

by the electrons changes in time as a linear function of  $\cos(n\omega^*t)$  and  $\sin(n\omega^*t)$ , where  $n = 0, 1, 2, \dots$ . This in turn will appear as a force acting on the electrons with the same time dependence. When Eq. (6) is fulfilled,  $\omega^* = \omega_{ce}$ ; otherwise  $\omega^*$  represents a spectrum of radian frequencies. Under the resonant condition of Eq. (5), one expects to get radiation having radian frequencies  $\omega_{ce}, 2\omega_{ce}, 3\omega_{ce}, \dots$ . However, when Eq. (6) is not valid, this effect will tend to widen the frequency distribution. Because of this nonlinear interaction, rf radiation with wavelength  $\lambda$  can be produced from a bunch of electrons with a scale length greater than  $\lambda$ .

The theoretical considerations and experimental results described here suggest a new mechanism for coherent rf emission. This mechanism can be of some importance in explaining nonthermal rf radiation from celestial objects.<sup>15</sup> Some of the observed rf emissions from astrophysical phenomena cannot be explained by any simpler mechanism.<sup>16</sup> The rippled magnetic field can be produced in astrophysical phenomena by any one of several instabilities which have been detected in laboratory plasmas.

<sup>1</sup>A. P. Harvey, *Coherent Light* (Interscience, New

York, 1970).

<sup>2</sup>J. Nation, *Appl. Phys. Lett.* **17**, 491 (1970).

<sup>3</sup>R. Q. Twiss, *Aust. J. Phys.* **11**, 564 (1958).

<sup>4</sup>V. V. Zheleznyakov, *Astron. Zh.* **44**, 42 (1967) [*Sov. Astron. AJ* **11**, 33 (1967)].

<sup>5</sup>M. Friedman and M. Herndon, *Phys. Rev. Lett.* **28**, 210 (1972).

<sup>6</sup>R. M. Phillips, *IEEE Trans. Electron Devices* **7**, 231 (1960).

<sup>7</sup>C. E. Enderby and R. M. Phillips, *Proc. IEEE* **53**, 1648 (1965).

<sup>8</sup>A. F. Harvey, *Microwave Engineering* (Academic, New York, 1963).

<sup>9</sup>J. Nation, *Rev. Sci. Instrum.* **41**, 1097 (1970).

<sup>10</sup>W. B. Thompson, private communication.

<sup>11</sup>G. Bekefi, *Radiation Process in Plasmas* (Wiley, New York, 1966).

<sup>12</sup>A. A. Sokolov and I. M. Ternov, *Dokl. Akad. Nauk SSSR* **166**, 1332 (1966) [*Sov. Phys. Dokl.* **11**, 156 (1966)], and *Zh. Eksp. Teor. Fiz., Pis'ma Red.* **4**, 90 (1966) [*JETP Lett.* **4**, 61 (1966)].

<sup>13</sup>A. P. Slabospitskii, V. D. Ferdorchenko, and B. W. Ruthevick, in *Plasma Physics and Controlled Thermonuclear Fusion*, edited by K. D. Sinelnikov (Israel Program for Scientific Translation, Jerusalem, 1965), No. 4.

<sup>14</sup>E. W. Laing and A. E. Robson, *Plasma Phys.* **3**, 146 (1961).

<sup>15</sup>M. Friedman, to be published.

<sup>16</sup>V. L. Ginzburg and V. V. Zheleznyakov, *Comments Astrophys. Space Phys.* **2**, 107, 167 (1970).

## Ising-Model "Metamagnet" and Tricritical Susceptibility Exponent\*

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A spin- $\frac{1}{2}$  Ising-model "metamagnet" is studied by the method of high-temperature expansions. The phase boundary in the  $H$ - $T$  plane is obtained for a simple cubic lattice with in-plane ferromagnetic interactions but between-plane antiferromagnetic coupling, and a tricritical point is located. Along the critical line, the staggered susceptibility appears to have an exponent  $\frac{5}{4}$  (consistent with the universality hypothesis), while at the tricritical point, the direct susceptibility shows a tricritical exponent of  $\frac{1}{2}$ .

Griffiths<sup>1,2</sup> has recently called attention to the existence of "tricritical points" in the phase diagram of metamagnetic systems. At the tricritical temperature  $T_t$ , the phase transition changes abruptly from second to first order. Such behavior has been observed in real materials such as  $\text{FeCl}_2$ ,<sup>3</sup>  $\text{Ni}(\text{NO}_3)_2 \cdot 2\text{H}_2\text{O}$ ,<sup>4</sup> and dysprosium aluminum garnet.<sup>5</sup> The tricritical point is characterized by its own set of exponents; in particular, we may expect that critical exponents will change *discontinuously* at  $T_t$  from their values on the second-order phase boundary, along which they remain

constant.<sup>6,7</sup>

Previous theoretical work has been restricted to Landau's "classical theory"<sup>8</sup> or molecular theory,<sup>9</sup> and some very recent Monte Carlo studies of models with tricritical points.<sup>10</sup> Riedel<sup>11</sup> has presented a scaling theory near the tricritical point consistent with experimental work on  $\text{He}^3$ - $\text{He}^4$  mixtures,<sup>12</sup> a system whose phase diagram is thermodynamically closely analogous to that of metamagnetic materials.<sup>1</sup> An Ising Hamiltonian has been proposed for the  $\text{He}^3$ - $\text{He}^4$  system<sup>13</sup> and analyzed by series expansions,<sup>14,15</sup> although

the series work gives no values for tricritical exponents.

In this note we give the phase boundary and report an estimate for the tricritical susceptibility exponent for a three-dimensional model "metamagnet" using high-temperature expansions. The specific model we consider is an  $S = \frac{1}{2}$  Ising model on a simple-cubic (sc) lattice with lattice anisotropy and in the presence of an external field. The Hamiltonian is

$$\mathcal{H} = -J_{xy} \sum_{\langle ij \rangle}^{xy} s_i s_j - J_z \sum_{\langle ij \rangle}^z s_i s_j - \mu H \sum_i s_i, \quad (1)$$

where  $s = \pm 1$ , the first sum is over all nearest neighbor spins in the  $x$ - $y$  plane and the second is over nearest neighbors coupled in the  $z$  direction. To simulate a metamagnet, we take  $J_{xy} > 0$  (ferromagnetic), and  $J_z < 0$  (antiferromagnetic). High-temperature series to eighth order in inverse temperature were generated for the two-spin correlation functions  $C_2(\vec{r}) \equiv \langle s_0 s_{\vec{r}} \rangle - \langle s_0 \rangle \langle s_{\vec{r}} \rangle$  using a computer program based upon the renormalized

linked-cluster theory of Wortis, Jasnow, and Moore.<sup>16</sup> From the correlation functions, we calculate series for the reduced susceptibility,

$$\bar{\chi} = \sum_{\vec{r}} C_2(\vec{r}), \quad (2)$$

and reduced *staggered* susceptibility,

$$\bar{\chi}_{st} = \sum_{\vec{r}} \eta_{\vec{r}} C_2(\vec{r}). \quad (3)$$

Here  $\eta_{\vec{r}}$  is a staggering index which is +1 for lattice sites on even numbered planes and -1 on odd planes.

The coefficients of successive powers of  $\beta \equiv 1/k_B T$  for the spin- $\frac{1}{2}$  Ising model are finite polynomials in the variable  $\tanh^2(\beta\mu H)$ , so fixing  $h \equiv \beta\mu H$  and evaluating these polynomials enables one to obtain exact information in the external field. Thus  $\bar{\chi}$  and  $\bar{\chi}_{st}$  are of the form

$$\sum_{n=0}^{\infty} P_n(\tanh^2 h) \beta^n,$$

where  $P_n$  is a polynomial of degree  $n+1$  in  $\tanh^2 h$ . In Table I we list the coefficients  $a_n$  and  $b_n$  through

TABLE I. Coefficients  $a_n$  and  $b_n$  in the series (4) and (5) for the susceptibility and staggered susceptibility, respectively. Here the expansion variable  $X$  denotes  $\tanh^2 h \equiv \tanh^2(\mu H/k_B T)$ . Single asterisk denotes uncertainty in last digit. Double asterisk denotes uncertainty in last two digits.

$a_0 = 1 - X$	$b_0 = 1 - X$
$a_1 = 2 - 8X + 6X^2$	$b_1 = 6 - 16X + 10X^2$
$a_2 = -2 + 2X + 10X^2 - 10X^3$	$b_2 = 30 - 134X + 186X^2 - 82X^3$
$a_3 = -14 + 200X - 566X^2 + 576X^3 - 196X^4$	$b_3 = 150 - 880X + 1894X^2 - 1760X^3 + 596X^4$
$a_4 = -42 + 1026X - 5144X^2 + 10040X^3 - 8526X^4 + 2646X^5$	$b_4 = 726 - 5150X + 14600X^2 - 20536X^3 + 14258X^4 - 3898X^5$
$a_5 = -46 + 1960X - 16814X^2 + 56256X^3 - 88048X^4 + 65040X^5 - 18348X^6$	$b_5 = 3510 - 28848X + 99166X^2 - 182208X^3 + 188208X^4 - 103296X^5 + 23468X^6$
$a_6 = -90 - 1014X + 10434X^2 - 594X^3 - 105900X^4 + 237436X^5 - 200228X^6 + 59956X^7$	$b_6 = 16710 - 157342X + 635378X^2 - 1426906X^3 + 1924948X^4 - 1559284X^5 + 701564X^6 - 135068X^7$
$a_7 = 2 - 19304X + 320862X^2 - 1757952X^3 + 4597716X^4 - 6479120X^5 + 5015212X^6 - 1984256X^7 + 306840X^8$	$b_7 = 79494 - 843728X + 3932594X^2 - 10493024X^3 + 17509596X^4 - 18699904X^5 + 12478036X^6 - 4755072X^7 + 792008X^8$
$a_8 = -1402 - 28478X + 1155680X^2 - 10011488X^3 + 40107912X^4 - 88872520X^5 + 115745632X^6 - 88314336X^7 + 36561936X^8 - 6342936X^9$	$b_8 = 375174 - 4445438X + 23579488X^2 - 73392864X^3 + 147460232X^4 - 197953224X^5 + 177251104X^6 - 101949760X^7 + 34145680X^8 - 5070392X^9$

order  $n=8$  in the susceptibility series and the staggered susceptibility series, respectively,

$$\bar{\chi} = k_B T \chi / N \mu^2 = \sum_{n=0}^{\infty} a_n \tanh^n \beta, \quad (4)$$

$$\bar{\chi}_{st} = k_B T \chi_{st} / N \mu^2 = \sum_{n=0}^{\infty} b_n \tanh^n \beta, \quad (5)$$

where we have changed the expansion variable from  $\beta$  to  $\tanh \beta$ . To the best of our knowledge these are the first high-temperature series expansions for inequivalent bonds ( $J_{xy} \neq J_z$ ) in a magnetic field. Previous Ising-model high-temperature series have been for *isotropic* lattices in a finite field<sup>17,18</sup> or for anisotropic lattices in *zero* field.<sup>19</sup>

(i) *Location of critical line and staggered susceptibility.*—The strongly divergent quantity along the second-order line separating the anti-ferromagnetic and paramagnetic phases is the staggered susceptibility. Accordingly, the  $\chi_{st}$  series was evaluated for an extremely wide range of values of the parameter  $h = \beta \mu H$  and series analysis was carried out by Padé approximants and other methods. It was found that convergence improved after a bilinear transformation<sup>20</sup> was carried out on the original high-temperature expansion variable.

For all but the largest magnetic fields (specifically, for  $\mu H_c \lesssim 1.7$ ) we found the exponent describing the divergence of  $\chi_{st}$  to be very nearly 1.25, thereby corroborating the prediction of the universality hypothesis,<sup>6</sup>

$$\chi_{st} \sim [T - T_c(H)]^{-5/4}. \quad (6)$$

In order to obtain a better estimate of the location of the critical line, we assumed the validity of (6) for those larger fields for which the series were less regular. The resulting phase boundary is shown as the solid curve in Fig. 1, with precision generally better than the size of the points, and with best precision at small fields.

(ii) *Location of tricritical point and tricritical susceptibility exponent.*—The hooking around of our calculated phase line below  $k_B T_c \approx 2.60$  (cf. dashed curve in Fig. 1) is clearly unphysical and represents the failure of high-temperature series where the transition becomes first order. Simple energy arguments give the  $T=0$  critical field to be  $\mu H_c = q_z |J|$  ( $=2$  for the present problem), where  $q_z$  is the coordination number in the  $z$  direction. A curve from this point joins the second-order line smoothly at  $k_B T \approx 2.60 \pm 0.05$ , which is the region wherein we identify the tricritical point.

The direct susceptibility series are quite irregular and do not lend themselves to the usual methods of analysis. Padé approximants to  $\log \chi$  were fairly inconclusive, but more convincing evidence was obtained by raising the original series to various powers, and doing a full eighth-order Padé analysis on the resulting series. It was expected that the Padé approximants for the correct exponent at the tricritical point would converge well and reproduce the poles given by the  $\chi_{st}$  series along the same  $\mu H/k_B T$  path.<sup>21</sup> Accordingly, we raised the  $\chi$  series to a wide variety of powers, and found best consistency with a power of 2. In particular, the roots from the  $\chi^2$  series matched to within 1% the roots arising from the  $\chi_{st}$  series

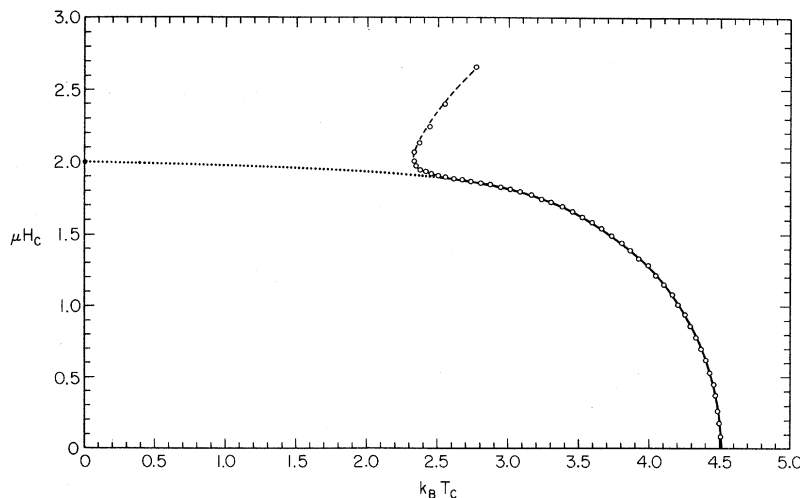


FIG. 1. Phase boundary for Ising-model metamagnet on sc lattice with in-plane interaction  $J_{xy} = +1$ , between-plane interaction  $J_z = -1$ . The second-order portion of the phase line is shown solid, the first-order portion is shown dotted, while the spurious hooking near the tricritical point is shown dashed.

analysis, while powers other than 2 led to roots which disagreed with the  $\chi_{st}$  series. Moreover, series convergence was best for  $\chi^2$ . Therefore we are led to conjecture that along the path  $\mu H/k_B T = \mu H_t/k_B T_t \cong 0.72$ ,

$$\chi \sim (T - T_t)^{-1/2}. \quad (7)$$

We briefly compare our results with certain predictions of mean-field (MF) theory.<sup>9</sup> Mean-field theory predicts that the phase boundary near the Néel temperature  $T_N$  is described by a square law,

$$(\mu H)^2 = Ak_B [T_N - T_c(H)], \quad (8)$$

with amplitude  $A^{MF} = 3.55$ . We find that the shape of the phase boundary predicted by series extrapolations is also described by (8) for  $\mu H_c \lesssim 0.8$ , with  $A^{\text{series}} = 3.57 \pm 0.05$ . Of course, the absolute values of  $T_N$  differ, with  $T_N^{MF} = 6$ , and  $T_N^{\text{series}} \cong 4.51$ . Moreover, despite the similar shape of the phase boundaries at small fields, we find the ratio of tricritical to Néel temperature to be considerably smaller than the prediction of mean field theory,

$$(T_t/T_N)^{\text{series}} = 0.58 \pm 0.01 \ll (T_t/T_N)^{MF} = \frac{5}{6}. \quad (9)$$

Finally, MF theory predicts a finite susceptibility as  $T \rightarrow T_t$  from above, in contrast to (7).

One would naturally wish to have a gauge for just how much confidence one should place in results of series expansions, especially when they are applied to a new situation such as the metamagnet (for which *no* other "nonclassical" calculations have been performed). To this end, we also applied the *same* methods to an "nnn" model with nearest-neighbor antiferromagnetic and next-nearest-neighbor ferromagnetic interactions.<sup>22</sup>

This model also exhibits tricritical behavior, and very recent Monte Carlo calculations<sup>10</sup> give a tricritical susceptibility exponent of  $0.29 \pm 0.18$ . Our high-temperature series give  $\gamma_t \cong \frac{1}{4}$ , in agreement with<sup>23</sup> the Monte Carlo work and *different* from the exponent for the metamagnet model. Otherwise, we find the same qualitative features for the nnn model as for the metamagnet. This fact, together with the fact that we find quantitative agreement with Monte Carlo calculations on the nnn model, leads us to believe that the methods used on the metamagnet are worthwhile.

We wish to thank Dr. Gerald Paul for having interested us in this problem and for kindly assisting us in the initial phases of this work. We gratefully acknowledge valuable conversations and correspondence with Professor Michael Wor-

tis. Professor David P. Landau kindly confirmed certain of our predictions, including our location of the tricritical point, using his Monte Carlo method on an  $8 \times 8 \times 8$  lattice. We also wish to thank David Lambeth, Douglas Karo, Richard Krasnow, and Luke Lui for numerous helpful discussions. Further impetus for this work has come from Dr. Robert Birgeneau and Dr. Gen Shirane, who have just initiated a study of tricritical points in metamagnets using inelastic neutron scattering.

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<sup>1</sup>R. B. Griffiths, Phys. Rev. Lett. **24**, 715 (1970).

<sup>2</sup>R. B. Griffiths, in *Critical Phenomena in Alloys, Magnets, and Superconductors*, edited by R. E. Mills, E. Ascher, and R. I. Jaffee (McGraw-Hill, New York, 1971), pp. 377-391.

<sup>3</sup>I. S. Jacobs and P. E. Lawrence, Phys. Rev. **164**, 866 (1967); R. Birgeneau and G. Shirane, private communication; see also the  $H=0$  work of W. B. Yelon and R. J. Birgeneau, Phys. Rev. B **5**, 2615 (1972).

<sup>4</sup>V. A. Schmidt and S. A. Friedberg, Phys. Rev. B **1**, 2250 (1970).

<sup>5</sup>D. P. Landau, B. E. Keen, B. Schneider, and W. P. Wolf, Phys. Rev. B **3**, 2310 (1971).

<sup>6</sup>R. B. Griffiths, Phys. Rev. Lett. **24**, 1479 (1970). See also L. P. Kadanoff, in Proceedings of the Varenna Summer School, Varenna, Italy (to be published).

<sup>7</sup>See also R. B. Griffiths and J. C. Wheeler, Phys. Rev. A **2**, 1047 (1970). Their postulates suggest a possible change of exponents at  $T_t$  from geometrical considerations of the coexistence surfaces in the phase diagram.

<sup>8</sup>L. Landau, Phys. Z. Sowjetunion **11**, 26 (1937), reprinted in *Collected Papers of L. D. Landau*, edited by D. ter Haar (Pergamon, New York, 1965), p. 193.

<sup>9</sup>R. Bidaux, P. Carrara, and B. Vivet, J. Phys. Chem. Solids **28**, 2453 (1967).

<sup>10</sup>D. P. Landau, Phys. Rev. Lett. **28**, 449 (1972).

<sup>11</sup>E. K. Riedel, Phys. Rev. Lett. **28**, 675 (1972).

<sup>12</sup>G. Goellner and H. Meyer, Phys. Rev. Lett. **26**, 1534 (1971).

<sup>13</sup>M. Blume, V. J. Emery, and R. B. Griffiths, Phys. Rev. A **4**, 1071 (1971). See also M. Blume, Phys. Rev. **141**, 517 (1966); H. W. Capel, Physica (Utrecht) **32**, 966 (1966).

<sup>14</sup>J. Oitmaa, J. Phys. C: Proc. Phys. Soc., London **4**, 2466 (1971).

<sup>15</sup>D. M. Saul and M. Wortis, *Magnetism and Magnetic Materials—1971, AIP Conference Proceedings No. 5* (American Institute of Physics, New York, 1972).

<sup>16</sup>M. Wortis, D. Jasnow, and M. A. Moore, Phys.

Rev. **185**, 805 (1969); see also the review by M. W. Wortis, in *Phase Transitions*, edited by C. Domb and M. S. Green (Academic, London, in press).

<sup>17</sup>A. Bienenstock and J. Lewis, Phys. Rev. **160**, 393 (1967); D. C. Rapaport and C. Domb, J. Phys. C: Proc. Phys. Soc., London **4**, 2684 (1971).

<sup>18</sup>D. Gaunt and G. A. Baker, Jr., Phys. Rev. B **1**, 1184 (1971); see also the very recent work of M. Ferer and M. Wortis, to be published, and references contained therein.

<sup>19</sup>G. Paul and H. E. Stanley, Phys. Lett. **37A**, 427 (1971), and Phys. Rev. B **5**, 2578 (1972); J. Oitmaa and I. G. Enting, Phys. Lett. **36A**, 91 (1971), and J. Phys. C: Proc. Phys. Soc., London **5**, 231 (1972).

<sup>20</sup>D. D. Betts, C. J. Elliott, and R. V. Ditzian, Can. J. Phys. **49**, 1327 (1971); M. H. Lee and H. E. Stanley, Phys. Rev. B **4**, 1613 (1971), and references contained

therein. M. Wortis, unpublished.

<sup>21</sup>The exponent obtained approaching the tricritical point along an  $H/T = \text{const}$  path should be the same as that along an  $H = \text{const}$  path provided the phase boundary is not parallel to the  $T$  axis at  $T_t$ .

<sup>22</sup>Previous Ising-model high-temperature series have been for *nearest-neighbor* antiferromagnets in a finite field (Ref. 17) or for *next-nearest-neighbor* antiferromagnets in *zero* field [G. Paul and H. E. Stanley, Phys. Rev. B **5**, 3715 (1972), and references contained therein].

<sup>23</sup>It should be noted that the Monte Carlo exponent is estimated along a constant magnetization path which need not ordinarily be expected to give the same exponent as obtained from a constant field path. We wish to thank Professor R. B. Griffiths for very helpful discussions concerning this point. See also Ref. 7.

## Vacancy-Impurity Binding Energy in Aluminum—1.7 at. % Zinc Using Positron-Annihilation Lifetimes\*

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Positron-annihilation lifetimes were measured in pure aluminum and in aluminum—1.7 at. % zinc at temperatures between 20 and 550°C. The data were analyzed using an extension of the trapping model to obtain the vacancy formation energy in aluminum ( $E_F = 0.62 \pm 0.02$  eV). Under certain restrictive assumptions the binding energy of vacancies to zinc atoms in aluminum was found to be  $E_B = 0.019 \pm 0.004$  eV. A relaxation of these assumptions yielded only an upper bound ( $E_B < 0.04$  eV).

A complete understanding of defects in metals requires knowledge of the sign and magnitude of the interaction between vacancies and impurities. Until recently it has been generally assumed that vacancies and impurities are bound to each other, but the experimental values of the binding energy in a given alloy can vary from 0.0 to 0.5 eV. There has been a growing feeling that the larger values are incorrect because they were often obtained by quenching techniques where vacancy and impurity clustering effects could lead to higher apparent values for the binding energy. On the other hand, the only equilibrium measurements to date have been performed at high temperatures where vacancy clustering causes difficulties.<sup>1,2</sup>

Positron-annihilation methods eliminate the problems mentioned above because they are performed in equilibrium at temperatures where the vacancy concentration is much lower. Positrons have been shown to be quite sensitive to vacancy-type defects in metals,<sup>3-6</sup> and details of the annihilation process have been used recently to deduce

the vacancy formation energy in aluminum ( $E_F = 0.66 \pm 0.04$  eV) using 2- $\gamma$  angular-correlation methods.<sup>7</sup>

In this Letter we report the use of positron-annihilation lifetimes to measure the binding energy of vacancies to zinc atoms in aluminum. We show for the first time that it is possible to extend the trapping model to include the effects caused by the presence of vacancy-impurity complexes. As a consequence of this inclusion, the vacancy-impurity binding energy is extracted from the analysis. Because of the low concentration of vacancies at the temperatures employed, analysis of the data also has been extended to include the contribution to positron trapping that arises from dislocations.

Standard delayed-coincidence lifetime measurements were made using  $\frac{3}{4}$ -in. cylinders of KL-236 plastic scintillator,<sup>8</sup> RCA 8575 photomultipliers, and integrated-circuit constant-fraction discriminators.<sup>9</sup> Instrumental resolution was typically 0.240 nsec full width at half-maximum for a Co<sup>60</sup> source with energy windows set at