Hyperfine Interact
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Personal recollections of 52 years of Mössbauer spectroscopy research

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Abstract The history of the scientific developments of the Mössbauer effect, through 52 years, is discussed in terms of personal observations and participation.

Keywords Paramagnetic relaxation · Intermediate valence · Bound diffusion · Superconductivity · Relativity

1 Introduction

I was exposed to Mössbauer spectroscopy (MES) early in 1959 when Solly Cohen and Shimon Ofer returned from the US to Jerusalem with the great promise for a new method of research, for low energy nuclear physics and hyperfine interactions in solids. I became the first MES student in Jerusalem. Two years later, immediately after accepting the Nobel Prize, Mössbauer the person, arrived directly from Stockholm to Jerusalem to present us, his Nobel Prize lecture. In the summer of 1970 I spent 4 months in Garching doing research with Mössbauer's group. Mössbauer visited Jerusalem several times. In one of his early visits he posed for a picture together with his hosts and Professor Felix Bloch (Bloch waves and NMR) who was also present (Fig. 1).

In the first few years MES served the nuclear physicists in their pursuit for new MES isotopes, their nuclear moments and radii. The isotopes ^{161}Dy and ^{160}Dy were discovered in Jerusalem [1].

Only a few experiments were devoted to general physics, like relativity [2, 3] interference, γ-ray optics [4], start time dependent MES [5] (all of great interest today with Synchrotron MES). The state of the art in the first four years was well documented in Frauenfelder's book [6].

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Fig. 1 From right to left: The Professors R. L. Mössbauer, S. Ofer, S. G. Cohen and Felix Bloch

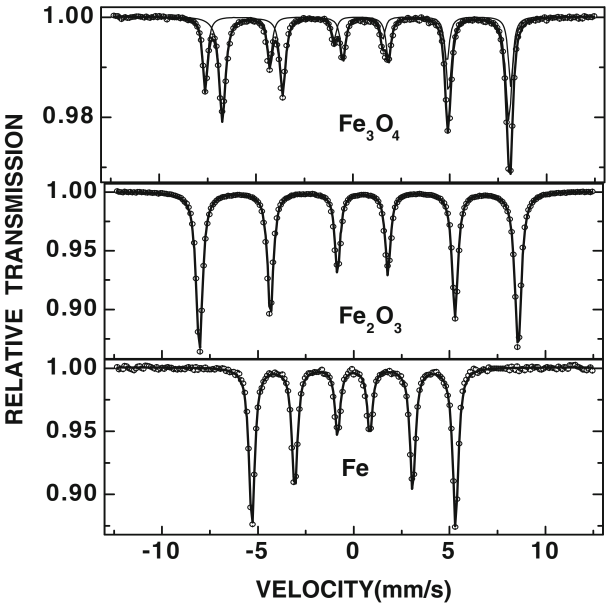
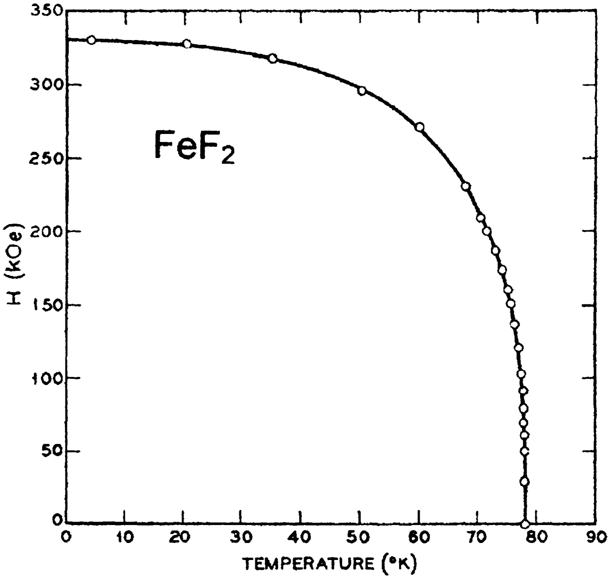


Fig. 2 Room temperature Mössbauer spectra of the most common magnetic materials

Very soon Physicists and Chemists joined with extensive studies of solid-state properties. All branches of Chemistry were investigated. The largest number of publications and books [7] are devoted to these subjects. All thermodynamic phase transitions were studied. New unexpected dynamic phenomena, directly observable in the spectrum like: spin relaxation [8, 9] in iron, and in all rare earths and actinides, spin crossover and transitions in iron compounds [10], intermediate valence and valence changes [11] in iron, europium, samarium and even neptunium, and bound diffusion [12], were all discovered in a very wide range of compounds.

Fig. 3 The temperature dependence of the internal magnetic field acting on Fe in FeF_2 [21]



Researchers interested in biological compounds, found in MES a very useful tool. They found that live biological systems are, in many characteristics, a unique phase of matter, in between solid and liquid. The associated theory explained well the observed phenomena [13, 14].

Superconductivity was always of interest for MES researchers. But only the discoveries of magneto-superconductors, in particular those with high T_c, made the MES research contribution significant [15, 16]. Coexistence of magnetic order and superconductivity is now well established.

During the last 52 years I was involved in some of the research projects mentioned above, performed in Jerusalem (with S. G. Cohen, S. Ofer, E. R. Bauminger, I. Felner and R. H. Herber), Bell Telephone Laboratories (with H. H. Wickman and R. L. Cohen), Argonne National Laboratory (with B. D. Dunlap), Technical University of Munich (with G. M. Kalvius) and the Free University of Berlin (with G. Kaindl and G. Wortmann)

In the present short review, the historical aspects and the scientists involved in the most interesting of the subjects mentioned above will be given. The recent studies of magneto-superconductors (with FeAs and FeSe) and the experiment for the discovery of a possible existence of a maximum acceleration in General Relativity [17] will be presented.

2 Detailed discussion

One of the early prominent studies concerned the subject of magnetism in solids. Just the discovery that one can measure so easily the magnetic Zeeman splitting of nuclear levels (magnetic hyperfine structure) was unbelievable earlier. The most

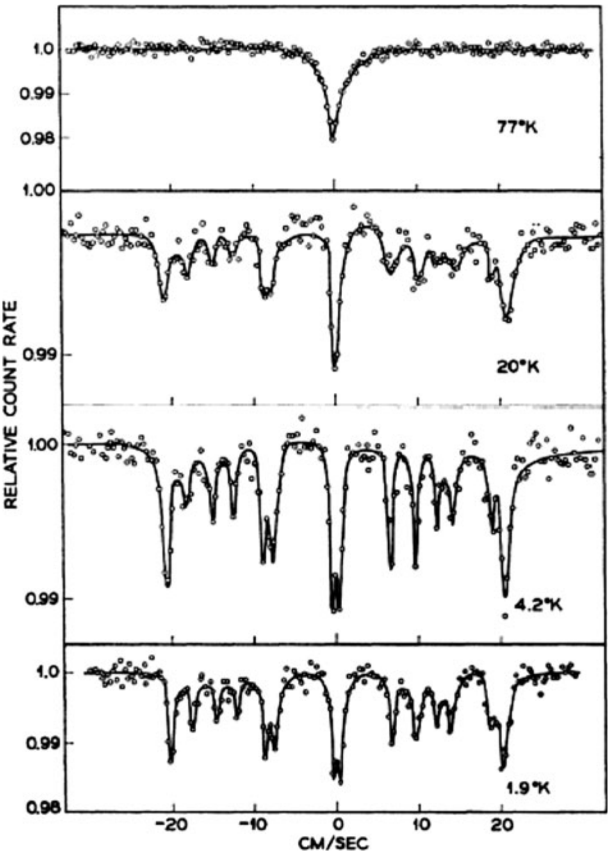
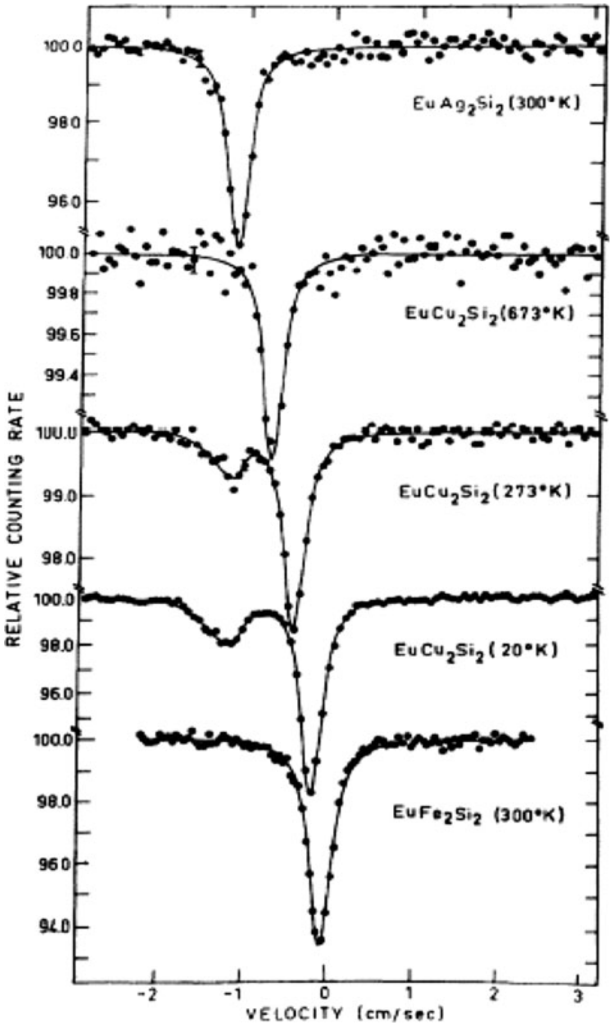


Fig. 4 The 26 keV Mössbauer spectra of ^{161}Dy in dysprosium aluminum garnet ($Dy_3Al_5O_{12}$) [22]

common magnetic iron compounds were immediately studied, Fig. 2. The discovery of many rare earth Mössbauer isotopes prompted studies of magnetic compounds of rare earths (this was the subject of my PhD thesis).

The high resolution of the ^{57}Fe Mössbauer 14.4 keV urged scientists to try to test General relativity in their laboratory. The successful measurement of the "gravitational weight" of a photon was sensational [2]. The story tells that another group made the same experiment. In an internal seminar the experiment was displayed, and an undergraduate student asked about the source and absorber temperature, when answered "why is it important?" he explained the importance, and published it [18]. He has discovered what is known today as the thermal shift, which is also the relativistic second order Doppler shift. The student was B. D. Josephson, who won later the Nobel Prize for much more important discoveries. W. Kündig performed an experiment [3] in which he tried to measure directly the second order Doppler shift, with an absorber moving perpendicular to the direction of the γ-ray. Quantum beats due to interference phenomena, were discovered by G. J. Perlow [19, 20].

Fig. 5 ^{151}Eu Mössbauer spectra of temperature dependent valence in $EuCu_2Si_2$ [11]



Among the various phase transitions the most investigated were the magnetic transitions, studied by recording the temperature dependence of the hyperfine field. One of the first was that by G.K. Wertheim [21] shown in Fig. 3.

The dynamic phenomenon of spin relaxation in paramagnetic compounds was discovered for iron and then also for many rare earth isotopes [9]. The phenomenon in magnetically ordered systems was very shortly also observed [8]. Typical relaxation spectra in paramagnetic $Dy_3Al_5O_{12}$ are shown in Fig. 4.

The phenomenon of intermediate valence in rare earth compounds [23] started with $EuCu_2Si_2$ [11], shown in Fig. 5.

The phenomenon of bound diffusion in biological systems [12], has been studied by P. Parak's group [14], by R. L. Mössbauer [24], and by our group. The theoretical

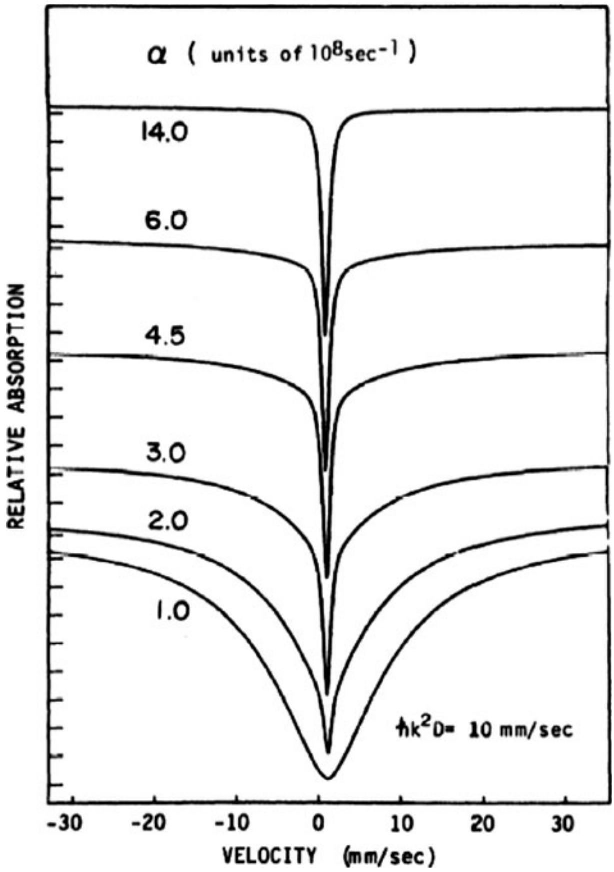


Fig. 6 Theoretical curves demonstrating how bound diffusion, with the decay rate (α) of a local deviation of a Mössbauer nucleus, to it's average position increases with temperature, leads from a narrow solid like absorption line to a very broad liquid like absorption line [13]

curves showing how a solid-like biological system, undergoing bound diffusion (relaxation rate increases with temperature), becomes liquid like, is shown in Fig. 6.

Mössbauer studies of magnetism in superconducting materials are very popular these days, due to the discovery of the FeAs [16] and lately the FeSe systems. From our studies we conclude that when the iron magnetic order is of spin density wave type, like in $EuFe_2(As_{1-x}P_x)_2$ [25], the appearance of superconductivity for certain x values destroys the spin density magnetism. However in the temperature rang in which the europium is magnetically ordered a transferred hyperfine field is observed even when the material is superconducting [25], Fig. 7. In the FeSe materials [26], the iron has a local moment and a large hyperfine field even at room temperature, and orders magnetically around 500 K. In this case the superconductivity and the magnetic order seem not affect each other.

Y. Friedman predicts [17] that General Relativity requires not only an absolute maximal velocity c, but also a maximal acceleration a_m. According to Friedman the second order Doppler shift for an absorber moving with velocity v and acceleration "a" in a direction perpendicular to the γ-ray is given by the formula: $-{}^1\!/_2 v^2/c^2 - {}^1\!/_2 a/a_m$. He argues that a correct analysis of the Kündig experiment [3] does not

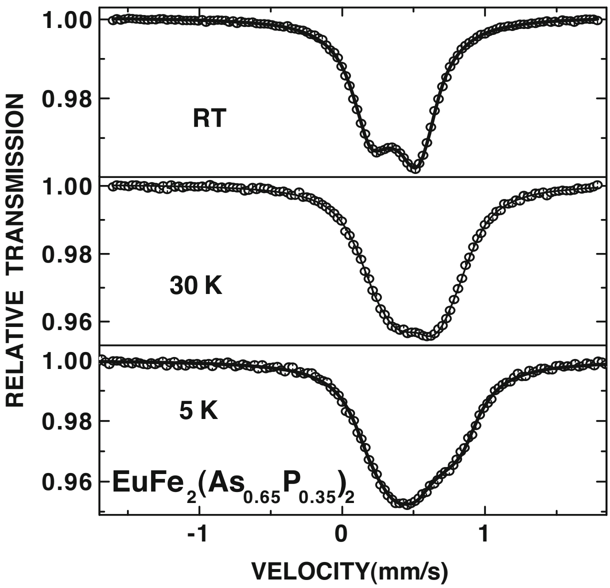


Fig. 7 ^{57}Fe spectra of $EuFe_2(As_{0.65}P_{0.35})_2$ at 5.0 K, in the superconducting state of the FeAs layers, at 30 K above T_M ~27 K of Eu, and at 297 K [25]

agree with the simple relativity formula and suggested an experiment in which the shift due to mechanical rotation will depend only on the centripetal acceleration. We are in the process of carrying out this experiment.

References

1. Bauminger, E.R., Cohen, S.G., Marinov, A., Ofer S.: Phys. Rev. Lett. **6**, 467 (1961)
2. Pound, R.V., Rebka G.A.: Phys. Rev. Lett. **4**, 337 (1960)
3. Kündig, W.: Phys. Rev. **129**, 2371 (1963)
4. Nowik, I.: Phys. Rev. **126**, 1878 (1962)
5. Lynch, F.J., Holland, R.E., Hamermesh, M.: Phys. Rev. **120**, 513 (1960)
6. Frauenfelder, H.: The Mössbauer Effect: A Review – with a Collection of Reprints (Frontiers in Physics Series), W. A. Benjamin (1962)
7. List of Mössbauer books is given in the web site www.mossbauer.org (2011). Accessed 15 Dec 2011
8. Nowik, I., Wickman, H.H.: Phys. Rev. Lett. **17**, 949 (1966)
9. Wichman, H.H., Wertheim, G.K.: Chap. 11 In: V.I. Goldanskii, R.H. Herber (eds.) Chemical Applications of Mössbauer Spectroscopy. Academic Press (1968)
10. Gutlich, P.: Struct. Bond. **44**, 83 (1981)
11. Bauminger, E.R., Froindlich, D., Nowik, I., Ofer, S., Felner, I., Mayer, I.: Phys. Rev. Lett. **30**, 1053 (1973)
12. Cohen, S.G., Bauminger, E.R., Nowik, I., Ofer, S.: Phys. Rev. Lett. **46**, 124 (1981)
13. Nowik, I., Cohen, S.G., Bauminger, E.R., Ofer, S.: Phys. Rev. Letters **50**, 1528 (1983)
14. Parak, P., Knapp, E.W.: Proc. Natl. Acad. Sci. Biophys. **81**, 7088 (1984)
15. Felner, I., Nowik, I.: Supercond. Sci. Technol. **8**, 121 (1995)
16. Nowik, I., Felner, I.: Phys. C –Supercond. Appl. **469**, 485 (2009)
17. Friedman, Y.: Ann. Phys. **523**, 408 (2011)
18. Josephson, B.D.: Phys. Rev. Lett. **4**, 341 (1960)
19. Perlow, G.J.: Phys. Rev. Lett. **40**, 896 (1978)
20. Monahan, J.E., Perlow, G.J.: Phys. Rev. A **20**, 1499 (1979)
21. Wertheim, G.K.: Phys. Rev. **161**, 478 (1967)

22. Nowik, I., Wickman, H.H.: Phys. Rev. **140**, 860 (1965)
23. Nowik, I.: Hyperfine Interact. **13**, 89 (1983)
24. Mössbauer, R.L.: Hyperfine Interact. **33**, 199 (1987)
25. Nowik, I., Felner, I., Ren, Z., Cao, G.H., Xu, Z.A.: J. Phys. Condens. Matter **23**, 065701 (2011)
26. Nowik, I., Felner, I., Zhang, M., Wang, A.F., Chen, K.H.: Supercond. Sci. Technol. **24**, 095015 (2011)