

# Disputed discovery: The beginnings of X-ray diffraction in crystals in 1912 and its repercussions<sup>1</sup>

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**Abstract.** The discovery of X-ray diffraction is reviewed from the perspective of the contemporary knowledge in 1912 about the nature of X-rays. Laue's inspiration that led to the experiments by Friedrich and Knipping in Sommerfeld's institute was based on erroneous expectations. The ensuing discoveries of the Braggs clarified the phenomenon (although they, too, emerged from dubious assumptions about the nature of X-rays). The early misapprehensions had no impact on the Nobel Prizes to Laue in 1914 and the Braggs in 1915; but when the prizes were finally awarded after the war, the circumstances of 'Laue's discovery' gave rise to repercussions. Many years later, they resulted in a dispute about the 'myths of origins' of the community of crystallographers.

## 1. Introduction

The discovery of X-ray diffraction in crystals a hundred years ago, and the ensuing birth of the new specialities of X-ray spectroscopy and X-ray crystallography, have been praised and reviewed on numerous occasions, most extensively half a century ago in P. P. Ewald's *Fifty Years of X-ray Diffraction* (Ewald, 1962). The pioneers were awarded with the Nobel Prize in physics as early as 1914 and 1915: Max von Laue, who had suggested in spring 1912 the – now famous – experiments performed by Walter Friedrich and Paul Knipping, 'for his discovery of the diffraction of X-rays by crystals'; William Henry Bragg and William Lawrence Bragg, father and son, 'for their services in the analysis of crystal structure by means of X-rays' (Nobel Prizes, 1914–1915). Few other discoveries received such swift recognition and widespread praise.

The reconstruction of the events that led to Laue's idea in 1912 have provoked critical scrutiny. The science historian Paul Forman challenged the 'clan of X-ray crystallographers' with a 'critique of the myths' that he discerned in their accounts of the discovery. Ewald's *Fifty Years Festschrift*, dedicated to the International Union of

Crystallography, and other celebratory reviews of the events in 1912, according to Forman, served the purpose of maintaining a disciplinary identity among the crystallographers. "This circumstance, and its evident social function of reinforcing a separate identity, strongly suggests that the traditional account may be regarded as a 'myth of origins', comparable to those which in primitive societies recount the story of the original ancestor of a clan or tribe" (Forman, 1969, p. 68). In turn, Ewald regarded this interpretation as "the myth of the myths". Forman failed, in Ewald's view, "to appreciate the vagueness of the physical information confronting Laue before the experiment. To make his interpretation plausible, the author repeatedly aggrandizes statements taken from the literature, especially from Laue's Nobel Lecture and from the *Festschrift* for the semicentennial of the discovery. These unjustified accents, needed in support of his main thesis, show that his scheme is pre-conceived, artificial, and unnecessary" (Ewald, 1969, p. 81).

From these passages it is apparent that a review of the discovery of X-ray diffraction in crystals in a short article cannot cover the disputed issues in a comprehensive manner. The complexity is due to historical as well as scientific aspects. The latter are concerned with the nature of X-rays and the space-lattice hypothesis of crystals as known before the discovery; the former concerns the lack of documentary evidence – letters, diaries, manuscripts – from the crucial period in spring 1912. The Nobel speeches of the discoverers, Ewald's voluminous *Festschrift* for the semicentennial, Forman's critical scrutiny together with Ewald's refutation, and further accounts (see Wheaton, 1983, pp. 199–220) provide insights from a variety of different perspectives – and yet there remain questions that cannot be resolved unequivocally from the available archival sources. We restrict this study to Laue's initial idea about the nature of the interference effect. We will not discuss Laue's views concerning the space-lattice hypothesis, which have also been disputed in the course of the Forman–Ewald controversy (Gasman, 1975; on the early history of crystallography, see Kubbinga, 2012). Central to the following review is the knowledge on X-rays in Arnold Sommerfeld's Institute for Theoretical Physics at Munich University, the site of the discovery, where Laue spent the early period of his career as *Privatdozent* and where Walter Friedrich was Sommerfeld's assistant.

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## 2. The quest for the nature of X-rays

Since his call to Munich in 1906 to one of the very few chairs of Theoretical Physics, Sommerfeld had struggled hard to meet the high expectations of Röntgen concerning the theoretical explanation of X-rays. “Isn’t it a shame that ten years after Röntgen’s discovery one still does not know what is going on with X-rays”<sup>2</sup>, Sommerfeld alluded to this expectation shortly before his call to Munich in a letter to Wilhelm Wien, who had become Röntgen’s successor at Würzburg and pursued fundamental investigations about the nature of X-rays (Sommerfeld, 1905). On Wien’s X-ray investigations see Pohl (1912). Sommerfeld had made a mark in this quest with an interpretation of earlier experimental observations by Hermann Haga and Cornelis H. Wind about the passage of X-rays through narrowing slits. Haga and Wind (1899) interpreted a diffuse broadening at the narrower end of the slit as a diffraction phenomenon. Sommerfeld regarded X-rays as a shower of electromagnetic square pulses and estimated from the experiments of Haga and Wind (1899) that the order of magnitude of the width of an X-ray pulse is about one ångström (= 0.1 nm). But he did not perceive such pulses as a superposition of waves; nor was the experimentally observed broadening conclusive enough to justify the assumption of a diffraction effect. The conclusion that X-rays were indeed electromagnetic waves with a wavelength of about 1 Å, therefore, was far from persuasive (Wheaton, 1983, pp. 35–40).

By 1908, Charles Glover Barkla had provided new evidence about the nature of X-rays. He arrived at the conclusion that X-rays appear in two varieties. One sort of X-rays was independent of the material from which they emerged and could be polarized; the other was like fluorescence radiation and could not be polarized. The latter was entirely dependent on the irradiated material and designated as ‘fluorescence’ or ‘characteristic’ radiation. Only the former could be perceived in terms of the electromagnetic pulse hypothesis. Sommerfeld (1909) elaborated this hypothesis so that he could account for the spatial distribution of X-ray intensity due to the deceleration of electrons (*‘Bremsstrahlen’*) on their impact upon the anode material of an X-ray tube.

Henceforth, both varieties of X-rays had a name and a distinct set of properties: *Bremsstrahlung* consisted of showers of rectangular electromagnetic pulses that were radiated away from an X-ray anode like electromagnetic waves from a radio antenna (with the direction of the antenna parallel to the beam of the electrons between the cathode and the anode). The characteristic radiation, on the contrary, displayed no dependency on the direction; its ‘hardness’ (that is today photon energy) grew with the atomic weight; furthermore, it was homogeneous, *i.e.* each characteristic radiation had a specific penetrating power. If X-rays consisted of waves, the characteristic radiation would have a line spectrum, in contrast to the continuous spectrum of the *Bremsstrahlung*. However, there were serious doubts about whether X-rays are waves. William Henry

Bragg, in a response to the *Bremsstrahlen* theory, presented Sommerfeld with an alternative view which became known as the neutral-pair hypothesis (Stuewer, 1971; Wheaton, 1983, pp. 87–90). Instead of assuming an electromagnetic interaction, Bragg speculated that an electron, on encountering an atom, may neutralize its charge and “takes the form of the X-ray or the  $\gamma$ -ray as the case may be: it may again lose the neutralising complement and become a secondary cathode or  $\beta$ -ray, the double transformation being accompanied by no very great change of speed” (W. H. Bragg, 1910). In another letter he referred to photographs in the recently invented cloud-chamber: “Have you seen any of C. T. R. Wilson’s pictures of the fog formed instantly after the passage of ionizing rays through a gas?” (W. H. Bragg, 1911). It seemed impossible to interpret these images other than by assuming a particle nature of the ionizing radiation.

The quest about the nature of X-rays, therefore, cannot be isolated from the riddles about radioactivity in the early years of the twentieth century. In a sequel to his *Bremsstrahlung* paper, Sommerfeld attempted to explain  $\gamma$ -radiation in a similar way to the X-ray *Bremsstrahlung*: He perceived the emission of  $\beta$ -rays from a radioactive substance as the ejection of electrons that become accelerated within short distances to almost the speed of light. The electromagnetic radiation caused by this acceleration would be radiated away in a similar manner to the X-ray *Bremsstrahlung* caused by the deceleration of electrons. If an electron is accelerated to almost the speed of light within a very short distance, the intensity of the electromagnetic radiation would be distributed within narrow forward-directed lobes. Couldn’t this be the  $\gamma$ -radiation observed together with  $\beta$ -rays? This “very strange view about the structure of  $\gamma$ -rays”, Sommerfeld admitted, was “the ultimate consequence of my view about the structure of X-rays” (Sommerfeld, 1911a, p. 3).

If  $\gamma$ -radiation and X-ray *Bremsstrahlung* are electromagnetic radiation produced by the acceleration of electrons, then the energy of these radiations is related to the energy of the electron. In the case of X-ray *Bremsstrahlung*, therefore, there should be a relation between the kinetic energy of the cathode-ray electron that is brought to a halt at the anode of an X-ray tube and the energy radiated away during this process. In order to derive this relation, however, Sommerfeld had to make further assumptions about the braking process. At this point he employed Max Planck’s quantum constant  $h$ . Because  $h$  has the dimension of an action, Sommerfeld assumed that in each elementary process, such as the stopping of a cathode-ray electron by an atom or the emission of a  $\beta$ -ray electron, the energy  $E$  of the electron and the duration  $\tau$  of its stopping obey the quantum rule  $E\tau = h$ .

From this ‘ $h$ -hypothesis’ Sommerfeld derived an expression for the ratio of the energy contained in the *Bremsstrahlung* to that of the cathode ray. The latter could be estimated from the voltage of the X-ray tube, the former from the penetrating power of the X-rays. However, the comparison of theory and experiment was possible only if it was known how much of an electron’s energy on impact with the anode material went into the production of characteristic rays and *Bremsstrahlen*, respectively.

<sup>2</sup> „Es ist eigentlich eine Schmach, dass man 10 Jahre nach der Röntgen’schen Entdeckung immer noch nicht weiß, was in den Röntgenstr. eigentl. los ist.“

Hence it was necessary to discriminate experimentally the unpolarized (characteristic) from the polarized (*Bremsstrahlen*) radiation. The ‘*h*-hypothesis’ yielded an expression for the pulse width of the *Bremsstrahlen* as a function of the energy of the cathode-ray electrons, which could be compared with new experiments on the passage of X-rays through slits measured by Röntgen’s assistant Paul Koch (1912) using a new photometer.

These were the problems that Sommerfeld hoped to clarify in 1911. The prospect of an experimental test of the ‘*h*-hypothesis’ was the reason why Sommerfeld invited Walter Friedrich to join his institute. Friedrich, a student of Röntgen, had accomplished in 1911 his doctoral thesis on just such experimental questions: he had analysed the spatial distribution of X-rays emitted from a platinum anode and reported his work in the Munich colloquium (Physikalisches Mittwoch-Colloquium, 1911; Friedrich, 1912). Sommerfeld was by this time granted a second assistant position, and he was eager to employ Friedrich as his ‘experimental’ assistant (his ‘theoretical’ assistant at that time was Wilhelm Lenz) because he expected from polarization experiments further evidence for his ‘*h*-hypothesis’. With two assistants, a mechanic and a basement room equipped for experimental work, Sommerfeld had means and personnel at his disposal which seem unusual in retrospect for an institute dedicated to theoretical physics. His embrace of experimental research apparently also evoked some rivalry. “Röntgen wants Friedrich for himself”, Sommerfeld wrote to his wife shortly after he had offered Friedrich the position as his assistant. “R. is stupid enough to demand an immediate decision within two hours. Friedrich declines. I am very glad about it, not only because I need Friedrich but also because one does not like to loose in a showdown. It does no harm that R. will have the opposite feelings” (Sommerfeld, 1912a). However, the showdown did not result in a permanent feud. With Röntgen “everything is OK after it was unpleasant for a while”, Sommerfeld wrote in another letter. “He is really an excellent man” (Sommerfeld, 1912b).

In November 1911, Sommerfeld’s ‘*h*-hypothesis’ was also the subject of debates at the First Solvay Congress, dedicated to the theory of radiation and quanta. On this occasion, too, Sommerfeld revealed that he was waiting for experimental evidence. “Our theory of the quantum of action yields some strange consequences that are worth an experimental test”, he concluded after deriving a formula which related the pulse width of the polarized X-rays to the energy of the cathode rays. According to this formula, the penetrating power of the polarized X-rays should not depend on the material of the anode and be universally determined by the speed of the cathode rays on impact. A comparison of the polarized X-rays emitted from a light and heavy anode material, such as carbon and platinum, would provide a suitable test if the intensity of the polarized radiation were indeed the same. “The theory can only satisfy in its present form if these consequences are confirmed quantitatively. Experiments to this end are under preparation in my institute”<sup>3</sup> (Sommerfeld, 1911b, p. 266).

<sup>3</sup> „Unsere Theorie des Wirkungsquantums liefert einige merkwürdige Konsequenzen, die der experimentellen Prüfung wert sind. Nach

Friedrich’s experiments, however, were dragging on without conclusive evidence. When Hendrik Antoon Lorentz, who had participated in the Solvay Congress, asked Sommerfeld a few months later whether his expectations in the ‘*h*-hypothesis’ had become fulfilled, Sommerfeld responded with a verse by Goethe that it is like planting roses without knowing whether they will blossom. “The experiments with X-rays which are supposed to tell me something about the likelihood of the blossoming are not yet ready”<sup>4</sup> (Sommerfeld, 1912c).

### 3. Laue’s ‘flash of inspiration’

Such was Sommerfeld’s expectation concerning Friedrich’s experimental work when Max Laue, who had belonged to Sommerfeld’s group since 1909 as *Privatdozent*, proposed another experiment. Laue suggested firing X-rays at a crystal in order to obtain diffraction effects. The idea of scattering X-rays from a crystal was not new. Röntgen himself had already in his earliest investigations studied the scattering of X-rays by crystals. “I continued the experiments to which I referred already in my first communication about the transparency of plates of equal thickness that have been cut from a crystal along different directions”, Röntgen reported in his third communication on further observations about the properties of X-rays, published in 1897. “Again, no influence of direction on the transparency could be recognized” (quoted in Glasser, 1995, p. 324). Since then, crystals had often been the target of beams of X-rays – without the slightest indication of a diffraction effect. How could Laue, a theoretician, be so bold to suggest once more such an experiment?

There is no direct archival record – in the form of letters, diary or manuscript – from which Laue’s motivation would become clear. According to the standard narrative (Laue, 1915), Laue’s idea was provoked by a conversation with Paul Peter Ewald, then working on his doctoral dissertation on crystal optics. Ewald’s theory involved the calculation of the scattering of light by a regular three-dimensional arrangement of resonators. Portraying himself in the third person, Ewald gave the following account about a conversation with Laue that provoked the decisive idea: “Laue suggested that they meet the next day – it was probably late in January 1912 – in the Institute and discuss before and after supper at his home. They met as

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*Gleichung (16) müsste die Härte der polarisierten Röntgenstrahlen von dem Material der Antikathode unabhängig und universell bestimmt sein durch die Geschwindigkeit der auftreffenden Kathodenstrahlen. Dasselbe gilt nach (13) von der Energie der polarisierten Röntgenstrahlen. Z. B. ist bei Kohle die Gesamtemission ziemlich schwach, die Polarisation verhältnismäßig groß, bei Platin die Gesamtemission stark, die Polarisation relativ schwach. Nach dem qualitativen Anschein ist es wohl möglich, dass die polarisierte Intensität bei beiden Antikathoden ihrem absoluten Betrage nach gleich ist. Die Theorie kann nur dann in ihrer vorliegenden Form befriedigen, wenn sich diese Konsequenzen auch quantitativ bestätigen. Dahingehende Versuche werden in meinem Institut vorbereitet.“*

<sup>4</sup> „Da hilft nun weiter kein Bemühn, Sinds Rosen nun sie werden blühn.“ Die Versuche mit Röntgenstrahlen, die mich über die Wahrscheinlichkeit des Blühens näher unterrichten sollten sind noch nicht fertig.“

arranged and took a detour through the Englische Garten, a park whose entrance was not far from the University. After having crossed the traffic on the Ludwigsstraße, Ewald began telling Laue of the general problem he had been working on, because, to his astonishment, Laue had no knowledge of the problem. He explained how, in contrast to the usual theory of dispersion he assumed the resonators to be situated in a lattice array. Laue asked for the reason of this assumption. Ewald answered that crystals were thought to have such internal regularity. This seemed new to Laue. Meanwhile they were entering the park, when Laue asked: ‘what is the distance between the resonators?’ To this Ewald answered that it was very small compared to the wave-length of visible light, perhaps 1/500 or 1/1000 of the wave-length, but that an exact value could not be given because of the unknown nature of the ‘*molécules intégrantes*’ or ‘particles’ of the structure theory; that, however, the exact distance was immaterial for his problem because it was sufficient to know that it was only a minute fraction of the wave-length” (Ewald, 1962, pp. 40–41).

In his Nobel speech, Laue claimed that as soon as he was aware of the distances of the atoms in a crystal, “my intuition for optics suddenly gave me the answer: lattice spectra would have to ensue.” He asked Friedrich to undertake such an experiment, but “the acknowledged masters of our science, to whom I had the opportunity of submitting it, entertained certain doubts about this viewpoint. A certain amount of diplomacy was necessary before Friedrich and Knipping were finally permitted to carry out the experiment according to my plan, using very simple equipment at the outset. Copper sulphate served as the crystal, since large and regular pieces of it can easily be obtained” (Laue, 1915).

It is undisputed that the idea arose in discussions about Ewald’s doctoral work. According to Sommerfeld’s report to the faculty in February 1912, the aim of Ewald’s dissertation was to calculate “the dispersion and double-refraction in an ideal rhombic [*sic*] electron lattice” (Sommerfeld, 1912d). The circumstance that prompted Laue to ask for the distance of the resonators in a crystal, therefore, seems very plausible. But there is only retrospective recollection about Laue’s conversation with Ewald and its immediate consequences – and these recollections gave rise to controversy. Sommerfeld recalled that Laue’s ‘thought of discovery’ emerged in a discussion between Ewald, Laue and himself (Sommerfeld, 1924).<sup>5</sup> According to Laue’s Nobel speech, the decisive moment happened “one evening in February 1912” when “P. P. Ewald came to visit me”. He did not mention that Sommerfeld also was present. Nor did he explain why Sommerfeld was reluctant to charge Friedrich with the execution of his idea. When he learned about Sommerfeld’s version of the three-man conversation, he asked Ewald to interfere because he did not want different views about the origins of his discovery to be circulated. Ewald forwarded Laue’s complaint to Sommerfeld and added that his [Ewald’s] “poor historical sense” disqualified him as a witness. Ewald left

Munich after his doctoral examination in order to become David Hilbert’s assistant, so he could only report from hearsay what happened after his departure. “I think that the idea of interferences occurred to Laue for the first time during a discussion about my doctoral work between him and myself in his flat”, he vaguely agreed with Laue. But in the same breath Ewald scoffed at Laue’s insistence on the ‘flash of inspiration’. Laue could be “at least so proud for some of his [Laue’s] other work” (Ewald, 1924).

#### 4. A discovery based on misapprehensions

Beyond the different views about the exact circumstances of Laue’s ‘flash of inspiration’, there are obvious reasons why Sommerfeld was reluctant about the immediate execution of Laue’s idea. First, Sommerfeld had other plans for the experiments in the basement of his institute. He wished that Friedrich would investigate the *Bremsstrahlung* emitted by different X-ray anodes from which he expected the confirmation of his ‘*h*-hypothesis’, as he had written to Lorentz by the end of February 1912 (Sommerfeld, 1912c). Second, Sommerfeld had good reasons to deny Laue the interruption of his own experimental plans. Laue did not perceive the crystal as a diffraction grating for the primary beam of X-rays, but expected that the primary X-rays excite the atoms of the crystal to emit characteristic radiation. He imagined that it is the homogeneous X-rays of this characteristic radiation which produce a diffraction pattern due to the regular spatial arrangement of the centers from which it is emitted. “Because we thought at first”, the Munich discoverers wrote in their first publication about the beginnings of their experiments, “that we had to deal with a fluorescence radiation, a crystal had to be chosen that contained a metal of a considerable atomic weight in order to obtain an intensive and at the same time homogeneous secondary radiation that seemed most appropriate for the experiments. According to Barkla metals with an atomic weight of 50–100 came into consideration. Since initially we had no good crystal containing such metals, we used for the preliminary trials a fairly well-developed copper sulfate crystal” (Friedrich *et al.*, 1912, p. 314).

It must have been for this reason that a “certain amount of diplomacy was necessary before Friedrich and Knipping were finally permitted to carry out the experiment”, as Laue hinted to Sommerfeld’s reluctance in his Nobel speech. Laue could not expect that the primary beam, perceived as a shower of pulses that contained a mixture of different wavelengths, was able to produce an interference pattern. Monochromatic waves were only expected from the characteristic radiation. But how should these fluorescent X-rays that were emitted by the crystal atoms without phase relation produce an interference pattern? Laue must have asked himself this question. If he didn’t, Sommerfeld surely will have asked the same question – and Laue would not have been able to offer a satisfying answer. “Far from being a mere extension of an optical experiment from a two-dimensional to a three-dimensional transmission grating, this was an experiment without analogy or precedent”, Forman criticized this part of the myth of the

<sup>5</sup> „Bei einer Besprechung zwischen ihm [Ewald], von Laue und dem Ref. [Sommerfeld] zündete bei Laue der Entdeckergedanke . . .“

discovery of X-ray diffraction. “No wonder Sommerfeld refused machine time” (Forman, 1969, p. 64).

As Forman also noted in his critique, it is not clear how the very first experiments were actually performed. The publication of the discovery did not reveal how the photographic plates were arranged initially, nor why Knipping became involved in the experiment. In his Nobel speech Laue claimed: “Immediately from the outset the photographic plate located behind the crystal betrayed the presence of a considerable number of deflected rays, together with a trace of the primary ray coming directly from the anticathode. These were the lattice spectra which had been anticipated” (Laue, 1915; for the arrangement of the photographic plates as presented in the first publication, see Fig. 17 in Kubbinga, 2012). Friedrich, however, admitted in a recollection ten years after the discovery that they set up the plates at first parallel to the primary beam so that the expected interference pattern from the crystal’s characteristic radiation could be recorded without the primary beam, and that these plates displayed “only little characteristic blackening phenomena” (Friedrich, 1922, p. 366). Ewald even stated that the plate was at first placed “between the X-ray tube and the crystal on the assumption that the crystal would act like a reflexion grating”. When the expected result was not found, “Friedrich and Knipping came to the conclusion that better success might be achieved by placing the plate behind the crystal, as for a transmission grating. Knipping insisted on placing plates all around the crystal” (Ewald, 1962, p. 44). Abram F. Ioffe, a Russian physicist who used to collaborate with Röntgen at that time, also reported that the photographic plates were initially placed so that they would not be exposed to the primary beam. He described the discovery as a result of frustration: “And day after day the X-ray tube was tremendously sizzling, but the plate was not blackened. Knipping, a young physicist working in the same room, had to leave the laboratory within the next two or three weeks, but the uninterrupted noise of the X-ray tube disturbed his experiments. In order to record at least something on the photographic plate, he placed it so that it became exposed by the X-rays – and there was the great discovery” (Ioffe, 1962).

Many physicists were puzzled by the Munich discovery and the initial explanation as an interference of the crystal’s own characteristic radiation. Peter Debye, who had served as Sommerfeld’s assistant until 1911, remarked that “one should generally not trade merit against luck with such things” (Debye, 1912). First of all he congratulated Sommerfeld. “If you had not had such a longstanding interest in X-rays, if you had not offered the means of your institute in the most liberal manner and provided free insight in your thoughts to everyone at any time, it would not have occurred to Laue and, in particular, he would not have found the practically trained collaborators who were indispensable for the success” (Debye, 1912). Debye subsequently began to study how the diffraction spots were affected by the vibration of the atoms in the crystal lattice (Debye, 1913). Laue’s view about thermal motion of the lattice atoms, as briefly mentioned in the first publication of the discovery, must only have added to the puzzles about its causes (Friedrich *et al.*, 1912, p. 309).

Elsewhere the Munich discovery also puzzled the physicists. “The men who did the work entirely failed to understand what it meant, and give an explanation which was obviously wrong”, wrote Henry Moseley to his mother when he was beginning his own research on X-ray diffraction in autumn 1912 (quoted in Heilbron, 1974, pp. 194–195). Still more than half a year after the discovery Laue considered unresolved the question of where the monochromatic X-rays observed in the sharp diffraction spots come from (Laue, 1912, p. 244). When the Braggs learned about the Munich discovery, they too regarded Laue’s explanation as wrong. But their investigation of this phenomenon also started with a misapprehension. They were biased by W. H. Bragg’s long-held conviction about a particulate nature of X-rays. From the modern perspective of wave–particle dualism, we are used to regarding X-rays both as electromagnetic waves and particles, but by 1912 Bragg considered the phenomena in which X-rays displayed their particle nature as evidence against the wave nature. When Bragg father and son began their own experiments they expected at first that the primary beam of X-rays emitted ‘secondary pencils’ of X-ray particles that could only traverse the crystal through alleys of the regularly arranged crystal atoms (W. H. Bragg, 1912).

However, the Braggs soon became aware that the phenomenon is indeed a diffraction phenomenon of the primary X-rays. While W. H. Bragg hoped for an explanation that would finally account both for the wave and particle aspect of X-ray phenomena (Stuewer, 1971; Wheaton, 1983, p. 208), his son began to adopt the idea of a wave interference – even though entirely different from Laue. On 11 November 1912, Bragg junior reported at Cambridge about the Munich experiment. In this talk, published later in the *Proceedings of the Cambridge Philosophical Society*, he interpreted the outcome of the experiment as an interference of X-rays that are reflected on crystal planes – and introduced what became known as ‘Bragg’s law’ (W. L. Bragg, 1913). Many years later, he recalled how careful he was to present it so that his father would not feel offended. He titled the paper *The Diffraction of Short Electromagnetic Waves by a Crystal* because he was “unwilling to relinquish my father’s view that the X-rays were particles; I thought they might possibly be particles accompanied by waves” (quoted in Ewald, 1962, p. 62).

After this talk at Cambridge and on the advice of C. T. R. Wilson, Bragg junior confirmed his view by an experiment with mica. On 8 December 1912 he sent a short report about this experiment to *Nature*. “A narrow pencil of X-rays, obtained by means of a series of steps, was allowed to fall at an angle of incidence of 80° on a slip of mica about one millimetre thick mounted on thin aluminium”, he described the experimental arrangement. “A photographic plate set behind the mica slip showed, when developed, a well-marked reflected spot, as well as one formed by the incident rays traversing the mica and aluminium.” By changing the angle of incidence he confirmed the law of reflection. Furthermore, the experiment had the virtue of being simple. “Only a few minutes’ exposure to a small X-ray bulb sufficed to show the effect, whereas Friedrich and Knipping found it necessary to give an exposure of many hours to the plate”, he compared his ex-

periment with that of the Munich group (W. L. Bragg, 1912). Thus, by the end of the year, the younger Bragg had dismissed both Laue's initial explanation of the observed spots in terms of the crystal's characteristic radiation as well as his father's particle interpretation.

At first it seemed as if the Braggs were engaged in a harmonic family collaboration. In a joint paper they described how a crystal could be used as an X-ray spectrometer (W. H. Bragg, W. L. Bragg, 1913a). Rather soon, however, they went along separate paths. Bragg senior made X-ray spectrometry his favorite research topic – an investigation which Henry Moseley and Charles Darwin were pursuing at the same time at Rutherford's laboratory in Manchester. When Rutherford asked the elder Bragg to delay his publication on this subject so that his young men had a chance to proceed along the same line of research, Bragg gave in but “always felt it was not quite reasonable” (quoted in Jenkin, 2001, p. 383). In another joint work in summer 1913, Bragg father and son investigated the structure of diamond (W. H. Bragg, W. L. Bragg, 1913b, c). By then, the field was considered so fertile that the Second Solvay Congress, held in Brussels in October 1913, was dedicated to ‘The Structure of Matter’. However, only the elder Bragg was invited to present their work. Henceforth, the son struggled to emerge from the shadow of his father. Although they kept collaborating for a while at Leeds, where Bragg senior had built the first X-ray spectrometers, Willy, as Bragg junior used to be called in distinction of his father's first name William, began to stress his independence by designating himself ‘W. Lawrence Bragg’. The rivalry between father and son also became apparent when both co-authored a book on their early work, with the preface written by the father alone (W. H. Bragg, W. L. Bragg, 1915; Jenkin, 2001).

The three-man collaboration at Munich was even more ephemeral. They used to review their discovery in separate accounts. In September 1913, when both Friedrich and Laue presented papers on X-ray diffraction at a conference in Vienna, their discord surfaced in a discussion concerning the initial misapprehensions. By this time it was clear that the wavelengths observed in the interference pattern were selected by the crystal from the continuous spectrum of the primary beam (Friedrich, 1913). Laue admitted that he had never clearly expressed his thoughts “how the monochromacy of the diffracted rays comes about”, but he denied what Friedrich had ascribed to him as his view at one time or another, namely “that the spectrum of the primary X-rays is monochromatic”. This thought had “never” come to his mind, Laue claimed, and he chided Friedrich for “some-what denigrating me” (Laue, 1913). But it was still not entirely clear how Laue's and Bragg's theories fit together.

The clarification of how both approaches accounted for the experimental results was not achieved in a single stroke but in a process to which several authors contributed throughout the year 1913. Among them, Ewald deserves to be singled out as the prime architect of the modern theory of X-ray diffraction in crystals (Cruickshank *et al.*, 1992). In a paper published on 1 June 1913 in the *Physikalische Zeitschrift* Ewald showed that Bragg's reflection and Laue's diffraction approaches may be perceived in terms of a construction in a ‘reciprocal space’,

now known as the ‘Ewald sphere’ (Ewald, 1913; see also Authier, 2012). Furthermore, Ewald elaborated the details for Sommerfeld's presentation at the Second Solvay Congress in October 1913 in Brussels. On this occasion, Sommerfeld reconciled Bragg's and Laue's approach for the special case of the zinc blende structure. “*Je me base pour cela essentiellement sur les brillants travaux expérimentaux de M. W. H. Bragg et les magnifiques recherches théoriques de son fils W. L. Bragg, auxquelles nous devons entre autres la connaissance de la structure de la blende*”, Sommerfeld acknowledged the achievements of Bragg father and son (Sommerfeld 1913, p. 125). In January 1914, Ewald published further evidence that the structure models derived by the method of the Braggs are equivalent with Laue's theory: “The correctness of Braggs' models for Zinkblende and diamond is fully confirmed by the distribution of intensity among the spots” (Ewald, 1914).

Within less than two years, ‘Laue's discovery’ was cleared from its initial misapprehensions and brought in line with the ensuing discoveries of the Braggs. Both the interpretation of the phenomenon and its uses for X-ray spectroscopy and the analysis of crystal structures had been ascertained. By the beginning of 1914, the diffraction of X-rays in crystals was ripe for Nobel prizes.

## 5. The Nobel awards

What may appear in retrospect as a straightforward awarding – few scientific discoveries promised such beneficial applications – turned into a rather complicated process. The procedure of selecting a Nobel laureate involves several stages, beginning with the nomination of candidates by a select group of nominators, expected before February each year. The nominations are solicited from Swedish and foreign members of the Royal Academy of Sciences, previous winners of the Nobel prize, professors of physics at Scandinavian universities, and individual scientists who are invited to present proposals on the basis of *ad hoc* deliberations (Crawford, 2002). Based on the nominations received, the Nobel committee would select a few candidates for closer scrutiny and ponder their merits in special reports. By summer, the Nobel committee would issue its recommendation to the Swedish Academy of Science in the form of a report. The procedure would be completed by the election of the laureates in November and the final award ceremony in December.

In 1914, however, the Great War interrupted this routine procedure. When the first nomination for the discovery of X-ray diffraction in crystals reached the Nobel committee on physics in January 1914, the course of events seemed to proceed as usual. Adolf von Baeyer, winner of the 1905 Nobel prize in chemistry, nominated Laue “for his work on X-rays” (von Baeyer, 1914). Apparently he considered the discovery so well known that he did not offer an explanatory statement. Another nomination for the same discovery came from Emil Warburg, the president of the Berlin Physikalisch-Technische Reichsanstalt, who was a little bit more explicit (Warburg, 1914). Warburg suggested to award Laue and W. H. Bragg together “for the

discovery of X-ray diffraction on crystals and for the application of this phenomenon for the investigation of crystal structures". Both Baeyer and Warburg belonged to a group of nominators whose recommendations were given considerable attention. Baeyer's nominations had already resulted in several Nobel prizes, three for chemistry: in 1904, 1906 and 1912 for William Ramsay, Henri Moissan and Victor Grignard; and one for physics in 1906 for Joseph John Thomson. Warburg's recommendations had contributed to five Nobel prizes in physics so far, in 1901, 1904, 1906, 1911 and 1913 with the awards for Wilhelm Conrad Röntgen, John William Strutt (Lord Rayleigh), Joseph John Thomson, Heike Kamerlingh Onnes and Willy Wien. Although Baeyer's and Warburg's nominations ranked high for the five members of the Nobel committee on physics, they were only two out of 23 for the physics prize of the year 1914; among the other recommendations were three for Lorand Eötvös, two for Albert Einstein, two for George Ellery Hale and two for Charles Fabry (Crawford, 2002).

With the recommendations on their desk, the Nobel committee could enter the stage of deliberation. By July 1914, they issued the special report with the recommendation that Laue should be awarded the Nobel prize (Gullstrand, 1914). However, before the final stage for the prize awards would take place, Sweden's neutrality in the war called for a postponement (Friedman, 2001, Chapter 5). Although the Academy endorsed the recommendation of the physics committee and elected Laue as the 1914 Nobel Prize winner, the decision was not made public. The postponement was scheduled until June 1915 and affected only the communication and the award of the 1914 prizes, not the prize decisions. Therefore, the Academy proceeded as usual and solicited nominations for the 1915 Nobel Prizes. Adolf von Baeyer, ignorant about the decision of preceding year, nominated again Laue "for his achievements in the field of physics" (von Baeyer, 1915). Henry Andrews Bumstead, the director of the Sloane Laboratory at the Yale University, too, regarded Laue as fit for the prize. "In my opinion, the most notable recent discovery in experimental physics is that of Laue upon the Diffraction of X-rays by Crystals", Bumstead (1915) argued, adding that if the prize should be awarded for this discovery one "might perhaps consider whether some share of it should not be awarded to W. H. and W. L. Bragg, for their services in the development and application of the method". Another nomination for Laue and the Braggs came from Stefan Meyer, head of the Vienna Radium Institute (Meyer, 1915). While these nominations ranked Laue first, Theodore William Richards, President of the Wolcott Gibbs Memorial Laboratory at Harvard University, nominated only the Braggs. Both "have risen into remarkable prominence on account of the highly interesting work which they have done upon the intimate structure of crystals as investigated with the help of the Röntgen ray", Richards justified his recommendation. "This work seems to me so important, so intelligently and sanely carried out, and so sound in its conclusions, that I now hereby venture to nominate these two men for the prize in Physics in 1915, proposing that each be awarded half of the prize in the manner more than once effected in the past" (Richards, 1914).

Although there were again competing nominations which deserved serious consideration (e.g. three for Hale and two for Max Planck), the committee had no qualms in focusing on the Braggs for the 1915 prize. After Laue had been elected in 1914 for the discovery of the effect, the Braggs should be rewarded for its application, the committee concluded its special report in June 1915 (Gullstrand, 1915). The Academy endorsed the recommendation and, without plans for a further postponement of the Nobel awards, voted in November for the Braggs as the physics Nobel laureates of 1915. At the same time, the decision for Laue in 1914 was confirmed. But the prospect of a Nobel ceremony with laureates from Germany and England shaking hands on neutral Swedish soil appeared unlikely, although not impossible (Crawford, 1992, Chapter 3). When Laue learned about the prize decisions in November 1915, he asked a member of the Nobel committee to forward his congratulations to the Braggs – and he was glad to learn *via* the same channel about the reciprocal response by the Braggs (Friedman, 2001, p. 91). However, such private feelings were short-lived and had no impact on the further course of events. The Nobel prize, and with it the Academy, was in the limelight of the national and international press. In order to avoid hostility from one or another side, the Nobel awards were postponed until after the war.

Even then it took years to overcome former resentments. When Laue finally received his prize at the first Nobel ceremony since 1913, held in June 1920 in Stockholm, the Braggs were absent (Friedman, 2001, p. 115; Crawford, 1992, p. 66). Bragg junior claimed to be prevented from attending by other circumstances. His father declined because, as he revealed to Rutherford, "I believe that several Germans are going" (quoted in Jenkin, 2001, p. 389). Indeed, five German Nobel laureates were traveling to Stockholm to receive their prizes. Planck and Johannes Stark had been awarded the Nobel prizes for physics for the years 1918 and 1919; Richard Willstätter and Fritz Haber were awarded the Nobel prizes for chemistry for 1915 and 1918 (Metzler, 1996). Bragg senior never presented a Nobel speech; his son fulfilled this ritual only two years later in September 1922 (W. L. Bragg, 1915).

## 6. Repercussions

Laue's Nobel speech brought the persistent discord about the Munich discovery once more to the surface. With his 'diplomacy' remark Laue alluded to Sommerfeld's reluctance to interrupt the scheduled experiments in his institute – but he did not mention that Sommerfeld had good reasons to regard Laue's plan as doomed to failure. In his critique of the myths about the discovery of X-ray diffraction, Forman quoted a most revealing passage from a letter in which Laue explained to Ewald in 1924 how he had persuaded Friedrich to undertake the experiment despite Sommerfeld's reluctance: "By March–April 1912 it seemed that Friedrich wanted to postpone the interference experiments. So I asked Dr. Knipping to take care of the matter. And then it went like in [Schiller's drama] Wallenstein: *Wenn es denn doch geschehen soll und muss, so mag*

*ich's diesem Pestaluz nicht gönnen.*' I would appreciate if you would show this letter occasionally to Sommerfeld"<sup>6</sup> (Laue, 1924).

By the time of this correspondence in 1924, Laue and Sommerfeld had already made peace. How deep their discord had been becomes apparent from earlier letters. "I am afraid my well-being would suffer a great deal from his presence",<sup>7</sup> Sommerfeld had written in 1916 on one occasion about Laue (Sommerfeld, 1916). The tension between Sommerfeld and Laue must have begun before the events in spring 1912, as Laue revealed in a letter to Sommerfeld in the wake of his Nobel speech: "Of course, you could justly point to some occasions when my behaviour was not always correct towards you, in particular shortly after my arrival in Munich [in 1909]. But you knew about my nervous problems. Had you allowed for 'mitigating circumstances', our personal relationship would surely have improved a lot." Laue's 'diplomacy' in order to divert Friedrich from Sommerfeld's plans must have brought the discord to a climax. When Friedrich and Knipping obtained indeed the diffraction pattern, Sommerfeld celebrated this result without Laue. "Why did you exclude me when you celebrated the discovery of X-ray diffraction with Friedrich and Knipping and the younger colleagues?" Laue confided his bitterness to Sommerfeld in the same letter. "But let us lay the past to rest, let's say 'never mind'. I felt always deeply hurt to be in an awkward relationship with a colleague whose accomplishments I have to value so high. I will be very relieved when this is going to change now"<sup>8</sup> (Laue, 1920).

Whatever had originally caused their personal discord, Laue's respect for Sommerfeld was sincere. He nominated him five times (1917, 1919, 1929, 1932 and 1933) for the

<sup>6</sup> „Ich erwähne die Zweifel, welche anfangs die anerkannten Meister unserer Wissenschaft, die ich zu befragen Gelegenheit hatte, gegen den Gedanken der Kristall-Interferenzen gehabt hätten. Dabei habe ich allerdings auch an Sommerfeld gedacht, aber nicht minder an W. Wien und in gewissem Sinne auch an Röntgen, der ja nicht einmal nach den ersten Versuchen von Friedrich an die Interferenz-Natur der Punkte glauben wollte. Und dass ein wenig Diplomatie erforderlich gewesen wäre, um den Beginn der Versuche im Sommerfeldschen Institut zu erreichen, das ist allerdings richtig. Denn um die Wende März–April 1912 sah es so aus, als wollte Friedrich die Interferenzversuche zunächst noch zurückstellen. Da veranlasste ich Dr. Knipping, sich der Sache anzunehmen; und dann ging es wie im Walenstein: ‚Wenn es denn doch geschehen soll und muss, so mag ich's diesem Pestaluz nicht gönnen.‘ Es wäre mir lieb, wenn Sie auch von diesem Briefe Sommerfeld gelegentlich in Kenntnis setzten.“

<sup>7</sup> „und hoffentlich kommt Laue nicht. Ich fürchte, dass mein Behagen sehr unter seiner Anwesenheit leiden würde.“ The remark refers to an invitation to Wien's country house in the Alps, where the Munich physicists used to meet for skiing.

<sup>8</sup> „Warum haben Sie mich ausgeschlossen, als Sie mit Friedrich und Knipping und den anderen jüngeren Fachgenossen die Entdeckung der Röntgenstrahlinterferenzen feierten? Nun könnten Sie natürlich mit Recht darauf hinweisen, dass ich Ihnen gegenüber nicht immer korrekt aufgetreten war, namentlich kurz nach meiner Übersiedelung nach München. Aber Sie wussten doch, in welchem Gemütszustande ich kam. Hätten Sie mir „mildernde Umstände“ bewilligt, so hätten Sie jedenfalls unsere persönlichen Beziehungen sehr wesentlich gebessert. Doch lassen wir das Vergangene ruhen; sagen wir „Schwamm darüber“. Es hat mich immer tief geschmerzt, mit einem Fachgenossen nicht gerade gut zu stehen, dessen Leistungen ich so hoch bewerten muss. Es wird mir eine große Erleichterung sein, wenn das jetzt anders wird.“

Nobel prize. From their correspondence it is obvious that their relationship considerably improved after 1920 and eventually became quite friendly.

## 7. Conclusion

The momentous nature of the event that gave birth to X-ray crystallography and X-ray spectroscopy tended to belittle the disputes about 'Laue's discovery' as mere historical side-aspects. Even Sommerfeld, who had excluded Laue in 1912 from the celebration of the discovery, abstained from a historical account that would have exposed the erroneous expectations with which Laue must have presented his 'flash of inspiration' to him. Sommerfeld finally regarded "Laue's discovery", as he wrote in 1926, as "the most important scientific accomplishment in the history of the institute" (Sommerfeld, 1926, p. 291). Thus he rated the discovery of X-ray diffraction in crystals even higher than his own achievements in atomic theory and those of his disciples Pauli and Heisenberg in quantum mechanics.

Decades later, X-ray crystallography had become a science of its own right. By now Laue regarded the idea that led to the discovery of X-ray diffraction in crystals "so self-evident" that he "never understood the astonishment which it caused among the experts" (Laue, 1961, p. VII). From Laue's perspective it is understandable that he did not like to recall the initial misapprehension with which he pursued his 'flash of inspiration' against all odds. But even when Laue's flawed reasoning with the characteristic X-rays was mentioned, such as in Ewald's *Fifty Years* recollection, it was excused as a consequence of stress. "The persistence of this misapprehension at the end of a period of the most strenuous and successful work is like a sign of exhaustion" (Ewald, 1962, p. 45 and p. 55).

However, misapprehensions, rivalry and discord are part and parcel of the scientific enterprise. To blank them out not only distorts the historical account but also denies the role of doubt and uncertainty in the process of discovery. Recalling these facets together with the celebrated discoveries, therefore, does not diminish the merits of the discoverers but rather adds to a better understanding of their accomplishments. In the case of the discovery of X-ray diffraction in crystals, weakness of memory became the rule, so that the 'Laue experiment' is commonly recalled as if it was intended originally how it entered our textbooks – with the crystal as a three-dimensional diffraction grating that selects from the continuous X-ray spectrum of the primary beam the monochromatic X-rays for interference in the Laue spots. A hundred years later, it is time to correct this recollection.

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