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[15] The Gaussian Quadrature with n positive roots of a function $f(x)$ over the interval -1 to 1 is exact if $f(x)$ is a polynomial in x of degree less than $4n$. Thus, by approximating the displacement field according to Eq. (8), we are in effect integrating exactly an expanded form for the integrand. In particular, the integrand is effectively expanded in a Taylor series of order $4n-1$ in each of the variables, x, y, and z, about either $+\frac{1}{2}\pi$ or $-\frac{1}{2}\pi$ depending on whether the variable is positive or negative. As $\vec{l}$ increases, the oscillatory behavior of the integrand becomes more pronounced until at some point the effective expansion breaks down; and this breakdown, when it occurs, takes place very sharply.

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Kohler's Rule and the Transverse Magnetoresistance of Very Dilute *Cu*Mn, *Cu*Fe, and *Cu*Co Alloys

C. M. Hurd and J. E. A. Alderson
Division of Chemistry, National Research Council of Canada, Ottawa K1A OR9, Canada

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The transverse magnetoresistance has been measured at 4.2 °K in polycrystalline alloys of Cu containing Co, Fe, or Mn, three solutes of respectively increasing magnetism. The solute concentrations are in the range 9–4000 at. ppm. Deviations from Kohler's rule in the low-field condition confirm the relative anisotropies of the solute potential scattering previously obtained from the Hall effect, and incidentally lead to a value of τ_B/τ_N of 0.9 for Co in Cu. The dependence upon solute concentration of the negative magnetoresistance in the *Cu*Mn alloys (23–271 ppm) shows an unexplained property: It is concentration independent and in agreement with previous work for the more concentrated samples, but becomes dependent upon concentration below about 70 ppm where no previous measurements exist. The importance of sample size effects in such measurements upon very dilute alloys is illustrated.

I. INTRODUCTION

According to Kohler's rule, the galvanomagnetic effects in a metal are functions of H/ρ_0, where H is the applied field strength and ρ_0 is the electrical resistivity when $H=0$. The circumstances under which this rule breaks down as the temperature or purity is altered have been discussed frequently in the literature.[1] These include cases where an effective change in the topology of the Fermi surface is produced (as, for example, by magnetic breakdown or localized interorbital electron scattering) or where the anisotropy of the average electronic relaxation time (τ) is changed (as can happen when two or more comparable scattering processes of different anisotropies coexist). A Kohler diagram can therefore frequently be a convenient presentation for discussing the contribution of these effects to the field dependence of a given galvanomagnetic property.

It is the purpose of this paper to discuss the application of a Kohler diagram to the isothermal transverse magnetoresistance of very dilute alloys of Cu containing solutes possessing varying degrees of magnetism. Following some recent work by us[2] showing evidence of possible solute clustering effects in such systems, we are particularly interested in their behavior as the solute's concentration is reduced to zero. The paper therefore deals with results obtained for very dilute alloys where the measurements inevitably encompass the intermediate-field region, i.e., between the low-field region ($\omega\tau \ll 1$), where the dominant scattering process governs the behavior of the magnetoresistance, and the high-field region ($\omega\tau \gg 1$), where only the topology of the Fermi surface and the degree of compensation are important. (ω is here the cyclotron frequency corresponding to H.)

The work has involved the measurement at 4.2 °K of the transverse magnetoresistance of polycrystalline samples of Cu containing Mn, Fe, or Co with an approximate concentration in the range 9–4000 at. ppm. The results lead to information on the relative anisotropy of the Coulomb-potential scattering of the solute ions (Sec. III), on the concentration dependence of the negative magnetoresistance in *Cu*Mn (Sec. IV), and on the importance of the electron scattering by the surfaces of the sample in wires of dilute alloys (Sec. V).

II. EXPERIMENTAL

The polycrystalline samples were prepared as described previously[3] from Cu of 99.9999-at.% pure grade[4] with a residual resistance ratio (RRR) of about 2000, and solute metals of 99.99 at.% or equivalent purity.[5] Each sample was chill cast twice in vacuum, and sections from the top and bottom of the second cast were chemically analyzed[6] to test the homogeneity. (In the case of the more concentrated *Cu*Co alloys, three castings were necessary to achieve acceptable homogeneity.) The results of these analyses, together with the electrical resistances measured at 4.2 °K, are given in Fig. 1.

In the cases of the *Cu*Zn, *Cu*Fe, and *Cu*Co alloys the measurements were made upon samples prepared for Hall effect studies. They were therefore in the form of flat plates with the shape and size described previously[3] and into which pointed potential probes were pressed, being aligned along the longitudinal axis of the sample with a separation of about 3 cm. This shape is not optimum for the accurate determination of the form factor, but we find the precision of our magnetoresistance data sufficient to illustrate the points under consideration (the precision of a given ordinate of Fig. 1 varies between 0.2 and 4.0%, with the maximum imprecision occurring at the lowest field strength). In the case of *Cu*Mn the samples were in the form of wires about 57 mm in length and with the diameters given in Fig. 4. The form factors of these samples were obtained from their weight and the density of the sample. The error in the final result is indicated by the error bars of Figs. 2 and 3. In the majority of cases the results for *Cu*Mn shown in Fig. 1 were obtained from wires of 0.040 in. (0.1 cm) diam; the least concentrated sample was the exception where, to avoid the size effects discussed in Sec. V, its diameter was increased to 0.080 in.

All measurements were made with a dc potentiometric arrangement having a resolution of 1×10^{-9} V. A primary current of 10 A was used in most experiments, but this had to be reduced to 2 A for the more concentrated alloys to avoid excessive Joule heating. Field strengths up to 15.17 kOe were employed. The magnetoresistance was determined from the mean of results obtained from the four permutations of applied electric and magnetic field directions.

Before measurement, the *Cu*Zn, *Cu*Mn, and the least concentrated examples from the *Cu*Fe and *Cu*Co series were each annealed in vacuum for a minimum of 24 h at 550 °C before the container was air cooled to room temperature. The rest were annealed in vacuum for a minimum of 24 h at 1000 °C before being rapidly quenched into iced brine. After quenching, the more concentrated *Cu*Co samples were immediately installed in the apparatus and cooled to 77 °K to minimize metallurgical complications, and in all cases the measurements were made within 6 h of quenching. The nonlinear dependence between the analytical solute concentration and $\rho_{4.2\,^\circ\mathrm{K}}$ for the more concentrated *Cu*Fe alloys of Fig. 1 suggests that not all of the solute was in solid solution in these cases.

III. POSITIVE MAGNETORESISTANCE COMPONENT

Fig. 1 shows in a Kohler diagram the field dependence of the transverse magnetoresistance measured at 4.2 °K in field strengths up to 15.17 kOe. The results cover the region between the high-field condition, where a linear dependence of $\Delta\rho$ upon H is expected[7] and observed for polycrystalline alloys, and the low-field condition where the corresponding expected[7] quadratic dependence upon H is observed.

An appreciable negative component is observed only in the case of the *Cu*Mn alloys. This is connected with the fact that in passing along the series *Cu*Co, *Cu*Fe, *Cu*Mn, we are known to be dealing with three systems showing, respectively, greater magnetism. Firstly, individual Co atoms are probably nonmagnetic in Cu and only show magnetism when there is appreciable solute-solute interaction (i.e., for concentrations above about 1%).[8] We therefore do not expect any evidence of magnetism in the dilute alloys represented in Fig. 1. (We have confirmed that alloys of Cu + 1 and 2% Co do show a negative component.) Secondly, with a Kondo temperature of about 16 °K, the individual moments in the *Cu*Fe system have suffered a significant Nagaoka compensation[9] at 4.2 °K. Fe is therefore only weakly magnetic in Cu at this temperature and is known to show only a small negative magnetoresistance compared with Mn.[10]

Leaving aside until Sec. IV the negative component in *Cu*Mn, it can be seen from Fig. 1 that the data for the transition series *Cu*Mn, *Cu*Fe, and *Cu*Co superimpose and therefore obey Kohler's rule to within experimental error (the abscissas are displaced for clarity). This is expected in the high-field condition where the scattering by the solute (and hence the nature of the solute) becomes irrelevant, but its persistence into the low-field condition implies that the Coulomb

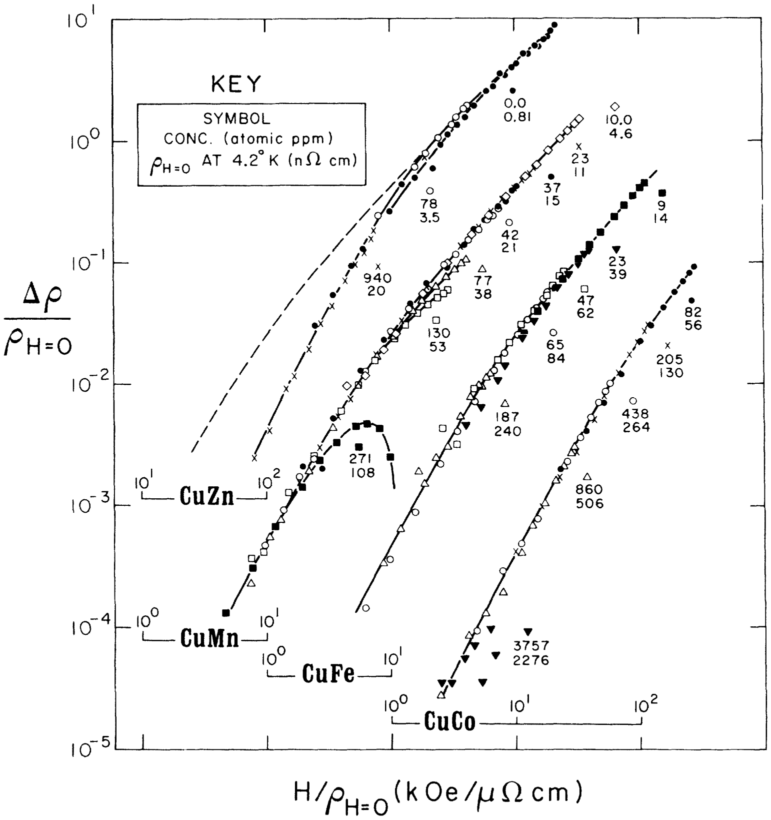

FIG. 1. Field dependence of the transverse magnetoresistance measured at 4.2 °K in fields up to 15.17 kOe for the polycrystalline samples indicated. Apart from the negative component shown by the more concentrated *Cu*Mn alloys, the data for *Cu*Mn, *Cu*Fe, and *Cu*Co superimpose and thus obey Kohler's rule to within experimental error (the abscissas are displaced for clarity). The low-field results therefore indicate that the Coulomb scattering has equivalent anisotropy in each system. The dashed line contrasts this behavior with that observed for a nontransition element Zn.

potential scattering of these transition solutes has the same anisotropy (to within experimental error) in each case. This is contrasted in Fig. 1 with the behavior shown by Zn, which probably can be taken as a typical nonmagnetic solute since the series of heterovalent metal solutes is known from previous work generally to obey Kohler's rule when dissolved in a given group-IB metal, viz., *Ag*(Cd, In, Sb) [7] and *Cu*(Zn, Sn). [11] The figure shows the significant deviation in the low-field region between the results for the transition solutes (dashed line) and Zn, indicating the differing anisotropies between these cases.

This is confirmed by what is known about the anisotropy of the Coulomb scattering from other experiments. The low-field Hall effect leads[12] to values of τ_B/τ_N (in the usual terminology) of 1.00, 0.90, and 0.87, respectively, for Zn, Mn, and Fe in Cu. Since the estimated uncertainty was ± 0.04 in each case, these data also give no detectable differences in τ_B/τ_N measured for Mn and Fe in Cu, and we conclude from this and from Fig. 1 that the corresponding value of τ_B/τ_N for Co in Cu must therefore also be approximately 0.9.

IV. NEGATIVE MAGNETORESISTANCE IN *Cu*Mn

The interpolated positive magnetoresistance of *Cu*Mn shown in Fig. 1 provides a "background" behavior representing the case where the combination of applied field strength and solute concentration is small enough that no detectable effects of spin scattering exist, and yet the concentration is large enough that the solute remains the dominant scattering center. [13] The negative component of $\Delta\rho/\rho_{H=0}$ at any given value of $H/\rho_{H=0}$ is therefore taken to be the vertical separation in Fig. 1 between this background and the total measured value of $\Delta\rho/\rho_{H=0}$. (We have pointed out elsewhere[2] that this empirical determination of the negative component automatically accounts for the effect of the high-field/low-field transition and therefore avoids possible error due to this cause incurred by assuming some fixed field dependence for the positive component.) If we wish to identify the negative component with the contribution arising from spin scattering, it should be noted that this implies Matthiessen's rule is obeyed by the Coulomb potential and spin resistivity contributions to the total resistivity. It has been suggested[14] that in general Matthiessen's rule is not obeyed in the presence of a magnetic field, but the subject has not been fully investigated either theoretically or experimentally. We shall subsequently return briefly to this point.

Figure 2 shows the field dependence of the

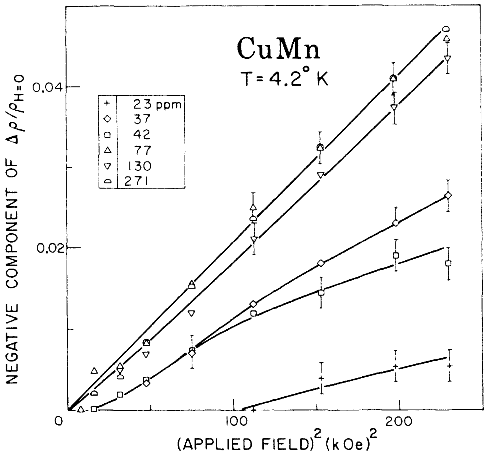

FIG. 2. Field dependence at 4.2 °K of the negative component of $\Delta\rho/\rho_{H=0}$ obtained as described in the text from the *Cu*Mn data of Fig. 1. It is only for the more concentrated samples that the expected concentration-independent linear dependence upon H^2 is observed.

negative component of $\Delta\rho/\rho_{H=0}$ obtained[13] as described above at 4.2 °K from the data in Fig. 1. The ordinate is therefore effectively the scattering power per solute ion arising from the presence of spin, and the figure shows that for sufficiently concentrated alloys this component becomes a concentration-independent linear function of the square of the applied field strength. It is well known[11,15,16] that this behavior is expected if the negative component is taken to arise from the freezing out of spin-flip scattering caused by the alignment of the individual noninteracting spins in the magnetic low-field condition (i.e., $g\mu_B H/kT < 2$, where the quantities have their usual meanings[16]). In this condition the field dependence of the spin-flip scattering is found to vary as the square of the magnetization in the sample and to dominate completely the field dependence of the conduction electron scattering amplitudes.[16] This situation presumably therefore exists for the data of Fig. 2 so that the observed variation with H^2 is not unexpected, but what we think is new is the evidence that this behavior is modified (for *Cu*Mn at least) for solute concentrations below a certain critical value (~70 ppm).

This is seen clearly in Fig. 3 which shows the concentration dependence of the negative component of $\Delta\rho/\rho_{H=0}$ for $T = 4.2$ °K at $H = 15$ kOe. The open circles are the data from Fig. 1, while we have also included results taken directly from Muto *et al.*[11] (70 ppm) and from Gerritsen's data[17] for the total magnetoresistance (0.042, 0.15, 0.33, and 3.2%). In the case of the three most concentrated alloys in the latter's series the positive contribution to $\Delta\rho/\rho_{H=0}$ is insignificant and can be safely neglected to give the negative component directly, but we estimated from our own results that this would not be a safe procedure for the alloy containing 0.042%. Since we could not obtain the corresponding positive contribution from the data given by Gerritsen, the best compromise seemed to be an estimation from our own results ($\rho_{H=0}$ can be obtained corresponding to Gerritsen's concentration, hence $H/\rho_{H=0}$ and the corresponding positive part from Fig. 1). This turned out to be + 0.02, and when combined with the - 0.02 given by Gerritsen for the total magnetoresistance gives the negative component shown in Fig. 3. In the case of Monod's data shown in Fig. 3, the absolute value of the negative component is obtained from Fig. 2 of Béal-Monod and Weiner[16] and the corresponding value of $\rho_{H=0}$ from Monod.[10] (Note that a value of $\Delta\rho/\rho_{H=0}$ at 15 kOe interpolated from Monod's data would be somewhat smaller than the corresponding values from other workers represented in Fig. 3. We believe this is associated with Monod's method[10] of treating the data, but since we have discussed this point in detail elsewhere,[2] we shall not elaborate here.)

Figure 3 shows that a breakdown of the concentration independence of $\Delta\rho/\rho_{H=0}$ occurs both at higher concentrations (between 0.33 and 3.2%, presumably because of the effects of either solute magnetic ordering or of changes in the solvent

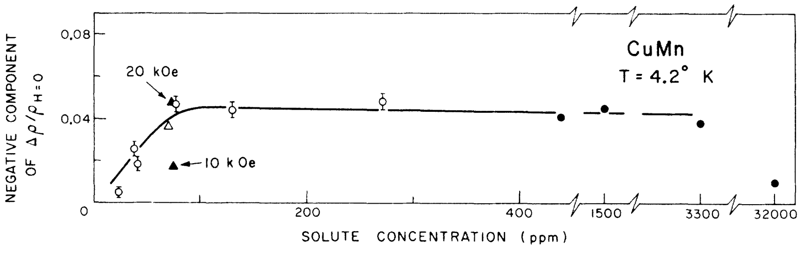

FIG. 3. Open circles: concentration dependence of the negative component of $\Delta\rho/\rho_{H=0}$ obtained from Fig. 1 for *Cu*Mn at 4.2 °K and 15.17 kOe (see also Ref. 13); closed circles: corresponding results at 15 kOe obtained, as described in the text, from Gerritsen (Ref. 17); open triangles: corresponding results at 15 kOe obtained by Muto *et al.* (Ref. 11); and closed triangles: two data at 10 and 20 kOe obtained by Monod (Refs. 10 and 16).

metal's band structure), and also at lower concentrations (below about 70 ppm). The latter effect is quite unexpected. If it is real, and not just some artifact of the experimental conditions or the treatment of the data, then clearly it indicates (for *Cu*Mn at least) that some solute-solute interaction is important even in the usual concentration-independent component of $\Delta\rho/\rho_{H=0}$ which has been taken as the foundation for theoretical development.[16] We can presently offer no certain explanation of this observation, but we think the following remarks are relevant:

(i) We have already referred to the fact that Matthiessen's rule is implicitly assumed in the separation of the negative component of $\Delta\rho/\rho_{H=0}$ from such as Fig. 1. We have made this assumption because it appears from the available study of this topic[14] that to do so will incur no appreciable error in the case of the very dilute alloys under consideration. However, it should be emphasized that there is presently no very comprehensive experimental or theoretical study of the validity of Matthiessen's rule in an applied field, and it must remain an open question whether the foregoing assumption is valid.

(ii) Metallurgical complications cannot be ruled out. It has previously been reported[18] that significant solute clustering effects may be evident in the low-temperature magnetic susceptibility of dilute *Cu*Mn alloys, and we have recently reported[2] similar evidence from the field dependence of the low-temperature Hall effect. In fact there is a correlation between the latter data and the behavior shown in Fig. 3; alloys of Cu+ 111- and 305-ppm Mn show evidence of a low-field saturating component in the Hall effect[2] (which is attributed to the presence of superparamagnetic clusters of solute), while alloys containing 10 and 19-ppm Mn do not. This supports the argument that perhaps the behavior shown in Fig. 3 reflects the formation of clusters as the concentration increases. The size of the cluster is independent of concentration above about 70 ppm (at least up to 3300 ppm), and with increasing concentration

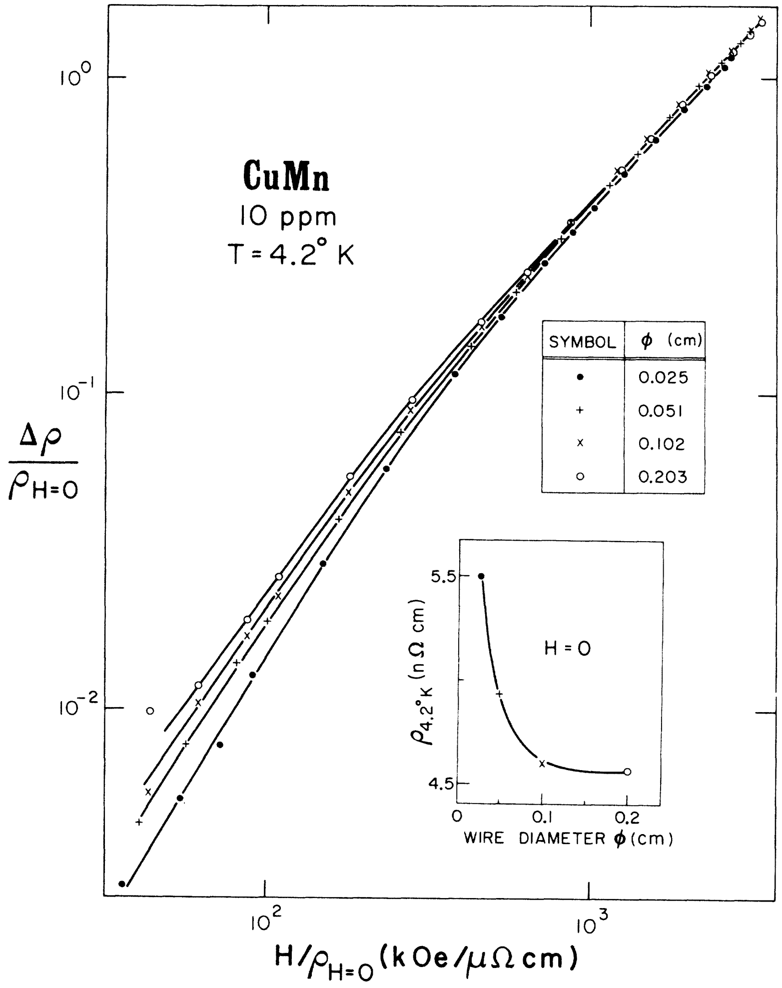

FIG. 4. Field dependence of the transverse magnetoresistance in a Cu+10-ppm Mn alloy measured at 4.2 °K for varying diameters ϕ. Deviations from Kohler's rule occur in the low-field condition when ϕ is less than about 0.2 cm because electron scattering from the sample's surfaces becomes comparable with that arising in the bulk.

only the number of such clusters is increased. (This proposed stability of the cluster's size as the concentration is varied is similar to the behavior already suggested for Co in Cu,[19] although there was no investigation in that work of the behavior when the solute concentration is reduced to zero.)

(iii) We find that to within experimental error $\rho_{H=0}$ measured at 4.2 °K shows a linear dependence upon solute concentration in the range 0–400 ppm with no evident anomaly around 70 ppm, such as might be expected if the above clustering is occurring. However, the reliability of this test is poor since it depends upon the accuracy of the analyses, which is low for the very dilute samples.

V. SIZE EFFECTS

We pointed out in the Introduction that Kohler's rule breaks down when two or more scattering processes of comparable magnitudes but differing anisotropies exist in an alloy. We show briefly in this section that such effects can arise in magnetoresistance measurements made upon very dilute alloys (i.e., typically with solute concentrations $< \sim 50$ ppm) in the form of wires when the diameter is reduced so that the scattering from the surfaces of the sample becomes comparable with that arising in the bulk. We describe experiments made upon a typical alloy of Cu + 10-ppm Mn which indicate that the effect in that case is appreciable in wires of up to ~0.2 cm diam and that, if not accounted for, could lead to an error in the interpretation of the data and particularly in the usual evaluation of the positive magnetoresistance. That such effects are not usually considered by experimenters is indicated by the failure in some recent cases[10,11] to even specify the dimensions of the samples used, although we emphasize that a rough extrapolation from our work suggests that at the concentrations used by these workers appreciable size effects would not be evident at 4.2 °K unless the minimum dimension of the sample was less than about 0.005 cm.

Figure 4 shows the field dependence of the transverse magnetoresistance obtained for the above alloy at 4.2 °K. (Free-electron theory leads to a corresponding electronic mean free path in this alloy of approximately 1.2×10^{-3} cm.) The diameter of the samples varies in the range 0.025–0.2 cm (0.010–0.080 in.), and the deviation from Kohler's rule at the lowest-field values is seen to amount to more than a factor of 2 in the value of $\Delta\rho/\rho_{H=0}$. We note from the results shown in Fig. 1 that the sample having the largest diameter of Fig. 4 (0.2 cm) gives results in agreement with the more concentrated alloys of the series, and is therefore typical of the bulk material. This is confirmed by the behavior of the residual resistance in zero field shown in the insert to Fig. 4.

Our conclusion from all of this is that, insofar as *Cu*Mn is a typical system, sample size effects in low-field galvanomagnetic effects sensitive to the anisotropy of a dominant relaxation time can be significant for a combination of temperature, solute concentration, and minimum sample dimension where it would not be expected from a simple free-electron calculation. Although we have not attempted any systematic study of the dependence of the size effect upon these parameters, we think that in view of the increasing tendency to study very dilute alloy systems it is worth drawing attention to the results of Fig. 4.

VI. SUMMARY AND CONCLUSIONS

The low-field transverse magnetoresistance measured at 4.2 °K indicates the anisotropy of electron scattering in *Cu*Mn, *Cu*Fe, and *Cu*Co systems to be equivalent and to correspond to a value of τ_B/τ_N of about 0.9.

The negative component of $\Delta\rho/\rho_{H=0}$ obtained at 15.17 kOe and 4.2 °K shows an anomalous solute concentration dependence in very dilute alloys of *Cu*Mn. Although the expected concentration-independent behavior is found for a wide range of solute concentrations (at least between 70 and 3300 ppm), an anomalous dependence upon concentration is observed below about 70 ppm. It is suggested that this behavior possibly arises from the formation of superparamagnetic clusters of solute atoms in which the number of atoms is independent of the solute concentration at least between about 70 and 3300 ppm. If this is the case, *Cu*Mn is evidently not a simple random alloy, and our results undermine the existing theories and interpretations which are entirely based upon the concept of localized individual solute moments being responsible for the negative magnetoresistance. Although there is supporting evidence from other experiments that such clustering can be significant in the *Cu*Mn alloys, there is clearly presently insufficient data to establish the above suggestion beyond doubt, and we have recently started further work to investigate the occurrence of the anomalous behavior in other Kondo systems.

ACKNOWLEDGMENTS

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Theory of Phase Transitions in the β-Tungsten Structure Induced by the Band Jahn-Teller Effect

E. Pytte
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

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A dynamic theory of the cubic-to-tetragonal transformation in the β-tungsten structure is presented starting with a Hamiltonian describing the interaction between static strain and acoustic phonons with the electrons in the triply degenerate *d* bands. For this model the elastic constants are strongly temperature dependent in the low-frequency limit. However, for frequencies in the neighborhood of the Debye frequency, they are nearly temperature independent.

I. INTRODUCTION

The cubic-to-tetragonal structural-phase transition in the intermetallic A_3B compounds with the β-tungsten (β-W) structure has been the subject of a number of recent theoretical and experimental papers.[1-37]

These phase transitions are believed to be induced by the interaction of a triply degenerate *d* electron band with the elastic strain.[6] The distortion from cubic symmetry splits the *d* bands into a singlet and a doubly degenerate band lying, respectively, below and above the bands in the undistorted structure. The energy gained by the increased occupation of the lower band is balanced by the increase in the elastic energy. The soft mode associated with the transition is an acoustic shear mode propagating in a [110] direction polarized in the $[1\bar{1}0]$ direction.

The transitions in the β-W structures have been described in terms of a one-dimensional linear-chain model to calculate the *d*-band structure in the tight-binding approximation.[6-10] The coupling of the electronic system to the elastic strain was discussed using a free-energy approach. A simpler constant-density-of-states model was introduced by Cohen, Cody, and Halloran[11] in terms of which good quantitative agreement could be obtained with experimental values.[11,12,29,38] Very recently Klein and Birman[14] revived an early suggestion by Anderson and Blount[5] that the transition might be driven by an optical-phonon instability.

In this paper a dynamic theory is presented for the band Jahn-Teller mechanism based on the elec-