

24, 1255 (1970).

<sup>3</sup>J. A. Lipa, C. Edwards, and M. J. Buckingham, *Phys. Rev. Lett.* **25**, 1086 (1970).

<sup>4</sup>By rounding we shall mean the systematic deviation of a measured quantity from its ideal behavior as  $T \rightarrow T_c$ .

<sup>5</sup>Grown by A. Linz, Center for Materials Science and Engineering, Massachusetts Institute of Technology.

<sup>6</sup>B. Golding, to be published.

<sup>7</sup>It is assumed that  $\text{RbMnF}_3$  exhibits the macroscopic properties of a cubic crystal in the transition region. We therefore have  $\beta = 3\alpha_L$ , where  $\alpha_L = (1/l)(dl/dT)$  is the measured linear thermal expansivity and  $l$  is a linear dimension of the crystal.

<sup>8</sup>See, for instance, M. E. Fisher, *Rep. Progr. Phys.* **30**, 615 (1967).

<sup>9</sup> $\beta$  and  $C_p$  are linearly related by the exact thermodynamic relation  $\beta = aC_p + b$ , where  $a = (VT)^{-1}(\partial T/\partial P)_t$  and  $b = -V^{-1}(\partial S/\partial P)_t$ . The partial derivatives are evaluated along a path of constant  $t = T_c - T$  and are only weakly temperature dependent [see M. J. Buckingham and W. M. Fairbank, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1961)]. Therefore,  $\beta$  and  $C_p$  will have the same asymptotic temperature dependence, i.e., both may be described by the critical exponents  $\alpha$  and  $\alpha'$ . The relative magnitudes of higher-order terms will not be the same generally, and the asymptotic regions for  $\beta$  and  $C_p$  may be considerably different.

<sup>10</sup>We emphasize that the minimum in  $\sigma$  has no funda-

mental or statistical significance. Significance is attached, however, to the rise in  $\sigma$  as  $\Delta T$  is decreased beyond this point. The minimum in  $\sigma$  is convenient, therefore, as an approximate delimiter of the "rounded" region.

<sup>11</sup>We also have  $A = 4.56$ ,  $B = 4.68$ ,  $A' = 5.49$ , and  $B' = 17.82$  in  $10^{-6} \text{ K}^{-1}$ . The error in  $T_c$  is stated relative to the expansivity measurements. The possible error in the absolute temperature scale is  $\pm 0.06 \text{ K}$ .

<sup>12</sup>B. Widom, *J. Chem. Phys.* **43**, 3892, 3898 (1965).

<sup>13</sup>R. B. Griffiths, *Phys. Rev.* **158**, 176 (1967).

<sup>14</sup>D. T. Teaney, V. L. Moruzzi, and B. E. Argyle, *J. Appl. Phys.* **37**, 1122 (1967).

<sup>15</sup>Because of the uncertainties in  $T_c$  and the extent of the rounding near  $T_c$ , a detailed investigation of the form of possible higher-order contributions has not proved fruitful. In liquid helium near the  $\lambda$  point, where  $T_\lambda$  can be measured very precisely and where the effect of the gravitational inhomogeneity on the specific heat can be easily calculated (Ref. 1), estimates of the magnitude of higher-order terms have been made and contribute significantly to  $C_p$  below  $T_\lambda$  in the region  $\epsilon \gtrsim 3 \times 10^{-4}$ .

<sup>16</sup>M. E. Fisher, to be published; see also the "Note added in proof" in Ref. 1.

<sup>17</sup>B. J. C. van der Hoeven, Jr., D. T. Teaney, and V. L. Moruzzi, *Phys. Rev. Lett.* **20**, 719 (1968).

<sup>18</sup>H. Y. Lau, L. M. Corliss, A. Delapalme, J. M. Hastings, R. Nathans, and A. Tucciarone, *J. Appl. Phys.* **41**, 1384 (1970).

## Lattice-Location Studies on Tl, Pb, and Bi in Iron, and the Hyperfine Field at Pb in Iron

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The channeling technique has been employed to study the lattice location of Tl, Pb, and Bi implanted into iron single crystals. The results, when compared with measured magnetic hyperfine fields acting on Pb in ferromagnetic iron, provide evidence for the association of high-field sites with Pb in nonsubstitutional positions.

Recent measurements of the magnetic hyperfine field at a Pb impurity in Fe, using the perturbed angular-correlation technique, are in disagreement. Field values measured for radioactive sources in which the parent nuclide had been implanted into the Fe host<sup>1,2</sup> were found to be a factor of 2 smaller than those obtained using metallurgically prepared sources.<sup>3,4</sup> We have investigated the lattice location in Fe of implanted impurities corresponding to the radioactive species used in the field measurements and have concluded that the observed difference in hyperfine fields is the result of dissimilar lattice positions of Pb in Fe as originally suggested by the authors of Ref. 1. Contrary to their proposal, however, we

find that the larger field value is to be associated with nonsubstitutional positions in the lattice.

Tl and Pb were implanted at 170 and 100 keV, respectively, by an isotope separator into carefully prepared Fe single crystals at room temperature to a dose of  $2 \times 10^{14}/\text{cm}^2$ . The volume concentration of the impurities in the implanted layer was less than 0.25%. The samples were then mounted on a goniometer surrounded by a copper can held at  $-300 \text{ V}$  to assure accurate beam-current integration and placed in the beam line of a 2-MV Van de Graaff accelerator.  $^4\text{He}^+$  ions back-scattered into an annular silicon surface-barrier detector were employed to locate crystal axes relative to the beam direction by the channeling

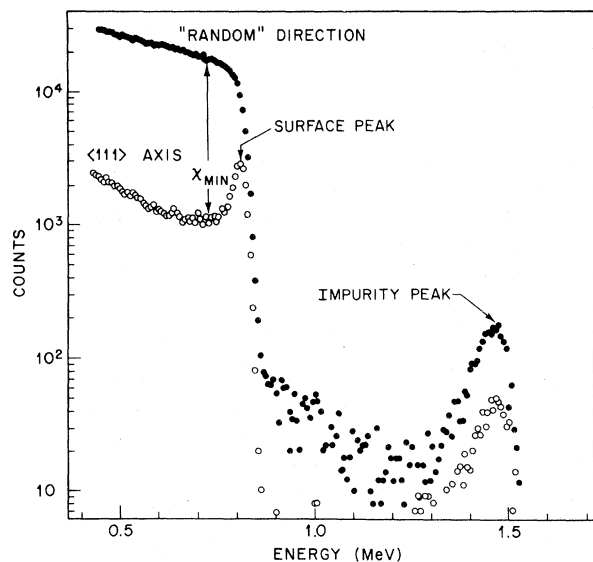


FIG. 1. Energy spectra of backscattered  $^{12}\text{C}^+$  ions incident at 2.0 MeV in channeling (open circles) and nonchanneling (closed circles) directions on an Fe single crystal implanted with  $2 \times 10^{14}$  Pb atoms/cm $^2$  at 100 keV.

technique.<sup>5</sup> The incident beam was defined to better than  $0.06^\circ$  angular divergence by suitable collimation.

Channeling has the property of suppressing the scattering of ions by atoms on regular lattice sites relative to those on irregular sites and can provide a quantitative measure of the lattice-location distribution of impurities in a single crystal.<sup>6</sup> The yields of 2.0-MeV  $^4\text{He}^+$  and  $^{12}\text{C}^+$  ions incident along major channeling directions and backscattered from the heavy impurities and host lattice were measured and compared with the corresponding yields for nonchanneled incident ions. Typical backscattered-ion energy spectra are shown in Fig. 1. The fraction of impurity atoms which are within  $\sim 0.1 \text{ \AA}$  of the row of host atoms parallel to the channeling direction is given by the relation  $(1 - R)/(1 - \chi_{\text{min}})$  where  $R$  is the ratio of the areas of the impurity peaks in the channeling and nonchanneling directions and  $\chi_{\text{min}}$ , a measure of the unchanneled beam, is a similar ratio for the Fe host measured just behind the "surface peak" as shown in Fig. 1. The quantity  $(1 - R)/(1 - \chi_{\text{min}})$ , denoted the substitutional fraction, for Tl and Pb is presented in Table I. It is clear from the data that the implanted Tl and Pb came to rest predominantly on substitutional sites.<sup>7</sup>

A dose of  $2 \times 10^{14}$  Bi atoms/cm $^2$  was implanted at 100 keV into each of several Fe crystals at room temperature. The samples were mounted

TABLE I. Results for Tl and Pb in Fe.

	Axis	Substitutional <sup>a</sup> fraction
Tl	$\langle 111 \rangle$	0.84(14)
Pb	$\langle 111 \rangle$	0.78(6)
	$\langle 110 \rangle$	0.84(6)
	$\langle 100 \rangle$	0.85(6)

<sup>a</sup>The uncertainties in the last digit are indicated by the figures enclosed in parentheses.

on a heating goniometer to perform lattice-location measurements as a function of annealing temperature in order to reproduce the high-temperature source preparation of  $^{207}\text{Bi}$  in Fe employed in Refs. 3 and 4. For temperatures above  $675^\circ\text{C}$  an external quartz-tube vacuum oven was used to anneal the samples. In this case, 1.9-MeV  $^{14}\text{N}^+$  ions were used as a probe.<sup>8</sup> The results, as listed in Table II and displayed in Fig. 2, represent averages over several runs. The onset of a marked decrease in the substitutional fraction is evident at an annealing temperature of  $600^\circ\text{C}$ . At and above this temperature, out-diffusion of Bi from the crystals was also observed. Out-diffusion is consistent with an increase in mobility of Bi in Fe and thus also with a change in lattice location.

TABLE II. Substitutional fraction for Bi in Fe for several annealing temperatures.

Annealing temperature <sup>a</sup> (°C)	Substitution fraction <sup>b</sup>		
	$\langle 100 \rangle$	$\langle 110 \rangle$	$\langle 111 \rangle$
25	0.81(6)	0.81(8)	0.77(6)
150			0.70(8)
300	0.91(8)		0.90(8)
450			0.83(8)
500		0.78(8)	0.83(8)
600	0.67(8)		0.70(8)
660			0.52(8)
675		0.54(8)	0.64(6)
700	0.61(8)		0.41(8)
725		0.39(10)	0.34(10)
750			0.37(10)
775		0.21(10)	0.31(10)
800	0.25(10)		0.39(10)
825		0.27(10)	0.32(10)

<sup>a</sup>Annealing temperatures were applied for 30 min. Heating and cooling times were approximately 15 to 20 min.

<sup>b</sup>The uncertainties in the last digit are indicated by the figures enclosed in parentheses.

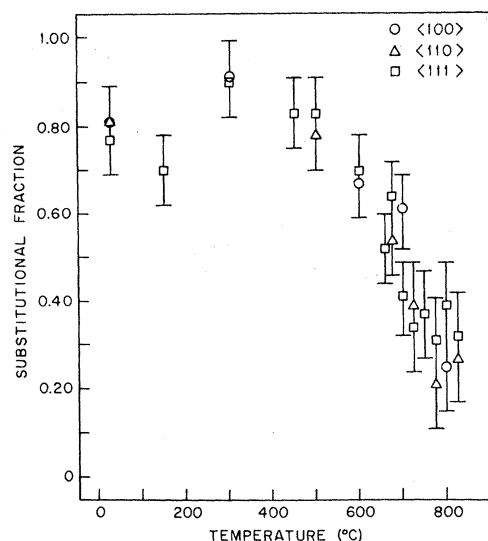


FIG. 2. Substitutional fraction (as defined in text) for implanted Bi impurity in Fe single crystals as a function of annealing temperature. The results shown are those presented in Table II.

We infer from these results that the implanted Tl and Pb parent nuclei, in the measurements which yielded the low field values,<sup>1,2</sup> were primarily on regular Fe lattice sites. On the other hand, Bi introduced metallurgically at temperatures at or above 600°C, for which the high field value was measured,<sup>3,4</sup> is largely at nonsubstitutional lattice locations.<sup>9</sup> Thus these data represent the first evidence of a stronger magnetic interaction *off*, as opposed to *on*, regular lattice sites.

Several theoretical descriptions of the origin of magnetic hyperfine fields at impurities in ferromagnets have appeared in the literature.<sup>10-16</sup> Systematic trends have been well reproduced, but accurate predictions in specific cases have not been possible because the fields result, in general, from detailed cancellation of the relatively large contributions from both conduction electron and core polarization. The phenomenological theory of Balabanov and Delyagin<sup>13</sup> predicts the field at Pb in Fe to be +247 kOe which agrees with the "substitutional" value. A slightly smaller conduction *s*-electron density or slightly greater overlap of Pb and Fe lattice wave functions at nonsubstitutional sites could easily account for the larger field observed there.<sup>16</sup> In fact, the observation of the higher field and nonsubstitutional lattice location in the samples subjected to high temperature may be indicative of the local formation of covalent bonds similar to those one expects in intermetallic compounds.

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<sup>1</sup>J. D. Bowman and F. C. Zawislak, Nucl. Phys. **A138**, 90 (1969). In this work the hyperfine field at <sup>208</sup>Pb in Fe was found to be 280 ± 70 kOe. <sup>208</sup>Tl, the immediate precursor of <sup>208</sup>Pb, was implanted by recoil from a preceding  $\alpha$ -particle emission in the decay chain of <sup>228</sup>Th.

<sup>2</sup>E. Bodenstedt has communicated, before publication, a measurement of ~300 kOe for the field at <sup>204</sup>Pb in Fe. In this case <sup>204m</sup>Pb was implanted by an isotope separator.

<sup>3</sup>In Ref. 1, a measurement of the field at <sup>207</sup>Pb in Fe yielded a value of 660 ± 45 kOe. The source of <sup>207</sup>Bi in Fe was prepared by diffusion at high temperature (~600°C).

<sup>4</sup>E. N. Kaufmann, Phys. Lett. **35A**, 165 (1971). This work confirmed the high field value found in Ref. 1 which had been in disagreement with an earlier measurement [G. C. Pramila, S. G. Cohen, and L. Grodzins, Phys. Lett. **24A**, 7 (1967)].

<sup>5</sup>J. Lindhard, Kgl. Dan. Vidensk. Selsk., Mat.-Fys. Medd. **34**, No. 14 (1965).

<sup>6</sup>The use of channeling as a tool for detecting the lattice location of impurities is most completely described in J. W. Mayer, L. Eriksson, and J. A. Davies, *Ion Implantation in Semiconductors: Silicon and Germanium* (Academic, New York, 1970).

<sup>7</sup>The correction factor  $1 - \chi_{\min}$  was always  $\geq 0.9$  as measured in this way but may, in fact, be somewhat smaller since it should be measured at an energy which corresponds to scattering at the depth of the impurity (not resolvable with our depth resolution). This correction would have no effect on the arguments presented here. The analysis does not take into account the recently observed phenomenon of certain interstitial sites displaying a greater than normal yield when the beam is incident in a channeling direction [see J. U. Andersen *et al.*, Radiat. Eff. **7**, 25 (1971); B. Domeij *et al.*, Radiat. Eff. **6**, 155 (1970)]. If properly accounted for, this effect would yield somewhat higher substitutional fractions than those given in Tables I and II and would thus have a minor effect on the room-temperature measurements. The agreement between the various axes in the high-temperature data also indicates that this effect is not sizable in this region.

<sup>8</sup>After each anneal the oxide-layer thickness was noted by observing the surface peak in the channeling spectrum. It was shown for all measurements that less than 10% of the implanted Bi resided in an oxide layer by using the area of the surface peak and assuming a Gaussian distribution for the impurity layer of width and position defined by the backscattering spectrum.

<sup>9</sup>The very small nuclear recoil energies after the  $\beta$  decay, electron capture, or  $\gamma$  emission are not expected to produce a change in the lattice location of the decay products. It has been shown for the case of  $^{212}\text{Bi}$  in silicon that  $\beta$  decay does not produce a change in the lattice location. [E. Uggerhoj, in *Radiation Damage in Reactor Materials* (International Atomic Energy Agency, Vienna, 1969), Vol. 1.]

<sup>10</sup>A. J. Freeman and R. E. Watson, *Phys. Rev.* **123**, 2027 (1961).

<sup>11</sup>E. Daniel and J. Friedel, *J. Phys. Chem. Solids* **24**,

1601 (1963).

<sup>12</sup>D. A. Shirley and G. A. Westenbarger, *Phys. Rev.* **138**, A170 (1965).

<sup>13</sup>A. E. Balabanov and N. N. Delyagin, *Zh. Eksp. Teor. Fiz.* **54**, 1402 (1968) [*Sov. Phys. JETP* **27**, 752 (1968)].

<sup>14</sup>I. A. Campbell, *Proc. Roy. Soc., Ser. A* **311**, 131 (1969).

<sup>15</sup>D. A. Shirley, *Phys. Lett.* **25A**, 129 (1967).

<sup>16</sup>M. B. Stearns, Scientific Research Staff Publication Preprint, Ford Motor Company, 28 January 1971 (to be published), and *Phys. Lett.* **34A**, 146 (1971).

## Measurement of the Variance of the Number of Neutrons Emitted in Fission of $^{252}\text{Cf}$ as a Function of the Fragment Mass and Total Kinetic Energy

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We have measured the variance of the number of neutrons emitted by individual fragments in fission of  $^{252}\text{Cf}$  as a function of the fragment mass and the total kinetic energy released. The variance does *not* show pronounced "sawtooth" structure as a function of the fragment mass. We deduce from this that the correlation coefficient between the excitation energies of complementary fission fragments is substantially less than unity for at least some portions of the fragment mass-ratio distribution.

The distribution of the average number of neutrons emitted in fission by the individual fission fragments provides a way of estimating the partition of the total excitation energy between the two fragments. A summary of the data on this distribution has been presented by Terrel.<sup>1</sup> From these data it is evident that the sawtooth dependence of the average number of neutrons  $\bar{\nu}(A)$  as a function of the fragment mass  $A$  is a general feature of low-energy fission. Data presented by Bowman *et al.*,<sup>2</sup> Milton and Fraser,<sup>3</sup> and Nardi and Fraenkel<sup>4</sup> show that the derivative of the average number of neutrons with respect to the total fragment kinetic energy  $E_K$ , denoted by  $(\partial\bar{\nu}/\partial E_K)(A)$ , also exhibits a sawtooth dependence as a function of the fragment mass. This parallel behavior of  $\bar{\nu}(A)$  and  $(\partial\bar{\nu}/\partial E_K)(A)$  indicates that fragments with high excitation energy are also more susceptible to receiving additional excitation energy. The results do not, however, provide any information about the *width* of the excitation-energy distribution or about any possible cross correlation between the excitation energies of the two fragments.

We have measured the variance in the neutron-number distribution of individual fission fragments emitted in  $^{252}\text{Cf}$  fission, as a function of the fragment mass and total kinetic energy. A  $^{252}\text{Cf}$  source of about  $2 \times 10^5$  fission/min deposited on a thin Ni backing was placed between two surface-barrier fission fragment detectors, denoted  $F_1$  and  $F_2$  in Fig. 1. These detectors together with the source were placed inside an aluminum vacuum chamber of 30-cm diam and 0.5-cm wall thickness. The fission fragment detectors  $F_1$  and  $F_2$  were placed at a distance of 5.5 cm from the source and subtended an angle of  $19^\circ$  with respect to it. The neutrons were detected by means of

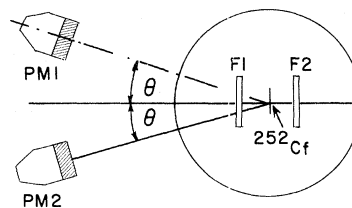


FIG. 1. Experimental arrangement.