Effects of self-interstitials and close pairs on the elastic constants of copper*

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Measurements of changes in the elastic constants of copper and their temperature dependences resulting from stage-I_C close pairs and stage-I_D isolated interstitials are used to investigate defect configurations. The relative elastic-constant changes below 4 K per unit concentration γ of isolated interstitials, $d\ln C/d\gamma$, are found to be 0 ± 1, -30, and -15 for the bulk modulus, C_{44}, and $C'=(C_{11}-C_{12})/2$, respectively. These results show that the bulk effect arising from the change in number and strength of interatomic bonds is small, but that there exist polarization effects expected from ⟨100⟩ split interstitials. The temperature dependence of the elastic constants provides evidence of thermally excited resonance modes of frequency $\omega=5\times10^{12}$ Hz, which are also evidence for ⟨100⟩ split interstitials. An elastic relaxation due to thermally activated stress-induced ordering of I_C defects is observed at 18 K in C_{44}, but not in C', providing symmetry information which leads to a model for the I_C close-pair configuration.

I. INTRODUCTION

In a previous paper,[1] hereafter referred to as paper I, measurements were reported of the changes introduced in a complete set of elastic constants of copper, irradiated below 4 K with thermal neutrons and subsequently annealed. It was found that the elastic constants are affected in several different kinds of ways by point defects. In the present paper, these data are interpreted in terms of various mechanisms by which defects affect elastic constants. The results are used to obtain evidence for the ⟨100⟩ split configuration of isolated interstitials and thermally excited resonance modes, and a model for the configuration of the I_c close-pair defect.

Much is known about interstitials in copper as a result of intensive research over the past two decades.[2] A prominent annealing stage near 40 K (stage I) obtained after radiation damage at helium temperature[3] is found to have substages[4] (labeled stages I_A, I_B, I_C, I_D, and I_E in order of increasing annealing temperature). The locations of stages I_A–I_D are independent of damage density, while stage I_E occurs at lower temperatures for larger damage doses. With a few exceptions, the same stages are found in other metals. Stages I_A, I_B, and I_C are interpreted[5] as arising from the recombination of close-pair interstitial-vacancy complexes, with activation energies which are substantially reduced from the free-interstitial migration energy by the influence of the nearby vacancy strain-field. Stage I_D is interpreted as arising from correlated recombination of an interstitial with its own vacancy over larger separation distances,[5] where the influence of the vacancy strain field is weak,[6-9] and stage I_E arises from the recombination of interstitials, freely migrating[5] in three dimensions,[6] with distant vacancies. The interstitial migration-activation energy measured in stages I_D and I_E is ~0.1 eV.

However, very little is known about the geometry of the interstitial. From symmetry considerations, several configurations can be specified,[10] and models for radiation damage have centered attention on three possibilities. The first has cubic symmetry and is located at the body-centered-cubic position. The second is a split interstitial with center of mass at a fcc lattice site but split along a cube direction. The third is the Crowdion configuration with axis along a ⟨110⟩ direction. Calculations for the free interstitial favor the ⟨100⟩-split-interstitial geometry.[11] Recently, Erhardt and Schilling[12] have excluded the Crowdion configuration for Al from measurements of diffuse x-ray scattering. No models have yet been firmly established for the close-pair configurations.

Elastic-constant changes and their temperature dependences are known to be especially well suited for determining the symmetry of defects. This is in contrast with most property-change measurements which merely count the number of defects. Several features of the measurements obtained in paper I allow one to separate the contributions from different kinds of effects, and permit one to extract information about particular defects. (a) Dislocation-pinning contributions were eliminated by a room-temperature fast-neutron irradiation of low-dislocation-density samples. (b) The measurements were made on a complete set (C_{11}, C_{44}, and C') of independent cubic-elastic constants.

This allows calculation of the changes to be expected in any cubic-elastic constant (bulk modulus, Young's modulus, etc.). Knowledge of these changes is useful in the comparison of different experiments, as well as in the separation of various effects. In addition, measurements of the behavior of a complete set of constants is important in determining the symmetry of the contributing defects. (c) The high degree of sensitivity which was achieved by use of the pulse-echo superposition technique permits a quantitative study of the annealing behavior of the irradiation-produced changes. This in turn allows investigation of the contributions from particular stage-I defects. (d) The simultaneous measurement of the resistivity and attenuation gives an accurate measurement of the concentration of irradiation-induced defects. Finally, (e) the measurement of the change in the temperature dependence of the elastic constants produced by the irradiation, and its annealing, allows further study of defect symmetries, and reveals the existence of an unexpected effect.

Point defects may change the elastic properties of a metal in several ways: (i) dislocation-pinning effects; (ii) a bulk effect due to the alteration in number and strength of interatomic bonds; (iii) polarization effects which occur if an anisotropic defect is strongly polarizable by an applied stress; and (iv) relaxation effects which involve the thermally activated reorientation of a defect under an external stress; (v) the thermal excitation of defect resonance modes is used here to explain certain of the temperature-dependent effects.

An early theoretical calculation by Dienes[13] indicated that the bulk effect would increase all the ordinary elastic constants of copper by about 10% per at.% interstitials, but decrease them by approximately 1% per at.% of vacancies. However, other estimates vary over two to three orders of magnitude and even differ as to the expected sign of the effect. For example, Zener[14] offers a semiquantitative treatment which predicts a decrease in the shear elastic constants of about 10% per at.% of either vacancies or interstitials, while Nabarro's[15] linear-elasticity result estimates 3.8 and −2.3% per at.% of interstitials and vacancies, respectively. On the basis of isotropic elasticity theory Townsend *et al.*[16] conclude that the bulk effect should be small [(~ 1-10)% per at.%], being positive for interstitials and negative for vacancies. On the other hand Melngalis[17] has given an anisotropic elasticity estimate of ~ −100% per at.%. A thermodynamic calculation by Holder and Granato[18] gives values of the order of a few percent or less.

These calculations are all concerned with the bulk effect. The importance of large polarization effects, which lead to corresponding large decreases in the elastic constants[19] has only recently been recognized. By using a computer simulation of the copper lattice, Dederichs *et al.*[20] have shown that the ⟨100⟩ split interstitial is strongly polarizable by an applied shear stress due to the large negative bending-spring constants which result from the highly compressed nature of the lattice in the vicinity of an interstitial defect. The large negative contributions from the polarizability effect will differ in magnitude among the various elastic constants according to the symmetry of the defect. These bending springs also introduce low-frequency resonant modes into the vibrational spectrum of the lattice.

A dynamic contribution to the elastic moduli will occur if a thermally activated relaxation between preferential defect orientations is induced by an applied stress. Characteristic of such an anelastic effect is a peak in the internal friction measurements and a concomitant decrease in the elastic constant. Any defect that introduces a strain field possessing lower symmetry than that of the lattice can give rise to such an anelastic effect. As the relative alignment of the applied stress and defect orientation is altered, the anelastic effects contribute in varying amount, thus revealing the symmetry of the interacting defect. These phenomena have been used extensively to investigate impurity effects in various materials[21] and two studies of the elastic moduli of copper at low temperatures have reported the existence of relaxation phenomena associated with a vacancy-interstitial complex.[22,23]

In summary, since many of these effects are sensitive to the symmetry of the defect, valuable clues to the geometrical configurations assumed by various defects in the damaged lattice may be obtained from elastic-constant measurements, particularly if individual contributions from particular kinds of defects can be separated out of the total irradiation-induced changes. This paper will focus on a discussion of the mechanisms by which point defects may alter the elastic constants of the host lattice and the symmetry information obtained from their measurement will be used to reach a number of conclusions about the geometrical nature of stage-I defects in copper.

In Sec. II different contributions from various stage-I defects are isolated in the experimental results from paper I. Results from other experiments are also used with the paper I results in further describing the nature of the defects interactions. In Sec. III the analysis of Sec. II is used to provide evidence for ⟨100⟩ split free interstitials with low-frequency resonance modes. A model for the I_C close-pair configuration is pro-

posed, and the expected effects of split interstitials on other physical properties are discussed.

II. ANALYSIS

A. Results at 3.6 K

The annealing results presented in paper I show that the irradiation-induced changes in all three elastic constants recover in a different fashion during isochronal annealing, and none of the constants recover linearly with the resistivity. This indicates that the magnitude of the elastic-constant change is different for different types of defects, and that it is therefore important to associate the elastic modulus recovery stages with the appropriate defect recovery stages characteristic of resistivity annealing.

The stage-I substages are not clearly isolated in the paper I resistivity measurements because the damage does not consist completely of simple isolated defects, and the resistivity measurements were not made with the highest definition possible. Nonetheless, the temperatures at which the substages are located can be determined accurately, since these temperatures depend only on the annealing rate, and numerous other data are available for this purpose.

Towards this end, the results of a number of detailed studies of resistivity annealing in this range are standardized to the conditions used for the measurements in paper I. For stages I_A–I_D the results obtained from three different studies[5,24,25] agree within $\pm\frac{1}{2}$ K when corrected[6] to the annealing rate ($\sim\frac{1}{3}$ K/min) used in paper I. The value of the temperature where the maximum rate of annealing of each stage occurs is $I_A = 16$ K, $I_B = 27$ K, $I_C = 32$ K, and $I_D = 41$ K. The dose dependence of I_E makes it more difficult to locate accurately. From studies of slow-neutron[26] and low-energy-electron damage[25] in which defect concentrations comparable to those introduced by the irradiations used in paper I were used, we estimate that I_E annealing should occur between 45 and 55 K in the measurements of paper I.

For further simplification of the discussion, the annealing results from paper I are shown in Fig. 1 as the percentage of the total irradiation-produced frequency change which remains at 3.6 K after various stages of the annealing program. The temperatures corresponding to the centers of the various stages determined in the manner discussed above, as well as a curve representing the average of the resistivity data, are also included in this figure.

Most of the theoretical work in this field has been concerned with the contribution to the elastic constants from isolated vacancies and interstitials. Since the total irradiation-produced changes include the effects of close pairs as well as more complex types of defects, they are not convenient for comparison with theory. Stage I_D is generally believed to arise from the recombination of interstitials with their own vacancies (the two defects being sufficiently separated that their mutual interaction can be considered negligible), so the ratio

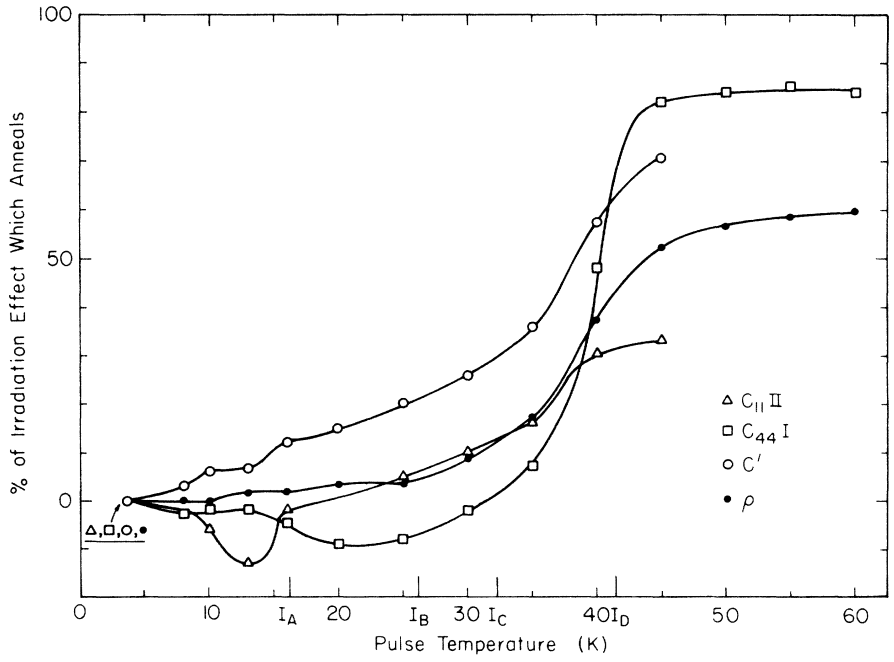

FIG. 1. Percentage of the total irradiation-produced frequency change measured at 3.6 K which remains at various stages of annealing for three independent modes measured in paper I. A curve representing an average of the resisitivity data is also shown. The temperatures corresponding to the maximum rate of annealing of the substages, which were determined in the manner discussed in the text, have been marked on the abscissa.

of the amount of elastic-constant recovery to that of the resistivity in stage I_D provides a direct means for determining the magnitude of the effects for isolated defects. The results for $d\ln C/d\gamma$ obtained by using the I_D annealing in the present experiments (taken to occur between 35 K and 45 K), are given in Table I.

The recovery of the bulk modulus, calculated from the measured changes in C_{11} and C', shows a behavior completely different from that of the other constants. The change in the bulk modulus during irradiation, $d\ln B/d\gamma$, is only -2, and this effect exhibits no significant recovery during the annealing through 45 K. In particular, no measurable change in the irradiation-induced bulk-modulus effect occurs during stage I_D. In contrast, the shear-modulus changes in stage I_D, given in Table I, are large and anisotropic, with the C_{44} change about twice as large as that for C'.

It would likewise seem possible to estimate the free-interstitial contribution to the elastic constants by using the changes observed during the annealing of stage I_E. However, the relatively small amount of I_E recovery which occurs, coupled with the aggregation and trapping of defects during the long-range migration of the interstitial, makes a quantitative measurement and interpretation more difficult. An accurate determination of the relationship between the recovery of the elastic constants and resistivity changes in stage I_E must await more detailed experimental results, preferably from studies of small-defect concentrations introduced by low-energy-electron irradiation.

B. Temperature-dependent effects

The measurements in paper I of the change in the temperature dependence of the C_{44} modulus induced by the thermal-neutron bombardment showed quite clearly that a strong dispersion was present at about 18 K, characteristic of a relaxation effect. However, it is important to realize that these results contain contributions from at least two other sources besides the observed relaxation: (a) some annealing of defects occurs at each temperature and (b) the defects which remain after the 45-K annealing pulse also give a temperature-dependent effect. These effects must be eliminated before extracting any quantitative information from the results.

TABLE I. The contributions from I_D defects to the measured set of elastic constants.

Run	$d\ln C/d\gamma$
C_{11}II	−2
C'	−15
C_{44}I	−29
C_{44}II	−33

The effects due to the first of these sources can be removed approximately by correcting each point of the original results for the annealing which was measured at 3.6 K. This annealing correction has practically no effect on the sharp dispersion near 18 K. The temperature dependence from the defects which remain after the 45-K annealing pulse is given by the upper curve of Fig. 6 in paper I. Consequently, we can remove the second contribution by considering only the difference between this curve and the total defect-induced change in temperature dependence. This difference, including the annealing correction, is presented in Fig. 2.

From Fig. 2, we conclude that a defect is created during the irradiation which introduces a sharp relaxation-modulus effect centered about 18 K and which anneals somewhere in the temperature interval from 25 to 40 K. Although a curve obtained by treating the data of C' in a similar fashion also gives evidence of temperature-dependent effects, the dependence is considerably weaker and no indication of a drop near 18 K is present. It is not possible to obtain a similar curve from the C_{11} results because of the different annealing program which was followed.

The results are consistent with kHz measurements by Nielsen and Townsend[22] (NT), who found a similar change in Young's modulus along the ⟨111⟩ crystal direction but no change for the ⟨100⟩ crystal direction. These effects would be expected for a defect having a ⟨111⟩ symmetry axis, as concluded by NT.

By combining the present 10-MHz results with

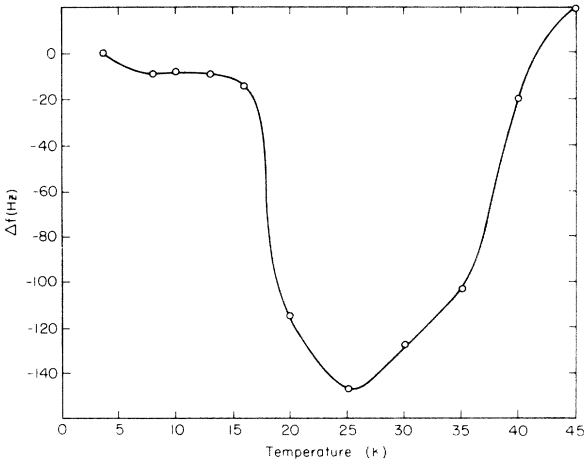

FIG. 2. Change in the temperature dependence of C_{44} produced by defects which anneal below 45 K.

the kHz measurements of NT, relatively more accurate values for the jump frequency and activation energy can be obtained. These results are $6 \pm 2 \times 10^{11}$ Hz and 0.017 ± 0.002 eV, respectively, in good agreement with those reported solely on the basis of the NT experiments ($8 \times 10^{11 \pm 1}$ Hz for the attempt frequency and 0.015 ± 0.002 eV for the energy of activation).

The annealing of the anelastic effect somewhere between 25 and 40 K found in paper I is also in agreement with NT. Although this annealing range is broad enough to include both the I_B and I_C stages, we note that in the present results the center of this annealing range lies within 1 K of the I_C resistivity peak and more than 5 K above that of I_B. Since NT were able to observe this annealing in detail, it is possible to determine more precisely which of the annealing resistivity substages should be associated with the recovery of the anelastic process in their measurements. In order to do this, we correct the resistivity data of various authors[5,24,25] for the differences in annealing rate.[6] The resulting temperatures ($\pm \frac{1}{2}$ K) at which stages I_B and I_C should occur in the NT measurements are 27.4 and 32.1 K, respectively. We note that the calculated annealing peak temperature for the I_C defect coincides almost exactly with the value of 32 K reported by NT for the maximum rate of recovery of the anelastic process. Furthermore, the annealing of the anelastic process does not begin until after the onset of I_B recovery and most of the annealing occurs at temperatures above the end of the I_B range. For these reasons, we conclude that I_C is the only recovery stage with which this relaxation may be identified.

The anelastic behavior of point defects is often discussed in terms of an elastic dipole,[27] since in many ways the effect is similar to that experienced by an electric dipole under the influence of an applied field. The second-rank tensor λ_{ij} used to describe the elastic dipole is the strain component ϵ_{ij} per unit mole fraction C of defects:

$$\lambda_{ij} \equiv \frac{\partial \epsilon_{ij}}{\partial C} . \tag{1}$$

For axially symmetric defects there are only two independent elements of this tensor, λ_1 and λ_2, and only the absolute value of the difference between these two, the so-called "shape factor" $|\lambda_1 - \lambda_2|$, can be obtained from measurements of the elastic moduli. However, the relationship of the shape factor to the observed change in C_{44} is a function of the defect orientation. For the $\langle 111 \rangle$ symmetry axis assumed by NT, the relation is given by[27]

$$-\Delta C_{44}/3C_{44}^2 = \tfrac{4}{27}(C_0 V_0/kT)(\lambda_1 - \lambda_2)^2 , \tag{2}$$

where C_0 represents the molar concentration of defects and V_0 is the molecular volume. In estimating C_0 associated with the 18-K relaxation we use the results of a review of the energy dependence of the total recovery of close pairs by W. Schilling,[28] indicating that about 8% of the total damage in the present experiment is comprised of I_C defects. Using this result and the change of 135 cycles in the resonant frequency indicated in Fig. 2, Eq. (2) gives a value of 0.5 for $|\lambda_1 - \lambda_2|$. This is in excellent agreement with the result which is obtained from the NT measurements, if their estimate for the concentration of defects is accepted. That estimate was based on the conclusion of Nilan and Granato[24] that the Seitz-Koehler[29] theory of damage production overestimates the number of displaced atoms by a factor of 5. Using this same estimate one obtains excellent agreement again between Townsend *et al.*[16] and Part I for the shear-elastic-constant change found during low-temperature irradiation.

In addition to the temperature dependence introduced in C_{44} by the I_C relaxation process, it was determined in paper I that a strong temperature-dependent effect was introduced in C' by the defects which anneal between 35 and 45 K (I_D) and in C_{44} by the defects which anneal between 45 and 60 K (I_E). This dependence shows a linear decrease of the elastic constant for temperatures above approximately 20 K; below this temperature, the effect gradually decreases, achieving zero slope below approximately 3 K.

III. MECHANISMS FOR ELASTIC-CONSTANT CHANGES AND DEFECT MODELS

A. Bulk and polarization effects

The changes in the elastic constants observed at 3.6 K are determined primarily by the bulk and polarization effects. These should be relatively independent of temperature, and are known as dielastic effects. The bulk effect can be estimated by determining the response to an applied stress, treating all atoms as if they were at centers of symmetry.

Polarizability effects can only occur when the defect symmetry differs from that of the host lattice. In this case, the atoms associated with the defect are no longer located at perfect-crystal-lattice sites. When a homogeneous external stress is applied, the defect atoms are displaced by an additional amount relative to the homogeneously deformed lattice.[19] This additional internal strain will cause a decrease in the measured elastic constant; the magnitude of the strain will be determined by the effective force constants in the im-

mediate vicinity of the defect. The decreases in the elastic constants should be anisotropic, where the nature of the anisotropy is related to the symmetry of the defect.

Since a hydrostatic stress does not alter the configuration of the defects in a cubic lattice, the bulk-modulus change should be free of relaxation and polarization effects, and thus provides the most direct measurement of the bulk effect. No evidence of any recovery of the small change in the bulk modulus created during irradiation is found in the annealing results through 45 K. This indicates that the bulk effect during irradiation is primarily due to defects which do not anneal in stage I, and that the magnitude of the bulk effect, $d\ln B/d\gamma$, from isolated vacancies and interstitials, is less than 1. In fact, this effect is so small in the paper I measurements that not even its sign can be determined. This indication of a small magnitude for the bulk effect is in agreement with the range of most of the reported theoretical estimates (~ 1–10% per at.%), and agrees best with the thermodynamic estimate[18] (~ 0.1% per at.%). The small effect could be a result of small contributions from both vacancies and interstitials, or else the consequence of nearly equal and opposite magnitudes.

If the bulk effect is indeed so small, the large decreases shown in Table I must be a result of the other types of contributions, such as polarization effects. In fact, this pattern of elastic-constant changes is just that expected on the basis of a recent calculation by Dederichs[30] for the polarizability of the ⟨100⟩ split dumbbell interstitial. Dederich's calculation indicates that certain of the force constants for the ⟨100⟩ split dumbbell interstitial allow large internal displacements, with consequently large decreases in certain of the elastic constants. In particular, he estimates the magnitude of the effect to be largest in C_{44}, less in C', and almost negligible in C_{11}. We see that this pattern is clearly obeyed by the present results. This behavior of the various elastic-constant changes produced by I_D defects is strong evidence for the split ⟨100⟩ interstitial configuration in copper. The likelihood of this is even stronger in view of quantitative results obtained below in discussing temperature-dependent effects.

B. Relaxation effects

Many attempts have been made to assign a particular lattice configuration to each of the three close-pair defects solely on the basis of their relative activation energies, but this approach leads to a wide variety of possible choices. Magnetic after-effect measurements provide more direct information for Ni, and will be discussed later. The I_C elastic relaxation in copper places further restrictions on these choices, and makes a new attempt more promising.

Relaxation can occur when an applied stress causes certain of the defect orientations which were equivalent in the unstressed crystal to become energetically favored in the stressed crystal. For isolated defects, the symmetry axis would be expected to lie along one of the symmetry axes of the crystal, and a C_{44} but no C' relaxation in a cubic crystal indicates trigonal symmetry.[22,27]

However, for a close-pair defect the symmetry axis need not lie along one of the symmetry directions, and the defect orientations whose relative energies are unaffected by a direct measurement of the C' elastic constant are all those in which the two orientations are symmetric about a (100) plane. Therefore, the type of motion which is compatible with the relaxation observed in the present case must possess mirror symmetry about a (100) plane. Since the two orientations should be equivalent in the unstressed crystal, the vacancy member of the close pair must also lie in the same (100) plane.

The criteria then which must be satisfied for the I_C defect are: (i) it must have a low activation energy for annealing (0.095 eV); (ii) an even lower activation energy for relaxation (0.017 eV); (iii) the relaxation must involve motion symmetric about a (100) plane; (iv) the vacancy member of the close pair must lie in the same (100) plane; (v) the relaxation must be induced by a measurement of the C_{44} elastic constant; and (vi) it must be characterized by a relatively large shape factor (0.5).

If we begin by considering the two interstitial types which appear to be the most energetically stable,[31] the ⟨100⟩-split and body-centered configurations, then only three arrangements appear to have both a low activation energy for annealing and the required mirror symmetry: (a) the body-centered interstitial with a vacancy at one of the unit-cell corners; (b) the ⟨100⟩-split dumbbell with a vacancy at its third-neighbor lattice site ($\frac{1}{2}\frac{1}{2}1$); and (c) the ⟨100⟩-split dumbbell with a vacancy in the diagonally opposite (⟨111⟩ direction) corner. Configurations (a) and (c) both have trigonal symmetry.

However, the identification of any of these schemes with the I_C defect is unlikely. In configuration (a), the arrangement of the pair is such that the interstitial and vacancy are next-nearest neighbors; thus only one closer lattice position, nearest neighbor, is available for both the stage I_A and I_B defects. Since the nearest-neighbor site is very likely unstable, the association of config-

uration (b) with I_C also seems unlikely for the same reason. Although arrangement (c) appears to be satisfactory from a proximity standpoint (there are five closer lattice positions), the requirement that the dumbbell must move two entire lattice spacings without annealing, and with an activation energy less than 0.02 eV, in order to relax into an equivalent lattice configuration, makes this arrangement highly improbable. This objection also applies, to a lesser extent, to configuration (b), which must move about one lattice spacing to reach an equivalent site.

In fact, it appears that any model which requires motion of the center of mass of the interstitial over a distance greater than the nearest-neighbor distance cannot satisfy criterion (ii). Consequently, we are led to the conclusion that it is not the entire vacancy-interstitial arrangement which relaxes, but rather only a reorientation (little or no motion of the center of mass) of the interstitial member that occurs. In this case, the symmetry information which is available from the observed relaxation is characteristic of the interstitial motion, and not of the entire close pair.

We first note that the flipping of a ⟨100⟩-split interstitial between different ⟨100⟩ orientations (without change of the center of mass) does not involve motion which is symmetric about a (100) plane, and therefore is not compatible with criterion (iii). Therefore the above requirement that the relaxation results from the reorientation of the interstitial leads to the conclusion that the interstitial member of the I_C close pair cannot have the ⟨100⟩-split configuration of the isolated defect.

However, the large relaxation strength calculated above ($|\lambda_1 - \lambda_2| = 0.5$) indicates that the interstitial still is characterized by some form of split configuration. In fact, the observed relaxation strength is even greater in magnitude than that estimated for the isolated ⟨100⟩-split configuration ($|\lambda_1 - \lambda_2| = 0.025$,[32] 0.21[33]). This large value may be understandable in terms of the nearby vacancy, which probably allows even further elongation of a split configuration interstitial. Therefore, we are looking for a simple reorientation of a split-configuration interstitial that involves motion which is symmetric with respect to the (100) plane containing the vacancy.

It was noted above that the value of the shape factor required to produce the measured modulus change depends on the orientation of the defect. The estimate given above assumed a ⟨111⟩-defect orientation. For the case of relaxation between equivalent sites symmetric about a (100) plane, the required value of the shape factor increases for interstitials whose axes are closer to either the (100) symmetry plane or a ⟨100⟩ direction than the ⟨111⟩ direction assumed above. Since the estimated value of 0.5 is already considerably larger than theoretical estimates for split interstitials, it seems unlikely that the axis of the I_C interstitial makes an angle with the (100) plane which is very much larger or smaller than does the ⟨111⟩ axis.

The remaining question concerning the I_C defect geometry is the location of the interstitial relative to its vacancy member. We use the results of a computer calculation of stability regions by Gibson *et al.*[11] as a guide in selecting this location. Their results for a (100) plane containing the vacancy are shown in Fig. 3. All stable positions have been marked with a large cross. Although these calculations are based on the geometry of a ⟨100⟩-split interstitial, we assume that the somewhat different configuration to be considered below can be approximated by these calculations.

Since the defect must possess a low activation energy for annealing (criterion 1), we eliminate all sites further removed from the vacancy than the site labeled 2 (site 3 and beyond). Site 2 lies along a ⟨100⟩ direction from the vacancy. This arrangement makes it unlikely that the interaction between the two members of the close pair would create more than one equivalent orientation for a split interstitial configuration which would be symmetric with respect to a (100) plane. On the contrary, it appears that a split interstitial located here would simply point directly towards the vacancy.

These objections do not apply to site 1, where a

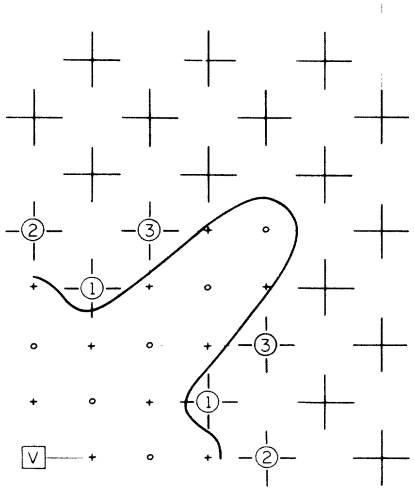

FIG. 3. Stability of a vacancy and a nearby interstitial in a (100) plane of copper. The vacancy is at the lower left-hand corner. All stable sites have been marked with a large cross [after Gibson *et al.* (Ref. 11)].

split-interstitial configuration which satisfies all the criteria may be located. The proposed model for the I_C defect is therefore shown in Fig. 4, with the center of mass of the split-configuration interstitial located at site 1 of Fig. 3; four of the 24 equivalent positions of the center of mass of the interstitial have been marked with a cross. The equivalent orientation into which the interstitial is believed to relax has been indicated by a dotted outline.

The configuration proposed here places the center of mass of the interstitial defect in the same lattice position with respect to the vacancy as does a model suggested previously by Peretto *et al.*[7] Their model was used to explain the observation of a magnetic after-effect for the I_C defect in nickel, and involved the motion of the center of mass of the interstitial between the four equivalent sites marked in Fig. 4. Their model assumes a ⟨100⟩-split interstitial configuration, so it is inconsistent with the symmetry requirements imposed by the elastic-constant measurements. However, the magnetic after-effect involves motion of the center of mass, and the model presented here would allow such motion. Thus the present model is consistent not only with all the criteria deduced from elastic-constant measurements, but is also consistent with the requirements necessary to explain magnetic after-effect measurements in nickel.

In addition to the I_C magnetic after effect, Peretto *et al.*[7] also found magnetic after effects associated with I_B and I_E defects. Their model for I_C as already discussed produced the magnetic after effect by motion between nearest-neighbor sites of an interstitial located at the vacancy's four equivalent fifth-neighbor lattice sites, enclosing a ⟨100⟩ axis. Their model for I_B similarly

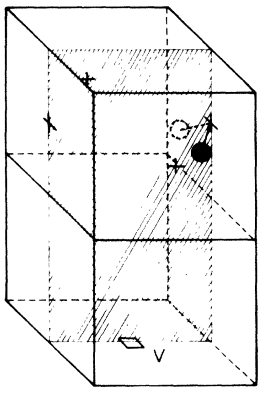

FIG. 4. Proposed model for the I_C defect. Four of the 24 equivalent sites for the center of mass of the interstitial have been marked with a small cross. The equivalent position into which the split-interstitial relaxes is shown by a dotted outline.

involved the migration of an interstitial in the vacancy's third neighboring site between the three equivalent sites enclosing a ⟨111⟩ direction. Krönmuller[34] has pointed out that a similar possibility exists for an interstitial located at the vacancy's seventh nearest neighbor, and suggested that the interstitials for stages I_B and I_C should be located at the third- and fifth-, or fifth- and seventh-nearest-neighbor sites, respectively. As we have used the fifth-neighbor site for stage I_C, this leaves the third-neighbor site for I_B. By using the relaxation effects observed in the elastic constants we have been able to show that the interstitial in stage I_C is not the ⟨100⟩-split configuration of the Peretto *et al.* model. However, no relaxation effects have been observed for I_B defects so the ⟨100⟩ orientation of the interstitial cannot be ruled out for the Peretto *et al.* I_B model.

The mechanisms responsible for magnetic after effects should also produce relaxation effects in elastic constants,[35] although very low frequencies (~1 Hz) would be required to provide measurable relaxation effects before the defects annealed. Such measurements in copper could provide crucial information concerning remaining uncertainties in the geometry of stage I_A and I_B defects.

C. Thermally excited resonance-mode effects

The temperature dependence of the contribution from the I_D defects to C', (Table III in paper I), and from the I_E defects to C_{44}, is not characteristic of a relaxation process. Also, it would be surprising if the polarization and bulk contributions from the individual defects carried a temperature dependence which is so much larger than that of the ideal-crystal elastic constants. On the other hand, the large observed temperature dependence could arise from only a small change in the vibrational contribution to the elastic constant from the entire lattice.

The simplest interpretation of a change in the vibrational spectrum is that the Debye characteristic temperature Θ_D has been shifted as a result of the irradiation. In the Debye approximation, the elastic constants vary as $T^4/\Theta_D{}^5$ in the low-temperature region. The required shift of the Debye temperature necessary to produce the observed change in the temperature dependence can therefore be found from the relation:

$$\frac{\Delta\Theta_D}{\Theta_D} = -\frac{1}{5}\frac{f_d(T_1) - f_d(T_2)}{f_p(T_1) - f_p(T_2)}. \tag{3}$$

Here, $f_d(T)$ is the irradiation produced change in the resonant frequency at temperature T, and $f_p(T)$ is the measured preirradiation value of the resonant frequency at temperature T. The present

results obtained in the interval from 3.6 to 25 K would then require a fractional change in the Debye temperature of about 4×10^{-3}. However, such a large change in Θ_D is not consistent with the observed elastic constant changes. In the Debye approximation, the velocity of sound, v, is related to the Debye temperature by

$$\Theta_D = (\hbar v/K)(6\pi^2 N/V). \tag{4}$$

Here N is the number of vibrational modes present in a volume V of the lattice. Therefore, the change in the Debye characteristic temperature produced by the irradiation is given directly by the change in the velocity of sound and of the volume:

$$\Delta\Theta_D/\Theta_D = \Delta v/v - \Delta V/V. \tag{5}$$

If the observed temperature dependence can be attributed to a shift of the Debye temperature, the relative change of the velocity of the sound wave produced by the I_D defects during irradiation would therefore also be on the order of 4×10^{-3}. The total velocity change which was measured during irradiation of this mode is less than 3×10^{-5}, clearly demonstrating that the radiation-induced temperature dependence is far too strong to be explained by a rigid shift of the lattice spectrum.

However, a strong change in the temperature dependence in the low-temperature region could be produced by a relatively small change in the vibrational spectrum due to the introduction of new low-frequency modes. The computer simulation by Dederichs *et al.*[20] predicts, in fact, the existence of such modes for the ⟨100⟩-split dumbbell interstitial, with characteristic frequencies of about $\frac{1}{7}$ the maximum cutoff value for the ideal lattice. Although Dederichs[30] was concerned with the excitation of these modes by an applied stress, resonant modes can be excited thermally as well leading to the large temperature-dependent effects.

The result of thermal excitation of resonant modes may be quantitatively analyzed by relating the temperature dependence of the elastic constants to the vibrational spectrum of the crystal containing defects. For this calculation we use the thermodynamic definition of the nth-order isothermal elastic constant, $C^T_{ijkl}\cdots$, in terms of the strain derivatives $\partial/\partial\eta_{ij}$ of the free energy F and volume V[36]:

$$C_{ijkl}\cdots \equiv \frac{1}{V}\frac{\partial^n F}{\partial\eta_{ij}\partial\eta_{kl}\cdots}\bigg|_T. \tag{6}$$

The free energy is the sum of the cohesive energy Φ, the total vibrational energy from all the vibrational modes α, and an entropy term $-TS$:

$$F = \Phi + \sum_\alpha \epsilon_\alpha - TS, \tag{7}$$

where

$$\epsilon_\alpha = \hbar\omega_\alpha/(e^{\hbar\omega_\alpha/kT}-1). \tag{8}$$

Defining the state of zero stress as that in the absence of all vibrations, the expression for the isothermal second-order elastic constants to first order in the vibrational part of the strain, η, is then given by[37]

$$C^T_{ijkl} = \tilde{C}^0_{ijkl} + \frac{1}{V^0}\sum_\alpha\left[\frac{1}{\omega_\alpha}\frac{\partial^2\omega_\alpha}{\partial\eta_{ij}\partial\eta_{kl}}\epsilon^0_\alpha - \frac{1}{\omega_\alpha^2}\left(\frac{\partial\omega_\alpha}{\partial\eta_{ij}}\right)_T\left(\frac{\partial\omega_\alpha}{\partial\eta_{kl}}\right)_T C^0_\alpha T\right] + \eta(\tilde{C}^0_{ijkl} + \tilde{C}^0_{ijklmm}). \tag{9}$$

The angular frequencies ω_α are the frequencies of oscillation about the static lattice; C^0_α represents the Einstein heat capacity for mode α and the $\tilde{C}^0_{ijkl}\cdots$ are the static lattice elastic constants. The change in the temperature dependence of the elastic constant resulting from the introduction of resonance modes by the N defects can then be obtained by considering the effects produced by the addition of a number N of Einstein oscillators, each of characteristic frequency ω_E. From Eq. (9) this change in the elastic constant is given by

$$\Delta C_{ijkl}(T) = \frac{\hbar N}{V_0}\frac{\partial^2\omega_E}{\partial\eta_{ij}\partial\eta_{kl}}\left(\frac{1}{2}+\frac{1}{e^y-1}\right) - \frac{\hbar N}{V_0\omega_E^2}\left(\frac{\partial\omega_E}{\partial\eta_{ij}}\right)_T\left(\frac{\partial\omega_E}{\partial\eta_{kl}}\right)_T\frac{ye^y}{(e^y-1)^2} + \Delta\eta(\tilde{C}^0_{ijkl}+\tilde{C}^0_{ijklmm}). \tag{10}$$

Here, $y=\hbar\omega_E/kT$ and $\Delta\eta$ is the change in the vibrational part of the strain. From the equilibrium condition, it is found to be[37]

$$\Delta\eta = -\frac{\hbar N}{3B}\left(\frac{1}{2}+\frac{1}{e^y-1}\right)\left(\frac{\partial\omega_E}{\partial V}\right)_T. \tag{11}$$

If we define K_1 and K_2 in the following manner,

$$K_1 = \frac{\hbar N}{V_0}\frac{\partial^2\omega_E}{\partial\eta_{ij}\partial\eta_{kl}} - \hbar N\frac{(C^0_{ijkl}+C^0_{ijklmm})}{3B}\frac{\partial\omega_E}{\partial V}\bigg|_T \tag{12}$$

and

$$K_2 = \frac{\hbar N}{V_0\omega_E}\left(\frac{\partial\omega_E}{\partial\eta_{ij}}\right)_T\left(\frac{\partial\omega_E}{\partial\eta_{kl}}\right)_T, \tag{13}$$

then we may write the contribution to the elastic constants from the addition of N resonance modes of characteristic frequency ω_E as

$$\Delta C_{ijkl}(T) = K_1\left(\frac{1}{2} + \frac{1}{e^y - 1}\right) - K_2 \frac{y}{(e^y - 1)^2} e^y. \quad (14)$$

Equation (14) shows that the introduction of resonant modes into the vibrational spectrum of the lattice produces both a zero-point and temperature-dependent effect. It predicts a linear decrease of the elastic constant with temperature for $\hbar\omega_E$ much less than kT, and an exponential temperature dependence for $\hbar\omega_E$ much greater than kT, achieving zero slope at 0K. In fact, the qualitative temperature dependence of the elastic constants given by this equation is precisely the behavior exhibited by the I_D defect contribution to the temperature dependence of the C' elastic constant (Table III of paper I).

The measured contributions from the I_D defects to the temperature dependence of the C' elastic constant obtained in paper I are shown in Fig. 5, along with a curve obtained from a least-squares fit of the temperature-dependent terms in Eq. (14) to this data, using $\omega_E = 5\times10^{12}$ Hz. Error bars are given on the figure which represent our own estimate of the experimental uncertainty. Although the curve obtained from the least-squares analysis fits the data points extremely well, it is difficult to assign error limits to the resulting values for the three adjustable parameters. It is possible to

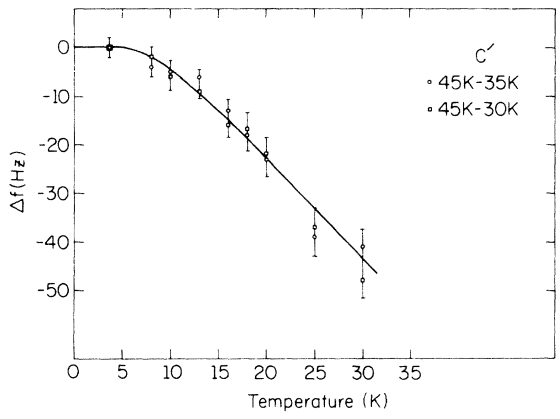

FIG. 5. Change in the temperature dependence of the C' resonant frequency produced by the defects which anneal in the 35–45-K temperature range (stage I_D). Since no significant change was observed between 30 and 35 K, both the difference between the 45- and 35-K results and between the 45- and 30-K results are shown. The solid curve represents a least-squares fit for $\omega_E = 5\times10^{12}$ Hz of the temperature-dependent terms in Eq. (14) to the results. The error bars represent our estimate of the experimental uncertainty of the results from paper I.

obtain satisfactory fits to the data with frequencies in the range $(1-9)\times10^{12}$ Hz. However, we may restrict this range further by considering the magnitudes of the frequency-strain derivatives appearing in K_1 and K_2.

In the range $\omega_E = (4-7)\times10^{12}$ Hz, the magnitude of $(1/\omega_E)(\partial\omega_E/\partial\eta_{ij})$ required for a satisfactory fit varies between approximately 40 and 100. The magnitude of $(1/\omega_E)(\partial^2\omega_E/\partial\eta_{ij}\,\partial\eta_{kl})$ is a very sensitive function of frequency in this region, ranging from about -10^4–$+10^4$ and becoming zero at a frequency slightly less than 6×10^{12} Hz. There are no theoretical values with which to compare these numbers, but $(1/\omega)(\partial\omega/\partial\eta)$ is approximately 2 for typical lattice modes when η represents a volume strain and $(1/\omega)(\partial^2\omega/\partial\eta^2)$ is of the order 1–10. However, if the values for the resonant modes were this small, then much smaller elastic-constant changes at 3.6 K would be expected, contrary to the experimental results. For shear strains, $(1/\omega)(\partial\omega/\partial\eta)$ is zero for lattice phonons by symmetry arguments with atoms at centers of symmetry. However, the split interstitial does not have this symmetry, and a first-order shear effect is to be expected for C'. Therefore the large values obtained from the present results imply that the resonant modes couple very strongly to shear strains. If we assume the first- and second-order strain derivatives progressively increase, as with typical lattice modes and higher-order elastic constants, the values for these parameters (~60 and ~10^3, respectively) required to fit the results for $\omega = 5\times10^{12}$ Hz are not unreasonable. However, very large values of the second-strain derivative, as much as three orders of magnitude greater than the first derivative for frequencies more than 30% different from 5×10^{12}, are not to be expected. Taking these considerations into account the range of frequencies which can be fit to the data is narrowed to

$$\omega_E = 5\times10^{12} \pm 30\%\ \text{Hz}. \quad (15)$$

This frequency compares very well with the prediction by Dederichs *et al.*[20] of approximately 6×10^{12} Hz for the ⟨100⟩-split dumbbell interstitial. The agreement provides even further evidence for the dumbbell configuration of the free interstitial.

The magnitude of the zero-point effect calculated from Eq. (14) for this frequency range is only a small fraction of the recovery of the C' elastic constant observed at 3.6 K between annealing pulses of 35 and 45 K. Hence no zero-point corrections to the results in Table I should be necessary before comparison with calculations of polarization effects.

The limited data obtained with the C_{44} mode permit only the temperature dependence which an-

neals in the interval between 45 and 60 K (stage I_E) to be separated in the manner discussed above. These results are shown in Fig. 6. Here again, we observe the kind of behavior which is expected if the defects introduce low-frequency resonance modes into the vibrational spectrum of this lattice. The fit to Eq. (14) for $\omega_E = 5\times10^{12}$ Hz obtained with the C' results has been included in this figure for comparison.

Although these results are also in agreement with the predicted behavior from the introduction of resonance modes, their interpretation is more difficult than in the C' case. As discussed previously, the small amount of annealing which occurs in this temperature range is further complicated by the clustering and trapping of defects during the long-range interstitial migration. Hence the change in the temperature dependence which occurs in this interval may be significantly affected by the new types of defects which are created. Therefore, the important question of whether or not the frequency found from the C_{44} data is exactly the same as the frequency obtained for the C' mode cannot be answered on the basis of the present results.

The preceding discussion indicates that low-frequency resonance modes cause a significant change in the normal temperature dependence of the elastic constants of irradiated crystals. The question then arises as to the magnitude of similar contributions to the temperature dependence of other physical properties. For example, the vibrational energy associated with a defect resonance mode should contribute to the stored energy of an irradiated solid. This energy (approximately 4×10^{-3} eV at 35 K for a frequency of 5×10^{12} Hz) is very small in relation to the value of about 5 eV for the formation energy of a Frenkel pair, and therefore would not be discernible in a measurement of the stored energy released during annealing. However, this effect would also contribute to the heat capacity of the irradiated solid at temperatures below the annealing range of the defects which possess the resonance modes. The magnitude of this contribution may be estimated by considering the change in the heat capacity at constant volume, ΔC_V, produced by N defects each with one resonance mode of frequency ω_E:

$$\Delta C_V = \frac{N}{k}\left(\frac{\hbar\omega_E}{T}\right)^2 \frac{e^{\hbar\omega_E/kT}}{(e^{\hbar\omega_E/kT}-1)^2}. \tag{16}$$

The relative contribution from a resonance mode at any given temperature to the specific heat is therefore determined only by its characteristic frequency. This calculation gives a maximum percentage change at a temperature near 9 K. At this temperature, it gives a change in the total heat capacity of copper of 2.2×10^{-4} J/mole K (approximately 0.5% of the total specific heat) for a resonance frequency of 5×10^{12} Hz and an atomic fraction of contributing defects of 10^{-4}. For frequencies of 4×10^{12} and 6×10^{12} Hz, and the same concentration of defects, the change in the total specific heat at this temperature would be approximately 0.8% and 0.3%, respectively. Changes in the heat capacity of this magnitude can be easily measured, [38] so that a measurement of the irradiation-produced change in the heat capacity at low temperatures should permit an accurate determination of the frequency of the resonance mode.

The presence of these modes should also change the temperature dependence of the lattice thermal expansion. The magnitude of this contribution is given by Eq. (11). However, since the value of $\partial\omega_E/\partial V$ is unknown, no numerical value can be given.

The temperature dependence of the electrical resistivity (deviations from Matthiessen's rule) should likewise be altered by the introduction of defect vibrational modes. In fact, the large resistivity effects observed by Magnuson, Palmer, and Koehler, [4] and which they interpreted as a large shift in the Debye characteristic temperature, are probably a consequence of the creation of resonance modes characteristic of stage I defects. However, the effect on the resistivity temperature dependence is difficult to analyze quantitatively, and no estimate will be given.

The presence of resonance mode effects could be expected for other defects as well as for the free dumbbell interstitial. For example, the frequency factor for the I_C relaxation process was found to be $\sim8\times10^{11}$ Hz. This factor includes an

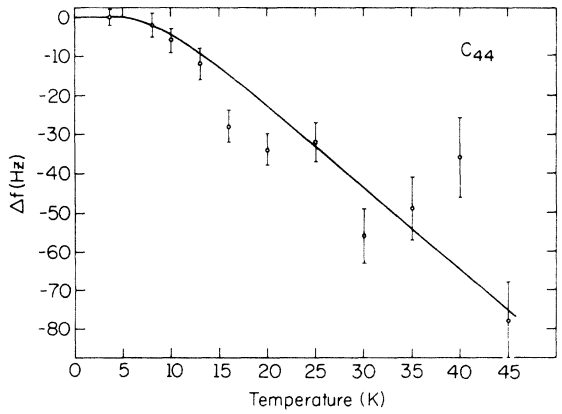

FIG. 6. Change in the temperature dependence of the C_{44} resonant frequency from the defects which anneal in the 45–60-K temperature range. The solid curve represents the least-squares fit of the temperature-dependent terms in Eq. (14) obtained with the C' data for $\omega_E = 5\times10^{12}$ Hz.

entropy factor, so the actual frequency involved could be much lower, and thermal excitation of this vibration could lead to measurable resonant-mode effects. In particular the relative specific-heat change, which depends only on the mode frequency, should be even larger at low temperatures than the I_D interstitial effects noted above. A measurement of such an effect would then provide a means for determining both the frequency and entropy factor of the I_C defect. Since the strain derivatives of the frequencies are not known there is no way to estimate the magnitude of the I_C resonant-mode contributions to the elastic-constant temperature dependence, and in fact no measurable effects were observed in the C' measurement of paper I (see Table III). However no measurements of their possible contributions to C_{44} were carried out.

In conclusion, the present results indicate that resonance modes should significantly change the temperature dependence of a number of physical properties. Since changes in elastic constants can be measured very accurately at low temperatures, and because their temperature dependence can be easily related to resonance-mode effects, the measurement of temperature-dependent effects of stage-I defects on the elastic constants provides a particularly convenient means for investigating the properties of resonance modes. A more detailed experiment especially designed for measuring irradiation produced changes in the temperature dependence of the elastic constants in the range from 3–25 K seems quite promising.

IV. SUMMARY

Measurements of changes in the elastic constants of copper and their temperature dependences resulting from stage-I_C close pairs and stage-I_D isolated interstitials are used to investigate defect configurations. The relative elastic constant changes at 3.6 K per unit concentration γ of isolated interstitials, $d\ln C/d\gamma$, are found to be 0 ± 1, -30, and -15 for the bulk modulus, C_{44}, and $C'=\frac{1}{2}(C_{11}-C_{12})$, respectively. The results can be understood in terms of four mechanisms.

(i) The bulk effect is a result of the change in number and strength of interatomic bonds caused by the creation of isolated vacancies and interstitials. Based on the results obtained for the change in the bulk modulus during annealing through stage I, this effect is so small relative to the other contributions, that not even its sign is known. This indication of a small effect is in agreement with the range of most theoretical estimates (-10 to $+10$).

(ii) Polarizability effects occur for defects whose atoms are not at perfect crystal lattice sites. In this case, an internal strain can be produced by the application of an applied stress, and this additional strain causes a decrease in the measured shear elastic constants. The calculated anisotropy depends upon the symmetry of the defect. The experimental results obtained at 3.6 K for the recovery of the three elastic constants during stage I_D are qualitatively in agreement with a calculation by Dederichs of the polarizability of the ⟨100⟩-split dumbbell interstitial.

(iii) Relaxation involves the thermally activated stress-induced ordering of preferential defect orientations, and therefore is a temperature-dependent effect. The relaxation process previously reported by Nielsen and Townsend has been observed. By combining the present results with those of NT, improved values have been obtained for the attempt frequency and energy of activation. We have attributed this relaxation to the I_C defect, and a model of this defect (Fig. 4) has been proposed. The model retains those features of the Peretto *et al.* model which are necessary to explain magnetic after effects in nickel as well. No other relaxation of stage-I defects has been observed.

(iv) The excitation of defect-resonance modes changes the vibrational spectrum of the lattice. This in turn introduces temperature-dependent changes in the elastic constants. The results obtained for the change in the temperature dependence of the C' elastic constant induced by the I_D defects (Fig. 5) provide evidence for ⟨100⟩-split interstitial-defect-resonance modes. The frequency of the resonance mode is found to be:

$$\omega = 5\times10^{12}\pm30\%\ \text{Hz}\,.$$

This frequency is in good agreement with the value of approximately 6×10^{12} Hz obtained by Dederichs *et al.* by a computer simulation of a ⟨100⟩-split interstitial in a copper lattice. The results for the temperature-dependence change in the C_{44} elastic constant introduced by the I_E defects (Fig. 6) are also consistent with the expected behavior from defect-resonance modes. However, the interpretation of the effect is more difficult in this annealing range, and it is not possible to say whether or not the frequency obtained is exactly the same as that from the I_D defects.

Based on the value of the frequencies obtained here, the excitation of resonance modes from both I_C and I_D defects should also contribute significantly to the temperature dependence of other physical properties. In particular, low-temperature elastic-constant and heat-capacity measurements should serve as useful tools with which to investigate these effects further.

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