[23] W-M Shyu, K. S. Singwi, and M. P. Tosi, Phys. Rev. (to be published).

[24] E. W. Kellerman, Phil. Trans. Roy. Soc. London A238, 513 (1940).

[25] S. H. Vosko, Phys. Letters 13, 97 (1964).

[26] See, for example, W. Cochran, *Inelastic Scattering of Neutrons* (International Atomic Energy Agency, Vienna, 1965), Vol. I, p.3.

[27] L. M. Falicov and V. Heine, Advan. Phys. 10, 57 (1961).

[28] D. J. W. Geldart and S. H. Vosko, Can. J. Phys. 44, 2137 (1966).

[29] P. Nozières and D. Pines, Phys. Rev. 111, 442 (1958).

[30] R. W. Shaw and N. V. Smith, Phys. Rev. 178, 985 (1969).

[31] Values are derived from first-order perturbation theory in Ref. 30, since second-order values for these metals are not given.

[32] W. J. L. Buyers and R. A. Cowley, Phys. Rev. 180, 755 (1969).

[33] G. Dolling (private communication).

[34] F. J. Kollarits and J. Trivisonno, J. Phys. Chem. Solids 29, 2133 (1968).

[35] N. W. Ashcroft and D. C. Langreth, Phys. Rev. 155, 682 (1967).

[36] N. W. Ashcroft and D. C. Langreth, Phys. Rev. 159, 500 (1967).

[37] D. J. W. Geldart and R. Taylor, Can. J. Phys. 48, 155 (1970); 48, 167 (1970).

[38] E. J. Woll and W. Kohn, Phys. Rev. 126, 1693 (1962); J. Björkman, B. I. Lundquist, and A. Sjölander, *ibid.* 159, 551 (1967).

[39] D. C. Wallace, Phys. Rev. 182, 778 (1969).

[40] M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Oxford U. P., Oxford, England, 1954).

[41] P. Lloyd and C. A. Sholl, J. Phys. C 1, 1620 (1968).

[42] E. G. Brovman, Yu. Kagan, and A. Holas, Zh. Experim. i Teor. Fiz. 57, 1635 (1969) [Soviet Phys. JETP 30, 883 (1970)].

PHYSICAL REVIEW B VOLUME 2, NUMBER 8 15 OCTOBER 1970

Magnetization Measurements of 0, 5, 10, and 20 at.% Al- Substituted First-Transition-Series Alloys

M. J. Besnus and A. J. P. Meyer

Laboratoire Pierre Weiss, Institut de Physique, Strasbourg, France

(Received 15 July 1969; revised manuscript received 4 May 1970)

Precision measurements on saturation moments of Cr-Fe, Fe-Co, Fe-Ni, and Co-Ni alloys with 0, 5, 10, and 20 at.% Al substitutions are reported; they show linear decrease of the mean atomic moment M at low Al concentration c. The negative slopes $-dM/dc$ vary with the electron to atom ratio $\mathfrak{z}$ of the transition metal: They show a maximum near $\mathfrak{z} \simeq 8.3$, as does the Slater-Pauling curve, and decrease to $-dM/dc \simeq 3.0\mu_B/$(Al atom) near pure nickel. At concentrations close to pure Fe, there is a sharp drop to $-dM/dc = 2.2\mu_B/$(Al atom). The moment decrease due to the Al solute is found to be in qualitative accord with $M = M_0(1-c) - c(V-n_s)\mu_B$, where $-(V-n_s)\mu_B$ is the moment induced by an impurity atom with valence V in the surrounding transition matrix with free-electron density n_s. This law takes for Ni as a particular case the known form $M = M_0 - cV\mu_B$. In the (Fe-Cr)-Al series the observed magnetic behavior is correlated with electronic-specific-heat data and critical concentrations for appearance of ferromagnetism.

I. INTRODUCTION

It is well known[1] that if Ni is alloyed with a nontransition metal which has V outer electrons, the mean atomic magnetic moment M decreases with the concentration c of the solute according to $M = M_0 - cV\mu_B$. Thus in the Ni alloys with Al, which has three valence electrons, $dM/dc = -3.0\mu_B/$(Al atom). On the other hand, if Fe is alloyed with Al or Si, the observed moment is known to decrease independently of the solute valence as $dM/dc = -2.2 = -M_0$, so that the effect on M seems to be only one of dilution. These two different kinds of behavior were also recently observed by neutron scattering measurements.[2,3] These observations would seem to indicate that a magnetic defect of roughly $-V\mu_B$ is formed in a Ni matrix around an Al impurity whereas an iron matrix is nearly unaffected by substituted Al atoms, which in this case behave as magnetic vacancies. On the other hand, magnetic investigations of several (Fe-Ni)-Al series lead to dM/dc values ranging from -2.5 to $-4.0\mu_B/$(Al atom).[4,5] In some series of these alloys, the moment decrease due to the Al impurity has values which are smaller than the expected valence value of $-3.0\mu_B/$(Al atom) observed in Ni-Al alloys.

Complementary conclusions indicating the complexity of the situation were reached by Srinivasan *et al.*[6] from electronic-specific-heat measurements on bcc alloys of 3*d* transition elements with 0, 10, and 20 at.% Al. They found that the curve of density of states versus the number $\mathfrak{z}$ of outer electrons per average transition atom, is nearly unaffected by addition of 10 at.% Al. However, 20 at.% Al addition mainly produces a shift of the density-of-states curve towards higher $\mathfrak{z}$ values, so that the γ peak in Cr-Fe alloys at $\mathfrak{z} \simeq 6.4$ is shifted to $\mathfrak{z} \simeq 6.7$ The authors concluded that in the 10 at.% Al alloys, the Al electrons do not interact with the *d* band, but that in the 20 at.% Al alloys there would be a removal of electrons from the *d* band. Until now, magnetic data were not available to allow one to check this model.

In view of these different results, it seemed advisable to study systematically the effect of Al substitution on the magnetic moment of first-transition-series ferromagnets, with the hope of obtaining additional information on the *d*-band change and electronic structure of these alloys. The rate of moment variation due to Al addition was thus determined experimentally at 15 different $\mathfrak{z}$ values of ferromagnetic-transition-element alloys ranging from $\mathfrak{z} = 6.4$ for Cr-Fe to $\mathfrak{z} = 10.0$ for Ni. For most series the Al contents used were 0 and approximately 5.0, 10.0, and 20.0 at.% Al, with intermediate values in some cases. The results which are presented in Sec. II show (i) that, except for pure iron, the mean atomic moment M of the alloys is always decreased by Al addition at a rate which is faster than a simple dilution, (ii) that the initial rate of moment decrease dM/dc per added Al atom is a function of the $\mathfrak{z}$ ratio of the transition-element matrix, and that only near nickel is $-dM/dc$ approximately equal to $3.0\mu_B$/(Al atom). The results are discussed in Sec. III. It is shown that they compare favorably with a moment decrease law which accounts additively (i) for a dilution of the transition atoms and (ii) for a moment decrease due to the nontransition impurity in accordance with either of two different existing electron transfer models. The second effect is found to be zero when Fe and most probably also Cr-rich (Cr-Fe) alloys are diluted with Al.

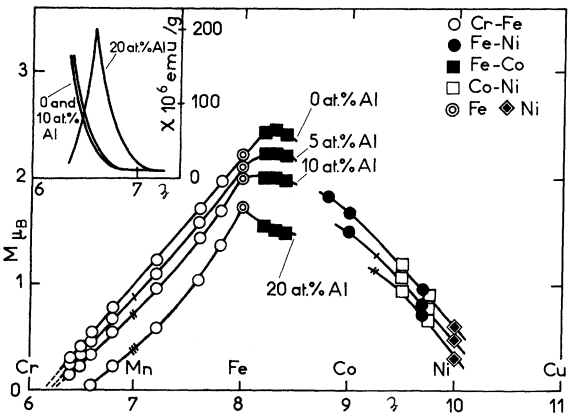

FIG. 1. Mean atomic moments M of first-transition-series alloys, without and with 5, 10, and 20 at.% Al, versus transition-metal ratio $\mathfrak{z}$ (o interpolated from data quoted in Ref. 8). Inset: Paramagnetic susceptibilities χ at 4 °K of 0, 10, and 20 at.% Al series for $6.4 < \mathfrak{z} < 7.2$.

II. EXPERIMENTAL

Alloys were prepared from at least 3*N* pure transition metals and 6*N* pure Al by induction melting under pure argon atmosphere in 99.5% pure Al_2O_3 crucibles. The ingots were annealed for 10 h at 1000 °C in vacuum and finally water-quenched when necessary in order to avoid formation of ordered phases. The samples with high Al concentrations were x-ray tested for phase homogeneity. Some (Fe-Ni)-Al samples which are recorded in Table I had to be rejected as showing appearance of bcc phase in addition to the fcc phase. The compositions were determined by weighting the constituent materials. The upper limit of weight loss at melting was 0.01%. For each alloy series of given $\mathfrak{z}$ value, the Al alloys were prepared from a single master alloy of the transition-metal components. The maximum relative error for Al composition is estimated to be less than ± 1% of the Al content. The measurements were done on prolate ellipsoidal samples with dimensions 8.0 ×4.0 mm.

The magnetizations were measured by the induction method[7] in fields up to 30 kOe at 4°K, or at 20 °K for alloys having Curie points above room temperature. Saturation magnetizations were obtained by $\sigma\,(H^{-2})$ or $\sigma^2\,(H/\sigma)$ extrapolations. The moments M are considered to have an absolute accuracy within ± 0.2%, and the relative accuracy within a given series is better than ± 0.1%. High superposed paramagnetic susceptibilities were observed in the Cr-rich (Cr-Fe) – Al alloys.

The measured average atomic moments M expressed in μ_B (moments per total number of atoms, that is, including Al atoms), are listed in Table I. They are shown in Fig. 1, together with data interpolated from previous investigations on Fe-*A*l alloys which are quoted in Ref. 8. As is seen from Fig. 1, the moment is decreased by Al addition at all $\mathfrak{z}$ values. The effect is especially great for high Al concentrations in the (Cr-Fe)-Al and (Fe-Co)-Al series.

In Fig. 2 moments have been plotted as function of Al concentration for some characteristic $\mathfrak{z}$ values. This figure shows that at constant $\mathfrak{z}$, the atomic moment M decreases linearly at low Al concentrations, whereas a departure from linearity is found at higher

TABLE I. Mean atomic moments M_0 of 3d transition-element alloys without and with various Al contents (c), and initial rate of moment change $-dM/dc$ in μ_B/(Al atom) at different electron-per-atom ratios $\mathfrak{z}$ of various transition-element alloys.

Transition-element alloy	$\mathfrak{z}$	M_0, μ_B	M, μ_B for (c) at.% Al	$-dM/dc$
($Fe_{0.2}Cr_{0.8}$)	6.40	0.293	0.255 (4.89), 0.148 (9.77)	0.78
($Fe_{0.25}Cr_{0.75}$)	6.50	0.403	0.386 (2.25), 0.333 (4.95), 0.220 (9.92)	0.75
($Fe_{0.3}Cr_{0.7}$)	6.60	0.545	0.514 (2.16), 0.457 (5.01), 0.346 (9.63), 0.054 (20.00)	1.43
($Fe_{0.4}Cr_{0.6}$)	6.80	0.777	0.673 (5.12), 0.548 (10.10), 0.413 (14.70), 0.227 (20.09)	2.03
($Fe_{0.6}Cr_{0.4}$)	7.20	1.231	1.099 (5.03), 0.958 (9.96), 0.798 (14.90), 0.597 (19.80)	2.62
($Fe_{0.8}Cr_{0.2}$)	7.60	1.720	1.585 (4.87), 1.438 (9.83), 1.044 (19.84)	2.77
($Fe_{0.9}Cr_{0.1}$)	7.80	1.967	1.967 (10.00, 1.433 (18.20)	2.70
Fe	8.00[a]			2.2
($Fe_{0.8}Co_{0.2}$)	8.20	2.432	2.248 (4.84), 2.021 (9.88), 1.546 (20.00)	3.80
($Fe_{0.7}Co_{0.3}$)	8.30	2.452	2.248 (5.05), 2.015 (9.97), 1.523 (19.96)	4.04
($Fe_{0.6}Co_{0.4}$)	8.40	2.412	2.221 (5.04), 1.991 (9.95), 1.494 (20.08)	3.79
($Fe_{0.5}Ni_{0.5}$)	9.00	1.676	1.610 (1.96), 1.503 (4.92), 2 phases (10.05), 2 phases (19.60)	3.37
($Co_{0.5}Ni_{0.5}$)	9.50	1.202	1.065 (4.96), 0.940 (9.99)	2.76
($Fe_{0.15}Ni_{0.85}$)	9.70	0.948	0.893 (1.94), 0.818 (4.81), 0.713 (9.89) 2 phases (20.02)	2.83
($Co_{0.25}Ni_{0.75}$)	9.75	0.920	0.775 (4.97), 0.670 (9.96)	2.92
Ni	10.00	0.616	0.565 (1.82), 0.464 (5.16), 0.398 (7.44)	2.94

[a] See Ref. 8.

Al contents, especially for (Fe-Cr)-Al alloys, as is also seen from Fig. 1. The slope dM/dc of the inital linear moment variation was determined graphically. The relative error of a slope is mainly determined by the relative error in Al compositions of the alloys and is estimated to lie within ± 2%.

The negatives of the initial slopes $-dM/dc$ are also listed in Table I and plotted in Fig. 3 along with values computed from other sources. In the (Fe-Ni)-Al series, our $-dM/dc$ values agree well with those deduced from the work of Bardos *et al.*[4,5] Figure 3 shows that the dM/dc values describe a single curve even in the $\mathfrak{z}$ range corresponding to different binary systems as Fe-Ni and Fe-Co, Co-Ni. It appears that the $-dM/dc$ curve exhibits a maximum near $\mathfrak{z} \simeq 8.3$ as does the Slater-Pauling curve which is the 0% Al curve of Fig. 1. Also, there is a sharp relative minimum centered on iron. The maximum value of $-dM/dc$ is approximately $4\mu_B$/(Al atom). Near nickel, the curve tends to $-dM/dc \simeq 3.0\mu_B$/(Al atom), whereas near Cr at $\mathfrak{z} \simeq 6.4$ it drops rapidly.

III. DISCUSSION

The results show first, that the moment decrease of $-3.0\mu_B$/(Al atom) anticipated from the valence of aluminum is only observed for Ni-Al. Further, they suggest that except for iron, $-dM/dc$ is always larger than the atomic moment M_0 of the matrix. Thus it appears at first sight that magnetic dilution behavior is essentially followed in Fe-Al alloys, and that at other $\mathfrak{z}$ values, the moment decrease is faster than by dilution. This leads us to assume that the moment decrease due to the Al impurity must involve (i) a dilution term accounting for the transition-metal substitution by the Al atoms and (ii) a term accounting for the change in the 3d band induced by the impurity atom. A well-known picture for this 3d-band change has been given by Mott in terms of an electron transfer in a rigid-band model. In systems like Ni-Al, following Mott's latest description,[1] (V-n_s) electrons, the difference between the V valence electrons of the solute and the free-electron density n_s of the matrix go into the unsaturated d band. We will suppose that this model applies to the ternary fcc alloys at $\mathfrak{z} \gtrsim 8.3$ with nickel-type d^- band structure where the d^+ band is thought to be filled while the d^- band remains incomplete up to $\mathfrak{z} \simeq 10.5$. If we account for the d-band contribution at a rate of one Bohr magneton per contributed electron the mean atomic moment M at constant is $\mathfrak{z}$ then given, as a function of the solute concentration c, by

$$M = M_0(1-c) - c(V - n_s)\,\mu_B. \qquad (1)$$

The second term of Eq. (1) may also describe screening effects of a local excess impurity charge ($V - n_s$) assuming that all the electrons which contribute to the screening go into the d band. As emphasized by Friedel,[9] these two concepts are the energetic and the spatial aspect of the same first-order approximation, and will therefore lead to the same second term in Eq. (1). Considering the spatial aspect of the model, Eq. (1) can be presented in a conceptual form

$$M=M_0[1-c(1+\alpha)]+\alpha c\left[M_0-\left(\frac{V-n_s}{\alpha}\right)\mu_B\right],$$

which states that αc atoms of the matrix around a nonmagnetic impurity have a mean moment M_0 decreased by a quantity proportional to $(V-n_s)$.

An alternative explanation has been proposed by Beeby.[10] Here the valence electrons from the impurity lead to an increase in the number of conduction electrons and by this to some *s*-*d* admixture which results in a reduction of *d*-band splitting through decrease in exchange strength near the impurity atom. The M (c) dependence is then given as

$$M=M_0(1-c)-ck(V-n_s)\mu_B\,, \tag{2}$$

where $k=A/3n_s$ is a function of the number of d subbands of the matrix. The numerical value of k is near unity.

In the following we will consider the first model and check in a rough approximation its validity as a function of $\mathfrak{z}$. It is immediately to be noted that for Ni, where $M_0\simeq n_s\mu_B$, Eq. (1) reduces to the known form

$$M\simeq M_0-cV\mu_B\,, \tag{3}$$

which appears as a particular case, and has thus not the extended validity which it is usually ascribed.

It is now of interest to consider the derivative of Eq. (1),

$$\frac{dM}{dc}=-M_0-(V-n_s)\mu_B\,, \tag{4}$$

and to examine its dependence on $\mathfrak{z}$. As n_s is a smooth function of $\mathfrak{z}$ and V is a constant, it appears that $-dM/dc$ will vary essentially in the same manner as the host magnetic moment M_0. Thus $-dM/dc$ will have a maximum corresponding to that of the Slater-Pauling curve at $\mathfrak{z}\simeq 8.3$. For $\mathfrak{z}>8.3$, one expects that $-dM/dc$ decreases and reaches $-dM/dc\simeq 3.0$ for nickel, as is effectively observed (see Fig. 3).

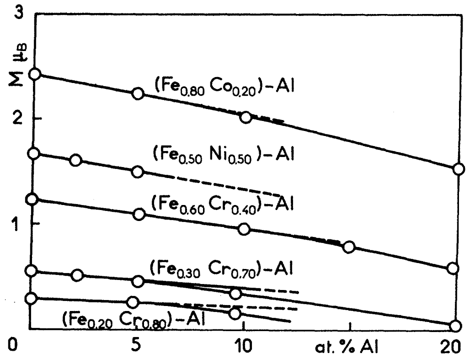

FIG. 2. Mean atomic moments M of some transition alloys series with Al substitution: the intitial moment decrease is linear.

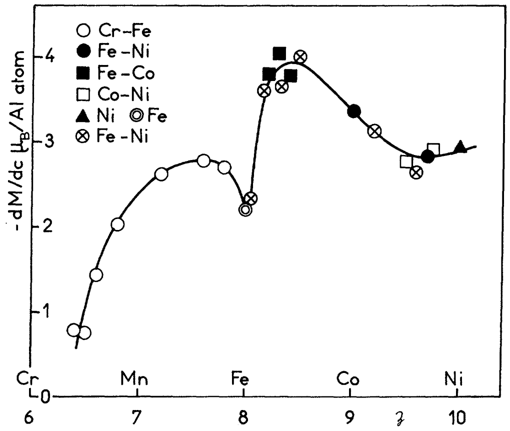

FIG. 3. Negative initial rate of change $-dM/dc$ of mean atomic moment with respect to Al concentration, plotted as a function of transition-metal ratio $\mathfrak{z}$ (⊗ calculated from Refs. 4 and 5; ○ from Ref. 8).

Figure 4 shows the apparent values of the free-electron density n_s around the impurity atoms as calculated from Eq. (4), using the experimental data of $-dM/dc$ and putting $V=3.0$. For $\mathfrak{z}>8.3$ (full line of Fig. 4), these calculated values are, excepting near Ni, significantly higher than the free-electron densities of the transition metals, which are generally admitted to increase from 0.6 for Ni to 0.9 for Fe. This may be understood on the basis of the discussed model as an increase of the electron density in nonmagnetic state in the vicinity of the Al atom. Thus the screening of the impurity charge would mainly but not completely be assured by d electrons. In Beeby's Eq. (2), the experimental data lead to k values $0.7\lesssim k\lesssim 1.0$.

In Fe-Al alloys, following the experimental facts presented in Sec. I, there is no electron transfer to the d band. Mott[1] pointed out that in this case the screening of the impurity excess electrons may be completely secured by s electrons.

In the bcc range of $6.0\lesssim\mathfrak{z}\lesssim 8.3$, according to usual concepts, the transition-element alloys have an iron-type d-band structure where both d^+ and d^- subbands are considered to be unfilled. In contrast to this model, Srinivasan *et al.*[6] propose that the Fermi level lies in one single d^+ subband at $5.8\lesssim\mathfrak{z}\lesssim 8.4$. In the usual model, the moment decrease due to Al atoms would imply a prevailing electron transfer to the minority d^- band as a consequence of the impurity screening by d electrons. As the density of states at the Fermi level is usually assumed to be much smaller in the d^- band as com-

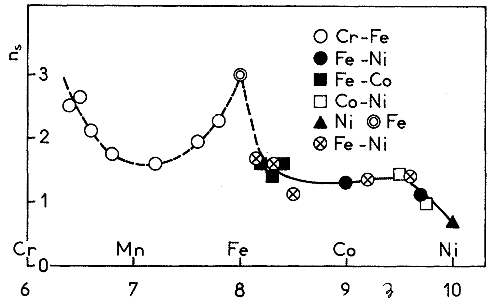

FIG. 4. From Eq. (4), calculated value n_s of apparent free-electron density in the Al impurity surrounding transition-element matrix as a function of transition-metal ratio $\mathfrak{z}$. For full line (fcc), and dotted line (bcc), see text (⊗calculated from Refs. 4 and 5; ○ from Ref. 8).

pared to the d^+ band, the accommodation of extra electrons in the d^- band should be due to a decreased d-band splitting. In this process, the filling up of the d^+ subband is not modified as shown by the electronic-specific-heat coefficients which remain constant up to 10 at.% Al addition.[6] This interpretation is now rather as in Beeby's model. The n_s value for $\mathfrak{z} \simeq 7.5$ is close to 1.5. This is again larger than the probable free-electron density of the transition metal which here should be near 1.0 since it must decrease from 2.0 at $\mathfrak{z} = 2$ to 0.6 at $\mathfrak{z} = 10$.

In the band model of Srinivasan *et al.*, the moment decrease caused by Al atoms would imply an electron removal from the only unfilled d^+ band, as pointed out by in Ref. 6 for alloys with 20 at.% Al.

Around Fe in the range $7.5 \lesssim \mathfrak{z} \lesssim 8.3$, the $-dM/dc$ anomaly is dominated by the dilution behavior of pure iron. Here Eq. (4) leads to $n_s \rightarrow V$ showing that the d band does not contribute at all to the impurity screening which, as pointed out by Mott, is secured by s electrons. As shown by the dotted line of Fig. 4, the calculated n_s values increase to 3.0 both at Fe and at $\mathfrak{z} \simeq 6.4$, leading to the idea that at $\mathfrak{z} \simeq 6.4$, the initial moment change may also be mostly of a dilution type.

The (Fe-Cr)-Al alloys show at low-temperatures high-paramagnetic susceptibilities. They have been determined as $\chi = dM/dH$ in fields $H > 10$ kOe, where in the case of large χ, the magnetization increases nearly linearly with H. These susceptibilities are plotted in the inset of Fig. 1. It appears that the χ values of the 0 and 10 at.% Al alloys are very close and that they are largely increased at $\mathfrak{z} \simeq 6.4$ while the 20 at.% Al alloys show a maximum at $\mathfrak{z} \simeq 6.6$. This behavior is related to that of the electronic-specific-heat coefficients γ. Figure 1 shows further that the critical concentration for ferromagnetism in the 20 at.% Al alloys is at $\mathfrak{z} \simeq 6.6$, whereas it is near $\mathfrak{z} \lesssim 6.4$ in both 0 and 10 at.% Al alloys. Thus it appears that the increased γ and χ values correspond to the critical concentrations for the appearance of ferromagnetism. In this case the observed γ and χ values will include enhancement effects which are known to occur in this concentration range.[11]

IV. CONCLUSION

The experimentally observed rate of moment decrease due to Al substitution in ferromagnetic bcc and fcc first-transition-series alloys, varies with the $\mathfrak{z}$ ratio of the transition metal. The rate of mean atomic moment decrease $-dM/dc$ due to an Al atom is qualitatively fitted to the experimental data using a moment decrease law as Eq. (1) or (2). These laws take into account (i) a passive dilution effect of the transition-element atoms of the ferromagnetic matrix by the nontransition impurity atoms and (ii) an active moment decrease due to the introduction of the nonmagnetic impurity atom which is proportional to $(V - n_s)$, the difference between the valence of the impurity and the free-electron density of the surrounding matrix. This second effect may be accounted for by either one of the mechanisms proposed by Mott[1] or Beeby.[10] If the dilution phenomenon (i) always has to be accounted for, the d-band contribution (ii) appears to vary as a function of $\mathfrak{z}$ and is zero in Fe-Al and becomes very small in (Cr-Fe)-Al near $\mathfrak{z} = 6.4$. An important conclusion is that the law $M = M_0 - cV\mu_B$ accounting for the moment variation in Ni-based alloys is only a particular case of Eq. (1), and does not have the significance of a contribution of the V valence electrons of the impurity to the d band of the ferromagnetic matrix. Finally, it has been shown that the electronic-specific-heat peak shift observed in the bcc first-transition-series element alloys with 0, 10, and 20 at.% Al are correlated with susceptibility peaks and to critical concentrations for appearance of ferromagnetism.

ACKNOWLEDGMENT

We want to thank Professor S. Brown for comments and assistance in the preparation of the manuscript.

---

[1] For a review, see N. F. Mott, Advan. Phys. 13, 325 (1964).

[2] T. M. Holden, J. B. Comly, and G. G. Low, Proc. Phys. Soc. (London) 92, 726 (1967).

[3] J. B. Comly, T. M. Holden, and G. G. Low, J. Phys. C 1, 458 (1968).

[4] D. I. Bardos, A. T. Aldred, and P. A. Beck, J. Appl. Phys. 38, 1260 (1967).

[5] D. I. Bardos, J. L. Beeby, and A. T. Aldred, Phys. Rev. 177, 878 (1969).

[6] For a review, see T. M. Srinivasan, H. Claus, R. Viswanathan, P. A. Beck, and D. I. Bardos, in *Phase Stability in Metals and Alloys*, edited by P. S. Rudman, J. Stringer, and R. I. Jaffee (McGraw-Hill, New York, 1967).

[7] A. Herr, thesis, Strasbourg, 1970 (unpublished).

[8] A. J. P. Meyer, Compt. Rend. 265, 448 (1967).

[9] J. Friedel, Nuovo Cimento Suppl. 7, 287 (1958).

[10] J. L. Beeby, Phys. Rev. 141, 781 (1966).

[11] For a recent review, see A. Hahn and E. P. Wohlfarth, Helv. Phys. Acta 41, 857 (1968).

PHYSICAL REVIEW B VOLUME 2, NUMBER 8 15 OCTOBER 1970

Structural Aspects of the Electrical Resistivity of Binary Alloys*

A. B. Bhatia and D. E. Thornton

Theoretical Physics Institute and Physics Department, University of Alberta, Edmonton, Canada

(Received 19 June 1970)

Using the weak scattering approximation and the Van Hove correlation-function technique, it is shown that the scattering function for a binary alloy (solid or liquid) is quite generally expressible in terms of three structure factors $S_{NN}(\vec{q})$, $S_{NC}(\vec{q})$, and $S_{CC}(\vec{q})$ constructed from the Fourier transforms of the local number density and concentration in the alloy. These structure factors have the property that at temperatures above the Debye temperature and in the long-wavelength limit $(q \to 0)$, $S_{NN}(0)$ and $S_{CC}(0)$ represent, respectively, the mean square thermal fluctuations in the particle number and concentration, and $S_{NC}(0)$ the correlation between these two fluctuations. Thermodynamic formulas for these fluctuations are given and their concentration and temperature dependence examined for various types of mixtures (regular, order-disorder type, athermal, etc.). It is concluded that the present formalism, because of its ready link with the thermodynamic properties of the alloy, can be helpful in interpreting the various experimental data and provides useful insight into the partial structure factors introduced in the Faber-Ziman theory of liquid alloys.

I. INTRODUCTION

In the current treatment of the problem of electrical conductivity of metals (liquid or solid), the relevant scattering function describing the scattering of electrons is given by the product of two factors: a structure factor which depends only on the relative positions of the ions in the metal, and the atom form factor or the pseudopotential matrix element which describes the scattering due to the individual ions in the metal.[1-5] The method depends on assuming that the total potential, responsible for scattering, in the metal may be written as the sum of the potentials due to individual ions in the metal and that it can be regarded as weak. Faber and Ziman[6] (FZ) have generalized this approach to discuss the electrical resistivity of liquid alloys. For a binary alloy the scattering function depends upon, in general, three independent partial structure factors which completely describe the structure of the alloy and the two form factors.

The transport properties of alloys have also been discussed on another, and seemingly unrelated, approach which is valid in the long-wavelength limit and which attributes the scattering of electrons – in analogy with the well-known theories[7,8] of light scattering in liquids and liquid mixtures – to the local thermal fluctuations in the density and concentration of the alloy. In particular, Krishnan and Bhatia[9] showed that the observed strong temperature dependence of the resistivity of an order-disorder type of alloy near its critical point may be attributed to the corresponding temperature dependence of the concentration fluctuations. A somewhat similar approach has been recently suggested by Takeuchi and Endo[10] for liquid alloys; see also Tomlinson and Lichter.[11]

The work of the present paper may, in a sense, be regarded as constituting a generalization of the above fluctuation approach so that it is applicable at shorter wavelengths and low temperatures. It is shown that in the aforementioned weak scattering approximation, the scattering function for a binary alloy (solid or liquid) is quite generally expressible in terms of three structure factors $S_{NN}(\vec{q})$, $S_{NC}(\vec{q})$, and $S_{CC}(\vec{q})$, which are derived from the Fourier transforms of the local number density and concentration of the alloy. For liquid alloys and for solids above the Debye temperature, these structure factors have the property that in the long-wavelength limit $(q \to 0)$, $S_{NN}(0)$ and $S_{CC}(0)$ represent, respectively, the mean square fluctuations in the particle