

Linde's Law and the Scattering Power of Associated Point Defects*

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Resistivity data taken in dilute alloys indicates that the scattering power of a point defect is relatively unaffected by the proximity of other defects. The scattering power of defect pairs as a function of separation is therefore investigated as a two-center scattering problem, and the theory is found to predict the observed insensitivity of scattering cross section to defect separation for all realizable separations of the defects.

I. INTRODUCTION

IN the interpretation of resistivity data obtained during quenching experiments, it has been surmised that the scattering cross section of a complex consisting of several defects is equal to the sum of the cross sections of the constituent point defects. Thus, for example, the resistivity associated with α atomic percent of divacancies has been taken as equal to 2α atomic percent of single vacancies.

The validity of this assumption is not at all clear on first examination since the resistivity due to point defects is well known to obey the equation¹

$$\Delta R = \frac{4\pi c}{k_F} \sum_{l=0}^{\infty} (l+1) \sin^2(\eta_l - \eta_{l+1}), \quad (1)$$

where c is the concentration, k_F the Fermi wave vector of the solvent lattice and η_l is the phase shift of an l -wave electron at the Fermi surface provoked by the scattering effect of a defect. One might speculate that with two defects in a small neighborhood of the lattice, the phase shifts induced in a particular partial wave would be roughly additive, so that the scattering power of a double defect would, by virtue of Eq. (1), be of the form of the square of the sum of the phase shifts of the individual defects. For like defects, one would therefore anticipate the resistivity to double if the defects associated into pairs.

That this is not the case is evident from resistivity data taken on dilute alloy systems, where a linear dependence of resistivity on solute concentration, is found up to concentrations at which it is certain that a considerable number of solutes occupy sites adjacent to other solute atoms. The objective of the present paper is to investigate the resistivity due to pairs of defects in the lattice as a function of their separation and in this way to demonstrate the physical mechanisms behind the approximate additivity of solute scattering power.

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¹ P. de Faget de Castrejlau and J. Friedel, *J. phys. radium* **17**, 27 (1956).

II. WAVE FUNCTIONS IN THE NEIGHBORHOOD OF A DEFECT

Following the work of Friedel *et al.*,^{2,3} it is now well established that valence differences between solute cells and the solvent lattice are screened out inside the solute cell, the resulting disturbance $\Delta\rho(\mathbf{r})$ of the electron density $\rho(\mathbf{r})$ in the surrounding lattice taking an oscillatory form which dies away with increasing distance from a point defect approximately as⁴

$$\Delta\rho(\mathbf{r}) \sim \rho(\mathbf{r}) \cos(2k_F r + \varphi_0), \quad (2)$$

φ_0 being a phase-shift dependent on the scattering power of the defect.

For the purpose of calculating resistivity data, it is convenient to treat the electron gas in terms of spherical eigenstates

$$\Psi_{lmn}(k_F, \mathbf{r}) = \psi_k(r_s) j_l(k_F r_1) P_l(\cos\theta) e^{im\varphi} \quad (3)$$

centered around the scattering cell. Here, $j_l(k_F r_1) \times P_l^m(\cos\theta) e^{im\varphi}$ is the usual solution of the free-electron wave equation for a spherical box, and $\psi_k(r_s)$ is the Wigner-Seitz wave function which makes the free electron waves into approximate Bloch waves, described by Eq. (3).

If one considers the electron gas at a lattice site neighboring a defect,⁵ then the eigenstates which describe the density are found to be mixed states

$$\Phi_n(k_F, \mathbf{r}) = \sum_{l,m} \gamma_{lmn}(k_F, r_1) \Psi_{lm}(k_F, \mathbf{r}), \quad (4)$$

with coefficients $\gamma_{lmn}(k_F, r_1)$ exhibiting an oscillatory decay to unity with increasing distance r_1 from the scattering center. However, the form of the mixing is such that the quantity

$$|\gamma_{lm}(k_F, r_1)|^2 = \sum_n |\gamma_{lmn}(k_F, r_1)|^2 \quad (5)$$

of any given partial wave in the electron gas near the defect site is very little different from that to be anticipated in the absence of the defect. Thus, in nuclear resonance experiments for example, the Knight shift⁶ found is only a few percent different from the

² J. Friedel, *Phil. Mag.* **43**, 152 (1952).

³ J. Friedel, *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1954), Vol. 3, p. 446.

⁴ W. Kohn and S. H. Vosko, *Phys. Rev.* **119**, 912 (1960).

⁵ C. P. Flynn, *Phys. Rev.* **125**, 881 (1962).

⁶ L. E. Drain, *Phil. Mag.* **4**, 484 (1959).

perfect lattice value even for nuclei in first-neighboring sites to a large valence solute.^{6,7} Thus, the s -wave content of the electron gas is virtually unaffected, as also are higher-order waves.

One sees, therefore, that a second defect situated in the proximity of a scattering center will be subject, at least at the Fermi surface, to a distribution of incident electrons which remains almost unmodified by the neighboring scattering center. The implication, therefore, is that the electron scattering by a defect is affected little by the presence of a second defect.

Nevertheless, one cannot assume that the scattering cross sections of the two defects simply add linearly to produce a total cross section, for it will be recalled that the partial waves of an electron scattered radially from the defect region will be additive, and since the scattering cross section is proportional to the square of these quantities, the resistivities would not be expected to add linearly in the manner experimentally observed.

III. SCATTERING BY DEFECT PAIRS

In order to find the scattering cross section of defect pairs, one must treat the scattering as a two-center problem. In accordance with the discussion in the previous section, the defects will be considered to scatter independently, the asymptotic form of the scattered waves resulting from an incident wave of unit amplitude

$$\psi_{\text{inc}} = \psi_{k_F}(r) e^{i\mathbf{k}_F \cdot \mathbf{r}},$$

being

$$\psi_1(r_1) = \psi_{k_F}(r) f_1(\theta_1) e^{ik_F r_1 / r_1} \quad (6)$$

and

$$\psi_2(r_2) = \psi_{k_F}(r) f_2(\theta_2) e^{ik_F r_2 / r_2},$$

$f_1(\theta)$ and $f_2(\theta)$ being the scattering amplitudes exhibited by the separate defects. As indicated in Fig. 1, the θ and r are scattering angles and radius vectors from the scattering centers to a point P with the subscripts distinguishing between the two defects. Also shown are the angles α and β , the former being the angle between the wave vector \mathbf{k}_F and the axis \mathbf{a} joining the defects, and β representing the angle between this axis and the scattering direction θ . We will use φ to denote the angle between the plane containing \mathbf{k}_F and \mathbf{a} and the plane containing \mathbf{r} and \mathbf{k}_F .

The asymptotic form of the complete wave may be written down for large r in the form

$$\psi = \psi_{k_F}(r) [e^{i\mathbf{k}_F \cdot \mathbf{r}_1} + (1/r_1) f_1(\theta_1) e^{ik_F r_1} + (1/r_2) f_2(\theta_2) e^{ik_F r_2}], \quad (7)$$

which, as P becomes remote from the defect sites, tends to

$$\psi = \psi_{k_F}(r) [e^{ik_F r \cos\theta} + (e^{ik_F r / r} \{ f_1(\theta) + f_2(\theta) e^{ik_F a (\cos\alpha - \cos\beta)} \})], \quad (8)$$

⁷ T. J. Rowland, Phys. Rev. **125**, 459 (1962).

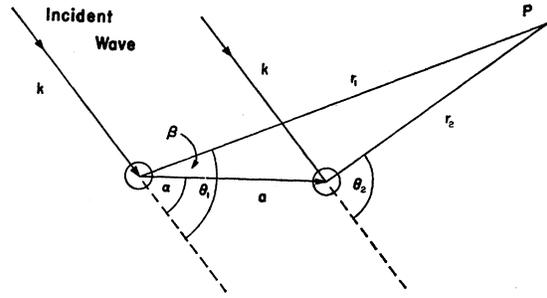


FIG. 1. The geometry of the scattering around a double defect.

with

$$\theta_1 = \theta_2 = \theta$$

and

$$r = r_1 = r_2 + a \cos\beta.$$

From Eq. (8) we can write down the differential scattering cross section of the defect pair:

$$|f(\theta)|^2 = |f_1(\theta)|^2 + |f_2(\theta)|^2 + \Delta|f(\theta)|^2, \quad (9)$$

where

$$2\Delta|f(\theta)|^2 = f_1(\theta) f_2^*(\theta) e^{ik_F a (\cos\alpha - \cos\beta)} + \text{c.c.},$$

is clearly the change in differential scattering cross section associated with the proximity of the defects, since the first two terms are the cross sections for the isolated defects. For spherically symmetric scattering centers producing phase shift η_l and ζ_n , respectively, in spherical waves of the type (3), the functions $f_1(\theta)$ and $f_2(\theta)$ are known to take the form⁸

$$f(\theta) = \frac{1}{k_F} \sum_{l=0}^{\infty} (2l+1) \exp(i\eta_l) \sin\eta_l P_l(\cos\theta), \quad (10)$$

so that the interaction part of the scattering cross section is

$$\begin{aligned} \Delta|f(\theta)|^2 = & \frac{1}{k_F^2} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} (2l+1)(2n+1) P_l(\cos\theta) P_n(\cos\theta) \\ & \times \sin\eta_l \sin\zeta_n [\cos(\eta_l - \zeta_n) \cos\{k_F a (\cos\alpha - \cos\beta)\} \\ & + \sin(\eta_l - \zeta_n) \sin\{k_F a (\cos\alpha - \cos\beta)\}]. \quad (11) \end{aligned}$$

The value of $\Delta|f(\theta)|^2$ is a function of the angles α and β as well as θ . In the bulk metal, the incident wave will be scattered by many defect pairs oriented in random directions to the wave vector. In order to continue with the calculation it is expedient to remove this dependency by obtaining an average cross section change due to the interaction of the defects, namely $\langle \Delta|f(\theta)|^2 \rangle_{\text{av}}$. This may be effected by means of the

⁸ P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill Book Company, Inc., New York, 1953).

standard substitutions

$$e^{ix \cos \xi} = \sum_{l=0}^{\infty} i^l (2l+1) P_l(\cos \xi) j_l(x) \quad (12)$$

and

$$P_l(\cos \xi \cos \gamma + \sin \xi \sin \gamma \cos \varphi) = \sum_{m=0}^l \frac{\epsilon_m (l-m)!}{(l+m)!} P_l^m(\cos \xi) P_l^m(\cos \gamma) \cos m \varphi. \quad (13)$$

On performing the averaging integrals the following expression is obtained for the average differential cross section due to the interaction:

$$\langle \Delta |f(\theta)|^2 \rangle_{\text{av}} = \frac{1}{k_F^2} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} (2l+1)(2n+1) P_l(\cos \theta) \times P_n(\cos \theta) \delta(\eta_l, \zeta_n) \sum_{m=0}^{\infty} (2m+1) P_m(\cos \theta) j_m^2(k_F a), \quad (14)$$

where

$$\delta(\eta_l, \zeta_n) = \sin \eta_l \sin \zeta_n \cos(\eta_l - \zeta_n).$$

Hence, making use of the relationship

$$\sum_{m=0}^{\infty} (2m+1) P_m(\cos \theta) j_m^2(k_F a) = \frac{\sin[2k_F a \sin(\theta/2)]}{2k_F a \sin(\theta/2)} = A(k_F a, \theta), \quad (15)$$

we obtain the final form

$$\langle \Delta |f(\theta)|^2 \rangle_{\text{av}} = \frac{1}{k_F^2} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} (2l+1)(2n+1) P_l(\cos \theta) \times P_n(\cos \theta) \delta(\eta_l, \zeta_n) A(k_F a, \theta). \quad (16)$$

The weighted cross section which is appropriate to the momentum transfer associated with resistivity is

$$Q = \int_0^{\pi} \langle \Delta |f(\theta)|^2 \rangle_{\text{av}} (1 - \cos \theta) \sin \theta d\theta, \quad (17)$$

and using the value of $\langle \Delta |f(\theta)|^2 \rangle_{\text{av}}$ giving in Eq. (16), the value of Q may be obtained analytically in useful form. It is interesting, first, to observe the behavior of Q in the limits of large and small $k_F a$. When $k_F a$ is small (say $k_F a < 1$) the value of $A(k_F a, \theta)$ is essentially unity, so that the value of $\langle \Delta |f(\theta)|^2 \rangle_{\text{av}}$ becomes

$$\langle \Delta |f(\theta)|^2 \rangle_{\text{av}} = \frac{1}{k_F^2} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} (2l+1)(2n+1) \times P_l(\cos \theta) P_n(\cos \theta) \delta(\eta_l, \zeta_n). \quad (18)$$

The expression for the total scattering power of the defect pair is identical with Eq. (18), provided $\delta(\eta_l, \zeta_n)$

is replaced by

$$\delta'(\eta_l, \zeta_n) = |f_1(\theta)|^2 + |f_2(\theta)|^2 + \delta(\eta_l, \zeta_n) = \sum_{q,p=1}^2 \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \sin \xi_{pl} \sin \xi_{qn} \cos(\xi_{pl} - \xi_{qn}), \quad (19)$$

where $\xi_{1l} = \eta_l$ and $\xi_{2l} = \zeta_l$, and use has been made of the standard equation for the cross section of a single spherical defect, namely,

$$|f(\theta)|^2 = \frac{1}{k_F^2} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} (2l+1)(2n+1) P_l(\cos \theta) P_n(\cos \theta) \times \sin \eta_l \sin \eta_n \cos(\eta_l - \eta_n). \quad (20)$$

Thus, $\delta(\eta_l, \zeta_n)$ is a perfect cross term between the cross sections of the separate defects, and the total resistivity for similar defects and small $k_F a$

$$\Delta R = \frac{16\pi c}{k_F} \sum_{l=0}^{\infty} (l+1) \sin^2(\xi_l - \xi_{l+1}), \quad (21)$$

as anticipated in the Introduction.

The behavior of $\delta(\eta_l, \zeta_n)$ at large values of $k_F a$ is dominated by the shrinking of $A(k_F a, \theta)$ by a factor $k_F a$. Because of the zero value of $(1 - \cos \theta)$ at $\theta = 0$, this results in a decay of amplitude $\sim (k_F a)^{-2}$ with increasing $k_F a$.

The integration involved in Eq. (17) has been performed to find the coefficients of $\delta(\eta_0, \zeta_0)$, $\delta(\eta_1, \zeta_1)$, and $\delta(\eta_0, \zeta_2)$ as a function of $k_F a$. These two coefficients are indicative of the form of the higher-order terms whose computation is made unnecessary by the rapid convergence of the phase shifts with increasing order.

The coefficients may be obtained in the form

$$\begin{aligned} \alpha_{00}(\mu) &= (8/\mu^4)[2\mu \sin \mu + (2 - \mu^2) \cos \mu - 2], \\ \alpha_{11}(\mu) &= (24/\mu^8)[(2880 - 1344\mu^2 + 74\mu^4 - \mu^6) \cos \mu \\ &\quad + (2880\mu - 384\mu^3 + 10\mu^5) \sin \mu \\ &\quad - 2880 - 96\mu^2 - 2\mu^4], \end{aligned} \quad (22)$$

$$\alpha_{02}(\mu) = -\frac{1}{2}\{\alpha_{00}(\mu) - \frac{1}{3}\alpha_{11}(\mu)\}, \quad (23)$$

with $\mu = 2k_F a$.

Expressions for the higher terms take the form of longer polynomials in μ multiplied by trigonometric functions as in Eqs. (22) and (23).

Figure 2 shows the coefficients of $\delta(\eta_0, \zeta_0)$, $\delta(\eta_1, \zeta_1)$, and $\delta(\eta_0, \zeta_2)$ plotted against μ . It will be observed that the amplitude undergoes a zero before reaching the first-neighboring site and thereafter oscillates with rapidly decreasing amplitude. There are therefore no sites at which the resistivity of a defect pair is different by more than a few percent from the resistivity associated with the separated defects, in agreement with the resistivity data obtained in dilute alloys. It will also be evident that in solvents of higher valence where the Fermi wave vector is larger with respect to the reciprocal lattice vector than in monovalent metals, the effect of

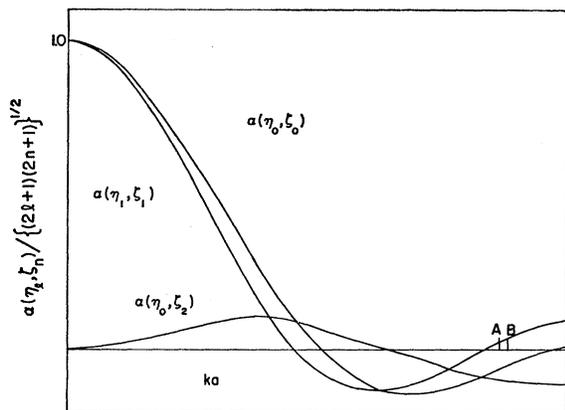


FIG. 2. The variation of various interaction terms with defect separation. The point *A* marks the interatomic distance in fcc lattices, and *B* that for bcc lattices.

the interaction term on the resistivity will be reduced still further.

IV. DIVACANCIES

While the results of the preceding treatment are readily applicable to dilute alloys, it is still difficult to make any precise statement as to the effect on residual resistance of the association of vacancies into multiple voids, since, in these cases, the scattering power is closely related to the considerable relaxation of the lattice around the defect area.

In the case of divacancies, it appears likely that the relaxation predominantly concerns the four common nearest neighbors of the two vacant sites.⁹ If the net relaxation around a divacancy is about twice that of a single vacancy, the form of the relaxation makes it reasonable to treat the divacancy as two vacancies whose individual scattering power is preserved, separated by roughly the undisturbed nearest neighbor distance. According to the results of Sec. III, the resistivity due to the double defect will then be close to twice that due to a single vacancy. However, this conclusion depends upon the validity of the conditions of lattice relaxation specified above. In general, it would be anticipated that for a smaller relaxation of the double defect, the resistivity would increase above that for the two single defects, and vice versa.

It is interesting to compare these conclusions with those obtained in other calculations of divacancy scattering power.

Asdente and Friedel¹⁰ have studied the resistivity due to spherical lattice voids of varying size, and find that for voids of $\sim 10^4$ vacancies, the scattering cross section tends to the classical value provided that proper account is taken of the charge repelled from the defect site. For small spherical voids corresponding

⁹ S. Yoshida and J. S. Koehler, *Acta Met.* **8**, 878 (1960).

¹⁰ M. Asdente and J. Friedel, *J. Phys. Chem. Solids* **11**, 115 (1959).

in volume to less than three or so vacancies, they find that the resistivity is almost proportional to the volume, with only a slight maximum at about twice the atomic volume. Thus they find, essentially, that for voids of this small size, the resistance is proportional to the number of vacancies contained, in agreement with the discussion of Sec. III.

Seeger¹¹ has performed calculations of the resistivity due to divacancies for two different assumptions concerning the shape of the defect. In the first, the divacancy was assumed to give rise to a spherically symmetric scattering potential, and in the second the potential was taken as ellipsoidal corresponding to the asymmetry of the actual defect. The calculations yielded the same resistivity to about two percent, demonstrating an insensitivity of scattering power to the shape of the scattering center which may, in any event, be anticipated from the fact that the non-spherical structure of the defect region is fairly small with respect to the Fermi wavelength.

Neither of the calculations of Asdente and Friedel nor that of Seeger takes account of the lattice relaxation around the defect regions and therefore taken together cannot be regarded as more than indicative of the actual electron scattering occurring at a divacancy. Nevertheless, to the extent that they predict only a small resistivity change on the association of two unrelaxed vacancies to form an unrelaxed divacancy, they are in agreement with the present theory, which predicts this result for association of any pair of defects whose individual scattering potentials are not affected by the association. Unfortunately, the case of a divacancy may not be in this category, because the effective scattering potentials could well be modified by changes in lattice relaxation. Moreover, the change in relaxation may well be different for vacancies associating in different lattices so that the resistivity change on association may also vary.

V. CONCLUSION

It has been shown that the total scattering power of a pair of defects in a metal lattice contains a contribution which is a rapidly-decreasing oscillatory function of the separation of the two defects, due to the interference between the partial waves of an electron scattered from the defect region. The interference becomes an efficient source of averaging only when the defects are separated by a quarter wavelength so that in the neighborhood of $k_F a = \pi/2$, the first minimum of the interaction term is found. Thereafter, with increasing separation of the defects, the averaging remains efficient so that the interaction term is close to zero.

However, the smallest interatomic distance with respect to the reciprocal Fermi wave vector which is found in metals—that in fcc lattices of monovalent metals—is given by $k_F a = 3.47$, so that in no case will

¹¹ A. Seeger, *J. Phys. Chem. Solids* **6**, 324 (1958).

the interaction term reach more than a small fraction of its potential value which could, in principle, double the infinite separation value of the total scattering power of a pair of identical defects. Thus, the linearity up to surprisingly high concentrations of the resistivity of dilute alloys is explained in terms of the insensitivity of total scattering cross section to defect separation. Further, the theory permits the prediction that, provided that the total lattice relax-

ation associated with the defect pair is not greatly modified in the process, the joining of two lattice vacancies into a divacancy will not result in a large fractional change in residual resistance.

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Ferromagnetism in EuH_2 ^{†‡}

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Magnetic measurements have been made on a Eu-H sample of composition represented by the formula $\text{EuH}_{1.86}$ over a temperature range extending from 4.2° to 300°K. The magnetization-temperature data for the sample, which for simplicity is termed EuH_2 , indicate a Curie point at 24°K. Field dependency studies in the liquid helium range reveal saturation effects and remanence, confirming that EuH_2 has become ferromagnetic at these temperatures. The observed saturation moment at 4.2°K is $\sim 6.0\mu_B$ (Bohr magnetons) per Eu atom. For $T > 30^\circ\text{K}$ the susceptibility shows a Curie-Weiss dependence on temperature and the effective moment is $8.4\mu_B$ per Eu atom. EuH_2 , the saturated hydride of Eu, is unlike the highest (i.e., saturated) hydrides of the other lanthanide metals which have been studied— GdH_3 , TbH_3 and HoH_3 —in that it alone exhibits magnetic ordering at low temperatures. As the hydrogen-saturated metals lack conduction electrons, the coupling between the localized $4f$ electrons is expected to be very weak and magnetic ordering is expected to occur only at very low temperatures if at all. Thus the behavior of the several trihydrides is in keeping with expectation whereas that of EuH_2 is not. The coupling mechanism in this case is not well understood; it may be due to direct overlap of the $4f$ orbitals.

STUDIES of lanthanide hydrides show that when they are hydrogenated to saturation, they cease to be metallic conductors.¹⁻³ This and other properties⁴ of these materials indicate that when hydrogen is taken into the metal, it absorbs electrons from the conduction band to form the H^- anion and, moreover, in the fully hydrogenated metal the conduction band is entirely depopulated. Acquisition of the supernumerary hydrogen electron from the conduction band rather than from the core electrons is confirmed by the fact that the magneton number is unaffected by hydrogenation.

The removal of the conduction electrons in the lanthanide metals should have strong implications as regards the cooperative magnetic phenomena which occur in these materials at low temperatures, since exchange in them is generally regarded as operating via the conduction electrons. Specifically, we might expect

that alignment in the hydrides will, if it occurs at all, develop at temperatures much lower than in the corresponding element. This expectation has been confirmed for the fully hydrogenated materials HoH_3 ,⁵ TbH_3 and GdH_3 ,⁶ which give no indication of either Curie or Néel points down to 4.2°K. In addition, for HoH_2 and TbH_2 , in which the conduction band is considerably depleted, the Néel points are quite low compared to the parent metal, occurring at 8° and 45°K, respectively.⁶ After observing these gratifyingly predictable behaviors, we decided to examine the hydride of the more complex metal europium. Among the lanthanides Eu is atypical in that (1) its structure is bcc, (2) it is divalent and hence at saturation forms only a dihydride, and (3) the question of whether its moments are aligned at low temperatures (i.e., $< 10^\circ$ to 15°K) is regarded by some as being as yet unsettled.⁷

Europium was hydrogenated using techniques previously employed in this laboratory.⁴ Despite very considerable effort the maximum hydrogen content

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[‡] This work was assisted by a contract with the U. S. Atomic Energy Commission.

¹ B. Stalinski, *Bull. acad. polon. sci.* **5**, 1001 (1957) and **7**, 269 (1959).

² J. C. Warf and K. Hardcastle, Office of Naval Research, Final Report August, 1961, (unpublished).

³ T. Peltz and W. E. Wallace, (unpublished measurements).

⁴ A. Pebler and W. E. Wallace, *J. Phys. Chem.* **66**, 148 (1962).

⁵ Y. Kubota and W. E. Wallace, paper presented at the Seventh Annual Conference on Magnetism, November, 1961 and submitted to *J. Appl. Phys.*

⁶ Y. Kubota, R. L. Zanowick, and W. E. Wallace (to be published).

⁷ See, for example, R. M. Bozorth, and J. H. Van Vleck, *Phys. Rev.* **118**, 1493 (1960).