Dislocation Paths in a Magnetic Field†

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Influence of a magnetic field on dislocation paths in crystals with paramagnetic obstacles is studied. The model incorporates, as the most important aspect, a dependence of the energy of dislocation−obstacle bonds on their spin multiplicity. An increase of the crystal plasticity in a magnetic field is explained by an increased population of high spin states with lower binding energy. The dependence of the average dislocation path length on the magnetic field accounting also for the hyperfine interaction between electron and nucleus spins is obtained. The theory agrees well with the available experimental data in a wide range of magnetic fields. The hyperfine interaction is shown to play a crucial role, especially at relatively low magnetic fields. The possible dependence of plastic properties of crystals on the value of nuclear spin may lead to a magnetic isotope effect in plasticity. The hyperfine interaction may effectively suppress the contribution of paramagnetic obstacles in the magnetoplastic effect if the external magnetic field is smaller than a certain value that is specific for each particular type of the obstacle.

I. Introduction

Variation of plastic properties of a crystal in a magnetic field has a general name of a magnetoplastic effect (MPE). Both positive and negative MPEs are observed, meaning that the magnetic field may either harden or soften crystals. The type of the effect observed in a particular system depends on the electronic structure of the crystal, types of defects, their magnetic properties, and concentrations. A positive MPE is typical of pure metals at low temperatures (see, reviews[1]). Such a behavior may be explained by the Kravchenko effect[2] according to which the electron component of the viscous dislocation drag grows in a magnetic field.

A negative MPE may be observed in all metals, dielectrics, and semiconductors.[3−16] If we restrict ourselves by considering only nonmagnetic materials then a conclusion can be made that a presence of paramagnetic obstacles pinning the dislocations is a necessary condition for a negative MPE. Such obstacles are always available in real crystals unless they have passed a special treatment. Contrary to the Kravchenko effect a negative MPE may be observed in a broad temperature range from helium to room temperatures, and sometimes even at higher temperatures. This feature makes one pay special attention to studying the MPE.

Our papers[17,18] propose a mechanism based on a model similar to that which has been successfully applied for an explanation of a magnetic field effect on radical chemical reactions.[19−21] The model considers bonds created between paramagnetic obstacles and passing dislocations. Each such bond is actually a radical pair formed due to interaction between a paramagnetic obstacle and dangling bonds of the dislocation core. (In metals we should be certainly more careful when speaking about a dangling bond. However, the model remains essentially the same.) The energy of the exchange interaction depends on the electron structure of atoms forming the dislocation−obstacle bond, on their valence, electronegativity, and so on.

However, the most important feature of such bond is that its binding energy strongly depends on the spin multiplicity of the pair. A strong bond is formed only in the singlet (S) state when spins of the two electrons are antiparallel. The binding energy in a triplet (T) state with parallel spins is very small, if any. Therefore, S−T transitions, induced by a magnetic field and favoring an occupation of weakly binding T states, may facilitate dislocation depinning and, hence, result an a plasticity growth.

It is worth noting here that the difference of the radii of the defect and host atoms (size factor) may lead to elastic strains. However, such elastic interaction results mainly in simultaneous shifts of the energies of the S and T states and is not capable of influencing significantly the mechanism described above.

The S−T model[17,18] allows one to explain such principal features of the MPE as an influence of a magnetic field on the internal friction of dislocations,[22] deformation hardening of crystals induced by a magnetic field.[23] The mysterious electroplastic effect, having remained without a consistent explanation for a quarter of a century, can be now understood much better.[24] These ideas about the role of the spin states formed the basis for our prediction made in 1996[23] that a resonance increase of plasticity may be observed in crossed permanent and microwave magnetic fields. The resonance takes place when the microwave frequency coincides with the ESR frequency of a dislocation−obstacle bond. Such a resonance was actually observed in 1998 by Golovin et al.[13−15] These experiments provide a direct evidence indicating the principal role of the spin states in MPE.

Another important idea, put forward for radical chemical reactions,[19−21] is that the hyperfine interaction (HFI) between the spins of the radical pair and the spin of the relevant nuclei produces an effective magnetic field which, similarly to the external magnetic field, may induce S−T transitions and thus influence the reaction rate. Our paper[25] discusses the role of HFI in MPE. The calculation shows that the HFI may actually

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change the plastic properties of crystals at relatively low magnetic fields.

The major part of available experimental results is devoted to studying the influence of a magnetic field on dislocation paths (see, e.g., refs 3−16 and references therein). This is an enormous amount of experimental data which still waits for a consistent interpretation, although some considerations as to the mechanism of this influence have been presented in refs 5 and 17. The aim of this paper is to propose a quantitative approach to this phenomenon with a proper account of the HFI. This approach can be applied to an interpretation of experimental data on the dislocation paths and, in particular, it will allow us to reveal the important role of magnetic nuclei in crystal plasticity.

II. Dislocation Paths in a Strong Magnetic Field

Plastic properties of real crystals with a large number of defects are controlled by pinning and depinning of dislocations from obstacles created by these defects. The defects may be roughly subdivided into two groups of strong and weak obstacles. The start of a dislocation motion is controlled by its freeing from strong obstacles. This becomes possible either due to etching of the surface strong obstacles or under the action of a strong enough mechanical stress. Thermal activation may be also of an importance for depinning from the strong obstacles.

Currently available magnetic fields may hardly influence depinning of dislocations from strong obstacles.[5] They are able to influence only depinning of dislocations from weak paramagnetic obstacles whose concentration is always larger than that of the strong obstacles. The role of the strong obstacles is usually played by the "forest" dislocations. The major part of the measurements is done in alkali−halide crystals where the role of magneto-sensitive weak obstacles is usually played by paramagnetic impurities (Ca, Mg, Ni, Li), color center, etc. Nonmagnetic impurities (Pb, Cu) do not contribute to MPE.

Dislocations, on getting free from strong obstacles, move under the action of external or internal stresses. Colliding with weak paramagnetic obstacles, the dislocations may form radical pairs (bonds) with them. An obstacle is able to pin a dislocation only if the radical pair is in the S state. If the radical pair is in a T state, the binding energy (if any) of the dislocation−obstacle bond is so small that the dislocation actually passes the obstacle without "seeing" it.

The Zeeman energy of the interaction of an electron spin with a magnetic field is usually negligibly small. That is why S−T transitions occur only if the S and T energies nearly coincide with each other. Such a situation takes place when the distances between the dislocation and the obstacle exceed 10^{-7} cm so that the exchange interaction in the radical pair is still weak and does not lift the degeneracy between S and T levels.[19] S−T transitions in our case take place when the dislocation approaches the obstacle but is still at a distance of two to three lattice spacings from it.

A magnetic field splits the triplet states into three magnetic sublevels, T_+, T_0, and T_- corresponding to three projections (+1, 0, −1) of the total spin of the radical pair. (We deliberately disregard here possible role of the electron orbital momentum which is not necessarily zero. It would not change qualitatively the MPE as described here, although it may add new interesting features which deserve a separate discussion.) If the magnetic field is strong enough, the T_+ and T_- states shift so strongly that they get out of resonance with the S state, and the spin conversion occurs due only to S−T_0 transitions. The latter are driven by the Δg mechanism[19−21] caused by a difference of the g factors of the two electrons in the radical pair and, hence, by the corresponding difference of the Zeeman frequencies. The Δg mechanism has been successfully applied in our papers[17,18,22−25] when considering various MPE related phenomena. This paper considers how this mechanism works in the theory of the dislocation paths in a magnetic field.

Let us now consider the following situation. Similar to ref 18, the spin relaxation time of the T_0 is assumed to be much shorter than that of the S state. Then after a transition from the S to T_0 state, induced by the Δg mechanism, the system rapidly relaxes to either the T_+ or T_- state, allowing for a free passage of the dislocation near the obstacle. This model holds under the assumption that the time of life of the relaxed $T_\pm$ states is longer than the passage time of the dislocation near the obstacle. This ad hoc assumption needs a special investigation which has not been carried out as yet. Currently, one can state only empirically that this condition seems to be valid in all known experimental situations.

The analysis of experimental data on the dislocation paths to be done below (see also ref 18) demonstrates that the passage time of a dislocation near an obstacle is of the order of 10^{-7} s, provided that the dislocation−obstacle radical pair is in a T state. A large number of paramagnetic centers are known whose spin relaxation times exceed 10^{-7} s, even at room temperature.[26] One may think that long living relaxed $T_\pm$ states appear due to strong deformations in the vicinity of the dislocation core. It is well known that[27] a distortion of strongly bound Jahn−Teller systems suppresses spin relaxation.

The forest dislocations may strongly pin moving dislocations. Therefore, the dislocation path lengths saturate when approaching the average distance between the forest dislocations. Below we shall consider paths which are shorter than this distance. Then one can be sure that the dislocation path length is controlled mainly by the interaction with weak obstacles.

To determine the dependence of the average dislocation path length on a magnetic field one should consider the average speed of the dislocation motion in a defect crystal. It is proportional to $1/\sqrt{C}$[28] where C is the concentration of the active obstacles. According to our model the obstacle is active, i.e., pins the passing dislocation, only in the S state. Then the concentration $C(H)$ of the active obstacles in the magnetic field H is proportional to the diagonal element $\rho_{SS}(H)$ of the density matrix which describes the population of the S state of an individual radical pair

$$C(H) = C_0\rho_{SS}(H)$$

with C_0 being the total concentration of the paramagnetic obstacles.

The average speed $v(H)$ of dislocations in a magnetic field is proportional to $1/\sqrt{\rho_{SS}(H)}$. Then one gets the average dislocation path length $L(H) = v(H)\Delta t$ during the time span Δt as a function of the magnetic field in the form

$$L(H) = L(0)\sqrt{\frac{\rho_{SS}(0)}{\rho_{SS}(H)}} \qquad (1)$$

Our calculation[18] assumes that initially dislocation−obstacle radical pairs are formed in one of the four states (S, T_0, T_-, T_+) with equal probability. After that the populations of these states evolve in the course of the dislocation passage, and it is the final values of $\rho_{SS}(H)$ which determine the pinning probability. Here we choose another initial condition that the pairs are formed in the binding S state. This empirical assumption is widely used in the theory of chemical reactions[29−31] where it

leads to a good agreement with the experiment. The calculation similar to ref 18 but with the new initial condition $\rho_{SS}(0) = 1$ results in

$$\rho_{SS}(H) = \frac{\left(1+\frac{T_1}{\tau}\right)\left(1+\frac{T_2}{\tau}\right)+\frac{H^2}{2H_m^2}}{\left(1+\frac{T_1}{\tau}\right)\left(1+\frac{T_2}{\tau}\right)+\left(1+\frac{\tau}{2T_1}\right)\frac{H^2}{H_m^2}} \quad (2)$$

where the field

$$H_m = \frac{\hbar}{\Delta g \mu_B \sqrt{T_1 T_2}}$$

characterizes the S−T transitions, T_1 and T_2 are the times of longitudinal and transverse relaxations of the T_0 state, and μ_B is the Bohr magneton.

When deriving eq 2 it was assumed (similarly to ref 18) that the time of the longitudinal spin relaxation of the S state is much longer than T_1, whereas the time of the transverse spin relaxation of the S state coincides with T_2. The region of the S−T resonance and the size of the obstacle are both of the order of interatomic spacing. The dislocations (if the pair is in a T state) move nearly freely both in the resonance region and closer to the obstacle. Therefore, more or less the same time τ determines both the passage time through the resonance region and the average passage time near the obstacle.

Equations 1 and 2 yield the dependence of the average dislocation path length on the magnetic field

$$L(H) = L(0)\sqrt{\frac{\left(1+\frac{T_1}{\tau}\right)\left(1+\frac{T_2}{\tau}\right)+\left(1+\frac{\tau}{2T_1}\right)\frac{H^2}{H_m^2}}{\left(1+\frac{T_1}{\tau}\right)\left(1+\frac{T_2}{\tau}\right)+\frac{H^2}{2H_m^2}}} \quad (3)$$

In a strong field $H \gg H_m$ the dislocation path length dependence (eq 3) saturates

$$L(\infty) = L(0)\sqrt{2+\frac{\tau}{T_1}} \quad (4)$$

Such a saturation at 20 to 30 T was observed in the NaCl crystals[12] after their illumination by a visible light. The high field limit $L(\infty)$ appears to be an order of magnitude larger than the path length $L(0)$ in the absence of the field. According to eq 4 such a ratio of $L(\infty)$ to $L(0)$ corresponds to the ratio $\tau/T_1 \sim 10^2$ of the characteristic times. A similar estimate has been obtained by us in ref 18 when analyzing other types of experiments.

Fitting eq 3 to the data,[12] an estimate $H_m \sim 5$ T is obtained which is rather close to $H_m \sim 3$ T of ref 18. This value of H_m allows one also to estimate the typical values of the spin relaxation times. Assuming $T_1 \sim T_2$ equation for H_m yields

$$T_1 \sim \frac{\hbar}{\Delta g \mu_B H_m}$$

At typical values of $\Delta g \sim 10^{-3}$ and $H_m \sim 5$ T one gets $T_1 \sim 10^{-9}$ s. This estimate is quite reasonable and corresponds to the range of typical spin relaxation times for impurity centers[26] and for color centers[32] in ionic crystals at room temperature.

The time τ is 2 orders of magnitude larger than T_1. At $T_1 \sim 10^{-9}$ s, the passage of a dislocation in the vicinity of an obstacle takes about $\tau \sim 10^{-7}$ s. Our model holds if the times of life of the $T_\pm$ states, allowing for a free passage of the dislocation, are at least not smaller than τ. An indirect evidence favoring such long living states in dislocations in NaCl are provided by the experiments[13−15] on the resonance increase of the plasticity of crystals in a microwave magnetic field where states with the time of life $\sim 10^{-7}$ s have been revealed.[13]

The characteristic field H_m appears to be very large. All of the experiments except for ref 12 were carried out at much lower fields. Then expanding eq 3 over small $(H/H_m)^2$ one gets

$$L(H) = L(0)\left(1+\frac{H^2}{H_0^2}\right) \quad (5)$$

where

$$H_0 = 2H_m\sqrt{\frac{T_1}{\tau}\left(1+\frac{T_2}{\tau}\right)} \quad (6)$$

The simple quadratic dependence eq 5 is observed in the range above ca. 0.5 T (e.g., refs 3−10). The experimental values of H_0 are about several tenths of Tesla which corresponds to a H_m of several Tesla if the time τ is 2 orders of magnitude larger than both T_1 and T_2.

III. Magnetic Isotope Effect in Plasticity

At lower magnetic fields ($\leq$ 0.5 T) strong deviations from the simple quadratic dependence eq 5 are observed. Sort of a crossover near $H \sim 0.2$ to 0.4 T appears below which the magnetic field hardly influences the MPE.[8,14] Our paper[25] explains this crossover as being connected to the hyperfine interaction (HFI) of the electron and nuclear spins. Here this model is applied to the dislocation paths in a magnetic field.

It is worth noting that a participation of nuclear spins in radical chemical reactions is a well known phenomenon.[19−21] The HFI between the electron and nuclear spins results in a dependence of chemical reaction rates on the value of the nuclear magnetic moments and, hence, on the isotope content of the reagents. This makes an observation of a magnetic isotope effect in chemical reactions possible. A similar influence of nuclear spins on MPE can also lead to a possibility of an isotope effect in plasticity, meaning that we may expect a change of the whole pattern of the MPE when intentionally changing the isotope content of the samples studied.

The results obtained previously in ref 25 are used now in our discussion of the influence of the nuclear magnetic moments on the dislocation paths in a magnetic field. The nuclear spin relaxation times are usually rather long. For example, the relaxation time for the ^{19}F nuclei in LiF crystals at room temperature exceeds 20 s.[33] This time becomes even larger at lower temperatures. This allows one to assume that the electron spin evolution proceeds in an effective field of the nuclei with fixed spins. The rate of the S−T transitions is now controlled by the sum of the external magnetic field and the nuclear effective field.

Similarly to ref 25, we shall assume that there is only one magnetic nucleus with a spin 1/2 in the dislocation−obstacle pair. The total field for nuclear spins parallel and antiparallel to the external field is $H + H_A$ and $H - H_A$, respectively. Here

$$H_A = \frac{A}{2\Delta g} \quad (7)$$

with A being the HFI coupling constant.

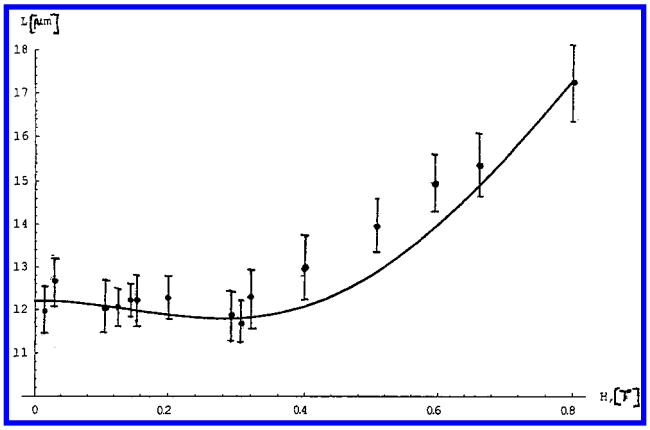

Figure 1. Dependence of the dislocation path length on a magnetic field: comparison of the theory (solid line) with the experimental data.[14]

The action of the external magnetic field on the nuclear spin orientation is negligibly weak, and we may assume that both spin orientations occur with equal probabilities. Hence, both orientations of the effective field H_A are also equally probable. Then the S state occupation is proportional to the density matrix element

$$\rho_{SS}^{HFI}(H) = \frac{1}{2}[\rho_{SS}(H + H_A) + \rho_{SS}(H - H_A)] \qquad (8)$$

Here the function $\rho_{SS}(H)$ is described by eq 2. Now eq 8 is substituted into eq 1 and one arrives at the dependence

$$L(H) = L(0)\sqrt{\frac{\rho_{SS}^{HFI}(0)}{\rho_{SS}^{HFI}(H)}} \qquad (9)$$

of the average dislocation path length on a magnetic field with an account of the HFI.

At $H < H_A$ the dislocation path length (eq 9) does not practically depend on H, since the spin transitions are controlled mainly by the HFI. A crossover to the conventional Δg mechanism occurs around H_A so that at higher fields the plasticity grows until its saturation.

The crossovers found in NaCl[8,14] yield $H_A = 0.2$ to 0.35 T. (The authors of these papers use the word "threshold" rather than "crossover". However, our analysis shows that we deal with a rather smooth transition between two regimes. That is why using the word "crossover" seems to us more appropriate.) Assuming a typical value of $\Delta g \sim 10^{-3}$, [19] eq 7 with such H_A value produces the HFI constant $A \sim 4$ to 7×10^{-4} T ~ 10 to 20 MHz. Such A values are quite reasonable and close to the HFI constants for various paramagnetic impurities and color centers in ionic crystals.[34−36]

Figure 1 presents a comparison of the theoretically calculated dependence of the dislocation path lengths (eq 9) with the experimental data for NaCl by Golovin et al.[14] Here $H_A = 0.35$ T and $H_m = 5$ T. The values $L(0) = 12.2$ μm and $\tau/T_{1,2} = 270$ are found by fitting the function (eq 9) at $H = 0$ and $H = 0.8$ T. One can see that the function $L(H)$ agrees well with the experimental data in the whole range of the H values.

The above theory accounting for the influence of a magnetic field on the transitions between different spin states of dislocation−obstacle bond describes well the observed dependence of the dislocation path lengths on the magnetic field. However, recent studies carried out by Golovin with co-workers[13] demonstrate that, in addition to the processes considered above, a decay of obstacles in a magnetic field may also contribute to the MPE. This decay is not included in the model discussed here. Unfortunately we still lack efficient experimental techniques of separating the contributions of the decay processes in MPE. That is why, despite seemingly excellent agreement between the theory and experiment, one should be more cautious by speaking only about a reasonable agreement.

IV. Suppression of MPE by Hyperfine Interaction

A paramagnetic obstacle may contribute to MPE only when the applied magnetic field exceeds the crossover field H_A of the corresponding radical pair. This fact puts essential limitations on the types of paramagnetic centers playing part in the MPE of a particular crystal. Crossovers near 0.2 to 0.4 T are most probably connected with paramagnetic centers characterized by a rather large localization radius of the electron. Since the HFI constant is proportional to the electron density on the relevant nucleus,[33] a larger localization radius of the electron favors lower values of the parameter A and, hence, a manifestation of the MPE in weaker magnetic fields. Symmetry may be also a factor acting in the same direction, e.g., d-electrons may have low density on the nuclei.

This point of view is supported by the experiments[7] in which a sharp rise of the number of the magneto-sensitive centers is observed after an X-ray irradiation of NaCl crystals. It is known[34] that such an irradiation results in the generation of a large number of electrons and holes. They are captured by shallow traps, e.g., by anion and cation vacancies and their complexes, and then form paramagnetic centers with large radii and low values of the HFI constant.

The opposite effect takes place when optically illuminating the crystals. In this case, impurity centers with energies lower than those of the absorbed quanta are ionized. The electrons excited to the conduction band are captured later by various traps. This capture holds for the traps with the depth exceeding the energy of the quanta. That is why a long time optical illumination leads to a transfer of electrons from shallow to deeper traps with smaller electron localization radii. Such traps are characterized by larger HFI constants, and, as a result, the optical illumination of crystals leads to a shift of the MPE crossover toward higher fields.

Such a shift has been revealed in dislocation paths in NaCl crystals subject to a prolonged optical illumination. Based on the first experiments[11] carried out in the fields up to 7 T, a conclusion of the existence of an optical quenching of the MPE had been made. However, later on, when the fields up to 30 T were achieved it became clear that the optical illumination did not kill the MPE completely. The crossover, discussed above, is shifted from a field a little bit larger than 0.3 to 0.4 T to the fields in the range from 7 to 10 T.[12] Now we understand that just this type of behavior of the crossover under optical illumination follows from the above discussion. It is emphasized also in reference 11 that this state is reversible and the MPE restores (the crossover field decreases) a few tens of minutes after the illumination has been switched off.

One may think that a high HFI value for Mn defects in alkali−halides prevents their active participation in the MPE. According to ref 10, magnetic fields up to 0.7 T do not influence the dislocation path lengths in NaCl, KCl, and LiF doped by Mn. Reference 37 presents the A value of about 1.6×10^{-2} T for Mn impurities in NaCl and KCl. If $\Delta g \sim 10^{-3}$, then such a center may become sensitive to a magnetic field only above 8 T, which is an order of magnitude larger than the fields used in ref 10.

V. Conclusions

Progress in the theory of plastic properties of crystals is accompanied by a gradual deepening of the level at which various dislocation events are considered. At the initial stage dislocations were treated within the framework of the elasticity theory of continuous media. Then a progress was achieved by accounting for the discrete atomic structure of crystals. This structure results in a periodic Peierls−Nabarro barriers for gliding dislocations, in formation of dislocation kinks when transferring between neighboring valleys of the potential landscape, in the appearance of Bordoni peaks in the internal friction of dislocations, and so on.[28] To explain an increase of plasticity caused by a superconducting transition, the introduction of an interaction between a moving dislocation and free conduction electrons becomes necessary.[1] The same interaction plays an important part in a decrease of the low-temperature plasticity of pure metals in a magnetic field.[2] Studies of the MPE in defect crystals indicate a necessity of accounting for spin states of electrons.[17,18] This paper demonstrates that a complete understanding of the influence of a magnetic field on the dislocation mobility in defect crystals can hardly be achieved without incorporating nuclear spin states as well.

We believe that a better understanding of the part played by magnetic nuclei in MPE can be gained if more experiments are carried out. The following experiments can be mentioned: (1) It has been outlined above that Mn impurities in alkali−halides are characterized by a high value of the crossover field, $H_A = 8$ T. It would be interesting to verify that at higher fields, $H > 8$ T, these defects would actually acquire a sensitivity to the magnetic field. (2) One may expect a resonance increase of plasticity under the action of a microwave magnetic field even if a permanent magnetic field is absent or very weak. In this case, the role of the permanent magnetic field is played by the HFI, which also lifts the degeneracy of the T states of the radical pairs. The HFI constant $A \sim 5$ to 10×10^{-4} T, obtained above, leads to the resonance in the fields with frequencies of about several tens of MHz.

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