

## Hyperfine Magnetic Fields Acting on $^{221}\text{Bi}$ in Ni, Co and Fe Lattices

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The gamma-gamma perturbed angular correlation technique has been used to determine the hyperfine magnetic fields acting on  $^{221}\text{Bi}$  nuclei in Ni, Co and Fe lattices.  $^{221}\text{Pb}$ , the immediate precursor of  $^{221}\text{Bi}$ , was implanted in the ferromagnets by recoil due to the preceding alpha emission from  $^{215}\text{Po}$ . The measurements have been performed at room temperature through the 704-405 keV gamma-gamma cascade in  $^{221}\text{Bi}$ . The results for the hyperfine fields are  $H(\text{BiNi}) = +160 \pm 30\text{kOe}$ ,  $|H(\text{BiCo})| = 440 \pm 80\text{kOe}$  and  $|H(\text{BiFe})| = 400 \pm 70\text{kOe}$ .

A técnica de correlação angular perturbada gama-gama foi usada para determinar os campos magnéticos hiperfinos atuando sobre núcleos de  $^{221}\text{Bi}$  em latitudes de Ni, Co e Fe.  $^{221}\text{Pb}$ , o precursor imediato de  $^{221}\text{Bi}$ , foi implantado nos ferromagnetos por recuo devido a precedente emissão alfa do  $^{215}\text{Po}$ . As medidas foram feitas à temperatura ambiente através da cascata gama-gama de 704-405 keV no  $^{221}\text{Bi}$ . Os resultados para os campos hiperfinos são  $H(\text{BiNi}) = +160 \pm 30\text{kOe}$ ,  $|H(\text{BiCo})| = 440 \pm 80\text{kOe}$  e  $|H(\text{BiFe})| = 400 \pm 70\text{kOe}$ .

### 1. Introduction

The determination of magnetic hyperfine fields acting on impurities embedded in ferromagnetic hosts has proved to be fruitful in the study of magnetic properties of metals and alloys. Recently, several papers have treated theoretically the existent experimental data for dilute alloys of diamagnetic impurities in ferromagnets. The fields on the elements of period V impurities in Fe, Co and Ni matrices are experimentally well known. A new version<sup>1</sup> of the local potential model of Daniel and Friedel<sup>2</sup>

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fits very well the hyperfine fields of the  $s-p$  impurities **Ag** to **Xe** in **Fe**. This model also predicts the reversal in sign of the hyperfine field between **Sn** and **Sb**.

Presently, it is still very difficult to obtain general conclusions concerning the origin of these hyperfine fields from first principles. Additional measurements are of interest in order to establish firm experimental systematics. Detailed experimental information on the fields acting at  $s-p$  elements belonging to period VI seems to be of considerable interest to test the model predictions. Here we present measurements of the average magnetic field acting on  $^{211}\text{Bi}$  nuclei as impurities in **Fe**, **Co** and **Ni** lattices.

## 2. Experimental Procedure

The work reported in this paper was performed through the perturbed angular correlation technique. The apparatus was a conventional fast-slow coincidence system with a TPHC identical to that described in Ref. 3, except that the gamma rays were detected by two  $7.6 \times 7.6$  cm **NaI(Tl)** scintillation spectrometers. The measurements were made on the 704-405 keV gamma-gamma sequence in  $^{211}\text{Bi}$ . The levels of  $^{211}\text{Bi}$  are populated by beta decay of the 36m  $^{211}\text{Pb}$  which in turn is fed by a sequence of beta and alpha decays starting from the 22y  $^{227}\text{Ac}$ . In all measurements the short-lived  $^{211}\text{Pb}$  activity was in equilibrium with  $^{227}\text{Ac}$ . Previous measurements<sup>4</sup> have shown that the angular correlation coefficients for this cascade in  $^{211}\text{Bi}$  are not affected by the complex low energy gamma spectrum of other daughters of  $^{227}\text{Ac}$ . Because the lifetime of the 405 keV level in  $^{211}\text{Bi}$  is much smaller than the 3ns resolving time of the coincidence system, the integral angular correlation method was used in all experiments.

The alloys of **Ac** were prepared by depositing carrier free  $^{227}\text{Ac}$  activity in dilute  $\text{HNO}_3$  on powders of pure host metals (**Cu**, **Fe**, **Co** and **Ni**). The mixtures were washed with distilled water. After drying, the powders were reduced in an **H<sub>2</sub>** atmosphere at about 600°C. The reduced powders were allowed to cool and immediately formed into circular right cylinders under a pressure of  $\approx 10^3$  atm. The sources were 5mm in diameter and 5mm high, all containing less than 0.01 atomic per cent of the impurity. Finally, the cylinders were annealed for one day at 800°C in **H<sub>2</sub>** atmosphere. The loss of activity during annealing was always less than 5%. The measurements described in next section were all made at room temperature.

### 3. Experimental Results

Initially, the directional angular correlation coefficients of the 704-405 keV cascade in  $^{211}\text{Bi}$  have been measured for a liquid, carrier free  $^{227}\text{Ac}$  activity in dilute solution of  $\text{HNO}_3$ . The distance between source and detectors was 10 cm and our result is  $A_2 = -0.098 \pm 0.005$  and  $A_4 = -0.001 \pm 0.006$ . These coefficients after correction for the geometry are in excellent agreement with recent measurements of the same correlation in liquid sources<sup>5</sup>.

Using the same geometry as above, the angular correlation function of the same cascade was measured for sources embedded in Cu, Ni, Co and Fe metals. The coefficients  $A_i$  obtained from these measurements are presented in Table I, without any correction for the geometry. A simple comparison of the  $A_i$  coefficients ( $A_i$  are very small) for the five different sources in Table I shows strong attenuation in the case of ferromagnetic hosts.

|         | Liquid             | BiCu               | BiNi               | BiCo               | BiFe               |
|---------|--------------------|--------------------|--------------------|--------------------|--------------------|
| $A_2^*$ | $-0.098 \pm 0.005$ | $-0.097 \pm 0.003$ | $-0.074 \pm 0.007$ | $-0.041 \pm 0.003$ | $-0.045 \pm 0.002$ |
| $A_4^*$ | $-0.001 \pm 0.006$ | $+0.002 \pm 0.004$ | $+0.002 \pm 0.005$ | $+0.002 \pm 0.004$ | $+0.001 \pm 0.003$ |

\*Uncorrected for geometry.

Table I - Angular correlation coefficients  $A_i$  for sequence 704-405 keV of  $^{211}\text{Bi}$  in five different environments.

When the resolving time of the coincidence system is larger than the lifetime of the nuclear level, the angular correlation function can be expressed by:

$$W(\theta, t = \infty) = 1 + A_2 G_2(\infty) P_2(\cos \theta),$$

where  $G_2(\infty)$  is the attenuation factor of the angular correlation function and  $A_2$  is assumed to be zero. For an unoriented ferromagnet, assuming that only magnetic interactions are present in the source,  $G_2(\infty)$  is expressed by

$$G_2(\infty) = \frac{1}{5} \left[ 1 + \frac{2}{1 + (\omega\tau)^2} + \frac{2}{1 + (2\omega\tau)^2} \right], \quad (1)$$

where  $\omega = g\mu_N H/\hbar$  is the Larmor frequency, and  $\tau$  = lifetime of the nuclear level.

This expression for  $G_2$  applies if the magnetic field acting on the nuclei is static and has a well defined value which is randomly oriented with respect to the counter directions when averaged over all nuclei. A typical example is an unoriented ferromagnet with an ensemble of static magnetic fields, pointing in all directions. This type of interaction does not produce any resultant rotation of the integral angular correlation function, only an attenuation.

The  $A_2$  coefficient obtained for the liquid source is assumed to be the unperturbed value for this angular correlation. This assumption is supported by the measurement for the source in a Cu host, which does not show any attenuation compared with the liquid source. As Cu is nonmagnetic and has a crystalline structure similar to that of the other three ferromagnetic metals, it is assumed that within the experimental errors a static quadrupole interaction is not present in our solid sources.

The experimental attenuation coefficients  $G_2(\infty) = A_2(\text{metal})/A_2(\text{liquid})$  for the metallic sources are presented in Table II. Expression (1) is used to calculate  $|\omega\tau|$  for the four solid sources and the result is also shown in table II. Using these values of  $\omega\tau$ , with the known magnetic moment  $\mu = +4.45 \pm 0.70$  n.m. (Ref. 4) and lifetime  $\tau = 460 \pm 30$  ps (Ref. 5) of the 405 keV level, the absolute values of the average magnetic fields acting on  $^{211}\text{Bi}$  in the three ferromagnets are obtained and are given in Table II.

|                             | <i>BiCu</i>     | <i>BiNi</i>        | <i>BiCo</i>        | <i>BiFe</i>        |
|-----------------------------|-----------------|--------------------|--------------------|--------------------|
| $G_2(\infty)$               | $+1.0 \pm 0.36$ | $+0.750 \pm 0.050$ | $+0.420 \pm 0.033$ | $+0.440 \pm 0.031$ |
| $ \omega\tau $<br>(radians) | 0               | $0.45 \pm 0.06$    | $1.21 \pm 0.13$    | $1.13 \pm 0.12$    |
| $ H $<br>(kOe)              | 0               | $160 \pm 40$       | $440 \pm 80$       | $400 \pm 70$       |

**Table II** - Experimental attenuation factor  $G_2(\infty) = A_2(\text{metal})/A_2(\text{liquid})$  for sequence 704-405 keV of  $^{211}\text{Bi}$  embedded in metallic hosts and extracted average precession angles  $|\omega\tau|$  and average hyperfine fields  $|H|$  (see text).

The sign of the hyperfine magnetic field on  $^{211}\text{Bi}$  in a Ni host was determined by the integral reversed field method of the perturbed angular

correlation technique. For the large mean precession angle observed in this case, the ratio defined as

$$\varepsilon(\theta, H) = \frac{W(\theta, +H) - W(\theta, -H)}{W(\theta, +H) + W(\theta, -H)},$$

with

$$W(\theta, \pm H) = 1 + \frac{b_2}{1 + (2\omega\tau)^2} (\cos 2\theta \pm 2\omega\tau \sin 2\theta),$$

is expressed, at  $\theta = \pm \frac{3}{4}\pi$  by

$$\varepsilon\left(\theta = \pm \frac{3}{4}\pi\right) = \mp \frac{2\omega\tau b_2}{1 + (2\omega\tau)^2},$$

where  $b_2$  is the coefficient of  $\cos 2\theta$  for the unperturbed angular correlation and is related to the measured  $b_2^m$  (without an aligning magnetic field) by  $b_2^m = b_2 G_2(\infty)$ .

The alloy with Nickel was placed between the pole-tips of a small, fully enclosed toroidal electromagnet described elsewhere<sup>6</sup>. In order to increase the coincidence rate both detectors were moved closer to the source. In this new geometry, angular correlation coefficients  $b_2 = -0.0365 \pm 0.0030$  and  $b_4 = +0.0010 \pm 0.0040$  are obtained, uncorrected for the geometry. In a second step the magnet was turned on applying an external aligning field of the order of  $3 \times 10^3$  Oe on the source and the ratio  $\varepsilon(\frac{3}{4}\pi)$  was measured to be

$$\varepsilon(\theta = \frac{3}{4}\pi) = -(2.40 \pm 0.11) \times 10^{-2},$$

yielding  $\omega\tau = +0.44 \pm 0.05$  rad, in excellent agreement with  $\omega\tau$  obtained from the attenuation measurement in the same source. This value of  $\omega\tau$  gives<sup>7</sup> for the hyperfine field on  $^{211}\text{Bi}$  in Ni,

$$H(\text{BiNi}) = (+160 \pm 30) \text{ kOe},$$

where the error comes mostly from the error in the magnetic moment measurement.

#### 4. Discussion

In accordance with this: systematics of hyperfine magnetic fields on “4d” elements in ferromagnetic metals, where the field changes sign between **Sb** and **Sn**, being positive after **Sb**, the elements belonging to the “5d” series also show such a change of sign between **Tl** and **Pb** (Ref. 8). The next element after **Pb** is **Bi**, and our result of a positive sign of the field on **Bi** in **Ni** and larger than the field on **Pb** in **Ni** agrees with what was expected.

With very few exceptions, the fields on a diamagnetic impurity in Fe, **Co** and **Ni** hosts have the same sign and are proportional to the magnetic moments of the host metals. Here again, systematics would indicate that the fields in **Co** and **Fe**, for which we measured the absolute values only, should be positive. Concerning the proportionality with the atomic magnetic moments of the matrices ( $\mu = 2.2, 1.7$  and  $0.6$  for Fe, **Co** and **Ni** respectively) the value of  $\omega\tau = 1.13 \pm 0.12$  radians for **Fe** is about 30% lower than the expected value in terms of this proportionality when comparing with **Co** and **Ni**. In fact, our result (Table II) shows that  $\omega\tau$  has the same values, within the experimental errors, in Fe and **Co** hosts, consequently giving hyperfine fields of the same order on  $^{211}\text{Bi}$  in both ferromagnets. It is interesting to observe that in a recent measurement of the fields on  $^{208}\text{Pb}$  in Fe, **Co** and **Ni** (Ref. 9),  $\omega\tau$  was also measured to be the same in value and sign for Fe and **Co**. The fields on  $^{208}\text{Pb}$  are of the order of 300 kOe and that on  $^{211}\text{Bi}$  are of the order of 400 kOe in both ferromagnets. The technique of source preparation was the same in both cases,  $^{211}\text{Bi}$  was studied by alloying  $^{227}\text{Ac}$  with the ferromagnets and  $^{208}\text{Pb}$  was studied in an alloy of  $^{228}\text{Th}$  with the ferromagnets.

Recent lattice location studies of **Pb** and **Bi** implanted in Fe (Ref. 10) using the channeling technique show that at room temperature a large fraction of the impurity atoms are located substitutionally in regular Fe sites. In our sources the levels of  $^{211}\text{Bi}$  are populated by beta decay from  $^{211}\text{Pb}$ . The nuclei of  $^{211}\text{Pb}$  are reached by an alpha decay of 7.3 MeV from  $^{215}\text{Po}$  and consequently suffer a recoil of approximately 100 keV through the metallic lattice before beta decaying to  $^{211}\text{Bi}$ . This means that independently of the location of the parent  $^{227}\text{Ac}$  in the metal,  $^{211}\text{Pb}$  is implanted by  $\alpha$ -recoil with an energy of 100 keV. It is shown in Ref. 10 that the two previously measured hyperfine magnetic fields on **Pb** in Fe of +650 kOe and +300 kOe (Ref. 9) correspond respectively to **Pb** atoms located off and on regular lattice sites. As systematics would indicate a larger field for **Bi** than for **Pb** in Fe, our result of  $\sim 400$  kOe (absolute

value) agrees with this trend if one assumes that Bi nuclei are located substitutionally in Fe metal.

|                     | $\omega\tau$<br>in radians      | $H$<br>in kOe  | Reference |
|---------------------|---------------------------------|----------------|-----------|
| $^{208}\text{PbFe}$ | $-1.59 \pm 0.09 \times 10^{-2}$ | $+ 280 \pm 70$ | 9         |
| $^{208}\text{PbCo}$ | $-1.57 \pm 0.14 \times 10^{-2}$ | $+ 280 \pm 70$ |           |
| $^{208}\text{PbNi}$ | $-0.71 \pm 0.15 \times 10^{-2}$ | $+ 125 \pm 35$ |           |
| $^{204}\text{PbFe}$ |                                 | $+ 262 \pm 6$  | 11        |
| $^{204}\text{PbCo}$ |                                 | $+ 262 \pm 8$  |           |
| $^{204}\text{PbNi}$ |                                 | $+ 135 \pm 3$  |           |
| $^{211}\text{BiFe}$ | $1.13 \pm 0.12$                 | $400 \pm 70$   | present   |
| $^{211}\text{BiCo}$ | $1.21 \pm 0.13$                 | $440 \pm 70$   |           |
| $^{211}\text{BiNi}$ | $+ 0.44 \pm 0.05$               | $+ 160 \pm 30$ |           |

**Table III** - Resumé of PAC data for  $^{208}\text{Pb}$ ,  $^{204}\text{Pb}$  and  $^{211}\text{Bi}$  impurities in Fe, Co and Ni hosts. The large errors of the fields on  $^{208}\text{Pb}$  and  $^{211}\text{Bi}$  come mostly from the errors in the g-factors.

In Table III, the data of PAC measurements on  $^{208}\text{Pb}$ ,  $^{204}\text{Pb}$  and  $^{211}\text{Bi}$  impurities in the three ferromagnets are presented. For  $^{208}\text{Pb}$  and  $^{211}\text{Bi}$  the errors in  $\omega\tau$  are smaller than in the obtained hyperfine fields. The anomaly of the fields in Fe and Co hosts is evident for the three impurities. The hyperfine fields on  $^{208}\text{Pb}$  and  $^{211}\text{Bi}$  are determined after implantation by a-recoil; we do not know the technique of source preparation'' in the case of  $^{204}\text{Pb}$ . It seems that channeling lattice location studies of Pb and Bi impurities in Co and Ni hosts can provide additional important information for the interpretation of these results.

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