DEUTSCHES ELEKTRONEN-SYNCHROTRON DESY

DESY SR-76/09
April 1976

Precise Interferometric Measurement of the Ni K-edge Forward Scattering Amplitude with Synchrotron X-Rays

by

U. Bonse and G. Materlik
Institut für Physik, Universität Dortmund

2 HAMBURG 52 · NOTKESTIEG 1

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
2 Hamburg 52
Notkestieg 1
Germany

Precise Interferometric Measurement of the Ni K-edge Forward Scattering Amplitude with Synchrotron X-Rays*

U. Bonse and G. Materlik

Institut für Physik, Universität Dortmund

We present the first high precision measurement of the complete wavelength dependence of the K-edge dispersion including the whole range of the Kossel and Kronig structure. Synchrotron X-rays of DESY, Hamburg, have been used providing a highly collimated and intense beam with continuous spectral distribution for interferometric measurement. In comparing the result with other experimental and theoretical f' values and also with the absorption spectrum, quite good agreement is obtained.

There exists a group of quite important experiments the success of which is essentially dependent on the availability of precise values of the real part of the coherent anomalous forward scattering amplitude $f = f_o + f' + if''$. We mention here (1) the method of anomalous dispersion in structure determination[1], (2) the determination of surface electron densities by total reflection[2], (3) the precision measurement of electron densities[3], and (4) the absolute measurement of diffracted intensities from powder samples[4].

Furthermore, the continuous measurement of the wavelength dependence of f' in the vicinity of absorption edges opens the way for the development of a so-called dispersion spectroscopy which in the future might compete with the well-known absorption spectroscopy which has been practiced for nearly 50 years by now and which played a major role in the understanding of the electronic structure of matter[5].

In the frame work of an one electron model the coherent forward scattering amplitude for X-rays is given by[6]

$$f(\omega) = \sum_I f_{II} - \frac{1}{m} \sum_I \sum_Z \left\{ \frac{\left| (\vec{P}_I \cdot \vec{\mathcal{E}}\, e^{i\vec{k}\vec{r}_I})_{IZ} \right|^2}{E_{IZ} - E - i\hbar\Gamma_{IZ}/2} + \frac{\left| (\vec{P}_I \cdot \vec{\mathcal{E}}\, e^{i\vec{k}\vec{r}_I})_{IZ} \right|^2}{E_{IZ} + E} \right\} \qquad (1)$$

$\vec{P}_I$ is the momentum operator of the electron in state $|I\rangle$ which at position $\vec{r}_I$ interacts with a photon of energy $E=\hbar\omega$, polarization vector $\vec{\epsilon}$ and wave vector $\vec{k}$. E_{IZ} is the energy difference of initial state $|I\rangle$ and intermediate state $|Z\rangle$, Γ_{IZ} its natural line width.

f_o is determined by the first term of equation (1). The resonance scattering and thus f' and f" are determined by the second term. Separately for each electron f' and f" are connected by a Kramers-Kronig relation[6-7].

$$f_I'(\omega) = \frac{2}{\pi} P \int_{\omega_I}^{\infty} \frac{\omega' \cdot f_I''(\omega')}{\omega^2 - \omega'^2} d\omega' \qquad (2)$$

Consequently f' contains just as f" information about the various electron states involved, namely about the deep initial states, the empty states near the Fermi edge (Kossel structure) and about those intermediate states which are modulated by photoelectrons that are backscattered from neighbouring atoms (Kronig structure, EXAFS).

X-ray interferometry as developed by Bonse and Hart[8] offers the possibility of measuring f' with very high precision. The strength of the method had first been shown in the region of normal dispersion[9-10] and then also near an absorption edge[11-13], however for a set of some discrete wavelength only. In the latter case, because of the drastically increasing absorption, for a precision measurement over 2 continuous wavelength range a more sophisticated experimental version for instance one using synchrotron X-rays is needed.

The successful use of synchrotron X-rays in the interferometric f' measurement was first reported by Bonse and Materlik[14] who developed the X-ray analogue to the direct measurement of anomalous dispersion in light optics (see Wood[15]). The experimental arrangement is shown in Fig.1 and has been described in detail elsewhere[14, 16]. The essential features

of this technique are:

(1) the separation and at-will selection of harmonics and of polarization states in the incident beam, (2) the possibility to employ the two wavelength method to eliminate the sample thickness, (3) the feasibility of f' measurements in the wavelength interval extending at least between 0.3 to 3.0 Å and for all materials independent of state, (4) the possibility to measure f' and f" simultaneously of the same sample.

The first result obtained is shown in Fig. 2. It is the anomalous dispersion spectrum of Ni measured over the complete K-edge wavelength region with a spectral resolution $\Delta\lambda/\lambda \leq 2 \times 10^{-4}$ at each point. On the long wavelength side of the edge a one parameter least square fit of Hönl's theory[17] is also drawn in Fig. 2. In the simple picture where a correction of Hönl's frequency dependence of the oscillator density is omitted the fit gives a total K-oscillator strength of 1.105 electrons. However, from the difference between measured data and Hönl's theory near the edge and above 1.57 Å we conclude that the frequency dependence must indeed be corrected. The discrepancy near the edge is mainly due to negligence of Γ_{IZ}, whereas disregard of L, M, ... -dispersion causes an error everywhere in the theoretical curve[18] what was also more recently discussed by Wagenfeld[7].

For the CuK α_1 wavelength the most reliable f' values are presented in Table I.

TABLE I. Comparison of measured (BM, DMR) and theoretical (W, CL) f' values for λ-CuK α_1.

BM[a]	DMR[b]	W[c]	CL[d]
$-3.014 \pm 2.3\%$	-3.081	-2.979	-2.956

[a] This work

[b] See Ref. 12.

[c] H. Wagenfeld, private communication.

[d] D. T. Cromer and D. Libermann, J. Chem. Phys. 53, 1891 (1970).

The error of 2.3% of the value measured in this work is that of a single measured point, although the value -3.014 is probably much more accurate because it was obtained with λ-averaging from the Hönl fit.

On the short wavelength side of the edge in Fig. 2 we have indicated also the fine structure measured in absorption by Couchois and Manescu[19]. K_1 marks the onset and K_2 the inflection point of the absorption edge. The positions of their maxima $\alpha, \beta, \gamma, \delta, \varepsilon, \zeta$ (full lines, sequence beginning near K_2) and their relative minima within β and within γ (point-dashed lines) and their minima: 1st min, 2nd min, A, B, C, D, E (dashed lines) correspond very well to the clearly visible fine structure of the measured f' spectrum.

It is concluded that as a result of the very good harmonization[20-21] of X-ray interferometry and synchrotron radiation a method has been established whereby it is possible to measure the complete coherent photoabsorption cross section for X-rays with high precision. In comparison to pure absorption spectroscopy which recently also became a more handy tool by the use of synchrotron radiation[22-23] the f' measurement is performed with a simpler spectral window and the simultaneous measurement of f' and f" can answer question about the influence of the spectral window on the measured f" spectrum. The importance of a simultaneous measurement of f' and f" has also recently been pointed out by Fukamachi and Hosoya[24], who measured f' and f" of Ga in GaP in a very narrow region near the Ga K-edge.

We thank the European Molecular Biology Laboratory (EMBL) group for generous hospitality in their experimental site at DESY in Hamburg and C. Kunz and E. E. Koch of the Synchrotron Radiation Group for technical advise.

* Research partially supported by funds of Bundesminister für Forschung und Technologie through DESY, Hamburg, Az. PH/SS 04 DO 1.

† Currently at Department of Applied and Engineering Physics, Clark Hall, Cornell University, Ithaca, N.Y. 14853.

Fig. 1 Experimental arrangement for interferometric f' measurement with synchrotron X-rays.

Fig. 2 Wavelength dependence of dispersion correction f' on either side of the K-edge of nickel. K_1 onset, K_2 inflection point of absorption edge. Full lines indicate positions of maxima, dashed lines those of minima of the absorption spectrum given in Ref. 19. Note the correspondence of the measured fine structure of f' with that of the absorption spectrum.

[1]Anomalous Scattering edited by S. Ramaseshan and S.C. Abrahams (Munksgaard, Kopenhagen, 1975).

[2]L.G. Parratt, Phys. Rev. 95, 359 (1954).

[3]R.W. James, The Optical Principles of the Diffraction of X-rays (G. Bell & Sons, London, 1958).

[4]P. Suortti and L.D. Jennings, Acta Cryst. A 31 S 3, S 218 (1975)

[5]L.V. Azároff, X-ray Spectroscopy (Mc Graw-Hill, New York, 1974).

[6]J.J. Sakurai, Advanced Quantum Mechanics (Addison Wesley, Reading Massachusetts, 1973).

[7]H. Wagenfeld in [1] page 13.

[8]U. Bonse and M. Hart, Appl. Phys. Lett. 6, 155 (1965).

[9]U. Bonse and H. Hellkötter, Z. Physik 223, 345 (1969).

[10]D.C. Creagh and M. Hart, Phys. Stat. Sol. 37, 753 (1970).

[11]U. Bonse and G. Materlik, Z. Physik 253, 232 (1972).

[12]V. B. Drozdovskij, I. P. Michajljuk and M. D. Raranski, Ukr. Fiz. Zur.19, 1215 (1974).

[13]C. Cusatis and M. Hart, Acta Cryst. A 31 S 3, S 254 (1975).

[14]U. Bonse and G. Materlik in [1] page 107.

[15]R. W. Wood, Physical Optics (Macmillan, New York, 1911).

[16]G. Materlik, Ph. D. thesis (Dortmund, 1975).

[17]H. Hönl, U. Physik 84, 1 (1933).

[18]H. Eisenlohr and G. L. Müller, Z. Naturforschg. 8a 429 (1953).

[19]Y. Couchois and I. Manescu, J. chim. phys. Physico-Chim., Biol. 47, 892 (1950).

[20]U. Bonse and G. Materlik, Acta Cryst. A 31 S 3, S 232 (1975).

[21]U. Bonse, G. Materlik and W. Schröder, J. Appl. Cryst., submitted.

[22]B. M. Kincaid and P. Eisenberger, Phys. Rev. Lett. 34, 1361 (1975).

[23]R. W. Fairchild, Ph. D. thesis (Ithaca, 1975).

[24]T. Fukamachi and S. Hosoya, Acta Cryst. A 31, 215 (1975).

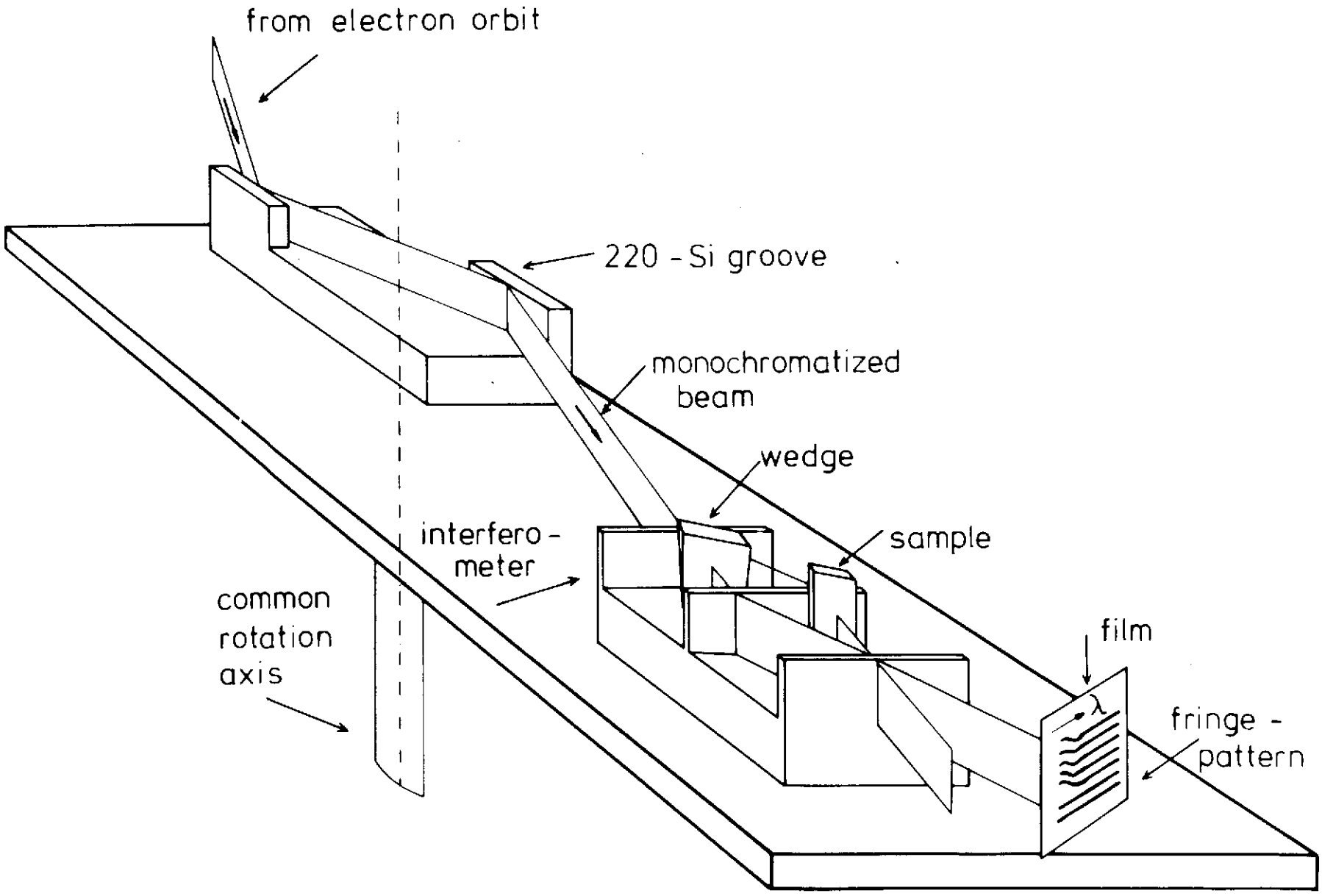
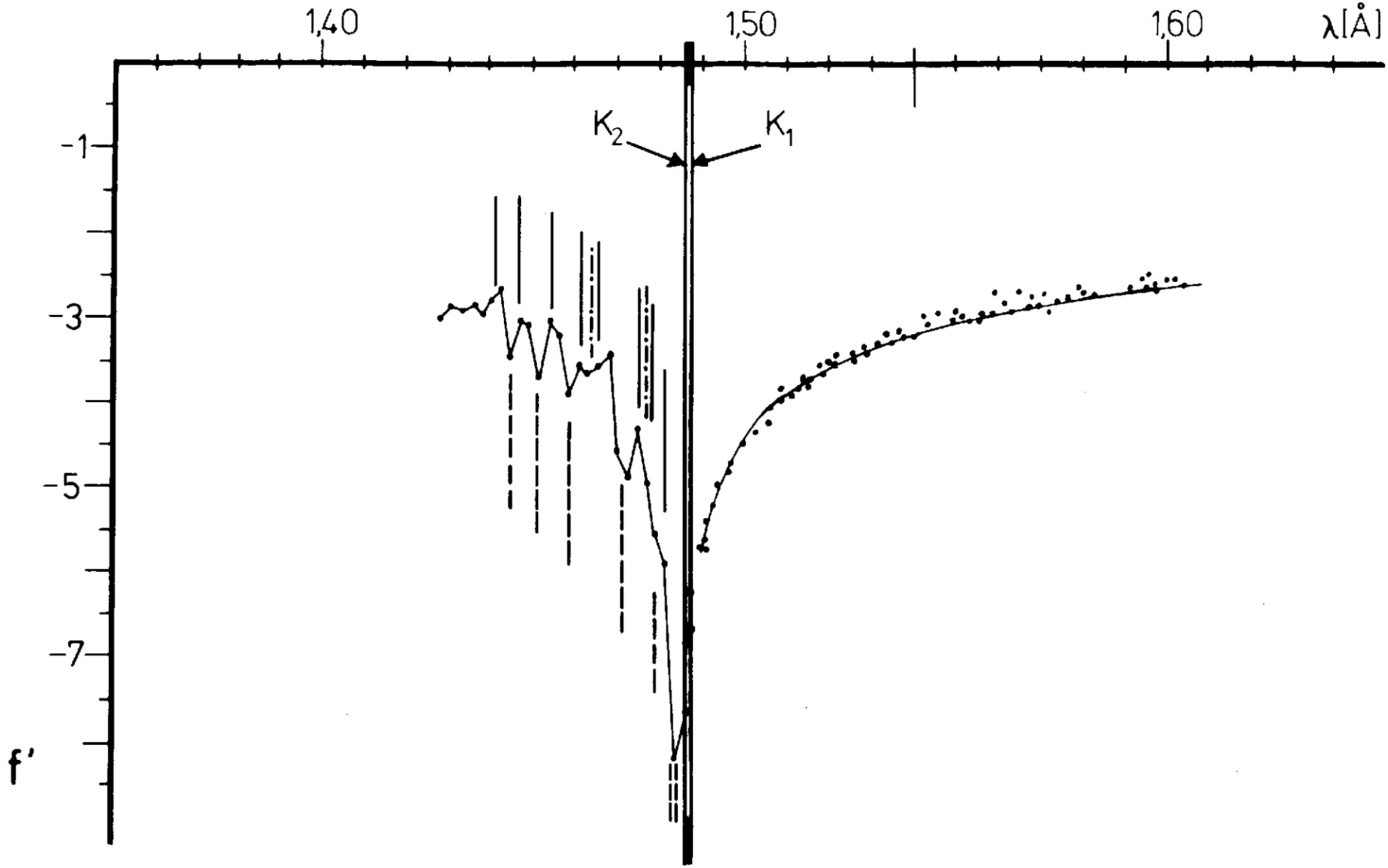

from electron orbit
220 - Si groove
monochromatized beam
wedge
interfero - meter
sample
common rotation axis
film
λ
fringe - pattern

Fig. 1


1,40
1,50
1,60
λ[Å]
K2
K1
-1
-3
-5
-7
f'