Exchange Mediated Interaction of Dislocations and Deformation Hardening of Invar Alloys

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We propose an explanation of anomalies observed in the behavior of deformation hardening of Invars. An extremely strong volume magnetostriction, typical of Invars, results in an enhancement of the exchange contribution to the energy of the dislocation system by about three orders of magnitude with respect to its value in conventional ferromagnetics. Both the self energy of individual dislocations and the interaction between them, determined usually only by the elastic deformation, in Invars is strongly diminished by the contribution of the exchange energy. This fact explains a strong suppression of the deformation hardening rate in the second stage as well as its strong temperature and content dependence in Invars.

I. INTRODUCTION

Quite a few very unusual physical properties characterize FCC ferromagnetic Invar alloys. We can mention nearly zero thermal expansion coefficient, giant values of the volume magnetostriction, a strong pressure dependence of the Curie temperature and of the saturation magnetization, anomalies in the temperature and magnetic field dependence of the elastic constant and other. The origin of the Invar magnetic and structural instabilities has been intensively studied during the last decade (see [1–9] and references therein).

Since actually all physical properties of Invars are anomalous, one should not wonder that their plastic properties also strongly differ from those of the conventional FCC alloys [10–16]. We can mention here the critical resolved shear stress in Invars which grows with decreasing temperature with a rate, which is tens times larger than that of the normal alloys. An explanation of this effect has been recently proposed in our paper [17] where the exchange interaction of the dangling d-states of the dislocation cores and solute atoms (e.g., Ni atoms in FeNi Invar alloys) was considered. The current paper addresses the problem of the deformation hardening of Invars by considering the long range interaction between dislocations which is mediated by the exchange energy variation induced by the elastic strains in the vicinity if the dislocations. It will be demonstrated below that the type of interactions appears to be anomalously strong in Invar alloys.

A study of dislocation properties in ferromagnetic alloys started more than half a century ago by Brown [18] who discussed the influence of edge dislocations on the magnetization of ferromagnetics in a strong magnetic field. This direction of study was continued by Seeger and Kronmüller [19,20] (see also [21–23]). However, only linear magnetostriction caused by the shear stresses in the vicinities of dislocations were taken into account in those studies. As for the volume magnetostriction, it was generally neglected. This neglect seems to be well justified in conventional ferromagnetics (such as, say, Fe or Ni), since their volume magnetostriction is two or three orders of magnitude smaller than the linear one [24]. Considering the Invar alloys one should keep in mind that their volume magnetostriction is two orders of magnitude larger than that of the conventional ferromagnetics [24,25] and, hence, the neglect can be hardly justified.

Ferromagnetics are characterized by a very large contribution of the exchange interaction in the internal energy of the crystal. This contribution is proportional to the squared magnetization [23]. Dislocations induce hydrostatic strains in their vicinities which result in variations of the magnetization, and hence, of the exchange energy. In Invars, with their anomalously strong volume magnetostriction, the variation of the exchange energy appears to be anomalously strong. One should expect, as a result, an anomalously strong contribution of the exchange effects in the interaction between dislocations.

Introducing the notion of the exchange interaction between dislocations and paramagnetic obstacles has allowed the authors of this paper to explain the principal features of the electro- and magnetoplastic effects [24,25]. Recent studies demonstrate that sort of a "chemical" bonding between dislocation and obstacles may play an important part in plasticity of alloys [29].

The exchange effects appear to be exceptionally strong in Invars, as has been demonstrated in our previous paper [17], in which the anomalously strong temperature dependence of critical resolved shear stress of Invars has been
explained. We plan to calculate in the current paper the dislocation self energy and their interaction energy accounting both for the elastic and exchange contributions. It will be shown that anomalously strong volume magnetostriction in Invars results in a large exchange energy variation, which becomes comparable with the elastic energy. This relates both to the energy associated with individual dislocations and to the interaction between the dislocations. This mechanism will allow us to explain the anomalous behavior of the deformation hardening of Invars.

II. EXCHANGE INTERACTION CONTRIBUTION TO THE DISLOCATION SELF ENERGY IN INVARS

We calculate in this section the change of the total energy of a crystal resulting from an introduction of an edge dislocation. This change may be called the dislocation self energy. There are two principal contributions to the dislocation self energy in a ferromagnetic. First, there is an elastic energy, associated with the strain field induced by the dislocation in its vicinity. Second, there is an exchange energy, appearing due to the fact that this strain changes direction and absolute value of the local magnetizations $M$. The calculation of the elastic energy can be found in literature (see, e.g. [23,30]). Here we shall concentrate on the calculation of the exchange energy of a dislocation. In particular, it will be shown below that the variation of the magnetization direction makes a contribution negligible as compared to that of the absolute value variation. The same can be said about the exchange energy variation due to the linear magnetostriction.

The density of the exchange energy $w_{ex}$ in a ferromagnetic is presented by many authors and contains two terms (see, e.g., [25])

$$w_{ex} = -\frac{\omega M^2}{2} + \frac{\omega d^2(\nabla M)^2}{z}.$$  

(1)

The first term here depends only on the absolute value of the magnetization $M$, whereas, the second, gradient, term accounts for the space variations of $M$. Here

$$\omega = \frac{3k_B T_C}{np_{eff} \mu_B}$$

(2)

is the molecular field constant. $d$ is the interatomic spacing, $z$ is the first coordination number, $T_C$ is the Curie temperature. $p_{eff}$ is the effective number of the Bohr magnetons per one atom, $n$ is the density of atoms, $k_B$ is the Boltzmann constant, $\mu_B$ is the Bohr magneton.

An edge dislocation creates a hydrostatic pressure [30]

$$p(\rho, \varphi) = \frac{\mu b}{3\pi(1-\nu)} \frac{1 + \nu \sin \varphi}{\rho}$$

slowly decaying with the distance $\rho$ from the dislocation core. Here $\mu$ is the shear modulus, $\nu$ is the Poisson coefficient, $b$ is the value of the Burgers vector, $\varphi$ is the angle counted from the direction of the Burgers vector. The pressure $p(\rho, \varphi)$ induces a variation of the magnetization in the vicinity of the dislocation which in the linear, in the local pressure, approximation, can be represented as

$$\Delta M(\rho, \varphi) = \alpha \overline{M} p(\rho, \varphi).$$

Here $\overline{M}$ is the uniform magnetization of the ferromagnetic in the absence of dislocations, $\alpha$ is a proportionality constant known empirically for various ferromagnetics (see, e.g., [31] for Invars).

Now we can calculate the variation of the exchange energy of the ferromagnetic caused by an edge dislocation. The variation of the first term in equation (1), depending only on the absolute value of the magnetization, reads

$$\Delta w_{ex}^m = -\frac{\omega \overline{M}^2}{2} [\alpha^2 p^2(\rho, \varphi) + 2\alpha p(\rho, \varphi)].$$

Integrating this expression over a plane perpendicular to the dislocation axis, one finds that the dislocation exchange energy per the dislocation unit length is

$$W_{ex}^m = -\frac{\omega \overline{M}^2 \alpha^2 b^2 \mu^2}{18\pi} \left( \frac{1 + \nu}{1 - \nu} \right)^2 \ln \frac{R}{r_0}.$$  

(3)
The calculation of the integral \( I \) was carried out under the same approximations as those used in the calculation of the elastic energy of the dislocation in a continuous medium \([30]\). The integral is cut off at small distances, \( \rho > r_0 \sim d \), and at large distances, \( \rho < R \), with \( R \) being of the order of the average distance between the dislocations.

The gradient part of the dislocation exchange energy (the second term in \([1]\)) is calculated in a similar way,

\[
W_{ex}'' = \frac{\omega M^2 \alpha^2 b^2 \mu^2}{9 \pi z} \left( \frac{1 + \nu}{1 - \nu} \right)^2 \tag{4}
\]

The quantity \( \ln \frac{R}{\rho} \) is about 10 for typical concentrations of dislocations (\( \sim 10^7 \) to \( 10^8 \) cm\(^{-2} \)), while \( z = 12 \) in FCC lattices. Therefore, the gradient term \([4]\) is about \( 10^{-2} \) of the dislocation exchange energy \([3]\).

This difference of the two contribution follows mainly from the fact that the variation of the absolute value of the magnetization, \( \Delta M(\rho, \varphi) \), decays only as \( \rho^{-1} \) with the distance from the dislocation core, whereas the gradient, \( \nabla M(\rho, \varphi) \) decays more rapidly as \( \rho^{-2} \). We should also consider the contribution of the linear magnetostriction appearing due to the shear deformations induced by the dislocations. The change of the local magnetization is proportional the gradient of the angle \( \vartheta \) defining the direction of the atomic spins. It means that the dislocation self energy due to the linear magnetostriction has generally the structure similar to that of the gradient term in \([1]\) and is also small. We shall neglect these two small contributions in what follows and omit the superscript \( m \) in the exchange energy \([3]\).

We need also the elastic energy per dislocation unit length which in the same approximation \([23,30]\) reads

\[
W_{el} = \frac{\mu b^2}{4 \pi (1 - \nu)} \ln \frac{R}{r_0}. \tag{5}
\]

Now the total self energy of a dislocation (per its unit length) containing both elastic and exchange contributions can be represented as

\[
W = W_{el} + W_{ex} = f(T)W_{el} \tag{6}
\]

where

\[
f(T) = 1 - \frac{\omega M^2(T) \alpha^2 E}{9} \left( \frac{1 + \nu}{1 - \nu} \right). \tag{7}
\]

Here the equality

\[
E = 2\mu(1 + \nu)
\]

connecting the Young modulus \( E \) with the shear modulus \( \mu \) has been used.

Since the magnetization \( M(T) \) is a function of temperature, the exchange contribution to the total energy \( W \) is also temperature dependent. As for the elastic contribution \( W_{el} \) its very weak temperature dependence can be neglected. In order to estimate the temperature dependence of the total energy we may use the mean field approximation in the theory of the second order phase transitions, according which the magnetization is

\[
M(T) = M_0 \sqrt{1 - \frac{T}{T_C}} \tag{8}
\]

where \( M_0 \) is the spontaneous magnetization of the ferromagnetic at zero temperature. Then the factor \([6]\) can be represented as

\[
f(T) = 1 - \frac{\omega M_0^2 \alpha^2 E}{9} \left( \frac{1 + \nu}{1 - \nu} \right) \left( 1 - \frac{T}{T_C} \right). \tag{9}
\]

The simple formula \([8]\) for the magnetization works rather well in usual ferromagnetics in a wide range below the Curie temperature. As for Invar alloys, it holds up to the temperatures which are 20 to 30K below \( T_C \) \([1]\). Closer to \( T_C \) the magnetization does not go to zero but becomes very small and falls down slowly with the increasing temperature.

Now we demonstrate that only in Invars the contribution of the exchange energy may be of importance. Really, the exchange correction in the factor \([7]\) is very small in conventional ferromagnetics and \( f \) is close to one. For example, Ni is characterized by the following parameters: \( M_0 = 0.510 \) kG, \( E = 3 \times 10^{12} \) dyn/cm\(^2\), \( \nu = 0.276 \), \( \omega = 13800 \) \([24]\). As for the quantity \( \alpha \) its value according to \([31,32]\) is \( -3 \times 10^{-13} \) (dyn/cm\(^2\))\(^{-1} \). Then one finds that the exchange correction in \([7]\) does not exceed \( 2 \times 10^{-4} \) and, hence, can be neglected in normal ferromagnetics.
The situation changes dramatically in Invar alloys in which the quantity $\alpha$ is one to two orders of magnitude larger than in normal ferromagnetics \[1\]. For example, the Fe$_{0.65}$Ni$_{0.35}$ Invar is characterized by $\alpha = -1.1 \times 10^{-11} (\text{dyn/cm}^2)^{-1}$ \[3\] which is nearly 40 times larger than in Ni. As a result, the exchange energy contribution, proportional to $\alpha^2$, becomes one thousand times larger. The factor $f$ for the Fe$_{0.65}$Ni$_{0.35}$ alloy can be estimated. First, the molecular field constant $\omega$ can be calculated using equation (3). It is known for this alloy that $T_C = 503K$ \[1\], $p_{eff} = 1.851$ \[3\], $n = 8.72 \times 10^{22} \text{cm}^{-2}$ which results in $\omega = 8110$. Then using the values $M_0 = 1.4kG$, $E = 1.4 \times 10^{12} \text{dyn/cm}^2$, $\nu = 0.3$ \[3\], one finds that $f = 0.35$ at low temperatures ($T \ll T_C$), meaning that the exchange effects are able to decrease the total dislocation self energy \[6\], by a factor of three.

III. EXCHANGE ENERGY CONTRIBUTION TO THE INTERACTION BETWEEN DISLOCATIONS IN INVARS

We may distinguish two types of interaction between dislocations connected with the magnetic structure of the ferromagnetic. The direct magnetic interaction between local magnetizations in the vicinities of parallel edge dislocations was calculated by Krey \[33\]. This interaction appears to be three orders of magnitude weaker than the elastic interaction even in Invar alloys with their anomalously strong variations of the local magnetization. This allows us to neglect in what follows the direct magnetic interaction of dislocations.

However, a special attention should be paid to the second type of interaction, i.e., the interaction between the dislocations caused by the exchange effects, which can be much stronger than the direct magnetic interaction. Considering the simplest case of two parallel edge dislocations in the same sliding plane, when they have either the same or the opposite mechanical signs, we can calculate the exchange energy contribution to the interaction between the dislocations.

Let $a$ be the vector connecting two dislocations. Then the local pressure induced by these two dislocation in a point $r$ is

$$p(r) = p_1(r) + p_2(r - a)$$

where $p_1$ and $p_2$ are the partial local pressures induced by the first and the second dislocations, respectively. Now repeating the calculations of the previous section with the pressure distribution \[10\] one arrives at the total change of the exchange energy of the ferromagnetic caused by the two dislocation. Then subtracting twice the self energy $W_{ex}$ \[9\] of the individual dislocations one may get the contribution of the exchange energy into the interaction between the dislocations,

$$W_{ex} = \pm \frac{\omega M^2 \alpha^2 b^2 \mu^2}{9\pi} \left( \frac{1 + \nu}{1 - \nu} \right)^2 \left( \frac{1}{2} + \ln \frac{R}{a} \right).$$

The signs correspond either to the same mechanical signs (−) or to the opposite mechanical signs (+) of the dislocations. The force due to the exchange interaction acting between the dislocations per their unit lengths is

$$F_{ex} = -\frac{\partial W_{ex}}{\partial a} = \pm \frac{\omega M^2 \alpha^2 b^2 \mu^2}{9\pi a} \left( \frac{1 + \nu}{1 - \nu} \right)^2.$$  \(11\)

The exchange contribution to the interaction between the dislocations has the sign opposite to that of the elastic interaction. Contrary to the elastic interaction, the exchange interaction results in an attraction of the dislocations with the same mechanical signs and a repulsion of the dislocations with the opposite signs.

The elastic force between two parallel edge dislocations \[12\] is

$$F_{el} = \pm \frac{\mu b^2}{2\pi(1 - \nu)a}.$$  \(13\)

Therefore, the total force acting between the two dislocations is

$$F(a) = F_{el}(a) + F_{ex}(a) = f(T)F_{el}(a)$$  \(14\)

where $f$ is the same factor \[5\] introduced in the previous section. If the configuration of the two dislocations is more complicated than that considered here, the expressions for the elastic and exchange contributions to the interaction will differ. However, we believe that the relation \[14\] will generally hold. Hence, the discussion carried out in the previous section relates also to the forces acting between the dislocations. In conventional ferromagnetics the total force \[13\] nearly coincides with the elastic force \[12\] ($f \approx 1$). However, in Invars the interaction due to the exchange energy becomes anomalously strong at low temperatures and the factor $f$ may become essentially smaller than one. Meaning that the total interaction between the dislocations becomes essentially smaller than the elastic one.
IV. DEFORMATION HARDENING OF INVAR ALLOYS

The analysis of the interaction between dislocation creates a ground for discussing some specific features of deformation hardening in Invar alloys. The deformation hardening in normal FCC metals and alloys is well studied (see, e.g., review [34]). The hardening can be subdivided into three stages. The hardening rate $\theta_{II}$ in the second stage, usually called the rapid stage of the deformation hardening, is approximately ten times larger than the hardening rate in the first stage. That is why one can easily distinguish between them. The quantity $\theta_{II}$ hardly depends either on the temperature or on the alloy content.

According to the experiments [11,13] none of these features hold in Invar alloys. The hardening rate $\theta_{II}$ rapidly falls down with decreasing temperature and strongly depends on the alloy content. At low temperatures ($T \ll T_C$) its value is two to three times smaller than the value $\mu/300$ typical for conventional FCC alloys. Therefore, the first and the second stages are separated not well enough.

The result of the previous two sections allow us to understand this drastic change of the behavior of the deformation hardening in Invar alloys. The hardening rate $\theta_{II}$ is proportional to the interaction between dislocations [23]. Therefore, accounting for the exchange contribution to the interaction between the dislocations, the hardening rate becomes

$$\theta_{II} = f(T)\theta_{II, el}$$

(15)

where $\theta_{II, el}$ is the hardening rate caused by the elastic interaction only.

As discussed above the factor $f(T)$ is nearly one in conventional ferromagnetics, whereas in Invars this factor becomes at low temperatures essentially smaller than one. $f(T)$ in Invars decreases with the decreasing temperature which should lead to a corresponding decrease of the deformation hardening rate $\theta_{II}$. Such a temperature dependence was really observed in [4]. In case of the Fe$_{0.65}$Ni$_{0.35}$ the factor $f(T)$ can become as small as $f = 0.35$ at $T \ll T_C$, meaning that the hardening rate $\theta_{II}$ in the Invar should be approximately three times smaller that its pure "elastic" value $\theta_{II, el}$.

The exchange contribution to the dislocation interaction becomes negligible at temperatures above $T_C$, meaning that $\theta_{II}$ measured at high temperatures is purely elastic one, $\theta_{II, el}$. This allows us to use the experimental data (above and below $T_C$) presented in references [11,13] and estimate the ratio $\theta_{II, Invar}/\theta_{II, normal}$ as lying in the range from $1/3$ to $1/2$. It agrees very well with our theoretical estimate of the $f$ value.

The parameter $\alpha$ depends on the Invar alloy content. Even a slightest deviation from the typical content Fe$_{0.65}$Ni$_{0.35}$ causes a strong drop of the $\alpha$ value and, hence, much stronger drop of the exchange effect contribution. Just, for example, changing the Ni concentration in this alloy from 35% to 38% results in a decrease of $\alpha$ by a factor of two [32]. Then, according to [4], the coefficient $f$ changes from 0.35 to 0.84. The exchange effects, as a result, are essentially suppressed and the deformation hardening rate in such an alloy is much closer to that of a normal alloy. The variation of the coefficient $f$ with the alloy content may provide an explanation of a strong dependence of the deformation hardening rate on the Invar alloy content, observed in references [11,13].

In order to demonstrate it, we calculate the dependence of the deformation hardening rate $\theta_{II}$ on the concentration dependence in FeNi Invar alloys (see figure 1). Unfortunately the available experimental information is very limited. The value of the coefficient $\alpha$ at room temperature is known only for the Fe$_{0.65}$Ni$_{0.35}$ alloy [4]. Reference [32] provides the $\alpha$ values for the Ni concentrations, 34.7% and 44.9%, but only at low temperatures, 4.2 and 20.4 K. To the best of our knowledge there are no other data on the concentration dependence of this coefficient. That is why we assume the concentration dependence of $\alpha$ measured in [4] at low temperatures and scale it to the room temperature value of $\alpha$ [31]. This rather rough approximation can be readily improved when new data on the $\alpha$ concentration dependence will appear.
The choice of the remaining parameters is straightforward. The deformation hardening rate, $\theta_{II, el}$, due to elastic strains is $\mu/300$ [11]. The elastic constants $E = 1.4 \times 10^{11}$ dyn/cm$^2$ and $\nu = 0.3$ do not depend on the alloy content at room temperature [33,35]. The Curie temperature for Invar alloy is very high, so we may assume that the magnetization has arrived at saturation at room temperature, i.e., $M = n p_{eff} \mu_B$ [6]. Then using equations (2) and (7) one gets

$$f = 1 - \frac{1}{3} k_B T_c \alpha^2 E \frac{1 + \nu}{1 - \nu}$$

(16)

The dependence of the Curie temperature on the alloy content is available in [33]. Then using equation (15) we are able to calculate the concentration dependence of the deformation hardening $\theta_{II}$ for FeNi alloy presented by the solid line in figure 1. The theory correctly predicts an increase of the deformation hardening with the increasing Ni concentration. It is also in good quantitative agreement with the experiment for the Fe$_{0.65}$Ni$_{0.35}$ alloy for which we have reliable experimental value of the coefficient $\alpha$, which may be the cause of this deviation. This calculation demonstrates a necessity of additional measurements of various characteristics of Invar alloys with varying contents in order to verify the model proposed in this publication.

V. CONCLUSIONS

This study demonstrates that the peculiar behavior of the deformation hardening of Invars cannot be explained by applying the conventional approach which considers only the elastic strain mediated interaction of dislocation. It is important to incorporate also the influence of the exchange effect on the interaction between dislocations which is anomalously strong in Invars. The exchange mediated interaction between dislocations has the sign opposite to the elastic contribution and may essentially diminish the total interaction at temperatures well below the Curie temperature. This decrease of the interaction is responsible for the lower rate of the deformation hardening in the second stage. This mechanism allows one also to explain the strong temperature and content dependence of the deformation hardening rate observed experimentally. An interesting case when the exchange interaction is so strong that it changes the sign of the total interaction needs a special attention and will be studied elsewhere.

A more detailed experimental study of the temperature and content dependence of the deformation hardening rate in Invars is necessary in order to verify the theoretical model developed in this paper. Equations (6) and (13) indicate that the temperature dependence of the deformation hardening rate $\theta_{II}(T)$ is directly connected to temperature dependence of the Invar magnetization $M(T)$. An experimental observation of such a connection would provide a strong support for our model.
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