氏名　趙　際　勇

学位（専攻分野）　博士（学術）
学位記番号　総研大甲第175号
学位授与の日付　平成8年3月21日
学位授与の要件　数物科学研究所　放射光科学専攻
学位規則第4条第1項該当
学位論文題目　Study of nuclear resonant Bragg scattering from
synthetic 2.2% and 95% $^{57}$Fe hematite single
crystals

論文審査委員　主査 教授 宮原恒啓
教授 安藤正海
教授 郭 須 奎一郎
教授 大隅一政
教授 松下正
教授 戸田惺志（東京大学）
助教授 高橋敏男（東京大学）
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 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%-57Fe were studied. During the experiment, both pure NBS and NBS accompanied by electronic scattering were concerned.

Crystal perfection 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%-57Fe 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%-57Fe) and an isotopically enriched (95%-57Fe) α-Fe2O3 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.
超際勇君の博士論文の内容は（1）2種類の濃度の$^{57}$Feメスバウアー応答をもつα-ヘマタイト単結晶をX線トポグラフィ図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。

超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微動場の取扱い理論・実験ともにまだなく回折結晶学的に示唆を与え研究に評価される。ヘマタイト単結晶（α-Fe$_2$O$_3$）を研究対象に考えた場合、空孔数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の濃度の関数であるであろう。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変えるかを調べる有力な手段になる。超際勇君は共鳴核$^{57}$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、その範囲を最確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。

超際勇君は実験的に（1）X線トポグラフィ図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、超際勇君は666、777および888反射のすべてを観察することができた。実験結果の観察は、（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ること（3）95%$^{57}$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^{-3}である。以上の結果超際勇君は次のような結果を得た。（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。超際勇君の実験からは濃度50%前後の回折波についての観測をしていくことは難しい。（3）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。（4）濃度50%前後の回折波についての観測しつけることは難しい。したがって（6）
50%前後の18F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。