FEMTOSECOND ELECTRON DIFFRACTION AND ITS APPLICATION FOR BEAM CHARACTERIZATION AT THE PAL*

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Abstract

We are proposing a femtosecond electron diffraction (FED) proof of principle experiment at the Pohang Accelerator Laboratory (PAL). The proposed experiment will be performed at the PAL High-Brightness R&D facility using an S-band photocathode RF gun. This paper addresses one of the challenges of FED using photocathode RF gun – smearing of the diffraction patterns due to the beam divergence. Simulation studies for the testing sample -Aluminium are presented. It shows that, diffraction will be detectable if the beam divergence is smaller than 0.3 mrad. Further more, our studies also show that, the 50 μrad beam divergence can be easily observable using the Al diffraction pattern, which could be explored for high-brightness electron beam emittance characterization. The Richardson-Lucy algorithm was used to demonstrate the feasibility of structure re-construction with FED.

INTRODUCTION

FED will open up the vistas in our understanding of structure dynamics. Investigations on the temporal evolution of the dynamical behaviour in fs time scale are essential for elucidating the function of the complex chemical and biological systems. The FED signal is the Fourier transform of the nuclei and electron density distribution of the samples. By studying the diffraction pattern variation as a function of time, one is capable of investigating the temporal evolution of the molecular behaviour of the sample. Time-resolved low energy electron diffraction (below 10 KeV) and high energy electron diffraction (10-200 KeV) has already been used to determine the transient structure of molecules with a state-of-art temporal resolution of one picosecond (ps) [1]. Besides the constraint on the pulse length, another requirement is that the pulse should be sufficiently bright to provide a discernible diffraction pattern. We are proposing to perform FED using an 2 MeV electron beam produced by a photocathode RF gun at PAL [2,3]. The MeV FED is expected to have 100 fs temporal resolution and unprecedented brightness (one million electrons per pulse) [2].

Comparing to the proposed X-Ray Free Electron Laser (XFEL), FED has the following advantages:

- Large interaction cross section - 6 orders of magnitude larger than X-Ray [4].
- Compactness - 'Table-top' scale.
- Less radiation damages to the sample.

To break through the ps time resolution barrier, 3 basic conditions should be satisfied: fs pump laser, fs MeV electron beam and fs synchronism. The total temporal resolution of a FED pump-probe system is [5],

$$\Delta t^2_{total} = \Delta t^2_{laser} + \Delta t^2_{e-beam} + \Delta t^2_{VM}, \tag{1}$$

Where Δt_{laser} and Δt_{e-beam} is the duration of the pump laser and probe electron beam respectively, and Δt_{VM} is the temporal broadening caused by velocity mismatch between photons and electrons. While fs laser system is commercially available, it is a challenge to make Δt_{e-beam} and Δt_{VM} less than 100 fs. Since the gas-phase sample is typically several hundred microns thick, Δt_{VM} is always larger than 1 ps if the beam's energy is less than 1 MeV. The proposed MeV FED will benefit from the relative high energy, and Δt_{VM} is estimated to be less than 100 fs if electron beam energy is higher than 1 MeV.

Space charge effects and initial energy spread are the main sources of bunch lengthening. The temporal broadening due to initial energy spread can be estimated by [6],

$$\Delta t_{IES} = \sqrt{2m_e \Delta\varepsilon} / eE, \tag{2}$$

where m_e is the electron mass, $\Delta\varepsilon$ is the FWHM initial energy spread, and E it the electric field near the photocathode. By a proper choice of photocathode material and laser wavelength, the initial energy spread can be minimised. Additionally the electric field on the photocathode for the RF gun is at least a factor of 10 higher than the present DC gun based electron diffraction system, which will further reduce the temporal broadening. For the proposed FED system at the PAL, a single Ti:sapphire laser system will be used for both pumping the sample and producing the femtosecond electron beam. This will minimize the timing jitter between the pump laser and probe electron beam. By choosing the copper cathode and the frequency tripled Ti:sapphire laser, the initial energy spread can be reduced to less than 0.1 eV. This temporal broadening is estimated to be significantly less than 100 fs for the proposed FED. The temporal broadening due to space charge during the transportation process in a drift space is given by [7],

$$\Delta t_{SC} = 2Qz^2 / I_a R l_z \gamma^4, \tag{3}$$

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where Q is the beam charge, z is the drift length, R is the beam radius, I_a = 17 kA is the Alfven current, l_z is the bunch length and γ is the Lorentz factor. Eq.(3) clearly shows the difficulty of preservation of fs beam in KeV energy and emphasizes the preference of MeV beam. In summary, we concluded that, in order to achieve fs temporal resolution, the electron beam energy has to be increased to MeV. Further more, higher beam energy will make it possible to have more than one million electrons/pulse, which is about two orders of magnitudes higher than present DC gun based electron diffraction system. The photocathode RF gun based FED would also make it possible to perform the single-shot study of the ultrafast dynamical behaviours.

One of the main challenges of realizing the FED based on a photocathode RF gun – smearing of diffraction pattern is addressed in this report. We also explore the possibility of using electron diffraction with a known sample to characterize the electron beam emittance.

BEAM DIVERGENCE EFFECT

To simplify our calculation, we made following assumptions. First, we assume that, the interaction between the electron and sample does not depend on the incident angle of the electron. Second, electrons incident upon the sample in different directions are incoherent. Third, the observed diffraction pattern (ODP) is the clean diffraction pattern (CDP) for a single electron convolved with the beam's distribution at the detection plane where the specimen is absent. The main difference between conventional KeV FED and the proposed MeV FED is the Bragg angle for the MeV FED is approximately one order smaller. So in order to make diffraction pattern discernable, the distance between the sample and the detector have to be lengthened. Thus the ODP could be highly smeared by the beam's divergence. Since most of the theory for structure analysis starts from the CDP, restoring it from the distorted ODP is necessary.

Consider a conventional FED (beam energy ~30 KeV) with a polycrystalline Aluminium sample, the ODP and beam's distribution at the detection plane when the sample is absent is shown in Fig. 1.

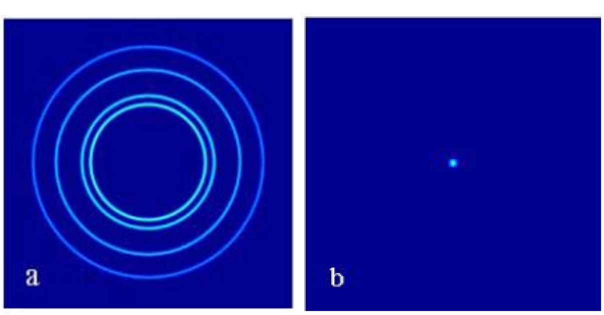

Figure 1: ODP of Al (a) and the beam's distribution (b) at the detection plane for a 30 KeV FED.

The diffraction pattern consists of four concentric rings represent the four diffraction plane 111, 200, 220 and 331 The radius of the ring are given by,

$$r = \lambda (k^2 + l^2 + m^2)^{1/2} L / a_0 , \quad (4)$$

Where a_0 = *4.05* Å is the lattice constant, L is the distance from the sample to the detector, λ is the De Broglie wavelength of the electron, and k, l, m are the Miller indices.

The relation between ODP and CDP can be described by,

$$O(r) = C(r) \otimes D(r) , \quad (5)$$

where $O(r)$ is ODP, $C(r)$ is CDP and $D(r)$ is the distribution of the electrons at the detection plane with all things being equal except the absence of the specimen. For the conventional FED, the Bragg angle is much larger than beam's divergence, so $D(r)$ is almost a Delta function and the distortion to the ODP is negligible as can be seen from Fig. 1.

As for the proposed high energy FED, λ is about one order of magnitude shorter, and hence the Bragg angle becomes comparable to beam's divergence. Lengthening the distance between the sample and the detector in order to make ODP observable will inevitably make $D(r)$ far from Delta function, so ODP may deviate from CDP significantly. The beam's divergence is mainly determined by thermal emittance; because for a low charge and short bunch, the contribution from space charge and RF force is negligible. For the proposed FED, we will adopt a scheme to further reduce thermal emittance in which the laser illuminates the cathode at oblique incidence [8]. Even when the divergence can be reduced to 0.2~0.3 mrad, it is still large enough to cause considerable distortion to the ODP.

The ODPs under various beam divergence are shown in Fig. 2. The distance between the sample and the detector is 2 m.

Fig. 2 shows that, ODPs strongly depend on the beam's divergence, which can be explored for the beam divergence and emittance characterizations. Rather roughly we may conclude if the inner two rings are distinguishable with eyes the beam divergence should be less than 50 μrad, and if the outer two rings are distinguishable with eyes the beam divergence should be less than 0.15 mrad. It is interesting to note that, one can judge beam divergence with naked eyes directly from images without any further calculations. Also from Fig. 2a and Fig. 2b, it is obvious that the resolution of beam divergence measurement based on FED pattern could be much higher than 50 μrad. It is worth mentioning that the FED pattern has higher sensitivity to divergence when the beam divergence is small. Thus this technique could be useful for ultralow emittance characterizations.

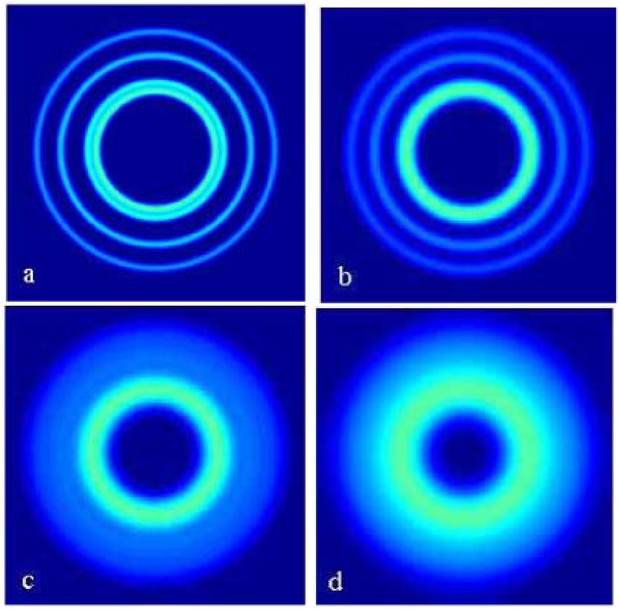

Figure 2: ODP of Al for the proposed ultrahigh energy FED. The rms divergence is 0.05, 0.1, 0.2 and 0.3 mrad for a, b, c and d respectively.

TEST OF EFFECTIVENESS FOR THE CDP RESTORATION

In principle, deconvolution of the ODP should allow retrieval of the CDP which will be further used for ultrafast dynamical behaviour investigations. Thus the effectiveness of the pattern restoration is crucial to the validity of the subsequent ultrafast process study. Generally speaking, the inverse problem is not easy and sometimes does not have a stable solution in the presence of noise for which a noise term should be added to Eq.(5),

$$O(r) = C(r) \otimes D(r) + N(r)\,, \qquad (6)$$

Normally the inversion is performed in Fourier domain to take advantage of the fact that the deconvolution in this domain is simply a division,

$$C(r) = F^{-1}\left[C(\omega)\right] = F^{-1}\left[\frac{O(\omega)}{D(\omega)} - \frac{N(\omega)}{D(\omega)}\right], \qquad (7)$$

The problem is that ODP and CDP are generally band limited and thus would decrease with increasing ω, but the noise does not behave this way. So high frequency noise may be amplified in the inversion process and the errors in the restored CDP may be too large to guarantee its validity.

For our case, we have adopted the well-known Richardson-Lucy [9-10] algorithm for the image deconvolution instead of linear inversion algorithm in which the noise amplification may occur. The restored CDP for Fig. 2c is shown in Fig. 3 for different iteration number.

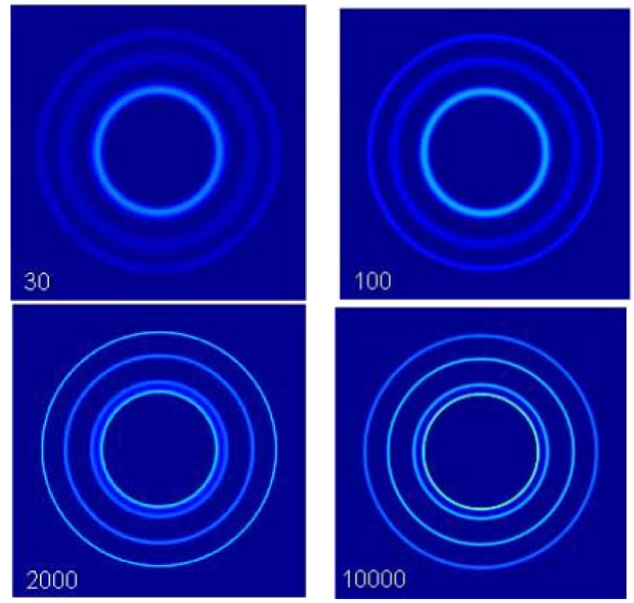

Figure 3: Restored CDP from Fig. 2c with Lucy-Richardson algorithm for various iterations.

A comparison between Fig. 1a and Fig. 3 shows satisfactory effectiveness and high precision of the restoration. This strongly supports the feasibility of the proposed ultrahigh energy FED facility. A test-of-principle experiment is under preparation and will be performed in Fall of 2005.

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