Title

THE INFLUENCE OF A RANDOMLY ORIENTED MAGNETIC FIELD ON ANGULAR CORRELATIONS

Authors

Matthias, E.
Rosenblum, S.S.
Shirley, D.A.

Publication Date

1964-12-01

UCRL-11820

University of California
Ernest O. Lawrence Radiation Laboratory

THE INFLUENCE OF A RANDOMLY ORIENTED MAGNETIC FIELD ON ANGULAR CORRELATIONS

Berkeley, California

DISCLAIMER

Submitted to Physical Review Letters

UCRL-11820

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

THE INFLUENCE OF A RANDOMLY ORIENTED MAGNETIC FIELD ON ANGULAR CORRELATIONS

E. Matthias, S. S. Rosenblum and D. A. Shirley

December 1964

THE INFLUENCE OF A RANDOMLY ORIENTED MAGNETIC FIELD ON ANGULAR CORRELATIONS

E. Matthias, S. S. Rosenblum, and D. A. Shirley

Department of Chemistry and
Lawrence Radiation Laboratory
University of California
Berkeley, California

December 1964

The study of hyperfine magnetic fields at nuclei of atoms in magnetic lattices has received much attention recently. In some cases the origins of these fields are not even qualitatively understood, and only for certain favorable examples do quantitative calculations exist. On the other hand a systematic knowledge of such fields is of considerable practical importance, particularly in connection with a "universal" method, discovered by Samoilov and co-workers,[1] for orienting atomic nuclei at low temperatures. This method involves inducing large hyperfine fields at the nuclei of various atoms by dissolving them in magnetic lattices such as iron. Unfortunately these induced fields are among those most difficult to predict theoretically, and for some time studies have been underway in this Laboratory to obtain enough information about induced fields that systematic correlations could be made and used to predict the field that could be expected in a given case.

Several techniques, including nuclear polarization, specific heats, NMR, and Mössbauer spectroscopy, have been employed to measure hyperfine fields. None of these is very generally applicable at present, and new methods are badly needed. In this letter we discuss the measurement of hyperfine fields by observation of the time dependence of perturbed angular correlations. Although it has long been known that internal fields influence these correlations, the power of the method has not previously been fully appreciated. Two features of special interest are: (1) No external polarizing field is necessary, i.e.,

the sample can be completely demagnetized, and (2) For a demagnetized sample the angular correlation will be modulated by at least two frequencies, the Larmor frequency and its first harmonic.

For a demagnetized source the magnetic domains may be taken to be randomly oriented, and the theory is similar to that of a polycrystalline source.[2] The angular correlation function for each domain is given by[3]

$$W(\vec{k}_1,\vec{k}_2,t) = \sum_{\substack{k_1k_2\\N_1N_2}} A_{k_1}(1)\, A_{k_2}(2)\, G^{N_1N_2}_{k_1k_2}(t)\, [(2k_1+1)(2k_2+1)]^{-\frac{1}{2}}$$

$$\times Y^{N_1}_{k_1}{}^{*}(\theta_1,\phi_1)\, Y^{N_2}_{k_2}(\theta_2,\phi_2) \qquad (1)$$

The perturbation factor has the general form

$$G^{N_1N_2}_{k_1k_2}(t) = \sum_{m_am_b} (-1)^{2I+m_a+m_b}\, [(2k_1+1)(2k_2+1)]^{\frac{1}{2}} \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix}$$

$$\begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2 \end{pmatrix} \langle m_b|\Lambda(t)|m_a\rangle \langle m_b'|\Lambda(t)|m_a'\rangle^{*} \qquad (2)$$

where $\Lambda(t)$ is the time-evolution operator describing the change in population of the substates $|\, m_a\rangle$ and $|\, m_b\rangle$ with time. This operator is given by the interaction Hamiltonian $\mathcal{H}$ according to

$$\Lambda(t) = e^{-(i/\hbar)\mathcal{H}t}. \qquad (3)$$

The effect of a random magnetic interaction can be calculated by choosing a reference frame in one domain and averaging over all directions (θ,ϕ) of the detectors with respect to this frame. The relative angle between the two

detectors is kept fixed under the averaging process. Using the orthogonality properties of the rotation group and the addition theorem of spherical harmonics, Eq. (1) reduces to

$$W(\theta,t) = \sum_k A_k(1)\, A_k(2)\, G_k(t)\, P_k(\cos\theta) \quad , \tag{4}$$

with $G_k(t) = \frac{1}{2k+1} \sum_N G_{kk}^{NN}$. The reference system was chosen in such a way that its z-axis was the quantization axis for the interaction Hamiltonian:

$$\langle m_b | \Lambda(t) | m_a \rangle = e^{-(i/\hbar)E_m t}\, \delta_{m\, m_a}\, \delta_{m\, m_b} \tag{5}$$

This simplifies the perturbation factor in Eq. (2) to the expression

$$G_{kk}^{NN}(t) = (2k+1) \sum_m \begin{pmatrix} I & I & k \\ m' & -m & N \end{pmatrix}^2 e^{-(i/\hbar)(E_m - E_{m'})t} = e^{-iN\omega_L t} \quad , \tag{6}$$

where we have used properties of the 3 j symbols and the definition of the Larmor frequency, $\omega_L = -g H \frac{\mu_N}{\hbar}$. We therefore obtain for the real part of the perturbation factor $G_k(t)$ in Eq. (4)

$$G_k(t) = \frac{1}{2k+1} \sum_N \cos N\omega_L t \quad . \tag{7}$$

Thus, in a time-differential measurement one observes a superposition of k frequencies with equal amplitudes. In Fig. 1 a the perturbation factors G_2 and G_4 are shown as functions of time. Note that because of the equidistant magnetic splitting the perturbation factor in Eq. (7) is <u>independent</u> of the nuclear spin I, in contrast to the situation for random electric quadrupole perturbations.

The integral attenuation factor $\overline{G}_k$ can be obtained from Eq. (6) and is given by

$$\overline{G}_k = \frac{1}{2k+1} \sum_N \frac{1}{1 + (N\omega_L \tau)^2} \qquad (8)$$

The theoretical behavior of $\overline{G}_k$ as a function of the interaction strength $(\omega_L \tau)$ is demonstrated in Fig. 1 b. In the limit of a very strong interaction the attenuation factor approaches a hard core value of $\frac{1}{2k+1}$ which is independent of the nuclear spin, in contrast to the hard-core values for polycrystalline sources.

To demonstrate experimentally the effect of a random magnetic interaction on the angular correlation we performed a time-differential measurement with a sample of In^{111} dissolved in Ni (less than 1 part in 10^{10}). The result is shown in Fig. 2. The large anisotropy of the 172-247 keV cascade in Cd^{111} confirms clearly the prediction of two frequencies for k = 2 (Eq. (7)). A least square fit of the data yields a Larmor frequency $\omega_L = (0.995 \pm 0.010) \times 10^8$ sec^{-1} which gives with a g-factor of $g = -0.318 \pm 0.007$ [4] a magnetic field for Cd dissolved in Ni of

$$|H| = 65.3 \pm 1.6 \text{ kgauss}$$

The accuracy of this value is limited by the uncertainty of the time calibration (1%) and by the fact that the g-factor is only known within 2%.

Several features of this method are worth pointing out: (1) The presence of a low-frequency (ω_L) component allows the measurement of fields twice as large as would otherwise be possible, with a given instrumental time resolution. (2) Very small fields are also accessible. In the present experiment any field between 5 and 500 kgauss could have been observed. (3) Fields

are measured throughout the sample, not just in domain walls. (4) Measurements may be made at any temperature and pressure, provided that the spin-correlation time is long compared with $1/\omega_L$. (5) Induced fields in antiferromagnets may also be measured. (6) Polarization in an external field may be followed independently of frequency shifts by observing the disappearance of the low-frequency component in the correlation function.

Aside from the famous case of Cd^{111} it appears that there are quite a few isotopes available which would allow the investigation of internal magnetic fields with the aid of this method.

ACKNOWLEDGMENT

One of us (E.M.) gratefully acknowledges a fellowship from the Miller Institute for Basic Research in Science, University of California.

This work was performed under the auspices of the U. S. Atomic Energy Commission.

REFERENCES

1. B. N. Samoilov, V. V. Sklyarevskii, and E. P. Stepanov, JETP 36, 448 (1959).
2. A. Abragam and R. V. Pound, Phys. Rev. 92, 943 (1953).
3. R. M. Steffen and H. Frauenfelder, Chap. I in "Perturbed Angular Correlations" ed. by E. Karlsson, E. Matthias, and K. Siegbahn, North-Holland Publishing Company, Amsterdam, 1964.
4. E. Matthias, L. Boström, A. Maciel, M. Salomon, and T. Lindquist, Nucl. Phys. 40, 656 (1963).

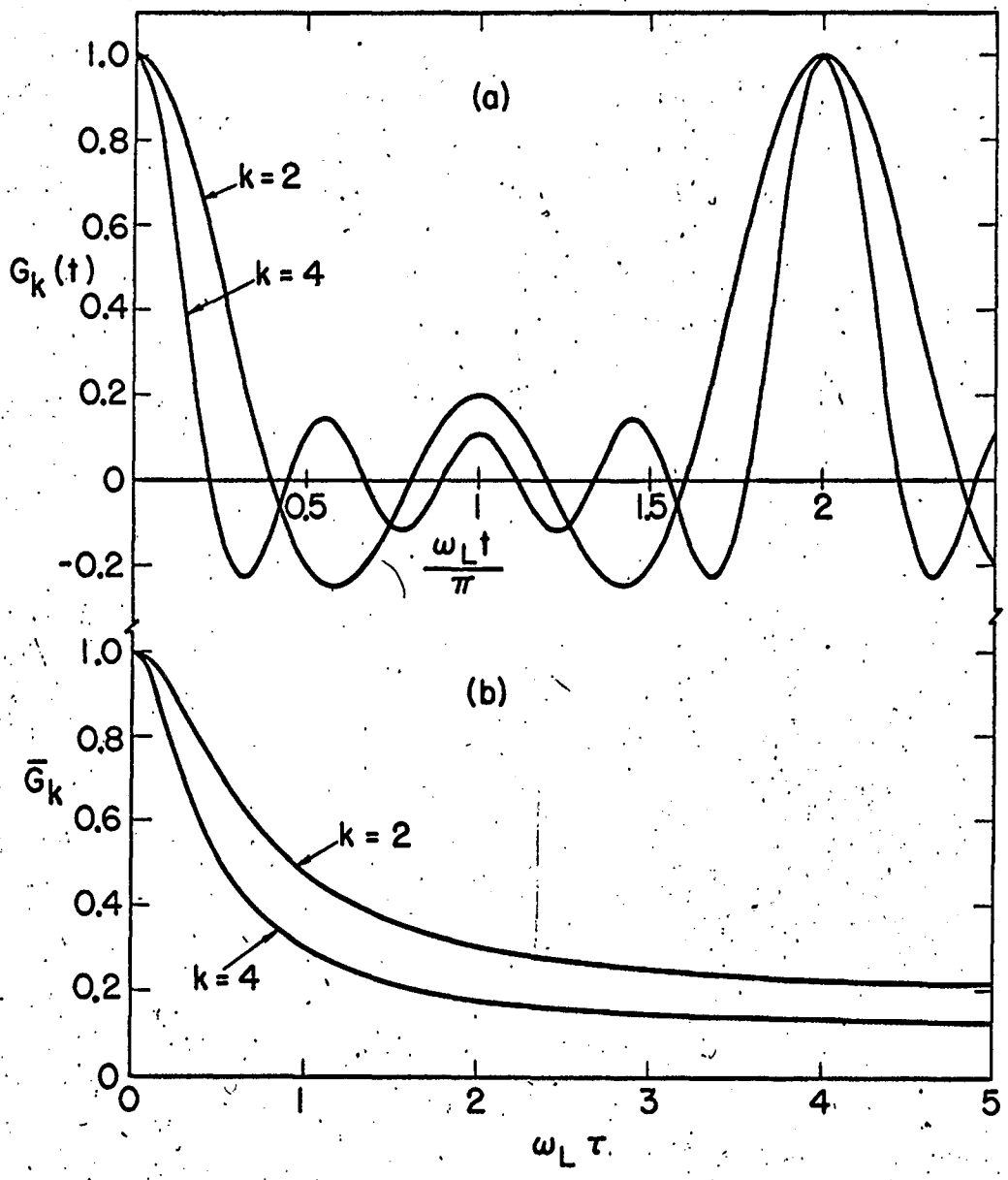
FIGURE CAPTIONS

Fig. 1a. Theoretical behavior of the time-differential perturbation factors G_2 and G_4 as a function of time.

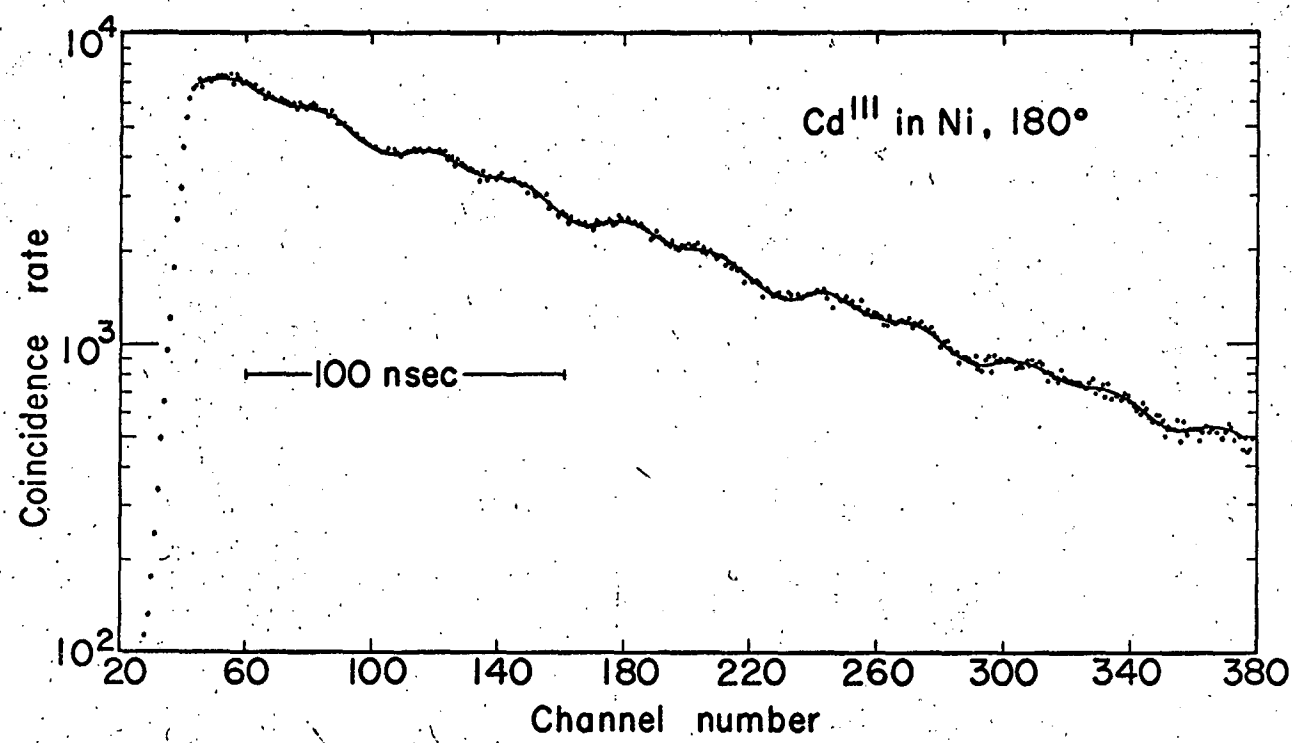
b. Integral attenuation factors $\overline{G}_2$ and $\overline{G}_4$ plotted as a function of the interaction strength.

Fig. 2. Time-differential measurement of a random magnetic interaction with a source of In^{111} dissolved in Ni. The solid curve represents the best fit of the points to the function

$$F(t) = Ne^{-\lambda t}\left(1 + a\left(1 + 2\cos\left(\omega_L t + \phi\right) + 2\cos 2\left(\omega_L t + \phi\right)\right)\right) + C.$$


(a)
1.0
0.8
0.6
0.4
0.2
0
-0.2
$G_k(t)$
k = 2
k = 4
0.5
1
1.5
2
$\frac{\omega_L t}{\pi}$
(b)
$\bar{G}_k$
k = 2
k = 4
0
1
2
3
4
5
$\omega_L \tau$

MUB-4657


10^4
10^3
10^2
Coincidence rate
Cd^{111} in Ni, 180°
100 nsec
20
60
100
140
180
220
260
300
340
380
Channel number

NUB-4658