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学位論文題目	Study of nuclear resonant Bragg scattering from synthetic 2.2% and 95% ^{57}Fe hematite single crystals
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Abstract

As well known, there are two popularly used theories for dealing with the x-ray scattering of crystals, the dynamical theory and the kinematical theory. The main frames of these two theories were developed in 1920's and 1930's. Generally, they are applicable to two extreme scattering processes, scattered by an ideal perfect crystal and by an ideal imperfect crystal, respectively. As the actual crystals are neither ideal perfect nor ideal imperfect, the theoretical estimation has to be modified when analyzing the diffraction from real crystals. Since 1970's, some theoretical tendencies have been done to bridge the kinematical and dynamical theory. Trials of developing more general x-ray diffraction theory have been extensively done, but not so successful yet until now. The experiment to verify these developing theories has met serious problems in preparing proper samples, as it seemed almost impossible to introduce defects in a crystal without inducing influence to the perfection of the crystal lattice.

Activities in the study of nuclear-resonant Bragg scattering(NBS) of synchrotron radiation(SR) have strongly increased since the theoretical prediction by Ruby in 1974 and the first experimental success in 1980. Similar to the concept of the x-ray dynamical diffraction theory, the multiple scattering process is introduced in the dynamical theory of NBS. If resonant nuclei in a crystal are highly concentrated, the NBS process has a close analogy to the dynamical procedure in ordinary atomic scattering of x-ray in a perfect single crystal. As NBS comes from the resonant nuclei, the behavior of NBS depends on the concentration and distribution of isotopes in a crystal. In an extreme case, the multiple scattering probability should be negligible when the resonant nuclei are so rare. It seems possible to adjust the NBS behavior from a dynamical scattering to a kinematical scattering by altering the isotopical abundance in the case of a perfect arranged lattice. It is thought to be basically different with the conventional method to adjust a diffraction process from a dynamical process to a kinematical process, e.g., by making crystal slices thinner or choosing a weak diffraction.

In order to verify the this idea, study of NBS from perfect single crystal with various isotopic abundance are essentially important. In this research, 2 kinds of hematite nearly perfect single crystals with resonant nuclei abundance of 95% and 2.2%-^{57}Fe were studied. During the experiment, both pure NBS and NBS accompanied by electronic scattering were concerned.

Crystal perfectness was characterized by x-ray topography and rocking curve measurement by using the laboratory x-ray generator. Nearly perfect sample

regions were selected from the measurement. The effective sample areas are about 1mmx4mm(for 95%-^{57}Fe sample) and 1mmx10mm (for natural abundant sample).

Wavelength dependent integrated intensities of hematite single crystals were studied by using the electronic scattering with the incident photon energy of 0.86A, 0.71A and 0.36A. It was found that the samples were nearly perfect. All of the electronic scattering from 2 samples could be explained in the frame of the dynamical diffraction theory.

Behaviors of nuclear-resonant Bragg scattering (NBS) from a natural abundant (2.2%-^{57}Fe) and an isotopically enriched (95%-^{57}Fe) α-Fe_2O_3 single crystals have been studied by adopting a time-resolved method using synchrotron radiation at the beam line AR-NE3, National Laboratory for High Energy Physics (KEK). The beam conditioner in the experiment included a water cooling double-crystal pre-monochromator with Si 1,1,1 symmetric reflections in the (+,-) setting upstream in the beam line, a narrow bandpass high resolution monochromator composed of two channel-cut silicon blocks with asymmetric 4,2,2 reflections and symmetric 12,2,2 reflections arranged in the (+,+) setting. Monochromatic x-ray after this high resolution monochromator has an energy bandwidth of about 6meV. It is a significant factor in measurement to obtain the nuclear resonant delay signal when the nuclear resonant scattering was accompanied by a strong electron scattering. The high resolution monochromator effectively suppressed the electronic prompt to an extent that the prompt had no significant influence to the delayed signal.

The time spectrum of a 7,7,7 pure NBS diffraction of an isotopically enriched sample were measured when the angles of an incident beam were set at the exact Bragg angle and the positions deviating from the Bragg condition. All of the time decay profiles were found to be speeded up compared to the decay of isolated nuclei, furthermore the decay were slowed down when the deviation from the Bragg condition became larger.

The time spectrum of a 7,7,7 pure NBS diffraction of a natural abundant hematite sample was first time measured when the incident angle was set at the exact Bragg angle. A time decay profile with the life time of about 100 nsec was obtained experimentally. Compared with the time spectrum from the isotopically enriched sample, a drastic change of the time decay has been found when the isotopic abundance became rare.

It is thus proved that the collective radiation phenomenon of nuclei from a crystal can be studied not only by changing the Bragg condition but also by adjusting the isotopical abundance.

The angular dependent integrated intensity from h,h,h reflections both of odd orders which are pure nuclear-resonant scattering, and of even orders which are accompanied by predominant electronic scattering were measured in the experiment. On the contrary to the reflections from the isotopically enriched specimen in which all of the nuclear resonant scattering behaved as dynamical scattering, the 7,7,7 diffraction of the natural abundant single crystal was proved as a kinematical scattering. In order to explain the integrated intensity of the even order reflections (h=6,8) of this sample, accompanied electronic scattering has to been taken into account.

It has been found that the isotopic abundance is a controllable parameter to regulate the pure NBS phenomenon in a perfect single crystal from a dynamical scattering to a kinematical scattering. During this regulation, the collective radiation could be changed drastically. An unique experimental method to justify the statistical dynamical theory of diffraction has been first time proposed.

論文の審査結果の要旨

趙際勇君の博士論文の内容は（１）２種類の濃度の^{57}Ｆｅメスバウアー核をもつα-ヘマタイト単結晶をＸ線トポグラフィとロッキングカーブにより結晶学的評価を行ったことおよび（２）２種類の濃度の^{57}Ｆｅメスバウアー核をもつα-ヘマタイト単結晶中の回折波の挙動の研究を行ったことである。

趙際勇君の研究は完全結晶格子の中で散乱体の数が格子点の数よりも少ない場合の波動場の取扱いは理論・実験ともにまだなく回折結晶学的に示唆を与える重要な研究と評価される。ヘマタイト単結晶(α-Ｆｅ$_2$Ｏ$_3$)を研究対象に考えた場合、空孔子点などの格子欠陥がない場合、電子散乱の観点からは完全度の高い結晶といえる。さらにこの単結晶の構成要素であるＦｅに同位元素の概念を導入すると^{56}Ｆｅと^{57}Ｆｅの化学的差異はないので化学的にはα-Ｆｅ$_2$Ｏ$_3$結晶の完全性は保たれているといえる。一方原子核による核共鳴散乱を考えると、回折強度は共鳴核^{57}Ｆｅの濃度の関数になるであろう。このことを利用すると完全格子中に散乱体が確率的に存在した場合に波動場がどのようになるかを調べる有力な手段になる。趙際勇君は共鳴核^{57}Ｆｅの濃度により多重散乱あるいは一回散乱が成立すると考え、その範囲を見極めることを目的として研究を行った。試料は２．２％の天然濃度のα-Ｆｅ$_2$Ｏ$_3$単結晶と９５％にエンリッチされたものを用いた。

趙際勇君は実験的に（１）Ｘ線トポグラフィとロッキングカーブの手法による結晶の評価（２）２結晶分光器により高輝度放射光から取り出した１４．４ＫｅＶ単色Ｘ線をα-ヘマタイト単結晶に照射することによって得られるメスバウアー回折Ｘ線の強度の観察（３）９５％^{57}Ｆｅと２．２％^{57}Ｆｅを含むα-ヘマタイト単結晶に対し電子散乱振幅と核共鳴散乱振幅の両方をもつ６６６および８８８反射のパーセント反射率・反射幅および積分反射強度の測定（４）核共鳴散乱振幅のみをもつ７７７反射のパーセント反射率・反射幅および積分反射強度の測定（５）これらの回折強度の時間変化の観察（６）回折角度と回折強度の時間変化の関連の観察を行った。その結果、趙際勇君は６６６、７７７および８８８反射のすべてを観測することができた上で次のような事実を見い出した。（１）７７７反射強度に関しては９５％^{57}Ｆｅの時間衰退は早く、２．２％^{57}Ｆｅの時間衰退は単原子的に遅いこと（２）９５％^{57}Ｆｅの７７７反射強度の回折角度を中心回折角度からはずしていくと減衰は遅くなり単原子の現象に近くなること（３）^{57}Ｆｅを２．２％含むα-ヘマタイト単結晶の６６６反射強度は９５％含む７７７反射強度の$２×１０^{-2}$である（４）^{57}Ｆｅを２．２％含むα-ヘマタイト単結晶の７７７反射強度は９５％含む７７７反射強度の$６×１０^{-4}$である（５）^{57}Ｆｅを２．２％含むα-ヘマタイト単結晶の８８８反射強度は９５％含む７７７反射強度の$５×１０^{-2}$である。以上の結果趙際勇君は次のような結論を導いた。（１）２．２％^{57}Ｆｅの濃度のα-ヘマタイト単結晶からの核共鳴散乱強度の時間スペクトルを世界で初めて観測した。（２）９５％^{57}Ｆｅから２．２％^{57}Ｆｅへ向かうこと、ないし回折角度を変えるときの時間スペクトル構造の変化は協力現象の観点から説明でき、常識の範囲内の結果であった。（３）２種類の濃度での実験からは濃度５０％前後での回折波についての見通しをつけることは難しい。（４）９５％は動力学的散乱現象にのり、２．２％は運動学的散乱現象にのると考えて今のところ矛盾はない。（５）濃度５０％前後での回折波についての見通しつけることは難しい。したがって（６）

５０％前後の^{57}Ｆｅの濃度の試料を作製して同様の実験を行う。

以上の研究は実験装置の立ち上げ、結晶の評価、本研究の主要部分の実験、解析からなるが、いずれの段階においても新しい考えを導入し積極的に研究を推進し、新しい分野を切り開く能力を十分に示した。