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Hyperfine field of francium in iron

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The hyperfine field of francium in iron has been measured for different isotopes using the method of low-temperature nuclear orientation and samples prepared by ion implantation at $T \leqslant 1$ K. For ^{208}Fr a nuclear magnetic resonance was observed. From the center frequency $\nu_L(B_{\mathrm{ext}}=0\ \mathrm{T})=490.7(4)$ MHz, we derive the corresponding magnetic hyperfine field as $B_{\mathrm{hf}}(\mathrm{Fr}Fe)=94.9(4)$ T. [S0163-1829(98)05118-2]

I. INTRODUCTION

Although many hyperfine fields (B_{hf}) for impurities in iron have been determined over the years,[1,2] for some elements no experimental data are available as yet, while for a number of other elements existing data are in disagreement. The hyperfine fields of the alkaline elements are especially important to test different theoretical approaches since the simple electronic structure of alkaline impurities is supposed to facilitate the interpretation of experimental values. For francium two measurements of the hyperfine field in iron have been published up to now,[3,4] with conflicting results. In both cases the method of low-temperature nuclear orientation (NO) was used.

NO is particularly well suited to study hyperfine fields. First, very low doses ($\leqslant 10^{11}$ atoms/cm^2) can be used, thereby limiting the damage created in the host lattice. Furthermore, since the impurity atoms can be brought into the host using ion implantation at low temperature (i.e., $\leqslant 1$ K), the created defects cannot migrate anymore. As a consequence, large fractions at substitutional sites can be obtained even for atoms which are insoluble in the host lattice in thermal equilibrium. Indeed, although channeling experiments[5] suggested that large substitutional fractions can only be obtained for elements with an enthalpy of solution $\Delta H_{\mathrm{sol}} \leqslant 200$ kJ/mol, it was shown later that for francium—which is the largest atom in nature and has $\Delta H_{\mathrm{sol}}=525(70)$ kJ/mol (Ref. 3)—a large substitutional fraction can be obtained with ion implantation: channeling measurements[6] revealed a substitutional fraction of 67(13)% after implantation at 120 K with a dose $<10^{12}$ ions/cm^2, while NO measurements yielded a fraction of 78(3)% for a sample implanted at $T<1$ K with a dose $<10^{11}$ atoms/cm^2.[7] Furthermore, for Cs [$\Delta H_{\mathrm{sol}}=512$ kJ/mol (Ref. 5)], the second largest alkaline atom, a high substitutional fraction of 63(1)% has been observed with NO for a sample implanted at room temperature with a dose of $\sim 3\times 10^{13}$ ions/cm^2.[8]

The first two measurements of the hyperfine field of francium in iron published up to now yielded $B_{\mathrm{hf}}(\mathrm{FrFe})=70(5)$ T with ^{208}Fr (Ref. 3) and $B_{\mathrm{hf}}(\mathrm{Fr}Fe)=131(4)$ T with ^{221}Fr (Ref. 4), respectively. Although the lowest temperature that was reached in both experiments (i.e., ≈ 16 mK) did not allow for a full decorrelation between the two parameters that were fitted, i.e., B_{hf} and a temperature-independent parameter, it is clear from the data that this correlation is insufficient to explain the difference between the two values. In order to clarify this situation, we have performed several NO experiments using different francium isotopes. Among these are also the two that were used in the previous experiments. In addition, we have carried out a nuclear magnetic resonance experiment on oriented ^{208}Fr nuclei (NMR/ON) as well. Whereas NO is an integral method, yielding the value of the average hyperfine field experienced by the nuclei, NMR/ON probes the hyperfine field corresponding to a unique lattice site and this with a precision typically at least 10 times better than static nuclear orientation.

II. EXPERIMENTS AND RESULTS

$B_{\mathrm{hf}}(\mathrm{Fr}Fe)$ was measured for the light isotopes ^{208}Fr ($I^\pi=7^+$, $T_{1/2}=59.1$ s), ^{211}Fr ($I^\pi=9/2^-$, $T_{1/2}=3.1$ min), and ^{213}Fr ($I^\pi=9/2^-$, $T_{1/2}=34.6$ s) and for the heavier ^{221}Fr ($I^\pi=5/2^-$, $T_{1/2}=4.9$ min). All activities were produced at the ISOLDE isotope separator at CERN, with a 1 GeV proton beam of about 2 μA.[9] For ^{208,211,213}Fr a thorium-di-phtalo-cyamine+ThC$_2$ target was used. These isotopes were produced directly and one after the other implanted on line (at 60 keV) into the same iron foil (purity 99.998%, thickness 50 μm) that was soldered to the cold finger of the NICOLE ^{3}He-^{4}He dilution refrigerator.[10] The ^{221}Fr source was prepared by implanting ^{221}Rn ($T_{1/2}=25$ min) produced on a ThC$_2$ target. After implantation the ^{221}Rn β^- decayed to ^{221}Fr. In all experiments the implantation dose was kept below 2×10^{11} ions/cm^2 and the temperature during implantation was <1 K. To determine the temperature, ^{54}Mn*Ni* and ^{57}Co*Fe* nuclear orientation thermometers were used. The iron implantation foil was magnetized in an external magnetic field of $B_{\text{ext}}=0.5$ T, which was afterwards reduced to 0.2 T for the NO measurements. In the NMR/ON experiment with ^{208}Fr, an external field of 0.1 T was applied.

For ^{208,211,213}Fr the alpha radiation was detected with two particle detectors (i.e., 500 $\mu\text{m}\times 50\ \text{mm}^2$ Si *p-i-n* diodes) installed at angles of 18° and 51° with respect to the quantization axis. For ^{221}Fr three of these detectors were employed, this time mounted at angles of 16°, 50°, and 84°, respectively. To correct for variations of the beam intensity during the on-line implantations, only ratios of count rates observed by detectors at different detection angles can be used to determine the α anisotropies, which are defined as

$$R(\theta_1,\theta_2,T)=W(\theta_1,T)/W(\theta_2,T)-1,$$

with

$$W(\theta,T)=1+f\sum_{k=2,4,\ldots} A_k B_k(\nu_{hf}/kT)Q_k P_k(\cos\ \theta)$$

The coefficients B_k describe the orientation of the oriented nuclear state with hyperfine frequency ν_{hf} at the temperature T, and the A_k are the angular distribution coefficients of the observed α transition. The Q_k correct for the finite dimensions of the source and the detector, and $P_k(\cos\ \theta)$ are the Legendre polynomials, with θ being the angle between the orientation axis and the emission direction of the radiation.[11,12] Experimentally, one has

$$W(\theta,T)=\frac{N(\theta,T\leqslant 100\ \text{mK})}{N(\theta,T\cong 1\ \text{K})},$$

with $T\cong 1$ K corresponding to an unoriented sample.

The experimental α anisotropies versus inverse temperature for ^{208,211,213}Fr and ^{221}Fr are shown in Figs. 1 and 2, respectively. The NO data were evaluated in a simple two-site model, where a fraction f of the nuclei experiences the full hyperfine interaction and the rest $(1-f)$ is not oriented at all. This model has been proven to be quite reasonable from a large number of experiments on a wide variety of elements implanted at low dose and at low temperature.[13] The fraction f was derived from the analysis of the anisotropy data for the 325, 636, and 778 keV γ transitions in the decay of ^{208}Fr, yielding $f=32(10)\%$. It is not clear why this value is so small. A possible explanation for this could be the presence of a thin ice layer on the implantation foil. Since the range of Fr ions with $A=210$ and energy of 60 keV is only 56(6) nm,[24] a very thin layer would be sufficient to explain the observed effect. The large relative error in f is due to different results for different γ-detector combinations. Whereas the anisotropy data of one of the two γ detectors with angle 18° relative to the orientation axis normalized to the data of the γ detector at 90° suggested $0.22\leqslant f\leqslant 0.26$, data from the second detector at 18° with the same normalization suggested $0.33\leqslant f\leqslant 0.42$. Here the uncertainties in the theoretical $A_k U_k$ coefficients were already taken into account. As the reason for this discrepancy could not be resolved and alternative methods to derive f did not result in a more precise value, the existing uncertainty is represented in the quoted error.[14] For ^{221}Fr the fraction $f=79(2)\%$ was derived from several γ transitions in the decay of At isotopes. These were obtained through the β decay of Rn isotopes implanted into the same iron foil as ^{221}Rn (the precursor of ^{221}Fr). Note that apart from the low value of 32(10)% which was obtained in one single experiment (the isotopes ^{208,211,213}Fr, lines 4–6 of Table I, were all implanted into the same iron foil) and which is believed to be due to a technical problem (see above) and nothing specific to the hyperfine interaction of francium in iron, all values for f listed in Table I are consistent with implantation systematics. It is well known[13] for impurities that are insoluble in the host, as is the case for francium in iron,[5] that cold implantation at low dose ($<10^{13}$ atoms/cm^2) may result in fractions as high as 70%, while implantation at room temperature (and especially at higher doses) results in considerably lower fractions of the order of 40%.

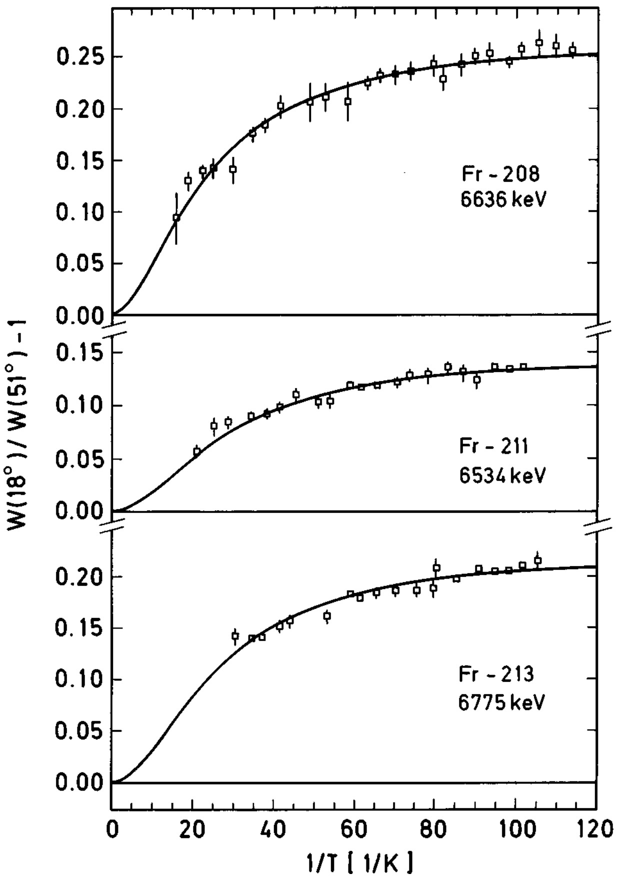

FIG. 1. Anisotropy $W(18°)/W(51°)-1$ vs inverse temperature for the 6636, 6534, and 6775 keV α lines in the decay of ^{208}Fr, ^{211}Fr, and ^{213}Fr, respectively.

From the α anisotropy data for ^{208,211,213}Fr, the hyperfine field was obtained from a two-parameter fit, adjusting to the data simultaneously the hyperfine interaction μB_{eff} (with $B_{\text{eff}}=B_{\text{ext}}+B_{\text{hf}}$) and fA_2. Since in this experiment only two detectors were available, it was not possible to include higher-order terms in the analysis as well. This, however, had only a rather small effect (i.e., $\leqslant 5\%$) on the result for B_{hf}.[14] For ^{221}Fr the $k=4$ term has been included in the analysis because data from detectors at three different detection angles were available.

For the NMR/ON experiment the isotope ^{208}Fr was cho-

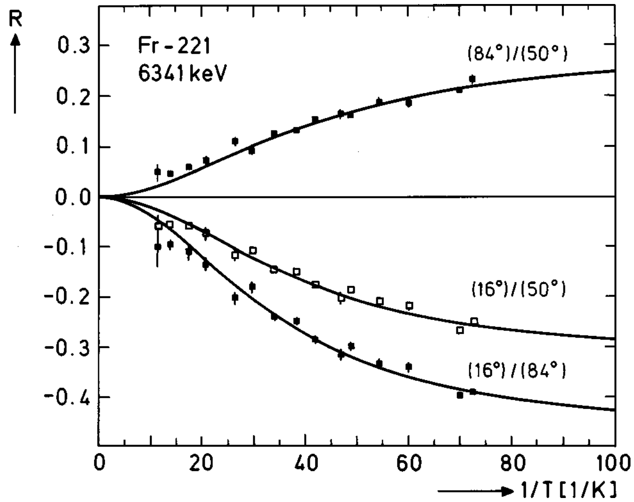

FIG. 2. Observed anisotropies $R=W(\theta_i)/W(\theta_j)-1$ (with $\theta=16°, 50°, 84°$) vs inverse temperature for the 6341 keV α line in the decay of ^{221}Fr.

sen because it has the largest α anisotropy of all light Fr isotopes which have been investigated with NO. The frequency range from 388 to 618 MHz was scanned four times, both upwards and downwards, in steps of 3 MHz with counting intervals of 50 s per point while monitoring the counting ratio $N(18°)/N(51°)$ of the 6636 keV α line in the decay of ^{208}Fr. The rf was frequency modulated (modulation width ± 3 MHz). The anisotropy of the ^{54}Mn*Ni* thermometer was monitored at the same time to look for indications of rf-power resonant heating. A small but clear destruction of α anisotropy was found at ν_L ($B_{\text{ext}}=0.1$ T)$=491.2(4)$ MHz. In Fig. 3 the count rate ratio versus rf frequency for the 6636 keV α line of ^{208}Fr and the count rate for the 835 keV γ ray of the ^{54}Mn*Ni* thermometer are shown for the frequency range around the resonance. If the effect on the α line were due to rf heating, a similar (but positive) effect with an amplitude of about 300 counts above background, i.e., slightly larger than the statistical errors, should be visible on the 835 keV count rate. No indication of such an effect is seen. This fact and, especially, the narrow width of the full width at half maximum (FWHM)$=6.7(12)$ MHz, i.e., about 1.4% of the center frequency, of the observed destruction of α anisotropy secure that it has to be interpreted as a nuclear resonance signal. The integrated destruction of anisotropy is $S=5(1)\%$.

From the center frequency, corrected for the external magnetic field, the hyperfine field was deduced as $B_{\text{hf}}(\text{Fr}Fe)=94.9(4)$ T, using $\mu(^{208}\text{Fr})=4.75(2)\mu_N$.[15] No correction for the Knight shift and for diamagnetism has been applied. No other narrow resonance signal with $S\geq 1\%$ was observed in the frequency region corresponding to fields between 75 and 119 T.

III. DISCUSSION

In Table I the results from the NMR/ON and NO measurements of this work, as well as of previous NO experiments with Fr isotopes, are listed. The value of $B_{\text{hf}}(\text{Fr}Fe)$ derived from static NO data of the lighter francium isotopes (the results for ^{221}Fr will be discussed at the end of this section) are in the range from 70 to 93 T, i.e., from at most 26% below the NMR/ON value up to values which are in accord with NMR/ON within errors. This fits into the systematics of such measurements.[13] It must further be noted that only such NO measurements which cover a wide temperature range and which do not suffer from correlation of fit parameters have an appreciable higher precision. In addition, the accuracy of results from static NO data is only high if the hyperfine interaction model used for data evaluation is matched to the case, which makes further information necessary. Keeping this in mind together with the fact that the static NO data were evaluated in a very simple site distribution model, which certainly is a simplified description of the real hyperfine interaction distribution after cold implantation, this result is quite satisfactory.

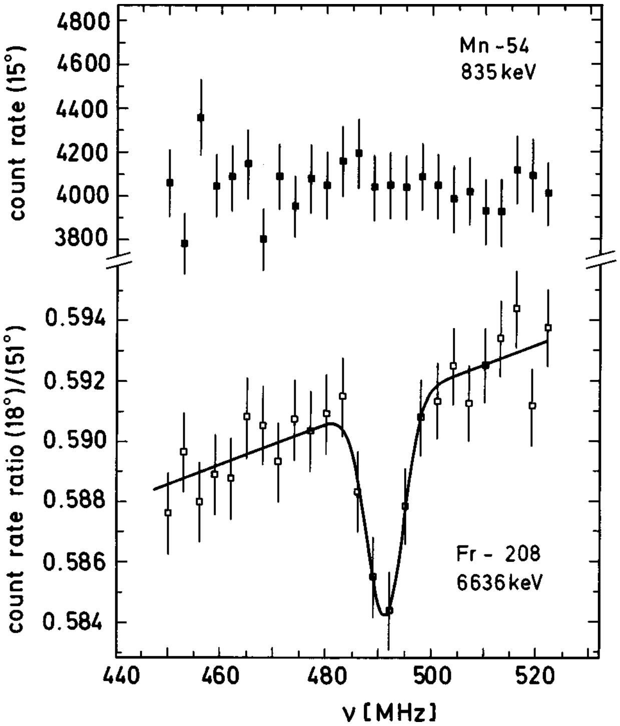

FIG. 3. Lower part: counting ratio $N(18°)/N(51°)$ vs rf frequency ν for the 6636 keV α line in the decay of ^{208}Fr. The center frequency was determined to be 491.2(4) MHz with $B_{\text{ext}}=0.1$ T. Upper part: count rate $N(15°)$ vs rf frequency ν for the 835 keV γ line of the ^{54}Mn*Ni* thermometer, showing no indication for a heating effect in the region of the nuclear resonance.

The shape and width of the resonance signal suggest that the NMR/ON field corresponds to francium nuclei in a site with a unique hyperfine interaction. Although only 5% of the nuclei which contribute to the anisotropy were resonated, it is probable that the field from NMR/ON is the field experienced by francium nuclei on unique substitutional lattice sites in host iron. The reason for this conclusion is the large substitutional fraction of 67(13)% for francium in iron found in emission channeling experiments after cold implantation at low dose,[6] i.e., conditions similar to those of the work reported here. The fact that (i) the results from our integral NO data on light francium nuclei implanted into the same iron foil agree with the NMR/ON result (see Table I) and that (ii) the same is true for the hyperfine field that was observed in earlier experiments on Fr*Fe* (Refs. 3 and 16) (in

TABLE I. Magnetic hyperfine field of Fr in iron from NMR/ON and different NO experiments. Note the observation of two groups of values for B_{hf}.

A	E_α (keV)	μ[a] (μ_N)	B_{hf} (T)	f (%)	Dose (ions/cm^2)	Remarks
208	6636	4.75(2)	94.9(4)		$<2\times10^{11}$	on-line implantation, NMR/ON[b]
207	6768	3.89(8)	91(8)	78(3)	$<10^{11}$	on-line implantation[c]
208	6636	4.75(2)	70(5)	78(3)	$<10^{11}$	on-line implantation[d]
208	6636	4.75(2)	93(4)	32(10)	$<2\times10^{11}$	on-line implantation[b]
211	6534	4.00(8)	77(5)	32(10)	$<2\times10^{11}$	on-line implantation[b]
213	6775	4.02(1)	86(5)	32(10)	$<2\times10^{11}$	on-line implantation[b]
221	218[e]	1.58(3)	130(4)	42(4)	$<7\times10^{13}$	^{225}Ra off-line sample[f]
221	6341	1.58(3)	141(10)	42(4)	$<7\times10^{13}$	^{225}Ra off-line sample[f]
221	6341	1.58(3)	121(6)	37(5)	$\sim3\times10^{13}$	^{225}Fr off-line sample[g]
221	6341	1.58(3)	109(4)	70(7)	$<2\times10^{11}$	^{225}Rn off-line sample[h]
221	6341, 6126	1.58(3)	124(6)	79(2)	$<2\times10^{11}$	^{221}Rn on-line implantation[b]

[a] From Ref. 15.

[b] This work.

[c] New analysis of data from experiment reported in Ref. 16.

[d] Reference 3.

[e] γ-ray energy.

[f] Reference 6.

[g] Reference 19.

[h] Reference 20.

which a high value for f, i.e., 78(3)%, was obtained) seems to indicate that in all these experiments always the same (unique) lattice site with a significantly large B_{hf} was preferentially populated. Further support of this idea is given by the fact that we did not find any other resonance in the frequency range corresponding to fields between 75 and 119 T.

The experimental integrated destruction of anisotropy gives only a lower limit for the fraction of nuclei at the site that was resonated, since it was not possible to attempt to increase the destruction by increasing the rf power because of limited beam time at the isotope separator. The low value for the destruction may also be due to the fact that the Fr atoms, being much larger than the Fe atoms, can collect vacancies in their vicinity during the thermal spike regime. This could lead to a large distribution of the hyperfine field such that for a large part of the Fr nuclei the resonance frequency is shifted a few MHz from the frequency of undisturbed lattice sites. However, the sensitivity of the experiment was not high enough to detect such a possible broad background signal.

The systematics of magnetic hyperfine fields of heavy elements in host iron is shown in Fig. 4. The experimental values were taken from Refs. 1 and 2, while the value for francium is the one reported here. The line connecting the points was drawn to guide the eye. It shows the smooth trend from negative fields for the $5d$ atoms over large positive fields for $6sp$ atoms, with a maximum around astatine, to again negative values for the actinide atoms. The value of francium on the steep back slope of the curve fits very well in this picture. Because of their simple electronic configuration, the alkaline atoms should be ideal test cases for the theoretical understanding of the hyperfine fields of impurities in iron. The case of Cs*Fe* has been measured by NMR/ON as $B_{hf}(\mathrm{Cs}Fe)=27.8(2)$ T.[8] The ratio of 3.41 for the francium and cesium hyperfine fields in iron compares very well with that of the free-atom hyperfine fields, i.e., 3.14. The latter could be rather well reproduced by relativistic calculations,[17] which, taking into account nuclear-size effects, resulted in a ratio of 3.27. Akai *et al.*[18] have performed microscopic self-consistent hyperfine-field calculations for impurities with $Z\leqslant56$ in host iron. Although their results for light atoms are very promising, their calculations fail to reproduce experimental values for atoms of the fifth period of the periodic table, the heaviest they treat in their paper. The authors state that one of the reasons for the shortcomings of their calculations is the neglect of lattice expansion through the larger atoms. This effect is expected to be even more important for francium. The availability of accurate experimental values for hyperfine fields of alkaline atoms in iron should be an incentive for theoretical calculations with consideration of the above-mentioned effects. At least for the alkalines with their simple electronic shell it should be possible to reproduce the experimental values quantitatively for all Z.

The results of the five static NO measurements for $B_{hf}(^{221}\mathrm{Fr}Fe)$ which are listed in Table I are all considerably higher—up to 38(5)% if one disregards the less precise value from α detection of Ref. 6—than the NMR/ON result of this work. While the experiments with the lighter francium isotopes were all performed with on-line implantation, in the experiments with ^{221}Fr the activity was produced via various decay chains after both off- and on-line implantation. Also, in the ^{221}Fr measurements, the implantation doses differed significantly for different experiments and the fraction in good sites varied by a factor of 2. Because of these facts, it is difficult to believe that the reason for the higher average hyperfine fields of the ^{221}Fr*Fe* NO experiments should lie in a common feature of the sample preparation, although it can-

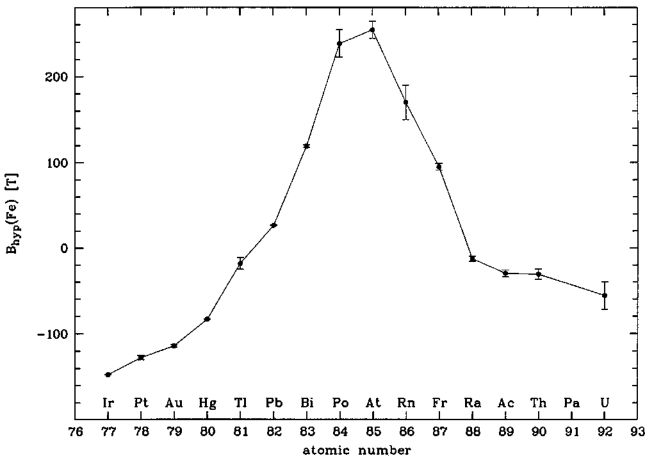

FIG. 4. Systematics of the experimental hyperfine field values in iron for impurities with $Z=77-92$ (Refs. 1 and 2).

not be excluded that the fact that the ^{221}Fr samples were obtained through the decay of precursor nuclei which were implanted in the iron foil may be responsible for this. Another possible reason could lie in a correlation of the parameters $B_{\rm hf}$ and f in the least-squares data evaluation, because even at the lowest experimental temperatures, saturation of orientation was not fully reached. Such a parameter correlation effect should, however, be rather small in our case because in two experiments[19,20] temperatures below 10 mK with considerable saturation of orientation could be reached. Also, the good simultaneous fit of theoretical anisotropy functions to the data for the three detector combinations in this work (see Fig. 2), where the $k=4$ term was not neglected, is in disagreement with a considerable correlation effect.

Because of the short half-lives of the francium isotopes considered in this paper, ranging from 14.8 s for ^{207}Fr to 4.8 min for ^{221}Fr, the possibility of effects due to incomplete spin-lattice relaxation has to be discussed. No experimental relaxation data for francium in the iron host are available. By scaling relaxation data for ^{206}Bi*Fe* (Ref. 25) (the nearest isotope for which experimental relaxation data in iron are available) in the low-temperature limit,[26] an effective Korringa constant $C_K=387$ mK s is obtained for ^{221}Fr, resulting in an effective relaxation time $T_1'(^{221}\mathrm{Fr})\approx 7$ s. Using the empirical relation for the relaxation of impurities in iron,[26] derived from experimental data for a large number of isotopes from different elements, a similar value is found. This short time guarantees that ^{221}Fr should be fully relaxed. Thus the observed small deviation of the anisotropy data points at the highest temperatures from the theoretical curves in Fig. 2 cannot be due to relaxation effects. For the shortest lived of the light francium isotopes, i.e., ^{207}Fr, an effective relaxation time of about 1.5 s is estimated, implying that relaxation effects should be negligible in this case as well. Also, the finding that for the light isotopes, with quite different half-lives (ranging from 14.8 s for ^{207}Fr to 3.1 min for ^{211}Fr), within errors always the same low hyperfine field value is found (see Table I) excludes the influence of relaxation.

Since in NO experiments only the product of the magnetic moment and hyperfine field is determined, a simple explanation for the fact that different hyperfine fields are observed in the measurements with light francium isotopes and those with ^{221}Fr would be the use of incorrect magnetic moments in the derivation of the hyperfine fields. The magnetic moments of the francium isotopes of interest in this paper have all been measured by high-resolution laser spectroscopy[15] with ^{212}Fr as common reference and ^{211}Fr as common calibration isotope. The $5/2^-$ ground state of ^{221}Fr is believed to be a decoupled proton $h_{9/2}$ state[21] which, because of the antiparallel coupling of spin and orbital momentum, should show some hyperfine anomaly[22,23] relative to ^{208}Fr. However, only the difference of the hyperfine anomaly in the laser experiment $^{221}\Delta^{208}_{\rm las}$ from that in the different environment iron $^{221}\Delta^{208}_{\rm Fe}$ can be responsible for the difference in hyperfine fields displayed in Table I.[23] It is difficult to believe that such a hyperfine anomaly effect could account for a larger part of the observed difference in fields.

IV. SUMMARY

Both NO and NMR/ON measurements were carried out on a number of francium isotopes in order to determine the hyperfine field of francium in iron. The data for the light isotopes ^{208,211,213}Fr are all in agreement with one another as well as with previous results obtained with ^{207,208}Fr (Refs. 3 and 16) and with the NMR/ON experiment on ^{208}Fr, which yielded $B_{\rm hf}(\mathrm{Fr}Fe)=94.9(4)$ T. From a comparison with the results of emission channeling experiments with francium in iron,[6] it is concluded that this field probably corresponds to francium atoms at substitutional lattice sites. All present and previous NO measurements on ^{221}Fr yielded a significantly larger result for the effective value of the hyperfine field. At present, there is no simple explanation for this observation. It should be mentioned that a similar situation exists with cesium isotopes.[8,13]

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