited by the x-ray wavelengths. We believe these results pave the way for the development of atomic resolution 3D x-ray diffraction microscopy.

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The development of imaging techniques has played a crucial role in understanding the microscopic world. One of the recent innovations, the invention of the tunneling microscope in 1982 [1], spawned a wealth of development of probe microscopes which have found wide ranging applications. These forms of microscopy, however, are primarily sensitive to surface information. By employing innovative aberration-correction techniques, transmission electron microscopes can probe the structure of thin film samples at atomic resolution, but the samples have to be thinner than $\sim$50 nm to avoid the multiple scattering effects [2]. To nondestructively probe thicker samples (>0.5 μm), x rays are more ideal due to their longer penetration length compared to that of electrons. However, x rays are more difficult to focus than electrons. By using Fresnel zone plates, the smallest x-ray focal spot currently achievable is around 30 nm for soft x rays [3] and 150 nm for hard x rays [4]. To achieve better resolution, diffraction methodology such as x-ray crystallography, with the resolution typically limited by the quality of sample diffraction, is the method of choice. X-ray crystallography has had a tremendous impact in materials sciences, chemistry, structural biology, and other areas, but is applicable only to structures with periodic repeats (most typically crystals). Many samples, however, such as amorphous and disordered materials including polymers, strains and defects in crystals, and some inorganic nanostructures, cannot be accessed by this approach. In biology, structures such as whole cells, subcellular structures, and viruses are very often noncrystalline or nonrepetitive. At the molecular level, somewhere around 20%–40% of all of the protein molecules including most of the important membrane proteins are difficult to crystallize and are, hence, not currently accessible by x-ray crystallography.

One form of microscopy, the combination of the coherent x-ray diffraction with the oversampling phasing method [5,6], has the potential to overcome these limitations (another potential approach is to use the transport of intensity method [7]). When a finite specimen is illuminated by coherent x rays, the weakly scattered x-ray photons form a continuous diffraction pattern in the far field. This continuous pattern can be sampled at spacing finer than the Nyquist frequency (i.e., the inverse of the size of the specimen), which corresponds to surrounding the electron density of the specimen with a no-density region [8]. The higher the sampling frequency, the larger the no-density region. When the no-density region is larger than the electron density region, the phase information is, in principle, available from the diffraction pattern itself and can be directly retrieved by using an iterative algorithm [8,9]. The first demonstration experiment of this form of microscopy was carried out by using coherent soft x rays in 1999 [10]. More recently, it has been extended to image the shapes of nanocrystals by using hard x rays [11,12]. The potential applications of this approach to imaging magnetic materials [13,14], nanocrystals [15], and biomolecules [16] have also been pursued. However, the experiments that have been carried out thus far have been limited to imaging 2D samples, and the highest resolution achieved until now is around 70 nm [10,11] . In this Letter, by using coherent x rays with a wavelength of 2 Å, we report the successful imaging of a 2D buried structure at 8 nm resolution. For the first time, we also report the reconstruction of the 3D structure of a noncrystalline sample at 50 nm resolution from a series of diffraction pattern projections.

The oversampling phasing method is strongly correlated to the coherence of the incident x rays. Since oversampling a diffraction pattern corresponds to generating a no-density region surrounding the electron density of the specimen, the coherence length of the incident x rays must be longer than or equal to the overall size of the electron density and the no-density region. Typically, the higher the oversampling degree, the higher the required degree of coherence.

The required spatial and temporal coherence for the oversampling method are expressed as $\Delta\theta \leq \lambda/2Oa$ and $\lambda/\Delta\lambda \geq Oa/d$, where $\Delta\theta$ is the divergence or convergence angle of the incident x rays, λ is the x-ray wavelength, O is the oversampling degree defined as the ratio of the radius of the electron density and the no-density region to the radius of the electron density region, a the sample size, and d the desired resolution.

The sample, fabricated by electron beam lithography, consists of two single-layered Ni patterns (each with a size of $2.5 \times 2 \times 0.1$ μm) rotated relatively 65° to each other in-plane and separated by a distance of 1 μm. The sample is supported by a thin silicon nitride membrane window. Figure 1(a) shows a scanning electron microscopy (SEM) image of the sample. Because of the 1 μm separation of the two layers, the SEM image shows the pattern in the top layer, and the pattern in the bottom layer is visible only as a soft blur. The experiments were carried out on an undulator beam line at SPring-8 with a wavelength of 2 Å. To achieve the desired coherence for the imaging experiments, a 150 μm horizontal slit was placed at a distance of 27 m upstream of the sample for obtaining the spatial coherence of $\Delta\theta < 1.5 \times 10^{-5}$ rad and a Si (1,1,1) double crystal for temporal coherence of $\lambda/\Delta\lambda \sim 7500$ [17]. Two-dimensional patterns were recorded from the sample. To improve the resolution, a CCD detector was shifted in both X and Y directions to record a set of diffraction patterns at different resolutions. These patterns were then tiled together to form a high-resolution diffraction pattern. Figure 1(b) shows a diffraction pattern with 1760×1760 pixels and a resolution of 8 nm at the edge. The total exposure time of the diffraction pattern is about 45 min using unfocused x rays from the undulator beam line. Since a beam stop is used to block the direct beam, the diffraction pattern has an area of missing data at the center with a size of 60×60 pixels, which was filled in by a patch of the intensities calculated from the magnitude of the Fourier transform of an x-ray microscopy image of the sample [18]. To convert the diffraction pattern to a high-resolution image, a reconstruction was carried out by using a random phase set as an initial input and a 2.8×2.6 μm square as the finite support (which is used to separate the electron density and no-density regions). Figure 1(c) shows the reconstructed image in which a line scan through an edge indicates a resolution of 8 nm. The top and bottom layered patterns are clearly seen as overlapped in this 2D image projection, and the variation of the electron density on the nanometer scale is also visible. Five more reconstructions with different random initial phase sets were done and the reconstructed images consistently and faithfully reproduced the original sample pattern. Because of the longer penetration length of hard x rays than of electrons, it can be seen that this form of microscopy can image much thicker specimens at very high resolution. This is probably beyond the capability of both scanning probe microscopy and transmission electron microscopy.

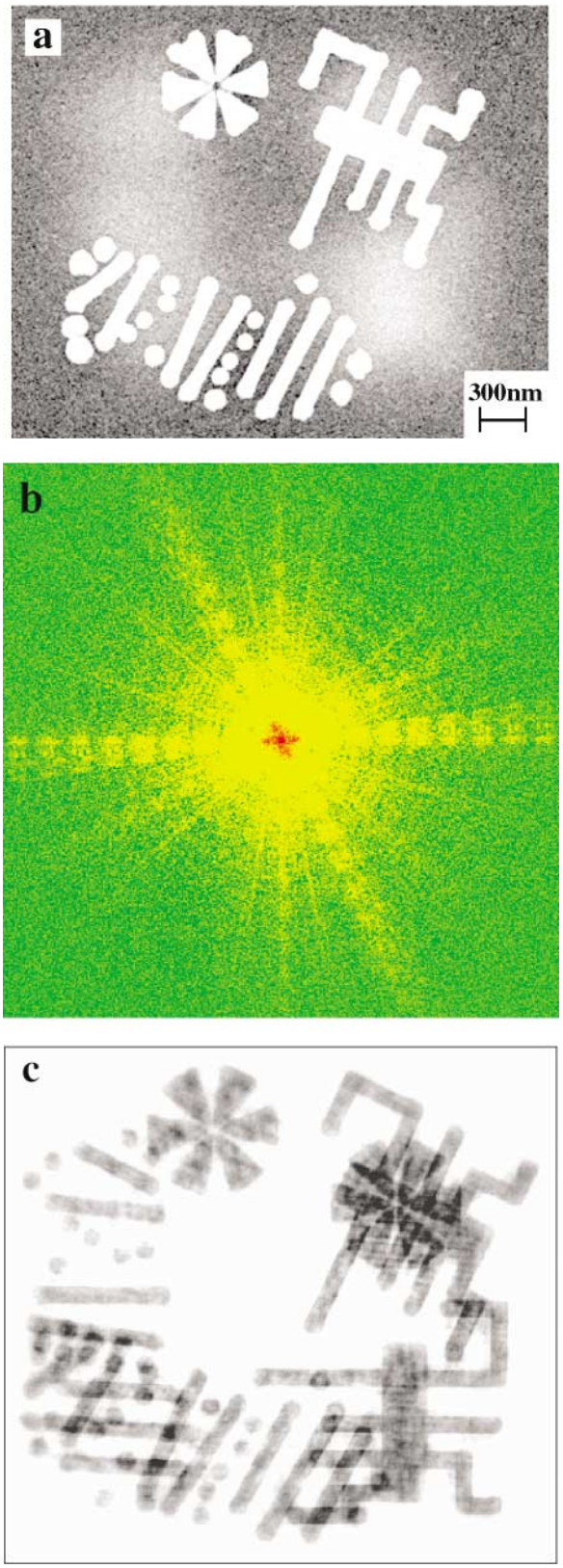

FIG. 1 (color). (a) A SEM image of a Ni sample with buried structures. (b) A high-resolution diffraction pattern (a 1760×1760 pixel array) recorded from the sample. (c) A high-resolution image reconstructed from (b).

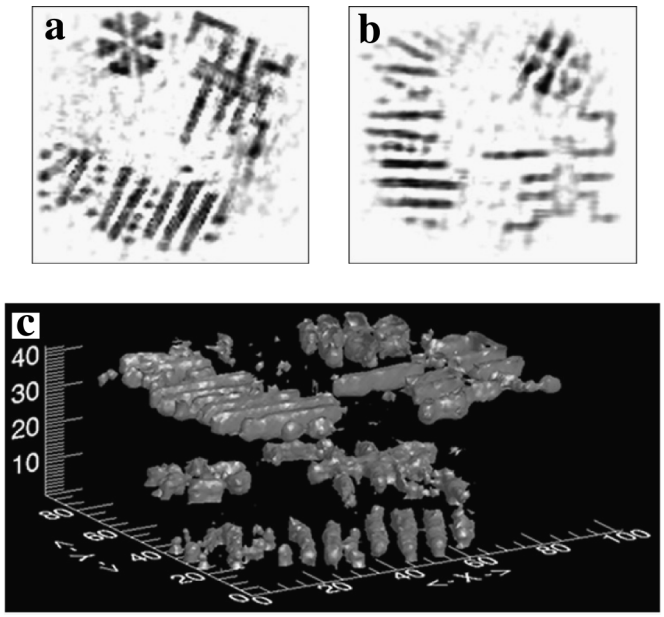

FIG. 3. The reconstruction of a 3D nanostructured material at 50 nm resolution. (a), (b) The reconstructed top and bottom layered pattern. (c) The reconstructed 3D structure displayed in isosurface rendering.

shown in Figs. 3(a) and 3(b), respectively. The two patterns are rotated relatively 65° to each other as in the original sample. Figure 3(c) shows a 3D isosurface rendering of the reconstructed image. The finest division in the z axis corresponds to 25 nm and the distance between two patterns is about 1 μm, which is consistent with the known characteristics of the sample.

We believe this form of microscopy will have wide applications in both materials and biology. For materials science samples, which are less sensitive to radiation damage, this form of microscopy can, in principle, achieve atomic resolution in three dimensions by either having a long exposure time or using higher flux coherent x-ray sources. In biology, this form of microscopy can be applied to image the 3D structures of whole cells, cellular organelles, and supramolecular structures at high resolution, while the resolution will be mainly limited by radiation damage to the specimens [19]. To alleviate the radiation damage problem, biological samples need to be frozen at the temperature of liquid nitrogen [20]. With the prospects of the x-ray free electron lasers (X-FEL) [21] providing ultrashort and extremely intense pulses, the radiation damage problem could possibly be circumvented by recording the diffraction pattern from a single biomolecule before it is destroyed by a single shot [22]. In combination of a simulated X-FEL with this form of microscopy, it has been shown that a 3D diffraction pattern calculated from 10^6 identical rubisco molecules can be successfully converted to a high quality electron density map with a resolution of 2.5 Å [16].

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