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Thermoelectric Power of Dilute Cu-Ni Alloys in a Magnetic Field

C. L. FOILES*

Physics Department, Michigan State University, East Lansing, Michigan

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The thermoelectric powers (TEP) of four dilute CuNi alloys and one CuCo alloy have been measured in the presence of a transverse magnetic field. The presence of the field did not induce any anomalous behavior in the TEP. There seems to be no systematic field effect in the temperature range of 4.2 to 20°K, and the maximum change due to a 10-kOe field was $<0.05\ \mu V/°K$. The temperature dependence of the absolute TEP of the CuNi alloys suggested that the diffusion component might follow a Gorter-Nordheim relation. An analysis consistent with this relation and assuming an unsplit virtual state is presented.

I. INTRODUCTION

It is well known that the addition of 3*d* transition metal solutes to copper can strongly affect the thermoelectric power (TEP).[1] The "giant" effects of some solutes (Mn, Fe) which also display a localized magnetic moment have overshadowed the weaker but pronounced effects of those (Co, Ni) which do not display a magnetic moment. The theory for all of these effects is still in the throes of development and generally concentrates upon those alloys displaying a localized magnetic moment.[2] However, at the present, two things seem clear. The terms from perturbation theory which cause the low-temperature anomalies in resistivity are related but not identical to those causing the TEP effects. The percentage change relative to normal behavior is greater for the TEP effects.

The possibility of observing the change in TEP as a magnetic moment forms offers a promise of insight into the problem. One possible technique for achieving this result is the measurement of TEP in a magnetic field. An analysis by Anderson[3] suggests that for dilute amounts of nickel in copper the conditions for the formation of local moments on the nickel are nearly satisfied. This suggestion is strengthened by the anomalous behavior of the susceptibility of this alloy as a function of nickel concentration[4] and by the density of states data of Pugh and Ryan.[5] Also, previous work of MacDonald, Pearson, and Templeton[6] indicates that a field of 11 kOe may change the TEP of Cu+0.44% Ni by some $0.4\ \mu V/°K$ near 20°K.

This paper reports work done on the TEP of dilute Cu-Ni alloys and one Cu-Co alloy in a magnetic field. A crude model is used to calculate the TEP due to the presence of the impurity, and the experimental results are discussed in relation to the predictions of this model.

II. EXPERIMENTAL PROCEDURE

The samples were prepared using 99.999% copper obtained from the American Smelting and Refining Company, nickel sponge obtained from Johnson Matthey, and 99.3% cobalt. The constituents were weighed (stated compositions are in at. % calculated from wt %), placed in an outgassed graphite crucible, and melted in a vacuum by induction heating. Each sample was melted and chill cast in copper molds a minimum of two times. Past experience has shown that two chill casts eliminate the need for a homogenizing anneal. The samples were rolled between pure copper

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* This work was supported by a National Science Foundation grant.

[1] The review by G. J. van den Berg, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Co., Amsterdam, 1964), Vol. IV, includes a consideration of TEP anomalies.

[2] See the review by M. Bailyn, Advan. Phys. **15**, 179 (1966).

[3] P. W. Anderson, Phys. Rev. **124**, 41 (1961).

[4] A. R. Kaufmann and C. Starr, Phys. Rev. **63**, 445 (1943); E. W. Pugh, B. R. Coles, A. Arrott, and J. E. Goldman, Phys. Rev. **105**, 814 (1957).

[5] E. W. Pugh and F. M. Ryan, Phys. Rev. **111**, 1038 (1958).

[6] D. K. C. MacDonald, W. B. Pearson, and I. M. Templeton, in *Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry, Madison, Wisconsin, 1957*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, Wisc., 1958).

(or later, hardened Cu-Be) plates to a thickness of 25 to 40 mil. Samples 7 cm in length were then cut from the alloy sheet.

Two samples from previous work by Schroeder *et al.*[7] were also used. These samples had been prepared in the same manner but formed differently. They had been drawn into wires of 10-mil diam and then annealed. A bundle of 7-cm lengths of these wires formed a sample for the present work.

A block diagram of the measurement circuit is given in Fig. 1. The two copper posts A and B each have an individual manganin heater which is supplied with a dc current. The sample connects the two posts and is soldered to them with a low melting solder. A length of copper wire connects one post to the He bath. Power to the heater of the latter post controls the temperature of the sample and power to the other heater controls the temperature gradient across the sample.

The temperature and the temperature difference were measured by Au-Fe–Chromel thermocouples. The Au-Fe was 0.03 at. % Fe. The work of Berman *et al.*[8] indicated that for the conditions of this experiment no correction due to the presence of the magnetic field was necessary for these thermocouples. A Au-Fe–Chromel thermocouple was calibrated against a platinum resistance thermometer from 4.2°K to room temperature. Platinum resistance thermometers have low sensitivity below 20°K, and an additional check for the 4.2 to 20°K range was made by measuring the TEP of Pb versus Nb_3Sn. The results were in excellent agreement with those of Christian *et al.*[9] and are taken to be a verification of the Au-Fe–Chromel calibration. The differential signal was amplified by a Leeds and Northrup null detector followed by a Philbrick operational amplifier. The output was displayed on a meter. The temperature signal was amplified by a Leeds and Northrup dc amplifier. The output was read from the meter of the instruments.

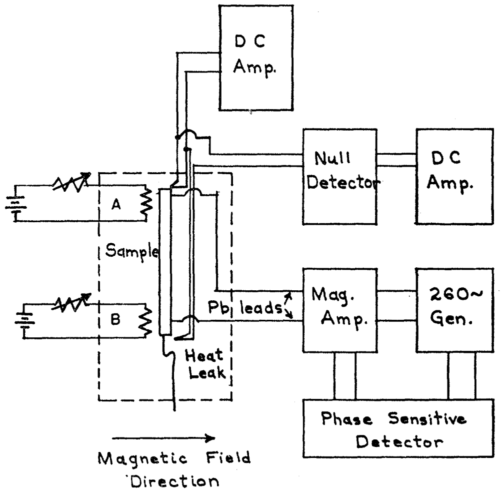

FIG. 1. Schematic diagram of the measurements.

The voltage measurement was made through the use of a magnetic amplifier. A detailed description of this circuit and measurement procedures is given elsewhere.[10] Briefly, the operation is as follows. The Pb leads and the sample form the thermocouple which supplies the magnetic amplifier with a dc signal. The magnetic amplifier gives an ac signal output (the sum of several harmonics) which is proportional to the dc signal. The phase sensitive detector measures the amplitude of the second harmonic and thus the magnitude of the dc signal. Using a reference signal at 260 cps, a sensitivity of 5×10^{-9} V was achieved.

The portion of the circuit enclosed by the dotted lines in Fig. 1 was mounted within a copper can which was immersed in the liquid He. The Pb leads and the thermocouple leads were continuous from the inner portion of the can to the region in the He bath where they made their connections. The leads were soldered into feedthroughs mounted in the flange of the copper can. The can was sealed through the use of a Pb O-ring. Data were taken even in cases where the can leaked, provided the pressure was not so large as to prevent easy establishment of a temperature gradient. Measurements made on Pb with and without a leak indicated that the absolute error in this procedure was <0.04 μV/°K.

The magnetic amplifier was mounted in the He bath and had to be shielded from the magnetic field. This was done by mounting the amplifier in a can with a lead foil covering. This covering was in turn covered with a Mumetal sheet. The 10-kOe limit on the applied magnetic field was dictated by the shielding of the amplifier.

III. EXPERIMENTAL RESULTS

The absolute TEP for the Cu-Ni alloys are plotted in Fig. 2. In determining these values, the Pb results of Christian *et al.*[9] and of Steele[11] were used; both sets of results were reproduced to within 0.05 μV/°K by using a Nb_3Sn sample and fields up to 7.5 kOe. The scarcity of zero-field points in the 6 to 8°K range resulted from avoiding the superconducting transition of Pb. The alloy measurements were reproducible to within 0.03 μV/°K. It seems clear that the magnetic field had little affect upon the TEP (certainly <0.05 μV/°K). Thus, these results are not consistent with any attempt to interpret the MacDonald, Pearson, and Templeton data[6] as arising from a magnetic field dependence of the TEP of Cu-Ni samples.

Two features of the present measurements should be

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[7] P. A. Schroeder, R. Wolf, and J. A. Woollam, Phys. Rev. **138**, A105 (1965).

[8] R. Berman, J. C. F. Brock, and D. J. Huntley, Cryogenics **4**, 233 (1964).

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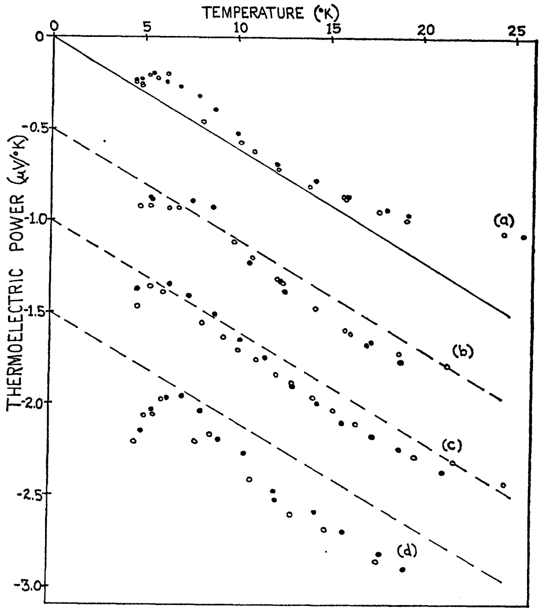

Fig. 2. Absolute TEP of the Cu-Ni alloys: (a) 0.1% Ni, (b) 0.35% Ni, (c) 0.63% Ni, (d) 0.85% Ni. For the purpose of display, the curves (b), (c), and (d) have been displaced by -0.5 μV/°K, -1.0 μV/°K, and -1.5 μV/°K, respectively. The straight lines represent a TEP of $-0.061\ T$ μV/°K displaced by the same amounts. The open circles represent the zero-field data and the solid circles represent the data at 7.5 kOe for (b) and (d) and the data at 10 kOe for (a) and (c).

noted. First, from 5 to 20°K the TEP of each sample can be represented to within ±0.1 μV/°K by a straight line having a negative slope. This linear temperature variation is highly suggestive of an impurity dominated scattering. (N.B. The residual resistivity of the most dilute sample should be $\simeq 0.125$ μΩ cm, which is more than 150 times the resistivity of pure copper at 20°K.) The apparent negative deviation from linearity at 5°K is believed to be due to Fe impurity introduced via the nickel sponge. Although the necessity of introducing weighting factors [see Eq. (2)] and ensuring questions as to valid, explicit forms for these factors when magnetic scattering occurs do not permit an unambiguous verification of this belief, two points can be mentioned in support: (1) The work of Gold *et al.*[12] indicates that pure Cu containing ~1 part per million (ppm) Fe can have a TEP between -1.5 to -4.5 μV/°K below 5°K. The specifications of the Ni sponge list are ~10 ppm Fe and thus the most concentrated Ni sample has a minimum of ~0.1 ppm Fe. The deviation at 5°K for this sample is -0.3 μV/°K. (2) The magnitude of the negative deviations at 5°K appears to increase as the Ni concentration increases.

Second, the total TEP is often the sum of a diffusive component S_d due to a nonequilibrium part of the electron distribution and a phonon drag component S_g due to the electron interaction with the nonequilibrium part of the phonon distribution. In copper the phonon drag component is positive and reaches a maximum near 60°K. The systematic positive deviation from linearity for the 0.1 and 0.35 at. % Ni alloys as the temperature increases is believed to be partially due to a phonon drag contribution. This interpretation is strengthened by the fact that measurements by Schroeder *et al.*[7] on the same 0.35 at. % Ni sample found the TEP at 80°K to be about -0.95 μV/°K with a negative temperature slope. The present work gives a value of -1.3 μV/°K at 20°K for this alloy. Since the difference between these values is more than the combined experimental errors, a smooth fit of the data requires the TEP to reach a minimum value and then rise to a maximum value as the temperature varies from 20 to 80°K. A positive phonon drag contribution is the most reasonable explanation for such a behavior. Assuming the alloying does not enhance the phonon drag component, the work of Crisp *et al.*[13] indicates that the contribution of this term at 20°K could vary from 0 to 0.3 μV/°K. This range of values is consistent with the present results.

The absolute TEP of the single Cu-Co alloy is plotted in Fig. 3 and the same conclusion of no appreciable field effect is indicated.

IV. DISCUSSION

The concept of bound or virtually bound states has become increasingly useful in understanding the behavior of alloys containing dilute amounts of transition metal solutes and it seems worthwhile to determine if this concept can provide some insight into the temperature dependence of the TEP of dilute Cu-Ni alloys. To this end a crude calculation of the diffusion TEP is attempted. The calculation is based upon the model presented by Daniel and Friedel.[14]

Assuming Matthiessen's rule and a well-defined relaxation time due to the impurities, the free-electron expression for the diffusion TEP (S_d),

$$S_d=\frac{\pi^2k^2T}{3e}\left[\frac{\partial\ln\rho}{\partial\epsilon}\right]_{\epsilon=\epsilon_F}, \tag{1}$$

reduces to a Gorter-Nordheim[15] relation of the form

$$S_d=\frac{\rho_{\text{th}}}{\rho_T}\left[\frac{\pi^2k^2T}{3e}\left(\frac{\partial\ln\rho_{\text{th}}}{\partial\epsilon}\right)_{\epsilon=\epsilon_F}\right]+\frac{\rho_0}{\rho_T}\left[\frac{\pi^2k^2T}{3e}\left(\frac{\partial\ln\rho_0}{\partial\epsilon}\right)_{\epsilon=\epsilon_F}\right]$$
$$\equiv\rho_{\text{th}}S_{\text{th}}/\rho_T+\rho_0S_0/\rho_T, \tag{2}$$

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[12] A. V. Gold, D. K. C. MacDonald, W. B. Pearson, and I. M. Templeton, Phil. Mag. **5**, 765 (1960).

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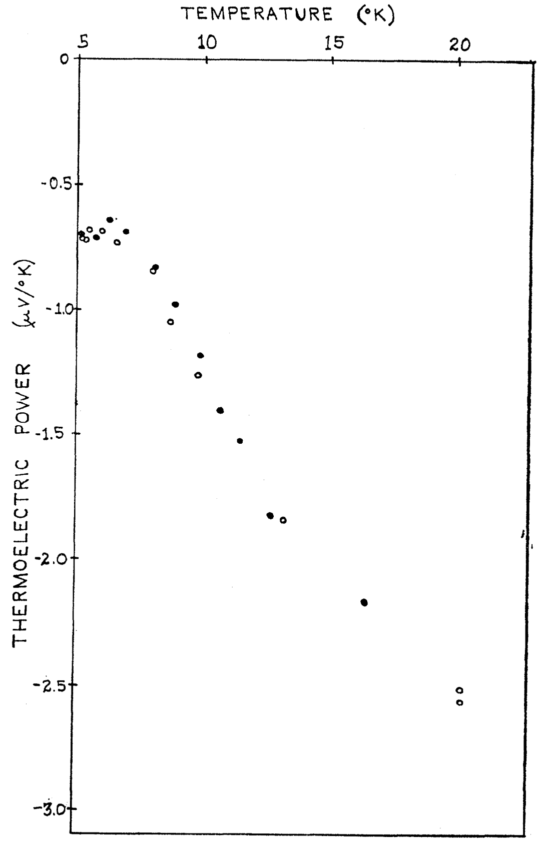

FIG. 3. Absolute TEP for the 0.1% Co Cu-Co alloy. The open circles are the data at zero field and the solid circles are the data at 10 kOe.

where k is Boltzmann's constant, T is the absolute temperature, e is the electronic charge, ρ is the resistivity as a function of electron energy ϵ, ϵ_F is the Fermi energy, ρ_{th} is the resistivity of the pure metal, ρ_0 is the resistivity due to impurity scattering, and ρ_T is the total resistivity of the alloy. A standard result from elastic scattering in a one-electron model[16] is

$$\rho_0=\frac{4c\hbar\pi}{100ne^2K}\Big[\sum_{l=0}^{\infty}(2l+1)\sin^2\delta_l -2l\sin\delta_{l-1}\sin\delta_l\cos(\delta_{l-1}-\delta_l)\Big], \qquad (3)$$

where c is the impurity concentration in at. %, n is the number of free electrons per atom, K is the magnitude of the electron wave vector, l is the quantum number associated with the angular momentum, and δ_l is the phase shift of the lth partial wave. The Friedel sum rule[17] is

$$N=\frac{2}{\pi}\sum_{l=0}^{\infty}(2l+1)\delta_l, \qquad (4)$$

where N is the valence difference between the pure metal, and the impurity results from requiring the scattering potential to be self-consistent.

The above equations are sufficient to permit the calculation of S_d if the energy dependence of the phase shifts can be determined. The process is greatly simplified by the fact that $3d$ transition metal impurities in copper seem to give rise to a resonant scattering, so that only $l=0$ and $l=2$ terms are important. The single problem remaining is to give a quantitative description of the resonance and this is done by using a model of the density of states. The density of states is assumed to consist of the usual conduction-band term plus a Lorentzian term n_d expressed as

$$n_d(\epsilon)=\frac{A}{\pi}\,\frac{\Delta}{(\epsilon-\epsilon_d)^2+\Delta^2}, \qquad (5)$$

where Δ characterizes the width and ϵ_d the location of this extra term. A is a normalization factor and is related to the total number of states available in this extra term. For the purposes of the present calculation A was set equal to 10, the total degeneracy for a d shell. The density of states was related to the scattering process by differentiating Eq. (4) with respect to energy and using the relation

$$\left(\frac{\partial N}{\partial\epsilon}\right)_{\epsilon=\epsilon_F}=n_d(\epsilon_F)=\frac{2}{\pi}\left[\left(\frac{\partial\delta_0}{\partial\epsilon}\right)_{\epsilon=\epsilon_F}+5\left(\frac{\partial\delta_2}{\partial\epsilon}\right)_{\epsilon=\epsilon_F}\right]. \qquad (6)$$

Since Eq. (6) is not the only way in which the density of states can be related to the scattering, it seems appropriate to comment upon the reasons for choosing this particular relation. First, the case of resonant scattering from a virtual state is characterized by a phase shift which is highly energy-dependent. This energy dependence has a maximum at the energy which represents the location of the state. Second, the more usual approach of calculating $\Delta\rho_0/\Delta\epsilon$ from ρ_0 values for different values of ϵ_F is difficult to apply in the present case. The difficulty arises from the dual requirement of having a resonant scattering and satisfying the Friedel sum for the different ϵ_F values. This is equivalent to requiring $(\partial N/\partial\epsilon)_{\epsilon=\epsilon_F}=0$ and results in $\partial\delta_0/\partial\epsilon>\partial\delta_2/\delta\epsilon$, which is difficult to interpret physically or requires additional phase shifts to be considered. The proper apportionment between these additional phase shifts is unclear without further specification. Finally, the approximations which lead to Eq. (1) are based upon the density of states and its first energy derivative evaluated at ϵ_F. The use of Eq. (6) is consistent with those approximations and does not constitute a violation of the Friedel sum rule.

The above considerations provide sufficient conditions to evaluate S_0 which now assumes the explicit

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[16] K. Huang, Proc. Roy. Soc. (London) 60, 161 (1948).
[17] J. Friedel, Phil. Mag. 43, 153 (1952).

form

$$S_0=\frac{\pi^2k^2T}{3e}\left[-\frac{1}{2\epsilon_F}+\frac{\partial\ln(\sin^2\delta_0+3\sin^2\delta_2)}{\partial\epsilon}\right]_{\epsilon=\epsilon_F}. \quad (7)$$

In the calculation of S_0 it was assumed that the $l=0$ partial wave was scattered by a perturbation charge equal and opposite to the amount of charge not in the d virtual state. Explicitly, a nickel atom has a total of 10 electrons in its $3d$ and $4s$ shells. If the virtual state contained M electrons, then the excess charge seen by the $l=0$ partial wave was $9-M$ which was defined as Z. The valency difference used in the Friedel sum was $M+Z$ and thus was 9.

The actual calculations can proceed in one of two directions; (a) the parameters Δ and ϵ_d can be specified with the subsequent calculation of the number of electrons in the virtual state M, ρ_0, and S_0 or (b) M and S_0 can be specified and ρ_0, Δ, and ϵ_d calculated. The latter procedure was chosen with the value for S_0 being that of the straight lines in Fig. 2 ($S_0=-0.061\ T$ μV/°K).

The results for Cu-Ni are listed in Table I. A close relation between the number of electrons in the virtual state and the width of the state is depicted with the state becoming increasingly narrow as it fills. The value of Δ for purely resonant scattering (i.e., $\delta_0=0$) is reasonably consistent with the estimate of $\Delta=0.3$ eV made by Klein and Heeger[18] from specific heat and resistivity data.

Although the general alloying behavior of Ni would lead one to expect $M>9$, the increasingly small values of Δ, as M increases beyond 9, are not consistent with an unmagnetized state. The relation between Δ, M, and the specific assumptions used can be illuminated by considering alternate expressions for $n_d(\epsilon)$. The use of a smaller normalization factor A—and hence an implicit assumption that not all the d electrons of the impurity are involved in forming the virtual state or that more than one virtual state is formed and a limited number of states are not filled—for the Lorentzian $n_d(\epsilon)$ causes the value of Δ to increase for a given value of M. The general pattern of Δ decreasing as M increases does not change. It should be noted that the use of $A<10$ is not easily justified by or related to the splitting of d electron states into t_{2g} and e_g symmetries. Friedel[19] points out that in band-structure calculations for transition metals having bcc structures the division of the density of states curve into peaks and valleys does not represent a one-to-one correspondence with d states and that "the t_{2g} and e_g characters are rather evenly distributed throughout the d band." This same general result would be expected for Ni in Cu.

TABLE I. Residual resistivity and virtual state parameters for Cu-Ni as a function of screening. M = number of electrons in the virtual state. Z = number of excess electrons in the conduction band. $\rho_0=1.25\ \mu\Omega$ cm (experimentally).

		Calculated	Lorentzian		Gaussian	
M	Z $(\equiv 9-M)$	ρ_0 ($\mu\Omega$ cm)	Δ (eV)	$\epsilon_F-\epsilon_d$ (eV)	σ (eV)	$\epsilon_F-\epsilon_d$ (eV)
8.6	+0.4	2.78	0.34	0.72	1.31	1.42
8.8	+0.2	2.22	0.28	0.73	1.37	1.61
9.0	0	1.81	0.24	0.74	1.38	1.76
9.2	−0.2	1.56	0.16	0.64	1.24	1.74
9.3	−0.3	1.53	0.12	0.55	1.08	1.60
9.4	−0.4	1.53	0.086	0.45	0.90	1.41
9.5	−0.5	1.59	0.054	0.34	0.72	1.19

Rather than merely altering the normalization of $n_d(\epsilon)$, its basic form can be changed. Using a Gaussian form, $n_d(\epsilon)\propto e^{-(\epsilon-\epsilon_d)^2/2\sigma^2}$, and normalizing so that 10 electrons can be accommodated, the above analysis was repeated and the results are listed in Table I. The Gaussian form yields larger values of $\epsilon_F-\epsilon_d$, and σ for some values of M is nearly an order of magnitude greater than Δ. Moreover, σ does not monatomically decrease over the range of $8.6\leq M\leq 9.5$. Alteration of the normalization factor, so that less than 10 electrons can be accommodated, increases σ.

With the assumptions used in the calculation, S_0 should be independent of solute concentration and the Gorter-Nordheim relation should be valid for temperatures above $\frac{1}{2}\Theta_R$ (where Θ_R is the resistance Debye temperature and $\frac{1}{2}\Theta_R\sim 160$°K). Figure 4 contains a comparison of the predicted temperature dependence with the measured dependence for several alloy concentrations. The predicted dependence was obtained by using the stated value of S_0, the experimental value of ρ_0 (1.25 $\mu\Omega$ cm/at. %),[20] $S_{\text{th}}=0.0055\ T\ \mu$V/°K,[12] and the resistivity data of White and Woods[21] for ρ_{th}. Above 300°K, ρ_{th} was determined from the curve in Wilson.[22]

Since only one Cu-Co sample was measured, it does not seem justifiable to present a detailed analysis for this system. However, two general features of such an analysis seem worthy of note: (1) S_0 for Cu-Co must be greater than for Cu-Ni, and (2) the values for Δ and σ are smaller than the corresponding values for Δ and σ in the Cu-Ni system. Both of these features are consistent with the general belief that Co in Cu is more nearly magnetized than Ni in Cu.

The above calculation indicates that using a virtual state having parameters consistent with those deduced from other properties can provide a qualitative understanding of the temperature dependence of the TEP of dilute Cu-Ni alloys. Although the approximations used do not seem to justify small refinements which could provide a more quantitative fit to the experi-

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[18] A. P. Klein and A. J. Heeger, Phys. Rev. **144**, 458 (1966).

[19] J. Friedel, in *Theory of Magnetism in Transition Metals*, edited by W. Marshall (Academic Press Inc., New York, 1967), Chap. 8.

[20] J. O. Linde, theses; taken from table of F. J. Blatt, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), Vol. 4, p. 318.

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[22] A. H. Wilson, *The Theory of Metals* (Cambridge University Press, Cambridge, 1953), p. 276.

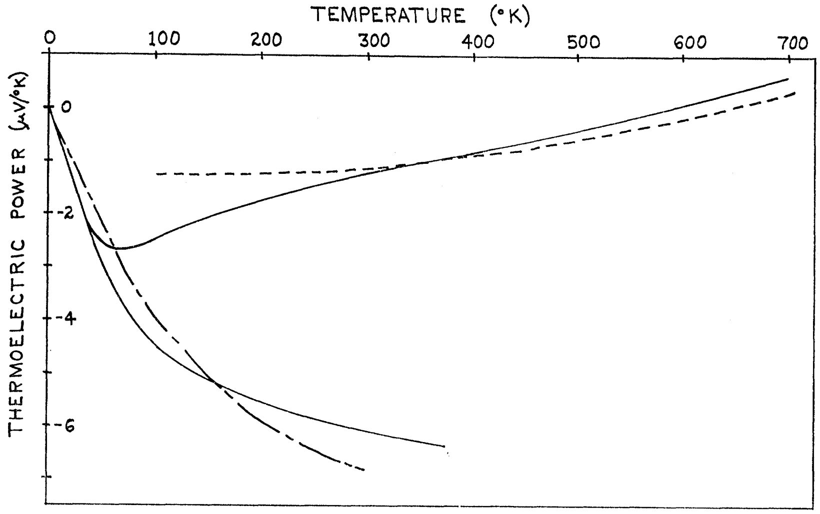

FIG. 4. The temperature dependence of the diffusion TEP for several Cu-Ni alloys. The solid curves are the calculated behavior and the dashed curves are the experimental results of Schroeder *et al.* (Ref. 7) and of Christenson [E. L. Christenson, J. Appl. Phys. 34, 1485 (1963)].

mental TEP data, several general features seem significant enough to merit explicit consideration:

(a) As expected, the unsplit virtual state can have a strong affect upon the TEP. An adequate description of the virtual state requires (or alternatively, provides) a more detailed understanding of how the solute enters into solution. This last point is graphically illustrated in Table I.

(b) This strong affect upon the TEP should also be present when the virtual state is split. Moreover, being a function of δ_0 and δ_2, the sign of this contribution can change. Such a contribution may prove important in understanding the temperature dependence of the TEP for alloys having split virtual states.

The lack of any significant field dependence of the TEP for these alloys is not convincingly ascribed to any single and/or combination of causes. Although several qualitative arguments which lead to the expectation of an effect could be advanced, the explanation of the absence of an effect requires more quantitative arguments which consider such points as (1) valid expressions for the weighting factors which appear in a Gorter-Nordheim relation, (2) the possibility of interference between scattering mechanisms, (3) the relation between spin degeneracy and TEP, and (4) self-consistent formation of a virtual state in the presence of a magnetic field. Such an explanation will not be attempted and, instead, the reader's attention is directed to an investigation completed subsequent to the above. Caplin *et al.*[23] studied the resistivity, the susceptibility, and the TEP of ϵ phase Cu-Zn. Although a moment forms on the Fe as the Cu content of the alloy host is increased, the TEP does not display any anomalous behavior and does not correlate with the magnitude of the moment. The general behavior for this system can be understood by invoking the same considerations used in the above Cu-Ni analysis. The lack of an anomalous contribution to the TEP as a moment forms is consistent with the lack of field dependence reported here.

ACKNOWLEDGMENTS

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[23] A. D. Caplin, C. L. Foiles, and J. Penfold, 1967 International Congress on Magnetism (to be published).