Nonlinear acoustic response from precipitate-matrix misfit in a dislocation network

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An analytical model is presented for the effects on the acoustic nonlinearity of the interaction of matrix dislocations with precipitate coherency strains in metallic alloys. The acoustic nonlinearity parameters are shown to depend linearly on the dislocation density, volume fraction of precipitates and precipitate-matrix misfit parameters, and on the fourth power of the dislocation loop length. The model predicts changes in the acoustic nonlinearity parameter associated with the growth of precipitates during the artificial aging of aluminum alloy 2024 that are in good agreement with experimental measurements reported in the literature. [S0021-8979(98)04222-4]

I. INTRODUCTION

Cantrell and Yost[1] have investigated the effect of precipitate-matrix coherency strains on the generation of acoustic second harmonics in metallic alloys. They developed a model for evaluating the change in the acoustic nonlinearity parameter β due to the effects of coherency strains on the elastic constants of the material and applied the model to the precipitation aging of aluminum alloy 2024 from the T4 to the T6 temper. For the purposes of model expediency and tractability, the precipitates are assumed to be spherical, elastically isotropic, and randomly distributed throughout an isotropic matrix. The coherency strains used in the calculations are obtained in Ref. 1 by averaging the strains along a radius connecting nearest neighbor precipitates. Such an averaging procedure neglects potentially important off-axis contributions to the averaging which must be considered. We thus propose in Sec. II a more realistic volume averaging of the coherency strains and find the change in the nonlinearity parameters due to stress-induced changes in the elastic constants to be considerably lower than that predicted in Ref. 1 using line averaging. The resulting discrepancy between model predictions using volume averaging of strains and experimental measurements of the nonlinearity parameters of aluminum alloy 2024 leads to a consideration in Sec. III of the effects of the coherency strains on the dislocations in the matrix. We show that the effects of such coherency strain-dislocation interactions are, indeed, substantial and account quite well for the experimentally measured changes in the nonlinearity parameters reported in Ref. 1.

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II. ACOUSTIC NONLINEARITY AND COHERENCY STRAINS IN DISLOCATION-FREE MATERIALS

A. Elastic nonlinearity and precipitate-matrix coherency strains

Let us denote the position of a material particle in some arbitrary stress-free configuration by the position vector $\mathbf{X}$. If the material is subjected to a local deformation at $\mathbf{X}$, the position of the same material particle in the deformed position is denoted by the position vector $\mathbf{x}$. The particle displacement vector is defined by $\bar{\mathbf{u}}=\mathbf{x}-\mathbf{X}$. We write the nonlinearity parameter in the zero stress state of the material as[1]

$$\beta_0=-\left(\frac{3\bar{C}_{11}+\bar{C}_{111}}{\bar{C}_{11}}\right), \tag{1}$$

where $\bar{C}_{11}$ and $\bar{C}_{111}$ are the second- and third-order Brugger elastic constants (written in Voigt contracted notation) referred to the zero stress state. The change in the nonlinearity parameter resulting from the elastic strains $\bar{u}_{ij}=\partial\bar{u}_i/\partial\bar{X}_j$, where u_i are the Cartesian components of the particle displacement vector, is given by

$$\begin{aligned}\beta-\beta_0=&\left(3-\frac{\bar{C}_{111}}{C_{11}}-\frac{\bar{C}_{1111}}{C_{11}}+\frac{\bar{C}_{111}^2}{C_{11}^2}\right)\bar{u}_{11}\\&+\left(3\frac{\bar{C}_{12}}{C_{11}}-\frac{\bar{C}_{1112}}{C_{11}}+\frac{\bar{C}_{12}\bar{C}_{111}}{C_{11}^2}+\frac{\bar{C}_{112}\bar{C}_{111}}{C_{11}^2}\right)\\&\times(\bar{u}_{22}+\bar{u}_{33}),\end{aligned} \tag{2}$$

where β is the acoustic nonlinearity parameter referred to the stressed state.

We assume that the average shape of the randomly orientated precipitates is spherical and that the precipitate has an effective misfit parameter equal to an appropriate average of misfit parameters associated with each precipitate-matrix

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interface plane of the actual precipitate. We thus consider an isotropic matrix embedded with isotropic spherical precipitates. For expediency, we assume that the elastic moduli of the precipitate and matrix material are approximately equal. The effective misfit parameter δ of a spherical precipitate is equivalent[2] to that associated with placing a sphere of natural radius $r_0(1+\delta)$ into a spherical hole of radius r_0. Both the sphere and the hole relax to a final radius $r_1=r_0(1+\epsilon)$. The displacement field around such a spherical precipitate is given as[2]

$$\overline{u}_i=\epsilon r_1^3\frac{\overline{X}_i}{r^3},\quad r^2=(\overline{X}_1^2+\overline{X}_2^2+\overline{X}_3^2),\quad i=1,2,3 \tag{3}$$

for $r\geqslant r_1$. For $r<r_1$, we write

$$\overline{u}_i=(\epsilon-\delta)\overline{X}_i,\quad i=1,2,3. \tag{4}$$

The relationship between ϵ and δ is determined by the condition that in the relaxed state the inward and outward pressures developed on the sphere and hole surfaces are equal. Such a condition leads to the expression[3]

$$\epsilon=\frac{(1+\nu)}{3(1-\nu)}\,\delta, \tag{5}$$

where ν is Poisson's ratio.

B. Contributions to elastic nonlinearity from precipitate and matrix material

We consider separately the contribution to the change in the nonlinearity parameter from the matrix material and the precipitates. We assume

$$\beta-\beta_0=f_p\Delta\beta_p+(1-f_p)\Delta\beta_m, \tag{6}$$

where f_p is the volume fraction of precipitates, $\Delta\beta_p$ is the change in the nonlinearity parameter of the precipitates, and $\Delta\beta_m$ is the change in the nonlinearity parameter of the matrix. To obtain the volume averaged strains in the matrix, consider the strain contributions along three mutually orthogonal radial directions from the center of the precipitate, say $\overline{X}_1$, $\overline{X}_2$, and $\overline{X}_3$, at equal distances into the matrix. Summing the contributions in Eq. (2) from these directions, we obtain for an isotropic matrix

$$\Delta\beta_m=\frac{1}{3}\left[\left(3-\frac{\overline{C}_{111}}{C_{11}}-\frac{\overline{C}_{1111}}{C_{11}}+\frac{\overline{C}_{111}^2}{C_{11}^2}\right)+2\left(3\frac{\overline{C}_{12}}{C_{11}}-\frac{\overline{C}_{1112}}{C_{11}}+\frac{\overline{C}_{12}\overline{C}_{111}}{C_{11}^2}+\frac{\overline{C}_{112}\overline{C}_{111}}{C_{11}^2}\right)\right]\times(\overline{u}_{11}+\overline{u}_{22}+\overline{u}_{33}). \tag{7}$$

It is straightforward to show from Eq. (3) that $(\overline{u}_{11}+\overline{u}_{22}+\overline{u}_{33})=\nabla\cdot\overline{\mathbf{u}}=0$. Thus, the displacement field is nondilatational. Since we have assumed an isotropic matrix, the results hold in general throughout the matrix and we obtain $\Delta\beta_m=0$. This null result is a direct consequence of the assumption of longitudinal wave propagation in an isotropic material in which the precipitate-matrix misfit produces pure shear strains in the matrix.

In the precipitate, Eq. (4) reveals that $\overline{u}_{11}=\overline{u}_{22}=\overline{u}_{33}=(\epsilon-\delta)$. If we assume that the Hiki–Granato relations[4] $\overline{C}_{11}=2\overline{C}_{12}$, $\overline{C}_{111}=2\overline{C}_{112}$, $\overline{C}_{1111}=2\overline{C}_{1112}$ hold for the elastic constants and that[5,6] $C_{111}/C_{11}\approx\overline{C}_{1111}/\overline{C}_{111}$, then we obtain from Eqs. (2) and (6) that the fractional change in the acoustic nonlinearity parameter is

$$\frac{\beta-\beta_0}{\beta_0}\approx\frac{6}{\beta_0}f_p(\epsilon-\delta)=-\frac{4(1-2\nu)}{\beta_0(1-\nu)}f_p\delta, \tag{8}$$

where the last equality follows from Eq. (5). We thus find that only the precipitates contribute to the change in the nonlinearity parameter as the result of coherency strains in this model.

For aluminum alloy 2024-T4, the GP zones have an estimated average value[1] of $\langle\delta_{\mathrm{GP}}\rangle\approx-8\%$ for the misfit parameter, $\beta_0\approx4.1$, and $\nu=0.33$. The volume fraction of GP zones in AA2024-T4 is more difficult to ascertain for stock material. We estimate the volume fraction by noting that the value $(f_p)_{\mathrm{GP}}\approx0.003\%$ is calculated from experimental measurements[7] of Al-4% Cu binary alloy. The addition of Mg in AA2024-T4 traps vacancies in the vicinity of the relatively large Mg atoms. The trapped vacancies accelerate and intensify the formation of GP zones in that alloy by allowing the movement of Cu atoms into GP zones.[8] Studies[8] of GP zones in AA7075 aged at room temperature for 25 years reveal a volume fraction of GP zones of approximately 2%. From a consideration of the equilibrium $CuAl_2$ phase in Al–Cu–Mg alloys,[9] we infer that the volume fraction of GP zones in AA2024-T4 after similar aging times at room temperature is of the order of 1%–4%. The exact number depends on the Cu to Mg ratio and the presence of impurity elements among other factors. Thus, the volume fraction of GP zones in a typical AA2024-T4 specimen must lie between that of the Al-4% Cu binary alloy and that of long-aged AA2024-T4. AA2024-T4 exhibits exceptional hardening compared to Al-4% Cu binary alloy. Since GP zones are in large measure responsible for the increased hardness, it is reasonable to assume that the volume fraction of GP zones in AA2024-T4 lies closer to that of the long-aged AA2024-T4 values. We thus assume the order of magnitude estimate $(f_p)_{\mathrm{GP}}\approx0.3\%$ for stock AA2024-T4 specimens.

Using the value $(f_p)_{\mathrm{GP}}=0.3\%$, we calculate from Eq. (8) the fractional change in the nonlinearity parameter $[(\beta-\beta_0)/\beta_0]_{\mathrm{GP}}\approx0.01\%$. We note that if $(f_p)_{\mathrm{GP}}$ were equal to the long-aged value 4%, then the maximum fractional change in the nonlinearity parameter would be approximately 0.13%. For S' precipitates in AA2024, we assume[1] $\langle\delta_{S'}\rangle\approx-1.2\%$ for the misfit parameter and that $(f_p)_{S'}\approx7\%$. The fractional change in the nonlinearity parameter for S' precipitates is estimated from Eq. (8) to be $[(\beta-\beta_0)/\beta_0]_{S'}\approx0.04\%$. All these values are two to three orders of magnitude lower than that measured from the experiments reported in Ref. 1.

III. ACOUSTIC NONLINEARITY FROM COHERENCY STRAIN-DISLOCATION INTERACTIONS

Although the average strain in the matrix is found here to be zero, the local values of strain are nonzero. Indeed, the

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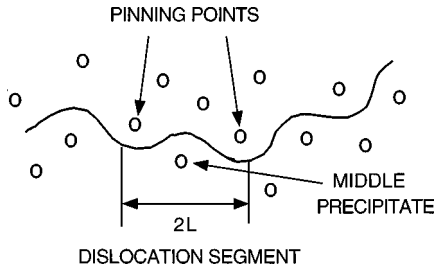

FIG. 1. Bowing (three-point bending) of dislocation segments on a matrix slip plane resulting from local precipitate coherency stresses. The dislocation is represented by the solid curve and the random array of precipitates is represented by circles. The average distance between the pinning points of the bowed dislocation segments is $2L$.

local strains can be quite large, becoming sufficiently large to nucleate misfit dislocations when the precipitates grow to a critical size. We thus consider the effect of local strains on the dislocation network in the matrix. We assume some density Λ of isolated single dislocations in the material. For definiteness, we assume edge dislocations lying in arbitrary slip planes in grains of arbitrary orientation in a polycrystalline solid. For a given dislocation, a longitudinal stress $\bar{\sigma} = \bar{\sigma}_1$ (Voigt notation) applied to the solid and referred to the zero stress configuration will give rise a shear stress $\bar{\tau}$ at the site of the dislocation of magnitude $\bar{\tau} = R\bar{\sigma}$ along its slip direction, where R is the Schmid or resolving factor. If the dislocation is pinned at points a distance $2L$ apart, the length of dislocation between the pinning points (dislocation segment) will bow out under the action of the resolved shear stress like an arched string. The movement (bowing) of the dislocation produces a plastic shear strain $\bar{\gamma}$ in the material. We assume that the relationship between the plastic strain $\bar{\gamma}$ and the longitudinal strain $\bar{u}_{11}$ is $\bar{u}_{11} = \Omega\bar{\gamma}$, where Ω is the conversion factor from shear strain to longitudinal strain. It is straightforward to show from the model of Hikata *et al.*[10] that the change in the nonlinearity parameter resulting from a longitudinal stress $\bar{\sigma}$ of magnitude $|\bar{\sigma}|$ imposed on a network of dislocations of density Λ is given by

$$\Delta\beta^{\text{disloc}} = \frac{24}{5}\frac{\Omega\Lambda L^4 R^3 \bar{C}_{11}^2}{G^3 b^2}|\bar{\sigma}|, \tag{9}$$

where G is the shear modulus of the material and b is the Burgers vector.

We assume that the dislocations in the network are generally straight in the absence of precipitates as the result of the dislocation line tension. The growth of precipitates causes the dislocations to bow under the action of local precipitate stresses. The radial stress in the matrix from a spherical precipitate is given by[11]

$$\bar{\sigma}_{rr} = -\frac{4G\epsilon r_1^3}{r^3} = -\frac{4(1+\nu)G\delta r_1^3}{3(1-\nu)}\frac{1}{r^3}, \tag{10}$$

where the last equality follows from Eq. (5). For ease of calculation, it is reasonable to assume that the bowed dislocation line crudely follows contoured paths radially around the interrupting precipitates,[2] as illustrated in Fig. 1. We approximate the stress experienced locally by a dislocation segment as that occurring roughly at the midpoint between two nearest-neighbor spherical precipitates. In regions where the dislocation does not bisect a line between two precipitates, the dislocation line tension tends to straighten the dislocation, forcing that segment of the dislocation slightly closer to its nearest precipitate. We assume that the magnitude of σ at the decreased radial distance from the nearest precipitate may be crudely approximated as equal to the magnitude of σ at the midpoint between two nearest-neighbor precipitates.

We assume that the nearest-neighbor precipitates are separated radially by the distance $(N+1)(2r_1)$ on center, where N is a nonnegative number. We have shown previously[1] that the factor $(N+1)$ is related to the volume fraction of precipitates as $(N+1) = f_p^{-1/3}$. Evaluating Eq. (10) at the midpoint $r = (N+1)r_1$ and accounting for contributions at the midpoint from each of the two precipitates involved, we obtain

$$\bar{\sigma} = 2\bar{\sigma}_{rr} = -\frac{8G(1+\nu)f_p\delta}{3(1-\nu)}. \tag{11}$$

Equations (9) and (11) reveal that the change in the nonlinearity parameter resulting from the interaction of the precipitate coherency stresses with matrix dislocations depends linearly on the dislocation density, volume fraction of precipitates and precipitate-matrix misfit parameters, and on the fourth power of the dislocation loop length.

We assume that the bowing of a given dislocation segment involves three precipitates such that the middle precipitate serves to bend the dislocation segment relative to the two end precipitates. That is, the three precipitates produce a ''three-point bending'' of the dislocation segment as illustrated in Fig. 1. For ease of calculation, we assume that the three precipitates are crudely collinear and consider the end precipitates to be ''pinning'' sites for the dislocation segment at a distance $2L = 4(N+1)r_1 = 4f_p^{-1/3}r_1$ apart. We note, as an aside, that for Al-4% Cu alloy the radius of the GP zones $r_1 \approx 2.1$ nm and $f_p \approx 0.003\%$. We calculate from these values that $L = 2f_p^{-1/3}r_1 \approx 135$ nm. This number is in very good agreement with the average spacing 100–110 nm measured between GP zones in Al-4% Cu alloy[7] and lends credibility to the expression for L. Substituting this expression for L together with Eq. (11) into Eq. (9), we write the fractional change in the acoustic nonlinearity parameter $(\Delta\beta^{\text{disloc}}/\beta_0)$ resulting from precipitate coherency stresses on the matrix dislocation network as

$$\frac{\Delta\beta^{\text{disloc}}}{\beta_0} \approx 205\frac{\Omega R^3 \Lambda r_1^4 \bar{C}_{11}^2(1+\nu)|\delta|}{\beta_0 G^2 b^2 f_p^{1/3}(1-\nu)}. \tag{12}$$

IV. COMPARISON TO EXPERIMENT AND CONCLUSION

Experimental measurements of the acoustic nonlinearity parameter as a function of heat treatment time during the artificial aging of AA2024 from the T4 to the T6 temper reveals the appearance of two distinct, rather sharp, peaks.[1] The first peak occurs at 72 min of heat treatment time and is associated with the precipitation and reversion of GP zones. The peak value of the nonlinearity parameter is roughly 11% larger than the adjacent minimum value. The second peak

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occurs after approximately 430 min of heat treatment and is associated with the formation of S' precipitates. The peak value for the S' precipitate is roughly 8% higher than the adjacent minimum value.

We now calculate the fractional change in the acoustic nonlinearity parameter $(\Delta\beta^{\mathrm{disloc}}/\beta_0)$ from Eq. (12) for GP zones and S' precipitates in artificially aged aluminum alloy 2024. We assume that the values $G=28.6$ GPa, $\bar{C}_{11}=109$ GPa $R=\Omega=0.33$, and $b=0.4$ nm, previously measured[12] for AA2024-T4, hold throughout the heat treating process to the T6 temper. A reasonable estimate of the dislocation density for the extruded rod specimens used in Ref. 1 is[2] $\Lambda\approx 8\times 10^{13}$ m^{-2}. We use the value $r_1\approx 2.1$ nm obtained from experimental measurements of Al-4% Cu alloys for the average radius r_1 of the GP zones in AA2024-T4. We assume the value $\delta_{\mathrm{GP}}\approx -8\%$ for the misfit parameter.[1] We note that the fractional change in the nonlinearity parameter is weakly dependent on the volume fraction of precipitates f_p, since f_p for a fixed value of r_1 appears in Eq. (12) as a cubic root. This implies that exact knowledge of f_p is less critical to the calculations than many of the other parameters in the equation and that order of magnitude estimates would likely suffice for a reasonable determination of $(\Delta\beta^{\mathrm{disloc}}/\beta_0)$. We thus assume, as above, that the presence of Mg in AA2024 promotes the formation of GP zones that are roughly two orders of magnitude larger than that in the Al-4% Cu binary alloy and write $(f_p)_{\mathrm{GP}}\approx 0.3\%$ for AA2024-T4. We thus calculate from Eq. (12) that the fractional change in the acoustic nonlinearity parameter from the GP zones $(\Delta\beta^{\mathrm{disloc}}/\beta_0)_{\mathrm{GP}}\approx 9.2\%$.

A reasonable estimate[1] of the misfit parameter for S' precipitates is $\delta_{S'}\approx -1.2\%$ and $(f_p)_{S'}\approx 7\%$ for the volume fraction. The precipitate radius can be estimated from the coherency length $l_c=b/|\delta|\approx 33$ nm by noting that this length corresponds to the circumference of the great circle circumscribing a sphere of radius $r_1=l_c/2\pi\approx 5.3$ nm. This value of r_1 corresponds then to the radius at which precipitate coherency is lost and thus the radius at maximum material strength. From the above values, we calculate from Eq. (12) that the fractional change in the acoustic nonlinearity parameter from the S' precipitates $(\Delta\beta^{\mathrm{disloc}}/\beta_0)_{S'}\approx 8.4\%$.

The calculated values $(\Delta\beta^{\mathrm{disloc}}/\beta_0)_{\mathrm{GP}}\approx 9.2\%$ and $(\Delta\beta^{\mathrm{disloc}}/\beta_0)_{S'}\approx 8.4\%$ are in good agreement with the corresponding values 11% and 8% measured experimentally for the fractional changes in the acoustic nonlinearity parameters. The present model emphasizes the importance on the matrix dislocation network of local stresses resulting from the precipitate-matrix coherency strains. We point out that the present model does not account for differences in the elastic constants of the precipitates and matrix, nor does it account for variations in the elastic constants of the matrix resulting from the change in matrix composition due to precipitation. Although these factors may contribute to changes in the nonlinearity parameters, the exact contribution is difficult to assess because of the lack of available elastic constant data pertaining to these effects. The agreement between present model calculations and experimental measurements suggests that the effect of precipitate-matrix misfit strains on the matrix dislocation network is likely the dominate mechanism responsible for the change in the nonlinearity parameter during precipitation aging of AA2024.

Finally, we point out that the basic conclusions given in Ref. 1 remain valid within the context of the present model. The present model primarily serves to shift the emphasis of the strain mechanism responsible for observing changes in the acoustic nonlinearity parameter from an averaged strain in the matrix to a local strain (stress) at a given dislocation site in the matrix.

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