

^{129}I magnetic hyperfine structure in the Heusler alloys Pd_2MnSb and PdMnSb

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(Received 17 January 1978)

Hyperfine fields at ^{129}I sites in the Heusler alloys Pd_2MnSb and PdMnSb have been measured and are shown to form part of a systematic trend of fields observed at $5sp$ elements. Theoretical estimates of these fields based on a homogeneous spin-polarized electron-gas model are compared to the observed field systematics.

I. INTRODUCTION

We report results of Mössbauer-effect (ME) experiments for ^{129}I magnetic hyperfine structure in the Heusler alloys (HA) Pd_2MnSb ($X_2\text{MnY}$) and PdMnSb (XMnY). Combined with measurements of fields at other $5sp$ elements¹⁻⁷ substituted at the Y site in these hosts, the present work provides the most extensive study of hyperfine-field systematics in any HA to date. The experimental results are analyzed using a homogeneous spin-polarized electron-gas model. In this paper, we show that the principal features of the field systematics are reproduced remarkably well by the present model, and thus we suggest that conduction-electron polarization (CEP) is the dominant mechanism for hyperfine fields in HA. In contrast to the field systematics between PdMnSb and Pd_2MnSb , the experiments reveal a salient role of Pd in modifying the spin polarization transferred to the Y site. The present calculations fail to reproduce this feature of the experiments quantitatively, and we suggest that this is probably due to a band-structure effect in Pd_2MnSb . The experiments also exhibit profound heat-treatment effects in Pd_2MnSb , and these are indicated to be consequences of a vacancy-associated defect trapped during a thermal quench of the material.

The Cd,¹ Sn,^{2,3} Sb,^{4,5} and Te^{6,7} hyperfine fields in Pd_2MnSb and PdMnSb have recently been measured and have revealed a monotonic increase with valence of the sp element. This trend of fields can be understood in terms of CEP effect as developed in two theoretical models, one due to Blandin and Campbell⁸ (BC) and the other due to Jena and Geldart⁹ (JG). In the Te-I-Xe region, both these models predict fields to saturate and then decrease in value. In the present work, we have focused on I-field measurements to see if this predicted behavior is observed.

The experimental procedure used in the present Mössbauer measurements and an interpretation of

the data appear in Secs. II and III. Section IV summarizes the trends of hyperfine fields at $5sp$ elements in the HA's PdMnSb and Pd_2MnSb . We have analyzed these field systematics using the model of JG. In that section we also provide a critical comparison of some theoretical models that have been invoked to explain hyperfine fields in HA's. Section V gives a summary of conclusions arising from the present work.

II. EXPERIMENTAL PROCEDURE

Sources of $\text{Pd}_2\text{MnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ and $\text{PdMnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ were prepared by induction melting neutron-activated $^{124,128}\text{Te}$ with 99.99%-pure Pd and Mn, and 99.9999%-pure Sb crystal chips in a partial pressure of Ar as follows: (a) Melts were taken to 1500 °C for 2 min, kept at 1400 °C for 10 min, and then allowed to cool slowly (15 min) to room temperature. (b) The ingots were powdered and sealed in evacuated quartz tubings which were back filled with a partial pressure (10 Torr) of He exchange gas. The source materials were kept at 1000 °C for 24 h and then quenched in water. (c) A final step consisted of vacuum annealing the sources at 300 °C for 16 h. Sources of $\text{PdMnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ were prepared following steps (a)-(c) outlined above, except that the quoted temperatures were reduced by 150 °C. Spectra of the sources were recorded at 4.2 °K after indicated heat-treatment steps and the results appear in Figs. 1 and 2. The spectra were fitted by a pure magnetic interaction $E_m = -g\mu_N H_{\text{eff}} m$, taking the ratio g_e/g_0 to be⁶ $-0.2270(15)$ for ^{129}Te and¹⁰ $1.496(5)$ for ^{129}I .

III. RESULTS AND INTERPRETATION

Incorporating Te as an impurity in Pd_2MnSb by melting the elements leads to its occupancy at the Sb site in the $L2_1$ structure of the host. The evidence is based on the identity of the ^{129}Te magnetic interaction in Pd_2MnSb , using Mössbauer parents

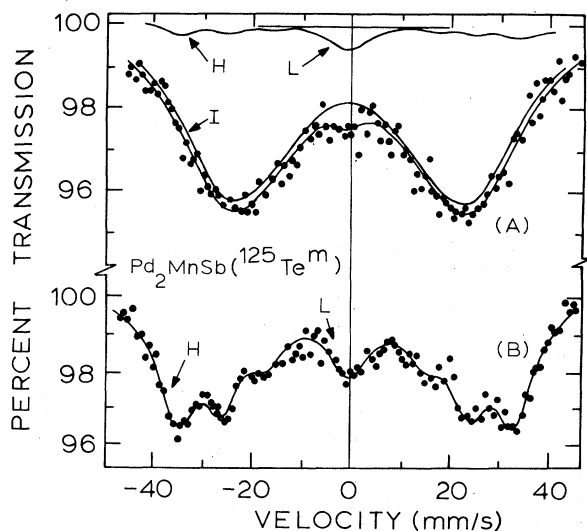


FIG. 1. Spectra of $\text{Pd}_2\text{MnSb}({}^{125}\text{Te}^m)$ source taken after (A) heat-treatment (b) and (B) heat-treatment (c). The top spectrum shows a fit to three sites: a high (*H*), intermediate (*I*), and low (*L*) field site of 856(9), 680(25), and ~ 0 kOe, with intensity ratio 5:90:5, respectively. ${}^{125}\text{TeZn}$ 12-mg/cm² thick was used as an absorber in the measurements.

of ${}^{125}\text{Sb}$ and ${}^{125}\text{Te}^m$. The magnetic field at a dilute Te impurity substituting at the Sb site was established to be 857 ± 9 kOe,⁶ using ${}^{125}\text{Sb}$ sources diffused in Pd_2MnSb platelets. Figure 1(B) shows a spectrum of a $\text{Pd}_2\text{MnSb}_{0.995}({}^{125}\text{Te}_{0.005}^m)$ source taken following heat treatment (c). The Te field of 856 ± 9 kOe resulting from the six-line spectrum agrees almost identically with our previous measurement⁶ using an ${}^{125}\text{Sb}$ source. We take this identity of fields to indicate that Te atoms in a $\text{Pd}_2\text{MnSb}_{0.995}({}^{125}\text{Te}_{0.005}^m)$ source following heat treatment (c) are present substitutionally at the Sb site in the host structure.

${}^{129}\text{I}$ magnetic structure observed in a $\text{Pd}_2\text{MnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ source following heat treatment (c) [Fig. 2(C)], in analogy to the ${}^{125}\text{Te}$ experiments mentioned above, must likewise be attributed to a ${}^{129}\text{Te}$ parent substitutional at Sb. It follows that the field of 502 ± 10 kOe deduced from this spectrum represents the field at I substitutional at Sb in Pd_2MnSb . Support for this identification arises independently from the trend of isomer shifts (IS), and we discuss this point below.

The IS of various ${}^{129}\text{I}$ sites observed in present HA experiments along with the shifts in some reference hosts are projected on a line plot in Fig. 3. An *sp* element substitutional at Sb in Pd_2MnSb sits at the bcc position of a Pd cube (inset of Fig. 4) and its IS in this host may be compared to that

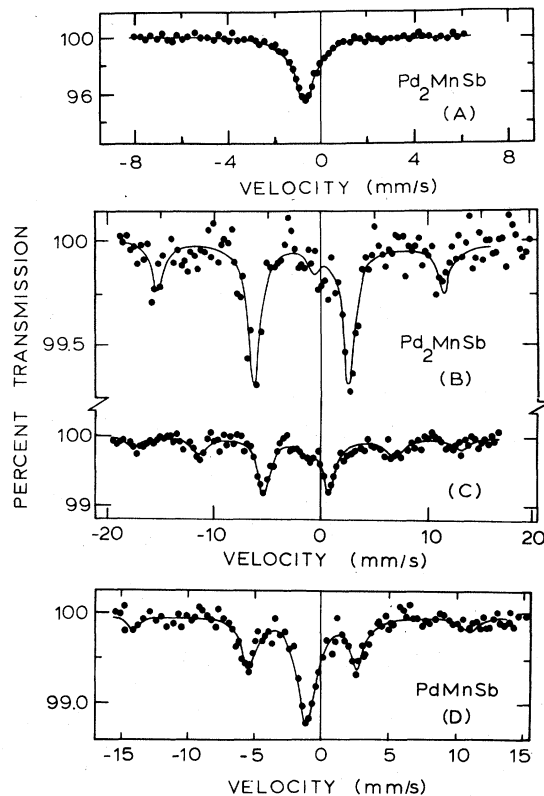


FIG. 2. Spectra of $\text{Pd}_2\text{MnSb}({}^{129}\text{Te}^m)$ source taken after (A) heat treatment (a), (B) heat treatment (b), and (C) heat treatment (c). (D) Spectrum of $\text{PdMnSb}({}^{129}\text{Te}^m)$ source taken after heat treatment (b). ${}^{129}\text{Ina}$ 12-mg/cm² thick was used as an absorber in the measurements.

in fcc Pd metal. ${}^{129}\text{I}$ IS of the substitutional site in Pd_2MnSb is more positive than that in Pd metal (see Fig. 3). Since $\Delta\langle r^2 \rangle / \langle r^2 \rangle$ for ${}^{129}\text{I}$ is known¹¹ to be positive, one concludes that the electron density at ${}^{129}\text{I}$ in Pd_2MnSb is larger than in the Pd metal. Observed ${}^{121}\text{Sb}$ IS in Pd_2MnSb ,^{4,5} relative to Pd metal, also show that the electron density at ${}^{121}\text{Sb}$ in Pd_2MnSb is larger than in Pd metal. Finally, we would like to point out that recent measurement³ of Sn impurity in Pd_2MnSb also shows that the electron density in Pd_2MnSb is larger than in Pd metal. From the observed IS's and known $\Delta\langle r^2 \rangle / \langle r^2 \rangle$ values, we have calculated $\Delta|\psi(0)|^2$ between hosts of Pd_2MnSb and Pd metal for the three elements, and find $\Delta|\psi(0)|^2_{129\text{I}} : \Delta|\psi(0)|^2_{121\text{Sb}} : \Delta|\psi(0)|^2_{119\text{Sn}} = 0.74(19) : 0.84(13) : 1.00$. There thus exists a pattern to the electron densities, namely, the presence of a systematically larger contact electron density at a 5*sp* element in Pd_2MnSb relative to Pd metal. We believe this pattern is most likely a manifestation of two com-

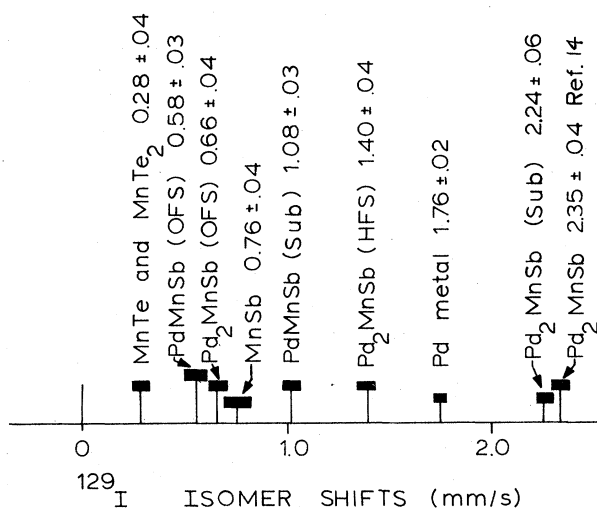


FIG. 3. ^{129}I IS of various sites observed in Pd_2MnSb and PdMnSb are shown in the form of a line plot. The energy of the γ ray increases to the right. OFS, zero-field site; HFS, high-field site; Sub, substitutional. The IS in Pd metal is taken from Ref. 14. All IS are quoted relative to Na^{129}I .

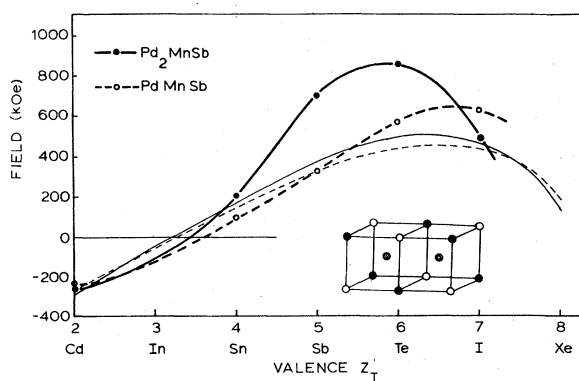


FIG. 4. Hyperfine fields at $5sp$ elements in indicated hosts plot as a function of element valence Z_I . Experimental fields (bold lines) are taken from Refs. 1-7. Theoretical fields (thin lines) are predictions based on JG model. In the crystal structure of Pd_2MnSb , open and filled circles represent Pd sites. For PdMnSb , filled circles represent Pd sites, while open circles represent Pd vacancies.

peting effects: a compression effect¹² of the local valence-charge density induced by confining the $5sp$ guest atom to a smaller atomic volume,¹³ and a second more prominent effect due to an increased conduction-electron concentration in the HA host relative to Pd metal.

The profound role of heat treatment on spectra of Pd_2MnSb sources may be elaborated. Following heat treatment (a), spectra of $\text{Pd}_2\text{MnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ sources showed narrow single lines [see Fig. 2(A) and Table I], a result which is due to the apparent small Te solubility in Pd_2MnSb at room temperature. Following heat-treatment (b), spectra of $\text{Pd}_2\text{MnSb}_{0.995}({}^{125}\text{Te}_{0.005}^m)$ [Fig. 1(A)] and $\text{Pd}_2\text{MnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ [Fig. 2(B)] sources exhibited magnetic splitting. Heat treatment (b) clearly permits trapping of the dissolved Te fraction at 1000°C by a thermal quench of the source material to room temperature. We suggest that rapidly cooled Pd_2MnSb samples possess a defective structure, and that the magnetic structures observed following heat treatment (b) be identified with defect ^{125}Te and ^{129}I sites. In our view, first-order changes in IS and H_{eff} (Table I) following heat treatment (c) result from detrapping of a Pd multiple-vacancy defect from the immediate environment of the resonant isotope. Spectra of Figs. 1(A) and 2(B) are both indicative of dominant ^{125}Te and ^{129}I sites, which are characterized by fields of 680 ± 25 and 729 ± 15 kOe, respectively. For Te impurity ($Z_I=6$) in Pd_2MnSb we note that H_{eff} at the defect site is smaller than at the substitutional site, while for I impurity ($Z_I=7$) in this host just the opposite result is observed. This trend of fields at the defect site in Pd_2MnSb is, in fact, closely similar to the one observed at the substitutional site in PdMnSb . We show later that

TABLE I. Summary of ^{125}Te and ^{129}I Mössbauer-effect parameters (hyperfine field H_{eff} , isomer shift δ) on indicated sources. Isomer shifts are quoted relative to $^{125}\text{TeZn}$ or $^{129}\text{I}^m\text{Na}$.

| Source | Heat treatment | H_{eff} (kOe) | δ (mm/sec) |
|---|----------------|------------------------|-------------------|
| $\text{Pd}_2\text{MnSb}({}^{129}\text{Te}^m)$ | (a) | 0 | 0.66(4) |
| | (b) | $729(10)^a$ | 1.40(10) |
| | (c) | 502(10) | 2.24(6) |
| | Ref. 14 | 513(6) | 2.35(4) |
| $\text{Pd}_2\text{MnSb}({}^{125}\text{Te}^m)$ | (b) | $680(25)^a$ | 0.06(10) |
| | (c) | 856(9) | 0.03(9) |
| $\text{PdMnSb}({}^{129}\text{Te}^m)$ | (b) | 660(10) | 1.05(10) |
| | (c) | 681(6) | 1.08(6) |

^a Dominant site in the spectrum.

these general results are manifestations of a reduced electron-spin density at the sites in question relative to the substitutional site in Pd_2MnSb . Finally, we would like to point out that the heat-treatment effects in Pd_2MnSb reported here are not peculiar to ^{125}Te and ^{129}I Mössbauer spectroscopy alone, and, in fact, closely resemble heat-treatment effects on this HA observed using ^{119}Sn (Ref. 3) and ^{121}Sb Mössbauer spectroscopy. Taken together, all these heat-treatment effects on Pd_2MnSb represent, to our knowledge, one of the first few instances when the presence of a local defect trapped near the guest atom modified the magnetic hyperfine interaction in first order. We discuss these defect aspects of Pd_2MnSb in a forthcoming paper.

Figure 2(D) shows the spectrum of a $\text{PdMnSb}_{0.995}({}^{129}\text{Te}_{0.005}^m)$ source taken after heat treatment (b) along with a fit of the data in terms of two ^{129}I sites; a high-field site of 660 ± 20 kOe and a low-field site of ~ 0 kOe. Spectra of PdMnSb sources taken following heat treatment (c) yielded results similar to those obtained after heat treatment (b). From these spectra, we identify the high-field site (681 ± 6 kOe) with $^{129}\text{Te}^m$ (^{129}I) substitutional at Sb and the zero-field site with undissolved $^{129}\text{Te}^m$ in PdMnSb . We note the lack of annealing effects and a smaller Te solubility displayed in PdMnSb . Both these characteristics of PdMnSb sources are in sharp contrast to those of Pd_2MnSb sources and, we believe, are probably due to a more compact lattice of PdMnSb .

IV. SYSTEMATICS OF HYPERFINE FIELDS IN PdMnSb AND Pd_2MnSb

The present magnetic field at ^{129}I substitutional in Pd_2MnSb is in agreement with a recent report by deWaard *et al.*,¹⁴ who have observed a high-field site of 514 ± 6 kOe (Table I) in samples of Pd_2MnSb prepared by sintering the elements. The IS's of the two sites in question also agree with each other very well (see Fig. 3). In Fig. 4, we have projected the present ^{129}I field results along with fields observed at other $5sp$ elements. We have assumed a positive sign for the I fields as suggested by the systematics.

We analyze these hyperfine-field systematics using theoretical models developed recently by several authors. In the model proposed by BC,⁸ the HA is regarded as a nonmagnetic host containing two kinds of impurities—magnetic Mn atoms and nonmagnetic (X, Y) atoms, where hyperfine fields are being measured. The scattering of electrons from these two types of impurities embedded

in a homogeneous electron gas is then calculated, and the hyperfine field at the nonmagnetic atom site determined. The model neglects interference effects between different impurities (in particular the magnetic ions), neglects the effect of spin splitting of the conduction band through s - d exchange interaction, and further uses asymptotic form for the electron-spin density even for the first near neighbors. It should be pointed out that the interaction¹⁵ between magnetic ions and the deviation of the electron charge and spin distribution from the asymptotic form¹⁶ for near-neighbor distances are large. In a recent paper Campbell and Blandin¹⁷ made a phenomenological adjustment to account for the preasymptotic behavior of spin-density oscillation, although its fundamental justification appears to us to be lacking. Jena and Geldart¹⁸ have discussed these points and the reader is referred to their paper for details.

Jena and Geldart⁹ (JG) have proposed an alternate model, based on a generalization of the model of Daniel and Friedel, to interpret the hyperfine-field systematics at nonmagnetic ion sites in HA. It is believed that a basic understanding of hyperfine fields at nonmagnetic ion sites in ferromagnets requires detailed treatment of the nonmagnetic ion's electronic structure in a spin-polarized background. The HA is regarded as a ferromagnetic host, where the interaction between the magnetic ions splits the conduction band of the alloy into a majority- and minority-spin band. The electron-spin density distribution around a given magnetic ion is inhomogeneous and exhibits the Ruderman-Kittel-Kasuya-Yosida (RKKY) oscillation. In order to calculate the perturbation of the host electron-spin density due to the presence of a nonmagnetic ion (where hyperfine field is measured), JG make use of the local-density approximation. In this approximation electrons within the volume element of a nonmagnetic atom respond to the total field, acting as if these were part of an infinite homogeneous electron gas of the same density. The model thus includes inhomogeneity of the ambient electron charge and spin density of the host. It assumes that the electron response to a perturbation is determined by the unperturbed (ambient) electron charge and spin density at the impurity site. This information clearly depends on the location of the impurity in the lattice.

In the absence of adequate experimental information, JG treat the ambient spin polarization by a band-splitting parameter Δ . The electron-spin density n_0 of the "homogeneous" medium is determined by assuming that each atom in the HA contributes a certain number of sp electrons to the conduction band. In the present calculation we have taken Pd to contribute 1.3 sp electrons¹⁹ to the con-

duction band. The contribution of the nonmagnetic Sb ion to the conduction electron density was determined by its number of valence electrons, i.e., 5 in this case. The contribution from the magnetic ion was determined from a knowledge of the local magnetic moment. The potential of the nonmagnetic ion was approximated by a square well of range a and depth V_0 . The range a is assumed to be the Wigner-Seitz radius of the nonmagnetic ion in its pure state, since in a metallic environment the ion is screened within the Wigner-Seitz cell. The depth V_0 is determined self-consistently by satisfying the Friedel sum rule. The conduction-band spin splitting is determined once and for all by normalizing the calculated field at the Sb site in PdMnSb with experiment. For Pd₂MnSb, the band splitting Δ was determined by scaling it linearly with the host magnetic moment. We then calculated the hyperfine fields at the $5sp$ element site by using the expression $H_{\text{eff}} = -\frac{2}{3}\pi\mu_B\alpha^2P(0)$, where $P(0)$ is the spin-polarization density transferred to the Y -site element, and α^2 is the Bloch enhancement factor determined by orthogonalizing the conduction-electron wave functions at the Fermi surface to the occupied atomic core orbitals. The results of these calculations appear in Fig. 4 as the thin continuous line (Pd₂MnSb) and the thin dashed line (PdMnSb).

The present measurements on I bring out a maximum in the fields near $Z_I = 6$ for Pd₂MnSb and beyond $Z_I = 6$ for PdMnSb. This feature along with the observed shape of the field systematics for both HA's, which leads to a crossing of the two curves near I, are prominent features of the data that is clearly reproduced by the present calculations. These are strongly suggestive that CEP effects are dominant in HA. In the JG model, following the spirit of the Daniel-Friedel model, one may understand the shape of the systematics in terms of field contributions arising from scattering and bound states in the impurity square-well potential. The crossover from negative to positive fields at $Z_I = 4$ and the saturation of fields near $Z_I = 6$ are the results of two competing contributions,⁹ one coming from scattering states, dominant at low Z_I , and the other from bound states which become increasingly important at high Z_I . In the theoretically calculated curves shown in Fig. 4, we note that the increased amplitude and frequency of the curve for Pd₂MnSb relative to PdMnSb lead to a crossing of the two curves near I. This result is due to an increase of k_F in Pd₂MnSb relative to PdMnSb,^{8,9} and is caused by an increase in Pd concentration.

The lack of a more quantitative agreement between the observed and calculated Pd₂MnSb fields raises interesting questions concerning the nature

of approximations made in the present theory and the role of other possible mechanisms for the origin of hyperfine fields such as core-overlap contribution. Aside from an indirect CEP mechanism, a direct overlap exchange polarization mechanism has been invoked to account for the existence of large positive fields observed at $5sp$ elements in metallic Fe. For HA's, any direct overlap of the spin-polarized d orbitals localized on Mn with the Y -site s orbitals is believed to be small, since the Mn sites happen to be second near neighbors of the Y site in the HA structure. We believe that the close similarity in the shape of the predicted and observed field systematics for Pd₂MnSb makes it plausible that the basic CEP mechanism invoked for HA is appropriate, but that the present calculations involve approximations that may in fact be quite poor for this host. The JG and BC models both assume a free-electron-like conduction band for these HA's. It appears to us that there exists a sharp density of d -like states of Pd at E_F (Fermi level), and that the addition of Pd to PdMnSb increases E_F and the consequent density of states at E_F . This then leads to a strong dependence of H_{eff} on Pd concentration. The significant change in H_{eff} in going from the defect to the substitutional site in Pd₂MnSb is an interesting point. It suggests that the Pd-induced density of states effect alluded to here is, in fact, predominantly a short range or local effect. Compositional variation³ of Sn fields in the HA Pd₂MnSn_{1-x}Sn_x contrasted to those in Cu₂MnIn_{1-x}Sn_x provides an independent clue that the Pd-based system may not have a simple band structure.

The models of both BC and JG are based on a charge screening model, and the conduction electrons are treated in a nearly free-electron-like scheme. Although different in approach, we believe that the basic physics contained in these two models is the same, i.e., if the approximations made by BC and JG can be eliminated, both approaches, in principle, would yield the same hyperfine field at a given nonmagnetic ion site. On the other hand, the approximations made in these theories are rather severe in the sense that the effects of band structure of the alloy are completely neglected. Therefore, a *quantitative* comparison between theory and experiment is not very meaningful. One hopes that the models predict the systematic trend of the hyperfine fields (change of sign and saturation) at nonmagnetic ion sites well. The agreement between the theory and experiment in the present calculation is therefore gratifying.

Recently, Stearns²⁰ has proposed a "volume misfit" model to account for hyperfine-field systematics at nonmagnetic ion sites in ferromagnetic hosts. In this model the host and impurity contri-

butions to the hyperfine field are separated. By subtracting s -conduction-electron polarization of the host from the measured hyperfine field, Stearns examines the remaining impurity contribution. She is then able to fit this contribution in terms of the difference between the volume occupied by the impurity and host ion. This approach ignores the fact that the host is metallic and that the electrons would have to screen the impurity ion. The volume misfit model does not take into account the physical concept of electron behavior in metals. We would like to mention that the effect of different volumes occupied by the nonmagnetic ions, having different charges, is actually contained explicitly in the JG model through the use of different Wigner-Seitz radii that simulate the range of the impurity potential. It is clear that a more realistic calculation involving the band structure of the ferromagnetic Heusler alloy is necessary to understand the hyperfine field *quantitatively*. Such band calculations may also provide further insight into the ferromagnetism of these HA.

Systematics of hyperfine fields at $5sp$ elements in metallic Fe host has been well documented,²¹ although our understanding of these systematics as they relate to the various mechanisms on the origin of fields continues to be a subject of some controversy. In contrasting the shape of the field systematics in Fe to those in the present HA, one is struck by the remarkable similarity of the curves from Cd ($Z_I=2$) to Sb ($Z_I=5$), and the equally remarkable dissimilarity in the curves from Te ($Z_I=6$) to Xe ($Z_I=8$). A possible way to understand these field systematics in Fe host could be to regard these fields to be made up of two significant contributions: (a) a CEP part important at low Z_I in the range $2 \leq Z_I \leq 6$ and (b) a direct overlap exchange polarization part dominant at high Z_I in the range $6 \leq Z_I \leq 8$, and contributing large positive fields that have been observed at Te, I, and Xe.

V. CONCLUSIONS

The principal results emerging from the present work may be summarized as follows: (i) Using $^{129}\text{Te}^m$ sources alloyed in Pd_2MnSb and PdMnSb , magnetic fields at ^{129}I substitutional at the Sb site in the two hosts have been measured and are found to be, respectively, 502 ± 10 and 681 ± 6 kOe at 4.2 °K. (ii) These fields are shown to form part of a systematic trend of fields observed at $5sp$ elements in these HA hosts. One can qualitatively understand this trend of fields in terms of a mechanism involving conduction-electron polarization. Calculations of fields based on a homogeneous spin-polarized electron-gas model, when compared to the experimental field results in Pd_2MnSb , reveal a systematic quantitative deviation. It is speculated that the origin of this quantitative deviation may rest in a Pd-induced band-structure effect. (iii) We have elucidated the metallurgy of a dilute Te impurity alloyed in Pd_2MnSb , using $^{125}\text{Te}^m$ and $^{129}\text{Te}^m$ labeled sources, and have shown the presence of profound heat-treatment effects. In particular, we have shown that both Te and I impurities in Pd_2MnSb become trapping centers for a defect produced on a thermal quench of the host material. This defect is believed to be associated with a Pd multiple vacancy and it is shown to be thermally activated by annealing at 300 °C. Following this heat treatment, Te and I impurities become substitutional at the Sb location in the host structure.

ACKNOWLEDGMENTS

We acknowledge the encouragement of Professor S. S. Hanna and Professor B. B. Triplett, and the generous assistance of C. C. Thomas in providing neutron irradiations. M. Marcuso and M. Blizzard assisted in the experimental work and data analysis. This work was supported in part by grants from the American Philosophical Society, the Department of Energy, and the NSF.

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