The Big and Little of Fifty Years of Mössbauer Spectroscopy at Argonne

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by
Catherine Westfall
Office of the Director, Argonne National Laboratory

September 2005

Scientists whose careers span 50 Years of Mössbauer spectroscopy in the lobby of Argonne's Advanced Photon Source. Back row, left to right: Michael Hu, Gopal Shenoy, Ercan Alp, Philip Mannheim, Tom Toellner, Harry Lipkin. Sitting, left to right: Caroline L'abbe, Wolfgang Sturhahn.

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THE BIG AND LITTLE OF FIFTY YEARS OF MÖSSBAUER SPECTROSCOPY AT ARGONNE¹

by

Catherine Westfall

1 INTRODUCTION

Using radioactive materials obtained by chance, a turntable employing gears from Heidelberg's mechanical toy shops, and other minimal equipment available in post World War II Germany, in 1959 Rudolf Mössbauer confirmed his suspicion that his graduate research had yielded ground-breaking results. He published his conclusion: an atomic nucleus in a crystal undergoes negligible recoil when it emits a low energy gamma ray and provides the entire energy to the gamma ray.²

In the beginning Mössbauer's news might have been dismissed. As Argonne nuclear physicist Gilbert Perlow noted: "Everybody knew that nuclei were supposed to recoil when emitting gamma rays – people made those measurements every day." If any such effect existed, why had no one noticed it before? The notion that some nuclei would not recoil was "completely crazy," in the words of the eminent University of Illinois condensed matter physicist Frederich Seitz.³

Intrigued, however, nuclear physicists as well as condensed matter (or solid state) physicists in various locations – but particularly at the Atomic Energy Research Establishment at Harwell in Britain and at Argonne and Los Alamos in the U.S. – found themselves pondering the Mössbauer spectra with its nuclear and solid state properties starting in late 1959. After an exciting year during which Mössbauer's ideas were confirmed and extended, the physics community concluded that Mössbauer was right. Mössbauer won the Nobel Prize for his work in 1961.

In the 1960s and 1970s Argonne physicists produced an increasingly clear picture of the properties of matter using the spectroscopy ushered in by Mössbauer. The scale of this traditional Mössbauer spectroscopy, which required a radioactive source and other simple equipment, began quite modestly by Argonne standards. For example Argonne hosted traditional Mössbauer

¹ The author greatly appreciates the help of the following scientists in the preparation of this manuscript: Ercan Alp, Bobby Dunlap, Frank Fradin, Hans Frauenfelder, Don Geesaman, Stanley Hanna, Gregors Hansen, Michael Kalvius, Clyde Kimball, Harry Lipkin, Walter Potzel, Gilbert Perlow, John Schiffer, Gopal Shenoy, Dean Taylor and Rich Valentin. Special thanks goes to Charles Johnson for many hours of help and encouragement. All interview transcripts are at Argonne National Laboratory, Argonne, Illinois.

² Rudolf Mössbauer, "The Discovery of the Mössbauer Effect," *Hyperfine Interactions*, 126 (2000), 1-12.

³ Private communication, Perlow, 26 Apr 2002; as quoted in Lipkin, "The Early History of the Mössbauer Effect," *Hyperfine Interactions*, 72 (1992), 3-11.

spectroscopy research using mostly existing equipment in the early days and equipment that cost \$100,000 by the 1970s alongside work at the \$50 million Zero Gradient Synchrotron (ZGS) and the \$30 million Experimental Breeder Reactor (EBR) II. Starting in the mid-1990s,⁴ Argonne physicists expanded their exploration of the properties of matter by employing a new type of Mössbauer spectroscopy – this time using synchrotron light sources such as Argonne’s Advanced Photon Source (APS), which at \$1 billion was the most expensive U.S. accelerator project of its time.

Traditional Mössbauer spectroscopy looks superficially like prototypical “Little Science” and Mössbauer spectroscopy using synchrotrons looks like prototypical “Big Science.” In addition, the growth from small to larger scale research seems to follow the pattern familiar from high energy physics even though the wide range of science performed using Mössbauer spectroscopy did not include high energy physics. But is the story of Mössbauer spectroscopy really like the tale told by high energy physicists and often echoed by historians? What do U.S. national laboratories, the “Home” of Big Science, have to offer small-scale research? And what does the story of the 50-year development of Mössbauer spectroscopy at Argonne tell us about how knowledge is produced at large laboratories?⁵

In a recent analysis of the development of relativistic heavy ion science at Lawrence Berkeley Laboratory I questioned whether it was wise for historians to speak in terms of “Big Science,” pointing out that Lawrence Berkeley Laboratory hosted large-scale projects at three scales, the grand scale of the Bevatron, the modest scale of the HILAC, and the mezzo scale of the combined machine, the Bevalac. I argue that using the term “Big Science,” which was coined by participants, leads to a misleading preoccupation with the largest projects and the tendency to see the history of physics as the history of high energy physics.⁶ My aim here is to provide an additional corrective to such views as well as further information about the web of connections that allows national laboratory scientists working at a variety of scales to produce both technological and scientific innovations. I shall pursue this theme by looking first at the circuitous path that led to the discovery and understanding of the Mössbauer effect, then outlining the spread of Mössbauer spectroscopy, and finally describing episodes that highlight three generations of Mössbauer spectroscopy at Argonne.

⁴ For more information on the ZGS, see Elizabeth Paris, “Do You Want to Build Such a Machine: Designing a High Energy Proton Accelerator for Argonne National Laboratory,” ANL/HIST-2; for more information on EBR-II, see Catherine Westfall, “Vision and Reality: The EBR-II Story,” *Nuclear News*, Feb. 2004, 25-32.

⁵ See Robert Seidel’s seminal work, “A Home for Big Science: The Atomic Energy Commission’s Laboratory System,” *Hist. Phys. Biol. Scien.*, 16 (1986), 135-175.

⁶ Westfall, “Rethinking Big Science: Modest, Mezzo, Grand Science and the Development of the Bevalac, 1971-1993,” *Isis*, 94 (2003), 30-56.

2 DISCOVERY AND UNDERSTANDING

2.1 MÖSSBAUER'S ROAD TO DISCOVERY, 1953 TO 1958

In 1953 Heinz Maier-Leibnitz, a specialist in nuclear spectroscopy, one of post-war Germany's eminent nuclear physicists, and a newly named professor of technical physics at the Technical University of Munich, gave an assignment to his new graduate student, 24-year-old Rudolf Mössbauer. Mössbauer's subject for both his master's and doctoral work would build on recent research by British physicist Philip B. Moon and Swedish physicist K. G. Malmfors.⁷

Moon and colleagues at the University of Birmingham had first observed nuclear resonance absorption, that is, that a nucleus absorbs γ -rays at a well-defined energy corresponding to a specific transition. This phenomenon had been predicted as early as 1929, when Werner Kuhn considered it in analogy to the optical absorption. Both nuclear and atomic resonances were measured with an experimental set-up that included a radiation source and an absorber made of the same nuclei or atoms. Quanta emitted by the source would produce resonant absorption at the same energy in the absorber. The nuclei or atoms excited in the absorber would then decay to their ground state, in the process producing radiation with the same energy. Two detectors were placed, one to register the re-emitted radiation (called fluorescence) and the other to measure corresponding reduction in transmitted intensity (called absorption)⁸ (Figure 1).

The nuclear resonance phenomenon was not observed for decades. Comparison of the nuclear and atomic cases shows why. When a photon is emitted by an atom or nucleus, most of the resulting energy is taken up by the photon, but part is lost through the kinetic energy of the recoiling atom or nucleus. As a result there is an energy deficit between absorption and emission lines (Figure 2). As Kuhn pointed out, although in the atomic case the recoil effects are small, these effects are large in the nuclear case since the recoil effects scale with photon energy, which is higher for the nuclear γ -transitions.⁹

Observation of nuclear resonance required either a compensation of energy deficit between source and absorber, or an increase in Doppler broadening, both of which would increase the overlap between emission and absorption states. Nuclear resonance was finally observed in 1951 when P.B. Moon and his Birmingham group managed to obtain measurements

⁷ Paul Kienle, "Obituary," *Physics Today*, 54:8 (2001), 65; P. B. Moon, "Resonant Nuclear Scattering of Gamma Rays: Theory and Preliminary Experiments," *Proc. Phys. Soc.*, 64 (1951), 76-82.

⁸ Mössbauer (ref. 2), 1-4. For more on the Mössbauer effect and its discovery, see Rudolf Mössbauer, "Recoilless Nuclear Resonance Absorption of Gamma Radiation," Nobel Lecture in Physics, 1942-1962 (Amsterdam, 1964), 584-601; Terry Sanderson, "Charles Johnson and the Liverpool Mössbauer Group." (MS thesis, Univ. of Liverpool, 2002); Klaus Hentschel, *Zum Zusammenspiel von Instrument, Experiment und Theorie: Rotverschiebung im Sonnenspektrum und Verwandte Spektrale Verschiebungseffekte von 1880-1960* (Hamburg, 1995).

⁹ Sanderson, *ibid.*, 3-4.

FIGURE 1 Rudolf Mössbauer's Original Experimental Set-Up, with Kind Permission of Springer Science and Media and Rudolf Mössbauer, Technical University Munich, D-85747, Germany

FIGURE 2 First observation of the Mössbauer Effect in the Case of the 129 keV Transition in ^{191}Ir , with Kind Permission of Springer Science and Media and Rudolf Mössbauer, Technical University Munich, D-85747, Germany

of 411 keV γ -radiation in the decay of ^{198}Hg to ^{198}Au by placing the source on an ultracentrifuge and applying the considerable speed of 670 m/s so that the linear Doppler effect compensated for the energy deficit between absorber and source caused by the recoil effect.¹⁰

Mössbauer's assignment in 1953 was to apply a method he learned from a paper by Malmfors, who in 1952 overcame the recoil effect by heating both the source and absorber, which increased the overlap between the absorption and emission lines. Aided by Maier-Leibnitz and reference material obtained from the Deutsches Museum library, which Mössbauer considered "the only reasonable library in Munich," the young graduate student decided to work on the 129 keV transition emitted when ^{191}Os decays to ^{191}Ir . The isotope seemed suitable for Malmfors' methods and the lifetime of that particular energy state was unknown and therefore would provide a new result for his thesis. Also crucial was the fact that the isotope appeared in the catalogue put out by the Harwell laboratory in Britain. At the time this catalogue was Maier-Leibnitz's only source for obtaining reactor-produced isotopes, since Germany had not yet received permission to build a reactor from the military government put in place when the Nazis were defeated. Mössbauer built a detector consisting of a bundle of 12 proportional chambers, which had an efficiency of about 5%. In Mössbauer's words, although this rate of efficiency marked a "world's record," his master's thesis showed that the detector was "entirely insufficient" to measure nuclear resonance.¹¹

In 1955, with the ink barely dry on his master's thesis, Mössbauer received the exciting offer to go to the Max Planck Institute for Medical Research in Heidelberg. He later remembered

¹⁰ Ibid. Moon, (ref. 7), 76.

¹¹ Mössbauer, (ref. 2), 4.

that he “accepted this offer to Heidelberg within less than a second because it was a dream for me.” Research life was better in Heidelberg than in Munich because the equipment was better. Mössbauer followed the example of Heidelberg colleagues and replaced his proportional counters with NaI counters which offered nearly 100% efficiency for his measurements. In addition, since the Heidelberg Institute’s budget was greater, he was able to buy electronic components that were much more suitable than the home-made variety.¹²

At this point Mössbauer also benefited from the unexpected kindness of a stranger. He needed radioactive sources and the only obvious place to get them was Britain. The military government’s regulations specified that Germans could only communicate with the British in such cases through the Max Planck Institute in Göttingen. Mössbauer became impatient with the process, and as he later admitted “wrote directly to England, which was strictly against the law.” To his good fortune, a woman who received his letter at one of the British companies who made materials for Harwell decided to help him “bypass regulations and manufacture the radioactive sources” he needed.¹³

Mössbauer’s luck would only improve. One crucial decision he had to make was how to control temperatures. Malmfors had increased the temperature of both the source and the absorber, and this was the method preferred by Maier-Leibnitz. But Mössbauer decided instead to lower the temperature. His reasons were strictly practical. He reasoned that “going down in temperature to the boiling point of liquid nitrogen” would have exactly the opposite “effect as going up in temperature,” consequently reducing the resonance overlap. This method was possible because Heidelberg had liquid nitrogen. Mössbauer also liked the idea of lowering the temperature because it seemed to him that it would be “much simpler to build a cryostat rather than a furnace.”¹⁴

In the long run, this decision led to the Nobel Prize. In the short run, Mössbauer faced considerable confusion. In measurements aimed at determining intensity differences, Mössbauer obtained a value that was correct, but the sign was wrong. He expected that decreased temperature would reduce the overlap of absorption and emission lines, thus producing an increase in transmitted intensity. Instead his measurements indicated there was a decrease, which seemed to indicate that the lines overlapped less. First, Mössbauer sought, in vain, for a dirt effect. Next the confused graduate student sought, in vain, for the answer from various German physics professors, including J. Hans Daniel Jensen.¹⁵

¹² Sanderson, 4-5. Hentschel notes that: “Energy resolutions of $E/\Delta E \approx 4 \times 10^{10}$, “which Mössbauer obtained, were at the time unheard of. “One of the few exceptions was Robert W. Dicke at Princeton who had a similar idea at the time but did not follow it up; according to his own later testimony: ‘I considered narrowing γ -radiation from radioactive nuclei in a solid in this way, but unlike Mössbauer did not think it was feasible.’” Hentschel, “Measurements of Gravitational Redshift between 1959 and 1971,” *Annals of Science*, 53 (1996), 269-295.

¹³ Mössbauer, (ref. 2), 5.

¹⁴ Ibid., 5-6. Hentschel, (ref. 12), 270.

¹⁵ Ibid.

In the end Mössbauer took inspiration from a 1939 paper by Walter Lamb that had previously been given to him by Maier-Leibnitz and had been subsequently suggested as a good source of information by Jensen. Lamb had developed, in Mössbauer's words "a theory for the resonance capture of slow neutrons in crystals." Mössbauer applied this theory "to the analogous problem of the resonance absorption of gamma radiation" and came to understand the effect that later bore his name.¹⁶

Mössbauer would often explain that this newly discovered phenomenon was like the situation of "a person throwing a stone from a boat. The majority of the energy is submitted to the stone, but a small amount goes into the kinetic energy of the recoiling boat. During summer time, the boat will simply pick up this recoil energy. If, however, the person throws the stone during winter time, with the boat frozen into the lake, then practically all the energy is going into the stone thrown and only a negligible amount is submitted to the boat. The entire lake will, thus, take up the recoil and this procedure occurs as recoilless process."¹⁷

After his initial understanding of the results of his first experiment Mössbauer "successfully performed a theoretical treatment." Two "incredibly narrow" emission and absorption lines overlapped "right at the resonance position." These lines later came to be called the recoilless absorption lines or Mössbauer lines. Now the sign he had observed in his thesis experiment, "which had been opposite to expectation, could be explained." Cooling the absorber and source had been crucial, for "with decreasing temperature, the extraordinarily narrow lines grew, increasing their overlap, thus increasing the absorption and by consequence, decreasing the transmission." Mössbauer published this result in his doctorate thesis.¹⁸

After submitting his thesis publication and sending two preprints to the two "popes in Doppler-shift experiments," Moon of Birmingham and F. R. Metzger at the Bartol Research Foundation in Pennsylvania, Mössbauer was struck with inspiration: he could measure the sharp lines by employing the linear Doppler effect to provide new insights into the properties of matter. "I was so excited," Mössbauer remembered, "I dashed across the hallway into the office of Professor Maier-Leibnitz without even knocking on the door and cried: I take the next train back to Heidelberg, because I forgot about the major experiment."¹⁹

Mössbauer was "panicked" that he would be scooped by the more experienced physicists, especially since making the measurements was "rather easy," once the Mössbauer effect itself was understood, since measurement involved "an effect of order 1%, while the previous measurement of cross section gave only an intensity change of order 0.01%."²⁰ The basic

¹⁶ Mössbauer, "Recoilless Nuclear Resonance Absorption of Gamma Radiation." (ref. 8), 591.

¹⁷ Mössbauer, (ref. 2), 8-9.

¹⁸ Ibid. Mössbauer, "Kernresonanzfluoreszenz von Gammastrahlung in ¹⁹¹Ir," *Z. f. Phys.* 151 (1958), 124-143. The other two papers that announced his discovery and explained the early experiments were: Mössbauer, "Kernresonanzabsorption von Gammastrahlung in ¹⁹¹Ir," *Naturwissenschaften* 45 (1958), 538-9 and Mössbauer, "Kernresonanzabsorption von γ -Strahlung in ¹⁹¹Ir," *Z. Naturforschung* 14a (1959), 211-16.

¹⁹ Mössbauer, (ref. 2), 10.

²⁰ Ibid.

elements of Mössbauer's experiment were those used by Moon in 1951; these same elements would also be used by the many who would perform subsequent Mössbauer's spectroscopy. The experimental set-up consisted of a source of γ -radiation mounted on a rotating disk, a stationary absorber, and a detector to measure the transmitted radiation. This equipment allowed measurement of the recoilless nuclear resonant absorption of γ -rays using Doppler shifts of the absorbing relative to the emitting nuclei (Figure 3).

Although easy in concept, the experiment was not without its challenges. To rotate the source, Mössbauer needed a turntable. To create one he needed a set of cone-shaped wheels. Since he was in a hurry and it "took about three days for a skilled guy to finish" just one wheel, Mössbauer "went to the mechanical toy shops of Heidelberg and bought" all their gears. Even though the turntable did not "work very smoothly" since the wheels did not fit together, he was able to measure the width of the first excited state of ^{191}Ir ; it was the first measurement using Mössbauer spectroscopy²¹ (Figure 4).

Mössbauer later noted that he wanted to make further measurements to "quietly cream off" the results of this initial work, an objective that he knew would be hard in light of the limited resources he had compared to those of his competitors. For that reason he published the spectra in *Naturwissenschaften*, "a journal as obscure as possible." He later insisted that the inability to obtain the necessary radioisotopes prevented him from immediately advancing to the next, obvious steps in this line of research. "I knew precisely what I had at my hands," he later explained. In any event, in 1961, just three years after publishing the spectra, Mössbauer would win the Nobel Prize for discovering the effect that bears his name. In the meantime, however, the action would shift from Germany as others further developed Mössbauer spectroscopy.²²

FIGURE 3 Principle and Actual Realization of the Observation of the Mössbauer Effect Using Doppler Shifts of the Emitting Relative to the Absorbing Nuclei, with Kind Permission of Springer Science and Media and Rudolf Mössbauer, Technical University Munich, D-85747, Germany

²¹ Ibid. Mössbauer, "Kernresonanzabsorption von Gammastrahlung in Ir^{191} ," (ref. 18), 538.

²² Mössbauer, (ref. 2), 9. Ibid., 10.

2.2 THE SCENE SHIFTS – AND WHY THE LATE DISCOVERY?

Mössbauer's hopes for obscurity were dashed within months. On a visit to see Maier-Leibnitz in an extended stay abroad, Hans Frauenfelder met Mössbauer and learned of his work and reported back to Harry Lipkin, a visitor from Israel who was leading Frauenfelder's group at the University of Illinois. Lipkin did not know what to think so he consulted Seitz.²³ In the meantime, John Schiffer, a nuclear physicist from Argonne, was visiting Darragh Nagle and others at Los Alamos. John Marshall had learned of Mössbauer's article and mentioned the odd result over lunch.²⁴

Soon experiments were underway to test Mössbauer's results at Los Alamos and Argonne. The Los Alamos work reportedly resulted from a bet that Mössbauer was wrong. Schiffer later remembered that when he returned to Argonne he read the *Naturwissenschaften* article for himself and concluded that "the effect seemed very far fetched." After "talking it over with several colleagues, including theorists, we decided that we should repeat it because it was probably wrong." After all, "it was not a good idea to have a wrong result stand in the literature."²⁵

The participants were later struck by the fact that the effect had so long been missed. Mössbauer was amazed that "it had been the delusion" of Jensen and Steinwedel, who had written a paper that followed up Lamb's work, "to assume that the recoilless lines could be approached in a classical steady way," instead of understanding them as "a typical quantum mechanical phenomenon, with the recoilless lines reached in an unsteady manner, shooting steeply out above a phonon background." (Ironically, Mössbauer felt it was his own "mediocre knowledge of quantum mechanics" that saved him from being misled by this error – he had simply been unable to "understand the contents of Jensen's paper.")²⁶

Schiffer would later judge that the Mössbauer effect should have been obvious for other reasons. "By the 1930s," he noted "any nuclear physicist should have realized that since x-rays

FIGURE 4 Position of the Emission and Absorption Lines at Room Temperature for the 129 keV Transition in Ir^{191} , Indicating the Common Region of Overlap and Assuming Nuclei Subject Only to Doppler Broadening, with Kind Permission of Springer Science and Media and Rudolf Mössbauer, Technical University Munich, D-85747, Germany

²³ Hans Frauenfelder, interview with Westfall, 26 Nov 2003; Lipkin, interview with Westfall, 16 Oct 2002.

²⁴ Frauenfelder, *The Mössbauer Effect* (New York, 1962), 12.

²⁵ Schiffer, "Memories of Early Mössbauer Work at Argonne." The author would like to thank Schiffer for providing a copy of his short memoir.

²⁶ Mössbauer, (ref. 2), *ibid.*, 6.

can be scattered by crystals in a recoil-free manner, as demonstrated by Bragg and explained by Debye and Waller, there should be recoil-free emission and absorption of nuclear gamma rays under the right conditions long before Lamb's work."²⁷

Lipkin would expand on this idea, noting that "all the physics to understand the Mössbauer effect" had been published by Lamb and others. In fact, the understanding "that photons could be scattered by atoms in a crystal without energy loss due to recoil was basic to all work in x-ray diffraction and crystallography. All the quantitative calculations, including the definition and evaluation of the Debye-Waller factor, were well known." The limitation was that "nobody interpreted this as a probability that a photon could be scattered by an atom in a crystal without energy loss due to recoil." After all: "The x-ray physicist worked entirely with the wave picture of radiation and never thought about photons."²⁸

At the same time: "Nobody noted that scattering the x-rays involved a momentum transfer and that coherence would be destroyed if there was any energy loss in the momentum transfer process. They did not see that the Debye-Waller factor also could be interpreted as the possibility that the scattering would be elastic and not change the quantum state of the crystal. They did not see that localizing the atoms to the extent needed to get phase coherence in the wave picture meant introducing an uncertainty in the momentum of the atom which was large enough to absorb the momentum transfer without energy transfer."²⁹

Lipkin concluded that the crux of the problem was a "communication barrier" that separated solid state and nuclear physicists who "spoke different languages and did not understand each other."³⁰ Certainly much of the subsequent development of the Mössbauer effect would be shaped by the difficult challenges and rich possibilities that came with the multidisciplinary nature of the work. Indeed, by mid-1960 researchers would discover that the ultra-high energy resolution of the Mössbauer resonance had the ability to resolve the so-called hyperfine interactions that connect nuclei with their surrounding electrons. Since these interactions connect the nuclear moments (nuclear monopole moment, the nuclear magnetic moment, and nuclear quadrupole moment) and corresponding solid state properties (electron charge distribution at the nucleus, magnetic field at the nucleus, and electric field gradient at the nucleus), understanding and exploiting the Mössbauer effect would require work at the intersection of nuclear and solid state physics and the skills and knowledge of both specialties – and Mössbauer spectroscopy would become an important tool for probing the properties of matter in a wide variety of specialties.

²⁷ Private communication, Schiffer, 12 Mar 2003.

²⁸ Lipkin (ref. 3), 4.

²⁹ Ibid., 4-5.

³⁰ Ibid., 5.

2.3 THE ROAD TO UNDERSTANDING THE MÖSSBAUER SPECTRUM, 1959-1960

On 3 August 1959 two papers were submitted to *Physical Review*, one by Schiffer and others at Argonne, another by Nagle and others at Los Alamos. (Schiffer later remembered that the Argonne group's measurements were first by a hair.)³¹

As the Los Alamos scientists noted, their paper reported “our repetition of Mössbauer’s measurements,” of ^{191}Os decaying to ^{191}Ir , “the extension of the temperature range to 1.5°K, measurements on various foil thicknesses, and a demonstration of semiquantitative agreement of the results with theoretical calculations by Visscher based upon a theory by Lamb.” The Argonne group, which worked with William Visscher’s calculations, also reproduced and extended Mössbauer’s measurements with ^{191}Ir and made a first stab at measurements with another isotope, ^{182}W , which was obtained from beta decay of ^{182}Ta . Thanks to these two papers, Mössbauer’s odd result had been vouched for by scientists at not one, but two major U.S. laboratories.³²

Argonne scientists also pointed to the possibilities of further research, noting that the new measurement technique would “provide an effective tool in measuring lifetimes in a region overlapping with fast electronic techniques.” They also speculated that: “It might also be possible to observe hyperfine structure effects splitting up the line into several components,” a suggestion independently provided by Mort Hamermesh and Mössbauer.³³

Lipkin later remembered, however, that despite such speculation, many physicists “insisted it was an unimportant curiosity. It was a nice exercise in quantum mechanics, but would not lead to anything new and useful. Nuclear physicists claimed that it would teach nothing new about nuclear structure. Solid-state physicists claimed that its use as a tool in solid-state physics could not compete with neutron scattering and nuclear magnetic resonance, which would get the same information more easily.” Lipkin, who had been interested in the new line of inquiry since hearing about Mössbauer’s work from Frauenfelder, was surprised that the Los Alamos work had not been reported at the annual APS meeting and that Argonne colleagues David Inglis and Maria Mayer had not thought to mention the Argonne work when discussing Lipkin’s upcoming summer appointment at their laboratory.³⁴

Lipkin was not the only one to remain interested in the newly discovered effect, however. That summer Schiffer was headed to Harwell for a year’s study funded by a Guggenheim Fellowship. Although he had originally planned to do other work with Harwell’s new tandem

³¹ L. L. Lee, et al., “Nuclear Resonance Absorption of Gamma Rays at Low Temperatures,” *Phys. Rev. Lett.* 3 (1959), 223-225; P. P. Craig, et al., “Nuclear Resonance Absorption of Gamma Rays in Ir^{191} ,” *Phys. Rev. Lett.* 3 (1959), 221-223. A later published form of William M. Visscher’s work can be found in Visscher, “Study of Lattice Vibrations by Resonance Absorption of Nuclear Gamma Rays,” *Annals of Physics* 9 (1960), 194-210.

³² Lee, Meyer-Schutzmeister, Schiffer, and Vincent, “Nuclear Resonance Absorption of Gamma Rays at Low Temperatures,” *Phys. Rev. Lett.* 3 (1959), 223.

³³ *Ibid.*, 225.

³⁴ Lipkin (ref. 3), 6-7.

accelerator, Schiffer would later remember that the “recent result with the Mössbauer effect was very much on [his] mind.” By fall 1959 others, including Harvard professors Robert V. Pound (a pioneer in nuclear magnetic resonance) and his graduate student Glen A. Rebka, had become interested in the new phenomenon. At this stage these researchers focused their attention on discovering whether other isotopes could be found that gave a larger effect and whether it could be embedded in crystals and compounds where the recoilless fraction could be increased.³⁵

To get advice in the search, Schiffer’s first step at Harwell was to contact Walter Marshall, who then headed Harwell’s theory group. Schiffer started by explaining Mössbauer’s results and “within an hour of conversation, Marshall understood the effect and its connection to the Debye-Waller factor in crystallography and explained to me what mattered most,” in the search for an isotope that would provide a large Mössbauer effect. A few months later, Marshall co-authored a note with Schiffer to lay out the same information for others. The note, which explained “that in the Mössbauer effect the fraction of recoilless gamma-rays can be calculated exactly within the framework of the Debye theory of solids by the Debye-Waller factor,” begins with the apology that it contained “nothing original” but did “clarify a perhaps trivially obvious point.” Schiffer later noted that the information was “only obvious to Marshall, a first-rate solid state physicist.” Schiffer “pushed Marshall to write it all down, because so many others were confused.”³⁶

As a result of his discussion with Marshall, Schiffer found a new direction. After the Argonne measurements he had discussions with theorists that led them to believe that they needed “a heavy massive nucleus and a high Debye temperature.” After talking to Marshall, Schiffer focused on the Debye-Waller factor and realized “the importance of low photon energies.” This led to “a systematic search of nuclear data compilation” in the Harwell library and “within a few hours,” Schiffer identified ^{57}Fe as an attractive candidate for further exploration and decided to create it through decay of ^{57}Co ³⁷ (Figure 5).

Within a few weeks Schiffer made the necessary ^{57}Co using the new Harwell tandem. A 15 December 1959 *Physical Review* article co-authored with Marshall explained: “A source of ^{57}Co has been prepared by bombarding 0.0005-in. Fe foil by 4-MeV deuteron from the Harwell Van de Graaf accelerator.” The researchers used “an absorber of a similar Fe foil” with “a xenon-filled proportional counter to detect the 14-keV radiation.” The idea was to measure the Mössbauer effect by observing the increase in the counting rate of γ rays between the stationary and the moving source. The results were impressive, despite what Schiffer would later call “this primitive method.” The “number of gamma rays emerging from the absorber with the source

³⁵ Schiffer (ref. 25). Henschel reports that Pound later judged that “his former experience with nuclear magnetic resonance and with perturbed directional correlations of short-lived isomeric nuclei ‘which involve much of the same physics’ had put him into a special disposition toward the new Mössbauer technology.” Henschel, (ref. 12), 273.

³⁶ Schiffer (ref. 25). Walter Marshall and Schiffer, “The Debye-Waller Factor in the Mössbauer Effect,” 7 April 1960. The author thanks Schiffer for providing this document.

³⁷ Schiffer (ref. 25).

FIGURE 5 Nuclear Decay Scheme for Co^{57} , with Kind Permission of Terry Sanderson, 1 Brimstage Close, Heswall, Wirral, Merseyside, England

moving with” a larger velocity “was approximately 20% larger than the number of gamma rays with the source stationary. This can be compared with the 1-2% absorption” reported in the ^{191}Ir measurements made by Mössbauer and the scientists at Los Alamos and Argonne.³⁸

As Schiffer and Marshall noted: “Since the magnetic field in Fe is expected to be approximately 200 kilogauss, one would expect the hyperfine splitting to be several times the line width of 4.5×10^{-9} eV even for the ground state magnetic moment of 0.05 nuclear magneton. The excited state presumably will have a larger magnetic moment and thus give rise to even larger splittings. It should be very easy to introduce ^{57}Fe into various alloys and compounds and obtain relative values of the hyperfine fields. These could also be measured easily as a function of temperature.” Schiffer and Marshall already had such experiments under construction.³⁹

Schiffer and Marshall also noted that T. E. Cranshaw had suggested “that the size of the absorption and the narrowness of the lines makes this an ideal case for measuring the gravitational red shift of a photon.” The same suggestion was made in a note by D. H. Wilkinson, by A. Boyle and S. Devons – and as a preliminary note by Pound in the

³⁸ Schiffer and Marshall, “Recoilless Resonance Absorption of Gamma Rays in Fe^{57} ,” *Phys. Rev. Lett.* 12 (1959), 556-557; Schiffer (ref. 25).

³⁹ Schiffer and Marshall, *ibid.*, 556.

previous month's edition of *Physical Review Letters*. The December 1959 edition of *Physical Review Letters* that contained the Schiffer and Marshall article also contained a paper with the reported effect in ^{57}Fe by Pound and Rebka which reported measurements to follow up the idea they had proposed the month before. The paper reported ^{57}Fe experiments with "the resonant scattering of a recoil-free γ ray" which appeared "to be sharp enough to be used for an experimental determination of the 'gravitational red-shift.'"⁴⁰

As the early experiments using the Mössbauer effect proceeded,⁴¹ the effort to better understand the ^{57}Fe spectrum continued. The Harvard work of Pound and Rebka showed three hyperfine spectral lines. In a paper submitted to *Physical Review Letters* in mid-December 1959 and published a month later, Frauenfelder and others at the University of Illinois made more exacting measurements and found four lines. This work gave qualitative evidence of the existence of magnetic hyperfine structure.⁴²

In early 1960, Stanley Hanna, Gilbert Perlow, and Richard Preston and others at Argonne published a paper to win "the race to interpret correctly the Mössbauer spectrum," in the words of British researcher Charles Johnson.⁴³ To accomplish this Argonne researchers built on their previous work which detected the polarization of γ rays. Fueled also by "the constant encouragement and discussion" with Hamermesh, they polarized γ rays by magnetizing the source and the absorber. Since this method allowed them to distinguish lines that were close together, the method allowed researchers to be the first to see the six transitions in the absorber and source⁴⁴ (Figure 6).

This identification of the entire hyperfine spectrum allowed a quantitative interpretation of the Mössbauer spectrum for the first time. As Schiffer later noted: "The trick," that the Argonne group mastered to get their results "was to get uniform enough large velocities to see the collection of lines, without any vibration (vibrational velocities had to be less than 1% of the uniform linear velocities (≈ 10 cm/sec) for the Doppler shifts, since the line widths were about 0.25 mm/sec, $\approx 1\%$ of the hyperfine splitting."⁴⁵

⁴⁰ Ibid., 556-557; R. V. Pound and G. A. Rebka, "Resonant Absorption of the 14.4-keV γ Ray from 0.10- μ sec Fe^{57} ," *Phys. Rev. Lett.* 12 (1959), 554-556.

⁴¹ Hentschel notes that others besides Pound, Rebka, Schiffer, and T. E. Cranshaw attempted to measure the gravitational red shift, including Robert Dicke at Princeton and Koichi Shimoda in Tokyo. However, these efforts were less successful. Hentschel (ref. 12), 273.

⁴² G. DePasquali, Frauenfelder, S. Margulies, and R. N. Peacock, "Nuclear Resonance Absorption and Nuclear Zeeman Effect in Fe^{57} ," *Phys. Rev. Lett.* 4 (1960), 71-73.

⁴³ Johnson, "Thirty Years of ^{57}Fe Mössbauer Hyperfine Spectrum," *Hyperfine Interactions*, 71 (1992), 15-30.

⁴⁴ S. S. Hanna, et al., "Observations on the Mössbauer effect in Fe^{57} ," *Phys. Rev. Lett.* 4 (1960), 28-9. Perlow, et al., "Polarization of Nuclear Resonance Radiation in Ferromagnetic Fe^{57} ," *Phys. Rev. Lett.* 4 (1960), 74-75. A compelling account of this and other early work is given in Hanna, "The Early Iron Age of the Mössbauer Era," *Hyperfine Interactions*, 90 (1994), 3-20.

⁴⁵ Schiffer (ref. 25).

FIGURE 6 Mössbauer Spectra of Co^{57} Source in Iron with an Iron Absorber, with Kind Permission of Springer Science and Media and Charles Johnson, Argonne National Laboratory

In a further experiment in spring 1960, Hanna, Perlow, Preston and others at Argonne applied an external field to the iron absorber and observed that the hyperfine field decreased. This showed that the hyperfine field was anti-parallel to the magnetization of the iron. This was surprising, since it had previously been assumed that the hyperfine field was parallel to the direction of the magnetization. Instead, a negative hyperfine field arises from the exchange interaction in the iron atom between the magnetic electrons (3d) and the inner electrons (3s) polarizing the inner electrons anti-parallel to the external field.⁴⁶

In April 1960 Brookhaven researchers O. C. Kistner and A. W. Sunyar produced another landmark paper, using the Mössbauer effect to measure the “resonant absorption of the 14.4-keV nuclear gamma ray of ^{57}Fe ” to “determine the quadrupole coupling for the $3/2^-$ excited state of ^{57}Fe bound in Fe_2O_3 , and to measure the energy shift of this nuclear gamma ray which is attributed to the effects of chemical binding.”⁴⁷ The paper thus presented two new features of the Mössbauer spectrum, the isomer shift (then called the chemical shift)⁴⁸ and quadrupole splitting.⁴⁹

⁴⁶ Hanna, et al., “Direction of the Effective Magnetic Field at the Nucleus in the Ferromagnetic Iron,” *Phys. Rev. Lett.* 4 (1960), 513-515.

⁴⁷ O. C. Kistner and A. W. Sunyar, “Evidence for Quadrupole Interaction of Fe^{57m} , and Influence of Chemical Binding on Nuclear Gamma-Ray Energy,” *Phys. Rev. Lett.* 4 (1960), 412-415.

⁴⁸ The isomer shift is a shift in energy levels that occurs between the ground state and the excited state of a nucleus. The ability to measure the isomer shift opened new spectroscopic possibilities. Like other hyperfine

Within months, important theoretical work also emerged. Lipkin presented basic concepts in a simplified form, increasing the understanding of the Mössbauer effect and publicizing its utility. In addition, the first comprehensive theory of the Mössbauer effect was published in June 1960 by two Argonne solid state physicists, Kundan S. Singwi and A. Sjölander.⁵⁰

In less than a year, researchers had gone from the first confirmation to a comprehensive theory of the Mössbauer effect, accumulating an impressive amount of new information in the process. The early measurements demonstrated how the Mössbauer spectrum arises and indicated how it could be used in application to solid materials. The work with ^{57}Fe further demonstrated that the Mössbauer effect could be used to measure the hyperfine field as a new way of probing magnetic materials. With the discovery of the isomer shift and quadrupole splitting, Brookhaven researchers showed that Mössbauer spectroscopy could provide new data on the electronic state of non-magnetic atoms (tin, antimony, xenon) as well as magnetic ones (iron, dysprosium), and thereby drew a new set of customers, chemists, to the field. As Argonne physicist Stanley Hanna later noted: the period was an “exciting, frenetic, but wonderful time during which so much was accomplished in such a short time.”⁵¹

With the advent of ^{57}Fe sources, the scientific possibilities of probing the properties of matter using the Mössbauer effect were all the more exciting because the equipment needed for Mössbauer spectroscopy was inexpensive and easy to use. As Frauenfelder pointed out in his 1962 review: “The ease with which the effect can be demonstrated with ^{57}Fe , its very large size, its persistence up to temperatures over 1000°C , and the very narrow line width, immediately changed this field of physics from one accessible to only a few laboratories to one in which even modestly equipped groups could compete.” In any often-cited table of the history of the Mössbauer effect Lipkin would dub this new era the “Iron Age,” and characterize it with the remark: “Wow!”⁵²

interactions, the isomer shift is a function of both electronic and nuclear properties. These properties are combined so that information for each separately cannot be gathered by Mössbauer spectroscopy alone. Nuclear parameters are constant; therefore hyperfine parameters can be used to compare electronic properties of different substances. Because the electronic properties are dependent on the electronic features of the atom, the isomer shift can provide information on a variety of information at the atomic level, such as atomic oxidation states and covalency effects. Sanderson (ref. 8), 233-233. Frauenfelder provides an explanation of isomer shifts. Frauenfelder (ref. 24), 53-57.

⁴⁹ Quadrupole splitting is the splitting of energy levels in the electric quadrupole interaction, which is the interaction between the electric field gradient and the nuclear quadrupole moment. Sanderson (ref. 8), 233. Frauenfelder provides an explanation of this Brookhaven work as part of his description of nuclear moments. Frauenfelder (ref. 24), 53.

⁵⁰ Lipkin, “Some Simple Features of the Mössbauer Effect,” *Annals of Physics* 9 (1960), 332-339; K. S. Singwi and A. Sjölander, “Resonance Absorption of Nuclear Gamma Rays and the Dynamics of Atomic Motions,” *Phys. Rev.* 120 (1960), 1093-1102.

⁵¹ Hanna, (ref. 44), 3.

⁵² Frauenfelder, (ref. 24), 12-13.

2.4 GRAVITATIONAL RED SHIFT EXPERIMENTS

Even though the understanding of the Mössbauer effect came quickly, exciting results were produced even more swiftly. John Schiffer, Ted Cranshaw, and A. Bruce Whitehead, working at Harwell, were the first to announce results for the gravitational red shift experiments. They began their February 1960 *Physical Review* article with an explanation of the phenomenon. The gravitational red shift, they noted, was: “the change in the frequency of spectral lines with gravitational potential.” This effect is “calculated from the time dilation in a gravitational potential which follows from the principle of equivalence.”⁵³ As historian Klaus Hentschel has described, Albert Einstein predicted the gravitational red shift in 1907 as part of a survey paper in *Jahrbuch der Radioaktivität und Elektronik* in 1907 and then repeated the prediction in later works, including his 1915-16 presentation of the general theory of relativity.⁵⁴

“From the point of view of a single coordinate system,” the article went on to explain “two atomic systems at different gravitational potentials will have different total energies. The spacings of their energy levels, both atomic and nuclear, will be different in proportion to their total energies. The photons are then regarded as not changing their energy and the expected red shift results only from the difference in the gravitational potential energies of the emitting and absorbing systems.”⁵⁵

Astronomical observations were made in the next few decades which “tended to confirm this effect,” although they were “somewhat ambiguous.” The discovery of the Mössbauer effect “suggested to several groups,” including Pound and Rebka, “the possibility of using this effect to measure the gravitational red shift.” For participants it was a heady time. As Schiffer later remarked, it was exciting to think of “measuring an effect that I had learned about in school that was one of the crucial experimental consequences of the General Theory of Relativity, mentioned in Einstein’s original paper that had never been observed before in a laboratory.”⁵⁶

Pound and Rebka published the results of their red shift measurements in *Physical Review Letters* on 1 April 1960, just six weeks after the Harwell work appeared. By this time the Harwell group also made another important measurement. As Schiffer later explained, Cranshaw and he had measured, “the analogous effect in a rotating system where the photons are shifted because of the centrifugal force (this is a second-order Doppler shift or the time dilation of Special Relativity).”⁵⁷

⁵³ Cranshaw, Schiffer, and A. B. Whitehead, “Measurement of the Gravitational Red Shift using the Mössbauer Effect in ^{57}Fe ,” *Phys. Rev. Lett.* 4 (1960), 163-164.

⁵⁴ Hentschel, (ref. 12), 269. This paper gives a detailed account of these and later gravitational red shift experiments. A general account of the gravitational red shift experiments written at the time can be found in Sergio De Benedetti, “The Mössbauer Effect,” *Scientific American*, 202 (1960), 73-80.

⁵⁵ Cranshaw, Schiffer, and Whitehead, “Measurement of the Gravitational Red Shift Using the Mössbauer Effect in ^{57}Fe ,” *Phys. Rev. Lett.* 4 (1960), 163.

⁵⁶ *Ibid.*, Schiffer, (ref. 25).

⁵⁷ *Ibid.* Pound and Rebka, “Apparent Weight of Photons,” *Phys. Rev. Lett.* 4 (1960), 337-341.

The work generated a lot of attention. Articles on the gravitational red shift experiments appeared in *The New York Times* in December 1959 and the *Observer* in February 1960.⁵⁸ Interest within the physics community led to a session at the American Physical Society meeting on 30 January 1960 that included preliminary results of the Harvard and Harwell red shift experiments as well as the Harwell measurement on a rotating system. Cranshaw and Schiffer summarized the session in an article published by *Nature* in March entitled: “Experiments to test Einstein’s Principle of Equivalence.”⁵⁹

The Harwell measurements used a drop of 12.5 meters (which was possible because of a water tower at the laboratory) and a 30 millicurie source of ^{57}Co .⁶⁰ As the *Nature* article explained, this intense source “was mounted on a moving-coil transducer also operated sinusoidally. A gramophone pick-up was placed so that the needle was moved by the rod holding the source, and the output from this pick-up enabled the movement of the source to be monitored continuously.” Measurements were made with “a proportional counter” filled “with krypton,” since this substance “has an absorption just below the 14.4-keV line, and so a comparatively low pressure can still give a high efficiency.”⁶¹

Although the experimental equipment was simple, elaborate care was taken to make accurate measurements. “Two counting channels were used, one fed with pulses from a pulse generator, and both were switched simultaneously in phase with the velocity of the source. The counts from the pulse generator were used to correct the counts from the proportional counter for inequalities of the time intervals. The results from two runs of counting were given, the source being always above the counter with an evacuated tube separating the two. In each run, cycles of four operations, including reversing the phase of the driving current in the transducer, were performed so that each part of the counting apparatus was used to count all four sets of pulses.”⁶²

The Harwell group made two experimental runs. “The average of these runs” the *Nature* article reported “was of the expected magnitude, but the fluctuations in the individual points were about twice as large as those expected from statistics.”⁶³

The experiments performed by Pound and Rebka differed in several respects. Their experiments had the benefit of a longer path – 22 meters, which was the full usable height of Harvard’s Jefferson Physical Laboratory. In addition, they had a radiation source that was more intense by a factor of 12.⁶⁴

⁵⁸ Hentschel notes that *The New York Times* article of 13 Dec 1959 discussed the work of Pound and Rebka; the *Observer* article in February 1960 focused on Schiffer and Marshall; Hentschel, (ref. 12), 273.

⁵⁹ Cranshaw and Schiffer, “Experiments to Test Einstein’s Principle of Equivalence,” *Nature* 185 (1960), 653-654.

⁶⁰ Hentschel, (ref. 12), 273.

⁶¹ Cranshaw and Schiffer, (ref. 59), 653.

⁶² Ibid.

⁶³ Ibid.

⁶⁴ Hentschel, (ref. 12), 273.

The experimental arrangement was also somewhat different. The Harvard source was “mounted on a piezo-electric transducer.” This transducer was “mounted on a hydraulic device” which could “produce velocities of the order of 0.0001 cm/sec for calibrating purposes. Two counting systems were used, one a sodium iodide crystal close to the source, the other a group of crystals to obtain a large area, operated at a vertical distance of about 75 ft.” In this case a “plastic bag containing helium was placed in the space between the source and the distant counters to reduce the absorption of the 14 keV γ -ray.”⁶⁵

As with the Harwell experiments, the Harvard researchers took care to ensure the accuracy of their measurements. “The transducer was operated sinusoidally, and the counts from the two counting-channels were switched into two pairs of counting systems by mercury switches, so that the pulses obtained with the source moving up were counted separately from those obtained with the source moving down. A dead time was provided to eliminate counts obtained during the switch-over period. The source on its transducer and the large counter were interchanged at intervals so that the expected sign of the asymmetry was changed.”⁶⁶

The March article noted that in initial experiments, the Harvard group measured “the asymmetries ... in the two counters in a series of two-hour runs taken in the six days previous to the April APS meeting.” Results, which Pound called “preliminary,” indicated that the “magnitudes of the asymmetries in the two counters fluctuated by larger amounts than could be accounted for by statistical fluctuations, but if averages were taken over all the runs, the measured difference in asymmetry between the counters was of the right magnitude.”⁶⁷

Cranshaw and Schiffer concluded their March article with a brief description of measurements with a rotating system. “The apparatus consisted of a source of iron-57 deposited on an iron cylinder 0.8 cm in diameter, clamped rigidly between two circular ‘Dural’ plates of 6 in. diameter. At a radius of 6.6 cm. near the circumference of the plates, a ‘Perspex’ strip was fixed carrying an iron-57 absorber.” The rotation came from a rotor from a neutron chopper. Schiffer had obtained this equipment by teaming up with Hal Hay and others from Harwell’s neutron chopper group. With this rotor “the whole system was rotated at speeds up to 500 cycles per sec., giving accelerations of 65,000 times terrestrial gravity. Any shift in the energy of the γ -rays would be observed as a decrease in their absorption, that is, by an increase in the counting rate.” In the end the group had impressive results: “The observed change in counting rate as a function of angular velocity followed the variation predicted by the theory of relativity with approximately 10 per cent accuracy due to counting statistics.”⁶⁸

By the time of the March publication – and before the April publication of Pound and Rebka – Cranshaw and Schiffer were aware of a potential problem with their experiment thanks to advice from an unexpected source. On 27 February Schiffer received a hand-written letter from a Cambridge undergraduate who “was very interested” in the red shift experiments but

⁶⁵ Cranshaw and Schiffer, (ref. 59).

⁶⁶ Ibid.

⁶⁷ Ibid.

⁶⁸ Ibid., Schiffer, (ref. 25).

noted that “there may be an additional frequency shift which must be taken into account, which is indirectly due to the reduction in mass of a nucleus when it emits a γ -ray.” He included some calculations to back up his claim, and humbly enough, a self-addressed, stamped return envelope for a response. In the young man’s words: “The relative shift for iron is 7×10^{-13} , and changes by 2×10^{-15} per $^{\circ}\text{C}$. This would mask the relativity effect if the temperatures of the emitter and absorber differed by more than a fraction of a degree. In addition there will be a small effect caused by the differential internal energy of the materials of the emitter and absorber, which enters into expression for the shift.” As Schiffer later summarized, the young man was pointing to “the temperature shift,” caused by “a second-order Doppler shift because of thermal motion.”⁶⁹

Schiffer remembers going “directly to Walter Marshall.” Marshall “immediately saw the point” and reacted by “hitting his forehead” and wondering “how he could have been so stupid to overlook this.” Marshall subsequently invited the clever undergraduate to visit them. To Schiffer the young man who emerged from the “shiny official car” provided by Marshall “looked extremely young ... about 16.” The young man was Brian Josephson, who would win the Nobel Prize in 1973 for the discovery of tunneling supercurrents. At the suggestion of Marshall and Schiffer, Josephson recorded his ideas about the temperature shift in an article to *Physical Review Letters*.⁷⁰

Josephson’s article appeared in the same April volume that carried Pound and Rebka’s follow-up to the measurements described in *Nature*. After a 10-day run the Harvard physicists confirmed the predicted gravitational red shift within a 10% error limit. Although their results were similar to those of their Harwell colleagues, Pound and Rebka insisted that “no conclusion can be drawn from the experiment of Cranshaw et al.” Their concerns were similar to those expressed by Josephson. They saw two sources of systematic error: “the temperature difference between the source and the absorber” and “the frequency difference inherent in a given combination of source and absorber.” As they pointed out: “only 0.6 $^{\circ}\text{C}$ of the temperature difference would produce a shift as large as the whole effect observed.” To avoid problems, Pound and Rebka inverted the position of the absorber and emitter in the course of measurements.⁷¹

Red shift measurements were an open question for the next several years. Pound published results in late 1960 with optimized equipment and obtained an error margin that was again about 10%. In 1964 Cranshaw and Schiffer published measurements and a more exact error analysis. While they revised their error margin to 20%, they defended their earlier results insisting that they had controlled temperature sufficient so that “a probability can be assigned to

⁶⁹ J. B. Josephson to Schiffer, 27 Feb 1960; Schiffer, (ref. 25). The author thanks Schiffer for providing this document.

⁷⁰ Schiffer, (ref. 25). Josephson, “Temperature-Dependent Shift of γ -rays Emitted by a Solid,” *Phys. Rev. Lett.* 4 (1960), 341-342. Josephson’s Nobel lecture can be found in Stig Lundqvist, ed. *Nobel Lectures, Physics 1971-1980* (Singapore, 1992.)

⁷¹ Pound and Rebka, “Apparent Weight of Photons,” *Phys. Rev. Lett.* 4 (1960), 337.

the validity of the assumption that a serious temperature difference did not exist.”⁷² All doubts about the confirmation of the red shift were extinguished in 1965 with measurements taken by Pound and Joseph Lyons Snider. This time the 14.4 keV transition was measured with an even more intense source – 1.25 curies of ^{57}Co – and with even more sophisticated techniques and data analysis. The new measurements confirmed the existence of the red shift within an error of less than 1%.⁷³

2.5 EXPANSION

The red shift measurements joined a flood of early work with Mössbauer spectroscopy. In June 1960 – the same month as the first comprehensive theory of the Mössbauer effect was published by Singwi and Sjölander – there was enough work to prompt Hans Frauenfelder to convene the first in a what would turn out to be a series of Mössbauer conferences that would reach into the 21st century.

Frauenfelder, in fact, organized the meeting in a mere five weeks, saying in the conference proceedings that the justification for the “hurry ... can be found in the importance and the rapid increase of experiments based on the Mössbauer effect.” Even though conference planning was complicated because he had broken his leg skiing, Frauenfelder was convinced to organize the conference without delay, feeling that “a direct exchange of information, while the field was still in the state of rapid expansion, would yield new ideas through cross fertilization of nuclear and solid state physics, and hopefully would prevent unnecessary duplication,” and in addition, “provide the contract granting agencies with a clearer impression of the possibilities and limitations of this new method.” To accommodate Frauenfelder’s limited mobility, the meeting was held at a location he could reach by car, an old estate-turned-conference-facility called “Allerton House,” about 20 miles from Urbana-Champaign where Frauenfelder taught.⁷⁴

Although the location was remote for some – and despite the late invitation – almost everyone who was invited came. The 90-member list of participants included those who were or were to become Nobel laureates (John Bardeen in addition to Mössbauer himself, who by then was working at Cal Tech). Also present were most of the researchers who had made early, major contributions.⁷⁵

In addition to discussion of the red shift work, talk at the conference focused on Mössbauer spectroscopy in nuclear and solid state physics. The proceedings report plenty of

⁷² Cranshaw and Schiffer, “Measurement of the Gravitational Red Shift with the Mössbauer Effect,” *Proc. Phys. Soc.* 84 (1964), 245-56. Pound and Rebka, “Variation with Temperature of the Energy of Recoil Free Gamma Rays from Solids,” *Phys. Rev. Lett.* (1960), 274-275. Hentschel gives a more complete account of this part of the story, (ref. 12), 276-277.

⁷³ Pound and Snider, “Effect of Gravity on Gamma Radiation,” *Phys. Rev.* B140 (1965), 788-803.

⁷⁴ Frauenfelder and Harry Lustig, “Mössbauer Effect: Recoilless Emission and Absorption of Gamma Rays,” Allerton House, University of Illinois (1960), iii. Frauenfelder, interview with Westfall, 26 Nov 2003.

⁷⁵ Frauenfelder and Lustig, *ibid.*, 73-76.

lively discussion and controversy. Frauenfelder remembers that the conference was “exciting.” James Mullen, who would later set up the Mössbauer lab in the Solid State Science Division at Argonne, attended as a graduate student. After retirement, he judged it to be one of the best conferences of his entire career.⁷⁶

Although the large laboratories (Argonne, Brookhaven, Los Alamos, Harwell) dominated the Allerton conference – and indeed, yielded most of the early, major contributions -- Mössbauer noted in his concluding remarks that large laboratories would not have a monopoly with the new spectroscopy (remarks that Frauenfelder would echo, as previously quoted, in 1962). In Mössbauer’s words: “This field is a fine example of a branch within experimental physics which does not require the application of much equipment, a fact which allows even the smaller research institutions to compete with large, well-equipped laboratories.”⁷⁷ The move to smaller institutions was accelerated because very quickly radiation sources – which had previously been available only to those with access to specialized equipment such as a large accelerator -- were purchased commercially. By the third Mössbauer conference, held in September 1963 in New York, the new trend was apparent. Of the over 213 participants, many came from universities (even some small colleges) and private corporate research laboratories.⁷⁸

The rapid expansion that prompted the Allerton conference was destined to continue. Participants came to the 1963 conference from 15 countries, including Japan and the Soviet Union, where Mössbauer spectroscopy was booming. The 1963 conference also sported a bibliography with over 600 entries, an amazing increase over the 52 bibliographic entries listed in the Allerton conference. (The 52 entries in June 1960 was itself an impressive increase, considering that in 1958, only Mössbauer was publishing on the subject.) By 1963, as the proceedings explained, work was being done in “physics, chemistry, and biology.”⁷⁹

Although smaller institutions became increasingly involved in Mössbauer spectroscopy after 1960, large institutions retained advantages that facilitated cutting edge research. In the words of Dean Taylor, who spent much of his career working on Mössbauer spectroscopy at Los Alamos, “the large laboratories never stopped being active participants in the development of Mössbauer spectroscopy.” Hentschel gave an example of this phenomenon in his study of the gravitational red shift experiments from 1959 to 1965. To explain why others besides Harwell and Harvard researchers did not “enter the race,” Hentschel explained: “to reduce systematic errors sufficiently it was indispensable to have strong and chemically pure sources of γ radiation: in the case of Schiffer and his team it involved exposing iron plates in the Birmingham cyclotron and subsequent chemical treatment by the Radiochemical Centre in Amersham. Pound and Rebka were provided with similar services by Oak Ridge National Laboratory, the Nuclear Science and Engineering Corporation and by Nuclear Metals Inc.” Having access to such resources was an important advantage during this period for all Mössbauer spectroscopy. In

⁷⁶ Frauenfelder, interview with Westfall, 26 Nov 2003, private communication, Jim Mullen, 6 Nov 2003. Ibid.

⁷⁷ Frauenfelder and Lustig, “Mössbauer Effect: Recoilless Emission and Absorption of Gamma Rays,” Allerton House, University of Illinois (1960); Dean Taylor, interview with Westfall, 30 Oct 2003.

⁷⁸ Alan Bearden, “Third International Conference on the Mössbauer Effect,” *Rev. Mod. Phys.* 36 (1964), 333-503.

⁷⁹ Ibid., 333.

particular, “the presence of a cyclotron or of a nuclear reactor was most convenient,” because such equipment was needed for “the preparation of strong sources.”⁸⁰

In the next five decades Mössbauer spectroscopy continued using radioactive sources. This “traditional” form included a variety of exotic applications, including the palm-sized Mössbauer spectrometer on the Mars Exploration Rovers. This spectrometer began analyzing soil and rock samples in early 2004⁸¹ (Figure 7).

Starting in the 1970s, researchers began exploring the feasibility of performing Mössbauer spectroscopy using synchrotron light sources. Although the idea was first conceived at Argonne by Stanley Ruby, Erich Gerdau was the first to successfully demonstrate this application in the mid-1980s at the German laboratory, DESY.⁸²

This new form of Mössbauer spectroscopy offered many advantages for those wanting a more complete picture of the properties of matter.⁸³ For one thing, synchrotron radiation based sources were a million times brighter than radioactive sources, opening the way to measure microscopically small samples, weighing only a few nanograms. Second, the incident energy was tunable over a large range, covering not only hyperfine interactions but also the entire phonon range and molecular vibrations. Third, synchrotron radiation is polarized linearly, and can easily be manipulated to be polarized circularly of either handedness (left or right) with the help of diamond quarter-wave phase plates. This, in turn, provides an avenue to measure element and isotope selective magnetization from buried monolayers in magnetic thin films. Finally, synchrotron radiation is pulsed with a pulse width less than 100 ps, and a separation of a few hundred nanoseconds or more. Thus, it is possible to study nanosecond time resolved phenomenon. All these properties were combined in later experiments.⁸⁴

This powerful new Mössbauer spectroscopy opened up new possibilities for illuminating the composition of materials. Those using this new tool were particularly interested in exploring

⁸⁰ Taylor, interview with Westfall, 30 Oct 2003; Hentschel, (ref. 12), 273-274.

⁸¹ “Mössbauer Spectroscopy Newsletter: Two Working Mössbauer Spectrometers on the Surface of Mars,” J. G. Stevens, et al. eds., *Mössbauer Effect Reference and Data Journal*, 27 (2004), 41-44.

⁸² E. E. Alp et al. “Nuclear Resonant Scattering Beamline at the Advanced Photon Source,” *Hyperfine Interactions* 90 (1994), 323-34. E. Gerdau et al., “Nuclear Bragg Diffraction of Synchrotron Radiation in Yttrium Iron Garnet, *Phys. Rev. Lett.* 54 (1985), 835-838.

⁸³ The new spectroscopy, which used x-rays, was not identical to the spectroscopy done with radioactive sources, which used gamma rays. One difference is that gamma rays from nuclear decay are unpolarized, while x-rays produced from light sources are polarized. However, gamma rays from nuclear decay are in many ways similar to x-rays produced in light sources, and everything (and more) that can be measured with traditional Mössbauer spectroscopy, that is, the hyperfine parameters of isomer shift, quadrupole splitting and magnetic hyperfine field strength, can be measured with light source Mössbauer spectroscopy. Therefore those using light source Mössbauer spectroscopy consider that their work continues the intellectual tradition begun with Mössbauer spectroscopy using radioactive sources.

⁸⁴ Caroline L’abbe, et al., “Nuclear Resonant Magnetometry and its Application to Fe/Cr Multilayers,” *Phys. Rev. Lett.* 93 (2004) 037201-037204 and private communication, Alp, 27 July 2004.

FIGURE 7 Mars Rover Spectrometer (NASA photo)

the diffraction of nuclear-resonant gamma rays by single crystals, which had no counterpart in standard diffraction theory due to unusual characteristics of nuclear resonance, namely, the extremely long lifetime of the nuclear excited state compared to atomic vibrations by several orders of magnitude. Indeed, the first observation of accelerated nuclear decay rate upon coherent diffraction created much excitement for physicists working with x-ray scattering. In addition, the elegant re-formulation of dynamical theory in the presence of nuclear excitation allowed the first meaningful generalization of multiple scattering theory of photons in many decades. These early experiments led to even more sophisticated ideas, like the arrest of photons during Bragg diffraction, and re-release of them by means of external switches of acoustic or magnetic fields.⁸⁵

While the coherent diffraction from a nuclear resonant medium was the main motivation for researchers at the point, the measurement of phonon excitations using the very narrow energy width of the Mössbauer isomers was also of great interest, since such excitations had long been mystifying. As had previously been the case, a variety of new technological advances converged, including improvements in silicon purification and fast silicon diode based detectors, as well as the development of the last generation of undulator based synchrotron sources. These combined advances made possible the first successful observation of phonon density of states. This observation, in turn, opened up the first real unification of neutron-based lattice-dynamics measurements with x-ray techniques. Furthermore, it generated a renewed interest in the dynamics of biologically important organic and inorganic compounds like proteins, porphyrins and enzymes, as well as geologically important parameters like sound velocity, and elastic modulus at extremely high-pressures of a few megabar, and high temperatures up to 3000 K, which mimics the conditions of earth's mantle and outer-core.

⁸⁵ This work is comprehensively reviewed in the two-volume set, Gerda, H. de Waard, eds., "Nuclear Resonant Scattering of Synchrotron Radiation," *Hyperfine Interactions*, 123/124 (1999).

Another motivation for using synchrotron radiation was to observe those nuclear resonances for which there was no suitable parent isotope, thus filling gaps conditions that previously could not be filled in the expanding knowledge of materials under extreme. For example, while iron can be a model for transition metals with incomplete 3d-shells, one needs a counterpart in 4d elements (Sn), in rare-earth elements (Eu, Dy, Sm), noble gases (Kr) or biologically relevant atoms like K. Some of these elements, like Kr or K, do not have convenient parent isotopes, and the science that requires observation of these elements could not have been carried out. This work allowed study of the dynamics of noble gases at high pressure and temperatures and allowed researchers to take advantage of the chemical inertness of Kr as a tag element in a complex medium like proteins.

As is typical in scientific investigations, advances in knowledge led to the pressure to advantage technology and further advances in technology further advanced knowledge. For example, the motivation to observe as many elements as possible inevitable led to isotopes with high nuclear transition energies like Ni at 67 keV, for which there is no known method of providing tunable high-resolution monochromatization. Such work prompted the development of exact back-scattering monochromators and stimulated the development of full-formalism of dynamical diffraction under exact Bragg backscattering conditions, which had previously been poorly understood.

At the turn of the 21st century, Mössbauer measurements were made at dedicated beam lines at Argonne's APS, Spring-8, near Osaka, Japan, and the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. Earlier work was performed mainly at a bending magnet beam line at HASYLAB on the DORIS III ring, and the wiggler beam lines at the Stanford Synchrotron Radiation Laboratory (SSRL), Stanford, California, at Cornell High Energy Synchrotron Source (CHESS), Ithaca, New York, and the National Synchrotron Radiation Facility (NSLS) Brookhaven National Laboratory, Upton, New York. These sources join the x-ray beam line on the PETRA ring at DESY, Hamburg, Germany and the undulator sources at the KEK Accumulator Ring in Tsukuba, Japan.⁸⁶ The primary purpose of the APS, ESRF, and Spring-8 was to measure molecular vibrations in biological molecules, including proteins as well as thermodynamic and elastic properties of materials under extreme pressure and temperatures (one megabar and 3000 K.)⁸⁷

By 2000 Mössbauer spectroscopy was a well-established, vibrant scientific enterprise. From 1960 to 2000, Mössbauer spectroscopy had more than 50,000 publications covering a large range of fields (Figure 8) and a wide variety of conferences.⁸⁸

⁸⁶ E. E. Alp, W. Sturhahn, and T. Toellner, "Synchrotron Radiation and the Mössbauer Effect: Recent Developments," Stevens, et al., eds., *Mössbauer Effect Reference and Data Journal*, 22 (1999) 167-170.

⁸⁷ Toellner et al., "Polarizer/analyzer Filter for Nuclear Resonant Scattering of Synchrotron Radiation," *Appl. Phys. Lett.* 67 (1995), 1993-1995 and private communication, Alp, 27 July 2004.

⁸⁸ More statistical information about Mössbauer spectroscopy can be obtained from John Stevens, Mössbauer Effect Data Center, North Carolina State University, Raleigh, North Carolina, 27695.

- Nuclear and Solid State Physics
- Biology
- Chemistry
- Materials Science
- Geology
- Archeology
- Forensic Science
- Paleontology
- Mineralogy
- Metallurgy
- Space Science (on Mars Exploration Rovers)

FIGURE 8 List of Fields that Use Mössbauer Spectroscopy

3 MÖSSBAUER SPECTROSCOPY AT ARGONNE

3.1 SMALL-SCALE SCIENCE IN A BIG LABORATORY: THE FIRST GENERATION OF MÖSSBAUER SPECTROSCOPY IN THE PHYSICS DIVISION IN THE EARLY 1960S

When Mössbauer spectroscopy got its start in the early 1960s, Argonne had (among other large machines) a 60-inch cyclotron operated by the Chemistry Division and the CP-5 research reactor operated by the laboratory's reactor program. In line with Hentschel's observation, the cyclotron was used to produce the strong, chemically pure γ -ray source (^{57}Co) which allowed researchers in Argonne's Physics Division to be a dominant force during this era. The reactor was also sometimes used to make such sources, and it came in handy in other ways as well. For example, Hanna, one of the leaders of the Mössbauer spectroscopy effort, later explained that they decided to "test a hunch" of Perlow "that a nonmagnetic atom combined with a magnetic one would become endowed with magnetic properties." In the 1960 Hanna and the others chose "neutron-irradiated tin containing metastable ^{119}Sn to use as a nonmagnetic source." Although the isotope would have been difficult to find elsewhere "such activity can inevitably be found stored around a reactor laboratory such as Argonne."⁸⁹

The Argonne experience also shows that large instruments such as reactors and accelerators were hardly the only special resources available for Mössbauer spectroscopy in the early 1960s at large laboratories. The Physics Division itself had much to offer. To get a jump start on the first experiments, Hanna asked Physics shop's foreman, Bruno Martinka, for the use of his most trusted lathe to rotate the source precisely. As time went on and detection equipment became more sophisticated, Argonne's Mössbauer spectroscopists benefited from detector specialists. For example, the Division's detector group made the thin NaI crystals needed to detect the low-energy γ -rays and with the help of this group, physicists devised instrumentation improvements, such as Perlow's 200-channel pulse-height analyzer. The group found it easy to scrounge small, surplus magnets useful for other parts of the experiment. They also relied on experts from other parts of the laboratory. In addition to obtaining radioactive sources from the Chemistry and Reactor Engineering Divisions, they drew assistance from the Metallurgy Division, where Frank Karasek expertly rolled the necessary very thin foils of enriched ^{57}Fe metal.⁹⁰

In addition to skilled hands and specialized machinery, Argonne physicists benefited from a wide array of intellectual resources. Gil Perlow's work with xenon compounds provides a vivid example of how the opportunities available to national laboratory scientists – including collaboration – could be used to great advantage.

⁸⁹ Hanna, "The Early Iron Age of the Mössbauer Era," *Hyperfine Interactions* 90 (1994), 3-20. For more on CP-5, see Jack M. Holl, *Argonne National Laboratory* (University of Illinois press, 1997), 116-117.

⁹⁰ Hanna, *ibid.*, 6; Gil Perlow, "Presentation for Mössbauer History Day," 1 Oct 2003.

Perlow remembers that his work on xenon compounds began as the result of an encounter with Argonne colleague, Marianne Schiffer, a structural biologist who told him about “some remarkable” work being done at the laboratory’s chemistry department, as well as a few other places, such as Lawrence Berkeley Laboratory. At Argonne: “John Malm, Cedrick Chernick, and associates had synthesized a number of compounds of xenon, in particular XeF_6 , XeF_4 , and XeF_2 as stable crystals, and XeO_3 as an explosively unstable polycrystalline material.”⁹¹ The Argonne physicist was amazed, because it had previously been thought that xenon was an inert gas that was completely unreactive chemically and therefore could not form compounds.⁹²

Perlow got the idea of using Mössbauer spectroscopy when he found that “the isotope table shows that ^{129}Xe and ^{131}Xe has low lying level structures suitable” for the technique. Argonne’s Chemistry Division was doing cutting edge work that dovetailed perfectly with the cutting edge Mössbauer spectroscopy being done next door in the Physics Division.⁹³

Perlow would not have to perform the specialized work alone. In addition to the expertise and exotic materials provided by Malm and his colleagues in the Chemistry Division, Perlow also benefited from an active collaboration with those outside Argonne. Argonne and Harwell had enjoyed an exchange of personnel for many years and an especially fruitful collaboration in Mössbauer spectroscopy that began at the time of the prestigious gravitational red shift experiments Schiffer performed as a visitor to Harwell. Like Schiffer, Perlow got help from Walter Marshall. Before the xenon measurements, in fact, Perlow had spent a year at Harwell where he learned to be, in his own words, “a materials scientist and theoretical chemist” thanks to working with the eminent solid state theorist. At Harwell Perlow worked with Charles Johnson, an exacting Mössbauer spectroscopist. Johnson, who had come to Argonne for a visit, would help make a variety of xenon measurements.⁹⁴

Perlow also had a secret weapon: his wife, Mina Rea. Mina Rea Perlow had a Ph.D. in chemistry from the University of Chicago. Although she was not paid, she had been aiding her husband’s efforts by making radioactive separations as needed – for example, she had helped in the preparation of ^{57}Co for the earlier Argonne work with Hanna. Mina Rea did the synthesis of the xenon compounds, a job that her husband later judged to be “specialized, elegant, and essential.”⁹⁵

In a paper in *Chemical Physics*, Perlow described the experimental procedure: “The technique is the Mössbauer effect of the 40-keV gamma ray emitted in the transition from the first excited state to the ground state of ^{129}Xe , following the beta decay of ^{129}I . The iodine is

⁹¹ Ibid., Perlow. et al., “Xenon Tetrafluoride,” *J. Am. Chem. Soc.* 84 (1962), 3593.

⁹² Private communication, Perlow, 26 Apr 2002.

⁹³ Perlow (ref. 90).

⁹⁴ Ibid.

⁹⁵ Ibid.; private communication, Perlow, 11 Nov 2004. C. L. Chernick, Johnson, Malm, Perlow and M. R. Perlow, “The Mössbauer Effect in Chemical Compounds of Xe^{129} ” *Physics Letters*, 5 (1963), 103-104; Perlow and M. R. Perlow, “The Mössbauer Effect in Xenon Compounds,” *Rev. Mod. Physics* 30 (1964), 353-357.

contained in $^{129}\text{KICl}_4 \cdot \text{H}_2\text{O}$. The resonant absorber is xenon ... as the hydroquinone clathrate. The experiments are done at 4.2°K.”⁹⁶

Later Perlow further explained: “In a sense, xenon makes the simplest of compounds, because the bonding is due to charge leaving the closed shells 6p and 6s,” which simplified the interpretation of results. “Mina Rea synthesized several sources containing beta-emitting ^{129}I in negative ions like IO_3^- and IO_4^- . They could be run against xenon contained in a clathrate, a sort of organic cage, forming an unsplit absorber. The IO_3^- yielded a velocity doublet, which had the same splitting as XeO_3 .” Noting this fact, the Perlows were able to use Mössbauer spectroscopy to examine XeCl_2 and XeCl_4 , produced by the decay of ^{129}I , by way of the analogues of the two compounds. “It was clear then,” in Perlow’s words “that in the process of beta decay the newly formed xenon atom captured its nearest neighbors and formed a compound.”⁹⁷

In a *Scientific American* article summarizing the application of Mössbauer spectroscopy to chemistry, R. H. Herber noted that by using this procedure, the Perlows were able to deduce “that the structure of XeCl_4 is square-planar (square and flat), whereas the structure of XeCl_2 is linear.” Although the second result was not surprising, the first result was news since “one might have thought that the true molecular state of XeCl_4 was tetrahedral.”⁹⁸

As Herber explained, with this work the Perlows elucidated “the structure of a compound that has not yet been synthesized in the laboratory by ordinary chemical means.” In his judgment, this work was “one of the most remarkable achievements of Mössbauer-effect spectroscopy.”⁹⁹

Although Mössbauer spectroscopy was breaking new ground in science, the first generation of Mössbauer spectroscopists worked at the same scale as experimentalists of the previous century. Their work contrasted, in particular, with that of their colleagues at Argonne’s High Energy Physics Division, who in 1965 began working with the \$50 million ZGS, which took 10 years to fund, design, and construct¹⁰⁰ (Figure 9). Although Argonne Mössbauer spectroscopists reaped the benefit of well-designed, expertly crafted instruments thanks to the supply of experienced technicians and well stocked shops that also facilitated ZGS experiments, Mössbauer spectroscopists in the early 1960s – unlike their high energy physics colleagues – mostly used simple equipment and services already at hand (Figure 10). Therefore, their costs were minimal – when something had to be paid for, it came from Physics Division director Mort Hamermesh’s discretionary budget. Experiments were also quickly implemented. Hanna and Perlow both remember discussing innovations at lunch in the Argonne cafeteria, then making the

⁹⁶ Perlow and M. R. Perlow, “Mössbauer Effect Evidence for the Existence and Structure of XeCl_4 ,” *Chemical Physics* 41 (1964), 1157-1158.

⁹⁷ Perlow, (ref. 90).

⁹⁸ R. H. Herber, “Mössbauer Spectroscopy,” *Scientific American* 225 (1971), 86-95.

⁹⁹ Ibid., 92.

¹⁰⁰ Paris, (ref. 4), 18; Holl, (ref. 89), 210-212. The Paris monograph provides a complete history of the development of the ZGS design, including the factors that led to the choice of a weak focusing design.

FIGURE 9 Aerial View of the Zero Gradient Synchrotron (Argonne, Slide #62c-20)

improved measurements later that day. The Mössbauer measurements of the time were, in the words of team member Carol Littlejohn (later Herzenberg) really “cheap-o” experiments. In the shadow of big physics at Argonne, they were doing “really little, little science.”¹⁰¹

3.2 NO BIGGER OR BETTER THAN IT NEEDED TO BE: THE SECOND GENERATION OF MÖSSBAUER SPECTROSCOPY AT ARGONNE IN THE MID-1960S-1970S

In the mid-1960s Mössbauer spectroscopy at Argonne entered a new phase that would provide another example of the development of small-scale research at a U.S. national laboratory. This new phase got started when Michael Kalvius was recruited to the Solid State Science Division to replace James Mullen, who had set up a Mössbauer spectroscopy laboratory

¹⁰¹ Carol (Littlejohn) Herzenberg, interview with Westfall, 11 Nov 2002. Hanna, (ref. 44); private communication, Perlow, 26 Apr 2002.

FIGURE 10 Gil Perlow, Charles Johnson, and John Oyler (Argonne, #201-5306)

but then left Argonne to take a professorship at Purdue. Kalvius came to Argonne from Case Reserve (later Case Western) in Ohio, but he got his start in Munich. Like Mössbauer, he was a student of Heinz Maier-Leibnitz; Kalvius did a thesis on Mössbauer spectroscopy just a few years behind Mössbauer.¹⁰²

By the time Kalvius came to Argonne in 1965, Hanna had left Argonne and had been replaced by a former Westinghouse physicist, Stanley Ruby (who would subsequently be the first to suggest using synchrotron light sources to do Mössbauer spectroscopy.) Argonne's Solid State Science Division included theorists Singwi and Sjölander, the ones who published the first comprehensive theory of the Mössbauer effect in 1960. Before the mid-1960s, however, the Solid State Science Division had not hosted a large effort in Mössbauer spectroscopy and experimentalists in the Physics and Solid State Science Divisions had not interacted much. This pattern changed when Kalvius and Ruby began working together. In the next two years the collaboration expanded to include others, including two young solid state physicists, Bobby Dunlap and Gopal Shenoy, who had earned his Ph.D. at the University of Bombay¹⁰³ (Figure 11).

Perlow's experience reveals that national lab physicists can get highly skilled next-door help; the Ruby-Kalvius-Dunlap-Shenoy collaboration showed the potential of intellectual cross-pollination when members of two national laboratory Divisions collaborate. By the mid-1960s, the team consisting of a nuclear-turned solid state physicist, a nuclear physicist from industry, and the two new, energetic and enthusiastic solid state physicists were in a prime position to build on Argonne's nuclear physics expertise to exploit the as yet unexplored nuclear physics as well as the rich solid state physics inherent in the hyperfine interactions that connect nuclear moments with solid state properties.

Their collaboration would also provide "a new turn" in Mössbauer spectroscopy at Argonne, as Shenoy noted and as division progress reports confirm. The unprecedented ultra-high resolution of Mössbauer spectroscopy gave solid-state physicists the opportunity to make a broad range of hyperfine structure measurements – Shenoy would remember later that they measured about 10,000 compounds. Solid-state physicists could add this hyperfine structure information, which Shenoy judged to be "the most important data point," to the mix of other data, such as measurements of electronic and magnetic properties and measurements of thermal and electrical conductivities, to piece together the vastly complicated picture of the magnetic, thermal, electronic, and mechanical properties of a large variety of materials. This new data was crucial because solid-state physicists depended upon a large quantity of high quality information to achieve the key goals of their field: delineating the basic properties of matter and finding

¹⁰²Michael Kalvius, interview with Westfall, 2 Oct 2003; private communication, Mullen, 6 Nov 2003.

¹⁰³B. D. Dunlap, "Actinide Research with Mössbauer Spectroscopy at Argonne National Laboratory," talk given at Mössbauer history celebration day at Argonne, 30 Sep 2003; "Physics Division Summary Report," 1 Apr 1967 – 31 Mar 1968, ANL-7481, p. 126; Gopal Shenoy, interview with Catherine Westfall, 30 Apr 2002; Kalvius, interview with Westfall, 2 Oct 2003.

FIGURE 11 Michael Kalvius, Gopal Shenoy, and Bobby Dunlap, about 1970

materials suitable for researchers in other fields as well as various industrial applications that they hoped would “revolutionize industrial science.”¹⁰⁴

As the stock of Mössbauer spectroscopy rose in the Solid State Science Division, it fell in the Physics Division, where many judged that the technique had already yielded the key insights of interest to nuclear physicists. Although Perlow, Ruby, and some other nuclear physicists continued using the technique, the mid-1960s marked the evolution, in Shenoy’s words, “from a discovery era” aimed primarily at exploring nuclear properties “using a few resonances in the Physics Division to an applications era using a large Mössbauer periodic table in the Solid State Science Division.” In the process: “the center of gravity moved” from the Physics Division, where work on Mössbauer spectroscopy dwindled, “to the Solid State Science Division,” where this work remained important through the 1970s.¹⁰⁵

¹⁰⁴Shenoy, interviews with Westfall, 30 Apr 2002, 31 May 2005. Shenoy, “Postdoctoral Research in the Solid State Science Division: A Personal Account,” talk given at Mössbauer history celebration day at Argonne, 30 Sep 2003.

¹⁰⁵Ibid., Shenoy. Shenoy, interview with Westfall, 31 May 2005. “Physics Division Summary Report,” 1 Apr 1967 – 31 Mar 1968, ANL-7481, p. 126; “Solid State Science Division Research Summaries,” 1 Oct 1971 – 30 Sep 1972; “Solid State Science Division Research Summaries,” 1 Oct 1974 – 30 Sep 1975, ANL-SSS-75-10; “Solid State Science Division Research Summaries,” 1 Oct 1977 – 30 Sep 1987, ANL-SSS-78-10.

The research pace increased: Shenoy would later say they had created a “Mössbauer factory” that produced a sort of “intellectual production line” of data that could be crafted into solid state theoretical as well as industrial solutions. Their production line included measurements from “nearly every known and newly discovered Mössbauer resonance – ^{57}Fe , ^{119}Sn , ^{121}Sb , ^{125}Te , $^{127,129}\text{I}$, ^{189}Pt , ^{191}Au , ^{149}Sm , $^{151,153}\text{Eu}$, $^{155,157}\text{Gd}$, ^{161}Dy , $^{166,168,170}\text{Er}$, ^{169}Tm , $^{170,171,172,174}\text{Yb}$, ^{193}Ir , ^{195}Pt , ^{197}Au , $^{234,238}\text{U}$, ^{237}Np , ^{240}Pu , and ^{243}Am .” As had been the case in the early 1960s, Argonne provided an impressive array of equipment and services. The CP-5 reactor prepared a wide variety of sources, including those as hot as many Curies for ^{166}Er and ^{170}Yb . The laboratory’s superior computing resources also came in handy: Kalvius remembered that “only the big laboratories had powerful computers.” Argonne’s computers, in fact, “made modern Mössbauer data analysis possible for the first time.” A particularly key computing component, in Shenoy’s opinion, was the state-of-the-art analyzers from which data spewed directly to IBM cards. “Thanks to Bob Whitman from Electronic Division, all these analyzers were kept in working condition.”¹⁰⁶

The second generation of Mössbauer spectroscopists, like their predecessors, also enjoyed the advantage of working in a laboratory with bountiful detection expertise and equipment. As Shenoy later noted: “It was not unusual to have four spectrometers simultaneously measure four distinct resonances on any given day. Measurements were routinely done from 1.5K to a few hundred degrees K and magnetic fields up to 5T. Photon counting was done using NaI, Li drifted Ge, and proportional counters. With hot sources for ^{166}Er , ^{170}Yb and ^{237}Np , the transmitted current was measured rather than single photons, and as a result high quality spectra were measured in minutes rather than days.” Argonne’s detection equipment was augmented by a high precision electro-mechanical device (double loudspeaker system) particularly well suited for feeding Doppler motion into a cryostat that Kalvius had brought with him.¹⁰⁷

Although Argonne researchers had access to this unusually wide array of equipment, the apparatus they used for Mössbauer spectroscopy was modest in size and expense – far closer to the size and scale of Perlow’s work than that of ZGS researchers. Kalvius would later estimate that their lab cost about \$100,000 to set up. As Dunlap explained: “Doppler drives were sometimes held in place on Dewar stands with C-clamps and soldered to drive rods: simple but effective techniques. Some Doppler drives were commercial, but in my first years at Argonne they were as likely as not to be handmade from stereo loudspeakers.” In short: “The equipment was good, but not better or bigger than it needed to be”¹⁰⁸ (Figure 12).

¹⁰⁶Ibid. Kalvius to Westfall, 17 Aug 2004. “Solid State Science Division Research Summaries,” 1 Oct 1971 – 30 Sep 1972; “Solid State Science Division Research Summaries,” 1 Oct 1977 – 30 Sep 1987, ANL-SSS-78-10. For more on computing at Argonne, see Charles Yood, “Argonne National Laboratory and the Emergence of Computer and Computational Science, 1946-1992,” Ph.D. dissertation, Pennsylvania State University, 2005.

¹⁰⁷Shenoy (ref. 104). Kalvius, interview with Westfall, 29 Sep 2003. “Solid State Science Division Research Summaries,” 1 Oct 1974 – 30 Sep 1975, ANL-SSS-75-10

¹⁰⁸Dunlap (ref. 103). Kalvius, interview with Westfall, 29 Sep 2003.

FIGURE 12 Gopal Shenoy with Mössbauer Spectroscopy Equipment about 1970

With the resources at their disposal, the second generation of Mössbauer spectroscopists produced new scientific knowledge. Scientific highlights included the measurement of the electronic and magnetic properties of actinide compounds (Np, Am, U, Pu), rare earth compounds (Er, Dy, Eu, Tm), and compounds of the 5s-5p elements (Sn, Sb, Te, I, Xe).¹⁰⁹ The actinide research of the period provides a particularly dramatic example of how Mössbauer spectroscopy could be exploited at Argonne.

As had been the case with xenon compounds, actinides were ripe for exploration; as Shenoy put it, they were “hugely interesting because their physics was totally different than anything else.” As Dunlap and Kalvius further explained in a 1985 textbook, the actinides “form a series of elements with unique physical and chemical properties. This special behavior is due to their unusual electronic structure which combines features of the 3d transition series (the iron group) with those of the 4f transition series (the rare earths or lanthanides).” The behavior of the actinides was so mysterious, in fact, that uranium and thorium, which were the first to be studied because they had isotopes with long enough half-lives to be found in nature, were classified as 6d elements based on their chemical behavior. It was only when neptunium and plutonium isotopes of sufficient quantities were produced for detailed study that researchers understood that the 5f orbitals of these actinides were occupied.¹¹⁰

¹⁰⁹For more details, see Shenoy (ref. 104). “Solid State Science Division Research Summaries,” 1 Oct 1971 – 30 Sep 1972; “Solid State Science Division Research Summaries,” 1 Oct 1974 – 30 Sep 1975, ANL-SSS-75-10; “Solid State Science Division Research Summaries,” 1 Oct 1975 – 30 Sep 1976, ANL-SSS-76-10; “Solid State Science Division Research Summaries,” 1 Oct 1977 – 30 Sep 1987, ANL-SSS-78-10.

¹¹⁰Shenoy, interview with Westfall, 30 Apr 2002; Dunlap and Kalvius, “Mössbauer Spectroscopy on Actinides and their Compounds,” in A. J. Freeman and G. H. Lander, eds. *Handbook of the Physics and Chemistry of the Actinides*, Vol. 2 (North-Holland, 1985), 331-434.

As Dunlap later noted, the “scientific complexity” of the actinides means that they are unlike the rare earths which demonstrate “strong 4f electron localization” that “allows one (in most cases) to treat the valence electrons as atomic-like.” They are also unlike the transition metal systems in which “d electrons are strongly delocalized and can be treated in first approximation with a free-electron model.” Instead, the actinides “show intermediate localization where neither of these extreme approximations work.”¹¹¹

The scientific complexity of the actinides are further complicated because “spectroscopic studies show that various energies determining the ground state of an actinide ion (spin-orbit coupling, crystal field energy, exchange energy) are often of comparable size. This means that various approximations commonly used for d transition compounds or 4f lanthanides do not apply.”¹¹²

In Dunlap’s opinion, thanks to these complications the actinides “display a fascinating and confusing array of phenomena which are scientifically rich and theoretically challenging.” For one thing, in contrast to rare earths, which are mainly trivalent states, in actinides a “large number of valence states may be seen. For example, Np occurs in all charge states from Np^{3+} to Np^{7+} .” In addition: “Active bonding of 5f electrons occurs in chemical systems,” and “there are a large variety of magnetic effects in metallic systems, ranging from localized to delocalized electronic behavior.” Finally: “Mixed valence and ‘heavy fermion’ behavior is well-known in metallic systems.”¹¹³

Again, Mössbauer spectroscopy was the tool of choice. As Dunlap and Kalvius explained, the technique had “distinct advantages” which “make it a particularly useful technique for the investigation” of actinide compounds, which are hard to obtain in high quantities. “Only comparatively little sample material is needed (about 10 to 100 mg suffices). Single crystals are not required.” Also, when “compared with bulk measurements like transport properties or magnetic susceptibility, the Mössbauer effect proves to be relatively insensitive to sample purity or radiation-induced lattice defects,” a problem inherent in actinide compounds. Also, for basic magnetic measurements, “Mössbauer spectroscopy is easily adapted to special environmental conditions such as applied high pressure or strong magnetic fields.” With Mössbauer spectroscopy it is also “possible to determine unambiguously the charge state of the resonant atom or ion in compounds.”¹¹⁴

Argonne’s second generation of Mössbauer spectroscopists measured resonances in a long list of actinide isotopes: 84.2 keV in ^{231}Pa , 43.5 keV in ^{234}U , 45.3 keV in ^{236}U , 44.7 keV

¹¹¹Dunlap (ref. 103).

¹¹²Ibid.

¹¹³Ibid. The term “heavy fermion” is used in condensed matter physics to describe metallic materials having very large electronic mass enhancement arising from an antiferromagnetic interaction between conduction electrons and local magnetic moments (d- or f-electron type) residing on a sublattice of atoms in the metal.

¹¹⁴Ibid., Dunlap and Kalvius, (ref. 110), 332.

in ^{238}U , 59.6 keV in ^{237}Np , 57.3 keV in ^{239}Pu , 42.8 keV in ^{240}Pu , and 83.9 keV in ^{243}Am .¹¹⁵ Highlights included the thorough investigation of neptunium and neptunium compounds.¹¹⁶

Another important early contribution was the demonstration that Americium metal worked well as source material. As Dunlap explained: “The linewidth was much larger than intrinsic but still small compared to observed hyperfine splittings, and the specific activity was very high.” Argonne researchers performed most of the initial work that established the resonance as well as many early applications to physical questions.¹¹⁷

Argonne researchers obtained particularly rich results with ^{237}Np , as Dunlap explains, thanks to such considerations as “source strength, source and absorber availability” and “intrinsic linewidths.” This resonance, which was first observed at the Savannah River Laboratory in 1964 by W. L. Pillinger and J. A. Stone, was quickly adopted at Argonne by Kalvius, Ruby, and Merwyn Brodsky. As Dunlap later noted: “Fortunately, the Np resonance is one of the best of all Mössbauer resonances because of its very large hyperfine parameters (magnetic, electric quadrupole, and isomer shift).”¹¹⁸

In a 2003 review, Dunlap gave a number of Argonne research highlights with neptunium and neptunium compounds. Figure 13 shows “Mössbauer spectra for Np in trivalent, quadrivalent, pentavalent and hexavalent compounds.” NpF_3 “shows a pure electric quadrupole splitting,” NpO_2 “shows a single line (slightly broadened due to a controversial phase transition at 25K), and the other two spectra are split by mixed magnetic and quadrupole interactions. Isomer shifts, shown by the arrows, are large between valence states.”¹¹⁹

Figure 14 “shows a summary of isomer shifts for chemical compounds taken over several years time. One observes large isomer shifts between different valence states and a large spread of isomer shifts for their respective charge state, and the spread to positive values is due to bonding effects. Furthermore, strong covalency effects are present even for the pure halides.”¹²⁰

¹¹⁵W. L. Croft, J. A. Stone, W. A. Pillinger, “Mössbauer Effect in ^{231}Pa ,” *J. Inorg. Nucl. Chem.* 30 (1968), 3203-3208; R. D. Meeker, et al., “Hyperfine Interactions and Nuclear Moments,” *Nucl. Phys. A* 224 (1974), 429-436; S. L. Ruby, et al., “Nuclear Gamma-Ray Resonance Study of Hyperfine Interactions in ^{238}U ,” *Phys. Rev.* 184 (1969), 374-380; Dunlap, et al., “Isomer shift and Hyperfine Splittings of the 59.6-keV Mössbauer Resonance in ^{237}Np ,” *Phys. Rev.* 171 (1968), 316; J. Gal, et al., “Mössbauer Isomer Shift in ^{243}Am ,” *Phys. Letts. B* 29 (1969), 489-492; Kalvius, et. al., Mössbauer Resonance in ^{240}Pu ,” *Phys. Rev. B* 18 (1978), 4581-4587.

¹¹⁶Dunlap (ref. 103), Dunlap and Kalvius (ref. 110), 334.

¹¹⁷Dunlap (ref. 103). Dunlap and Kalvius (ref. 110), 335.

¹¹⁸Ibid., Dunlap (ref. 103).

¹¹⁹Ibid. See this reference for more details and more research highlights. B. D. Dunlap, et. al., “Isomer Shift and Hyperfine Splittings of the 59.6-keV Mössbauer Resonance in ^{237}Np ,” *Phys. Rev.* 171 (1968), 316-321.

¹²⁰Dunlap (ref. 103).

FIGURE 13 Mössbauer Spectra for Np in Trivalent, Quadrivalent, Pentavalent and Hexavalent Compounds

FIGURE 14 A Summary of Isomer Shifts

A great deal of work was also done with the magnetic properties of Np compounds. As Dunlap summarizes: “The Np resonance provides detailed magnetic hyperfine spectra that allow very accurate determinations of magnetic parameters. A particularly interesting case, which also shows how Mössbauer spectroscopy can be used to complement other analytical techniques, is NpP.” Neutron diffraction data had shown the presence of two magnetic moments for Np, but a detail of the neutron analysis made it difficult to establish the values unambiguously. Figure 15 presents an analysis which “gives two hyperfine fields which can be used with a known scaling between the hyperfine field and magnetic moment to resolve the uncertainty.”¹²¹

Although the second generation of Mössbauer spectroscopists at Argonne had equipment for measuring Mössbauer spectra that could have been duplicated at a small college, this work could not have been performed at many other places. For one thing, Argonne was one of the few worldwide facilities with the expertise and specialized equipment needed to work with the highly radioactive, toxic actinides. Argonne’s expertise with actinides reached back to the laboratory’s WWII predecessor, the Metallurgical Laboratory. In 1944, Glenn Seaborg (who before the war had created, with others, elements 93 and 94) was able to detect elements 95 (americium) and 96 (curium) at the Met Lab after samples of plutonium had been bombarded, respectively, by

¹²¹Ibid.

neutrons in the Hanford reactor and by helium atoms at the 60-inch cyclotron at Berkeley. The tradition of actinide chemistry continued after Seaborg left and Argonne was founded.¹²²

Thanks to decades of experience, Argonne researchers were among the lucky few to have access to actinides which were only allowed at places – like Argonne – with a special license and tracking and inventory procedures. Argonne also had gathered the necessary, specialized equipment for actinide work: by 1963, Argonne had a Chemistry Hot Laboratory with heavily shielded cells which allowed scientists to safely work with actinides and other highly radioactive substances using remote control¹²³ (Figure 16).

As before, intellectual resources were a key advantage for Argonne's second generation of Mössbauer spectroscopists. At this stage Kalvius, Ruby, Dunlap, and Shenoy not only had reactor specialists to prepare the necessary isotopes and chemists and metallurgists to help prepare high quality materials – they also had these (and other) specialists to help them. Dunlap later listed 12 specialists from seven groups outside the Solid State Science and Physics Divisions who helped with actinide Mössbauer spectroscopy¹²⁴ (Figure 17).

Collaboration was important in other ways as well. Kalvius, Ruby, Dunlap, and Shenoy had launched the second generation of Mössbauer spectroscopy at Argonne – and continued the intellectual tradition of Argonne Mössbauer spectroscopy – by bringing expertise from nuclear physics to facilitate solid state physics and by adding perspectives from many other fields to the mix. In Dunlap's opinion, this multidisciplinary approach – which was as apparent in other types

FIGURE 15 Analysis of Two Hyperfine Fields which can be Used with a Known Scaling between the Hyperfine Field and Magnetic Moment to Resolve the Uncertainty

FIGURE 16 Containment Boxes within the Heavily Shielded Cells of the Chemistry Hot Laboratory, Argonne, #120-7820

¹²²Holl (ref. 89), 456-464. A full history of the discovery of neptunium and plutonium can be found in J. L. Heilbron and Seidel, *Lawrence and His Laboratory* (University of California Press, 1989).

¹²³Dunlap (ref. 103). "Argonne National Laboratory Annual Report, 1963," Argonne National Laboratory Library.

¹²⁴Ibid., Dunlap (ref. 103).

- Mössbauer Spectroscopy – Mike Kalvius, Bob Dunlap, Gopal Shenoy, Stan Ruby, Lynn Soderholm, and many visitors and postdocs.
- Chemistry – Don Cohen, Sherman Fried, Bill Carnall
- Alloy Preparation and Phase Analysis – Dan Lam
- Magnetism and Transport – Merwyn Brodsky, Tony Aldred
- Neutron Scattering – Gerry Lander and Mel Mueller
- Nuclear Magnetic Resonance – Frank Fradin
- Electronic Structure Calculations – Dale Koelling, Art Freeman
- Optical Properties – Boyd Veal

FIGURE 17 Those Who Helped in the Mössbauer Effort with Transuranics

of Mössbauer work as it was in actinide research – was an important boon since it allowed Argonne scientists to get around the “tendency to view hyperfine data as self-contained and not to see how it relates to and compliments other results.” Since at “Argonne one could find an expert in a vast number of subjects, it was very easy to involve people with other interests and capabilities, and so to build a scientific story.” When working with a material as scientifically complex as the actinides this multidisciplinary advantage was all the more important.¹²⁵

As had previously been the case, the resources at Argonne – access to the CP-5 reactor, technical support, the capability of working with actinides – drew additional intellectual resources. Some came from the Chicago area, such as Northwestern University theorist Art Freeman, who was a foremost expert in magnetism and hyperfine interactions, and Northern Illinois University experimentalist Clyde Kimball, who made his mark in “understanding magnetism and superconducting systems and phonon anomalies.” Argonne also became a Mecca for visitors, post-doctoral fellows and students as Shenoy and Dunlap attracted students from 1974 to 1984 from a variety of countries including France, Sweden, Turkey, India, Brazil, and Greece.¹²⁶

In the process, Argonne’s pattern of collaborating with another large laboratory also continued; in this case, the partner was the Technical University of Munich where Mössbauer had done his influential graduate work. Argonne’s relationship with TU Munich began before Mössbauer arrived; Maier-Leibnitz, had close ties with various U.S. nuclear physicists, including Hanna and Perlow. After a stay at Cal Tech, Mössbauer returned to Munich in the mid-1960s to found an institute in Mössbauer spectroscopy. Just before leaving Munich again, Mössbauer successfully convinced Kalvius to return to Munich in 1970. After Mössbauer’s second departure, Kalvius became head of the institute. Throughout the 1970s this institute was a thriving center of Mössbauer spectroscopy and TU Munich and Argonne had a lively exchange of personnel.¹²⁷

¹²⁵Ibid.

¹²⁶Shenoy, “Mössbauer History Celebration Day: Biographies of Argonne’s Mössbauer Scientists,” ANL-03/27.

¹²⁷Kalvius, interview with Catherine Westfall, 29 Sep 2003; Fritz Wagner, interview with Westfall, 1 Oct 2003; Shenoy, *ibid.*

One example of this exchange came shortly after Kalvius arrived in Munich, and Shenoy followed him. Once there he collaborated with many young physicists under the direction of Mössbauer and Paul Kienle. When Shenoy returned to a staff position at Argonne, many of these physicists visited him there. One fruit of this collaboration was an important reference work, *Mössbauer Isomer Shifts*, which Shenoy and Fritz Wagner published in 1978; it provided a cross-disciplinary guide to the isomer shift that proved especially useful to those in chemistry, biology, and nuclear physics.¹²⁸

As before, along with the exchange in personnel came an exchange in equipment expertise. In this case Perlow developed a drive system at Argonne using the piezoelectric effect in quartz single crystals. In the process of mutual education between Perlow and TU Munich's Walter Potzel on the subtleties of the piezoelectric effect in quartz, this piezo-drive was improved, first still at Argonne and thereafter at Munich by rearranging the crystals and making the arrangement more compact and stable against unwanted vibrations. In Potzel's opinion, the combination of "experimental details" which he obtained at Argonne as well as "German craftsmanship" allowed the researchers "to successfully perform a whole series of experiments on fundamental physics" at both institutions including work with special Mössbauer transitions, like the high-resolution resonance in ^{67}Zn .¹²⁹

Research life was not that different for Argonne's first and second generations of Mössbauer spectroscopists apart from the shift from the Physics Division to the Solid State Science Division. Added to the continued supply of specialized resources was another constant: as before, ideas were hatched at the Argonne cafeteria and tested quickly. Kalvius later remembered: "We had a group that used to get together every day at lunch from various divisions. If anyone had a new idea, no matter how ridiculous it might first seem, we discussed it." Hanna and Perlow remembered that ideas in the early 1960s that were discussed at lunch would be carried out later that day; by the later period the turnaround time was only slightly longer. "Many times ideas that were discussed would result in an experiment in the next week," in Shenoy's words. At this stage the multidisciplinary nature of the work as well as the quick pace made research life exciting. "There was a sense of freedom, of few intellectual barriers, of getting things done." It was a time when the Mössbauer factory in the Solid State Science Division was fueled "by the factory of ideas" in the cafeteria: The diverse group of experts considered on-going measurements and developed new theoretical insights that in turn led to new research directions.¹³⁰

Although the work of Mössbauer spectroscopists remained much the same through the 1970s, by the early 1980s the days of traditional Mössbauer were clearly numbered. As Dunlap later noted: they "all went on to do other things. Mike moved to muon spin resonance work, Gopal set off to design and build the Advanced Photon Source (APS)." For his part, Dunlap "did some bulk property measurements, and finally spent some years in administration."

¹²⁸Kalvius, interview with Westfall, 29 Sep 2003. Shenoy and Wagner, *Mössbauer Isomer Shifts* (Amsterdam, 1978). Walter Potzel, interview with Westfall, 29 Sep 2003.

¹²⁹Potzel, interview with Westfall, 29 Sep 2003; Shenoy, (ref. 126).

¹³⁰Kalvius, interview with Westfall, 29 Sep 2003; Shenoy, interview with Westfall, 30 Apr 2002.

Frank Fradin, a material scientist who worked closely with Kalvius, Shenoy, and Dunlap on research into actinides later judged that by this time “the cream” of results from Mössbauer spectroscopy “had been skimmed off the top.”¹³¹

Mössbauer spectroscopy did not die out at Argonne, however. Building on decades-long expertise in the construction of large accelerators, Shenoy, Fradin (who became division director of the Materials Science Division in 1985), and others began work on the Advanced Photon Source, which became a reality along with plans for an APS Mössbauer beam line.¹³² At this point the apparatus for Mössbauer spectroscopy got bigger, opening the question of whether earlier patterns would persist as Mössbauer spectroscopy continued to develop at Argonne.

3.3 THE BIG (?) SCIENCE OF THE APS MÖSSBAUER BEAM LINE AND THE THIRD GENERATION OF MÖSSBAUER SPECTROSCOPY AT ARGONNE, 1980S-2005

In January 1986, the Department of Energy (DOE) announced it would build the Advanced Photon Source at Argonne. Argonne researchers finished a Conceptual Design Report just two months later. The synchrotron light-source would include a number of beam lines that could be used for basic and applied research in a wide range of fields, including physics, materials science, chemistry, biological science, geophysical, environmental, and planetary science, as well as x-ray instrumentation. Despite a funding crisis in 1988, the synchrotron light source began operation in 1996. The eventual cost of the APS, including construction of beam lines, was almost \$1 billion, making it the DOE’s most expensive accelerator facility at the turn of the 21st century¹³³ (Figure 18).

Argonne’s Mössbauer beam line – and Argonne’s third generation of Mössbauer spectroscopy – began with Ercan Alp. As a student, Alp had worked with traditional Mössbauer spectroscopy at the Middle East Technical University in Ankara Turkey and had also spent time with Gerdau at Hamburg on an International Atomic Energy Agency scholarship. In 1984, he came to Argonne to work with Shenoy as a post-doc. Although his original intent had been to continue with traditional Mössbauer spectroscopy, the Mössbauer laboratory was closing, so Alp switched to Extended X-ray Absorption Fine Structure Spectroscopy (EXAFS), which led to work at Stanford’s SSRL. Although Alp wanted to begin plans for an APS Mössbauer beam line as soon as he heard of Gerdau’s successful DESY experiments, funding uncertainties for the large accelerator forced him to bide his time.¹³⁴

¹³¹Dunlap (ref. 103), Frank Fradin, interview with Westfall, 15 Aug 2003.

¹³²Shenoy (ref. 126). Holl (ref. 89) describes the large accelerator building tradition that began with the ZGS and continued with the Intense Pulsed Neutron Source and the APS.

¹³³For more on the advent of the APS, see Holl, (ref. 89), 467-473. “Conceptual Design Report of a 6 GeV Synchrotron X-ray Source,” March 1986. Alan Schriesheim, “State of the Laboratory – 1996,” Argonne National Laboratory.

¹³⁴Alp et al., Nuclear Resonant Scattering Beamline at the Advanced Photon Source,” *Hyperfine Interactions* 90 (1994), 323; Alp, interview with Westfall, 13 Aug 2003.

FIGURE 18 Aerial View of the APS, Argonne

By 1989, however, the time was ripe to proceed: funding for the APS seemed secure and plans for beam lines were underway, and in the meantime, Alp had gained experience with synchrotron light source spectroscopy at SSRL and had become a permanent member of the Argonne staff. As a first step, Alp went to DESY to learn about doing nuclear resonance scattering using a synchrotron light source. When he returned to Argonne he consulted with Dennis Mills, who headed an APS x-ray optics group, and came up with the plan to design a beam line that would do both inelastic scattering and nuclear resonance scattering. He then tested his ideas at an experiment in 1991 at the Cornell's CHESS which measured the quadrupole splitting and isomer shift of Mohr's salt as a means to "assess and demonstrate the potential of this new technique." The layout of the experiment was reminiscent of previous Mössbauer spectroscopy – in addition to detectors and an absorber, there was a source, in this case x-rays produced by a synchrotron light source¹³⁵ (Figure 19).

As Alp and collaborators explained in a 1992 article, along with the new research possibilities of synchrotron light source Mössbauer spectroscopy came the particular challenge of designing and manufacturing new x-ray optics devices, in particular monochromators with an

¹³⁵Alp, Sturhahn, Toellner, "Synchrotron Mössbauer Spectroscopy of Powder Samples," *Nucl. Instr. Meth. in Physics Research B*, 97 (1995), 526-529.

FIGURE 19 Layout of Synchrotron Mössbauer Spectroscopy. Reprinted from *Nuclear Instruments and Methods in Physics Research B* 97, E. Alp et al., “Synchrotron Mössbauer Spectroscopy of Powder Samples,” Alp et al., 527, 1995, with permission from Elsevier.

energy resolution of $1\text{ }\mu\text{eV}$ -1 neV. Using the experience gained with the first CHES experiment, Alp returned to Cornell to face this challenge with an experiment aimed at measuring the Mössbauer resonance of ^{119}Sn using synchrotron radiation for the first time. As he later recalled, he “received about 20 celebratory messages from all around the world.” People were excited “because it was the first time something really exciting had happened in the field” since Gerda showed that Mössbauer spectroscopy could be done with a synchrotron light source 8 years before.¹³⁶

This success was possible because “we were the first ones to successfully develop the nested monochromator concept.” Although this concept had been proposed by T. Ishikawa and others, “nobody in the world had actually made it work.” As Alp and collaborators explained, their monochromator was used “as a beam conditioner for nuclear resonant scattering of synchrotron radiation to produce x-rays with μeV -neV resolution in the hard x-ray regime.” It was “designed to reduce the bandpass of synchrotron radiation to 1-5 meV level, without sacrificing angular acceptance”¹³⁷ (Figure 20).

Success, which was possible because of collaboration, also facilitated further fruitful collaboration. On the heels of the development of the monochromator and the successful ^{119}Sn measurement, Alp was able to recruit Wolfgang Sturhahn from the group at Hamburg and two talented young American scientists, Tom Toellner and Tim Mooney.¹³⁸

¹³⁶ Alp, interview with Westfall, 13 Aug 2003. Alp, et al., “Coherent Nuclear Resonant Optics for Third Generation Radiation Sources,” the International Society for Optical Engineering, proceedings reprint 1740 (1992), 95-102; Alp et al., “Time Resonant Nuclear Resonant Scattering from ^{119}Sn using Synchrotron Radiation,” *Phys. Rev. Lett.* 70 (1993), 3351-3354

¹³⁷ Alp, interview with Westfall, 13 Aug 2003; Toellner et al., “High Energy Resolution, High Angular Acceptance Crystal Monochromator,” *Optics for High-Brightness Synchrotron Radiation Beamlines*, the International Society for Optical Engineering, proceedings reprint 1740 (1992), 218-223. T. Ishikawa et al., “Construction of a Precision Diffractometer for Nuclear Bragg Scattering at the Photon Factory,” *Rev. Sci. Instrum.* 63 (1992), 1015-1018.

¹³⁸ Mooney went on to be the leader of another group at the APS, but Sturhahn and Toellner continue to work with Alp on the APS Mössbauer beam line in 2004. Alp, interview with Westfall 13 Aug 2003; Sturhahn and Toellner, interview with Westfall, 2 Oct 2003.

FIGURE 20 Monochromator. Reprinted with permission from Toellner, et al., “Optics for High-Brightness Synchrotron Radiation Beamlines,” *SPIE*, 1740, 1992.

In 1994 and 1995, before the APS was ready for use, this third generation of Mössbauer spectroscopists did experiments with Japanese collaborators at the NE3 undulator beam line of the accumulator ring at KEK in Japan which demonstrated the measurement of inelastic nuclear resonant scattering for the first time as well as the use of polarization-based optics.¹³⁹

The results of Alp’s group through the mid-1990s impressed Shenoy, Mills, and other APS managers. In Alp’s words “it was clear we no longer had an exotic application.” This support was crucial because, as Alp remembered, the estimated cost of the beam line was \$4 million which was considered “a huge investment in an experimental field.”¹⁴⁰

By the time he finished the Japanese experiments in 1995, the third generation of Mössbauer spectroscopists had gained experience, success, a good working collaboration, and support from senior APS scientists. The previous year the collaborators had developed plans for the APS beam line. The collaborators spent 1996 – the year the APS started operation – bringing the beam line to life.¹⁴¹

In January 1997, the Mössbauer beam line was complete and experiments begun, a feat made possible because, in Sturhahn’s words, they “were well prepared by our experiments at

¹³⁹Sturhahn et al., “Photon Density of States Measured by Inelastic Nuclear Resonant Scattering,” *Phys. Rev. Lett.* 74 (1995), 1993-5; Toellner et al. “Polarizer/analyzer Filter for Nuclear Resonant Scattering of Synchrotron Radiation,” *Appl. Phys. Lett.* 67 (1995), 1993-1995.

¹⁴⁰Alp, interview with Westfall 13 Aug 2003.

¹⁴¹*Ibid.*

other places before the APS was built.”¹⁴² Alp was very proud of their early work. A project on dynamical properties of nano-crystalline iron was the first *Physical Review Letter* based on APS research and also showed the group’s eagerness to engage in emerging fields, like nanoscience. As time went on, particular interests of the group included “high-pressure and high-temperature measurements on iron bearing materials for geophysical applications, vibrational studies on iron containing biomolecules and proteins, and the vibrational and magnetic behavior of thin films.”¹⁴³

In 2005, research life proceeded at the Mössbauer beam line according to the pattern set at the APS. As is the case with other APS beam lines, beam time on the Mössbauer beam line was allocated by the APS beam allocation committee. As in an arrangement typical when APS staff members control a beamline, Alp, Sturhahn, and Toellner maintained the beam line. Sturhahn focused on “the development and application of the novel experimental techniques using resonances of Mössbauer nuclei and synchrotron radiation” and Toellner continuing to address the many challenges of beam optics.¹⁴⁴

In addition, the group actively recruited users to use their beam line with the intent of making it a multi-purpose tool for the widest possible range of scientific applications. The users were not the only ones to benefit. As Sturhahn noted, “we reap scientific benefit from collaborating with them as well as the strategic benefit of having plenty of users.” Like users of other large machines, the group understood that continuing their research hinged on enlisting interested users. In their case, as Sturhahn put it, lack of interest in their beam line could mean that it would be replaced with another that will bring more users since “continued funding of the synchrotron light source requires showing that it has strong user backing.” Luckily for the group members, they were well-positioned to launch applications in biophysics, geophysics, and materials science and were judged by an editor of the *Mössbauer Effect Reference and Data Journal* to be the most productive group in the United States.¹⁴⁵

Those using the APS Mössbauer beam line in particular enriched knowledge about materials by developing phonon spectroscopy and expanding its application to geo-physics and bio-physics. In particular, APS Mössbauer spectroscopists developed sapphire back-scattering crystals as x-ray optical components, tunable monochromators with record-breaking resolutions and isotope-selective nuclear magnetometry. They also extended knowledge of the detailed behavior of dynamics of phonons in amorphous materials, and the systematics of the Boson peak, and made the first observation of nuclear lighthouse effect. All of these accomplishments

¹⁴²Sturhahn and Toellner, interview with Westfall, 2 Oct 2003. Alp. et al., “Nuclear Resonant Scattering Beamline at the Advanced Photon Source,” *Hyperfine Interactions* 90 (1994), 323-341 Alan Schriesheim, “State of the Laboratory – 1996.”

¹⁴³B. Fultz et al., “Phonons in Nanocrystalline ⁵⁷Fe,” *Phys. Rev. Lett.* 79 (1997), 937-940; Shenoy, (ref. 126).

¹⁴⁴Sturhahn and Toellner, interview with Westfall, 2 Oct 2003.

¹⁴⁵*Ibid.*; Stevens, remarks at Mössbauer history celebration day at Argonne, 30 Sep 2003. For more on the necessity of gathering interested users for large projects, see Westfall and Lillian Hoddeson, “Thinking Small in Big Science: The Founding of Fermilab, 1960-1972,” *Tech. and Cult.* 37 (1996), 457-492 and Robert W. Smith, *The Space Telescope* (Cambridge University Press, 1989).

have, in turn, motivated new generation of experimenters to adopt this method of nuclear resonant scattering with synchrotron radiation.¹⁴⁶

An important feature of the APS Mössbauer beam line was that it continued a long-term tradition of Mössbauer research at Argonne. A striking personification of the continuity of this tradition of Mössbauer spectroscopy at Argonne came with Harry Lipkin, a member of the first generation of Mössbauer spectroscopy. In his 1973 book on modern methods of quantum mechanics Lipkin published a chapter explaining how certain sum rules apply in governing the dynamics between the energy and momentum of the incident photon and the bound atoms in a solid. When Alp invited Lipkin to lecture to his group in 1994 Lipkin learned of some experiments involving phonon exchange using tunable high resolution monochromators and set out to calculate the third moment of the energy spectrum. In the process he pointed out that while the first moment was related to the recoil-free fraction, the third moment was related to the force constant. After the experiments in Japan, Sturhahn applied this knowledge to normalize the different spectra obtained from iron-containing samples, and he was able to extract the phonon density of states. Lipkin, who became a regular visitor to Alp's group, was quite happy to see the continuation of his early work and published his calculation. By 2005, a new generation of Mössbauer spectroscopists had emerged at Argonne. Representatives of four generations of Mössbauer spectroscopy, including Lipkin and younger scientists trained by Alp and his group, are shown in the title page.¹⁴⁷

¹⁴⁶G. Shen et al., "Photon Density States in Iron at High Pressures and High Temperatures," *Phys. and Chem. of Min.* 31 (2004), 353-359.

¹⁴⁷Lipkin, *Quantum Mechanics* (Amsterdam, 1973), 33-110; Sturhahn, et al., *Phys. Rev. Lett.* 74 (1995), 3832-3835; Lipkin, "Mössbauer Sum Rules for Use with Synchrotron Sources," *Phys. Rev. B* 52 (1995), 10073-10079.

4 CONCLUSION

The experience of Alp's group shows that although the intellectual goals of extending knowledge about matter with measurements of Mössbauer spectroscopy did not change, research life did change when experimental equipment included a billion dollar synchrotron light source instead of a radioactive source plus other equipment equaling \$100,000 or less. Whereas funding was a sidelight for earlier researchers, for Alp and his group, obtaining funding was a crucial theme: Alp could not proceed with his beam line until APS was secure, he had to worry about costs for the beam line, and group members thought about building a constituency to maintain the beam line's continued existence.

Research life was different for the third generation of Mössbauer spectroscopists in other ways as well. It took about 10 years for the synchrotron light source to proceed from the first funding announcement to operation of the accelerator (and several more years were spent in planning and designing it). The Mössbauer beam line took a year to build, and potential users had to first obtain permission from the APS beam allocation committee. Gone were the days when the cafeteria was the factory for ideas that could be implemented by all those interested later that day or in the next week.

At the same time, research at the APS was not the same as research at large-scale nuclear and high energy physics facilities. A vivid contrast, for example, can be drawn between research in 2005 using the Mössbauer beam line and research using the Colliding Detector Facility (CDF) at Fermilab, a high energy physics laboratory not far from Argonne. Even though Fermilab also supported a synchrotron that is about the same size and had an original cost (if adjusted to 1990s dollars) that was about the same as the APS, CDF otherwise operated at a scale vastly greater than the Mössbauer beam line: the Mössbauer beam line is a few yards long, CDF is the size of a warehouse; the Mössbauer beam line cost \$4 million, CDF cost \$37 million; experimental teams using the Mössbauer beam line had about a dozen collaborators, CDF had about 500 collaborators. Also, CDF is used only for high energy physics, while the Mössbauer beam line is used for applications as diverse as biophysics, geophysics, and materials science¹⁴⁸ (Figure 21).

Still, performing Mössbauer spectroscopy at the APS is in many ways similar to performing Mössbauer spectroscopy with radioactive sources. Some of the similarities, such as the continuities in patterns of collaboration, do not distinguish Mössbauer spectroscopy from CDF and other large-scale research. Indeed, Fermilab and Lawrence Berkeley Laboratory history show that it is typical for large laboratories to collaborate with each other and to have productive exchanges of personnel. However, the consistent team size over time and the broad multipurpose nature of the Mössbauer beam line make it not only different from CDF, but also different from other large-scale science, such as the modest and mezzo science of the HILAC and Bevalac, which were both used for types of nuclear physics research. This gives credence to Shenoy's

¹⁴⁸Westfall, "Collaborating Together: The stories of TPC, UA1, CDF, and CLAS," *Hist. Stud. Phys. Scien*, 32 (2001), 163-178. For the cost of Fermilab, see Westfall, "A Tale of Two More Laboratories: Ready for Research at Fermilab and Jefferson Laboratory," *Hist. Stud. Phys. Scien*, 32 (2002), 369-407.

FIGURE 21 Ercan Alp at the APS Mössbauer Beam Line, Argonne

view that Mössbauer spectroscopy at the APS is “not really a different type of Big Science,” but rather “a different type of little science.”¹⁴⁹

One of the lessons to be learned from 50 years of Mössbauer spectroscopy at Argonne is that large laboratories have much to offer small-scale research. Part of the advantage comes because large laboratories are the “Home” of large apparatus. A major theme in the story of Mössbauer research at Argonne is, in fact, the special access Argonne researchers had to reactor-produced and accelerator-produced radioactive sources, and eventually, to a \$1 billion synchrotron light source. The special capabilities of large machines are not the whole story, however. Access to exotic xenon compounds, powerful computers and highly restricted actinides also provided Argonne Mössbauer spectroscopists with research opportunities open to few others.

But the most distinctive aspect of the 50-year history of Mössbauer spectroscopy is that it consists of a multi-decade intellectual tradition that features people more than the massive instruments that so often monopolize the attention of historians. At center stage in this history are those who made contributions to science using equipment that was relatively modest in size and expense. This history highlights a fact that applies to national laboratory research at a variety of scales: that at base it is propelled by people from many places – from various parts of the laboratory, from other large laboratories, from universities, from industry, from foreign

¹⁴⁹Shenoy, interview with Westfall, 30 Apr 2002. The Bevalac was used for nuclear physics and biomedical research as well as radiation therapy. Although it had a wider scope of research than many high energy and nuclear physics accelerators, this scope was still narrower than that at the APS. With Hoddeson and Adrienne Kolb I argue that CDF in fact represents a new, larger scale of “megascience” that changes how physics research is conducted. Hoddeson, Kolb, and Westfall, *The Ring of the Frontier: From Big Science to Megascience: A History of Fermilab under Robert Wilson and Leon Lederman*, forthcoming.

countries. These people gather at a large laboratory to exploit its resources, which include smart people with varied expertise as well as specialized equipment and material. Once gathered these people produce better instruments, more ideas, greater and more varied expertise, research accomplishments, and at least in some cases, long-term, productive intellectual traditions. In the process, the laboratory's resources for research of many kinds at a variety of scales are continuously improved. Rather than thinking of large laboratories as the "home" of Big Science, it is therefore more useful to view such facilities as sites where knowledge is produced in a self-perpetuating process of wide-ranging collaboration.