

Formation of solute atmospheres around dislocations*

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Solute atoms form long-range atmospheres around dislocations when in thermal equilibrium. These atmospheres scatter phonons and can be studied through their effect on the thermal conductivity. Previous studies indicate that solute atmospheres are formed after plastic deformation at room temperature in Cu-Al but not in Cu-Ge. Ordinary diffusion at room temperature is too slow to permit atmospheres to form, but excess vacancies produced by plastic deformation can enhance diffusion. One can define a net time-integrated diffusion due to excess vacancies until the excess is exhausted in terms of R^2 , where R is the range over which the atmosphere attains equilibrium. It depends on the excess concentration and vacancy lifetime. The lifetime of the vacancies is calculated, assuming that the dislocations act as vacancy sinks. The resulting R is independent of the vacancy jump rate, and depends only on the initial vacancy concentration and on the dislocation density. The values of R thus calculated are much smaller than observed. However, the tendency for the vacancies to be associated with the solute atoms, and the possibility of dislocations not acting as perfect sinks of vacancies, may enhance the effectiveness of a vacancy in promoting solute diffusion and bring the value of R closer to the observed magnitude. Estimates of R are given for Cu-Al and Cu-Ge. The case of Al-Mg is also discussed.

I. INTRODUCTION

In substitutional alloys there is an interaction between the solute atoms and the strain field of a dislocation. There is, thus, a tendency for the concentration of solute atoms to be modulated by the strain field and for solute atmospheres to be formed around each dislocation. If solute and solvent atoms differ in size, the free energy is lowered by a modulation in the concentration. The local concentration $c(\vec{r})$, in equilibrium at a temperature T_0 becomes

$$c(\vec{r}) = c_0 \exp[-\Delta(\vec{r})K\delta V/kT_0], \quad (1)$$

where c_0 is the average solute concentration, $\Delta(\vec{r})$ is the local value of the dilatation due to the dislocation, K is the bulk modulus, δV is the difference in volume between solute and solvent atom, and k is the Boltzmann constant.

The presence of solute atoms changes the local value of the phonon velocity, so that solute atmospheres scatter phonons. Their contribution to the scattering amplitude is additional to that due to the inhomogeneous strain field of the dislocation itself. The solute atmospheres thus enhance phonon scattering by a dislocation relative to the value due to a "bare" dislocation, and thus cause a further decrease in the lattice thermal conductivity.¹

In order for solute atmospheres to occur it is not only necessary that they are favored configurations at thermal equilibrium, but it is also necessary that the atmospheres have an opportunity to attain their equilibrium distribution. The formation of solute atmospheres involves a diffusive motion of solute atoms around a dislocation, as discussed by Cottrell and Jawson.² The time required for the

diffusion is roughly of the order of the value of R^2/D , where R is the range of the atmosphere and D the diffusion coefficient. The "range" of the atmosphere, though not a well-defined quantity, may be understood as follows: The scattering of a phonon by a dislocation and by its associated atmosphere involves the properties of the solid to a distance of the order of a phonon wavelength from the dislocation core. Since the wavelength of the phonons which dominate the thermal conduction process at temperature T varies inversely as T , the range R also depends inversely on the temperature at which the lattice thermal conductivity is measured. In the measurements of Mitchell *et al.*³ and of Friedman *et al.*⁴ of the lattice thermal conductivity of deformed Cu-Al alloys the important range of measurements was 1 to 4 °K. The corresponding range R over which atmospheres were studied was thus of the order 100–400 Å. In these studies it was assumed that, after plastic deformation at room temperature, the atmospheres attained an equilibrium value according to (1) with $T_0 = 300$ °K. Subsequent annealing to higher temperatures was assumed to reduce the strengths of the atmospheres according to (1), when T_0 is now replaced by the annealing temperature.

However, the diffusion coefficient D decreases rapidly with decreasing temperature, so that no appreciable diffusion can take place over the range of interest in the time available. In fact, the experiments mentioned^{3,4} were based on the assumption that no diffusion took place at room temperature after the strength of the atmospheres was reduced by annealing, so that the equilibrium appropriate to the annealing temperature was frozen in. Yet it was also assumed that room-temperature

equilibrium was established after the initial plastic deformation. This inconsistency can only be resolved if one assumes that diffusion was much more rapid immediately after severe plastic deformation.

Since plastic deformation creates an excess of vacancies which in turn promote excess diffusion, this assumption is not unreasonable. It is the purpose of this paper to investigate whether the excess vacancies can promote enough diffusion of solute atoms to allow atmosphere formation to the degree observed.

II. MODEL

We have to consider two diffusion problems: the diffusion of the vacancies to the sinks, and the diffusion of the solute atoms around the dislocations. The vacancy concentration, determined in the first problem, governs the diffusion coefficient in the second problem, since the diffusion coefficient is given by

$$D = \alpha a^2 \nu C_v, \quad (2)$$

where α is a factor which includes correlation factor, a is the lattice parameter, ν is the vacancy jump frequency, and C_v is the vacancy concentration (fraction per atomic site). The vacancy concentration is composed of C_{0v} , the thermal-equilibrium concentration, and C_{ev} , the excess vacancy concentration due to plastic deformation. We shall assume that $C_{ev} \gg C_{0v}$, so that

$$C_v \simeq C_{ev}. \quad (3)$$

We shall assume that C_{ev} has a uniform value initially, that is, immediately after the cessation of plastic deformation, and that the dislocations act as vacancy sinks. As the vacancies diffuse towards the dislocations, C_{ev} decreases with time. We shall make a simplifying assumption that C_{ev} is independent of position, and only a function of time. This cannot be strictly true, as will be discussed below, and the vacancy concentration must be less in the immediate vicinity of the dislocation core. This makes the solute diffusion coefficient a function of position, and complicates the solute diffusion problem. However, this will mainly affect the time development of the solute atmosphere $c(\vec{r})$ in the immediate vicinity of the dislocation line.

The excess vacancy concentration depends on the applied plastic strain ϵ , according to the empirical relation

$$C_{ev} = B\epsilon^m. \quad (4)$$

Theoretical estimates⁵⁻⁸ and resistivity measurements⁹ give $m \simeq 1$ to 1.5 and (for $m = 1$) $B = 10^{-4}$. Thus, for 10% plastic strain,

$$C_{ev} = 10^{-4} \times (0.1)^1 = 10^{-5} \text{ per atomic site}. \quad (5)$$

A theoretical treatment given by Nabarro¹⁰ justifies (4) by considering the formation energy of a vacancy and the work done during plastic deformation.

The value of C_{ev} given by (4) and (5) is the initial vacancy concentration $C_{ev}(0)$ after plastic deformation ceases. It is now assumed that the vacancy concentration decays with time, according to

$$\frac{d}{dt} C_{ev} = C_{ev}/\tau \quad (6)$$

and that, after plastic deformation ceases,

$$C_{ev}(t) = C_{ev}(0) e^{-t/\tau}. \quad (7)$$

The decay constant τ depends on the rate at which vacancies diffuse to the sinks. The sinks are the dislocations, so that τ depends on the diffusivity of the vacancies and on the dislocation density.

III. VACANCY MOTION

Assuming that the excess vacancy concentration is uniform and has an initial value $C_{ev}(0)$, and that the dislocations act as sinks, one can calculate the lifetime τ by following the solution of the vacancy diffusion equation of Seidman and Balluffi.¹¹ It is assumed that the dislocation network can be approximated by a regular array of parallel dislocations. Each dislocation then dominates the diffusion in the cylindrical region of outer radius R_0 , where

$$R_0 = (\pi\rho)^{-1/2}, \quad (8)$$

ρ being the dislocation density. The assumption of a regular array is an idealization. Ham,¹² however, has shown that a regular and a random array behave similarly (to within 10%) when the dislocations act as sinks. In any case, the dislocation arrangement may well be nonrandom.

To obtain the flux of vacancies into the core, Seidman and Balluffi¹¹ solved the vacancy diffusion equation

$$\frac{\partial}{\partial t} C_{ev} = \nabla \cdot (D_v \nabla C_{ev}), \quad (9)$$

where D_v is the diffusion coefficient for vacancy motion. The boundary conditions are that the vacancy flux vanishes at $r = R_0$, and that the concentration vanishes at $r = r_0$, where r_0 is a capture radius for the capture of vacancies by the dislocation core.

The solution is given in terms of complicated Bessel functions¹¹⁻¹⁴

$$C_{ev}(r, t) \propto \pi \sum_{n=1}^{\infty} \left(J_1^2(R_0 \alpha_n) \times \frac{J_0(r_0 \alpha_n) Y_0(r \alpha_n) - J_0(r \alpha_n) Y_0(r_0 \alpha_n)}{J_0^2(r_0 \alpha_n) - J_1^2(R_0 \alpha_n)} \times \exp(-D_v t \alpha_n^2) \right), \quad (10)$$

where α_n are the roots of

$$Y_0(r_0\alpha_n)J_1(R_0\alpha_n) - J_0(r_0\alpha_n)Y_1(R_0\alpha_n) = 0. \quad (11)$$

The notation of the Bessel functions is that of Jahnke and Emde.¹⁵

The average vacancy concentration $\langle C_{ev} \rangle$ can be expressed in terms of its initial value by integrating $C_{ev}(r, t)$ over the volume of the cylinder, which gives

$$\frac{\langle C_{ev} \rangle}{C_{ev}(0)} = \frac{4}{R_0^2 - r_0^2} \sum \frac{J_1^2(R_0\alpha_n) \exp(-\alpha_n^2 D_v t)}{\alpha_n^2 [J_0^2(r_0\alpha_n) - J_1^2(R_0\alpha_n)]}. \quad (12)$$

Ham¹² has shown that the largest fraction of the concentration decays according to the first term in the eigenfunction expansion.

The fraction of total vacancies contributed by the n th term in Eq. (12) is given by

$$\frac{4J_1^2(R\alpha_n)}{(R_0^2 - r_0^2) \alpha_n^2 [J_0^2(r_0\alpha_n) - J_1^2(R_0\alpha_n)]}, \quad (13)$$

which yields a value of above 0.99 for the lowest order component α_1 , according to Ham.¹² Penning¹³ has also calculated, for various dislocation densities, the ratios of contributions from the first and the second terms of the eigenfunction expansion. According to these calculations, no more than 10% of the total contribution of vacancies can be ascribed to second- and higher-order terms. It is within this approximation, that we have considered C_{ev} independent of position. Hence Eq. (12) can be written

$$\langle C_{ev}(t) \rangle / C_{ev}(0) \approx \exp(-\alpha_1^2 D_v t), \quad (14)$$

where

$$\alpha_1^2 = 2(R_0^2 - r_0^2)^{-1} \ln(R_0/r_0 - \frac{3}{8}). \quad (15)$$

We are particularly interested in the case when $R_0 \gg r_0$. Since R_0 is related to the dislocation density by (8), Eq. (15) can then be written

$$\frac{\langle C_{ev}(t) \rangle}{C_{ev}(0)} \approx \exp\left(\frac{-2\pi\rho D_v t}{\ln(R_0/r_0)}\right). \quad (16)$$

It will be seen in Sec. IV that the degree of atmosphere formation is essentially independent of D_v . The solute diffusion coefficient, owing to (2) and (16), will be a function of time, but the range of the atmosphere formation over a long time will not depend on D_v , since it will depend on the time integral of the diffusion coefficient

$$\int D(t) dt = \langle Dt \rangle, \quad (17)$$

which will be independent of D_v .

This, however, assumes that the atmosphere formation can proceed without interruption until all excess vacancies are exhausted. It is thus necessary to estimate τ of (7) and to verify that is

less than the time that has typically elapsed between plastic deformation and the low-temperature thermal conductivity measurements, which indicate the presence or absence of solute atmospheres.

From (7) and (16)

$$\tau = \ln(R_0/r_0) / (2\pi\rho D_v). \quad (18)$$

After severe plastic deformation, the dislocation density is typically 10^{10} – 10^{11} lines/cm², so that $R_0 = 600$ Å and $\ln(R_0/r_0)$ is between 5 and 6. The vacancy diffusion rate in copper at room temperature is of the order of 10^{-13} cm²/sec; the diffusion actually takes place as divacancies.¹⁶ Thus τ is typically of the order of 10^3 sec if all dislocations act as sinks, 2×10^4 sec if only 5% of all dislocations act as sinks. Since the storage time of specimens between plastic deformation and thermal conductivity measurements is typically an order of magnitude larger, it seems justified to regard excess vacancies as exhausted and the atmospheres as fully formed as they will ever be before measurements are made.

IV. SOLUTE ATOM MOTION

If the solute concentration departs from its equilibrium value $c(\vec{r})$ of Eq. (1) by an amount $n(\vec{r})$, then solute diffusion will tend to decrease $n(\vec{r})$ according to

$$\frac{\partial}{\partial t} n(\vec{r}) = \nabla \cdot [D \nabla n(\vec{r})], \quad (19)$$

where the solute diffusion coefficient D , given by (2), is proportional to C_{ev} and is thus a function of time (7). If $n(\vec{r})$ is expressed as a linear superposition of Fourier components, i.e.,

$$n(\vec{r}) \propto \sum_{\vec{q}} n(\vec{q}) e^{i\vec{q} \cdot \vec{r}}, \quad (20)$$

each component satisfies the equation

$$\frac{d}{dt} n(\vec{q}) + D(t) q^2 n(\vec{q}) = 0. \quad (21)$$

The advantage of treating $n(\vec{r})$ in terms of Fourier components is that each component $n(\vec{q})$ governs the scattering of phonons through a wave vector \vec{q} , so that the time dependence of phonon scattering is governed by a simple equation¹⁷

$$n(\vec{q}, t) = n(\vec{q}, 0) e^{-t/\tau(\vec{q})}, \quad (22)$$

where

$$\tau(\vec{q}) = (Dq^2)^{-1}. \quad (23)$$

Since $\tau(\vec{q})$ increases with decreasing q , and since the high- q components of n describe mainly the solute atmosphere at small distances from the dislocation core, the range R over which the atmosphere is essentially complete in a time t_0 is given by

$$R \approx (Dt_0)^{1/2} \quad (24)$$

and the wave vector through which phonons are scattered by essentially equilibrated atmospheres is given by

$$q_0 = (Dt_0)^{-1/2} \approx 1/R \quad (25)$$

These considerations apply strictly only to the case of a constant diffusion coefficient D .

If the diffusion coefficient varies with position (due to a possible strain dependence), Eqs. (19) and (21) become complicated and can only be solved in an approximate manner. In the present case, however, we are concerned with the time dependence of the solute diffusion coefficient. In this case, Eqs. (22) and (23) must be generalized to

$$n(\vec{q}, t) = n(\vec{q}, 0) \exp\left(-q^2 \int_0^t D(t) dt\right) \quad (26)$$

and because of (17) and (16), and using (2),

$$n(\vec{q}, t) = n(\vec{q}, 0) \exp\left(-q^2 \alpha a^2 \nu C_{ev}(0) \frac{\ln(R_0/r_0)}{2\pi\rho D_v}\right) \quad (27)$$

Now, D_v , the vacancy diffusion coefficient, although a strong function of temperature, is related to ν , the vacancy jump frequency, by

$$D_v = \alpha a^2 \nu \quad (28)$$

so that (27) simplifies to

$$n(\vec{q}, t) = n(\vec{q}, 0) \exp\left(-q^2 C_{ev}(0) \frac{\ln(R_0/r_0)}{2\pi\rho}\right) \quad (29)$$

No distinction has been made so far between the motion of solute and solvent atoms, so that α is the same in (2) and (28). The necessary generalization will be discussed in Sec. V.

Atmosphere formation will be essentially complete for those Fourier components of the solute distribution for which terms in large parentheses in Eq. (29) are of the order unity. This implies that the range of the formed atmosphere R is given by

$$R^2 = C_{ev}(0) \ln(R_0/r_0) / 2\pi\rho \quad (30)$$

For a typical dislocation density of 10^{10} cm^{-2} and an initial vacancy concentration of 10^{-5} , the range of atmosphere formation R would be of the order of $3 \times 10^{-8} \text{ cm}$. Note that R depends only on the dislocation density and the initial vacancy concentration. It does not depend on ν or D_v ; the ultimate degree of atmosphere formation does not depend on the rate at which vacancies move but only on the number of steps a vacancy moves before reaching a sink and of course on the initial number of vacancies. This is why (30) contains, apart from $C_{ev}(0)$, only geometrical factors relating to sink density.

Our result [Eq. (30)] that the time required for

the formation is essentially independent of the vacancy diffusion rate D_v , is a consequence of our simplifying assumptions. Firstly, we have assumed that $C_{ev} \gg C_{ov}$, hence our theory would only hold at low temperatures. Secondly, we have assumed a unique vacancy diffusion coefficient D_v . In actual fact, there are single vacancies, divacancies and trivacancies, each with its own diffusion coefficient. The ratio of these species will initially differ from its mutual equilibrium value, and gradually attain mutual equilibrium. This makes the effective D_v time dependent. For an initial concentration of 10^{-5} , it takes 10^5 steps for a vacancy to reach a dislocation; it also takes 10^5 steps for a vacancy to reach another vacancy. Thus, for our dislocation density, our estimate of R^2 may well be in error by a numerical factor of the order of unity, but not likely to be in error by an order of magnitude.

V. DISCUSSION

A range of $3 \times 10^{-8} \text{ cm}$, as derived from Eq. (30), is not sufficient to account for the formation of any solute atmosphere, and certainly not for the atmosphere formation found in deformed Cu-Al alloys by means of low-frequency phonons.^{3,4} The low-temperature thermal-conductivity studies extended down to temperatures of 1.5 °K. Since the dominant phonons in thermal conduction have wave numbers around

$$q = (T/\Theta) (6\pi^2)^{1/3} a^{-1} \quad (31)$$

where a^3 is the atomic volume and Θ the Debye temperature, these experiments indicate that atmospheres are essentially complete for $q \approx 1/R$, where R is at least about $170 \times 10^{-8} \text{ cm}$ (taking $\Theta = 330 \text{ °K}$ and $a = 3 \times 10^{-8} \text{ cm}$). The value of R^2 required to account for the thermal conductivity measurements thus exceed the order given by Eq. (30) by a factor of about 3000.

It is quite likely that the present considerations underestimated the solute diffusion and hence the value of R^2 . The diffusion rate of the solute atoms differs from the self-diffusion rate, partly because there is a tendency for vacancies to be associated with solute atoms and partly because the jump frequency of the solute atoms may be different.

However, Manning¹⁸ has shown that the correlation factor which enters as a factor in α of Eq. (2) can be strongly temperature dependent when two interdiffusing species have substantially different jump rates. This result can also be understood to reflect the requirement that the sum of the fluxes must vanish. His treatment does not consider association between one species and the vacancy, and this association can lead to a large difference between the effective diffusion rate of the solute

and the self-diffusion rate of the parent metal.

It may be argued that if there is a strong association between vacancies and solute atoms, enhancing the diffusion of solute atoms, the same factor would also reduce the vacancy lifetime and be eliminated in (30), which is independent of the diffusion rate. However, the motion of vacancies to the sinks must be, in the aggregate, the motion of disassociated vacancies, otherwise the motion would increase the solute concentration at the vacancy sinks.

Thus the vacancy diffusivity D_v in Sec. III must be understood to be the diffusivity of disassociated vacancies. In dilute alloys this would be related to the self-diffusion coefficient of the parent metal by Eqs. (2) and (28).

The enhancement in the diffusion rate of the solute can be obtained from the ratio

$$E = D_s/D_0, \quad (32)$$

where D_s is the interdiffusion coefficient of the solute and D_0 is the self-diffusion coefficient of the parent metal. This is because the diffusion of solute atoms around the dislocation is an interdiffusion process, and the measured values of D_s include changes in jump frequency, association effects, and correlation effects. On the other hand D_0 is related to the diffusivity of disassociated vacancies through Eqs. (2) and (28).

The enhancement ratio E is the ratio of the diffusivities at room temperature, while D_s and D_0 are measured at higher temperatures, and must be extrapolated to room temperature. This extrapolation introduces some uncertainty.

From the interdiffusion data for Al in Cu by Rhines and Mehl and da Silva and Mehl quoted by Smithells,¹⁹ and the copper self-diffusion data quoted by Peterson,²⁰ the room-temperature value of E is 3.2×10^3 . This enhancement factor would make the value of R^2 agree with the thermal conductivity data. For $\rho = 10^{10} \text{ cm}^{-2}$, and $C_{ev}(0) = 10^{-5}$ atomic fraction, R would be increased to $170 \times 10^{-8} \text{ cm}$, which, as discussed earlier, is just the value needed to account for thermal resistance down to 1.5°K .

However, if the recent data of Oikawa *et al.*²¹ is used in conjunction with the same self-diffusion data,²⁰ one obtains an extrapolated value of $E = 3.4 \times 10^2$.

Hence it is probable that the diffusion enhancement factor does not fully account for the observed atmosphere formation in Cu-Al. There is yet an-

other factor which may modify the estimate of R^2 . We have assumed that all the dislocations act as perfect sinks for vacancies. If only a fraction η of all dislocations capture vacancies, or alternatively if the dislocations capture vacancies within a radius r_0 with an efficiency η , then the estimates of R^2 of (30) must be increased by a factor $1/\eta$, and R increased by a factor of $(1/\eta)^{1/2}$.

Unfortunately, we do not have an independent measure of this factor η . Mitchell *et al.*²² studied the motion of dislocations in deformed Cu-Al, and found that during plastic deformation, the majority of dislocations seemed unable to climb. Since a dislocation which cannot climb cannot accept vacancies, it may well be that η , the fraction of dislocations acting as vacancy sinks, is well below unity in this material. Whether it is low enough to account for the missing factor 10, is of course impossible to say at present.

The possible formation of solute atmospheres was studied by thermal conductivity also in the case of Cu + 7-at. % Ge by Friedman.²³ In this case no evidence for atmosphere formation was found up to 4.2°K . This means that in this system R must be less than $65 \times 10^{-8} \text{ cm}$. Using diffusion data by Reincke and Dahlstrom²⁴ on dilute Cu-Ge, and assuming that the activation energy varies with solute concentration similarly to Cu-Al,²⁵ E is about 130, so that R would be $36 \times 10^{-8} \text{ cm}$. It would thus appear that atmospheres should visibly affect the thermal resistivity of Cu-Ge only at and above 10°K . It would thus be of interest to extend lattice thermal conductivity studies of Cu-Ge alloys to higher temperatures.

Another system of interest is aluminum-rich Al-Mg. The association energy between a solute atom and a vacancy, as measured by an equilibrium method²⁶ is only about 0.08 eV. However, the apparent association energy as deduced from the interference of Mg on the motion of Cu is about 0.20 eV.²⁷ Unfortunately no reliable data on the interdiffusion is known to us at the present time. It would thus be of interest to see whether the Al-Mg system shows atmosphere formation in its temperature thermal conductivity.

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