01 Jan 1983

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Recommended Citation

M. Wuttig and L. Chun-Hung, "Twinning Pseudoelasticity In InTl," *Acta Metallurgica*, vol. 31, no. 7, pp. 1117 - 1122, Elsevier, Jan 1983.
The definitive version is available at https://doi.org/10.1016/0001-6160(83)90207-9

Acta metall. Vol. 31, No. 7, pp. 1117–1122, 1983
Printed in Great Britain.

0001-6160/83 $03.00+ .00

TWINNING PSEUDOELASTICITY IN In–Tl

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(*Received* 8 *November* 1982)

Abstract—Twin boundaries generally contribute significantly to the damping of a twinned crystal. In an In–19.1 at.% Tl single crystal the damping gives rise to automodulation if the internal friction studies are performed in the 10 Hz frequency range, with a strain amplitude larger than 0.05 and at temperatures lower than 10°C. The frequency of the automodulation is about 60 times slower than the frequency with which the reed type sample is driven. The automodulation frequency has a maximum at −6 C and is characterized by an activation energy of 29 kJ/mol (7 kcal/mol). This maximum can be understood by applying Lücke and Stüwe, and Cahn's theory of impurity diffusion controlled grain boundary motion to the motion of twin boundaries. On the basis of this analysis it is suggested that the twinning pseudoelasticity in In–Tl comes about because at low temperatures the Tl atoms in the vicinity of the core of the twin boundary cannot relax to their equilibrium configuration.

Résumé—Les joints de macle contribuent généralement notablement a l'amortissement dans un cristal maclé. Dans un monocristal d' In–19,1 at.% Tl, l'amortissement produit une automodulation si l'on effectue les études frottement intérieur pour des fréquences de l'ordre de 10 Hz, avec une amplitude de la déformation supérieure à 0,05 et pour des températures inférieures á 10°C. La frèquence de l'automodulation est environ 60 plus lente que la fréquence appliquée à l'échantillon. La fréquence d'automodulation passe par un maximum à −6°C et elle est catactérisée par une énergie d'activation de 29 kJ/mol (7 kcal/mol). On peut comprendre ce maximum en appliquant au mouvement des joints de macle la théorie de Lücke, Stüwe et Cahn du mouvement des joints de grains contrôlé par la diffusion d'une impureté. A partir de cette analyse, nous pensons que la pseudoélasticité de maclage dans In–Tl vient du fait qu'à basse température les atomes de Tl voisins du coeur du joint de macle ne peuvent pas se relaxer vers leur position d'équilibre.

Zusammenfassung—Zwillingsgrenzen tragen im allgemeinen beträchtlich zur Dämpfung in verzwillingten Kristallen bein. In einem Einkristall In–19,1 At.-% Tl führt diese Dämpfung zu einer Selbstmodulation wenn die Versuche zur inneren Reibung im Bereich von 10 Hz mit Dehnungsamplituden über 0,05 bei Temperaturen unter 10°C durchgeführt werden. Die Frequenz dieser Selbstmodulation ist etwa 60-mal niedriger als die Frequenz, die an der blattförmigen Probe anliegt. Sie hat ein Maximum bei −6°C und ist charakterisiert durch eine Aktivierungsenergie von 29 kJ/Mol (7 kcal/Mol). Dieses Maximum Läßt sich verstehen, wenn die Theorie der Verunreinigungs-kontrollierten Korngrenzenbewegung von Lücke und Stüwe und von Cahn auf Zwillingsgrenzen angewendet wird. Eine solche Analyse legt nahe, daß die Pseudoelastizität durch Zwillinge in In–Tl davon herrührt, daß bei niedrigen Temperaturen die Tl-Atome in der Nähe der Zwillingskorngrenze nicht in ihre Gleichgewichtskonfiguration erreichen können.

1. INTRODUCTION

The deformation of metals and alloys by the motion of interfaces is often reversible. Hence, it is referred as pseudoelasticity. Two categories of pseudoelasticity are distinguished, pseudoelasticity associated with the martensitic transformation and twinning pseudoelasticity (T.P.). Both have been recently reviewed [1]. Various causes for the T.P. have been suggested, such as a stabilization process in Au–Cd [2], elastic interaction in Cu–Al–Ni [3], disordering in Fe_3Be [4] and others [5, 6]. The cause for T.P. in In–Tl alloys has not yet been identified [7], however. In this paper the results of an internal friction study of the T.P. in an In–19.1 at.% Tl alloy will be reported. The results lend support to the idea that the T.P. is caused by the redistribution of Tl atoms [8].

The technique of internal friction is well suited to study metallurgical processes on an atomic scale [9]. Internal friction studies of T.P. have therefore been performed [10–12] including some on In–Tl alloys [13, 14]. All these measurements were made at amplitudes of oscillation of the order of 10^{-5}. If the T.P. in In–Tl is due to diffusion controlled redistribution it is of interest to perform large amplitude internal friction studies as the motion of the twin boundary must change as it is pulled away from the region in which the redistribution occurs. This breakaway of the twin boundary from this region has been observed and will be reported and discussed below.

2. EXPERIMENTAL

Alloys were prepared from 99.99% pure In and Tl. Amounts of the elements of the desired weight ratio were inductively melted under a protective argon atmosphere in a graphite crucible. The melt was well agitated to insure a uniform composition. Cast rods were sealed under vacuum in a glass tube for subsequent growth of single crystals by the Bridgman

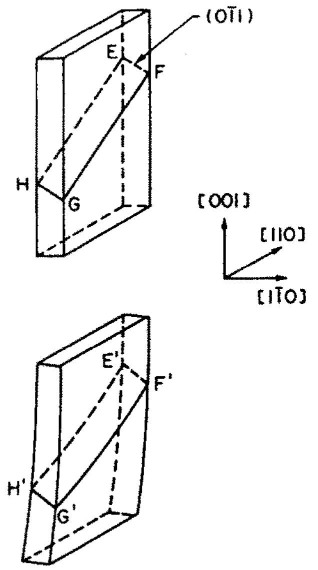

Fig. 1. Geometry before bending and after bending of the sample used in this investigation.

method. The single crystals were homogenized at a temperature of 140°C for one week and afterwards carefully removed using concentrated HF. The one used for this study contained 19.1 at.% Tl as determined by chemical analysis. The composition was uniform to within ±0.1%. The crystallographic orientation of the reed used for the present measurements is shown in Fig. 1.

The internal friction apparatus is shown schematically in Fig. 2. With it resonance curves could be recorded on an X-Y recorder. Furthermore the amplitude of vibration and phase could be monitored as a function of time, as described previously [15]. The temperature of the reed could be controlled to within ±0.1°C. Its resonance frequency could be varied by using different combinations of Sm–Co magnets and small ballast disks made of soft iron.

3. RESULTS

The key result of the study is that automodulations occur at temperatures below about 10°C if the reeds are driven in the 10 Hz frequency range to amplitudes of vibration in excess of a nominal surface strain of $x/l \approx 0.05$ (see Fig. 2 for the definition of the quantities x and l). This fact, together with some details of the automodulation, is shown in Figs 3–5.

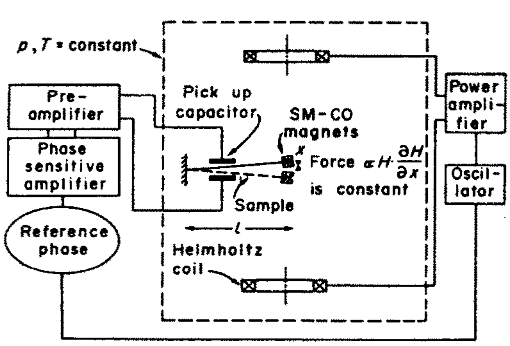

Fig. 2. Schematic of the internal friction apparatus.

Figure 3 illustrates that the automodulations set in at a critical amplitude of vibration of about 10 mV which is equivalent to $x/l \approx 0.05$. In this figure the automodulation manifests itself as an apparent noise which is present at large amplitudes of vibration only. It should be noted that the automodulations occur even though the resonance curve was scanned with a constant driving force.

The apparent noise in Fig. 4 is really a well developed periodic oscillation, as is evident from Figs 4 and 5. In Fig. 5 automodulations are presented which were observed at various points of the resonance curve shown in Fig. 4. It can be seen that the frequency and amplitude of the automodulation vary as the drive frequency is changed. The modulation frequency is largest at drive frequencies roughly halfway between the threshold and peak frequencies. The automodulation of the amplitude of vibration

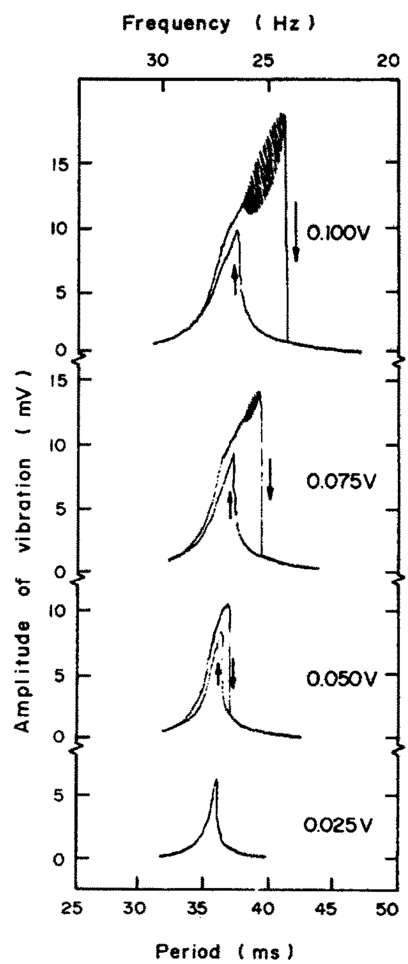

Fig. 3. Resonance curves of a reed made in In–19.1 at.% Tl single crystal oriented as shown in Fig. 1 at different driving forces (indicated in volts) measured at −8.0°C.

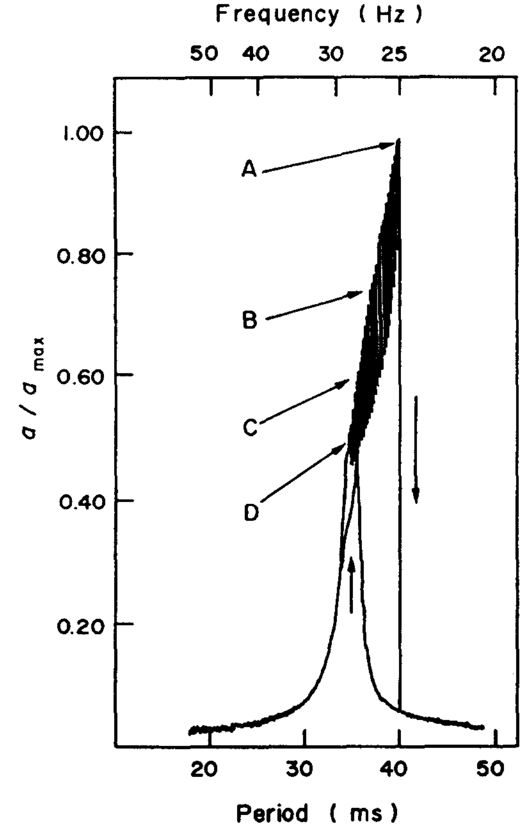

Fig. 4. The resonance curve of the In–19.1 at.% Tl reed taken at −15.7°C. The automodulations recorded at various drive frequencies are specified as A, B, C, D, and shown in Fig. 5.

varies from zero to 35%. The associated phase automodulation has been observed as well.

The variation of the frequency of the automodulation with temperature is very pronounced and shown in Fig. 6. For consistency the maximum automodulation frequency observed at each temperature has been plotted. It can be seen that the automodulation frequency of the In–Tl alloy displays a maximum at −6°C. The high and low temperature portions of the automodulation frequency can be fitted by an Arrhenius equation with an activation energy of 29 kJ/mol (7 kcal/mol). The significance of this result will be discussed below.

4. DISCUSSION

In this discussion it will be shown that the automodulation arises in the course of large amplitude oscillations if the deformation is predominantly due to twinning pseudoelasticity. The discussion will be couched in terms of the well known theory of impurity controlled grain boundary motion as applied to twin boundary motion. In essence, it will be proposed that the automodulation is the result of a periodic breakaway of the twin boundary from its self-generated potential well. The analysis based on this proposal will lead to a straight forward interpretation of the observed maximum of the automodulation frequency.

The main prediction of the continuum theory of impurity controlled grain boundary motion is contained in Fig. 7. In this figure, the force f required to move the grain boundary with the steady state velocity v, as given by Cahn [16] and Lücke and Stüwe [17]

$$f = \lambda v + \alpha c v/(1 + \beta^2 v^2) \qquad (1)$$

is displayed in a normalised fashion. In equation (1) the parameter λ signifies the background damping. The quantity α denotes the low velocity damping constant of a grain boundary in a metal containing an impurity of concentration c. The quantity β characterizes the damping at high velocities. In the context of this study it is of importance to note that the temperature dependence of α and β is predominantly controlled by the diffusivity D of the species which controls the boundary motion, $\alpha \propto D^{-1}$, $\beta \propto D^{-1/2}$ [16].

It can be seen from Fig. 7 that the boundary can move with two distinct velocities if $\epsilon > 8$. Lower velocities, $\dot{\eta} < \dot{\eta}_1$, are controlled by the diffusional characteristics of the boundary represented by the parameter $(\lambda + \alpha c)$, "loaded case" [16] whereas higher velocities, $\dot{\eta} > \dot{\eta}_4$, are controlled by the intrinsic boundary damping λ only, "unloaded case". In the region $\dot{\eta}_1 \leq \dot{\eta} \leq \dot{\eta}_4$ the velocity is multivalued and

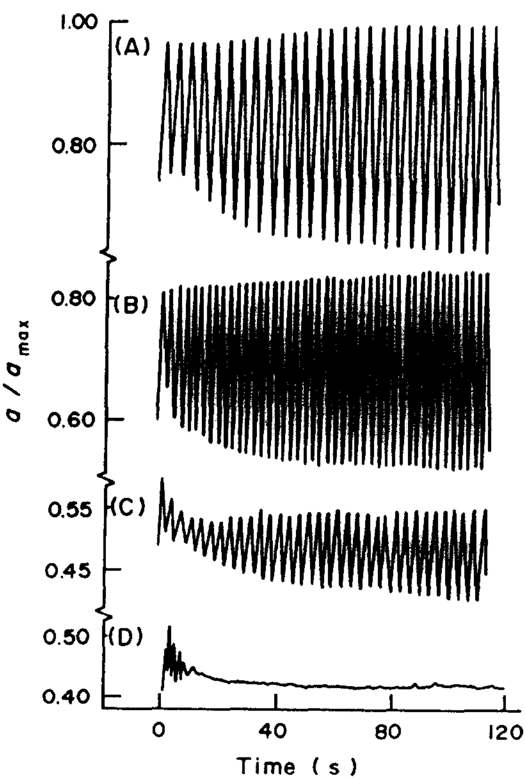

Fig. 5. The automodulation recorded at various drive frequencies as specified in Fig. 4.

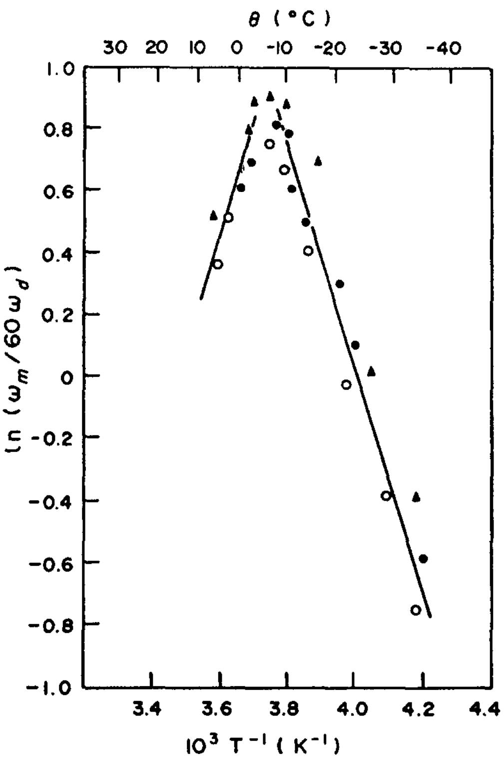

Fig. 6. Variation of the frequency of automodulation with temperature of the In–19.1 at.% Tl reed. The various sets of data points (●, ○, ▲) refer to different sets of measurements taken at different resonance frequencies.

a transition between the two velocity regimes can occur. This transition can give rise to automodulations [18, 19]. One cycle of this periodic motion is indicated schematically by arrows in Fig. 7. It should be noted that this cycle can only occur if the velocity exceeds the critical value, $\dot{\eta}_2$, i.e. at a critical amplitude of oscillation. This is exactly what is observed experimentally (see Fig. 3).

The drag force f can be used to construct an approximate equation of motion of a reed whose large amplitude oscillations are predominantly controlled by the motion of twin boundaries. This fact has been verified by visual observations of the reed oscillations by means of a light-microscope whose light source was strobed synchroneously with the oscillation. The equation of motion must therefore have the form

$$M\ddot{y} + [\lambda + \alpha/(1 + \beta^2\dot{y}^2)]\dot{y} + ky = E\sin(\omega_d t). \quad (2)$$

In this equation, M is the effective mass of the reed. The damping parameters λ and α now denote the total damping due to the twin boundary motion, with and without diffusional adjustment, respectively. The quantity β^{-1} is the characteristic velocity of the twin boundary. The force constant k designates the total pseudoelastic force constant of all twin boundaries. It will be commented on more fully below. The right hand side of equation (2) represented the externally applied force. In normalized form equation (2) reads

$$\ddot{\eta} + \lambda\omega^{-1}M^{-1}[1 + \epsilon/(1 + \delta^2\omega^2\beta^2\dot{\eta}^2)]\dot{\eta} + \eta = E'\sin(\omega_d t) \quad (3)$$

where the differentiation is performed with respect to the normalized time $\tau = \omega t$, $\eta = y/\delta$ is the normalized displacement and the constants ϵ, ω^2 and E' are given by $\epsilon = \alpha/\lambda$, $\omega^2 = kM^{-1}$ and $E' = E/(M\delta\omega^2)$. The quantity δ represents the half width of the stabilization potential of the twin boundary [16].

Equation (2) is highly nonlinear because of the nonlinear damping term as well as the nonlinearity necessarily associated with the force constant k. The damping term is strictly correct only for steady state boundary motion and thus cannot be expected to hold exactly under all experimental conditions. Its analytical form does describe the breakaway, however, and is therefore suitable for a semiquantitative analysis of the automodulation. The pseudoelastic force constant has the following features. If the velocity of the boundary v is large compared to the diffusional speed D/x (x denotes the boundary displacement) it is essentially given by the spatial derivative of the impurity distribution at the position of the boundary. In the opposite case, $v \ll D/x$, the constant k equals zero, i.e. the reed stays plastically deformed at high temperatures as is known. In an intermediate range, $v \approx D/x$, k will be time dependent. In the following analysis only the two limiting cases will be considered.

For a given frequency of oscillation the two limiting cases $v \ll D/x$ and $v \gg D/x$ characterize the high and low temperature regimes of the present experi-

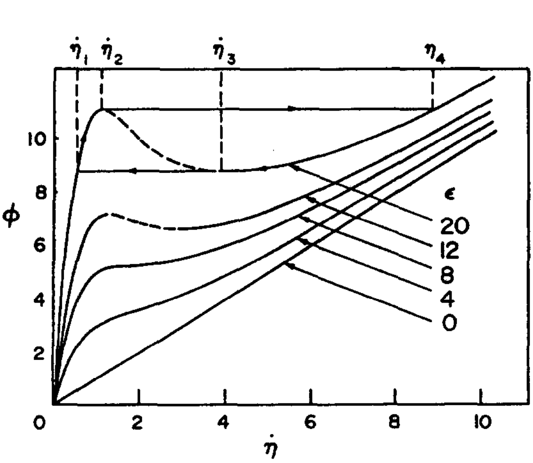

Fig. 7. The normalized drag force $\phi = \beta f/\lambda$ as a function of the normalized velocity $\dot{\eta} = \beta v$ for different values of the parameter $\epsilon = \alpha c/\lambda$ according to equation (1). The dashed line gives the unstable part of the curve and is of interest in this study. The arrows connecting $\dot{\eta}_2$ and $\dot{\eta}_4$ as well as $\dot{\eta}_3$ and $\dot{\eta}_1$ indicate the transitions from the "loaded" to the "unloaded" grain boundary configuration and the reverse. One cycle of the automodulation is indicated schematically by all four arrows.

ment. In the high temperature regime the force constant k can be neglected and the equation to be analyzed is

$$\ddot{\eta} + a[1 + \epsilon/(1 + b^2\dot{\eta}^2)]\dot{\eta} = 0 \tag{4}$$

where $a = \lambda\omega^{-1}M^{-1}$ and $b = \delta\omega\beta$. In this equation the external force has been omitted. It will be implicity taken into account by considering only values of $\epsilon > 8$, i.e. large amplitudes of oscillation. With the substitution $\zeta = \dot{\eta}$ equation (4) can be rewritten as

$$\mathrm{d}\tau = -a(1 + b^2\zeta^2)/(1 + \epsilon + b^2\zeta^2)\,\mathrm{d}\zeta \tag{5}$$

and integrated. The result is an estimate for the period of the automodulation.

$$\tau_m = 2(1+\epsilon)^{-1}\ln[(1+\epsilon+b^2\zeta^2)/\zeta^2] - 2\ln[1+\epsilon+b^2\zeta^2]\Big|_{\zeta_{max}}^{\zeta_{min}}, \tag{6}$$

where $\zeta_{min} = \dot{\eta}_2$ and $\zeta_{max} = \dot{\eta}_2 + \Delta\dot{\eta}$, and where $0 < \Delta\dot{\eta} < \dot{\eta}_4 - \dot{\eta}_2$ (see Fig. 7). The temperature dependence of τ_m is given by the quantity ϵ. If the temperature dependence of the slowly varying logarithmic term is neglected expression (6) leads to

$$\tau_m \approx k_1(1+\epsilon)^{-1} + k_2 \tag{7}$$

where k_1 and k_2 are constants and $|k_2| > |k_1|$. Automodulation can only occur if $\epsilon > 8$ (see Fig. 7), thus $\epsilon + 1 \approx \epsilon \gtrsim 10$. It follows that in the high temperature regime the frequency of the automodulation $\omega_m = \tau_m^{-1}$ is proportional to ϵ and, since $\epsilon = \alpha/\lambda$ and $\alpha \propto D^{-1}$,

$$\omega_m \propto D^{-1}. \tag{8}$$

The frequency of the automodulation in the low temperature regime may be analyzed by observing that now the pseudoelastic constant exerts a predominant influence. Proceeding as was done for the high temperature regime this leads to the analysis of spontaneous oscillation of the equation of motion

$$a[1 + \epsilon/(1 + b^2\dot{\eta}^2)]\dot{\eta} + \eta = 0. \tag{9}$$

Dimensionally, the period of the automodulation is given by [20]

$$\tau_m = \int \mathrm{d}\eta/\dot{\eta} \tag{10}$$

which, in conjunction with equation (9) leads to

$$\tau_m \approx 4a \cdot \ln(\dot{\eta}_{max}, \dot{\eta}_2) + \epsilon f(\dot{\eta}_{max}, \dot{\eta}_2). \tag{11}$$

It can be seen from this equation that in the low temperature regime $\tau_m \propto \epsilon$ and therefore

$$\omega_m \propto D. \tag{12}$$

The observed temperature dependence of the automodulation shown in Fig. 6 can now be readily interpreted. Since at high temperatures $\omega_m \propto D^{-1}$ and at low temperatures $\omega_m \propto D$ a maximum of the function $\omega_m = f(T)$ must exist at an intermediate temperature as is actually observed at around -6°C. Furthermore in the limiting temperature regions an Arrhenius plot $\ln \omega_m = f(Q/RT)$ should be a straight line with equal activation energies Q but of opposite slope. This is observed as well. A least square analysis yields a value of $Q = 29$ kJ/mol (7 kcal/mol).

The idea that the automodulation represents a periodic breakaway of the twin boundary from its equilibrium position leads to a semiquantitative understanding of the temperature dependence of the automodulation frequency and yields an activation energy of 29 kJ/mol (7 kcal/mol). This value is based on a number of assumptions; some of which have already been stated. In addition, the simple triangular shape of the potential well of the boundary [16] and the spatial independence of the diffusivity leading to expression (1) are also embodied in the estimates (8) and (12). The significance of the value of the activation energy, 29 kJ/mol (7 kcal/mol), must, therefore, be carefully assessed. Numerically, it may be compared to the activation energy of twin boundary stabilization, 49 kJ/mol (12 kcal/mol) [21], the activation energy for tracer diffusion of Tl in In, 65 kJ/mol (15 kcal/mol) [22] and the activation energy derived from small amplitude internal friction measurements, 67 kJ/mol (16 kcal/mol) [23]. It is interesting to observe that the activation energy of the automodulation equals about half the activation energy of Tl diffusion. This might support the suggestion that the pseudoelasticity due to twin boundary motion in In–Tl is caused by the diffusion controlled rearrangement of Tl atoms. The ratio of about 1/2 then indicates that the rearrangement occurs at or near the core of the twin boundary. At first sight this interpretation seems to be inconsistent with the assumption of the spatial independent of the diffusivity. However, if the diffusivity is a sharply peaked function most of the damping results from the core region. In this case the spatial dependence of the diffusivity $D(x)$ can be approximated by a delta function, $D(x) = D\delta(x)$, leading again to $\alpha \propto D^{-1}$ [16] which was used to interpret the Arrhenius plot of the automodulation frequency. The rearrangement of Tl atoms, or, rather, the lack of it in the core of the twin boundaries is therefore a likely cause for the twin boundary pseudoelasticity in In–Tl, in agreement with an earlier suggestion [8].

In summary, the observation of automodulations associated with twinning pseudoelasticity lends qualitative support to the concepts used in the theory of impurity controlled grain boundary motion [16, 17]. In conjunction with this theory it yields much insight into the dynamics of twin boundary motion, how they break away from and the possible nature of their "impurity atmosphere". The temperature dependence of the automodulation suggests that a diffusion controlled rearrangement of Tl atoms occurs in the core of twin boundaries. This, in turn, implies that the distribution of the alloying elements depends on the state of stress, which is not unexpected.

Acknowledgement—The support of the National Science Foundation is gratefully acknowledged.

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