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January 1987

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Prepared for the U.S. Department of Energy
under Contract DE-AC03-76SF00098

Center for Advanced Materials

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Strengthening Contributions of Strong Ordered Precipitates

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Abstract

The Hanson and Morris solution for the critical resolved shear stress of a random array of point obstacles is modified to consider the case of strong ordered precipitates. The solution approximately accounts for the effects of dislocation self-interactions and elastic anisotropy. The modified solution leads to a simple relationship between the antiphase boundary energy and the minimum looped precipitate diameter. This relationship is used to predict antiphase boundary energies for Al_3Li and Ni_3Al of 57 ± 15 mJ/m^2 and 102 ± 35 mJ/m^2. These values are in good agreement with recent determinations of the antiphase boundary energy by other techniques.

I. Introduction

The contribution of ordered precipitates to the critical resolved shear stress of precipitation-hardened alloys has been extensively studied (Ardell, 1985; Glazer, 1986). However, most analyses use theories premised on the assumption that the strengthening obstacles are relatively weak. This paper will develop a theory of order hardening by strong precipitates in the context of the random array theory of dislocation glide. Two examples of such precipitates in systems of technological interest are the $L1_2$ precipitates Ni_3Al (γ'), present in Fe- and Ni-based superalloys, and Al_3Li (δ'), found in aluminum-lithium alloys. These precipitates are sheared by dislocations when they are small and are bypassed by Orowan looping at larger sizes. The focus of this paper is the relationship between the minimum radius at which the precipitates are looped and the energy of the antiphase boundary created when they are sheared.

A theory for the critical resolved shear stress of a material hardened by shearable precipitates must include an expression for two parameters that describe the force required to bypass a precipitate: the force required to shear a precipitate as a function of its size (F_s), and the force required for Orowan looping (F_{loop}), the effective upper limit to the force with which a precipitate can resist the passage of a dislocation. These parameters will be discussed in turn below.

The force required to shear spherical (or cuboidal) ordered precipitates is generally measured either in terms of the energy of the antiphase boundary (γ) created when the precipitate is sheared at its widest point or by the precipitate diameter (d_{loop}) at which Orowan looping first becomes preferable. It can be shown easily that the shear force is given by

$$F_s = \gamma d = F_{loop}(d / d_{loop}) \quad (1)$$

where d is the precipitate diameter. The looping diameter is a convenient obstacle strength parameter since it can be determined unambiguously from transmission electron micrographs of deformed material. The antiphase boundary energy can be determined from phase diagram information (e.g. Khachaturyan and Morris, 1986) or from measurements of dislocation pair spacings (e.g. Huang and Ardell, 1986). In either case, the antiphase boundary energy is determined indirectly and its usefulness depends on the accuracy of the theory used to calculate it from the experimental evidence. Perhaps not surprisingly, the values of the antiphase boundary energy for both Ni_3Al and Al_3Li have been in debate.

Orowan looping occurs when the size of the precipitate exceeds a critical value, the looping diameter d_{loop}. At this size, the force required to shear the precipitate is greater than the force required to loop it. The value of the critical resolved shear stress at the looping diameter is a function of the ratio of the applied force to the dislocation line tension. In a simple model that neglects any dislocation self-interactions, the precipitates are looped when the applied force is equal to twice the line tension. However, it has been known for some time that the attraction between the arms of a dislocation as it bows about a particle of finite size significantly lowers the applied force required for looping (Bacon, Kocks and Scattergood, 1973). Although this decrease is often neglected, it has a correspondingly large effect on the critical resolved shear stress whether the theory is phrased in terms of γ or in terms of d_{loop}.

This paper develops a model for the critical resolved shear stress of alloys hardened by ordered precipitates in the context of the random array model for dislocation glide developed by Hanson and Morris (1975a and b). The strength of the precipitates is written in terms of the average diameter of the precipitates and the looping diameter. The effect of dislocation self-interactions is approximately included. Finally, the antiphase boundary energies *predicted* by the theory are compared with the best available values determined by other methods.

II. Critical resolved shear stress for a random array of strong obstacles

II.1 The Hanson and Morris random array theory

Most investigations of strengthening by discrete obstacles have developed the consequences of a single model of dislocation glide (Ardell, 1985; Glazer, 1986). The most important assumptions of this model are as follows. The dislocation glides in an idealized single crystal microstructure characterized by the mean square obstacle spacing l_s of a random distribution of point obstacles. The obstacles are mathematical points whose interaction with the mathematical dislocation is the same as the interaction between the physical obstacle and dislocation. The strength of the obstacle in athermal glide is given by the peak in the force-distance curve for the physical dislocation-precipitate interaction. In the absence of dislocation self-interactions, the force exerted by a dislocation on an

obstacle is defined as $F = 2T \cos(\Psi/2)$ where T is the dislocation line tension and Ψ is the bow-out angle defined in Fig. 1. The strength of the obstacle is given in dimensionless form by $\beta = F/2T = \cos(\Psi/2)$ where F is now the force required to bypass the obstacle either by shearing or looping. The maximum effective particle strength, achieved when the particle is looped under the applied force F_{loop}, is β_c. The value of the line tension depends on whether the dislocation is edge or screw in character; in discussions of real materials, in which both types of dislocations contribute to deformation, the line tension of the dislocation type that appears to control the process is used.

In order to calculate the critical resolved shear stress, most models assume isotropic elasticity and a Poisson's ratio of zero. The line tension is also assumed to be constant along the length of the bowing dislocation. Essentially all of the analytic and computer simulation solutions to this model lead to an equation for the critical resolved shear stress τ in terms of the dimensionless value

$$\tau^* = \tau l_s b/2T \tag{2}$$

where b is the Burgers vector in the glide plane. The analytic solution for the critical resolved shear stress is given by

$$\tau^* = Q\beta^{3/2}. \tag{3}$$

The various solutions account differently for the statistical randomness of the array, resulting in values of Q between 0.7 and 1.0.

Hanson and Morris (1975a) consider explicitly how the statistics of dislocation-obstacle interactions along the dislocation line affect the critical resolved shear stress. The distinguishing feature of the solution is that the dislocation is described in terms of its configuration, the unique set of pinning points along its length. The unit interaction is with the configuration rather than with isolated obstacles. In this representation, the critical resolved shear stress for an entire array is defined simply as the stress required to bypass the weakest point in the strongest line, or configuration, within the array. This solution has been used for both analytic and computer simulations of strengthening behavior in real materials (Melander and Persson, 1978a and b; Altintas and Morris, 1986a and b; Glazer, Edgecumbe and Morris, 1985). The Hanson and Morris solution for an infinite array leads to equation (3) above with a value of Q of approximately 0.9 for values of β less than 0.7. The solution also relates the critical resolved shear stress for a mixture of obstacles to that for identical obstacles. The effect of a mixture of obstacle strengths is important in real materials and will be discussed in a later paper.

II.2 The Effect of Dislocation Self-Interactions

The term dislocation self-interaction is used here to describe the elastic interaction between different segments of a dislocation that is bowed about an obstacle so that the elastic strain fields of the segments overlap. If the arms of the dislocation bow toward one another, the interaction lowers the total line energy of the dislocation. This self-interaction

force is additive to the applied shear force and helps the dislocation to bypass the obstacle. The effect can be easily seen for the case of Orowan looping, which occurs when the arms of the dislocation are antiparallel. The force required to reach this condition if the dislocation bows in a circular arc is considerably greater than the force needed if the arc is distorted elliptically by the attraction between the bowing arms. Conversely, if the elliptically bowed arc that defines the actual looping criterion is approximated by the circular arc assumed in most solutions for the critical resolved shear stress, then looping occurs before the arms of the dislocation are antiparallel. In the terms of the Hanson and Morris model, this effect can be described by a value of β_c less than one.

Bacon, et al. (1973) have quantified the effect of dislocation self-interactions on the stress for Orowan looping. They model their results for impenetrable obstacles by treating them as penetrable obstacles in the constant line tension approximation. The model is most useful when the ratio of particle diameter to particle spacing is small. Bacon, et al. find an expression for the critical cusp angle for Orowan looping

$$\beta_c = \cos(\Psi/2) = (\ln D + 0.7) / \ln l \tag{4}$$

where $D = [d^{-1} + l^{-1}]^{-1}$,

and d and l are the average particle diameter and spacing, respectively, expressed in units of the inner cutoff radius of the dislocation, which is on the order of the Burgers vector. Since point obstacles are in many ways the geometric equivalents of fully penetrable obstacles, Bacon et al. use this result to correct β_c in some early solutions for the critical resolved shear stress of a random array of point obstacles.

The same technique can be applied to modify the Hanson and Morris solution for dislocation self-interactions. Equating l to the mean square obstacle spacing in these units, l_s/b, in equations (2) and (4) and comparing the resulting equation for the critical resolved shear stress to equation (3) leads to an expression for the line tension T

$$T = (Gb^2 / 4\pi K) \ln (l_s / b). \tag{5}$$

where K is 1 for edge dislocations and (1 - ν) for screw dislocations. It should be noted that this equation for the line tension differs from the standard form derived by deWit and Koehler (1959). The identification of variables within the logarithmic term is somewhat arbitrary. Since the line tension is not very sensitive to the exact values, the simpest possible choices have been made. When the obstacles are small and well-spaced, β_c can be approximated by (ln d/ln l). The values of β for precipitates that are still small enough to be shearable scale with β_c. The remaining difficulty is assigning a value to β_c. For impenetrable obstacles, Bacon et al. (1973) found that the value of β_c increases as the ratio d/l increases because the increased separation between the arms of the dislocation lessens the strength of their interaction. However, for penetrable obstacles such as the ordered precipitates in question here, the distance separating the arms will be considerably less than the diameter of the precipitate. An exact solution to this problem does not exist, but Hanson and Morris suggest that $\beta_c = 0.7$, which implies $d << l_s$, is a reasonable choice.

For ordered spherical precipitates, these considerations lead to a relatively simple formula for the critical resolved shear stress. By analogy to equation (1), the precipitate strengths can be defined in terms of their diameters as

$$\beta_c = c d_{loop}/2T_{loop}$$
$$\beta = \beta_c(d / d_{loop})(T_{loop}/T) \tag{6}$$

where T_{loop} refers to the sample for which d_{loop} was measured and c is a constant. For a given material, the ratio of the line tensions reduces to a ratio of ln (l_s/b). The critical resolved shear stress can be written using equation (3) as

$$\tau^* = 0.9[\beta_c(d / d_{loop})(T_{loop}/T)]^{3/2} \tag{7}$$

where τ^* is defined in equation 4, T in equation 5 and β_c is 0.7. This equation is precise if all the precipitate cross-sections in the glide plane have diameter d. In any real material, there will be a distribution of precipitate sizes and a distribution of cross-sectional diameters for each precipitate. The effect of this distribution is discussed elsewhere (Munjal and Ardell, 1976; Glazer and Morris, 1986; Glazer, 1986).

Because the precipitates are ordered, matrix dislocations are coupled as superdislocations that are total dislocations in the precipitate crystal structure. For the $L1_2$ structure, the dislocations are paired. Force balances for the ordered precipitate situation are given in the reviews by Brown and Ham (1971) and Ardell (1985). A simple pileup of two uncoupled dislocations doubles the applied stress on the obstacles (Friedel, 1964). When ordered precipitates are present, the antiphase boundary area between the two dislocations further increases the stress on the lead dislocation by an amount that depends on the configuration of the second dislocation as well as the first. The statistics of this interaction have not been studied. However, the strong line assumption predicts that the case in which the applied stress is least magnified (i.e. by a factor of two), not the average situation, controls the critical resolved shear stress. Consequently, τ^* (equation 7) should be divided by two to account for the effect of the superdislocations pairs on the critical resolved shear stress.

Equations (6) and (7) were derived under the assumption of isotropic elasticity. This assumption is good for aluminum, but it is quite poor for nickel or iron. Scattergood and Bacon (1975) showed that the effect of elastic anisotropy on the Orowan stress could be modeled with reasonable accuracy in the constant line tension approximation by simply replacing the average values of G and ν with the anisotropic values appropriate to the slip system. For face-centered cubic materials, these are the values for {111} planes. Transformation equations to calculate G_{111} and ν_{111} from the cubic elastic constants c_{11}, c_{12} and c_{44} are given by Ardell (1986).

III. Determination of antiphase boundary energies

III.1 Relationship of the antiphase boundary energy to the looping diameter

The minimum precipitate size at which Orowan looping occurs is directly related to the looping diameter. If self-interactions are neglected, the antiphase boundary energy is defined by the simple force balance

$$F_{loop} = 2T' = d_{loop}\gamma' \tag{8}$$

where T' is the deWit and Koehler line tension and γ' is the predicted value of the line tension when self-interactions are neglected. In the terms of the modified Hanson and Morris solution described in Section II.2, the force balance is

$$F_{loop} = 2T_{loop}\beta_c = d_{loop}\gamma \tag{9}$$

For screw dislocations these relations imply that

$$\gamma = [0.7 / (1 + \nu)]\gamma' \tag{10}$$

For a typical metal with a Poisson's ratio near one-third, equation (10) says that the antiphase boundary energy is overestimated by approximately a factor of two if self-interactions are neglected.

III.2 Sample calculations of the antiphase boundary energy

One measure of the validity of the theory for the critical resolved shear stress given above is whether the antiphase boundary energies predicted by equation (9) are reasonable. Calculations for Ni_3Al and Al_3Li are given below. Both of these coherent precipitates have sufficiently low misfit strains that it is reasonable to assume that most of the peak dislocation-precipitate interaction is due to the ordered structure of the precipitate.

Ni_3Al : Both Ni_3Al and $Ni_3(Al,Ti)$ precipitates have been studied in Ni, Fe-Ni-Cr and Co-Ni-Cr alloys. The minimum looped diameter of the precipitates is 15 ± 3 nm (Raynor and Silcock, 1970; Thompson and Brooks, 1982; Munjal and Ardell, 1976; Chaturvedi, Lloyd and Chung, 1976). The variation in chemical composition probably contributes to the size of this range. Where the looping diameter was not explicitly measured, it has been estimated to be 1.5 times the average precipitate diameter when the strength began to peak. Previous work on the effect of the precipitate size distribution on the critical resolved shear stress suggests that perhaps 10% of the precipitates exceed the looping diameter at this point (Glazer and Morris, 1986). Measurements of precipitate size distributions in aluminum-lithium alloys by Gu, Liedl, Kulwicki and Sanders (1985) and in Ni-Al alloys by Munjal and Ardell (1976) suggest that about 10% of the precipitates are greater than one and a half times the average diameter. This argument gives an approximate value for the looping diameter consistent with those that were explicitly stated. The value of l_s can be calculated using the volume fraction (generally about 0.1) and an

average precipitate diameter of about 35 nm. Thompson and Brooks (1982) give an average value of the shear modulus, 73.9 GPa. The Burgers vector varies slightly with composition, but is about 0.255 nm. Edge dislocations apparently control deformation (Raynor and Silcock, 1970; Chaturvedi, et al. 1976; Ardell, Munjal and Chellman, 1976).

Assuming elastic isotropy, the values above lead to a value for the antiphase boundary energy of 177 ± 40 mJ/m^2. However, these alloys are anisotropic and this value should be improved if proper values of G and ν are used. In their solution, Scattergood and Bacon assume that the elastic constants of the matrix and the precipitate are identical. However, it seems reasonable to assume that the exact solution for different elastic constants will be bracketed by the homogeneous solutions that assume the elastic constants of the precipitate and the matrix, respectively. Ledbetter (1984) gives the elastic constants for an Fe-Ni-Cr alloy similar to those studied by Raynor and Silcock (1970) and Thompson and Brooks (1982) at room temperature as $c_{11} = 2.07$, $c_{12} = 1.32$ and $c_{44} = 1.23$, all in units of 10^{11}Pa. Douin, Veyssiere and Beauchamp (1986) cite elastic constants for Ni_3Al at 650°C: $c_{11} = 1.781$, $c_{12} = 1.193$ and $c_{44} = 1.0232$, also in units of 10^{11}Pa. These values may be transformed to determine G_{111} and ν_{111}. For γ-Fe, these are 48 GPa and 0.362, which lead to a value for γ of 93 ± 25 mJ/m^2; for Ni_3Al, they are 38.6 GPa and 0.404, and the corresponding value of the antiphase boundary energy is 112 ± 30 mJ/m^2, where the uncertainty comes from the error in the looping diameter. Averaging these values leads to a final estimate for the antiphase boundary energy of 102 ± 35 mJ/m^2.

Douin, et al. (1986) have probably made the best direct measurements of γ_{111} in Ni_3Al polycrystals. These calculations are based on glide dissociation distances measured by weak beam transmission electron microscopy on isolated dislocations and include the effect of elastic anisotropy on the dislocation line energy. Their previous calculations that did not include the effects of anisotropy led to a value for the antiphase boundary energy of 180 ± 30 mJ/m^2 (Veyssiere, Douin and Beauchamp, 1985). The anisotropic calculations led to a revised value of 111 ± 15 mJ/m^2. These values are extremely close to the values determined here of 177 ± 40 mJ/m^2 (isotropic elastic constants) and 102 ± 35 mJ/m^2 (anisotropic elastic constants). While the extent of the agreement may be fortuitous, it does suggest that the antiphase boundary energy values predicted by analysis of the critical resolved shear stress given here are reasonable. It should be noted that numerous other values given in the literature calculated by a variety of methods are considerably higher than the anisotropic value of 111 mJ/m^2 ; these range from 135 mJ/m^2, which is reasonable, to 380 mJ/m^2, which is not (see for example, Ardell, 1980; Raynor and Silcock, 1970; Singhal, 1971).

Al_3Li in Al: The looping diameter of the Al_3Li precipitate in binary aluminum-lithium alloys has been measured by a number of investigators (de Hosson, Huis in't Veld, Tamler and Kanert, 1984; Sainfort and Guyot, 1985; Miura, Matsui, Furukawa and Nemoto, 1985; Furukawa, Miura and Nemoto, 1985). A survey of the data suggests that the looping diameter is 50 ± 10 nm. However, these data are measured in alloys of widely differing mean square obstacle spacings, so T_{loop} must vary considerably. Since aluminum is nearly isotropic, average values of G and ν of 30 GPa and 0.35, respectively, are reasonable. The Burger's vector is approximately 0.29 nm. In contrast to superalloys,

deformation in aluminum-lithium alloys appears to be controlled by screw dislocations (Miura et al., 1985; Huang and Ardell, 1986). The data are given in Table 1. Substituting these values into equations (5) and (9) lead to values for T and γ for each case. Although the looping diameters are quite different, each data set leads to an antiphase boundary energy of ~57 mJ/m^2. The uncertainty on this value is approximately ± 15 mJ/m^2.

Most other values of the antiphase boundary energy for Al_3Li have been calculated from dislocation pair spacings. These values range from 130 to 195 mJ/m^2 (Sainfort and Guyot, 1985; de Hosson et al., 1984; Tamura, Mori and Nakamura, 1970; Huang and Ardell, 1986). However, Khachaturyan and Morris (1986) have shown recently that the antiphase boundary energy can be calculated from thermodynamic information to good accuracy. Their calculations, which are based on second near neighbor interactions, predict an antiphase boundary energy of 77 mJ/m^2, which is close to the value determined here from the looping diameter. This calculation should provide an upper limit for the antiphase boundary energy since it assumes perfect order in the precipitate and an infinitely sharp interface. A number of other theoretical calculations based on the order-disorder temperature that lead to different values of the antiphase boundary energy are discussed by Khachaturyan and Morris (1986).

III.3 Discussion

There has been considerable controversy over the value of the antiphase boundary energy for both Ni_3Al and Al_3Li. The values determined here agree with two of the most recent calculations done by completely different techniques. This agreement lends credibility both to the antiphase boundary energies and to the assumptions of the basic theory. Since there are many other values in the literature, it is worth considering whether these values are physically reasonable.

It has been known for some time that that antiphase boundary energy scales with the order-disorder temperature and that the order-disorder temperature must be less than the melting points of the pure components of the ordered phase. Thus, it would be very surprising if the antiphase boundary energy of Ni_3Al did not significantly exceed the antiphase boundary energy for Al_3Li.

According to the analysis given in Section II, the numerical value of the antiphase boundary energy is strongly affected by the character of the dislocations that control deformation through the value of the line tension. In view of their common matrix and precipitate crystal structures, it seems surprising that deformation at room temperature is dominated by edge dislocations in superalloys, but by screw dislocation in aluminum-lithium alloys; nonetheless, the experimental evidence seems relatively clear. A simple analysis suggests that since the critical resolved shear stress is proportional to the interaction force and inversely proportional to the dislocation line tension, deformation of an order-hardened alloy should be controlled by edge dislocations because their line tension is lower (Glazer, 1986). The reason for the behavior observed in aluminum-lithium alloys is not known. If the assumption of screw control is incorrect, the antiphase boundary energy of Al_3Li would be even lower.

Finally, it seems appropriate to comment on the discrepancy between the antiphase boundary energies determined here from the minimum loop diameter and those determined from the dislocation pair spacing in underaged material. The pair spacing method has several drawbacks. It is generally assumed that the only important forces controlling their spacing are the repulsion between the dislocations and the attractive force from the antiphase boundary area between them. However, if this were the case the second dislocation should follow the contour of the lead dislocation, whereas transmission electron microscopic investigations suggest that the radii of curvature of the dislocations is different (De Hosson, et al., 1984; Furukawa, et al., 1985; Nembach, Suzuki, Ichihara and Takeuchi, 1985). These micrographs suggest that the second dislocation assumes a local strong line configuration. If this is the case, the dislocation pair spacing calculations leave out important forces that could significantly affect antiphase boundary energy predictions.

Conclusions

A modified version of the Hanson and Morris theory for the critical resolved shear stress has been developed that includes dislocation self-interactions and the effect of elastic anisotropy. The modified version can be used to predict the critical resolved shear stress of materials hardened by strong obstacles. For strong ordered precipitates, the theory predicts a simple relationship between the minimum looped precipitate diameter and the antiphase boundary energy. This relationship accounts for the effect of the mean square obstacle spacing on the minimum looping diameter through the dislocation line tension. The predicted antiphase boundary energies for Ni_3Al of 102± 35 mJ/m^2 and Al_3Li of 57 ± 15 mJ/m^2 are in good agreement with recent determinations by other techniques.

Acknowledgment

The authors would like to thank Prof. A. J. Ardell for helpful discussions of this work. This study was jointly supported by the Aluminum Company of America and the Director, Office of Energy Research, Office of Basic Energy Science, Material Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098. J. Glazer is supported by an AT&T Bell Laboratories Fellowship.

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Table 1 Antiphase boundary energies calculated from experimental data on binary aluminum-lithium alloys.

Reference	Minimum looped diameter (nm)	Volume fraction	Mean precipitate diameter (nm)	l_s (nm)	T (J/m)	γ (mJ/m^2)
deHosson et al (1984)	30	0.05	140	900	2.4×10^{-9}	56
Furukawa et al (1985)	25	0.18	80	275	2.0×10^{-9}	57
Sainfort and Guyot (1985)	17	0.25	11	35	1.4×10^{-9}	58

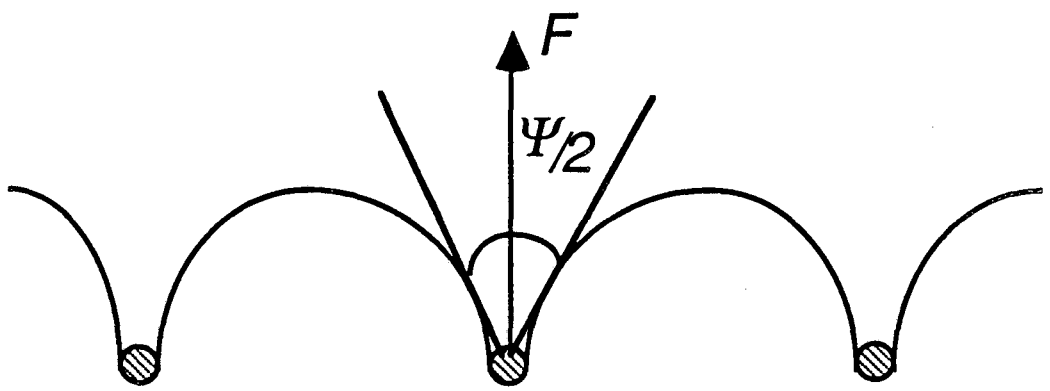

Figure 1 The force the dislocation exerts on the obstacle is defined as $F = 2T \cos(\Psi/2)$ where T is the line tension.

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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