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Deformation of MgO by Vickers microindentation tests

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Résumé. — La microdureté Vickers a été mesurée à température ordinaire dans des faces (100), (110) et (111) de MgO, en faisant tourner la diagonale de la pyramide. Les empreintes peuvent être déformées, probablement en raison de la fissuration. Des comparaisons ont été faites avec NiO et CoO, de même structure cristallographique, mais de plasticité et fragilité différentes.
Il faut être circonspect quand on considère l'anisotropie de dureté qui est généralement expliquée par la déformation plastique. L'anisotropie des rosettes de dislocations est faible, et incohérente avec celle de dureté.
Les valeurs de dureté dans les diverses faces cristallographiques de MgO, NiO et CoO ne satisfont pas un modèle unique.

Abstract. — Vickers microhardness tests have been performed at room temperature on (100), (110), and (111) MgO faces for various orientations of the pyramid diagonal. The indentations may be deformed probably because of cracking. Comparisons have been made with NiO and CoO which have the same crystal structure, but different plastic and fracture properties. One must be cautious when considering hardness anisotropy which is usually explained by plastic deformation. Dislocation rosette anisotropy is small, but inconsistent which that of hardness. The change of hardness with crystallographic faces in MgO, NiO and CoO cannot be uniquely described.

1. **Introduction.** — Hardness tests are the simplest and cheapest ones to assess the mechanical properties of materials. A large amount of data has been accumulated [1], however, their interpretation remains uncertain, due to the complexity of the stress tensor under an indentor which may induce deformation by different mechanisms e.g. elastic deformation, dislocation slip and work hardening, crack and/or twin propagation. The relation with plastic/brittle behaviour of solids is still a subject of interest [2].

Microhardness tests have often been used as a tool to calibrate the yield stress variations in crystals subjected to microstructural changes [3]. This shows that hardness values H_v can be related to dislocation mobility. Such a relation has also been used to explain the hardness anisotropy when the indenter is rotated in a crystalline face [1]. The slip systems can be deduced from the anisotropy of Knoop hardness [4] which is also observed for Vickers hardness H_v, provided that the periodicity (the lowest common multiple of plane symmetry and indenter symmetry) is not too high [5].

Such a behaviour has been observed in NiO by changing the indenter symmetry [5]; it is not so clear for tests performed in (100), (110) and (111) faces of TiC [6]. Moreover, the anisotropy for NaCl cannot be explain on the basis of $\{ 110 \} \langle 1\bar{1}0 \rangle$ slip [7].

The correlation between hardness anisotropy and plastic deformation has been calculated by Brookes assuming that dislocations glide under a stress described by tensile forces acting parallel to the line of maximum slope in the indenter facet ([1], p. 149, p. 199). This allows to calculate an effective Schmid factor (ESF) and then should explain most of the features of H_v anisotropy. There is a number of limitations for the application of this model that we have discovered in the study of Vickers microhardness and plastic deformation of NiO, MgO, CoO [3, 8, 9]. The shape of indents may be far from square. This may be due to crack formation and propagation and may explain part of H_v anisotropy. We have also studied the anisotropy of dislocation rosette lengths revealed by etch pitting and made comparison between H_v in various crystallographic faces; the concept of effective Schmid factor (ESF) is too simple to account for all our observations.

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2. **Experimental techniques.** — Transparent MgO crystals provided by Spicer, have been used. X-ray Laue back reflection was used to cut (110) and (111) face specimens. (100) faces were obtained by cleavage. The specimens were mechanically polished and then, chemically polished in H_3PO_4 to remove surface damage; they were annealed in air at 1 200 °C for at least one day. The crystals of NiO and CoO, grown in an arc image furnace, are those used in a previous study [3, 9]; only undoped crystals were used.

A difference between H_v on (100) cleaved and polished faces was not found. However, slightly shorter dislocation rosette lengths were measured on cleaved surfaces; this may be due to an an experimental artefact or to lower dislocation mobility.

Microhardness tests have been performed using a Zeiss microscope with loads between 40 g and 200 g applied during 5 to 300 s. Dislocation etch-pitting has been performed on MgO using HNO_3 (90 °C during 15 s for (100)) and H_3PO_4 (60 °C, 60 s for (110)). The sizes of the indentation diagonals and dislocation rosettes 2 l were measured using the device attached to the Zeiss microscope. Hardness values H_v were determined from the tables in the instruction manual.

It must be noted that the measurement of the diagonal length of the indentation may depend on various factors, in particular human factors. Focussing of the microscope change the shape and the size of the image of the indentation, giving an error especially at small loads. A discrepancy of 10 % on H_v value is not surprising between two experimentalists.

A current observation is that a Vickers indentation is not square [10]; this is obviously a source of spurious H_v values. H_v is equal to the pressure of the pyramid during indentation, i.e. to the load divided by the contact surface. If the indentation is square, its diagonal gives the surface. If it is not, the diagonal gives the contact surface only if it is identical to the one during contact, before unloading; this is difficult to assess ([1], p. 453). In view of this difficulty, we have rather made an estimation of a mean surface by adjusting the square in the ocular of the microscope with the shape of the indentation. This may change H_v by 15 to 20 %, which in many cases is as large as the observed H_v anisotropy (see below Fig. 3). Dislocation rosette lengths are then more reliable physical characteristics.

3. **Results.** — 3.1 INFLUENCE OF LOAD AND TIME. — The observed indentations were sometimes surrounded by cracks which appeared systematically on (100) MgO faces [10].

The diagonals of the indentations d were measured for various loads P. Kick's law, $P = ad^2$ [11] was verified for (100), (110) and (111) faces of NiO [8], CoO and MgO. Values for H_v are independent of load; they are reported in table I. The diagonal of the indenter was paralell to ⟨ 100 ⟩ for (100) and (110) faces. The relation between H_v and the time t under load has been examined for the 3 faces in MgO; for t larger than 20 s, H_v was always constant.

Chemical etch pitting has been performed on (100) and (110) faces of MgO. Dislocation rosettes in (100) faces have their usual features; pits for screw dislocations are along ⟨ 100 ⟩ and are along ⟨ 110 ⟩ for edge dislocations ([1], p. 28). Dislocation rosette lengths 2l were measured for screw and edge bands. Etch pitting of (110) faces shows quite different rosettes (Fig. 1) in agreement with early observations [12]. After chemical etching, indentations are deformed along ⟨ 100 ⟩ and four dislocation arms are revealed. This does not allow to measure 2 l for screw and edge dislocations which are mixed in the same arms.

For both faces, 2 l was found independent of time for t larger than 30 s. The variation with load could be fitted with a parabolic law (Fig. 2).

3.2 HARDNESS ANISOTROPY. — Following the well established use [1], hardness anisotropy is the effect which results from rotating the crystal around an axis perpendicular to its surface. The orientation of the Vickers indenter is referred to its diagonal. We have studied H_v anisotropy in (100) NiO and (100), (110) and (111) MgO faces. The results are shown in figure 3, together with results from the litterature.

For NiO, our values agree well with those of [3]. They are slightly larger than those of [8] (Table I) and lower than those of [5]. The major difference with the latter is that we find no anisotropy for H_v (Fig. 3). For MgO, our results at 50 g and 200 g fall in between values from the litterature (Fig. 3), except for a curve at 50 g ([1], p. 180); this high H_v is attributed to a dependence of H_v with load ([1], p. 181) that we did not find.

Table I. — H_v values for different oxides. For MgO, we show the anisotropy of H_v according to [5].

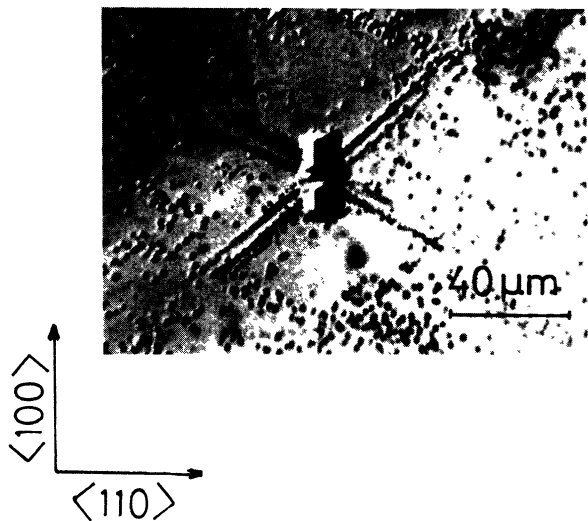| Compounds	Hardness values (MPa)			Ref.
	(100)	(110)	(111)	
NiO	4 800	7 060	6 160	[8]
CoO	2 970	2 800	2 750	[9]
MgO	7 600	7 800	9 850	This work
MgO	5 280/7 190	7 190/8 350	7 930/8 580	[5]

Fig. 1. — Scanning electron micrograph of a (110) MgO face after Vickers indentation (Load 50 g) and chemical etch pitting.
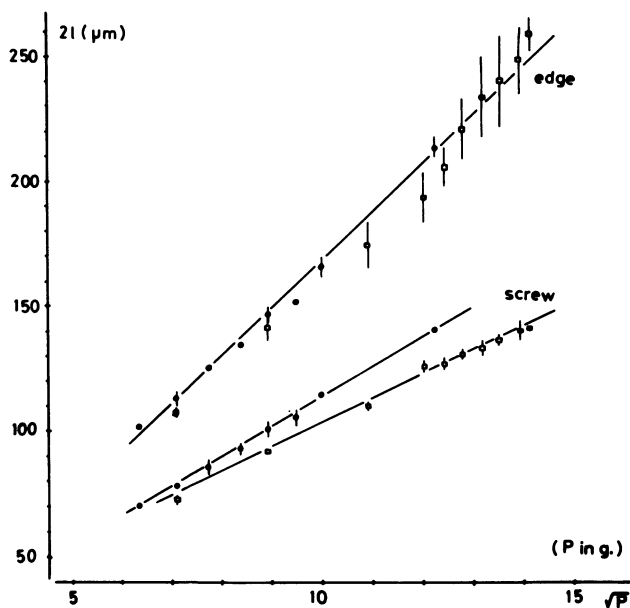

Fig. 2. — Variation of the dislocation rosette length (2 l) with square root of the load for (100) MgO face. Square points are for cleaved surface and circle points for polished one.

For (100) as well as for (110) and (111) faces, we found virtually no H_v anisotropy (Fig. 4). When we measured the indentation diagonal rather than a mean area, we found an anisotropy in H_v similar to that of the other authors; this is shown at 200 g (Fig. 3) where two curves have been plotted for the two types of measurements. This raises the question of the nature of the H_v anisotropy. Is it a characteristic of plastic properties or of experimentalist ? We believe that dislocation rosette lengths 2 l which are connected to plastic deformation, should show a more meaning-full anisotropy than H_v.
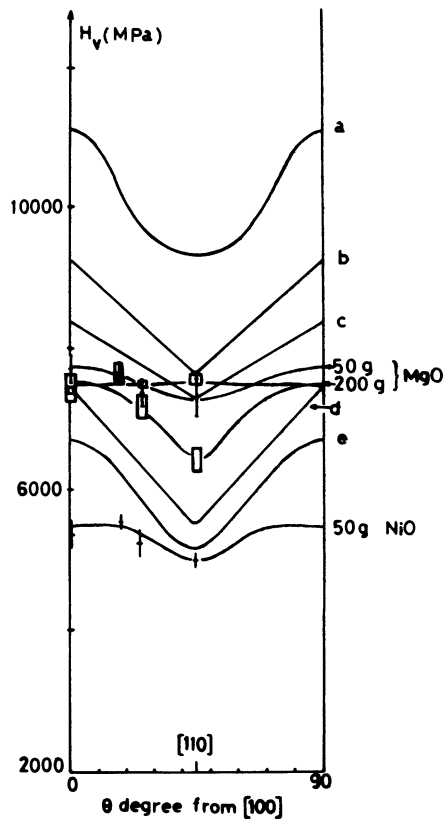

Fig. 3. — Vickers hardness (H_v) for different orientations of the pyramid on (100) faces of MgO and NiO. θ = angle between indentation diagonal and $\langle 100 \rangle$ direction. Data from the litterature : MgO; a, 50 g, ([1], p. 180); b, load unknown, [15]; c, 300 g, [10]; d, 100 and 300 g, ([1], p. 180); NiO; e, 500 g, [5]. Large open rectangles : our results, under a load of 200 g, using the two ways to measure indentation surface. Vertical bars : our results under 50 g.

3.3 DISLOCATION ROSETTE ANISOTROPY. — Etch pitting could be performed in (100) and (110) MgO (Fig. 1); we studied the anisotropy rosette length 2 l. The results are shown in figure 4 for indentation loads of 50 g and 200 g. For screw dislocations, 2 l is insensitive to rotation of indentor in (100) MgO. Edge dislocations show some anisotropy in their motion; they run a shorter distance when the indentation diagonal is parallel to $\langle$ 110 $\rangle$ (Fig. 4). No variation of 2 l can be detected in (110) MgO (Fig. 4).

3.4 HARDNESS IN VARIOUS CRYSTALLOGRAPHIC PLANES. — For MgO, we have checked that there is no H_v anisotropy when rotating the pyramid in (100), (110) and (111) (Fig. 4), an observation probably also valid for NiO and CoO (verified for (100) NiO, see Fig. 3). Values for H_v in (100), (110) and (111) faces of NiO, CoO and MgO are reported in table I. When showing a variation, the hardness is the smallest for (100) faces (Table I). However, the three oxides show very different behaviour; H_v is insensitive to the tested face for CoO, and it is the largest in (110) for NiO and (111) for MgO. Such different behaviours are unexpected for similar compounds.
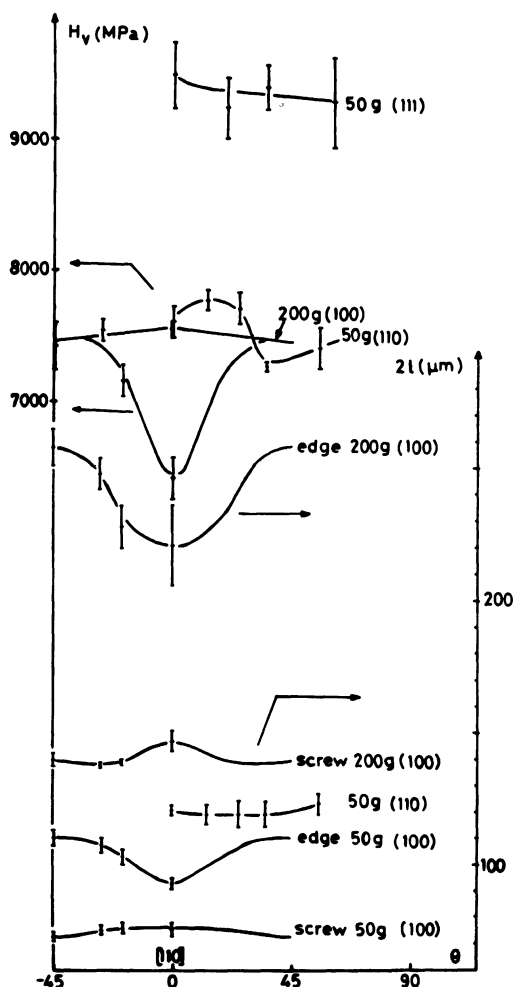
Fig. 4. — Vickers hardness (H_v) and dislocation rosette $(2 \, l)$ for different orientations of the pyramid on (100) and (110) faces of MgO; θ = angle between indentation diagonal and $\langle 110 \rangle$ direction. Also shown H_v for (111) MgO face.

4. Discussion. — 4.1 H_v MEASUREMENTS. — Because of the experimental factors, absolute hardness values cannot be obtained; for example, Armstrong [10] found that H_v can shift from 7 300 MPa to 5 300 MPa by using two different testers. Only comparisons can be made for experiments performed by the same person and the same tester.

We have measured H_v values for different loads and loading time. We have found constant H_v for NiO [8], CoO [9] and MgO. These observations are expected, but disagree with several publications for MgO ([1], p. 174 and [10]). These observations, at variance with ours, are obviously correlated to the different ways of measuring the indentation surface. Our technique seems better as it gives H_v values constant with load.

We also found H_v constant for loading times between 20 s and 300 s. The absence of indentation creep is not in agreement with ([1], p. 268-9); creep is very sensitive to environment and can be induced by humidity ([1], p. 378), an uncontrolled factor.

Microhardness anisotropy has been widely used to study dislocation slip in crystals especially in brittle ones [4]. In MgO, an unexpected opposite hardness anisotropy was found using Knoop and Vickers indentors ([1], p. 180). These opposite anisotropies are compatible with the theory of Brookes [15]. However, two other explanations have been put forward, which take into account workhardening and cracking due to multiple slip [10]. Before any such attempt, it is important to be sure that H_v anisotropy does exist. Most authors follow strictly the use which consist to calculate H_v from the diagonal length of indentation ([1], p. 3); this is without problem for perfect square indentations. As noted by Armstrong [10] for MgO, the indentation can be greatly deformed (Fig. 5). Use of the diagonal length in such cases gives H_v values by defect (Fig. 3). The deformation of the indentation (Fig. 5) can be due to three mechanisms;

a) Surface raising by plastic deformation [10]. This has also been observed in aluminium ([1], p. 457-463), giving the same pattern as in MgO; H_v values are lowered for indentation diagonal parallel to $\langle 110 \rangle$ and increased for indentation diagonal parallel to $\langle 100 \rangle$, giving an apparent H_v anisotropy.

b) Elastic recovery is often invoked to explain the deformation of the indentation. The diagonal may be either affected by this recovery [11], or unaffected ([1], p. 460). When using the indentation diagonal to determine the contact surface area, and therefore the H_v value, an unknown error is made which may affect the H_v anisotropy because the deformation of the indentation depends on the orientation of the Vickers pyramid.

c) If cracking occurs, there may be elastic opening of the crack and the diagonal length may be affected ([1], p. 461) giving uncorrect H_v.

There is no cracking around indentations in CoO (Fig. 5) which is the softest crystal under hardness testing (Table I) and the hardest under conventional uniaxial testing [9]. The relation between hardness and deformation by dislocation glide is not straightforward. The strength of NiO is between that of MgO and CoO (Table I) [9]. Irregular cracking occurs around indentations in NiO with no deformation of the indentation (Fig. 5). MgO has a low yield stress and a low failure stress. It has large cracks along $\langle 110 \rangle$ around indentations (Fig. 5) [10] which are strongly deformed for this orientation of the pyramid. It may then be dangerous to conclude that H_v is anisotropic in MgO type oxides. It is interesting to note that cracks appear along $\langle 110 \rangle$ in MgO and along $\langle 100 \rangle$ in NaCl [10] and that the Knoop anisotropies are opposite for these two compounds, suggesting a relation between fracture and hardness anisotropy.

The existence of H_v anisotropy due to plastic deformation in MgO is then questionable; it may be entirely a consequence of errors in measuring H_v values. The Brookes theory was found compatible with H_v aniso-

MgO   NiO   CoO
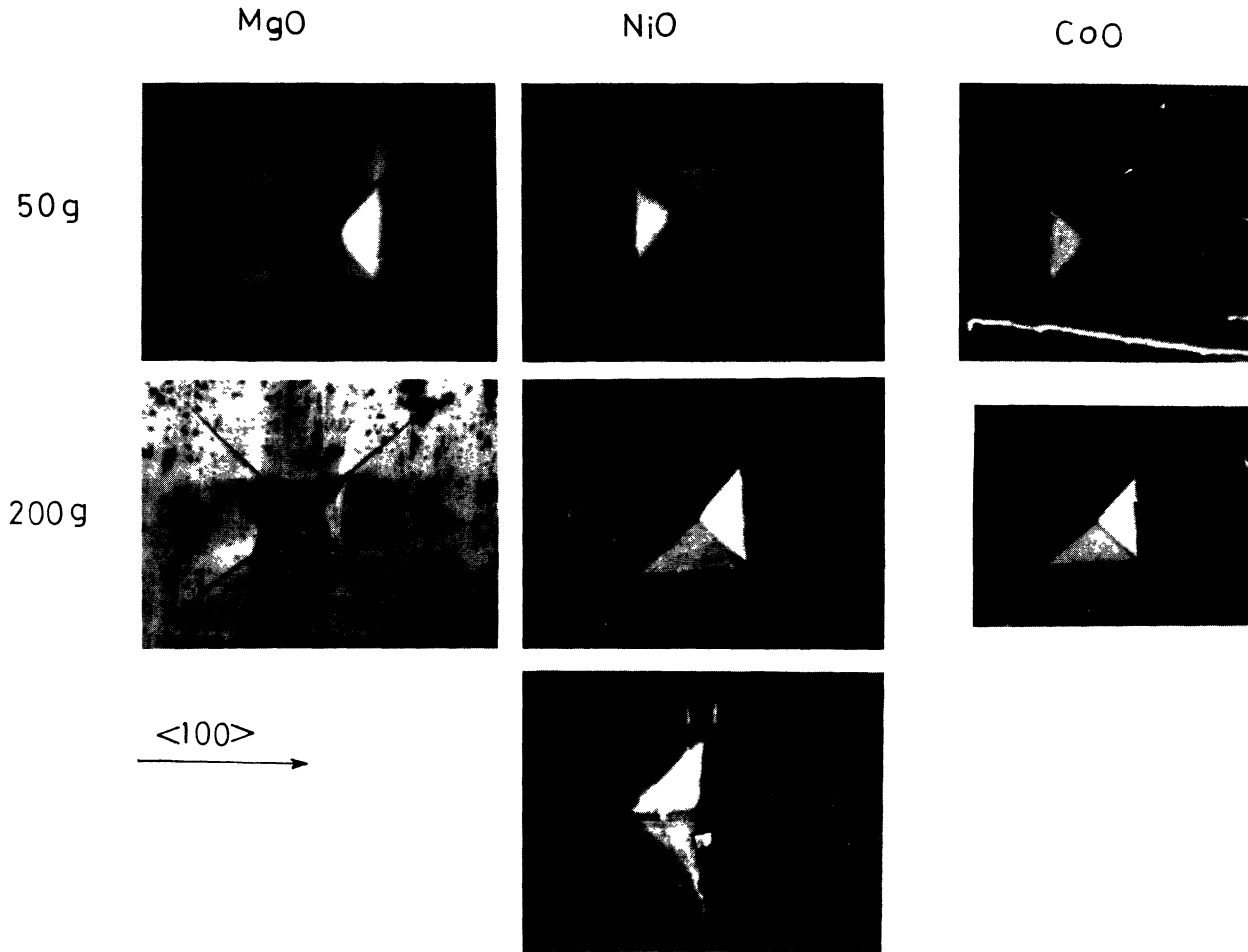
50 g

200 g

<100>

Fig. 5. — Scanning electron micrograph of Vickers diamond indentations in (100) faces for MgO, NiO and CoO. Loads 200 g and 50 g. All indentations have been magnified to the size of the one on MgO under 200 g.

tropy in MgO and NiO [5, 15]. It does not mean that the anisotropy is due to plastic deformation.

4.2 DISLOCATION ROSETTE MEASUREMENTS. — Again, the absence of water seems to preclude any variation of 2 l with indentation time ([1], p. 387), in agreement with our results.

The relation between 2 l and P is parabolic (Fig. 2). These observations are consistent with the model of Gridneva et al. [13] who show that for a dislocation velocity V proportional to τ^m (τ = resolved shear stress) ·

$$l \propto P^{m/(2m+1)} t^{1/(2m+1)} .$$

Large m values describe our results, in agreement with what is generally observed in ionic crystals [13]. In MgO around room temperature stress velocity measurements give m equal to about 10 for pure crystal by reanalysing figure 9 of [14], p. 981 and equal to 11 for doped crystals ([14], p. 990). Dislocation rosette lengths 2 l are more characteristic of plastic deformation during indentation. It is then expected that 2 l is anisotropic. It sounds sensible to believe that H_v

increases when 2 l decreases [3]. Indeed, for ionic crystals it has been observed that l/d or $l.H_v^{1/2}$ is constant (d = diagonal of indentation), a result explained by the model of Gridneva [13]. We then expect opposite anisotropies for 2 l and H_v, if any. Experimental observations are just the contrary (Fig. 4); 2 l and H_v are minimum for the indenter diagonal parallel to $\langle 110 \rangle$ (Fig. 4). There is an inconsistency between two experimental characteristics that should be related to the same physical mechanism. This, again, suggests that various competing deformation processes must be involved, which influence in different ways the H_v and 2 l values. The Brookes theory, based or dislocation slip, should then be able to explain 2 l anisotropy. However, its conclusions are not compatible with the observations (Fig. 4).

4.3 APPLICATION OF EFFECTIVE SCHMID FACTOR (ESF). — Brookes theory ([1], p. 149, p. 199) calculates an effective Schmid factor (ESF) to explain H_v anisotropy. We have used the ESF concept to try to explain the dislocation rosette observations in MgO and the variation of H_v in (100), (110) and (111) faces.

For indentation in (100) faces of MgO, we have seen (§ 4.4) that the Brookes theory, which is compatible with H_v anisotropy, is not compatible with 2 l anisotropy. Moreover, we have found an ESF smaller for these slip planes containing dislocations with an edge character at their emergence, than for these with a screw character. This is an indication that edge bands should be shorter than screw bands in the dislocation rosette, at variance with observations (Fig. 4 [1], p. 28).

The comparison of H_v in (100), (110) and (111) faces of NiO, CoO and MgO (Table I) already suggests that the use of ESF is unable to explain the observations. It is not surprising that we found maximum ESF values of 0.36 in the 3 faces allowing to explain only similar H_v. A fairly large variation of H_v with crystallographic faces is observed, except for CoO. It has to be due to deformation controlled by other mechanisms besides the primary slip system used to compute ESF. Dislocations with a great variety of Burgers vectors have been observed around indentation in MgO [16]. This is an indication that other slip systems can be activated during indentation. Cracking and/or workhardening behaviour are also probably able to explain the different behaviours reported in table I.

5. **Conclusions.** — Several inconsistencies have been found for hardness behaviour of MgO type oxides.

We do not agree that a large anisotropy of Vickers hardness exists when rotating the indenter on a (100) face although the Brookes theory, based on dislocation slip in the primary glide planes, gives a good account of it. The way to measure indentation area and the role of other deformation mechanisms may be decisive in this issue. The study of dislocation rosette lengths did not clarify the situation; they are not correlated with hardness values and did not fit in the frame of the Brookes theory.

This shows that microhardness, although an easy and cheap tool to study deformation, is still far from a full understanding. In particular, studies are generally limited to surface observation; TEM may bring usefull information on the volume under the surface [16]. Cracking, which may considerably alter the hardness measurement, is very important, in particular because hardness technique is preferrably used in brittle materials [2, 17, 18]. Complex stress tensor and multiple deformation mechanisms during hardness tests show that it is an uneasy tool to assess mechanical properties.

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