A SUMMARY OF THE INTERNATIONAL CONFERENCE ON
HYPERFINE INTERACTIONS STUDIED BY NUCLEAR
REACTIONS AND DECAY*

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(A talk presented to the conference on June 15, 1974, in Uppsala)

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Five days ago the conferees assembled in Uppsala to attend this International Conference on Hyperfine Interactions Studied in Nuclear Reactions and Decay. Any conference with a title of this length can be assumed to have had several predecessors. The direct forebears of this Conference were the 1963 Uppsala Conference on Perturbed Angular Correlations [1], the 1967 International Conference on Hyperfine Interactions Detected by Nuclear Radiations at Asilomar [2], and the 1970 International Conference on Hyperfine Interactions Detected by Nuclear Radiation at Rehovoth and Jerusalem [3]. Other major topical conferences have included those on Mössbauer spectroscopy [4] and the 1972 International Conference on Nuclear Moments and Nuclear Structure at Osaka [5]. To summarize the present conference effectively we must fit it into the context established by these earlier meetings. Before embarking on a detailed discussion of advances reported at this conference, it therefore seems appropriate to note in broad overview a few of the evolutionary mainstreams of this field. Five trends are noted below:

(i). The quality and sophistication of measurements is improving significantly. Time-differential studies are displacing time-integral ones, and more effort is being devoted to working with well-characterized systems. Longer-lived levels, with narrower natural widths, are receiving more attention.

(ii). The center of gravity of interest has shifted from nuclear physics to extranuclear phenomena. This seems inevitable in view of the numbers of cases in the two areas: One might say that nuclear moments will be "used up" before hyperfine fields. At the same time accurate measurements of nuclear moments continue at a very substantial rate, and the field retains a strong nuclear base.

(iii). There is some shift of attention from static to dynamic phenomena such as relaxation times.

(iv). The proportion of Mössbauer spectroscopy studies in this type of conference is decreasing. This implies no decrease in the application of Mössbauer spectroscopy (the contrary is certainly true!), but simply that this technique has reached maturity and become routine faster than have the various angular correlation methods.

(v). An earlier trend from nuclear decay to nuclear reactions has slowed down. Many of the studies reported in this conference were based on nuclear decay, especially involving the old standby ^{111}Cd .

Let us turn now to a discussion of specific conclusions reached in this conference. The chronological sequence of the conference sessions provides a convenient logical sequence, which is followed below.

In the first session I. Lindgren [6] described the calculation of hyperfine fields in atomic systems. Although hyperfine interactions have been known for half a century, truly quantitative (1%) calculations of hyperfine interaction strengths in even the "simple" alkali atoms are not yet readily accessible. Unrestricted Hartree-Fock calculations, even with the inclusion of relativistic effects (RUHF) still fall ~ 10-20% short of the experimental values of a_{ns} . Clearly configuration interaction must be considered, and theoretical interest is now being focussed on developing new perturbational approaches; e.g., linked-diagram expansions. These methods appear promising because they are designed explicitly to solve hyperfine-structure problems. We may expect real progress in this important fundamental area to be reported in future conferences.

T. P. Das [7] described hyperfine structure calculations in metals. Two aspects of this subject deserve comment. First, conventional electron band-structure calculations are made using a local-potential approximation to the exchange interaction, usually the Slater exchange potential or one of its variants. Now Hartree-Fock-Slater (HFS) results can be at best only comparable to HF theory (i.e., with optimized exponents); thus ordinary band-structure calculations are simply not sufficiently sophisticated to give more than a rough estimate of hyperfine-structure parameters. An outstanding problem in band-structure theory is to bring the basic calculations up to the Hartree-Fock self-consistent field level. It may then be feasible to include the subtler effect of electron-electron correlation that will be needed for quantitative estimates of hyperfine parameters.

The calculation of electric field gradients in metals has received new attention since experimental determinations of the sign of the quadrupole coupling constant, e^2qQ , have become available. It should be noted in this context that both Mössbauer spectroscopy and nuclear orientation yield the sign of e^2qQ readily. Both methods have been used to this purpose by Kaindl and co-workers [8,9] and others. Raghavan, et al. [10] have shown that angular correlations can also give the sign of e^2qQ if the nuclei are first polarized, by using β - γ correlations. From a theoretical point of view, studies of e^2qQ are very reliable in improving our physical picture of electrons in metals. The reason for this is that there are two main contributors to the field gradient eq: ion cores and electrons in valence bands (sometimes inaccurately called conduction electrons). In most cases these two contributions are of opposite sign and tend to cancel, because the valence electrons still have considerable atomic character and tend to concentrate near the ion cores. For free-electron metals such as Be and Mg, calculations show that the ion cores' contributions to e^2qQ dominate [7]. This can be attributed to the diffuseness of the valence-electron distribution in free-electron metals. For a uniform charge distribution $q \equiv 0$. Metals such as Cd and Zn, on the right side of the periodic table, have more localized valence electrons and large, easily polarizable closed shells. For these metals calculations show that the total field gradient should have opposite sign from that of the ion cores alone [7]. Experimentally most sign measurements have been made on solute atoms (e.g. ^{111}Cd) in host lattices. This introduces an additional complication. For experiments performed to date, it appears that the solute is very important. Thus for ^{111}Cd , the sign of e^2qQ is

opposite to that of the lattice in all the above metals - Be, Mg, Cd, and Zn - while in the free-electron metal titanium they have the same sign [11].

A related problem also received some attention in this conference. This is the question of the effective charge to be assigned to atoms in host metals in crystalline-field situations. X-ray photoemission experiments on the d-bands of Au and Ag can be interpreted in terms of a crystal-field model only if the neighboring atoms have negative effective charges (as in Cd and Zn above). The sign of the $A_4^0 < r^4 >$ term is also consistent with negative charges on Ag and Cu atoms in ErCu and ErAg as determined by inelastic neutron scattering [12] and on Sb and Bi in LaSb, LaSb and LaBi, as determined by electron spin resonance [13]. It is already clear that point-charge models will not give consistent results for $A_4 < r^4 >$ and $A_6 < r^6 >$ however [13], presumably because different radial moments of the electron charge distribution are involved. Conversely, additional information about this distribution can be obtained by determining these parameters separately. We can expect much activity in this interesting area in the near future.

Physical theory makes heavy use of the "test charge" concept, in which a small charge is used to probe electromagnetic fields. In hyperfine interactions the nucleus serves as a convenient test charge. Two other probes of the inner regions of atoms were discussed in this conference, as well as one probe of interatomic regions.

Polarized negative muons are readily captured into atomic 1s orbitals that lie inside the occupied electronic orbitals, where their precession can be studied by PAD techniques. T. Yamazaki and co-workers

have pointed out that a captured μ^- thereby shields electrons from one unit of nuclear charge [14], leading to a local electron distribution in element Z characteristic of element Z-1 (e.g. Mn in Fe). As yet no effect has been observed in magnetic materials, but a intensive search is underway by the Tokyo-Berkeley group [14].

Core electrons also serve as test probes of the interiors of atoms, because core orbitals are highly localized. Crystal-field and local-moment effects can be detected in photoemission spectra, and there appear to be several points of contact between hyperfine interactions and x-ray photoemission (ESCA) spectroscopy [15].

Polarized positive muons come to rest and decay in interstitial regions, where they sense a very different region of charge distribution [16]. Our usual hyperfine concepts do not apply very readily to μ^+ studies, and it is too early to make general conclusions about this new technique, except to note that it has one characteristic necessary for success: it yields beautiful data.

A brief digression seems appropriate at this point. In searching for weak signals in precession data it is useful to Fourier transform the data into frequency space: most workers now use this approach. Another more powerful technique has been widely overlooked, however; this is the autocorrelation of the data before Fourier transforming them. This two-step process, well-known in other areas of data analysis, has been found to be much more effective in analyzing angular correlation data than Fourier transformation alone [17].

The second conference session has devoted to recoil into vacuum,

gas, and solids and to radiation damage studies on implanted sources. The third session was concerned with nuclear moments, and the results have already been ably summarized by R. Kalish. In this summary we shall therefore omit the nuclear moment results and comment on the techniques employed in Sessions II and III taken together.

The recoil-into-vacuum studies are turning out very well. We must single out for special mention the time-differential studies on short-lived states in light nuclei, in which time is measured by the recoil-distance method. When applied to one-electron atoms this method was given the acronym RED/ONE (for REcoil Distance/ONE-electron atoms) [18]. Two applications of this beautiful technique were reported at this conference: a measurement of the g-factor of the 1^+ state in ^{16}N and a measurement of the hyperfine interval in lithium-like ^{19}F . This method enjoys the natural superiority of time-differential techniques, and it should find wide application to light elements in the future.

M. Goldberg [18] pointed out the advantages of MAD/TWO methods (MAGnetic Decoupling/ $n = 2$ shell) for heavier elements with hyperfine frequencies too large for the RED/ONE approach. Although many optical levels are available to ions with unpaired 2s electrons, magnetic decoupling greatly reduces the complexity of the system. Arguments can be made that the effective magnetic field can be deduced without understanding all the details of the ionic configurations. The accuracy of MAD/TWO-derived magnetic moments remains to be assessed, but we note that for the 3^- level at 6.13 MeV in ^{16}O the RED/ONE and MAD/TWO values coincide [19,20]. We can expect more such comparison cases to be studied.

It appears that the RED/ONE method may be applicable up to elements somewhere between Na ($Z = 11$) and K ($Z = 19$) where the MAD/TWO method may take over.

The evolution of the IMPAC technique is almost a closed chapter in the story of hyperfine interactions, and it has been a very interesting chapter indeed. The discovery and explanation of transient magnetic fields at nuclei of ions moving through ferromagnets has stimulated considerable thought, and the Lindhard-Winther theory [21] gives a satisfactory semiquantitative description of the transient fields. The inherent difficulty and inaccuracy of the IMPAC method preclude its use for measuring g-factors to the accuracy usually required to give definitive answers to nuclear structure questions, but the internal discrepancies are slowly being resolved [22]. D. E. Murnick reported that corrections for radiation damage in heavy-ion beams brings most of the existing data onto a smooth ϕ/g -vs- Z curve [23], while J. L. Eberhardt [24] showed that the g-factor of the 1.37 MeV, 2^+ state in ^{24}Mg , from RED/OIE measurements, falls nicely onto this curve.

Implantation studies have moved to a higher level of sophistication. A separate meeting was held at Dieppe, on the subject of radiation damage and hyperfine interactions, immediately preceding the present conference. H. Bernas [25] and H. de Waard [26] have summarized the state of the art generally and the Dieppe meeting specifically, and a further summary here would be redundant. The take-home lesson that they left with us is that many supposed hyperfine-structure studies on sources prepared by ion implantation have in fact been radiation-damage studies. The doses of $\sim 10^{14}$ atoms cm^{-2} that have often been used are far too

large to produce intrinsic samples. Much lower doses are needed: just how low is currently being investigated by a combination of conventional radiation damage techniques (e.e., electron microscopy) and hyperfine techniques. The latter are becoming more sophisticated, making use, for example, of time-differential studies of field gradients due to the presence of defects. C. Abromeit and H. Gabriel have made a theoretical investigation of these field gradients [27], finding time-differential curves that are tantalizingly similar to the experimental data for the AgCd case [25].

It is interesting to review the history of hyperfine measurements with implanted samples, including both source and IMPAC work. At first some investigators who thought they were doing nuclear physics were in reality measuring extranuclear properties of their samples. Attention then focussed on the solid-state aspects of the measurements as investigators realized that extranuclear properties could not simply be described in terms of a unique, stationary effective magnetic field. It has become clear that the problems actually under study involved ions passing through a polarized, degenerate Fermi gas in the case of IMPAC, and radiation damage in the implanted-source case. It is gratifying that hyperfine interactions in excited nuclei can be applied so effectively to the study of metallurgical problems, but unfortunate that this application was approached in such a roundabout way!

Before leaving the subject of nuclear moment measurements, two predictions of nuclear-structure theory should be noted. K. Kumar has suggested [28] that a new quantum number κ , rather than K , may be a good quantum number for spherical nuclei. This model, which requires admixing

of particles not in time-reversal conjugate states, would explain the $0^+, 2^+, 4^+$ sequence and the large value of $Q(2^+)$ in these nuclei. It would predict high values of $Q(\text{intrinsic})$ for high-spin states and nonzero spectroscopic quadrupole moments in 3^+ states. Neither of these predictions appears to be testable with present experimental techniques on known states, but discovery of states with accidentally long lifetimes could change this conclusion.

Ward, et al. [29] have raised an interesting question on the basis of their vacuum deorientation work on states in ^{150}Sm and ^{156}Gd . They found no evidence for increase of deorientation with I , suggesting that the results of Nordhagen et al. [30] might have arisen in part from decreasing g -factor with increasing I , due to "backbending" of the moment of inertia. This is an important open question.

Session IV was devoted to solid-state studies, and it became apparent that some very sophisticated measurements have been made with NMR and angular correlation methods. J. Budnick [31] reported spin-echo studies on ferromagnetic Fe_3Si and alloys derived from this compound by small changes in composition (e.g. $\text{Fe}_{0.77}\text{Si}_{0.23}$) and/or addition of small amounts of impurities. The NMR spectra are profoundly affected by these changes, indicating great sensitivity and showing that the spin-echo method will be very useful for studying spin-exchange effects in ferromagnets.

The Uppsala angular correlations group reported a detailed study of the PAC pattern of $^{111}\text{CdNi}$ during magnetization [32]. They found no evidence for a large "cone angle" between the external field and the impurity hyperfine field, in contrast to earlier results with other systems [33]. Perhaps this is because the ^{111}Cd hyperfine field arises

from conduction electron polarization, while in systems with localized moments a cone angle is possible. Further work on this perplexing problem is clearly needed.

C. Hohenemser gave a critical discussion of the possible probe dependence of critical exponents [34]. There is no convincing evidence for a probe dependence of β in the equation

$$M \propto \left(\frac{T-T_c}{T_c} \right)^\beta$$

for the nickel lattice. The few experiments that yield β values appreciably different from 0.38 all suffer from a depression of T_c by ~ 10 degrees Celsius, from analysis of data over a narrow range of $(T-T_c)/T_c$, or both. Clearly great care is needed in these sophisticated experiments.

N. J. Stone and co-workers at Oxford have carried out some elegant NMR/ON experiments based on the existence of very small electric field gradients, collinear with H_{hf} , at impurities in ferromagnets [35,36]. The effect of the field gradient is to make the level spacings unequal for a given nucleus. Thus a single-passage experiment successively inverts the level populations pairwise, and cyclically permutes the populations. Fast passage NMR/ON can be used to study esoteric relaxation effects and to measure quadrupole effects that are much too small to resolve directly. This work greatly extends the versatility of NMR/ON.

P. Heubes et al. [37] reported and discussed the fact that quadrupole resonance frequencies in solids vary as

$$v_T = v_0 + AT^{3/2}.$$

The $T^{3/2}$ dependence was followed so accurately for several lattices over a wide temperature range as to imply that it should follow from some simple model of lattice dynamics. Quitmann, et al [38] have proposed a thermal-vibration model in which the ion cores are screened by conduction electrons. Using values of $\langle x^2 \rangle$ from x-ray scattering they have calculated the temperature dependence of v in iridium quite accurately. It would be premature to infer that this is the final explanation, but it appears quite promising.

The above examples were selected for special comment from among 57 papers contributed to Session IV. Several other sets of papers could have been chosen. Rather than discuss further examples, however, let us note a general theme. Hyperfine interactions in solids permit the study of atom-neighbor interactions on a microscopic scale. This is of particular value in metals, to which other spectroscopic techniques are not readily applicable. Magnetic hyperfine interaction are useful for studying atom-neighbor effects in magnetic materials, because of the great sensitivity of exchange coupling strengths to distance. In non-magnetic materials, magnetic hfs constants are of little diagnostic value for atom-neighbor effects. The electric quadrupole interaction is very sensitive to defect and solute effects in these materials, however. In favorable TDPAC cases it is possible to resolve several frequencies. We believe that one of the main advances reported in this conference is the ability to use quadrupole coupling TDPAC studies to study atom-neighbor effects. A

number of papers in Session IV reported such studies,

Session V was devoted to relaxation. One theme that emerged was the wide applicability of isomeric nuclear states for studying time and/or temperature regions not accessible to conventional techniques. Thus D. Riegel [39] showed that PAD measurements with the 20 msec state of ^{71}Ge in liquid gallium were consistent with a diffusive quadrupolar relaxation mechanism. H. Ackermann, et al. studied the β -decay asymmetry from polarized ^8Li in Li metal as a function of applied field and temperature [40]. Their results were consistent with mono-vacancy diffusion at low temperatures, in agreement with results on stable lithium, but showed evidence for a divacancy mechanism at higher temperatures.

A. Nacath described relaxation processes affecting nuclei of atoms possessing local moments [41]. Again as in several other talks in the conference, basic questions about local moments and crystal fields in metals were raised and partially answered. Thus the relaxation rates for the system ^{59}Co are consistent with $S = 3/2$, as expected for Co^{2+} , while the low value of H_{hf} is presumably due to orbital admixtures arising through crystal-field effects, which resolve the ^4F term into $^2\text{A}_2$, etc., levels. Finally, the signs of the effective charges on Mo and Au lattice sites appear to be positive and negative, respectively, which is qualitatively consistent with expectations based on quadrupole-coupling studies.

This brief summary was made to pull together some of the main points of the conference. The conferees will have many opinions about which topics should have been selected. With 146 contributed papers to choose from, a consensus is unlikely. Nor is one desirable; each participant should make his own conference summary. One primary point on which we

can all agree, however, is that the field of hyperfine interactions studied by nuclear reactions and decay is flourishing and that it is certain to provide exciting material for future meetings.

References

1. "Perturbed Angular Correlations," edited by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland, 1964).
2. "Hyperfine Interactions and Nuclear Radiations," edited by E. Matthias and D. A. Shirley (North-Holland, 1968).
3. "Hyperfine Interactions in Excited Nuclei," edited by G. Goldring and R. Kalish (Gordon and Breach, New York, 1971).
4. Reviews of Modern Physics, Vol 36, No. 1, Part 2 (1964).
5. J. Phys. Soc. Japan, Volume 34 Supplement, (1973).
6. I. Lindgren, invited paper, Session I, this conference.
7. T. P. Das, invited paper, Session I, this conference.
8. G. Kaindl, D. Salomon, and G. Wortmann, Phys. Rev. Letters 28, 952 (1972).
9. G. Kaindl, P. Bacon, and A. J. Soinski, LBL-1960 (submitted to Phys. Letters B)
10. R. S. Raghavan, P. Raghavan, and E. N. Kaufmann, Phys. Rev. Letters 31, 111 (1973).
11. E. N. Kaufmann, P. Raghavan, and R. S. Raghavan, paper IV. 29, this conference.
12. P. Morin, J. Pierre, J. Rossat-Mignod, K. Knorr, and W. Drexel, Phys. Rev. B9, 4932 (1974).
13. P. Davidov, E. Bucher, L. W. Rapp, Jr., L. D. Longinotti, and C. Rettori, Phys. Rev. B9, 2879 (1974).
14. T. Yamazaki, Invited paper, Session I, this conference.
15. D. A. Shirley, Invited paper, Session I, this conference.
16. Positive-muon studies were reported in papers I-1, I-2, I-4, and I-5, this conference.

17. E. Matthias and D. A. Shirley, Nucl. Instr. Methods 45, 309 (1966).
18. M. Goldberg, Invited paper, Session III, this conference.
19. W. L. Randolph, N. Ayres de Campos, J. R. Beene, J. Burde, M. A. Grace, D. F. H. Start, and R. E. Warner, Phys. Letters 44B, 36 (1973).
20. C. Broude, M. B. Goldberg, G. Goldring, M. Hass, M. J. Renan, B. Sharon, Z. Shkedi, and D. F. H. Start, Nucl. Phys. A215, 617 (1973).
21. J. Lindhard and A. Winther, Nucl. Phys. A166, 413 (1971).
22. A. Gelberg, Invited paper, Session II, this conference.
23. G. K. Hubler and D. E. Murnick, paper II. 21, this conference;
G. K. Hubler, H. W. Kugel, and D. E. Murnick, Phys. Rev. C9, 1954 (1974).
24. J. L. Eberhardt, R. E. Horstman, H. W. Heeman, and G. van Middelkoop, paper III. 8, this conference.
25. H. Bernas, Invited paper, Session II, this conference.
26. H. de Waard, Invited paper, Session II, this conference.
27. C. Abromeit and H. Gabriel, papers II. 11-12, this conference.
28. K. Kumar, Invited paper. Session III, this conference.
29. D. Ward, H. R. Andrews, R. L. Graham, J. S. Geiger, and Neils Rad, Paper II. 4, this conference.
30. R. Nordhagen, G. Goldring, R. M. Diamond, K. Nakai, and F. S. Stephens, Nucl. Phys. A142, 577 (1970).
31. J. Budnick, Invited paper, Session IV, this conference.
32. E. Karlsson, B. Lindgren, and B. Jonsson, Paper IV. 1, this conference.
33. K. Krane, B. T. Murdoch, and W. A. Steyert, Phys. Rev. Letters 30, 321 (1973), and references therein.
34. C. Bohnenmeyer, Paper IV. 14, this conference.

35. P. T. Callaghan, P. D. Johnston, and N. J. Stone, Paper IV. 30, this conference.
36. P. T. Callaghan, P. D. Johnston, W. M. Lattimer, and N. J. Stone, Paper IV. 55, this conference.
37. P. Heubes, G. Hempel, H. Ingwersen, R. Keitel, W. Klinger, W. Loeffler, and W. Witthuhn, Paper IV. 33, this conference.
38. D. Quitmann, K. Nishiyama, and D. Riegel, private communication.
39. D. Riegel, invited paper, Session V, this conference.
40. H. Ackermann, D. Dubbers, H. Grupp, M. Grupp, P. Heitjans, W. Hell, and H. J. Stöckmann, Paper II. 13, this conference.
41. A. Narath, invited paper, Session V, this conference.