## THEN & NOW

## Max von Laue and the discovery of X-ray diffraction in 1912

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From today's perspective, only a few discoveries are regarded to be as exciting and as ground-breaking as the discovery of X-ray diffraction by crystals. "The undersigned are engaged since 21 April 1912 with experiments about the interference of X-rays passing through crystals," a one-page report announced the discovery to the Bavarian Academy of Science. It was signed by Walter Friedrich, Paul Knipping and Max Laue (then still without the "von") and deposited by Arnold Sommerfeld on 4 May 1912 in order to ascertain the discovery before it was officially communicated in the form of an elaborate paper. "The guiding idea was that interferences arise in consequence of the space lattice structure of the crystals, because the lattice constants are ca. 10 times greater than the conjectured wavelengths of the X-rays." A simple drawing sketched the experimental arrangement. As evidence for the discovery some exposures were attached.1

The discovery was immediately recognized as a sensation. In England, William Henry Bragg and William Lawrence Bragg, father and son, developed an alternative method by which they confirmed the discovery. Within about a year it became clear that the discovery gave birth to two new sciences,

Figure: The discovery of X-ray diffraction by crystals was made in April 1912 in the Institute of Theoretical Physics at the University of Munich. The experiment was proposed by Max von Laue and performed by Walter Friedrich and Paul Knipping. The

photograph shows the experimental apparatus used for the discovery. It is on exhibit at Deutsches Museum in Munich. (Source: Deutsches Museum, Archiv, Bildnummer 103; reproduced with permission of Deutsches Museum).

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<sup>&</sup>lt;sup>1</sup> Deutsches Museum Munich, Archive (henceforth abbreviated as DM), document 1951-5. Reprinted in [1].



X-ray crystallography and X-ray spectroscopy. Few discoveries were so swiftly awarded Nobel prizes. Max von Laue, who had suggested the Munich experiment, received the 1914 Nobel Prize "for his discovery of the diffraction of X-rays by crystals", the Braggs received the 1915 prize "for their services in the analysis of crystal structure by means of X-rays".<sup>2</sup>

In retrospect, Laue's idea that prompted the discovery appears straightforward: Send a beam of Xrays through a crystal, and the regular three-dimensional arrangement of crystal atoms will sort out those that are seen in the Laue spots from the mixture of wavelengths in the primary beam by interference. Thus, the experiment is evidence for the wave nature of X-rays and the space lattice of crystals at the same time. This is how the discovery entered the textbooks.

Yet, the historical events were not as straightforward as it seems in retrospect. Laue revealed in his Nobel speech that the "acknowledged masters of our science" - he meant in particular Sommerfeld, in whose institute he was Privatdozent - "entertained certain doubts" about his idea. "A certain amount of diplomacy was necessary before Friedrich and Knipping were finally permitted to carry out the experiment according to my plan".<sup>3</sup> Indeed, it is not clear what Laue had originally suggested and why the "acknowledged masters" were opposed to his

plan. But it is clear that the discovery happened against all odds and remained disputed for some time [2]. Peter Debye, for example, remarked that "one should generally not trade merit against luck with such things".<sup>4</sup> Another pioneer of X-ray research, Henry Moseley, believed that the Munich discoverers "entirely failed to understand what it meant, and give an explanation which was obviously wrong".<sup>5</sup>

The first publication by Friedrich and Knipping reveals indeed that the early experiments were based on a misapprehension. It is disclosed by the argument as to which crystal should be used in the diffraction experiment. "Because we believed at first that we had to deal with fluorescence radiation, a crystal had to be chosen that contained a metal with a considerable atomic weight," Friedrich and Knipping presented as the argument why they chose copper sulphate as a crystal [3, p. 314]. In other words: The crystal was not imagined to act as a threedimensional diffraction grating for the primary beam of X-rays, but as an emitter of the so-called characteristic X-rays. Laue apparently expected that if this characteristic radiation originates from the regularly arranged points of the crystal's space lattice, then they should be subject to interference.<sup>6</sup> Accordingly, the photographic plates on which the diffraction pattern was recorded were placed left and right and in the back of the crystal [3, Fig. 1], so that

the primary beam would not disturb the expected effect.

If this was the plan presented to the "acknowledged masters" in order to divert Sommerfeld's assistant Friedrich from his other tasks, Sommerfeld had good reasons to object. There is no phase relation between the characteristic radiations emitted at different points in the crystal, and so the condition for interference is not met. "No wonder Sommerfeld refused machine time," Paul Forman concluded in 1969 in a critical analysis about the "myth of the discovery of X-ray diffraction" [1, p. 63–64].

Faced with Sommerfeld's objection, Laue must have had some pains to persuade Friedrich to perform the experiment. Apparently Laue addressed Knipping, a doctoral student of Röntgen, in order to overcome Friedrich's hesitance. And then Friedrich reacted like in Schiller's Wallenstein, Laue revealed later: "Wenn es denn doch geschehen soll und muss, so mag ich's diesem Pestaluz nicht gönnen".<sup>7,8</sup> When the first experiments produced no result, "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," Paul Ewald reconstructed the course of events many years later [4, p. 44]. Abram F. Ioffe, a Russian physicist who used to collaborate with Röntgen at that time, described the discovery as the result of Knipping's frustration: "In order to record at least something

<sup>&</sup>lt;sup>2</sup> http://www.nobelprize.org/nobel\_prizes/physics/laureates/.

<sup>&</sup>lt;sup>3</sup> http://www.nobelprize.org/nobel\_prizes/physics/laureates/1914/laue-lecture.pdf.

<sup>&</sup>lt;sup>4</sup> P. Debye to A. Sommerfeld, 13 May 1912. DM, HS 1977-28/A,61.

<sup>&</sup>lt;sup>5</sup> Quoted in J. L. Heilbron, H. G. J. Moseley: The life and letters of an English physicist, 1887–1915. University of California Press, Berkeley 1974, 194–195 (the letter is dated 4 November 1912).

<sup>&</sup>lt;sup>6</sup> See also the discussion of this argument in [1, p. 63–64].

<sup>&</sup>lt;sup>7</sup> M. von Laue to P. P. Ewald, 1 May 1924. Quoted in [1, p. 64].

<sup>&</sup>lt;sup>8</sup> engl: Yet, if it has to be and should be done, I don't want Pestalutz to be the one.

on the photographic plate, he placed it so that it became exposed by the X-rays – and there was the great discovery" [5, p. 40].

The diffraction spots that surrounded the central spot of the primary beam could be explained by Laue as an interference pattern due to the crystal's space lattice: each spot was caused by Xrays that corresponded to a certain lattice constant and wavelength. But it remained mysterious how the monochromaticity observed in the "Laue-spots" came about. It was clear that it was not due to the crystal's characteristic X-rays but had to come from the primary beam because crystals like diamond had no characteristic radiation but nevertheless produced "Laue spots". It was known that the X-rays that are produced in the anticathode of an X-ray tube come in two varieties: one was polarized and could be described as electromagnetic pulses due to the braking of electrons ("Bremsstrahlen"); the other was the unpolarized fluorescence X-rays that seemed to be characteristic for the anticathode material. It was left to William Lawrence Bragg to demonstrate that the diffraction pattern was due to the reflection of the "white" Bremsstrahlen of the primary beam on the crystal planes that selected certain wavelengths for the diffraction pattern by what became known as the "Bragg condition". However, it took a few more months until the equivalence of Laue's and Bragg's approaches became clear.

Further evidence came from the first X-ray spectra from an-

ticathodes produced by Moseley and Darwin in 1913. These spectra showed the continuous "white" Bremsstrahlen spectrum and the sharp peaks of the characteristic radiation. But Laue was still hesitant to accept Bragg's explanation. If all wavelengths were present in the "white" Bremsstrahlen, then the Laue or Bragg equations for interference would be satisfied for any angle - with the result that the photographic plate should be totally blackened. It was left to Sommerfeld and Ewald to show that the Bremsstrahlen spectrum was limited so that it did not contain arbitrarily short wavelengths. Only with this restriction did the modern interpretation of "Laue's discovery" become clear [6, 7].

What at first sight appears in retrospect as a splendid discovery based on Laue's "flash of inspiration", therefore, displays with closer historical scrutiny a complex maze of misapprehensions and uncertainty. Laue's "diplomacy" was followed by a grave discord with Sommerfeld. "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 1920. He apologized that he had not always behaved correctly towards Sommerfeld but also complained that Sommerfeld had had little patience with his problems [2, p. 37]. The discord, however, did not prevent Sommerfeld from praising "Laue's discovery" a few years later as "the most important scientific accomplishment in the history of the institute" [8, p. 291].

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