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学 位 論 文 題 目	Dielectric Anisotropy of Cobalt Crystals near K-Absorption Edge Measured by Using an Energy-Tunable X-Ray Polarimeter with a Phase Retarder
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論文内容の要旨

A new-type x-ray energy-tunable polarimeter based on Siddons-Hart-Amemiya's x-ray polarimeter, into which Hirano-Ishikawa-Kikuta's diamond phase retarder was introduced, has been newly developed. The new x-ray polarimeter consists of polarizer, analyzer and phase retarder crystals. The polarizer and analyzer are similar channel-cut silicon crystals giving four consecutive 422 reflections with Hart-Rodringues' offset mechanism, owing to which a high extinction radio over 10^8 is achieved in a photon-energy range of about 350 eV in the vicinity of the cobalt *K*-absorption edge(7709 eV). The analyzer was rotated around the beam axis in a range of ±2 degree from the crossed nicol position to analyze the polarization state of x-rays(elliipticity and rotation of polarization). The third optical device, Hirano-Ishikawa-Kikuta's phase retarder is a diamond crystal giving 111 asymmetric Laue reflection whose plane of incidence is incidence is inclined by 45 degree from the horizontal plane. The phase retarder produces elliptically polarized x-rays from linear polarization in terms of diffractive birefringence. Energy scan was performed by changing Bragg-reflection angles of the polarizer, analyzer and phase retarder crystals simultaneously.

Using this x-ray polarimeter, x-ray linear birefringence and linear dichroism in an *hcp* cobalt crystal has been detected and quantitatively measured simultaneously for the first time near the cobalt *K*-absorption edge. The *hcp* cobalt single crystal sample (1120 foil 12 μm thick; c-axis lying on the foil surface)was placed between the phase retarder and the analyzer with it's optic axis(c-axis) being inclined by 45 degree from the horizontal plane. An anisotropy of refraction index of *hcp* cobalt, $\Delta n(=n_c-n_a$, where n_c and n_a are refractive indices for polarizations in the directions of c- and a-axes)was obtained from ellipticity spectrum of polarization, while an anisotropy of absorption, $\Delta\mu(=\mu_c-\mu_a$, where μ_c and μ_a are absorption coefficients for polarizations in the directions of c- and a-axes)was obtained from rotation spectrum of polarition. Not only the magnitude but also the sign of Δn was obtained from an increase or decrease of ellipticity of elliptical polarization incident on the sample crystal.

X-ray anisotropic absorption in uniaxial crystal has been observed by Templeton and Templeton for the first time using a technique nowadays known as the polarized XAFS. They detected absorption anisotropy of uniaxial crystals with their optis axis set parallel and perpendicular to the polarization of horizontally polarized synchrotron x-radiation. Although this was a extremely pioneering work and this method is simple and convenient, it has the following two week points; first, anisotropy of refractive index of sample cannot be directly measured while only absorption anisotropy is obtained, and seconly, difference of absorption coefficient $\Delta\mu$ cannot be precisely discussed because the identity of crystal thickness is not exactly guaranteed when the optic axis being set parallel and perpendicular to the polarization plane of the incident x-rays. In the case of the present work, the thickness of sample *hcp* cobalt crystal was $12\mu m \pm 1\mu m$, that is, the inhomogeneity of the sample thickness was up to ten percent. In spite of that, we could precisely obtain Δn and $\Delta\mu$ from spectra of ellipticity and rotation of polarization, respectively, due to a feature of the polarimeter as a *polarization interferometer*, which is a concept the present author proposed in 1993.

In x-ray *polarization interferometry*, Δnt and $\Delta\mu t$, where t is the sample thickness, are directly measured as difference in phase and in amplitude between c-axis and a-aixs polarizations interfering with each other being reflected by the analyzer, instead of only $\mu_c t$ and $\mu_a t$ being measured seperately as in the polarized XAFS.

The Bonse-Hart x-ray interferometer has been regarded as a unique way to obtain phase information of wave in the x-ray frequency region for 30years since 1965. It's typical phase detection limit is about $2\pi/100$. Magnitude of phase difference due to the linear birefringence of cobalt crystal measured in the present work did not much exceed $2\pi/100$ at it's extremum. Therefore, it can be concluded that the birefringence of cobalt crystal could not be detected by using the Bonse-Hart interferometer. Phase detection limit of hte x-ray *polarization interferometer* (the x-ray polarimeter)has been estimated to be $2\pi/10,000$, a hundred times as small as that of the Bonse-Hart interferometer. This value is an estimation in the present stage using a normal bending magnet beam line BL4A of the Photon Factory. In the main body of the present thesis, it is shown that an increase and decrease in ellipticity spectra corresponding to phase difference of $2\pi/1,000$ has been detected as an ezidently significant signal. Since the wavelength of x-rays near the cobalt K-absorption edge is about 1.6Å, this signal corresponds to a wave-front shift of 0.0016Å between c-axis and a-axis polarizations. Such a small wave-front shift of electromagneti cwave has not been measured in any field of metrology.

The Kramers-Kronig relation between Δn and $\Delta\mu$ has been clearly confirmed in the absolute scale. This is the first experimental confimation of the Kramers-Kronig relation between linear birefringence and linear dichroism in the X-ray frequency region. On the other hand, this established theory guarantees the precision of the present experiment.

The original motive of the present experiment was an unexpected peak found in a spectrum of ellipticity obtained by using an *hcp* cobalt polycrystalline sample. It has been confirmed, with incidence of elliptically polarized x-rays with plus and minushelicity, that the peak of 'ellipticity' for an *hcp* cobalt polycrystallie sample was not real ellipticity but a partially random state of polarization.

Furthermore, a-, b- and c-plate crystals were prepared cut from an identical cobalt compound crystal. This crystal belongs to the monoclinic crystal system and therefore has a biaxial asymmetry. Polarization analyses were performed for the a-, b- and c-plate samples in the same way as was done for the *hcp* cobalt single crystal. Completely different spectra of ellipticity and rotation of polarization have been obtained for a-, b- and c-plates. This is the first observation of triple refraction and triple absorption in the x-ray frequency region. The Kramers-Kronig relation for each pair of spectra of ellipticity and rotation of polarization was confirmed in the absolute scale for the a-, b- and c-plate crystals respectively.

論文の審査結果の要旨

沖津康平君の博士論文の内容の要約は以下の通りである。

1）X線偏光子とX線検光子からなるエネルギー可変型X線ポラリメータにダイヤモンド結晶からなる移相子を導入し、X線の偏光状態を符号を含めて完全に解析することのできる、エネルギー可変型X線偏光解析装置を開発した。この装置は、物質中を伝搬するX線の垂直偏光と水平偏光の位相差を1周期の1万分の1の精度で検出できる性能を有する。従来のX線干渉計の100倍の精度に相当する。

2）新しく開発したエネルギー可変型X線偏光解析装置を用いて、X線領域の電磁波に対して物質が示す直線複屈折と直線二色性を、同時、かつ、定量的に測定することに世界で初めて成功した。試料としてはコバルト単結晶(六方晶)を用い、K吸収端近傍での直線複屈折スペクトルと直線二色性スペクトルの測定を行った。

3）上記の直線複屈折と直線二色性の間にはクラマース・クロニッヒの関係が定量的に成り立つことを明らかにした。X線領域ではクラマース・クロニッヒの関係が成り立っていることを実験的に確認したのは、これが世界で初めてである。

4）さらに、2軸異方性を持つ単斜晶系のコバルト錯体が三重複屈折と三色性を示すことを予測し、実際に、それがK吸収端近傍で観察されることを示した。また、これらの三重複屈折と三色性の間にもクラマース・クロニッヒの関係が定量的に成り立つことを示した。

以上の研究を遂行する上で、沖津君は動力学回折理論に基づく移相子の特性評価、回転角度の定量計算を行った。また、シリコン単結晶で作られているX線偏光子とX線検光子における同時反射の問題を回避するために、その羅針盤ともいえる2次元マップ(グリッチマップ)の作成を行った。さらに、偏光解析を精密に行うためのソフトウエアーの製作も併せて行った。測定スペクトルから得られたコバルト試料の構造異方性についても考察を加えている。また、従来の偏光ＥＸＡＦＳに対して本研究で開発した手法が優れている点について定量的に議論している。また、偏光干渉計をいう概念でこの測定法を考察し、従来のX線干渉計との対比も行っている。

沖津君の研究は、放射光の特長である、大強度・連続波長・指向性・偏光性の特性を有効に利用した新しい測定手法の開発であり、放射光科学における未開の領域の開拓であるので、その意義は極めて大きい。

上記のように沖津君の研究は数物科学研究科放射光科学専攻 の博士学位論文としての内容に十分に値し、専門的にも総合的にも優秀な研究業績であると判断した。