

# THE ELASTIC CONSTANTS OF COPPER ALLOYS\*

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The elastic constants of single crystals of dilute solutions of Al, Si, Zn, Ga and Ge in Cu have been determined by the ultrasonic pulse method. Alloying causes a decrease in all the fundamental constants,  $C_{44}$ ,  $(C_{11}-C_{12})/2$  and  $(C_{11}+2C_{12})/3$ , the relative decrease in  $(C_{11}-C_{12})/2$  being the greatest. The changes in shear constants  $C_{44}$  and  $(C_{11}-C_{12})/2$  have been interpreted in terms of the known electrostatic and ionic contributions to these constants for copper. The large decrease in  $(C_{11}-C_{12})/2$  is attributed to the change in ion-ion interactions, upon alloying. The electrostatic shear stiffness of an alloy is found to be nearly equal to the electrostatic term for copper times the square of the electron atom ratio.

## LES CONSTANTES D'ÉLASTICITÉ DES ALLIAGES DE CUIVRE

On a déterminé les constantes d'élasticité de solutions diluées de Al, Si, Zn, Ga et Ge dans le cuivre, par la méthode de pulsation ultra-sonique. L'addition de ces éléments produit une diminution de toutes les constantes fondamentales,  $C_{44}$ ,  $(C_{11}-C_{12})/2$  et  $(C_{11}+2C_{12})/3$ ; la diminution relative de  $(C_{11}-C_{12})/2$  étant la plus importante.

Les changements des constantes de cisaillement  $C_{44}$  et  $(C_{11}-C_{12})/2$  furent interprétés en termes des contributions électrostatiques et ioniques, connues, à ces constantes pour le cuivre. La diminution importante de  $(C_{11}-C_{12})/2$  est attribuée à la modification des interactions entre les ions, lors de l'addition des éléments d'alliage. Il fut constaté que la rigidité électrostatique en cisaillement d'un alliage était approximativement égale au terme électrostatique pour le cuivre multiplié par le carré du rapport du nombre d'électrons au nombre d'atomes.

## DIE ELASTIZITÄTSKONSTANTEN VON KUPFERLEGIERUNGEN

Die Elastizitätskonstanten von verdünnten Lösungen von Al, Si, Zn, Ga und Ge in Cu wurden mit Hilfe einer Ultraschallimpulsmethode bestimmt. Legieren rief eine Verringerung in allen fundamentalen Konstanten,  $C_{44}$ ,  $(C_{11}-C_{12})/2$  und  $(C_{11}+2C_{12})/3$  hervor. Die relative Verringerung war in  $(C_{11}-C_{12})/2$  am grössten. Die Veränderungen in den Scherungskonstanten  $C_{44}$  und  $(C_{11}-C_{12})/2$  wurden auf Grund der im Kupfer bekannten Beiträge der elektrostatischen und Ionenkräften zu diesen Konstanten gedeutet. Die starke Abnahme von  $(C_{11}-C_{12})/2$  wird den beim Legieren erfolgenden Änderungen in den Ion-Ion Wechselwirkungen zugeschrieben. Es wurde gefunden, dass die elektrostatische Scherungssteifheit einer Legierung nahezu gleich dem elektrostatischen Glied für Kupfer mal dem Quadrat des Elektron: Atom Verhältnisses ist.

## Introduction

The elastic constants of a single crystal are of interest because of the information they give about the nature of the binding forces in solids. The elastic constants of some simple crystals have been investigated both theoretically and experimentally.

Of interest here is the calculation [1] of the elastic constants of copper from fundamental principles and the recent dynamic [2] measurement of these same constants. More recently, one of the authors [3] has investigated the elastic constants of a Cu-4 per cent Si single crystal and has interpreted the change in shear constants starting from Fuchs' theory of the elastic constants of copper. The present work is an extension of this type of analysis to other copper-alloy single crystals.

## Experimental Procedure

Copper alloy single crystals were prepared in a manner similar to that previously described [4].

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Briefly, the method, which is a modification of the Bridgman technique, is to slowly cool a hydrogen atmosphere furnace through the solidus point of the alloy, the alloy charge, contained in a graphite crucible, being placed in a region of high temperature gradient.

The alloys were prepared by melting the solvent in a large graphite crucible and then rapidly adding the solute. Except when adding the solute, the carbon crucible was covered, thus ensuring a reducing atmosphere. After holding the charge at an elevated temperature for some time, a portion of it was cast into one of the small crucibles used for crystal growth. The cast ingot was cleaned in nitric acid and the inside of the graphite crucible was polished smooth before the ingot in its crucible was placed in the crystal growing furnace.

After removal from the crystal growing furnace, the ingots were etched in nitric acid to develop any grain boundaries. A back reflection Laue photograph taken with the X-ray beam parallel to the cylinder axis of the ingot determined the orientation of the ingot. A small cylinder to be used for the acoustic measurement was then sawed from the ingot using a thin water-cooled abrasive wheel. The end faces of this cylinder were surface ground

until flat and parallel and then given a metallographic polish. Two small disks were also sawed from the ingot adjacent to each face of the cylinder. Chemical analyses of these disks by the James H. Herron Company gave an accurate determination of the composition of the cylinder.

The longitudinal and transverse acoustic wave velocities were measured by the pulsed ultrasonic method [5]. The crystal cylinders were cut from selected ingots with their axes as close as convenient to the [110] direction. Near this direction the fundamental shear constants are measured nearly directly and the numerical reduction of the data converges rapidly.

After the velocity measurements had been completed on a crystal cylinder, one of its polished faces was electropolished to remove the cold worked surface layer. A back reflection Laue photograph was then taken from this polished face and orientation of the crystal cylinder then determined using the least squares method [4].

The elastic constants of the crystal were then calculated using the approximation method [6]. The results of these calculations for various copper alloys are shown in Table I, where Zener's notation for the fundamental shear constants  $C = C_{44}$  and  $C' = (C_{11} - C_{12})/2$  has been used. Also included in Table I are the elastic constants for copper [2].

In order to calculate the elastic constants for a given crystal, one must also know its density. The densities used to obtain the elastic constants in Table I were computed from the composition as determined by chemical analysis and the lattice parameter-composition data in the literature [7].

Several things are evident upon examination of Table I. In general, all three fundamental elastic constants,  $C$ ,  $C'$ ,  $K^{-1}$  decrease upon alloying. This is in qualitative agreement with the work of Smith [8], and Koster and Rauscher [9] in which Young's modulus was found to decrease when a solute was introduced into copper. The constant most affected is  $C'$ , where the percentage change upon alloying is about five times as great as for the other constants. The elastic anisotropy,  $C/C'$ , increases with composition upon alloying, increasing relatively faster for the higher valence solutes.

Measurement of the acoustic wave velocities is felt to be good to 0.5 per cent. The elastic constants appearing in Table I are then thought to be good to 1.0 per cent, since the square of the velocity enters into the calculation of the elastic constants.

TABLE I  
ELASTIC CONSTANTS OF COPPER ALLOYS IN UNITS OF  $10^{12}$   
DYNE CM<sup>-2</sup>

(The elastic constants of pure copper as measured by Lazarus (2) are included for comparison.)

Alloy	Atom composi- tion (per cent)	$C = C_{44}$	$C' =$ $(C_{11} - C_{12})/2$	$K^{-1} =$ $(C_{11} + 2C_{12})/3$	Aniso- tropy $C/C'$
Cu	0	0.756	0.237	1.396	3.19
CuZn	4.59	0.742	0.221	1.339	3.36
CuAl	4.81	0.749	0.221	1.363	3.39
	9.98	0.766	0.209	1.316	3.66
CuGa	1.58	0.743	0.229	1.345	3.25
	4.15	0.741	0.221	1.357	3.36
CuSi	4.17	0.748	0.218	1.387	3.43
	*5.16	0.749	0.210	1.328	3.56
	7.69	0.741	0.197	1.395	3.67
CuGe	1.03	0.750	0.228	1.362	3.29
	1.71	0.750	0.224	1.332	3.35

\*The elastic constants of this alloy crystal have been reported previously [3]. However a new chemical analysis shows that the composition is actually different than previously stated. The composition is used to calculate the density and hence the elastic constants so these quantities have been changed correspondingly in the current listing.

In the course of growing these alloy crystals some crystals resulted in which segregation was evident. These segregated crystals were all characterized by the following:

1. A wood-grain like appearance on the sides of the ingot and appearance of a fine rectangular grid on the polished faces of the crystal cylinders. This grid etched in nitric acid to a corresponding system of etch pits on such crystals.

2. Attenuation so high that accurate velocity measurement was impossible.

3. Uniformly poor back reflection Laue photographs.

These segregated crystals were useless for the elastic constant work.

Many specimens were produced, however, *without* these three obvious characteristics although there was no major change in growing technique; such good specimens were used for the present work. It may be commented in this connection that the acoustic test for segregation may be expected to be sensitive. A stiffness variation results from a composition variation and while the former is small, the visible periodicity of the composition

variation in bad crystals was around one-half millimeter which is just the acoustic wave length. High acoustic scattering leading to high attenuation, as observed in bad crystals, is just what one expects. On the other hand the good crystals exhibited attenuation which was indistinguishable from that of pure metals. It is felt, therefore, that the results of this paper are descriptive of alloy crystals of at least small segregation.

It is proper to enquire, however, what would be the effect of segregation on the measured elastic constants and therefore on the conclusions of this paper. For this purpose one may compare an ideal alloy crystal of uniform composition  $x$  with a model of a segregated crystal. The latter may be taken for simplicity as consisting half of material of composition  $2x$  and half with composition zero. The velocities which would be "observed" may be calculated by assuming the conclusions made in this paper, and the two "crystals" will be found to be indeed different. The observed velocity in these experiments is a time average whereas what one would like to have is the space average, or better, the square of the velocity averaged over space. Since *averages* are involved, however, one does not expect more than a second order effect and this turns out to be the case. The detailed arithmetic for the comparison described above shows that the error arising in segregation is less than the ordinary experimental uncertainty of velocity measurement which has been stated.

The large scale homogeneity of the alloy crystals is best measured by the chemical analyses. The first crystals measured were the low and high composition Si alloys. From the three inch cropped ingots four thin disks were taken for analysis at roughly even intervals along the length. For the low Si crystal the chemical results were 1.84, 1.93, 1.88, 1.76 and for the high Si crystal, 3.44, 3.45, 3.64, 3.74, expressed in weight per cent Si. This homogeneity was felt to be good enough and subsequently the analyses were confined to disks cut adjacent to each end of the acoustic specimen, which, it should be noted was only one centimeter long. As it turned out most of the other crystals appeared to be better than the figures just given for Si. Thus the analyses quoted in pairs for the various specimens were as follows: Al, 2.09–2.10, 4.48–4.49; Ga, 1.69–1.77, 4.51–4.54; Ge, 1.16–1.18, 1.90–1.78; Si, 1.84–1.93, 3.45–3.64. For the reason named later the germanium analysis was questioned and so the whole acoustic specimen of the higher composition was analyzed, yielding the value 1.95,

which was used. The middle Cu–Si specimen was a gift to this laboratory and was in the form of a large diameter disk. Our own analysis of a single piece taken from the specimen next to the acoustic path yielded the value quoted. The Cu–Zn specimen was a gift from Horizons Incorporated of Cleveland, Ohio, and our single analysis agreed with theirs at 4.72 weight per cent. The uncertainty in composition indicated by these analyses is insignificant in its effect on the elastic constants of the specimens.

### Interpretation

Fuchs' theoretical calculation [1] of the elastic shear constants starts by writing the energy per atom of the crystal as:

$$(1) \quad U = U_E + U_I + U \text{ (volume only)}$$

Here  $U_E$  represents the electrostatic potential energy of an f.c.c. structure of positive point charges imbedded in a uniform negative charge distribution. The term  $U_I$  is the repulsive exchange interaction energy between closed shell ion cores.

The shear constants  $C = C_{44}$  and  $C' = (C_{11} - C_{12})/2$  are calculated by imagining an appropriately strained crystal, calculating the energy in the strained state, and differentiating twice with respect to the shear strain. Only the energy terms  $U_E$  and  $U_I$  contribute to the elastic shear constants since the other terms are assumed to be functions of volume only. The resulting four terms have been called the electrostatic stiffnesses  $C_E$ ,  $C'_E$  and the ionic stiffnesses  $C_I$ ,  $C'_I$ . The electrostatic stiffnesses  $C_E$ ,  $C'_E$  are the contributions from the electrostatic energy to the elastic shear constants  $C$  and  $C'$ . In the actual calculation of the electrostatic stiffnesses, the only contribution [10] is from the lattice of positive point charges, the uniform negative charge distribution not contributing to the shear constants. The ionic stiffnesses are the contributions to  $C$  and  $C'$  from the ion exchange energy. The numerical results of Fuchs are  $C_E = 0.257$ ,  $C'_E = 0.0286$ ,  $C_I = 0.63$ ,  $C'_I = 0.225$ , all in units of  $10^{12}$  dyne  $\text{cm}^{-2}$ .

The analytical results of Fuchs for the electrostatic stiffnesses are

$$(2) \quad C_E = 0.9478(2e^2/a^4) \quad C'_E = 0.1058(2e^2/a^4)$$

where  $a$  is the lattice parameter.

The ionic stiffnesses may be written as

$$(3) \quad C_I = 2a^{-3}y(y-3)W \quad C'_I = a^{-3}y(y-7)W$$

where  $W$  is the repulsive energy of a single pair of

ions which has been assumed by us to have the exponential form ( $W = Ae^{-r/\rho}$ ).

Some simple assumptions [3], about the effects of alloying have been made previously in considering the elastic constants of a copper-silicon alloy. The electrostatic stiffnesses contain the charge squared, and it is reasonable to assume that these terms for the alloy will be given by equation (2), multiplied by a factor  $Z^2$ . This statement may be regarded as a definition of  $Z$  which for the moment may be regarded as an empirical parameter.

Equation (3) shows that the ionic stiffnesses are proportional to the bond energy  $W$ . Simply then, in the dilute range, the effect on these terms of adding solute atoms would be proportional to the number of solute atoms added, that is to the atomic concentration,  $x$ . The proportionality constant, denoted  $\alpha$ , will then be an empirical parameter which measures the ionic effect. Since the exponential  $W$  will control both these terms a single parameter  $\alpha$  has been assumed to be adequate.

According to the above then, the shear constants for the alloy will be given as

$$(4) \quad \begin{aligned} C &= C_E Z^2 + C_I(1 + \alpha x), \\ C' &= C'_E Z^2 + C'_I(1 + \alpha x). \end{aligned}$$

The changes in elastic constants due to alloying will be, of course,

$$(5) \quad \begin{aligned} \Delta C &= C_E(Z^2 - 1) + C'_I \alpha x, \\ \Delta C' &= C'_E(Z^2 - 1) + C'_I \alpha x. \end{aligned}$$

However, all of the solutes considered expand the copper lattice upon alloying, some more than others. From equation (2) it may be shown that the changes in the electrostatic stiffnesses due to a change in lattice constant are

$$(6) \quad \delta C_E / C_E = -4\delta a/a \quad \delta C'_E / C'_E = -4\delta a/a$$

Similarly [3], the changes in the ionic stiffnesses are

$$(7) \quad \delta C_I / C_I = -17.8\delta a/a \quad \delta C'_I / C'_I = -17.3\delta a/a$$

The change in the shear constant due to a change in lattice constant would then be the sum of the changes in electrostatic and ionic stiffnesses. The difference between this calculated change owing to lattice parameter change and the observed change in elastic shear constant is ascribed to the effect of alloying per se, i.e., the insertion of foreign atoms into the lattice. These latter differences are the quantities  $\Delta C$  and  $\Delta C'$  which have been used in equation (5). Similarly, the ionic and

electrostatic stiffnesses introduced into equation (5) are those for an expanded lattice, calculated according to equation (6) and equation (7).

Thus, in recapitulation, the values for  $\Delta C$ ,  $\Delta C'$ ,  $C_E$ ,  $C'_E$ ,  $C_I$ ,  $C'_I$  are all adjusted for the change in lattice parameter upon alloying. When these values are introduced into equation (5)  $Z$  and  $\alpha$  may be determined. Then the values of  $Z$  and  $\alpha$  which appear in Table II are expected to be characteristic of introducing solute atoms into an expanded copper lattice. Thus they describe the effect of alloying alone.

From inspection of Table II, it is clearly evident that the empirical parameter  $Z$  very nearly equals the electron atom ratio  $q$ , and one is justified in concluding on an *experimental* basis that these quantities are equal. One would expect the parameter  $\alpha$  to be nearly independent of composition for the specimens in a given alloy system. We feel that Table II shows that  $\alpha$  is indeed independent

TABLE II  
THE PARAMETERS  $Z$  AND  $\alpha$  FOR COPPER ALLOYS

Alloy	Atom composition (per cent)	$\alpha$	$Z$	$q$ , electron-atom ratio
CuZn	4.59	-1.0	1.08	1.05
CuAl	4.81	-0.9	1.11	1.10
	9.98	-0.8	1.25	1.20
CuGa	1.58	-1.4	1.03	1.03
	4.15	-1.0	1.09	1.08
CuSi	4.17	-2.5	1.13	1.13
	5.16	-3.0	1.18	1.16
	7.69	-3.0	1.25	1.23
CuGe	1.03	-3.6	1.06	1.03
	1.71	-3.3	1.09	1.05

of composition within the experimental error. Thus the reasonable behaviour of both parameters is held to be justification for the present empirical analysis.

The experimental values of  $Z$  and  $\alpha$  are a synthesis of a large number of physical quantities coming from both experimental and theoretical sources. It hardly seems profitable to discuss in detail the accuracy of the final parameters other than to say that it is felt that  $(Z^2 - 1)$  and  $\alpha$  are good to about five per cent as implied in Table II. The chemical composition is probably the most important single measurement entering

into these results appearing as it does in the density, as a coefficient of  $\alpha$ , and in  $q$  the quantity with which  $Z$  is compared. In this connection it will be noted that in the Cu-Ge alloys  $Z$  departs most markedly from  $q$ , and that  $\alpha$  is exceptionally high. A higher Ge composition would put *both* parameters more nearly in line with those for the other alloys. The chemical analysis of these specimens was however repeated with essential agreement with the first analysis and the results are reported as found.

This analysis has been carried out with Fuchs' theoretical stiffnesses as the starting point. The electrostatic terms are reasonably sure both physically and in respect to agreement with experiment, notably in the case of sodium. The theoretical ionic terms seem to us to be quite questionable, however, both from the point of view of the original theory, and because they do not in fact give the actual elastic constants of copper at room temperatures. (The agreement at absolute zero is probably better.) However, there seems to be little doubt that physically the two effects, electrostatic and ionic, are present and that the ionic effect enters in about the *relative* magnitudes of the Fuchs values, and that it varies rapidly with  $r$  as supposed here. These properties are all that are actually required by the present analysis. It actually makes little difference, for example, whether one makes the lattice parameter correction or not, or whether one uses the Fuchs ionic stiffnesses or empirical ones obtained by subtracting the electrostatic terms from the experimental constants of copper.

### Discussion

It was first pointed out by Jones [11] and Hume-Rothery [12] that the electron-atom ratio  $q$  is the important quantity determining the limit of solid solubility in these alloys and in others. The electron-atom ratio here again appears to have an important role, this time determining the electrostatic stiffnesses of these alloys.

The electrostatic shear stiffnesses of pure copper were calculated by Fuchs on the basis of an assumed model of positive point charges imbedded in a *uniform* valence electron charge distribution. Our result, that  $Z \cong q$ , can be shown to be reasonable, as a first approximation, by extending the Fuchs model to the substitutional alloy. The alloy picture then is that of an array of positive point charges imbedded in again a negative charge distribution which is uniform at a density now of  $q$

electrons per atomic volume. The positive charges statistically have the value  $V_A = 1$ , the solvent valence, and  $V_B$  the solute valence. As previously stated the only contribution to the shear stiffnesses is from the point charges when the negative charge is uniformly distributed. The Coulomb energy of the positive charges will be a sum of terms  $V_i V_j / R_{ij}$  which must be differentiated twice to give the stiffnesses. If we consider all pairs of ions at a given spacing  $R_{ij}$  and in a given orientation, it is easy to show that  $\overline{V_i V_j} = q^2$ . This conclusion holds for all sets of pairs and hence the electrostatic stiffnesses of the model are given directly by  $C = C_0 q^2$ , and therefore  $Z = q$ . The important restriction of this argument is that the valence charge must be uniform.

With the experimental result that  $Z \cong q$  in mind it is tempting to reverse the above reasoning and conclude that the valence electron charge is indeed uniformly distributed in these copper alloys. The validity of the reverse reasoning is questionable, and the resulting conclusion is obviously false on physical grounds. It has been long recognized that in a substitutional alloy the valence charge must pile up in the vicinity of the more highly charged solute ions, effectively screening their excess charge in part [13]. With the solute ion charge partially screened one would be led to expect as a second crude approximation of the model for the alloy that  $Z$  should be less than  $q$ . However such considerations take no account of redistribution of the screening charge and the resulting possible stiffness contribution when the crystal is sheared. In any case the present results indicate that if anything  $Z$  is slightly greater than  $q$ . The problem of interpreting the observed  $Z$ 's in any but a crude manner would appear to be difficult. We are inclined to avoid a definite conclusion, merely commenting that the present result suggests to us a more uniform valence charge distribution than we had supposed beforehand.

The values of  $\alpha$  appearing in Table II are all negative. This behaviour is reasonable since it is the *ion core* interaction which must be considered in the stiffness equations, and the solute ion cores  $\text{Zn}^{+2}$ ,  $\text{Ga}^{+3}$ , and  $\text{Ge}^{+4}$  must surely be smaller than  $\text{Cu}^{+1}$ . For this series of ion cores the electron configuration is identical with only the nuclear charge changing in the series. The ion cores  $\text{Al}^{+3}$  and  $\text{Si}^{+4}$  have a different electron configuration from  $\text{Cu}^{+1}$  but the net ion charge makes for a smaller core with these solutes also. Hartree cal-

culations have been carried out [14] for the ions  $\text{Cu}^{+1}$ ,  $\text{Ga}^{+3}$ ,  $\text{Al}^{+3}$ ,  $\text{Si}^{+4}$  and confirm these statements. The charge distributions for all three solute ions are smaller than  $\text{Cu}^{+1}$ ;  $\text{Si}^{+4}$  is smaller than  $\text{Al}^{+3}$ ; and furthermore  $\text{Al}^{+3}$  is smaller than  $\text{Ga}^{+3}$ .

The parameter  $\alpha$  is increasingly negative in the sequence Zn, Ga, Ge and in the sequence Al, Si as previously predicted [3] on the basis of just the reasoning quoted above. However,  $\alpha$  also decreases in the order Al, Ga and in the order Si, Ge, contrary to the previous prediction and contrary to what one would expect simply from the sizes of the Hartree charge distributions for  $\text{Al}^{+3}$  and  $\text{Ga}^{+3}$ .

One model used by Smith and Burns for estimating the empirical parameter  $\alpha$  is to assume that the solute-solute and solute-copper interactions are all zero and that there are no local distortions. Then  $\alpha$  simply measures the number of broken bonds and should equal  $-2$  for this model. The value  $-2$  is then a lower limit on  $\alpha$ ; considering the solute-solute and solute-copper ion interactions to be only weakened, rather than broken, would have the effect of raising  $\alpha$  above this lower limit.

In fact, the values of  $\alpha$  for the solutes Si and Ge surpass this lower limit. In itself, this is not too surprising, in view of the simple picture presented. In this picture, moreover, the low values of  $\alpha$  can be easily understood as being due to movement of the nearest neighbors of the solute ion [3].

The ideas presented here represent a somewhat different approach to understanding alloys. In contrast to the idea of atomic diameter, one thinks instead of ion core diameter. That this is actually different can be seen when one considers substituting a solute atom such as Zn into the copper lattice. In the usual atomic picture, the Zn atom which is supposed to be larger than Cu would produce local compressions, whereas the ion core is expected to be smaller than the Cu ion and would then produce a local tension. Thus the nearest neighbor movements would be opposite in the two cases.

## Summary

The elastic constants of several copper alloy single crystals have been measured by the pulsed ultrasonic method. The changes upon alloying have been interpreted with regard to two parameters,  $Z$  and  $\alpha$ . The electron atom ratio and  $Z$  have been shown to be nearly equal experimentally. The ionic parameter  $\alpha$  is found to be smaller (larger absolute magnitude) as the solutes progress to the right in the periodic table.

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