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学位論文題目	Design, Fabrication and Performance of Monochromators and its Application for Si and GaAs Lattice Spacing Measurement using Synchrotron Radiation
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論文内容の要旨

X-ray optic systems have been developed for the study of the relative lattice spacings of Si-wafers using synchrotron radiation (SR). Since, unlike a X-ray tube, SR has no characteristic [wavelength] spectral lines, a new tool of (+,+) high resolution channel-cut monolithic monochromators (MM) are introduced in the systems as a wavelength selective device. Using two types of MM, two schemes are proposed and applied to the study of the lattice spacings of Si-wafers. The lattice spacing differences are determined in the range of sub ppm level, for example in scheme-1 we obtain 0.6 ppm and 0.2 ppm in scheme-2. One of the practical advantages of this system is that it can be applied for a fast and precise measurement of the lattice spacing changes due to the doping and defects in Si, GaAs and other single crystals.

Lattice spacing measurements of Si and other single crystals is of importance both for fundamental solid state physics and applications. For decades, especially Si is routinely used in the semiconductor industry as well as commonly employed in beamline optics of synchrotron radiation facilities and other x-ray experiments. Much of the works on lattice spacing measurement has been reported using well defined wavelength of laboratory x-ray sources. In contrast relatively few works have been reported using the state-of the art SR x-ray source.

Unlike X-ray tube data for which tables of wavelengths of the characteristic spectral lines are available, there are no lines in the synchrotron radiation (SR) spectrum. Thus it demands to make schemes and data available for precise determinations of lattice spacing which utilize the SR source. Thus one of our main motivation is to develop a high precision relative lattice spacing measurement system using SR for synchrotron radiation users. One of the main advantage of these systems is that in most of the applications purposes, one does not need high accurate absolute value, but only a relative high precision value of wavelength.

To this end, we have constructed two X-ray optics systems for relative high precision lattice spacing measurement of single crystal using synchrotron radiation. In our new optics energy selective (+,+) channel-cut monolithic monochromator together with higher angle resolution goniometer with a precision of 0.36 arc sec has been introduced. We have designed and fabricated several kinds of MM that give a fixed exit beam position and provide a convenient setting of the whole X-ray optics. With the current set-up it is simple to make another d-spacing measurement, if need be by simply replacing the monochromator with another. In addition to the measurements of Si crystals, measurements for other materials such as GaAs crystals can easily be performed. The monolithic double crystal monochromator is obtained from a single perfect crystal as a means of obtaining an X-ray beam of well defined wavelength. Monolithic monochromator (MM), is in effect a single perfect crystal where two sets of Bragg planes play the role of two seperate crystals. As the interplanar angle between the concerned diffraction plane is fixed in the MM therefore the wavelength emerging from this device is highly stable and is extremely stable against temperature variations. The two types of MM allow us to propose two schemes for the lattice spacing measurement. Approximately, the precision achieved in $\Delta d/d$ in these systems is in the range of 10^{-7} to 10^{-8}.

1. Monolithic Monochromator:

In order to obtain suitable wavelength for experiments from MM, we consider the following three equations:

$$\lambda_1 = \lambda = 2d_1 sin\theta_1(1 - \delta/\sin^2\theta_1)$$

$$\lambda_2 = \lambda = 2d_2 sin\theta_2(1 - \delta/\sin^2\theta_2)$$

$$\theta_1 + \theta_2 + \beta_0 = \pi$$

where δ represents the real part of refraction index and β_0 is the interplanar angle between the two planes, θ_1 and θ_2 are the Bragg angles for two diffraction planes d_1 and d_2 respectively.

(1) Solving the above equations the equations yield after simple algebra:

$$\lambda = \frac{2d_1 sin\beta_0}{[(\sqrt{(h_2^2+k_2^2+l_2^2/h_1^2+k_1^2+l_1^2)} - \cos\beta_0)^2 + \sin^2\beta_0]^{1/2}} \tag{1}$$

(2) if $d_1 = d_2 = d$ Eq. 1 further reduces to :

$$\lambda = \frac{2d\sin\beta_0}{[(1-\cos\beta_0)^2 + \sin^2\beta_0]^{1/2}} \tag{2}$$

Using Eqs. 1 and 2 a simulation code MMCD [Monolithic Monochromator Crystal Design] has been realized which can generate different wavelengths using (h_1, k_1, l_1) and (h_2, k_2, l_2) combinations . Our code relies on the Deslatte's (1973) lattice spacing of silicon wafer of d_{220} = 1.9201715±0.0000006 angstrom at 25^0C.

Two types of Monolithic monochromator [MM] have been designed and fabricated on the basis of Eqs. 1 and 2. These are notated by Type-1 and Type-2. Type-1 is used in the experimental set-up, refered to as scheme-1 and similarly Type-2 is utilized in the set-up called scheme-2.

Figs. 1 and 2 show the MM that have been made for the wavelength defined by Eqs. 1 and 2 respectively. Some of the simulated wavelengths from MMCD code is given in Table I for both types of MM with refraction correction δ, given by $\delta = 4.48 \times 10^{-6} n_0 \lambda^2$. Here n_0 is the number density, for example for Si $n_0 = 699nm^{-3}$, and λ is the wavelength of x-rays.

Table 1
Type 1 and type 2 simulated wavelength from *MMCD* computer code.

Type 1: different indexes. Type 2: same indexes.

$h_1k_1l_1$	$h_2k_2l_2$	HKL	β_0	θ_1	θ_2	λ	δ
Type 1							
1 5 5	$\bar{5}$ 3 1	$\bar{4}$ 8 6	69.2044	63.1437	47.6519	1.356950	0.57719×10^{-5}
3 5 5	$\bar{3}$ 3 1	0 8 6	70.8196	75.8031	33.3772	1.370938	0.58915×10^{-5}
3 3 5	$\bar{3}$ 5 1	0 8 6	73.5280	57.1730	49.2990	1.391940	0.60734×10^{-5}
1 1 3	$\bar{5}$ $\bar{3}$ 5	$\bar{4}$ $\bar{2}$ 8	74.0515	25.2230	80.7255	1.395642	0.61057×10^{-5}
1 1 3	$\bar{3}$ $\bar{5}$ 5	$\bar{2}$ $\bar{4}$ 8	74.0515	25.2230	80.7255	1.395642	0.61057×10^{-5}
1 5 5	5 1 5	6 6 10	46.6641	66.6680	66.6680	1.396621	0.61143×10^{-5}
1 1 3	$\bar{5}$ 3 1	$\bar{4}$ 4 2	104.7631	25.3769	49.8599	1.403594	0.61755×10^{-5}
3 5 5	1 5 $\bar{1}$	4 10 4	54.8119	83.0088	42.1792	1.403614	0.61757×10^{-5}
1 1 5	1 1 7	2 2 12	42.4407	68.3502	69.2091	1.410846	0.62686×10^{-5}
Type 2							
1 3 5	3 $\bar{5}$ $\bar{1}$	4 $\bar{2}$ 4	119.0593	30.4704	30.4704	0.931039	0.27172×10^{-5}
3 5 5	$\bar{5}$ $\bar{3}$ 5	$\bar{2}$ 2 10	94.8614	42.5693	42.5693	0.956631	0.28686×10^{-5}
1 3 5	5 $\bar{1}$ $\bar{3}$	6 2 2	111.8037	34.0981	34.0981	1.029302	0.33210×10^{-5}
3 3 5	$\bar{3}$ 5 $\bar{3}$	0 8 2	102.0815	38.9593	38.9593	1.041527	0.34004×10^{-5}
1 5 5	$\bar{1}$ $\bar{5}$ 5	0 0 10	91.1235	44.4382	44.4382	1.064914	0.35548×10^{-5}
1 3 5	$\bar{5}$ $\bar{3}$ 1	$\bar{4}$ 0 6	104.9006	37.5497	37.5497	1.118970	0.39249×10^{-5}
1 5 5	$\bar{5}$ 5 $\bar{1}$	$\bar{4}$ 10 4	72.8954	53.5523	53.5523	1.223493	0.46924×10^{-5}
3 3 5	$\bar{3}$ $\bar{3}$ 5	0 0 10	80.6311	49.6854	49.6854	1.263037	0.50006×10^{-5}
1 3 5	$\bar{5}$ 3 $\bar{1}$	$\bar{4}$ 6 4	91.6373	44.1814	44.1814	1.279591	0.51325×10^{-5}
1 5 5	5 5 1	6 10 6	46.6641	66.6680	66.6680	1.396621	0.61143×10^{-5}
1 3 5	$\bar{3}$ 5 $\bar{1}$	$\bar{2}$ 8 4	78.4630	50.7685	50.7685	1.223493	0.46924×10^{-5}
3 3 5	$\bar{3}$ $\bar{3}$ 5	0 0 10	80.6311	49.6854	49.6854	1.422186	0.63402×10^{-5}
1 1 5	$\bar{1}$ 5 $\bar{1}$	0 6 4	92.1226	43.9387	43.9387	1.450516	0.65953×10^{-5}
1 3 5	$\bar{1}$ $\bar{3}$ 5	0 0 10	64.6231	57.6885	57.6885	1.551732	0.75479×10^{-5}
1 3 5	5 3 1	6 6 6	57.1216	61.4392	61.4392	1.612607	0.81517×10^{-5}

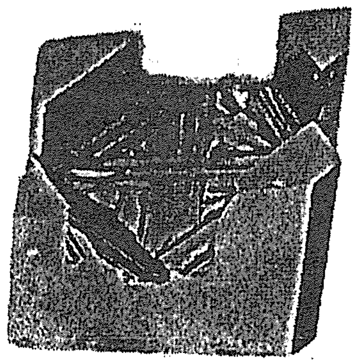

Figure 1
Type 1 monolithic monochromator designed for a wavelength of 0.1410 nm with index (1, 5, 1), (1, 1, 7), ($\bar{1}$, $\bar{5}$, $\bar{1}$), ($\bar{1}$, $\bar{1}$, $\bar{7}$).

Table
Relative lattice-spacing values measured using scheme 1 for Si and GaAs samples.

Sample number	Sample type	Sample index	MM wavelength (mm)	Average $\Delta d/d$ ($\times 10^{-7}$)
Si	FZ	(444)	0.1542	6.1
Si	CZ	(800)	0.1356	6.3
GaAs	CZ	(800)	0.1356	8.0

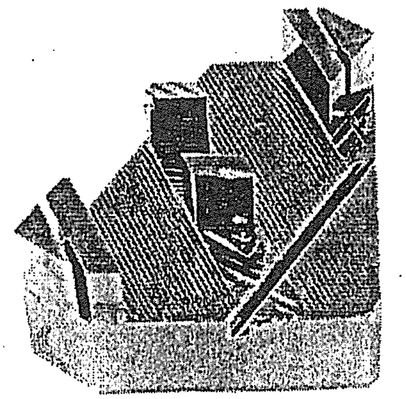

Figure 2
Type 2 monolithic monochromator for a wavelength of 0.1612 nm with index (5,1,3),(1,5,3),($\bar{5}$,$\bar{1}$,$\bar{3}$),($\bar{1}$,$\bar{5}$ $\bar{3}$).

As mentioned above, two relative lattice spacing measurement methods using two types (i.e scheme-1 and scheme-2 based respectively on the diffraction plane conditions $d_1 \neq d_2$ and $d_1 = d_2$) of (+,+) energy selective MM with SR have been developed. Results of the two schemes are summarized in this section.

In scheme-1 we applied our method to several MM. The novelty of the method is that in each case MM and sample can be changed. For the Si sample grown by FZ and for the plane (444) a MM wavelength of 0.1410 nm is utilized. Nine measurements were taken at different position of Si wafer and the measured d_{444} value obtained on the average from nine measurement points is 0.078390564 nm $\pm 2 \times 10^{-8}$. The average value of $\Delta d/d$ is 6.2×10^{-7}. Table III shows the results of our lattice spacing measurement for Si grown by CZ, and FZ methods where the planes (800) and (444) are considered in addition to GaAs grown by CZ method for the plane (800). We can see that the average value of $\Delta d/d$ is higher for GaAs compared to that of Si. Temperature and refraction corrections have been taken into account. In scheme-2 two equivalent planes and a few arc sec rotation (D) of the samples provide two quasi-simultaneous Bragg diffraction. The results of seventy measurements taken at different positions of Si(153) FZ prepared wafer showed that the approximate average value of $\Delta d/d$ is 1.1×10^{-7}.

Temperature variation has been carefully monitored. The true value of d is given by $d_{obs} + \Delta d_r + \Delta d_t$, where Δd_r and Δd_t are the refraction and temperature corrections respectively. The refraction correction is calculated for wavelength .16126 nm as 0.00000749Å and temperature correction was0.00000291Å to the d-value. The measured d_{153} value obtained on the average from seventy measurement points is 0.091801632 nm $\pm 2 \times 10^{-8}$. For 70 measurement the $\Delta d/d$ obtained from ΔD is within 0.2 ppm level. The standard deviation calculated from 100 measurement of differential peak difference D at one point was 2×10^{-8}.

In type-2 monochromating there is a possibility of third beam diffraction which may modify the intensity. Detailed calculation shows in our present study that there could be a Bragg-peak shift when we consider the 3-beam case and the shift is around 12 micro-radians with respect to the 2-beam. Table II shows some of the monochromator designed together with their characteristics parameter.

Further, it is hoped that the Monolithic monochromator and the systems developed will result in a more widespread use in the condensed matter research.

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Table 2
Monochromator and some of their design parameters.

Diffraction planes	Bragg angles θ_1, θ_2 (°)	Wavelength (nm)	$L \times B \times H$ (mm)	Beam gap (mm)	$\Delta\lambda/\lambda$ ($\times 10^{-6}$)	$\Delta\theta$ ($\times 10^{-6}$)
(5 9 11), $(7\,\bar{7}\,\bar{7})$	74.30, 50.77	0.0694067	45 × 40 × 40	7	0.5	1.6
$(3\,3\,\bar{3})$, $(\bar{3}\,\bar{5}\,\bar{3})$	47.53, 68.58	0.1542067	55 × 52 × 50	29	2.8	9.2
(1 5 1), (1 1 7)	42.44, 68.06	0.1410846	35 × 42 × 35	15	2.5	8.7
(5 1 3), (1 5 3)	61.43, 61.43	0.1612607	60 × 55 × 37	35	4.3	0.1
(1 5 5), $(\bar{5}\,3\,1)$	62.78, 47.65	0.1352569	40 × 30 × 35	0	2.3	6.2

論文の審査結果の要旨

シリコンなどの完全性の高い結晶では、僅かな量の不純物などの格子欠陥の存在により格子定数が変化する。すなわち、格子定数の変化を精密に測定することにより格子欠陥自体の研究に有用な研究手段となる。また、シリコン結晶の格子定数の測定は、アボガドロ数の実験的決定にも重要な一部分をなしており重要である。　また、非弾性散乱測定用には１０数 keV の放射光用を 1meV 以下のエネルギー分解能で分光するための分光結晶においては、結晶内での格子定数のばらつきが 10^{-8} 以下に抑えられていることが必要で、当然そのような微小な格子定数の変化を検出する方法が必要とされている。

Rahman 氏は単結晶内の複数個の格子面で逐次回折が起きることを利用した放射光用モノクロメーターを考案・開発し、単結晶の格子定数を高精度で測定できるシステムを完成した。すなわち、一体となっているシリコン単結晶ブロック内に反射表面を複数個もち、結晶内の複数の回折格子面で逐次反射を４回繰り返し、その幾何学的配置固有の波長を持つ単色・平行化したＸ線ビームを得、ボンド法と呼ばれている手法を用いて格子定数を 10^{-7} ～ 10^{-8} の精度で測定する方法を確立した。白色Ｘ線から高い分解能で単色Ｘ線を得るには、第２結晶からの反射ビームが第１結晶への入射ビームの方向から遠ざかる方向に向かうように配置する（+,+）セッティングが用いられ、各々独立した２枚の結晶を用いることが一般的であったが、結晶の相対的角度の安定性に問題があり、高精度の測定には限界があった。Rahman 氏は単一結晶ブロックに反射面を設け結晶固有に存在する格子面の組み合わせを見いだし長期的にも極めて安定性高くＸ線ビームを単色平行化出来ることを見いだし、複雑な形状の結晶モノクロメーターをデザイン、製作しその性能を確かめた。幾何学的配置により決まる格子面間角以外に、屈折率による補正、結晶内でおきる同時反射、光源位置の変動、結晶の温度変化などの測定精度に影響する因子について、考察と補正を行っている。特に、同時反射の影響については、３波近似の動力学的回折理論を用いて詳細な検討を行っている。

このモノクロメーターおよび計測システムの性能を確認するために、ボロンをドープしたシリコン単結晶および砒化ガリウム単結晶の格子定数測定を行い、所期の精度で格子定数を決定できることを示した。これらの測定結果にもとづき、この方法の今後の発展の可能性についても考察を行っている。

以上、本研究では、単一結晶ブロック内の複数の格子面において逐次反射をおこし、単色・平行Ｘ線を、これを用いて単結晶の格子定数を 10^{-7} から 10^{-8} の精度で測定できる計測法を確立し、シリコンや砒化ガリウム単結晶の測定例を示し、測定システムの有用性を示し、将来の様々な応用の可能性を示した。

本論文において開発された手法・測定システムは、学術上優れているものと認められる。従って、以上の研究は数物科学研究科物質構造科学専門課程の博士学位論文としてふさわしい内容をもつものであると総合的に判断した。