spin disorder acts qualitatively very much as though it were at zero frequency. But the fluctuating fields due to spin disorder occur at frequencies comparable to J/h, where J is a characteristic exchange coupling, and for this system we can take J to be of the order of 6°K (the specific-heat results of Franck, Manchester, and Martin show a maximum at this temperature for the three alloys they studied). The fluctuation time is therefore of the order of 10^{-10} sec, and this is too short for the nucleus to respond effectively. Therefore, if this rough estimate is correct, the Mössbauer experiment should see only the distribution of fields due to the geometrical effect.

We are unable to give any plausible explanation of why the width of our $p(H)$ curves increases with increasing temperature. However, it is worth noting that in our alloys the nearest-neighbor exchange interaction is playing a more important role than in the alloys where the specific-heat anomalies have been observed. For example, in the 2.6% alloy there is a 30% chance that an Fe atom has a nearest neighbor which is also Fe. Assuming this nearest-neighbor interaction is antiferromagnetic and stronger than the more distant interactions, it seems likely that our samples must contain a good deal of zero-point motion in the spin system. This qualitative idea is supported by the observation that the entropy deduced from the specific-heat results on an 0.2% alloy showed that the spins at 1.3°K were almost completely aligned, whereas our Mössbauer measurements on higher concentrations show that, at the same temperature, the majority of the spins are only partially aligned. We believe the present results indicate that the experiments could be profitably pursued at lower concentrations and lower temperatures.

CONTRIBUTED PAPERS FOR SESSION V

Magnetic Shielding and Local Moments of Fe^{57} Impurities*

R. D. Taylor, W. A. Steyert, and D. E. Nagle, *Los Alamos Scientific Laboratory*

The hyperfine field of Fe^{57} has been studied as a function of temperature and external magnetic field for sources composed of Co^{57} diffused into such host metals as Cu, Ti, Nb, Au, Pd, and Pt. In zero external field all of the sources, except Ti, showed a single narrow Mössbauer line at all temperatures from 300 to 4°K. Titanium, not being cubic, gave a broader line. At room temperature these sources showed little, if any, shielding of the Fe^{57} from the external field, i.e., the observed partially resolved Mössbauer hyperfine spectrum directly corresponded to the applied field. At 4°K a remarkable magnetic shielding is apparent for Cu, Ti, and Au, while little shielding was found for Nb, Pd, and Pt.

We define the shielding constant $a = (H - H_{\text{eff}} + H_i)/H$, where H is the applied field, H_{eff} is the field at the Fe^{57} impurity deduced from the Mössbauer spectrum, and, where present, H_i is the internal field. We find a (4°K) is 0.45 ± 0.04 for Cu, 0.23 ± 0.05 for Ti, and 1.0 ± 0.1 for Au. The value of a for a given source at 4°K is nearly constant, independent of H, up to 60 kOe, the maximum field used. For Cu, a was studied in detail as a function of temperature with $H = 60$ kOe. A reasonable fit to the data is given by $a = 8.1/(T + 13)$. Circular polarization measurements showed H_{eff} for Cu to be positive with respect to H.

Previous studies of the localized moment of Fe^{57} in Pd and Pt[1] assumed zero shielding of the impurity from the external field. A somewhat improved fit of the data is possible if one allows a to be 0.05 and 0.11, respectively. A similar treatment of new data on Fe^{57} in Au requires a to be 1.0 ± 0.1. The local moment observed is 2.0 ± 0.2 Bohr magnetons if $J = 1$ is assumed and the saturation internal field is -120 ± 2 kOe.

* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] P. P. Craig, D. E. Nagle, W. A. Steyert, and R. D. Taylor. Phys. Rev. Letters **9**, 12 (1962).

High Field Studies of Localized Moments in Metals

N. Blum and A. J. Freeman, *National Magnet Laboratory**
L. Grodzins, *Physics Department, Laboratory for Nuclear Science,† MIT*

In this paper we report on a series of Mössbauer experiments in high external magnetic fields. Such measurements have been shown to give new information concerning localized magnetic moment distributions in metals and alloys.[1–3]

The hyperfine field at the Fe^{57} nucleus in a number of alloys of the 3*d* and 4*d* transition metals was measured as a function to temperature and applied magnetic field over the range 1.5 to 300°K and up to 110 000 Oe in both longitudinal and transverse field geometries using high current Bitter solenoids. The spectrometer, which is similar to those described elsewhere,[4] uses a constant acceleration drive system to produce a linear velocity spectrum, and a multichannel analyzer for handling and storing the data. Results for 1% Fe^{57} in Ti and Sc are given; these show no additional contributions to the hyperfine field at any temperature studied. Alloys with 1% Fe^{57} in Mo and Rh exhibit large negative temperature-dependent contributions to the hyperfine field at low temperatures. These results contrast with recent experiments using Co^{57} in Mo and Rh as sources,[2] where no anomalous contributions to the hyperfine field were observed. Experiments using Co^{57} in Cu as a source also exhibit large temperature-dependent negative contributions to the hyperfine field at low temperatures. Details of the results are given, and the data are analyzed and discussed in terms of current theoretical views on localized magnetic distributions in metals.

* Supported by the U. S. Air Force Office of Scientific Research.
† Supported by the U. S. Atomic Energy Commission.

[1] N. Blum and L. Grodzins, Bull. Am. Phys. Soc. **7**, 39 (1962).

[2] P. P. Craig, D. E. Nagle, W. A. Steyert, and R. D. Taylor, Phys. Rev. Letters **9**, 12 (1962).
[3] A. J. Freeman, Phys. Rev. **130**, 888 (1963).
[4] H. Frauenfelder, *The Mössbauer Effect* (W. A. Benjamin, Inc., New York, 1962), p. 43.

On the Measurement of the Sign and Magnitude of Internal Fields*

N. Blum, *National Magnet Laboratory, MIT*
L. Grodzins, *Laboratory for Nuclear Science, MIT*

The sign of the spontaneous internal field on an iron nucleus has been traditionally determined by measuring the change in the position of the hyperfine lines upon application of an external field.[1] Other techniques are necessary for induced internal fields. We have determined the sign of the induced field in metal–iron alloys by comparing the internal field direction to that in metallic iron. The source and absorber —one of which is metallic iron—are placed in a longitudinal magnetic field [$H_{ext} \ll H_{int}$ (Fe)] and the 8-line absorption pattern observed. The hyperfine patterns for parallel and antiparallel fields are quite dissimilar.[2] For example, consider the doublet at the pattern extremity: If the field directions in source and absorber are parallel, the intensity ratio for the doublet lines is, ideally, 3:1; if antiparallel, the ratio is 1:3. The doublet separation appears to be a more sensitive, less ambiguous, measure of the unknown internal field than that obtained from the quartet- or sextet-line pattern which results from using a single exploratory line. The sign of the induced internal field in ~1% Fe alloys of Cu and Cr are positive.

* Supported by the U. S. Atomic Energy Commission and the Air Force Office of Scientific Research.
[1] S. S. Hanna, J. Heberle, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. Letters **4**, 513 (1960).
[2] H. Frauenfelder, D. E. Nagle, R. D. Taylor, D. R. F. Cochran, and W. M. Visscher, Phys. Rev. **126**, 1065 (1962).

Mössbauer Effect in Fe^{57} and Sn^{119} Metals in Large External Magnetic Fields*

J. Heberle, M. Schulhof, and S. S. Hanna, *Argonne National Laboratory*

With the successful operation of a superconducting magnet designed for Mössbauer research, it appeared of interest to study the Mössbauer spectra of pure tin (nonferromagnetic) and pure iron (ferromagnetic) in large magnetic fields. Several strange and interesting effects have been reported[1,2] in various alloys and compounds in magnetic fields. In a field of 48 kG (applied along the direction of the gamma rays), a sample of white tin produced a 4-line absorption spectrum (as expected). The positions of the lines were determined by fitting 4 Lorentzian lines with a least-squares program. The ratio of the magnetic moment of the excited state to that of the ground state was determined to be $\mu_{ex}/\mu_g = -0.68$. In an isotropic sample no first-order quadrupole effect is expected, and no significant one was observed. In addition, no significant change in the isomer shift was detected when the external field was changed from 0 to 48 kG. In the case of iron the two inner lines were measured in fields of 11 and 48 kG. Again, no significant change in the isomer shift was detected, and the increase in applied field decreased the internal field by 27 kOe.

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] N. Blum and L. Grodzins, Bull. Am. Phys. Soc. **7**, 39 (1962).
[2] R. W. Grant, M. Kaplan, D. A. Keller, and D. A. Shirley, Bull. Am. Phys. Soc. **7**, 601 (1962).

Manganese–Tin Alloys in Large External Magnetic Fields*

S. S. Hanna, J. Heberle, J. Diaz, and R. W. Reno, *Argonne National Laboratory*

The earlier investigation[1] of manganese–tin alloys with the Mössbauer effect showed that application of an external magnetic field decreased the Zeeman splitting of Sn^{119} in Mn_3Sn ($T \approx 300°K$) but increased it in Mn_2Sn ($T \approx 250°K$). In the latter case the temperature was only a few degrees below the Curie point T_c ($\approx 260°K$). By means of a superconducting magnet, these measurements have now been extended to high fields and low temperature. At 4.2°K the application of a field of 46 kG (along the direction of the gamma radiation) to Mn_2Sn produced a decrease in the Zeeman splitting in contrast to the increase observed near T_c. As pointed out by H. E. Hall (private communication from A. J. F. Boyle), the behavior near T_c may be attributed to an increase in saturation magnetization produced by the applied field. Near T_c this effect could cause an increase in the field at the nucleus which is larger than the applied field. The result at 4.2°K indicates that the internal field at the tin nucleus in ferrimagnetic Mn_2Sn is negative. This result makes the existence of *negative* internal fields even more prevalent. In Mn_3Sn no clear-cut result was achieved at 4°K. On application of an external field, the lines broaden (or become double). This broadening is perhaps related to the complex magnetic structure observed previously.[2,3]

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] L. Meyer-Schützmeister, R. S. Preston, and S. S. Hanna, Phys. Rev. **122**, 1717 (1961).
[2] K. Yasukochi, K. Kanematsu, and T. Ohoyama, J. Phys. Soc. Japan **16**, 1123 (1961).
[3] L. Meyer-Schützmeister, *The Mössbauer Effect, Second International Conference, Saclay, France, 1963* (John Wiley & Sons, Inc., New York, 1962), p. 190.

Systematics of Conduction-Electron Polarization of Impurity Atoms*

D. A. Shirley, R. W. Grant, and G. A. Westenbarger, *Department of Chemistry and Lawrence Radiation Laboratory, Berkeley*

Using the Mössbauer effect in Au^{197} the signs and magnitudes of magnetic fields induced at nuclei of Au atoms in very dilute solution in metallic Fe, cubic and hexagonal Co, and Ni were determined by direct comparison with an external magnetic field. The fields were all negative and of magnitudes 1420 (180), 980 (120), 990 (120), and 340 (60) kG, respectively. Errors are given parenthetically. These fields are roughly proportional to the saturation magnetization of the lattices. In nuclear polarization experiments on Ag^{110m} in Fe and Ni lattices, magnetic field–nuclear moment products of approximately -13×10^5 and -4×10^5 G-nm, respectively, were determined. Again the magnitude of the internal field is proportional to the magnetization of the host and is very roughly 500 kG in Fe. These fields, together with those for other heavy diamagnetic

atoms dissolved in iron, are shown, in the manner of Marshall and Johnson,[1] to be consistent with a "conduction" electron polarization of ≈7%, by comparison with hyperfine coupling constants from atomic spectroscopy. Contributions of conduction-electron polarization to internal fields are thus estimated for the simpler heavy metals (dissolved in iron) throughout the Periodic Table. In particular, this mechanism might be expected to contribute a component to the internal field from 6*s* electron polarization which increases in magnitude monotonically by about a factor of 10 from Cs to Au.

* Work sponsored by U. S. Atomic Energy Commission.
[1] W. Marshall and C. E. Johnson, J. Phys. Radium **23**, 733 (1962).

A Correlation of Isomer Shifts of Gold in Several Dilute Gold Alloys with the Alloy Residual Electrical Resistivities

Louis D. Roberts, Richard L. Becker, F. E. Obenshain, *Oak Ridge National Laboratory*
J. O. Thomson, *University of Tennessee*

The isomer shifts of the 77-keV γ of Au^{197} have been measured for a number of 1% gold alloys using a metallic Pt source at 4.2°K. These shifts are compared with the predictions of a simple theoretical model. In this model the modification of the electron distribution at the gold impurity is obtained from a perturbing potential. The *s*- and *p*-wave phase shifts are inferred from the residual electrical resistivity of the alloy together with the Friedel Sum Rule, including the effects of relative sizes and valences of the impurity and host atoms. A square well which reproduces the *s*-wave phase shift at the Fermi energy is used to obtain the continuation of the *s*-wave function inward to the impurity nucleus. The same well is assumed to hold for all wave numbers below the Fermi level. The isomer shift is then found to be proportional to $[Z(r_{Au}/r_h)^3 P_{av} - 1]$, where Z is the effective number of *s*-band electrons per host atom, r_{Au} and r_h are radii of gold and the host, and P_{av} is the average over the *s* band of the "charge polarization factor" $P(k)$ used at the Fermi surface by Daniel in a calculation of the Knight shift on an impurity. The experimental isomer shifts for 1 at. % Au^{197} in Ag, Pd, and Pt are 2.15, 2.4, and 1.37 mm/sec relative to pure gold. Determining a proportionality constant from the Ag alloy, the experimental shifts for Pd and Pt are obtained from the calculation assuming *s*-band fillings of 0.69 and 0.47, respectively. The calculated values are quite sensitive to the assumed *s*-band fillings.

The Magnetic Splittings and Relative Shifts of the 14.4-keV Fe^{57} Absorption Line in Host Lattices of Iron Isotopes

G. R. Isaak and G. Longworth, *University of Birmingham, England*

A search for an effect of isotopic mass of the host lattice on the magnetic field at the nucleus of an impurity atom is reported.

Differences in the hyperfine splittings of the 14.4-keV Mössbauer line of ^{57}Fe nuclei in lattices of iron isotopes were looked for using a sensitive Doppler-scanning technique.

The magnetic fields at the ^{57}Fe nuclei in metallic ^{54}Fe and ^{56}Fe foils prepared by identical processes are the same to within the experimental accuracy of about 5 parts in 10^4 (150 G).

Preliminary measurements of the relative shifts of the ^{57}Fe resonance line in lattices of the various isotopes, at 300 and 80°K, are reported.

Direct Measurement of the Internal Field in Nickel

F. E. Obenshain and H. H. F. Wegener,* *Oak Ridge National Laboratory*

The internal magnetic field H_e for the Mössbauer nucleus Ni^{61} in metallic nickel has been determined by application of an external magnetic field which ranged from 2.8 up to 24.0 kG. The hyperfine splitting of the Ni^{61} Mössbauer radiation shrinks when the external field is applied, indicating a negative hyperfine field. Comparing the observed changes in the absorption spectra with computer calculated absorption curves (taking into account absorber-thickness, etc.) yields $H_e = -(90 \pm 20)$ kG assuming additivity of the fields. This result is in contradiction with the older NMR results, but it agrees with the new NMR results published recently by Streever.[1] The previous value obtained at ORNL for the ratio of the nuclear moments is, of course, still valid and is $\mu_e/\mu_g = -0.47 \pm 0.08$; taking the new value $\mu_g = 0.70 \pm 0.04$ for the ground-state moment we obtain $\mu_e = -0.33 \pm 0.06$ for the first excited state. The magnetic moments for Ni^{61} in the ground and first excited states are therefore about 2.3 times larger than originally reported.

* Present address: University of Erlangen, Erlangen, Germany.
[1] R. L. Streever, Phys. Rev. Letters **10**, 232 (1963).

Isomer Shift and Electric Quadrupole Coupling Measurements in Several Hexagonal Close Packed Metals*

R. Segnan and Bernard Mozer, *Brookhaven National Laboratory*

We have measured room temperature Mössbauer spectra for Fe^{57} in the following hcp metals: Cd, Co, Dy, Gd, Mg, Ru, (Se), (Te), Ti, Zn. Co^{57} was diffused into these metals and used as the source. The absorber was 3.85 mg/cm² of ferrous potassium cyanide. The isomer shift ΔE and electric quadrupole coupling constant 2ϵ have been previously observed in Be by Kistner and Mozer,[1] who give the value $\Delta E = -0.11 \pm 0.01$ mm/sec and $2\epsilon = 0.58 \pm 0.02$. The isomer shift and the quadrupole splitting should yield information about the charge density and electric field gradient of the conduction electrons at the iron nucleus. The first effect arises from the electrostatic interaction between the nuclear charge and the electrostatic charge within the nuclear volume. The second effect results from the noncubic symmetry surrounding the iron nucleus. Possible systematics of these effects will be discussed.

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] O. C. Kistner and Bernard Mozer, Bull. Am. Phys. Soc., **7**, 505 (1962).

Superconducting Magnets for Mössbauer Experiments*

Juergen Heberle, *Argonne National Laboratory*

Three different magnets (designated as *M6*, *M7*, and *M8*) have been constructed, tested, and used. M6 has been described previously.[1] All three solenoids were wound on

aluminum formers with copper-plated Nb-25% Zr wire (0.010-in. diam.) with 0.001-in. Mylar between layers. Absorbers with an open diameter of 0.48 in. can be clamped in the central plane.

The winding of *M7* has an i.d. of 0.60 in., an o.d. of 2.45 in., and a length of 1.70 in. It has been reliably operated at 21.5 A, at which the field was over 50 kG. When the current is being increased, induced voltages cause current flow through the copper plating from turn to turn. Consequently, the current can be increased only rather slowly. Thus a whole hour is required to take *M7* from zero to 21.5 A.

In order to reduce this undesired effect, the resistance of the insulation between adjacent turns in *M8* was increased by covering the wire with an additional thin plating of zinc. *M8* has been operated at 21.0 A for a total of over 30 h and produces 48 kG. That these magnets are especially compact is indicated by the length of wire: The 10 385 turns on *M8* have a length of only 4000 ft.

In use the magnet is immersed in liquid helium with the axis of the solenoid horizontal. The gamma rays enter and leave the liquid-helium chamber through beryllium windows.[2]

* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] J. Heberle and R. W. Reno, Bull. Am. Phys. Soc. **7**, 431 (1962).
[2] J. Heberle, Rev. Sci. Instr. **33**, 1476 (1962).

Spin Density Wave Theory of Magnetic Shielding Phenomena*

R. M. Housley† and J. G. Dash, *University of Washington, Seattle*

It is shown how magnetic shielding effects observed for Fe^{57} nuclei in Cr, Cu, and other metals can be explained in a simple, consistent manner by spin-density wave theory. Results reported at this meeting in Cr and in Cu are interpreted in some detail. Areas in which further experiments should prove most interesting are indicated.

* This work was supported by the U. S. Air Force Office of Scientific Research, Contract No. AF 49(638)—1004.

† National Science Foundation Predoctoral Fellow.