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PHYSICAL REVIEW B

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Hyperfine Magnetic Field of Argon Implanted in Nickel

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The hyperfine field of argon in nickel is measured to be 280 ± 23 kG using the technique of time-differential perturbed angular correlations. The argon nuclei were recoil implanted in nickel by the ${}^{39}\text{K}(d, \alpha)$ ${}^{37}\text{Ar}$ reaction. The second excited state of ${}^{37}\text{Ar}$ at 1610 keV was used in this measurement. The unperturbed α_2 -1610-keV γ -ray angular correlation was studied with a copper backing for the target, and values $A_2 = 0.265 \pm 0.057$ and $A_4 = -0.222 \pm 0.098$ were obtained for the correlation coefficients. The amplitude of modulation of the time spectrum is found to be very small. This may be interpreted by assuming that the hyperfine field acts only on a small fraction of the implanted nuclei.

I. INTRODUCTION

At present, hardly any data exist about the hyperfine magnetic fields acting on nuclei with Z < 20 imbedded in ferromagnetic lattices. A knowledge of these fields is of interest not only because of their utility in measuring magnetic moments of shortlived nuclear excited states, but also for understanding the mechanism which produces these fields. We report here on a measurement of the magnetic field on Ar nuclei in nickel. This measurement is of particular significance because Ar is an inert gas with a closed electron shell. This simple electron configuration should facilitate the calculation of the magnetic hyperfine field, which is believed to be largely due to overlap polarization.

II. EXPERIMENTAL PROCEDURE AND RESULTS

We used the technique of time-differential perturbed angular correlation following recoil implantation in a nuclear reaction. The second excited state of ³⁷Ar at 1610 keV is suitable for such a measurement as it has a fairly long half-life of 4.5 nsec and its magnetic moment has been recently measured with quite good accuracy.¹ This state was excited in the ³⁹K(d, α) ³⁷Ar reaction. In the reaction process, the excited ³⁷Ar nuclei got implanted in the target backing material.

The deuteron beam from the Groningen Van de Graaff accelerator at a deuteron energy of 4.925 MeV was used in this experiment since a rather sharp resonance was observed at this energy for the yield of the α_2 group. The target consisted of $300-\mu g/cm^2$ KBr vacuum evaporated on a $1-mg/cm^2$ nickel backing. With this target thickness all ³⁷Ar nuclei recoiling in the forward direction with 2.25-MeV energy leave the target material and penetrate the backing which is just thick enough to stop them. Thus, the number of elastically scattered deuterons is kept as low as possible. The nickel foil used as a backing is obtained commercially and is stated to have a purity of better than 99.4%. The only major impurity is Co which is 0.5%. The reason for selecting nickel rather than iron was that in analogy to $xenon^2$ we expected the field on Ar in Fe to be of the order of 1 MG. For such a large field the Larmor frequency would have been too high to be measured by this technique. The target was mounted between the pole pieces of a permanent magnet which provides a field of 1.2 kG for magnetizing the nickel backing to saturation. It was observed that the target had a tendency to evaporate gradually during the bombardment and it was therefore necessary to change the target position slightly from time to time to get a new beam spot.

The α particles emitted in the backward direction were detected by an $80-\mu$ -thick annular silicon detector. The over-all energy resolution under experimental conditions was 80 keV which includes the effects of target thickness and kinematic broadening. Fast-timing pulses were obtained with a fast trigger circuit after the preamplifier. For the γ rays, a 4×4-in. NaI(Tl) crystal mounted on a XP 1041 photomultiplier was used. As the total γ counting rate was $\geq 10^5$ /sec, it was necessary to operate the photomultipier with the dynode from which the linear signal was derived at earth potential, and to stabilize the voltages of all higher dynodes by voltage-regulating gas discharge tubes. The fast pulses in this case were obtained with an avalanche transistor limiter circuit. A beam current of 100 nA could be used without pile-up problems in the electronics, or any serious deterioration of the target.

The delayed-time spectrum of the α_2 group gated by the 1610-keV region of the γ spectrum was recorded on one memory half of a 400-channel analyser. The prompt spectrum of the α_5 group gated by the same γ 's was recorded simultaneously on the second memory subgroup. This makes it possible to correct for a possible drift in the time spectrum. Such drifts were seen to be much less than 1 nsec over a period of several days. The time resolution of the prompt curve recorded under actual experimental conditions was $2\tau_0 = 1.8$ nsec. The time calibration of the time-to-amplitude converter was done in the usual way by length. The calibration was also checked with a crystal-controlled 100-MHz pulse train generator. The data were recorded with the γ detector at 135° with respect to the beam and the measurement was then repeated at 45° . The individual time spectra at the two angles are shown in Fig. 1. Counts were summed in groups of four channels, each group corresponding to a 0.8-nsec time interval. The curves correspond to a least-squares fit to the expression

$$\begin{split} N(t) &= e^{-t/\tau} \left[B_0 + B_2 \cos 2(\theta + \omega t + \alpha) \right. \\ &+ B_4 \cos 4(\theta + \omega t + \alpha) \right] + \text{const} , \end{split}$$

where B_0 , B_2 , B_4 , ω , α , and const are taken as variable parameters. The effect of the finite time resolution has been taken into account by representing the prompt curve by $N(t) = (1/2\tau_0) \cos^2(\pi t/4\tau_0)$ and integrating over it. The lower part of the figure shows the ratio

$$R(t) = \frac{N(t, 135^{\circ}) - N(t, 45^{\circ})}{\frac{1}{2}[N(t, 135^{\circ}) + N(t, 45^{\circ})]}$$

with the counts at the two angles suitably normalized and corrected for chance coincidences. This ratio has been fitted to

$$R(t) = 2B_2 \sin(2\omega t + \alpha) ,$$

where B_2 , ω , and α are the variable parameters. In this fit we have neglected the dependence on B_4 , since $B_4 <<1$.

The value of the Larmor frequency obtained from this measurement is $\omega_L = (0.512 \pm 0.031) \times 10^9$ rad/ sec. This gives H_{int} (ArNi) = (280 ± 23) kG using the value of $\mu = -(1.33 \pm 0.05)$ nm for the $\frac{7}{2}$ 1610keV level of ³⁷Ar as measured by Randolph *et al.*¹ The sign of this internal field can be determined from a knowledge of the sign of the *g* factor, the sign of R(t) near t = 0, and the sign of the B_2 and B_4 coefficients. These coeffecients must also be known to determine a possible attenuation of the angular correlation due to implantation of ³⁷Ar in nickel.

To find the unperturbed angular-correlation coefficients, the α_{2} - γ_{2} angular correlation was studied



FIG. 1. Time spectrum of the decay of 1610-keV level of 37 Ar recoil implanted in nickel in the 39 K(d, α) 37 Ar reaction. The prompt spectrum, recorded simultaneously, is shown by the dashed curve. The ratio

$$R = \frac{N(t, 135^{\circ}) - N(t, 45^{\circ})}{\frac{1}{2}[N(t, 135^{\circ}) + N(t, 45^{\circ})]}$$

is plotted in the lower part of the figure. The full curves correspond to least-squares computer fits.

with the target on a backing of 1 mg/cm^2 of copper. The γ spectra in coincidence with all the α groups were recorded at five angles with a PDP-9 on-line computer using a program for two-parameter analysis. The fast-slow coincidence set up was the same as in the previous measurement except that the gates on the α and γ sides were now made so wide as to include the entire α and γ spectra. Gates of 25-nsec width were also placed on the time spectrum for true and chance coincidence. From the α_{2} - γ coincidence spectra corrected for chance coincidences, the intensity of the 1610-keV photopeak at the five angles chosen was obtained. These intensities were then fitted to the angular-correlation function $W(\theta) = 1 + A_2 P_2 (\cos \theta) + A_4 P_4 (\cos \theta)$. The values found for the coefficients are $A_2 = 0.265$ ± 0.057 and $A_4 = -0.222 \pm 0.098$. No correction has been applied here for the finite solid angles of the detectors. The corresponding values of B_2 and B_4 are $B_2 = 0.126 \pm 0.051$ and $B_4 = -0.117 \pm 0.052$. We can now use this observed shape of the angular-correlation pattern to conclude that the sign of the hyperfine magnetic field of Ar in nickel is positive. Comparison of the unperturbed values with $B_2 = 0.031$ and $B_4 = -0.044$ obtained from the analysis of the time spectra shows that there is an attenuation of the angular correlation by almost a factor of 4.

III. DISCUSSION

Recently the hyperfine field on Ca in iron has been measured to be -92 kG using a similar technique.³ One thus sees that there is a very sharp fall of the hyperfine magnetic field as one goes from Z = 18 to Z = 20. A similar effect has been observed in the 5p-6s shell where the hyperfine field falls from the value of 1.4 MG for Xe in Fe^{2,4} to -85 kG for Ba in Fe.⁵ The value $H = (280 \pm 23)$ kG found for the magnetic hyperfine field of Ar in nickel confirms the trend of hyperfine fields observed in other groups of the Periodic Table. An empirical expression for these trends has been given by Balabanov and Delyagin⁶; this, however, is not applicable to impurities with Z < 18 in its present form.

If we assume that the hyperfine field is proportional to the magnetic moment of the host, as is frequently found for diamagnetic impurity atoms. we would expect the field of Ar in iron to be about 1020 kG. Large positive fields have also been observed for krypton⁷ and xenon implanted in iron. For the case of xenon, a reanalysis of the nuclear-orientation measurement of Niesen et al.² by Niesen,⁴ taking into account that only a part of the Xe nuclei experience a large hyperfine field, yields H(XeFe)= 1.4 \pm 0.2 MG. Following Shirley at al.⁸ we may write these hyperfine fields as a sum of contributions due to overlap polarization and conductionelectron polarization: $H_z = H_z^{OP} + H_z^{CEP}$. The last term may be estimated by using the empirical relation⁸ $H_Z^{CEP} = 0.054 H_{ns}^Z$ for solutes in iron, where H_{ns}^{Z} is the field due to one outer-shell s electron of a free atom with atomic number Z. Using the values for H_{ns}^{Z} given in or interpolated from Table II in the paper of Shirley and Westenbarger⁹ we have $H_{Ar}^{CEP} = -40$ kG and $H_{Xe}^{CEP} = -110$ kG and thus we estimate $H_{Ar}^{OP}/H_{Xe}^{OP} = 0.70 \pm 0.09$. This ratio is somewhat larger than that of the volume overlaps of argon and xenon atoms in iron, which, neglecting lattice relaxation, is 0.48 (using radii of 1.5 Å for iron, 1.91 Å for argon, and 2.2 Å for xenon¹⁰). An attempt at interpretation of the trends of hyperfine fields in terms of volume overlap has been presented by Stearns¹¹ but it appears from her data that the applicability of her model is questionable for the case of the noble gases. For a quantitative picture, a more complete calculation of the overlap integrals is required, taking into account the polarization of the different s shells of the noble gas and their contributions to the field at the nucleus.

Another interesting point is that the amplitude of modulation of the time spectrum is almost a factor of 4 smaller than one would expect from the unperturbed angular correlation. Such a large attenuation of the angular correlation has also been ob-

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served for fluorine¹² and calcium³ recoil implanted in ferromagnetic materials. In the latter case the atomic radius is too large to fit properly in the host lattice and one must expect that a large fraction of the implanted nuclei is in substitutional sites associated with one or more vacancies. This conclusion is based on the results of recent Mössbauer studies¹³ with Xe implanted in Fe which have shown a three-component spectrum indicating that the Xe atoms occupy three different types of sites with different hyperfine fields, isomer shifts, and recoilless fractions. These data all indicate that only about 30% of the implanted atoms appear to have no associated vacancies; the rest of the Xe atoms, associated with one or more vacancies, experience small hyperfine fields. Channelling experiments of Feldman and Murnick¹⁴ confirm that only about 35% of the xenon atoms implanted in iron are at substitutional sites. It therefore seems probable that in the case of argon in nickel also a considerable fraction of the implanted nuclei occupy sites where the field is much smaller than at regular substitutional sites. Such nuclei would precess with a much lower Larmor frequency which would result in the amplitude of modulation of the highest Larmor frequency being small.

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