The long-range order parameter, $S$, of pure Cu$_3$Au and two Cu$_3$Au alloys containing 0.64 and 1.5 at. % Co was determined over the temperature range 325 to 387 °C. In principle, the state of order in the ternary alloy $(\text{Cu}_3\text{Au})_1-x\text{Co}_x$ is described by two order parameters $S_1 = A_x - A_\beta$ and $S_2 = B_\beta - B_\delta$ where $A_x$ is the fraction of $\alpha$ sites occupied by $A$ atoms, etc. It is shown that $S_2 - S_1 = C_\alpha - C_\beta = 0$ as long as the Co atoms are randomly distributed on $\alpha$ and $\beta$ sites. The values of $S$ were measured at temperature using Warren's method [Warren, B. E. (1969). *X-Ray Diffraction*. Reading, Mass.: Addison-Wesley] applied to powder samples, employing the $\{110\}$, $\{210\}$, $\{211\}$, and $\{221,300\}$ superstructure and the $\{111\}$, $\{200\}$, $\{220\}$, $\{311\}$, and $\{222\}$ fundamental reflections. The results on pure Cu$_3$Au are in relatively good agreement with earlier measurements, but in poor agreement with theoretical predictions, especially at temperatures above 360 °C. Within the accuracy of the measurements, it is observed that cobalt, which is in solid solution in both alloys, has no effect on the experimentally determined values of $S$. The root-mean-square vibration amplitudes, $\langle u^2 \rangle^{1/2}$, of the Cu and Au atoms at 640 °K were also estimated from the data, and it appeared that $\langle u^2 \rangle_{\text{Cu}}^{1/2}$ decreased slightly with increasing Co content while $\langle u^2 \rangle_{\text{Au}}^{1/2}$ increased.

I. Introduction

As part of a project concerned with the precipitation hardening of ordered and disordered Cu$_3$Au by f.c.c. cobalt-rich precipitates, we believed it necessary to evaluate the influence of Co on the ordering characteristics of Cu$_3$Au. We anticipated that the presence of cobalt in solid solution might affect the long-range order parameter $S$, whereas Co precipitates (Ardell & Hovan, 1972) might influence the kinetics of antiphase domain (APD) growth, either of which could in turn affect the mechanical behavior of the alloys. For example, the addition of Al has already been shown (Chapman & Gillam, 1970) to affect the microhardness of Cu$_3$Au, and the kinetics of APD growth have been shown to be retarded by Al$_2$O$_3$ particles dispersed in Cu$_3$Au (Sastry & Ramaswami, 1974).

After having decided that such measurements were necessary, we felt it would be both interesting and instructive to determine absolute values of $S$ for the Cu$_3$Au-Co solid solutions, as well for pure Cu$_3$Au, without assuming that the thermal vibrations of the Cu and Au atoms are the same. Such measurements are possible, in principle, using Warren's (1969) method on powder samples (or single crystals), which yield values of $S$ and the Debye-Waller factors of the A and B atoms directly and simultaneously. In this paper we describe the results of our investigations of the ordering behavior of Cu$_3$Au containing up to 2 at. % Co in solid solution, exclusive of the effect of Co precipitates on the kinetics of APD growth, which will be published elsewhere. Our results are the only measurements to date of absolute values of $S$ and the Debye-Waller factors of the Cu and Au atoms based on Warren's method, which we compare with older measurements (Keating & Warren, 1951; Chipman, 1956; Schwartz & Cohen, 1965; Gehlen & Cohen, 1969) and existing theories (Cowley, 1950; Kikuchi & Sato, 1974).

II. Theoretical considerations

1. Long-range order in ternary alloys with the Cu$_3$Au type of ordered structure

In a ternary alloy at least two long-range order parameters are necessary to describe the state of order. This can be visualized quite readily from an estimate of the structure factor for a superstructure reflection, $F_S$, of a ternary alloy having the Cu$_3$Au type of ordered structure. Assuming that the atom fractions of A, B, and C type atoms ($X_A$, $X_B$, and $X_C$, respectively, where $X_A + X_B + X_C = 1$) are distributed over two types of sites (denoted here as $\alpha$ and $\beta$ sites with fractions $y_\alpha$ and $y_\beta$, where $y_\alpha + y_\beta = 1$), it can be shown that when $h, k$ and $l$ are mixed $F_S$ is given by

$$ F_S = (A_x - A_\beta)f_A + (B_\beta - B_\delta)f_B + (C_\alpha - C_\beta)f_C, \quad (1) $$

where $f_A$, $f_B$, and $f_C$ are the atomic scattering factors of A, B, and C atoms respectively, and $A_x$ is the fraction of $\alpha$ sites occupied by A atoms, etc. (Murakami, Nakashish & Kachi, 1971). Just as in binary systems (Warren, 1969), $A_x + B_\beta + C_\alpha = 1$, and $A_\beta + B_\delta + C_\beta = 1$. In addition, the fraction of sites occupied by A atoms must equal the fraction $X_A$ of A atoms. Thus (1) can be written as

$$ F_S = S_1 f_A - S_2 f_B + (S_2 - S_1) f_C = S (f_A - f_B) + AS (f_C - f_B), \quad (2) $$

where

$$ S_1 = A_x - A_\beta = (A_x - X_A)/y_\beta = S \quad (3) $$

$$ S_2 = B_\beta - B_\delta = (B_\beta - X_B)/y_\alpha = S_2 \quad (4) $$

$$ S = (S_1 + S_2)/2 \quad (5) $$
It is instructive to consider what the values of $S_1$ and $S_2$ would be in the ternary alloy $(A_xB_y)_{1-x}C_x$ where $X = X_C$ is the concentration of C atoms. It is obvious that $X_A = y_A(1 - X)$ and $X_B = y_B(1 - X)$. Thus we obtain in this case:

$$S_1 = \frac{[A_a - y_A(1 - X)]}{y_B}$$

$$S_2 = \frac{[y_B(1 - X) - B_a]}{y_B}.$$  

The maximum values that $S_1$ and $S_2$ can attain correspond to the case where all $A$ atoms are on $\alpha$-sites and all $B$ atoms on $\beta$ sites, so that $A_a = X_A/y_A = 1 - X$ and $B_a = 0$. Therefore

$$(S_1)_{\text{max}} = 1 - X = (S_2)_{\text{max}}.$$  

It follows from (5) that $C_a = X$ in this case. It is also true that as long as the $C$ atoms are distributed at random on $\alpha$ and $\beta$ sites, $C_a = X$, and the long-range order parameters $S_1$ and $S_2$ will be identical; we believe that this is a reasonable assumption for our $(Cu_3Au)_{1-x}Co_x$ alloys, which we will examine later. Thus (2) reduces to the binary case:

$$F_S = S(f_A - f_B),$$

where $S = A_a - A_B = B_B - B_a$ is Warren’s generalized LRO parameter.*

2. Warren’s method for the determination of $S$

With the use of various approximations discussed by Warren (1969), the diffracted intensity, corrected for multiplicity, Lorentz–polarization, and structure factors, can be written in expanded form for the fundamental ($I_F$) and superlattice ($I_S$) reflections of the Cu$_3$Au type of ordered structure, respectively, as:

$$\ln I_F = \ln K - 2(B - pB)(\sin \theta/\lambda)^2$$

and

$$\ln I_S = \ln K + 2\ln S - 2(B - gB)(\sin \theta/\lambda)^2.$$  

In (10) and (11) $K$ is a proportionality constant, $2B = B_{Cu} + B_{Au}$ and $2B = B_{Cu} - B_{Au}$ where $B_{Cu}$ and $B_{Au}$ are the Debye–Waller temperature factors of Cu and Au respectively, and $p$ and $g$ are functions of the atomic scattering factors as defined in Warren (1969).

* Other definitions of $S$ are, of course, possible. One of the more frequently used definitions is that due to Cowley (1950), denoted here by $S_c$, and given by the formula

$$S_c = \frac{y_A(X_A - X_B)}{X_A}.$$  

With this definition of $S_c$, $F_S$ can be determined from X-ray measurements using the formula (Schwartz & Cohen, 1965):

$$F_S = S_c(X_A X_B / y_A y_B)(y_A - f_A).$$

It is evident, on comparison with (10), that

$$S = S_c X_A X_B / y_A y_B.$$  

While $S_c$ has certain advantages in describing long-range order in binary systems, it cannot be readily generalized for ternary systems.

It is apparent that plots of $\ln (I_F)$ and $\ln (I_S)$ vs. $(\sin \theta/\lambda)^2$ should be linear if $p$ and $g$ are constant. In Cu$_3$Au, $g$ varies from 2.04 to 1.86 as the order of the superstructure reflection increases from $\{110\}$ to $\{221,300\}$. This variation is not large enough to affect the analysis, although it could introduce a small systematic error in the final result. The values of $p$ for the fundamental reflections up to $\{222\}$ are so small (<0.007) that the variation of $p$ with $2\theta$ can safely be ignored. Thus, if the experimentally measured intensities are plotted according to (10) and (11) and the curves are extrapolated to $(\sin \theta/\lambda)^2 = 0$, the difference between the intercepts at $(\sin \theta/\lambda)^2 = 0$, i.e. $(I_0)_S = \ln K + 2\ln S$ and $(I_0)_F = \ln K$, is $2\ln S$. The parameters $B_{Cu}$ and $B_{Au}$ can be calculated from the slopes, $q_F = -2(B - pB)$ and $q_S = -2(B - gB)$, respectively, of the $\ln (I_F)$ and $\ln (I_S)$ vs. $(\sin \theta/\lambda)^2$ plots.

III. Procedures and results

1. Sample preparation

Pure Cu$_3$Au and Cu$_3$Au–Co alloys containing 0.5, 0.64, 1.0, 1.5 and 2.0 at.% Co were prepared from elements of 99.999 % purity. To insure homogeneity, the alloy samples were cold-rolled and then remelted. For a final homogenization the alloys were annealed at 925°C in an H$_2$ atmosphere furnace for about three days.

Filings (~325, <44 µm) were solution-treated for 15 min at 925°C followed by a water quench. The loose powder samples were placed directly in a horizontal stationary high-temperature X-ray furnace mounted on a $\theta$–$\theta$ diffractometer (Wagner, 1969) and heated in an He–25 % H$_2$ atmosphere for 1 h at 410°C, which is above the critical temperature of ordering (~390°C) of Cu$_3$Au (Keating & Warren, 1951). After both heat treatments the Cu$_3$Au–Co filings were found to be non-magnetic, indicating either that the extent of Co precipitation was minimal or that any precipitates that grew were extremely small (< 20 Å in diameter).

Fig. 1. Variation of the lattice constant of Cu$_3$Au at 25°C with dissolved Co.
2. Lattice constant

In order to ascertain further that nearly all the Co (i.e., up to 20% Co) remained in solid solution, the lattice constants $a$ of the disordered alloys (after the second heat-treatment) were determined by plotting the values of $d_{\text{obs}}$ obtained from the Cu $K\alpha$ peak positions of the $\{311\}$, $\{222\}$, $\{331\}$ and $\{420\}$ reflections as a function of $\cos \theta \cot \theta$, and extrapolating the straight line through all data points to $\cos \theta \cot \theta = 0$.

The lattice constant $a$ is plotted as a function of Co concentration in Fig. 1. All points fall on a straight line which extrapolates to a value of $a = 3.58 \text{Å}$ at 100% Co, which is only slightly larger than that of f.c.c. Co (Barrett & Massalski, 1966). In view of the extremely large extrapolation involved, this result may be fortuitous. It nevertheless suggests that up to 20% Co can be dissolved in Cu$_3$Au at 925°C.

3. Critical temperature, $T_c$

The lattice constants of ordered and disordered Cu$_3$Au are significantly different (Keating & Warren, 1951). Upon heating the alloy through $T_c$ the lattice constant abruptly changes as the transformation to disorder occurs. This results in an abrupt change in the position of a convenient high-angle reflection at $T_c$, such as $\{420\}$, when using Cu $K\alpha$ radiation. The discontinuity in the curve of the diffraction angle, $2\theta$, vs $T$ corresponds to $T_c$. When the position of the $\{420\}$ peak maximum was plotted as a function of temperature, it was found that the critical temperature is 390 ± 2°C for all the alloys on which measurements were made. The disappearance of the $\{110\}$ reflections at 390°C confirmed this value of the critical temperature.

4. Long-range order parameter

The intensity profiles used in the analysis were those of the $\{110\}$, $\{210\}$, $\{211\}$, and $\{221, 300\}$ superstructure reflections and the $\{111\}$, $\{200\}$, $\{220\}$, $\{311\}$, and $\{222\}$ fundamental reflections and the corrected X-ray intensities were plotted according to (10) and (11). The values of $\ln (I_F)$ of the fundamental reflections measured at temperatures between 325 and 387°C fell on a straight line since the values of $p$ are small ($p < 0.1$) and $b = (B_{\text{Cu}} - B_{\text{Au}})/2 \approx 0.21$ (see Table 2) is also small. The values of $g$ are not small, i.e. $g \approx 1.9$ and $2gb(\sin \theta/\lambda)^2 = 0.82 (\sin \theta/\lambda)^2$. Nevertheless, the approximation

$$1 + 2gb(\sin \theta/\lambda)^2 = \exp [2gb(\sin \theta/\lambda)^2]$$

used in Warren's method is good to within 2% in the range $(\sin \theta/\lambda)^2 \leq 0.2$ considered in the present experiments, thus permitting us to draw straight lines through the values $\ln (I_F)$ of the superstructure reflections when plotted as a function of $(\sin \theta/\lambda)^2$. An example of these graphical procedures, illustrating the results on the 0.64% Co alloy is shown in Fig. 2. The results on the other alloys were similar. The scatter increased with increasing Co content (see Table 1).

The slopes, $q_F$ and $q_S$, and the intercepts, $(I_0)_F$ and $(I_0)_S$, were calculated by a least-squares analysis of all data points, and are given in Table 1, together with the values of the root-mean-square error of the slopes, $m_s$, the intercepts, $m_i$, and the correlation coefficient, $r$. The experimentally determined values of $S$, evaluated from the intercepts of the curves, are also given in Table 1 together with the error $\Delta S = S[(m_1)_F + (m_1)_S]/2$. The variation of $S$ as a function of temperature is shown in Fig. 3.
Table 1. Values of the slopes \((q_F \text{ and } q_s)\) and the intercepts \((I_0)_F \text{ and } (I_0)_s\)

Also given are the root-mean-square error of the slopes, \(m_s\), the intercepts, \(m_0\), the correlation coefficient, \(r\), evaluated by least-squares analysis, the values of the long-range order parameter \(S\) and the associated error \(\delta S\).

<table>
<thead>
<tr>
<th>Alloy (at. % Co)</th>
<th>Reflection*</th>
<th>(T (°C))</th>
<th>Slope (m_s)</th>
<th>Intercept (m_0)</th>
<th>(r)</th>
<th>(S)</th>
<th>(\delta S)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>(F)</td>
<td>325-387</td>
<td>-2.166</td>
<td>0.134</td>
<td>1.019</td>
<td>0.020</td>
<td>-0.994</td>
</tr>
<tr>
<td></td>
<td>(S)</td>
<td>325</td>
<td>-1.125</td>
<td>0.085</td>
<td>0.844</td>
<td>0.009</td>
<td>-0.999</td>
</tr>
<tr>
<td></td>
<td></td>
<td>365</td>
<td>-1.111</td>
<td>0.180</td>
<td>0.678</td>
<td>0.019</td>
<td>-0.975</td>
</tr>
<tr>
<td></td>
<td></td>
<td>380</td>
<td>-1.329</td>
<td>0.424</td>
<td>0.630</td>
<td>0.045</td>
<td>-0.911</td>
</tr>
<tr>
<td></td>
<td></td>
<td>387</td>
<td>-0.910</td>
<td>0.209</td>
<td>0.489</td>
<td>0.022</td>
<td>-0.951</td>
</tr>
<tr>
<td>0.64</td>
<td>(F)</td>
<td>325-387</td>
<td>-2.220</td>
<td>0.271</td>
<td>0.991</td>
<td>0.040</td>
<td>-0.978</td>
</tr>
<tr>
<td></td>
<td>(S)</td>
<td>325</td>
<td>-1.861</td>
<td>0.310</td>
<td>0.816</td>
<td>0.033</td>
<td>-0.973</td>
</tr>
<tr>
<td></td>
<td></td>
<td>350</td>
<td>-1.676</td>
<td>0.372</td>
<td>0.743</td>
<td>0.040</td>
<td>-0.954</td>
</tr>
<tr>
<td></td>
<td></td>
<td>365</td>
<td>-1.797</td>
<td>0.730</td>
<td>0.676</td>
<td>0.080</td>
<td>-0.865</td>
</tr>
<tr>
<td></td>
<td></td>
<td>380</td>
<td>-1.667</td>
<td>0.342</td>
<td>0.518</td>
<td>0.037</td>
<td>-0.960</td>
</tr>
<tr>
<td></td>
<td></td>
<td>385</td>
<td>-1.477</td>
<td>0.103</td>
<td>0.399</td>
<td>0.011</td>
<td>-0.995</td>
</tr>
<tr>
<td></td>
<td></td>
<td>387</td>
<td>-1.110</td>
<td>0.399</td>
<td>0.303</td>
<td>0.043</td>
<td>-0.890</td>
</tr>
<tr>
<td>1.5</td>
<td>(F)</td>
<td>325-385</td>
<td>-2.270</td>
<td>0.212</td>
<td>1.024</td>
<td>0.031</td>
<td>-0.887</td>
</tr>
<tr>
<td></td>
<td>(S)</td>
<td>325</td>
<td>-2.092</td>
<td>0.595</td>
<td>0.866</td>
<td>0.064</td>
<td>-0.927</td>
</tr>
<tr>
<td></td>
<td></td>
<td>365</td>
<td>-1.809</td>
<td>0.860</td>
<td>0.690</td>
<td>0.093</td>
<td>-0.830</td>
</tr>
<tr>
<td></td>
<td></td>
<td>380</td>
<td>-2.040</td>
<td>0.410</td>
<td>0.594</td>
<td>0.044</td>
<td>-0.962</td>
</tr>
<tr>
<td></td>
<td></td>
<td>385</td>
<td>-2.015</td>
<td>0.710</td>
<td>0.432</td>
<td>0.077</td>
<td>-0.895</td>
</tr>
</tbody>
</table>

* \(F\) – fundamental, \(S\) – superlattice.

Within the limits of the experimental errors of the present measurements, there seems to be no effect of Co on the critical temperature \(T_c\) (= 390°C) and the LRO parameter \(S\). The present data are in relatively good agreement with the earlier results of Keating & Warren (1951), also shown in Fig. 3, Chipman (1956) \((S=0.8 \text{ quenched from 380°C})\) and Schwartz & Cohen (1956) \((S=0.8 \text{ quenched from 383°C})\) close to the critical temperature. The variation of \(S\) with \(T\) is in poor agreement, especially at the higher temperatures, with the theoretical prediction of Cowley (1950), (see Fig. 3). At lower temperatures, our curve appears to merge closer with the predictions of the model of Kikuchi & Sato (1974), (see Fig. 3), which is based on the cluster-variation method.

5. Debye–Waller factors and atomic vibration amplitudes

The Debye–Waller factors \(B_{\text{Cu}}\) and \(B_{\text{Au}}\) were calculated from the slopes of the curves given in Table 1. The resulting values of the \(B\)'s are presented in Table 2 for pure \(\text{Cu}_{3}\)\(\text{Au}\) and the alloys containing 0.64 and 1.5 at. % Co. The parameters \(B_{\text{Cu}}\) and \(B_{\text{Au}}\) can be converted to the root-mean-square vibration amplitudes \(\langle u^2 \rangle^{1/2}\) using the formula \(B = 8\pi^2\langle u^2 \rangle\). The values of \(\langle u^2 \rangle^{1/2}\) are given in Table 2. It should be pointed out that the values of \(B\) in Table 2 are average values of all measurements made at temperatures between 325 and 380°C, and thus correspond to approximately \(T = 640°C\). It is of interest to calculate a value of \(\langle u^2 \rangle^{1/2}\) corresponding to \(T = 300°C\) \((i.e. \text{room temperature})\). This was done by assuming a linear dependence of \(B\) upon the absolute temperature, \(T\), thus neglecting the small dependence of the Debye function on \(T\) in this temperature range. The values of \(\langle u^2 \rangle^{1/2}\) at 300°C are also given in Table 2. We find for \(\text{Cu}_{3}\)\(\text{Au}\) the values \(\langle u^2 \rangle^{1/2}_{\text{Cu}} = 0.091 \text{ Å}\) and \(\langle u^2 \rangle^{1/2}_{\text{Au}} = 0.069 \text{ Å}\) which are in good agreement with those of Chipman (1956) \(\langle u^2 \rangle^{1/2}_{\text{Cu}} = 0.093 \text{ Å}\), \(\langle u^2 \rangle^{1/2}_{\text{Au}} = 0.069 \text{ Å}\), but slightly lower than those reported by Gehlen & Cohen (1969) \(\langle u^2 \rangle^{1/2}_{\text{Cu}} = 0.098 \text{ Å}, \langle u^2 \rangle^{1/2}_{\text{Au}} = 0.083 \text{ Å}\). As shown in Table 2, the values of \(\langle u^2 \rangle^{1/2}_{\text{Cu}}\) appear to decrease slightly with increasing Co content while the values \(\langle u^2 \rangle^{1/2}_{\text{Au}}\) increase. The effect is small, however, and just within the limits of our experimental errors.

IV. Discussion

Our results on pure \(\text{Cu}_{3}\)\(\text{Au}\) are in very good agreement with those of earlier investigators, thus demonstrating that Warren’s method can be used for reliable determinations of \(S\) and atomic vibration amplitudes. However, ordered \(\text{Cu}_{3}\)\(\text{Au}\) is a particularly favorable alloy for testing Warren’s method since the scattering factors of \(\text{Cu}\) and \(\text{Au}\), upon which the parameters \(p\) and

<table>
<thead>
<tr>
<th>% Co</th>
<th>(T (K))</th>
<th>(B_{\text{Cu}})</th>
<th>(B_{\text{Au}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>640</td>
<td>(\langle u^2 \rangle^{1/2}_{\text{Cu}})</td>
<td>0.132</td>
<td>0.128</td>
</tr>
<tr>
<td>300</td>
<td>(\langle u^2 \rangle^{1/2}_{\text{Cu}})</td>
<td>0.091</td>
<td>0.088</td>
</tr>
</tbody>
</table>
g depend, differ considerably. Because of this, the value of g is relatively small, thereby introducing only a small error into the analysis. While the method does not have the inherent accuracy of, for example, that used by Schwartz & Cohen (1965), it is far simpler to use and yields nearly identical results.

While there can be no quarrel with treating Cu₃Au-Co alloys as ternary alloys, as we have done, it is tempting to consider the consequence of regarding these alloys as quasi-binary because the alloys are dilute in Co. Since the scattering factors for Cu and Co are quite similar it is not unrealistic to treat the alloy as an off-stoichiometric one of the type (Cu-Co)ₙAuₙₓ, where \( X_n = (1-X)/4 \) and \( X_A = (3+X)/4 \). In this case there is only one value of S, denoted here by \( S_{q-b} \), that need be dealt with; according to Warren’s generalized definition of S, the maximum value of \( S_{q-b} = 1 - X \), which is identical to \( (S_1)_{\text{max}} \) and \( (S_2)_{\text{max}} \) according to (8). Even if we were to assume that all the Co atoms behave like Au in a quasi-binary scheme, which is less physically appealing, the maximum value of \( S_{q-b} \) would still be \( 1 - X \). What cannot be overlooked, however, is that \( S_1 = S_2 = S \) only when the Co atoms are randomly distributed among the Cu and Au sites; it is impossible for this condition and either of the quasi-binary descriptions discussed above to be satisfied simultaneously. In fact, it is not difficult to show that when the Co atoms are randomly distributed among the Cu and Au sites in a (Cu-Co)ₙ(Au-Co) quasi-binary alloy, the maximum value of S is unity. These considerations suggest that a ternary alloy of the type dealt with in our study, no matter how dilute, cannot be properly treated as a quasi-binary alloy.

In closing, we examine the validity of the assumption that the Co atoms are randomly distributed among the Cu and Au sites, which leads to the condition \( S_1 = S_2 = S \) used in our evaluation of the data. Presumably, if the Co atoms preferentially occupy one or the other sub-lattice, the value of S derived from the analysis would be artificial in the sense that it could not completely describe the state of order in the system. If we consider the effect on S of the two extreme cases, i.e., all the Co atoms occupying either Cu or Au sites, we find that the effect on the measured values of S is undetectable. It follows from (5) that, when all the Co atoms occupy Cu sites, \( (S_2 - S_1)_{\text{Cu}} = 4X/3 \), whereas, when all the Co atoms occupy Au sites, \( (S_2 - S_1)_{\text{Au}} = -4X \). In the 1.5% Co alloy \( (S_2 - S_1)_{\text{Cu}} = 0.02 \) and \( (S_2 - S_1)_{\text{Au}} = -0.06 \). In spite of the fact that these differences are not negligible, the effect on the measured values of S is. This can be seen on rewriting (2) as

\[
F_S = S(f_A - f_B) \left( 1 + \frac{\Delta S}{S} \frac{f_C - f_B}{f_A - f_B} \right).
\]

Taking \( \Delta S = 0.06 \), the term \( (\Delta S/S)[(f_C - f_B)/(f_A - f_B)] \approx 0.002 \). Therefore, it is clearly impossible to determine the disposition of Co atoms on the Cu and Au sites from our experimental results.

We are grateful to the National Science Foundation for financial support of this project under Grant No. GH-31818.

References