

# Effect of pressure on the exchange-enhanced band paramagnetism of palladium alloys

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The influence of hydrostatic pressure at various temperatures on the magnetic susceptibility of palladium-based alloys characterized by high Stoner factors is investigated experimentally. The magnitude of the magnetovolumetric effect is investigated as a function of concentration and temperature using simple empirical models of the change in the electron spectrum under pressure and due to alloying within the framework of the band approach, and estimates of the volume integrals are obtained for the primary spectral parameters and the effective exchange interaction  $I$ . The quantity  $d \ln I / d \ln V = -0.7 \pm 0.2$  differs substantially from the theoretical data for palladium, which suggests a weaker volumetric dependence of  $I$ .

Exchange-correlated enhancement of the spin paramagnetism of band electrons is one source of information on many-particle interactions in metals. The Stoner factor characterizing this enhancement is determined by the expression

$$\chi = S\chi_s = \chi_s (1 - I\chi_s)^{-1}, \quad (1)$$

where  $\chi_s$  is the unperturbed spin susceptibility,  $I$  is the effective exchange interaction parameter.  $S$  reaches values of order 10 in palladium and palladium-based alloys<sup>1</sup> and is convenient for investigating the relationship between  $I$  and the characteristics of the electron spectrum, which can be controlled by varying the atomic volume. Several studies have been devoted to finding the magnetovolumetric effect  $d \ln \chi / d \ln V$  in palladium and palladium-based alloys. Direct measurements of the pressurized susceptibility  $\chi(P)$  of pure palladium in the 4.2-300 K range<sup>2</sup> have yielded a relatively weak temperature dependence of the magnetovolumetric effect. Data from analogous research for pure palladium and a comparison of the magnitude of the effect in Pd-Rh alloys at  $T = 300$  K from Ref. 3-5 and  $T = 4.2$  K from volumetric magnetostriction research<sup>6,7</sup> indicate a temperature dependence of the magnetovolumetric effect that is stronger than in Ref. 2. The volumetric magnetostriction data in turn substantially magnify the low-temperature result from Ref. 3 for Pd<sub>0.95</sub>Rh<sub>0.05</sub>. The susceptibility of epitaxially-distended palladium films in Au-Pd-Au sandwiches corresponds to an even greater magnetovolumetric effect at low temperatures.<sup>8</sup> The inconsistency of existing experimental information has led to the further investigation of the temperature dependences of the magnetovolumetric effect.

Below we present results from systematic studies of the influence of pressure on the magnetic susceptibility of palladium alloys containing rhodium and silver at fixed temperatures of 20.4 and 78 K, and a quantitative analysis of the data is carried out within the framework of the band model. The baric derivative values for the primary characteristics of the electron spectrum and the effective exchange interaction parameter are compared to results from theoretical calculations.

## EXPERIMENTAL TECHNIQUE AND RESULTS

The influence of pressure on magnetic susceptibility was measured, as previously,<sup>4</sup> by the levitation technique:

Suspension of a specimen by a strong inhomogeneous magnetic field. The susceptibility in this case is determined by equating the magnetic force and the specimen weight (accounting for the hydrostatic support of the medium) when the levitation condition breaks down and the specimen falls. The low-temperature implementation of the method is described briefly in Ref. 3 and here we simply note that unlike Ref. 4 the field source in this case was a superconducting solenoid, with gaseous helium used as the medium pressurizing the specimen. The relative measurement accuracy (better than 0.03%) made possible reliable determination of  $\chi(P)$  up to 1.7 kbar at  $T = 20.4$  K and up to 2.5 kbar at  $T = 78$  K. The binary palladium alloys Pd<sub>1-x</sub>Rh<sub>x</sub> ( $0 \leq x \leq 0.2$ ) and Pd<sub>1-y</sub>Ag<sub>y</sub> ( $0 \leq y \leq 0.05$ ) were investigated in these conditions together with the ternary alloys Pd<sub>1-x-y</sub>Rh<sub>x</sub>Ag<sub>y</sub> ( $0 \leq x \leq 0.1$ ,  $y = 0.05$ ) containing a fixed quantity of silver. This study essentially used the same specimens as in preceding studies<sup>4,5</sup> or specimens fabricated by an analogous technology from identical raw materials.

The standard experimental relationships  $\chi(P)$  in Fig. 1 demonstrate the scale and linearity of the pressure effects. The baric susceptibility derivatives at  $T = 20.4$  and 78 K are shown in Table I together with analogous data at room temperature obtained previously.<sup>4,5</sup> Figures 2 and 3 show the

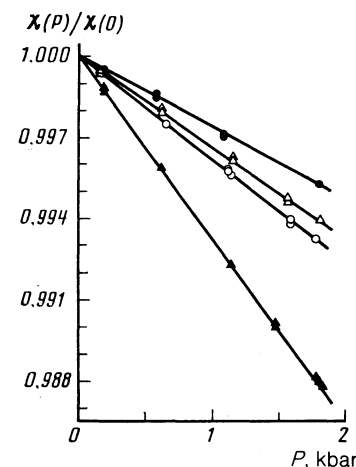


FIG. 1. Representative experimental dependences of susceptibility on pressure in palladium alloys at  $T = 20.4$  K: ●—Pd; △—Pd<sub>0.95</sub>Ag<sub>0.05</sub>; ▲—Pd<sub>0.95</sub>Rh<sub>0.05</sub>; ○—Pd<sub>0.80</sub>Rh<sub>0.20</sub>.

TABLE I. Experimental pressure derivatives of the susceptibility in palladium alloys at various temperatures.

Composition	$-d \ln \chi / dP, \text{ Mbar}^{-1}$		
	20.4 K	78 K	293 K *
Pd	2.7 (0.2) **	2.6 (0.1)	1.46 (0.1)
Pd <sub>99</sub> Rh <sub>1</sub>	3.9 (0.3)	3.7 (0.3)	—
Pd <sub>98</sub> Rh <sub>2</sub>	4.9 (0.4)	4.2 (0.2)	1.94 (0.15)
Pd <sub>95</sub> Rh <sub>5</sub>	6.7 (0.3)	5.4 (0.25)	2.64 (0.2)
Pd <sub>92</sub> Rh <sub>8</sub>	5.95 (0.3)	5.2 (0.3)	2.65 (0.12)
Pd <sub>91</sub> Rh <sub>9</sub>	—	5.3 (0.2)	2.80 (0.15)
Pd <sub>89</sub> Rh <sub>11</sub>	—	5.0 (0.3)	2.86 (0.1)
Pd <sub>85</sub> Rh <sub>15</sub>	—	4.5 (0.2)	2.88 (0.1)
Pd <sub>80</sub> Rh <sub>20</sub>	3.9 (0.3)	4.15 (0.2)	2.64 (0.2)
Pd <sub>99</sub> Ag	2.5 (0.3)	2.8 (0.3)	1.65 (0.15)
Pd <sub>98</sub> Ag <sub>2</sub>	—	2.6 (0.2)	1.40 (0.15)
Pd <sub>97</sub> Ag <sub>3</sub>	—	2.5 (0.3)	—
Pd <sub>95</sub> Ag <sub>5</sub>	3.3 (0.3)	2.6 (0.3)	—
Pd <sub>92</sub> Ag <sub>8</sub> Rh <sub>3</sub>	—	4.15 (0.2)	1.95 (0.15)
Pd <sub>90</sub> Ag <sub>5</sub> Rh <sub>5</sub>	5.3 (0.4)	4.7 (0.4)	2.15 (0.15)
Pd <sub>88</sub> Ag <sub>5</sub> Rh <sub>7</sub>	—	4.8 (0.4)	2.3 (0.15)
Pd <sub>86</sub> Ag <sub>5</sub> Rh <sub>9</sub>	—	4.55 (0.3)	2.2 (0.15)
Pd <sub>85</sub> Ag <sub>5</sub> Rh <sub>10</sub>	—	4.0 (0.3)	2.25 (0.15)

\*Data from Ref. 4, 5 corrected to account for the refined compressibility of the medium exerting pressure on the specimen.<sup>3</sup>

\*\*The error is given in parentheses.

magnetovolumetric effects in binary and ternary alloys as a function of concentration. A linear interpolation of the bulk compression moduli for pure components at the corresponding temperature<sup>9,10</sup> was used for the bulk compression moduli of the alloys; this is a quite reasonable approximation.<sup>11</sup> Clearly the magnitude of the effect grows by a factor of two to three when the temperature drops from 300 to 20 K, while the influence of temperature is substantial for the Pd–Rh alloys in the 20–78 K range as well.

The concentration relation of the magnetovolumetric effect in Pd–Rh alloys at 20.4 K qualitatively corresponds to the results from magnetostriction research at 4.2 K (Refs. 6 and 7). The cause of the substantial quantitative discrepancy is considered in Ref. 3 using the Pd<sub>0.95</sub>Rh<sub>0.05</sub> alloy. It arises in the analysis of the magnetostriction data as a result of ignoring the parasitic contribution to the susceptibility from the impurity magnetic moments which becomes substantial at low temperatures. The nature of the behavior of this contribution under pressure<sup>3</sup> suggests that the values found in the present study for the magnetovolumetric effect are left

virtually undistorted by the impurity moments and can be used for further quantitative analysis without additional correction.

#### DISCUSSION OF EXPERIMENTAL RESULTS

The dominant contribution to the magnetic susceptibility of palladium-based alloys can be attributed to the spin paramagnetism of the *d*-electrons described by expression (1) in which

$$\chi_s(T) = -\mu_B^2 \int_0^{\infty} N(E) \frac{\partial f}{\partial E} dE. \quad (2)$$

Here  $N(E) \propto \Delta_d^{-1}$  is the electron state density at  $T = 0$  K,  $\Delta_d$  is the width of the *d*-band,  $f$  is the Fermi distribution function.

The behavior of the electron state density under pressure on the Fermi level  $E_F$  is determined by the expression<sup>12</sup>

$$\frac{d \ln N(E_F)}{d \ln V} = \frac{\partial \ln N(E_F)}{\partial \ln V} + \frac{\partial \ln N(E_F)}{\partial q} \frac{dq}{d \ln V}, \quad (3)$$

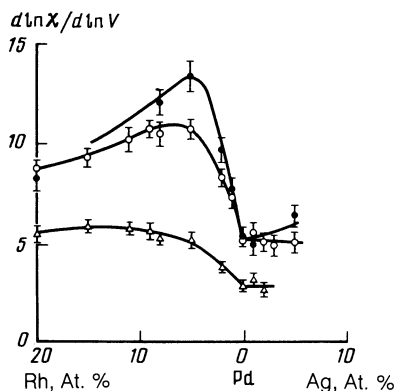


FIG. 2. Magnitude of the magnetovolumetric effect as a function of concentration in binary palladium alloys containing rhodium and silver at various temperatures: ●—20 K; ○—78 K; △—293 K.

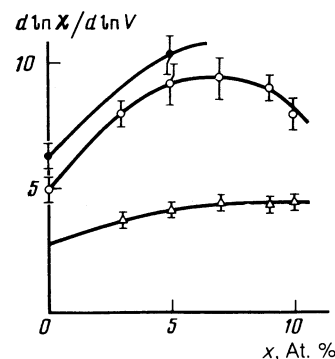


FIG. 3. Magnitude of the magnetovolumetric effect in ternary Pd<sub>0.95-x</sub>Ag<sub>5</sub>Rh<sub>x</sub> alloys at various temperatures: ●—20 K; ○—78 K; △—293 K.

where the first term on the right side reflects broadening of the  $d$ -band (the deformation contribution), while the second term reflects the change in the number of  $d$ -electrons  $q$  due to the relative displacement of the states  $s(p)$  and the  $d$ -bands on the Fermi level (the shift contribution). The  $d$ -band is assumed to be uniformly deformed, i.e.

$$\frac{\partial \ln N(E_F)}{\partial \ln V} = - \frac{d \ln \Delta_d}{d \ln V},$$

and the derivative  $dq/d \ln V$  is assumed to be constant in a limited energy range. Consequently for the magnetovolumetric effect we have from (1)–(3):

$$\frac{d \ln \chi}{d \ln V} = \frac{\partial \ln \chi_s}{\partial \ln V} + I \chi \left( \frac{\partial \ln \chi_s}{\partial \ln V} + \frac{\partial \ln I}{\partial \ln V} \right) + \frac{\partial \ln \chi}{\partial q} \frac{dq}{d \ln V}. \quad (4)$$

Taking into account (2) and the estimate  $d \ln \Delta_d / d \ln V = -5/3$  (Ref. 13), we obtain

$$\frac{\partial \ln \chi_s}{\partial \ln V} = - \frac{d \ln \Delta_d}{d \ln V} \left( 1 + \frac{\partial \ln \chi_s}{\partial \ln T} \right) = \frac{5}{3} \left( 1 + \frac{\partial \ln \chi_s}{\partial \ln T} \right). \quad (5)$$

After substitution of (5) into (4) we obtain the pressure effect in the following form:

$$\begin{aligned} \frac{d \ln \chi}{d \ln V} &= \frac{\partial \ln \chi}{\partial q} \frac{dq}{d \ln V} \\ &= \frac{5}{3} \left( 1 + \frac{\partial \ln \chi_s}{\partial \ln T} \right) + I \chi \left( \frac{5}{3} + \frac{d \ln I}{d \ln V} \right). \end{aligned} \quad (6)$$

At low temperatures when  $d \ln \chi / d \ln T$  is small the deformation term remaining on the right side of (6) is a linear function of  $\chi$  whose slope yields  $d \ln I / d \ln V$  (we assume  $\partial \ln I / \partial \ln V = d \ln I / d \ln V = \text{const}$  in accordance with the weak dependence of the calculated values of  $I$  on the electron concentration in the Rh–Pd–Ag series<sup>14</sup>).

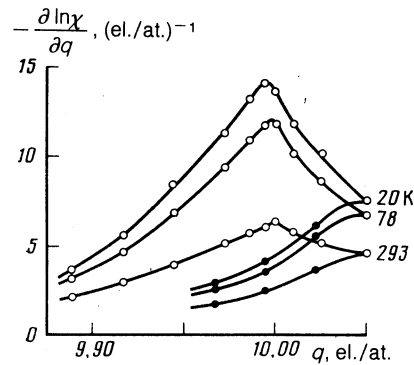


FIG. 4. The derivatives  $\partial \ln \chi / \partial q$  plotted as a function of electron concentration in binary (○) and ternary (●) palladium alloys containing rhodium and silver. Error: Less than 10%.

In order to calculate the contribution of  $(\partial \ln \chi / \partial q)(dq/d \ln V)$  to the left side of (6) due to electron overflow it is necessary to know the differential characteristics  $\partial \ln \chi / \partial q$  of the specific alloys. The difference in the susceptibility of the binary and ternary isoelectronic palladium alloys suggests a significant smoothing of the spectral fine structure due to electron scattering in a disordered alloy,<sup>15</sup> since the hard band model is not suitable for obtaining  $\partial \ln \chi / \partial q$  without a scattering correction. Such a correction can be incorporated empirically, without relying on theoretical models<sup>16</sup> whose accuracy is not adequate for the spectral fine structure which is important for the magnetic properties of palladium alloys. The scheme to account for scattering is based on using a grid of experimental susceptibilities of isoelectronic alloys with variable total impurity concentrations and different scattering parameters  $\Gamma^{5,15}$  which makes it possible to determine the  $\chi(q)_{\Gamma = \text{const}}$  relation at a fixed temperature. They can then be used to find the derivative  $(\partial \ln \chi /$

TABLE II. Parameters characterizing the spectrum and electron-electron interaction in palladium.

Parameter	Theory	Experiment
$\Delta_d$ , Ry	{ 0.43 [1, 18] 0.42 [17] 0.45 [19]	–
$N(E_F)$ , stat/at·Ry	{ 32.7 [1] 31.0 [14, 17] 34.7 [18] 29.5 [20] 33.96 [21]	–
$-\frac{\partial \ln N(E_F)}{\partial q}$ (el./at.) <sup>-1</sup>	{ 1.9 [1] 2.36 [17] 1.8 [21]	1.6 ± 0.2
$\chi(0)$ , 10 <sup>-6</sup> EMU/g	–	{ 6.77 [22] 6.9 [15]
$S$	{ 4.46 [14] 4.8 [20] 13.6 [18] 8.5 [21]	9.0 ± 1.3* ± 0.2
$J$ , 10 <sup>-3</sup> , Ry **	{ 25.0 [14] 26.7 [18, 20] 25.97 [21]	26.2 *
$I$ , 10 <sup>6</sup> (EMU/g) <sup>-1</sup>	{ 1.12 [14] 1.16 [19] 1.20 [18, 20]	1.17 ± 0.15* ± 0.03

\*Obtained by comparison within the framework of expression (1) of the experimental susceptibility at  $T = 0$  K to the most vigorous calculation of  $N(E_F)$  (Ref. 21). The error factor corresponds to the spread of calculated  $N(E_F)$ .

\*\* $J = (N_A/A)\mu_B I$ ,  $N_A$  is the Avogadro number,  $A$  is the atomic weight.

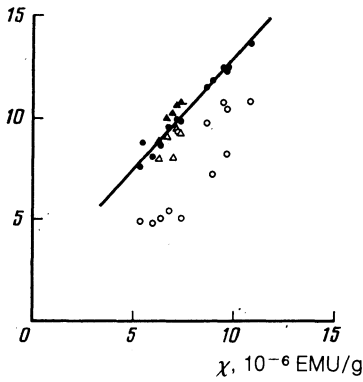


FIG. 5. The magnetovolumetric effect  $d \ln \chi / d \ln V$  (open symbols) and its deformation component  $d \ln \chi / d \ln V - (\partial \ln \chi / \partial q)(dq / d \ln V)$  (solid symbols) plotted as a function of magnetic susceptibility at  $T = 78$  K in binary (circles) and ternary (triangles) palladium alloys.

$\partial q)_{\Gamma = \text{const}}$  for a specific alloy with given  $q$  and  $\Gamma$  (the details of this routine are described in Ref. 5).

The behavior of the  $\partial \ln \chi / \partial q$  derivatives for the alloys under analysis in the present study at various temperatures is shown in Fig. 4 (their dependence on  $q$  was selected to simplify comparison and use). It is clear that all compositions under analysis belong to the decaying section of the state density curve  $N(E)$ . This curve has an inflection point just below the Fermi level in palladium and a maximum ( $\partial \ln \chi / \partial q = 0$ ) near 9.8 el./at. predicted by *a priori* theoretical calculations of the electron spectrum of palladium.<sup>1,17</sup> The quantity

$$\frac{\partial \ln N(E_F)}{\partial q} = S^{-1} \frac{\partial \ln \chi}{\partial q}$$

for  $T = 0$  K for pure palladium is in good agreement with theory if we set  $S = 9$  (see Table II). This agreement supports the reliability of these derivatives and indirectly supports a constant value of the parameter  $I$  in alloys.

Figure 5 plots  $d \ln \chi / d \ln V$  as a function of  $\chi$  for  $T = 78$  K. The data from Ref. 15, 22, 23 containing a correction for the parasitic contribution of the localized moments, were used as  $\chi$ . Also given here is the least-squares linearization of

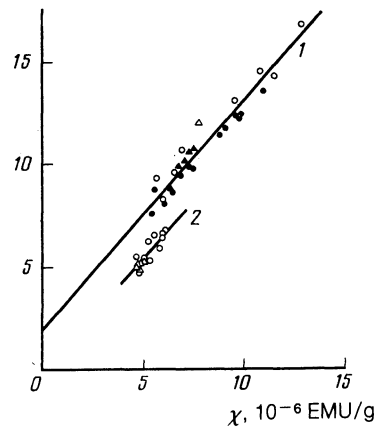


FIG. 6. The deformation contribution  $d \ln \chi / d \ln V - (\partial \ln \chi / \partial q)(dq / d \ln V)$  at various temperatures plotted as a function of the magnetic susceptibility in palladium alloys and its linear approximation: 1—Data for  $T = 20.4$  K (open symbols) and  $T = 78$  K (solid symbols); 2—data for  $T = 293$  K. The circles represent binary alloys while the triangles represent ternary alloys.

the dependences of the deformation contribution (6) on  $\chi$  by an exhaustive search of the electron overflow parameter  $dq / d \ln V$  using the data from Fig. 4. It is possible to represent the experimental results at all temperatures analogously (Fig. 6). At 20.4 and 78 K they are described by nearly the same curve. The corresponding value of  $dq / d \ln V$  and estimate of  $d \ln \Delta_d / d \ln V$  by the point of intersection of the curve with the  $X$ -axis are shown in Table III. The data are down-shifted in qualitative agreement with (6) at  $T = 293$  K, although the shift exceeds the anticipated value of  $5/3 \partial \ln \chi / \partial \ln T$ , which is  $-1.1$  for palladium.

The parameter  $dq / d \ln V$  in palladium can be estimated independently by the change under pressure in the Fermi surface cross-sections belonging to the nonhybridized spectral states. Specifically, the following relation follows from model (3) for the hole ellipsoid centered at point  $X$  in the Brillouin zone<sup>12</sup>:

$$\frac{dq}{d \ln V} = -\frac{2}{3} \xi N_d \left( \frac{d \ln \Omega}{d \ln V} + 1 \right). \quad (7)$$

TABLE III. Volumetric derivatives of parameters characterizing the spectrum and electron-electron interaction in palladium.

Parameter	Theory	Experiment
$-\frac{d \ln \Delta_d}{d \ln V}$	$\left\{ \begin{array}{l} 1.64 \text{ [24]} \\ 1.69 \text{ [19]} \\ 1.6 \text{ [20]} \end{array} \right.$	$2 \pm 1$
$\frac{d \ln N(E_F)}{d \ln V}$	$\left\{ \begin{array}{l} 1.23 \text{ [25]} \\ 1.22 \text{ [20]} \\ 1.12 \text{ [26]} \end{array} \right.$	$1.05 \pm 0.15$
$\frac{dq}{d \ln V}, (\text{el./at.})^{-1}$	$\left\{ \begin{array}{l} 0.3 \text{ [26]} \\ 0.2 \text{ [19]} \\ 0.6 \text{ [20]} \end{array} \right.$	$0.40 \pm 0.04$ $0.34 \pm 0.1^*$
$\frac{d \ln \chi(0)}{d \ln V}$	$\left\{ \begin{array}{l} 5.1^{**} \\ 5.4 \text{ [27]} \end{array} \right.$	$5.25 \pm 0.4$ $5.2 \pm 0.2 \text{ [7]}$ $4.5 \text{ [6]}$ $3.0 \pm 0.5 \text{ [2]}$
$\frac{d \ln I}{d \ln V}$	$-0.2 \text{ [27]}$	$-0.7 \pm 0.15$
$\frac{d \ln S}{d \ln V}$	$-0.18 \text{ [20]}$	
	$3.9^{**}$	$4.2 \pm 0.5$

\*Based on de Haas-van Alphen effect data under pressure.<sup>25</sup>

\*\*Estimate based on corresponding calculation data.<sup>20</sup>

With known values of the chemical potential for the ellipsoid  $\xi$ ,<sup>1</sup> the partial state density in the  $d$ -band  $N_d$  (Ref. 1) and the pressure dependence of the ellipsoid volume  $\Omega$  (Ref. 25) expression (7) yields a result similar to that obtained from the magnetic properties (Table III). Finally substitution of the values  $dq/d \ln V$ ,  $\partial \ln N(E_F)/\partial q$  and  $d \ln \Delta_d/d \ln V = -5/3$  into (3) yields  $d \ln N(E_F)/d \ln V$  for palladium, in agreement with *a priori* calculations.<sup>20,25,26</sup> The experimental parameter values given in Table III together with the theoretical broadening of the  $d$ -band therefore reliably describe the transformation of the electron state of palladium alloys under pressure and are correctly accounted for in their magnetic properties.

The parameter values from Tables II and III entering into expression (6) and the slope of curve 1 in Fig. 6 equal to  $(1.13 \pm 0.1) \cdot 10$  g/EMU are used to determine the derivative  $d \ln I/d \ln V$  (Table III). Accounting for the orbital paramagnetism—the greatest of the dropped contributions to the total palladium susceptibility<sup>28</sup>—and its pressure dependence<sup>12</sup> will reduce the absolute value of the derivative by less than 0.1, which is within the estimated error.

As we see from Table III the value of  $d \ln I/d \ln V$  found is substantially different from that calculated based on the functional of the local electron density<sup>20,27</sup> which describes  $I$  with sufficient accuracy both in palladium (Table II) and other metals<sup>14</sup>; the results from the study confirm the independence of  $I$  and  $d \ln I/d \ln V$  of the fine components of the electron spectrum. The observed  $d \ln I/d \ln V$  has the same relation to calculation<sup>29</sup> in palladium as in vanadium,<sup>3</sup> i.e., the discrepancy is fundamental. It is important to avoid confusion regarding the apparent correlation between the calculated<sup>20,27</sup> and experimental values of  $d \ln \chi/d \ln V$  in palladium, since in calculations<sup>20</sup> the differential between  $d \ln I/d \ln V$  and the experimental value is balanced by the difference of the calculated and experimental values of the Stoner factor (Table II), while the result of Ref. 27 does not follow from the calculated values of the quantities determining the result provided in this study.

The only band structure parameter that correlates with the behavior of  $I$  in palladium is the width of the  $d$ -band:

$$d \ln I/d \ln \Delta_d = 0.42 \pm 0.1.$$

Such a correlation is characteristic of the intraatomic electron interactions in the narrow bands.<sup>30</sup> In the simplest case<sup>30,31</sup> the intraatomic repulsion energy  $U$  of electrons with antiparallel forces dominates; this breaks down in metal due to screening<sup>30</sup>:

$$U_{\text{eff}} = U(1 - kU)^{-1}, \quad (8)$$

where  $k \sim \Delta_d^{-1}$ .  $N^*$  represents the density of states assigned to the spin and one of the three degenerate orbitals at the ceiling of the  $d$ -band, i.e.,  $N^* \approx 1/6 N(E_F)$  (Ref. 30). Taking into account that  $d \ln U/d \ln V = 0$  we find

$$d \ln I/d \ln \Delta_d = 1 - U_{\text{eff}}/U. \quad (9)$$

The resulting estimates of  $U_{\text{eff}}/U = 0.6$ ,  $U \approx 0.27$  Ry,  $U \approx k^{-1} \approx \Delta_d$  are of a reasonable order of magnitude,<sup>31</sup> although the lack of a reliable value of  $U$  either calculated or found independently does not allow determination of the possibility for a quantitative description of the exchange-correlated effects within the framework of the approach of

Ref. 30. Based on the strong coupling approximation, this approach is justified to some degree in palladium, i.e., in the case of a near-filled  $d$ -band when the concentration of  $d$ -holes is small and the degree of localization of their wave functions increases. It is far from clear whether or not it is suitable for vanadium. Further theoretical and experimental investigations of the problem of interelectron interactions in metals are of the highest importance.

A common property of the test metals, including certain intermetallic compounds,<sup>32–34</sup> is the relatively weak dependence of the Stoner factor on pressure due to the cancellation of variations in the exchange-correlation parameter  $I$  and the electron state density. This is what complicates the process of achieving ferromagnetism in Pd by varying the atomic volume and necessitates searching for the cause of the strong growth in the susceptibility of a Au–Pd–Au metal-film sandwich with homogeneous deformation of the epitaxial Pd layer<sup>35</sup> or in the specific nature of the film state of Pd itself.<sup>36</sup> The data provided in the present study will help to refine the range of ferromagnetism in the phase diagram of palladium alloys.

As anticipated<sup>4</sup> the investigation of  $d \ln \chi/d \ln V$  in palladium alloys at low temperatures yields more reliable values of parameters describing the behavior of the energy spectrum and electron interaction. We succeeded, albeit with only moderate accuracy, in independently isolating the broadening effect of the  $d$ -band whose role is suppressed as the electron degeneration temperature is approached. The actual suppression in the range 20–300 K (the relative vertical shift of curves 1 and 2 in Fig. 6) turned out to be greater than that predicted by the band model (5). This could be attributed to ignoring the temperature dependence of the chemical potential in (5) or the orbital contribution influencing the magnetism of the thermal lattice vibrations as well as spin fluctuations. In any case this fact, together with the existing exaggeration of the local deformability of the state density peak of Pd, explains the difference between the numerical estimates of the slope of curve 2 and  $d \ln I/d \ln V$  in Ref. 5 and the data from the present analysis. A study similar to Ref. 5 was carried out in Ref. 16; the latter study employed low-temperature magnetostriction data and the model spectrum of Pd–Rh alloys. For the reasons discussed above the results from Ref. 16 are largely qualitative.

In conclusion we note that the methods employed to account for changes in the electron state of palladium due to pressure and impurities may also be useful for other systems in analyzing a variety of thermodynamic properties associated with variations in the atomic volume.

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<sup>1</sup>O. K. Andersen, Phys. Rev. B. 2, 883 (1970).

<sup>2</sup>W. Gerhardt, F. Razavi, J. S. Schilling *et al.*, Phys. Rev. B. 24, 6744 (1981).

<sup>3</sup>A. S. Panfilov, Yu. Ya. Pushkar', and I. V. Svechkarev, Fiz. Niz. Temp. 14, 532 (1988) [Sov. J. Low Temp. Phys. 14, 532 (1988)].

<sup>4</sup>I. Ya. Dekhtyar, A. S. Panfilov, I. V. Svechkarev, and R. G. Fedchenko, Zh. Eksp. Teor. Fiz. 60, 340 (1971) [Sov. Phys. JETP 33, 185 (1971)].

<sup>5</sup>V. N. Mapchenko, A. S. Panfilov, and I. V. Svechkarev, Zh. Eksp. Teor. Fiz. 71, 2126 (1976) [Sov. Phys. JETP 44, 1118 (1976)].

<sup>6</sup>R. Keller, J. Ortelli, and M. Peter, Phys. Lett. A. 31, 376 (1970).

<sup>7</sup>V. Pluzhnikov and E. Fawcett, J. Phys. F. 12, 1467 (1982).

- <sup>8</sup>M. B. Brodsky, *J. Appl. Phys.* **52**, 1665 (1981).
- <sup>9</sup>D. K. Hsu and R. G. Leisure, *Phys. Rev. B* **20**, 1339 (1979).
- <sup>10</sup>K. A. Gschneidner Jr., *Solid State Physics Vol. 16*, edited by F. Seitz and D. Turnbull, N.Y.: Acad. Press, 275 (1964).
- <sup>11</sup>E. Walker, J. Ortelli, and M. Peter, *Phys. Lett. A* **31**, 240 (1970).
- <sup>12</sup>I. V. Svechkarev and A. S. Panfilov, *Phys. Stat. Sol. (b)* **63**, 11 (1974).
- <sup>13</sup>V. Heine, *Phys. Rev.* **153**, 673 (1967).
- <sup>14</sup>J. F. Janak, *Phys. Rev. B* **16**, 255 (1977).
- <sup>15</sup>A. Hahn and W. Treutmann, *Z. Angew. Phys.* **26**, 129 (1969).
- <sup>16</sup>S. Hirooka and M. Shimizu, *J. Phys. Soc. Jap.* **43**, 477 (1977).
- <sup>17</sup>F. M. Mueller, A. J. Freeman, J. O. Dimmock, and A. M. Furdyna, *Phys. Rev. B* **1**, 4617 (1970).
- <sup>18</sup>A. H. MacDonald, J. M. Daams, S. H. Vosko, and D. D. Koelling, *Phys. Rev. B* **23**, 6377 (1981).
- <sup>19</sup>D. G. Pettifor, *J. Phys. F* **7**, 613 (1977).
- <sup>20</sup>L. Fritsche, J. Noffke, and H. Eckardt, *J. Phys. F* **17**, 943 (1987).
- <sup>21</sup>K. L. Liu, A. H. MacDonald, J. M. Daams *et al.*, *J. Magn. and Magn. Mater.* **12**, 43 (1979).
- <sup>22</sup>A. J. Manuel and J. M. P. St Quinton, *Proc. Roy. Soc. A* **273**, 412 (1963).
- <sup>23</sup>F. E. Hoare, J. C. Matthews, and J. C. Walling, *Proc. Roy. Soc. A* **216**, 502 (1953).
- <sup>24</sup>G. M. Stocks, R. W. Williams, and J. S. Faulkner, *J. Phys. F* **3**, 1688 (1973).
- <sup>25</sup>H. Skriver, W. Venema, E. Walker, and R. Griessen, *J. Phys. F* **8**, 2313 (1978).
- <sup>26</sup>S. G. Das, D. D. Koelling, and F. M. Mueller, *Sol. St. Comm.* **12**, 89 (1973).
- <sup>27</sup>O. K. Andersen, J. Madsen, U. K. Poulsen *et al.*, *Physica. B* **86-88**, 249 (1977).
- <sup>28</sup>J. Benkowitsch and H. Winter, *Z. Phys. B* **50**, 217 (1983).
- <sup>29</sup>Y. Ohta and M. Shimizu, *J. Phys. F* **13**, L123 (1983).
- <sup>30</sup>J. Kanamori, *Progr. Theor. Phys.* **30**, 275 (1963).
- <sup>31</sup>N. D. Lang and H. Ehrenreich, *Phys. Rev.* **168**, 605 (1968).
- <sup>32</sup>L. S. Litinskaya, A. S. Panfilov, Yu. Ya. Pushkar, and I. V. Svechkarev, *Inst. Phys. Conf. Ser.*, No. 55, 185 (1981).
- <sup>33</sup>N. Buis, J. J. M. Franse, and P. E. Brommer, *Physica. B + C* **106**, 1 (1981).
- <sup>34</sup>J. J. M. Franse, P. H. Frings, F. R. de Boer, and A. Menovsky, *Physics of Solids Under High Pressure*, edited by J. S. Schilling and R. N. Shelton, North-Holland Publ. Co., 181 (1981).
- <sup>35</sup>T. Jarlborg and A. J. Freeman, *Physica. B* **107**, 69 (1981).
- <sup>36</sup>Y. Takahashi and M. Shimizu, *J. Phys. F* **15**, 71 (1985).

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