Volume dependence of magnetic hyperfine fields in Eu-intermetallic compounds

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High-pressure Mössbauer studies on ¹⁵¹Eu intermetallics (EuAl₂, EuAl₄, EuCu₂, EuCu₅ EuSn₃, Eu_{0.5}Yb_{0.5}Sn₃, EuZn₂, and EuPt₂) provide information about the volume dependence of the effective hyperfine field B_{eff} at the Eu²⁺ nucleus. These measurements have been performed at 4.2 K and pressures up to 65 kbar. The results show that (i) the volume dependence of the transferred hyperfine field B_{thf} is the dominating contribution to $\partial B_{\text{eff}}/\partial \ln V$; (ii) the magnitude of $\partial B_{\text{th}}/\partial \ln V$ can be explained only if one assumes the participation of s, p, and d conduction electrons in the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction responsible for B_{thr} ; and (iii) the exchange integrals for non-s conduction electrons ($J_{p,f}$ and/or $J_{d,f}$) are more sensitive to volume changes than that for s conduction electrons $(J_{s,f})$.

I. INTRODUCTION

The effective magnetic hyperfine field B_{eff} at the nucleus of $Eu^{2+}(4f^{78}S_{7/2})$ ions in metallic systems is commonly assumed to be composed of the following different contributions¹:

$$
B_{\text{eff}} = B_{\text{c}} + B_{\text{cep}} + B_{\text{thr}} + B_{\text{dip}} \quad ; \tag{1}
$$

 B_c represents the core polarization field, B_{cep} is the contribution from conduction-electron polarization by the magnetic ion itself, and B_{thf} the so-called transferred hyperfine field, stands for all other contributions from neighboring magnetic ions. The total dipolar field B_{dip} is the sum of the Lorentz field, the demagnetization field, and the contribution from neighboring magnetic dipoles; B_{dip} is usually small compared to the other three contributions. Such a model has been used to analyze the different contributions to B_{eff} in several Eu-intermetallic compounds, 2^{-4} assuming $B_c = (-34 \pm 2)$ T (Ref. 5) and B_{cen} to be independent of the Eu concentration in magnetically diluted systems.

A study of the volume dependence of B_{eff} in intermetallic systems by means of high-pressure Mössbauer experiments provides further information on the applicability of such a model. We have shown previously⁶ that the magnitude of B_{eff} increased in the intermetallic systems $EuSn₃$, $EuAl₂$, $EuAl₄$, EuCu₂, and EuCu₅ and decreased in EuZn₂ (Ref. 6) with increasing pressure. In the meantime we have performed additional high-pressure Mössbauer experiments on EuPt₂ and on magnetically diluted EuSn₃, i.e., on $Eu_{0.5}Yb_{0.5}Sn_3$. These experiments support the qualitative discussion of $\partial B_{eff}/\partial p$ made in our previous paper. Furthermore, the dilution experiment offers the possibility to separate the different contributions to $\partial B_{\text{eff}}/\partial p$ and to propose a model for the mechanism involved in producing B_{thf} in these intermetallic compounds. This model will be discussed in connection with the calculation of the partial density of states at the Fermi surface in the model compound LaSn₃.^{7,8}

II. EXPERIMENTAL DETAILS

The high-pressure Mössbauer experiments were performed with a Chester-Jones type high-pressure setup with B_4C anvils allowing Mössbauer transmission experiments at liquid-helium temperature with low γ -ray energies.⁹ The high-pressure cell is based on a pyrophylite ring supported from outside by a steel belt which provides an almost quasihydrostatic pressure at the absorber up to about 65 kbar. Absorbers were in the form of epoxy-cast pellets with a typical size of \simeq 4-mm diameter and \simeq 0.6–0.8-mm thickness. A superconducting lead manometer¹⁰ was employed in the pressure cell for in situ measurements of the mean value of the pressure and the pressure profile across the absorber.

The compressibility data were obtained with a high-pressure cell especially constructed for x-rayscattering measurements (Debye-Scherrer method) with low x-ray absorption (diamond anvils). NaCl was employed in the pressure cell for an *in situ* pressure calibration.

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Most of the Eu intermetallic compounds were the direct reaction of stoichiometri amounts of the elements in degassed and sealed molybdenum or tantalum (in the case of EuSn₃) crucibles. After about 1 h induction heating at temperatures between 900 and 1200 °C the samples were annealed for several days at $\approx 700^{\circ}$ C. EuZn₂ and Eu_{0.75}Sr_{0.25}Zn, were obtained by induction melting of the elements in a water-cooled copper boat in a pure argon atmosphere. EuPt, was prepared in the Phillips Research Laboratories, Eindhoven, The Netherlands. In all cases (with the exception of $EuSn₃$, see Fig. 1) a Eu^{3+} contamination was not detectable in the Mössbauer spectra. For all samples the x-ray powder patterns showed only lines belonging to the corretructures. Typical total absorb thicknesses were $40-70$ mg/cm². The Mössbauer abdependence of the line of the increased to the control of the interest of the opportunity of the control of the sinusoidally moved ¹⁵¹SmF₃ source (100 mCi; \simeq 4-mm active diameter). Both source and absorber were kept at 4.2 K during the Mössbauer runs.

III. RESULTS

Typical ¹⁵¹Eu Mössbauer absorption spectra of etallic compounds at different presanne compounds at direre $uSn₃$ and EuCu₅) have been fitted w
ically split hyperfine (hf) pattern (18 Lorentzian lines) assuming the theoretical ratios as expected for polycrystalline absorbers without texture. In the case of $EuSn_3$ (see Fig. 1) an additional small $Eu³⁺$ line, probably due to some $Eu₂O₃$ contamination, had to be added. A quadrupo lar interaction is clearly visible in the $EuCu_s$ spectrum (see Fig. 3); the observed electric field gradient of $V_{zz} = 2.5 \times 10^{-17}$ V/cm² did not change with pressure. In all other compounds the quadrupo igibly small. Due to the unresolved hf pat t_2 we had to make assumptions on th linewidth W. For $\frac{1}{2}W = 1.5$ mm/s, as is typical for
Eu-intermetallic compounds, we obtain $B_{\text{eff}} = 4.6$ ± 0.3 T at 0 kbar, a value which is slightly higher

FIG. 1. 151 Eu Mössbauer absorption spectrum of EuSn₃ at 4.2 K and different pressures. Doppler velocity v is given relative to $^{151}SmF_3$ at 4.2 K.

FIG. 2. 151 Eu Mössbauer absorption spectrum of EuAl₄ at 4.2 K and different pressures.

FIG. 3. 151 Eu Mössbauer absorption spectrum of EuCu₅ at 4.2 K and different pressures, ' The solid lines are the results of least-squares fits assuming a magnetically split hyperfine pattern plus a small quadrupolar interaction.

than that given by Kropp et al. from NMR measurements. $¹¹$ This discrepancy can be easily explained by</sup> a small Pt excess $(>= 5$ at. %) in our EuPt₂ sample.¹²

The pressure dependence of the effective magnetic hyperfine field B_{eff} at the ¹⁵¹Eu nucleus and of the 151 Eu isomer shift S (relative to the 151 SmF₃ source) are shown in Figs. 5 and 6. In all cases B_{eff} and S vary approximately linearly with applied pressure p . The isomer shift increases, i.e., the electron density $\rho(0)$ at the ¹⁵¹Eu nucleus increases in all compounds with pressure. The magnitude of B_{eff} increases in EuAl₂, EuAl₄, EuCu₅, and EuSn₃ but decreases in $EuZn₂$ and $EuPt₂$ with increasing pressure.

In order to get more information about the main contribution to $\partial B_{\text{eff}}/\partial p$ we have studied magnetically diluted EuSn₃, i.e., $Eu_{0.5}Yb_{0.5}Sn₃$, as a function of applied pressure. The results of this experiment are shown in Fig. 7: $\partial |B_{\text{eff}}|/\partial p$ is reduced from $\partial |B_{\text{eff}}|/\partial p = (6.5 \pm 0.5) \times 10^{-2}$ T/kbar for EuSn₃ to about half of that value for $Eu_{0.5}Yb_{0.5}Sn_3$ $[8]B_{\text{eff}}/8p = (3.0 \pm 0.6) \times 10^{-2}$ T/kbar, see Table I]; the isomer shift S, however, is almost unchanged.

FIG. 4. ¹⁵¹Eu Mössbauer absorption spectrum of EuPt₂ at 4.2 K and different pressures. The solid lines are the results of least-squares fits assuming a pressure independent quadrupolar interaction as given by Ref. 11 and a linewidth $\frac{1}{2}W = 1.5$ mm/s, independent of pressure.

FIG. 5. Effective magnetic hyperfine field B_{eff} at the $Eu²⁺$ nucleus as a function of applied pressure for several Eu intermetallics at 4.2 K as obtained from least-squares fits.

FIG. 6. 151 Eu isomer shift S as a function of applied pressure for several Eu intermetallics at 4.2 K. S values are given relative to $^{151}SmF_3$ at 4.2 K.

FIG. 7. (a) Pressure dependence of the effective magnetic hyperfine field B_{eff} at the Eu²⁺ nucleus for EuSn₃ and $Eu_{0.5}Yb_{0.5}Sn_3$. (b) Pressure dependence of the ¹⁵¹Eu isome shift S for EuSn₃ and Eu_{0.5}Yb_{0.5}Sn₃.

Compound	S (mm/s)	$B_{\rm eff}$ (4.2 K) (T)	$B_{\text{eff}}(0)$ (T)	$B_{\rm cep}$ (T)	B_{thr} (T)	κ	$\partial S/\partial p$ $(10^{-3} \text{ kbar}^{-1})$ $(10^{-2} \text{ mm/s kbar})$ $(10^{-2} \text{ T kbar}^{-1})$	$\partial B_{\rm eff} /\partial p$
EuAl ₂	$-8.94(1)$	$-27.0(3)$	$-28.1(4)$	$+10.5(2.0)^a$ $-4.5(1.0)^a$		1.11(6)	$+1.8(1)$	$+6.0(5)$
EuAl ₄	$-10.98(2)$	$-27.8(4)$	$-28.9(5)$	$+4(2)^a$	$-1(1)^{b}$		$+1.3(1)$	$+5.3(5)$
EuCu ₂	$-8.27(4)$	$-17.3(3)$	$-18.4(5)$	$+14(2)^a$	$0(1)^a$		$+2.0(2)$	$+1.3(5)$
EuCu ₅	$-8.26(2)$	$-27.4(3)$	$-27.5(4)$	$+7.6(5)^c$	$-0.5(5)^c$		$+1.0(1)$	$+2.7(5)$
EuSn ₃	$-10.04(4)$	$-28.1(7)$	$-28.4(5)$	$+6.2(4)^{b}$.	$-0.6(6)$ ^b	1.01(7)	$+1.7(1)$	$+6.5(5)$
$Eu_{0.5}Yb_{0.5}Sn_3$	$-10.09(7)$	$-27.6(7)$	$-27.9(8)$	$+6.2(4)$ ^b	$-0.3(6)^{b}$		$+1.5(2)$	$+3.0(6)$
EuZn ₂	$-9.1(1)$	$-23.8(3)$	$-24.4(4)$	$+7.6(6)$	$+2(2)$		$+2.3(1)$	$-3.5(1)$
EuPt ₂	$-8.6(1)$	$-4.6(3)$	$-4.6(3)$	$+10(5)^d$	$+20(5)^d$	0.53(4)	$+0.7(4)$	$-1.8(1)$
^a Reference 2.					^c Reference 4.			

TABLE I. Summary of high-pressure Mössbauer and compressibility (κ) data for Eu intermetallics. The numbers in parentheses represent numerical errors in the last digit; i.e., $x(y) \equiv x \pm y$.

^bReference 3.

'Reference 4. ^dReference 11.

FIG. 8. Density of states for LaSn₃ calculated by Hackenbracht (Ref. 8). The partial densities of states (Sn5s, Sn5p, and $La5d$) as well as the total density of states per unit cell are shown.

A summary of all hf data together with some compressibility data and values for B_{cep} and B_{thr} as far as known are given in Table I. All S and B_{eff} values measured at 0 kbar (Table I, columns I and 2) are in agreement with the experimental errors giver by other authors.^{2–4} $B_{\text{eff}}(0)$ is obtained from the measured value of B_{eff} at 4.2 K by extrapolating to $T = 0$ K using the known ordering temperatures and the Brillouin function for $S = \frac{7}{2}$. The value of B_{thf} for EuZn₂ has been obtained from B_{eff} in magnetically diluted $Eu_{1-x}Sr_xZn_2$ with $x = 0.25$ and by extrapolating to $x = 1$ as described in Ref. 1. Since the dipolar field B_{dip} is unknown for all compounds, with the exception of EuCu₅, it is included in the B_{thf} values given in Table I. For $EuCu₅$ the dipolar field $B_{\text{dip}}=0.4$ T (Ref. 4) has been subtracted.

IV. DISCUSSION

We will describe the effective hyperfine field B_{eff} at the $^{151}Eu^{2+}$ nucleus as given by Eq. (1). For the discussion of the pressure dependence of B_{eff} we assume B_c to be pressure independent. Since B_c is caused by the polarization of the inner s shells by. the 4f moment, any pressure effects on inner shells are neglected by this assumption. The pressure dependence of B_{dip} is generally given by $\partial \ln B_{\text{dip}}/\partial \ln V$ $=-1$, which is very weak (see below) and will be neglected. The pressure dependence of B_{eff} is now

reduced to

$$
\frac{\partial B_{\text{eff}}}{\partial p} \simeq \frac{\partial B_{\text{cep}}}{\partial p} + \frac{\partial B_{\text{thr}}}{\partial p} \quad . \tag{2}
$$

The positive values for B_{cep} in all compounds studied (see Table I) together with the usual assumption that the spin density of the conduction electrons increases with decreasing volume gives $\partial B_{\text{cen}}/\partial p > 0$ for all compounds. If $\partial B_{\rm cen}/\partial p$ would be the dominant contribution to $\partial B_{\text{eff}}/\partial p$ one should observe a negative value for $\partial B_{\text{eff}}/\partial p$ for all compounds $(B_{\text{eff}} < 0$, see Table I). Since a negative value for $\partial |B_{\text{eff}}|/\partial p$ is only observed for EuZn₂ and EuPt₂, we have to conclude that $\partial B_{\text{cep}}/\partial p$ is *not* responsible for the observed behavior of $\partial |B_{\text{eff}}|/\partial p$.

The influence of $\partial B_{\text{th}}/\partial p$ on $\partial B_{\text{eff}}/\partial p$ can be discussed as follows: B_{thf} is positive for EuZn₂ and $EuPt₂$ and negative for all other compounds (see Table I). If we assume that the magnitude of B_{thr} increases with decreasing volume (a detailed discussion of this point will be given later), we obtain $\partial B_{\text{thf}}/\partial p < 0$ for EuZn₂ and EuPt₂ and $\partial B_{\text{thf}}/\partial p > 0$ for all other compounds. Putting this pressure dependence of B_{thf} in Eq. (2) we obtain the experimentally observed sign for $\partial |B_{\text{eff}}|/\partial p$, namely, $\partial |B_{\text{eff}}|/\partial p < 0$ for EuZn₂ and EuPt₂ and $\partial |B_{\text{eff}}|/\partial p > 0$ for all other compounds. Thus we come to the conclusion, that $\partial B_{\text{thr}}/\partial p$ is responsible for the observed behavior of $\partial |B_{\text{eff}}|/\partial p$.

This conclusion is further supported by the results

obtained from magnetically diluted EuSn₃: in $Eu_{0.5}Yb_{0.5}Sn_3$ the value of B_{thf} is about half that for EuS n_3 . If our conclusion is correct the value of $\partial |B_{\text{eff}}|/\partial p$ in Eu_{0.5}Yb_{0.5}Sn₃ should be about half that for EuS n_3 . This is exactly what we have observed (see above).

In the following we discuss the observed volume dependence of B_{th} within the framework of the RKKY theory. The RKKY theory describes the long-range exchange interaction between localized spins \overline{S}_i and the conduction electrons which results in a conduction-electron polarization and thus in an effective transferred hyperfine field at the Eu^{2+} site. If the conduction electrons are described by s wave functions only, as it is usually done, one obtains 13

$$
B_{\text{th}}^{\text{RKKY}} = \frac{9\pi n^2 A(s)}{2E_F g_n \beta_n} J_{sf}(\vec{S})_s; (\vec{S})_s = \sum_i F(2k_F R_i) \vec{S}_i
$$
 (3)

 $A(s)$ represents the hyperfine coupling constant corresponding to the direct interaction of an unpaired 6s electron with the nucleus, g_n and β_n are the nuclear g value and the nuclear magneton, respectively, E_F is the Fermi-level energy, n is the conduction-electronto-atom ratio, J_{sf} denotes the effective exchange integral that describes the coupling between the $4f$ moments and the s conduction electrons, $\langle \overline{S} \rangle_s$ stands for the spin polarization of the s electrons, F is the so-called Ruderman-Kittel function, k_F is the Fermi vector, and R_i , the position of the *i*th localized spin \vec{S}_i .

In the free-electron model the sum $\sum_i F(2k_F R_i) \vec{S}_i$ is volume independent $(k_F \propto V^{-1/3}, R_i \propto V^{1/3})$. We assume that the volume dependence of $B_{\text{th}}^{\text{RKKY}}$ is mainly given by the volume dependence of the exchange interaction J_{sf} ,

 $\partial \ln B_{\rm th}^{\rm RKKY}/\partial \ln V \simeq \partial \ln J_{\rm sf}/\partial \ln V$.

It is expected that $\partial \ln J_{sf}/\partial \ln V$ is of the order of -1 or even smaller.¹⁴ From the measured pressure dependence of B_{eff} we obtain the following $\partial \ln B_{\text{thf}}/\partial \ln V$ values for the compounds whose compressibilities are known (see Table I): -1.7 for EuPt₂, -12 for EuAl₂, and -100 for EuSn₃. The value for EuPt, can be explained within the RKKY model as described above. The high values of $\partial \ln B_{\text{th}}/\partial \ln V$ for EuAl₂ and especially for EuSn₃, however, are physically unreasonable if they are discussed within this model. In order to explain the high $\partial \ln B_{\text{th}}/\partial \ln V$ values, B_{th} has to be an effective sum of at least two (or more) contributions with different signs and different volume dependences. Such a sum would result from the participation of non-slike conduction electrons in the RKKY interaction. Similar considerations have been made by other authors discussing the exchange interaction in interme
tallic rare-earth compounds.^{15,16} tallic rare-earth compounds.^{15, 16}

In order to confirm the importance of p and d conduction electrons for the RKKY interaction the partial density of states at the Fermi surface has been calculated for $LaSn_3$ (model compound for $EuSn_3$, which shows the largest $\partial \ln B_{\text{thf}}/\partial \ln V$ value) by Hackenbracht, 8 using the method given in Ref. 7. The result of this calculation can be seen in Fig. 1: the main contribution to the total density of states comes from the Sn5p electrons (\approx 60%) and La5d electrons (\approx 20%). A modification of $B_{\text{th}}^{\text{RKKY}}$, which takes non-s-like conduction electrons into account is therefore needed for $EuSn₃$ and all the other intermetallics which show high $\partial \ln B_{\text{thf}}/\partial \ln V$ values. In this case B_{thf} can be written as

$$
B_{\text{thf}}^{\text{RKKY}} = (B_{\text{thf}}^{\text{RKKY}})_s + (B_{\text{thf}}^{\text{RKKY}})_{\text{non-s}}
$$

=
$$
\frac{9\pi n^2}{2E_F g_n \beta_n} [A(s) J_{sf}(\vec{S})_s + A(p) J_{pf}(\vec{S})_p
$$

+
$$
A(d) J_{df}(\vec{S})_d + \cdots] , \qquad (4)
$$

where $\langle \vec{S} \rangle_{g}$ and $\langle \vec{S} \rangle_{g}$ stand for the spin polarization of the p and d conduction electrons, respectively. The effective exchange integrals J_{sf} , J_{pf} , and J_{df} are of the same order of magnitude.¹⁶ The hyperfin parameters $A(s)$, $A(p)$, and $A(d)$, however, differ in their signs and their magnitudes: $A(p) \approx A(d)$ \approx -0.1A (s).^{17,18} Using these values, Eq. (4) can be simplified in the following form:

$$
B_{\text{th}}^{\text{RKKY}} \simeq (\text{const})A \text{ (s)}
$$

$$
\times [J_{sf}(\vec{S})_s - 0.1(J_{pf}(\vec{S})_p + J_{sf}(\vec{S})_d)] \quad . \quad (5)
$$

Equation (5) shows that the magnitude and the sign of B_{thf}^{RKKY} is determined by the relative magnitudes of the s- and non-s (p,d) contributions. A small, negative B_{thf} is observed for all Eu-intermetallic compounds given in Table I with the exception of EuPt₂. One has to conclude, therefore, that the non-s conduction-electron contribution to B_{th} is slightly larger than the s conduction-electron contribution in these compounds. For $EuPt_2$ the value of B_{thf} is large and positive [(+20 \pm 5) T] which clearly shows the dominant s contribution to B_{thr} [see Eq. (5)]. This explains why the value of $\partial \ln B_{\text{thf}}/\partial \ln V$ for EuPt₂ fits within the RKKY model using only s conduction electrons.

The observed volume dependence of B_{thr} offers now the possibility to get an information about the volume dependences of J_{sf} and J_{pf} or J_{df} . The volume dependence of $B_{\text{thr}}^{\text{RKKY}}$ as deduced from Eq. (4) is

$$
\frac{\partial B_{\text{th}}^{\text{RKKY}}}{\partial \ln V} = \left(\frac{\partial B_{\text{th}}^{\text{RKKY}}}{\partial \ln V} \right)_s + \left(\frac{\partial B_{\text{th}}^{\text{RKKY}}}{\partial \ln V} \right)_{\text{non-s}} \quad . \tag{6}
$$

The measured value of $\partial B_{\text{th}}/\partial \ln V$ is negative for

EuPt₂ [(-35 ± 5) T] and positive (approximately the same magnitude) for the other compounds $[(+55 \pm 5)$ T for EuAl₂ and $(+65 \pm 5)$ T for EuSn₃]. Using Eq. (6) together with the fact that

$$
(B_{\text{thf}}^{\text{RKKY}})_s \simeq (B_{\text{thf}}^{\text{RKKY}})_{\text{non-s}}
$$

for all compounds, with the exception of $EuPt_2$ (see for all compounds, with the exception of μ_{th}
above) where $(B_{\text{thf}}^{\text{RKKY}})_{\text{non-s}} \approx 0$, we obtain

$$
(\partial \ln B_{\rm thr}^{\rm RKKY}/\partial \ln V)_{\rm non-s} \simeq 2(\partial \ln B_{\rm thr}^{\rm RKKY}/\partial \ln V)_{s}
$$

If we take into account that $\langle \vec{s} \rangle_s$, $\langle \vec{s} \rangle_p$, and $\langle \vec{s} \rangle_d$ do not change with volume (see above) and assume that $A(s)$ and $A(d)$ are essentially volume independent, we get

$$
\partial \ln J_{\text{non-s},f} / \partial \ln V \simeq 2 \partial \ln J_{s,f} / \partial \ln V
$$

We want to point out that this result does not depend on any assumption regarding the magnitudes of $J_{s,f}$, $J_{p,f}$, and $J_{d,f}$. A stronger volume dependence of the exchange integrals for non-s conduction electrons compared to that for s conduction electrons is expected from theoretical arguments¹⁹: the overlap of the non-s-like states with the $4f$ states is more sensitive to a volume change than that for s-like states.

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V. CONCLUSION

High-pressure Mössbauer studies of the effective hyperfine field B_{eff} at the Eu²⁺ nucleus in Eu-intermetallics offer the possibility to study the volume dependences of the different contributions to B_{eff} . We could show that (i) $\partial B_{\text{th}}/\partial \ln V$ is the dominant contribution to $\partial B_{\text{eff}}/\partial \ln V$ and (ii) the magnitude of $\partial \ln B_{\text{th}}/\partial \ln V$ can be explained only if one assumes the participation of p and d conduction electrons in the RKKY interaction responsible for B_{thr} . This was confirmed by a calculation of the partial density of states at the Fermi surface of $LaSn_3$ (representative for $EuSn₃$). Thus our high-pressure Mössbauer studies together with these calculations lead to the conclusion that the magnitude of B_{thf} is an effective sum of different contributions due to the participation of s, p , and d conduction electrons in the RKKY interaction. Furthermore, the volume dependence of B_{thf} shows that the exchange integrals for non-s conduction electrons $(J_{\text{pf}}$ and/or J_{df}) are more sensitive to volume changes than that for s conduction electrons $(J_{s,f}).$

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