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Hardness Measurement and Evaluation of Thin Film on Material Surface

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Abstract: A method for hardness measurement and evaluation of thin films on the material surface was proposed. Firstly, it is studied how to obtain the force-indentation response with a finite element method when the indentation is less than 100 nanometers, in which current nanoindentation experiments have not reliable accuracy. The whole hardness-indentation curve and fitted equation were obtained. At last, a formula to predict the hardness of the thin film on the material surface was derived and favorably compared with experiments.

Key words: thin film material; hardness; nanoindentation; finite element simulation

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摘 要: 给出了基于纳米硬度试验的表层薄膜的硬度测算方法。首先研究如何利用有限元计算弥补纳米硬度测量在压痕深度小于百纳米时的精度缺陷, 进而探讨薄膜- 基体材料系统的硬度随压痕深度变化的规律, 最后导出了根据实验曲线预测表层薄膜材料的硬度的公式, 并进行了实验验证。

关键词: 薄膜材料; 硬度; 纳米压痕; 有限元模拟
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To meet the requirements of wear-resistance, corrosion-resistance and fatigue-resistance, the materials with a thin film on the surface have attracted a great deal of attention and have been widely used in many industrial fields such as aeronautics, astronautics, vessel and automobile engineering, *etc*. For example, heat-resistance coating technology is adopted on the blade of an aeroengine in order to improve performance and increase its service life. In practice, how to obtain the mechanical properties of thin film materials is a general problem and evaluating the hardness with nanoindentation is a simple and effective method to solve the problem[1]. However, with the development of science and technology, many advanced requirements for materials are brought forward and many films with thickness of the order of 10^2 up to 10 nanometers are made. In this case, the hardness of thin films can hardly be directly determined through indentation because of the precision limitation of the indentation equipment. In fact, the hardness obtained from experiments shows the combined action of the thin film and substrate. Therefore, how to evaluate the hardness of thin films becomes an important problem to be overcome urgently.

A lot of literature[2-5] have investigated the measurement methods of hardness of thin film materials. In general, the "10 percent rule" is a fundamental rule to evaluate the hardness of thin films; *i. e.* when the depth of indentation is less than 10% of the thickness of a thin film, the influence of the substrate on the experimental result will be less than 2%. To some extent, the "10 percent rule" is effective, but the method obviously has its limitations[6]. When the thickness of the

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thin film is less than 1μm, according to the rule, the depth of indentation should be less than 100nm and the corresponding measurement data should be reliable, which will be beyond the ability of the present experiment technology.

From the point of view of mechanics, the process of nanoindentation is very complex. In the process, the material in different places under the indenter will undergo different procedures and be in different states, which is very difficult to handle with an analytical method. Numerical simulation has attracted a great deal of attention because of its distincted advantages over the analytical method[7], which is not restricted by the depth of indentation and can probably represent the whole process.

In the present paper, the geometrical shape of the indenter is precisely simulated and the process of nanoindentation is analyzed with a three-dimensional finite element method (FEM). The loading and unloading processes of the indentation experiment, as well as the hardness-displacement curve of nanoindentation are represented through the variation of the mesh scale. For film/substrate composite, the regularity of hardness to indentation depth is discussed. The fitting hardness formulae by which the hardness of thin film materials can be predicted according to the experimental curves are proposed and verified by experiments.

1 Simulation of Nanoindentation

In the present work, a three-dimensional finite element model and Berkovich triple pyramid indenter with the tip radius less than 100nm are adopted. The loading and unloading process with gliding contact can be simulated[8].

1.1 Geometrical shape and mesh

As shown in Fig. 1, Berkovich indenter, the standard indenter of nanoindentation equipment, has a geometrical shape of a triple pyramid and the height is set to be 3μm. The whole material is divided into two parts, *i. e.*, thin film and substrate. In the present model, the size of the material is assumed to be 25 times greater than the depth of indentation, by which the boundary effect can be neglected. According to symmetry, 1/6 of the indenter and film/substrate composite is considered.

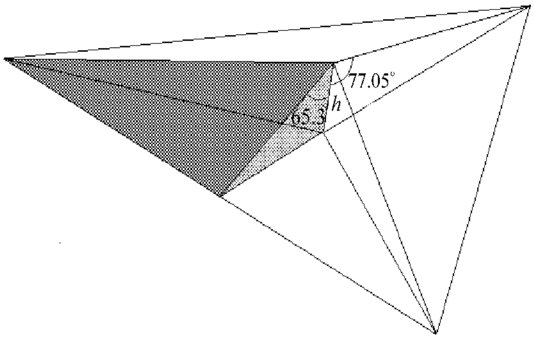

Fig. 1 Shape and parameters of Berkovich indenter

As shown in Fig. 2, a four-node-tetrahedron element is adopted. In order to guarantee calculation precision and save calculation costs, the mesh is fined locally rather than globally in the present finite element model. For instance, the size of the mesh in the contact region is smaller than that of the mesh in other regions.

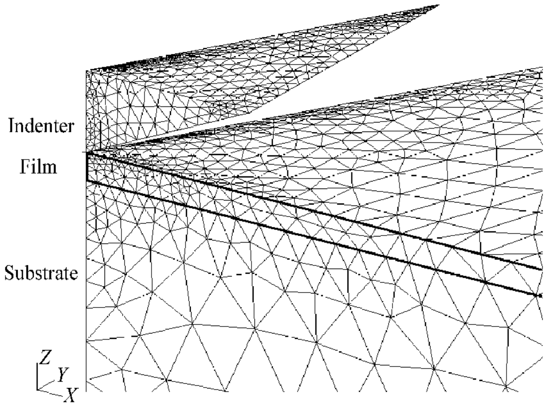

Fig. 2 Finite element model

1.2 Characteristics of material

The indenter should be handled as an elastic body because the hardness of the indenter and that of the film/substrate may be of the same order. In this paper, the Young's modulus of the indenter is set to be 1141GPa and the specimen is modeled as a von Mises solid with discrete yielding followed by linear, isotropic work hardening. Properties of the material of the specimen are shown in Fig. 3, in which E is Young's modulus, Y is yield strength, and E_T denotes hardening modulus.

The hardness of the film/substrate composite

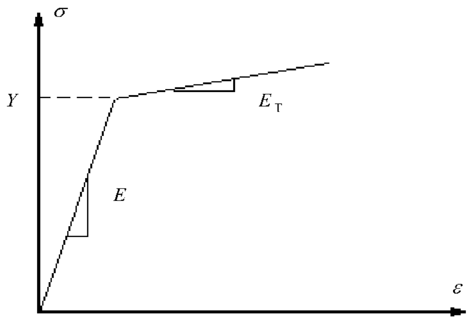

Fig. 3 Stress-strain curve of material

will vary with the depth of indentation because the properties of the film and substrate are different. To investigate the regularity of material hardness, it is the key point to analyze the influence of characteristic difference between the film and the substrate on the variation of hardness with the indentation depth. The influence is investigated through parameter combinations of the film and the substrate as shown in Table 1. In the table, Y_f and Y_s are yield strength, E_f and E_s denote Young's modulus, v_f and v_s are Poisson' ratio, and E_{Tf} and E_{Ts} represent hardening modulus, where subscripts "f" and "s" stand for film and substrate respectively.

Table 1 Yield strength ratios and Young's modulus ratios of film to substrate in models

Yield strengths of films Y_f/MPa	10	20	50	200	500	1000	1500	3000
Y_f/Y_s	0.1	0.2	0.5	2.0	5.0	10.0	15.0	30.0
Young's moduli of films E_f/GPa	25.6	64	256	384	640			
E_f/E_s	0.2	0.5	2.0	3.0	5.0			

(Y_s = 100MPa, E_s = 128GPa, $v_f = v_s$ = 0.3, $E_{Tf} = E_{Ts}$ = 2.6GPa)

1.3 Experimental verification of FEM results

To verify the reliability of the finite element model, Ti/Si material, a typical soft film on hard substrate, is selected as experimental material of nanoindentation. For the material, the hardness of the film is approximately equal to 1/3 of that of the substrate. The comparison of FEM and experimental results is shown in Fig. 4, which indicates that the results calculated by the present model are excellently in good agreement with the experimental ones. In addition, many times of experiments are done and the results show good repeatability. To be clear, only two experimental curves are given in Fig. 4.

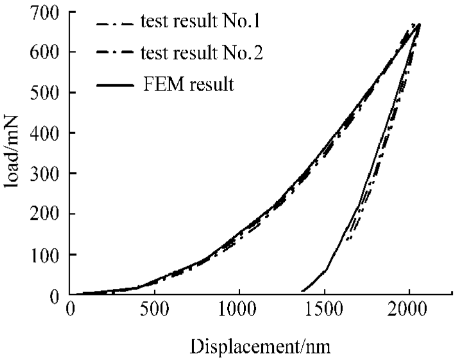

Fig. 4 Comparison of FEM and experimental results

FEM mesh scale can be controlled and then the hardness of the material can be calculated for nanoindentation. In general, the finite element analysis is not affected by the absolute size of the mesh, and the advantage is taken to investigate the variation of hardness when the depth of indentation is less than 200nm in the present study.

2 Regularity of Hardness Variation of Film with Indentation Depth

Sometimes, the thickness of a thin film is only of the order 10^2 up to 10 nanometers. In this case, the real hardness of the film can be obtained only when the depth of indentation is very small. Confined by the precision of the present indentation, the hardness of the material cannot precisely be determined when the depth of indentation is less than 200nm, especially 100nm.

Fig. 5 shows the hardness-displacement curves obtained by indentation for Ti/Si material. It can be seen that all the curves fluctuate severely when the depth of indentation is less than 150nm. This

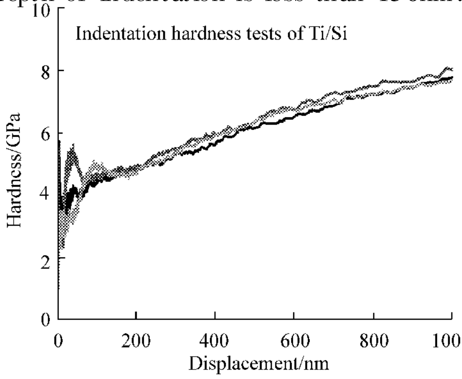

Fig. 5 The hardness-displacement curves obtained by indentation for Ti/Si material

indicates that the hardness of the thin film cannot be obtained by experiment.

To investigate the relationship of the hardness of the composite with that of the film and substrate, material models of the composite, film and substrate are independently established to calculate their hardness respectively. Fig. 6 gives the hardness-displacement curves, in which curve 1 reflects the film, curve 2 represents the composite, and curve 3 denotes the substrate. It can be seen that curves 1 and 3 vary flatly and straightly, which is in accordance with the previous research[9]. Curve 2 locates between curve 1 and curve 3, and it matches curve 1 for small indentation because the hardness is mainly determined by the film rather than by the substrate.

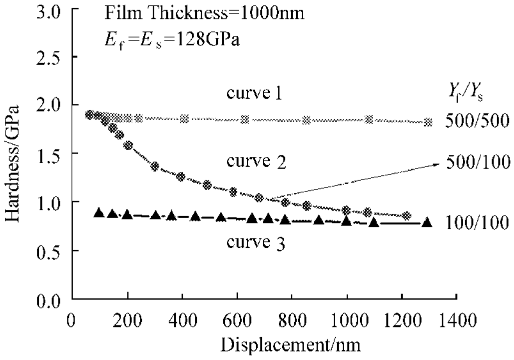

Fig. 6 The hardness-displacement curves obtained with FEM

3 Hardness Evaluation of Film Material

When the thickness of the film is less than 1μm, the hardness cannot precisely be determined by nanoindentation. In the present work, the load-displacement curve of nanoindentation is simulated with FEM, and the main factors that affect hardness are investigated. A hardness formula, by which the hardness of thin film material can be predicted according to the experiments, is derived by investigating a great deal of data of film/substrate composites.

3.1 Influences of material parameters on results

The load-hardness curve by which the mechanical properties of material can be represented is the basic result of indentation. In this paper, the influences on the results of the yield strength, Young's modulus, hardening modulus and Poisson's ratio of the film are analyzed and the main parameters are determined.

Different load-displacement curves can be obtained by changing the yield strength ratio of film to substrate shown in Table 1. Fig. 7 gives the curves when the yield strength ratio Y_f/Y_s changes. It can be seen that the yield strength ratio is a smart parameter that makes influence on the loading process.

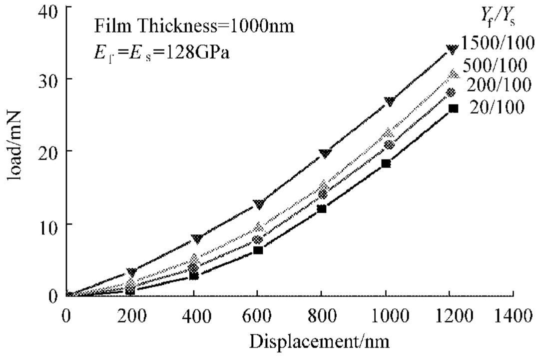

Fig. 7 Load-displacement curves of different yield strength ratio

The influence of the yield strength and Young's modulus of the film on the loading curve is shown in Fig. 8. It can be seen that the influence of Young's modulus is far less than that of the yield strength when the thickness of the film is small. In this case, the influence of Young's modulus on the results can be neglected.

Hardening moduli of many materials are far less than their Young's moduli, and the ratio is set to 0.0– 0.1 in the present work. Fig. 9 shows the

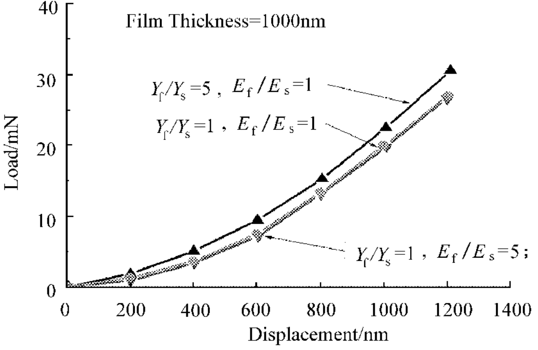

Fig. 8 Comparison of influence on load-displacement curves of Young's modulus and yield strength

influence of the hardening modulus ratio of the film to the substrate on the results. As shown in the figure, the influence of the hardening modulus of the film on the loading curve is insignificant. When the hardening modulus of the film increases by 5 times, the variance corresponding to the maximum displacement is no more than 10%. Therefore, the influence of the hardness modulus on the results is neglected in this paper.

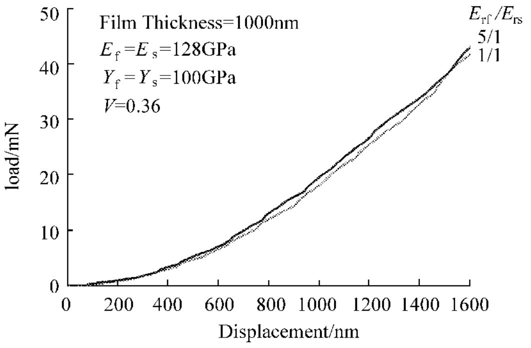

Fig. 9 Load-displacement curves of different hardening modulus ratio

In addition, previous research indicated that the influence of Poisson′ ratio on the results is very small, which is verified by the present work. In view of this, the influence of Poisson′ ratio is also neglected in the present analysis.

3.2 Fitting formula of hardness-displacement curves

As mentioned above, the hardness of a thin film cannot be directly obtained through indentation because of the limitations of experimental precision, so an effective method that can determine the hardness according to hardness tests is to be derived. In this paper, how to figure out the hardness of a thin film by fitting the hardness-displacement curve of the film/substrate composite according to the experimental data when the depth of indentation is large enough is investigated.

Figs. 10(a), (b) and (c) are the fitting hardness-displacement curves of the film/substrate. The data required to be acquired from indentation and the data provided by FEM are shown in the figures.

By fitting the results obtained through a great deal of parameter combinations, there is

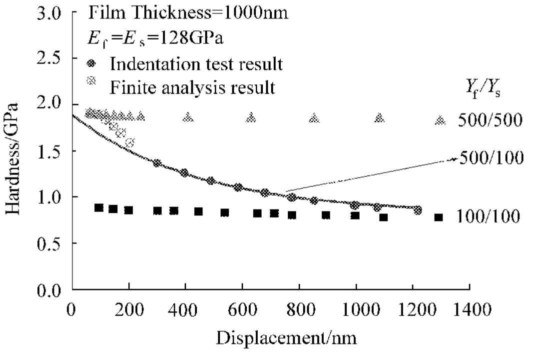

Fig. 10(a) The fitting curve when Y_f/Y_s is 5∶1

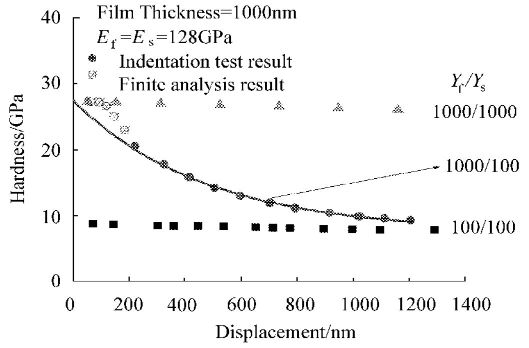

Fig. 10(b) The fitting curve when Y_f/Y_s is 10∶1

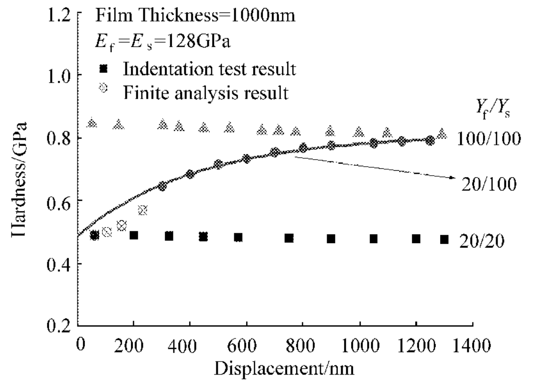

Fig. 10(c) The fitting curve when Y_f/Y_s is 1∶5

$$H = H_s + (H_f - H_s)\exp[-\gamma h/t] \quad (1)$$

where H_f and H_s are hardness of the film and substrate, and h and t are the depth of indentation and the thickness of the film, respectively. H is the measured hardness while the depth of indentation is h.

As shown in Fig. 10, H will tend to H_f when h approaches 0, *i.e.* the hardness calculated by Eq. (1) will approach the hardness of the film along with the reduction of the indentation depth.

3.3 Hardness of film

In Eq. (1), let $\gamma = 2.2$. Then

$$H = H_s + (H_f - H_s)\exp[-2.2h/t] \quad (2)$$

From Eq. (2)

$$H_f = H_s + (H - H_s)\exp(2.2h/t) \qquad (3)$$

Eq. (3) is the formula to calculate the hardness of the film. As long as the parameters t, H_s and H are measured by experiment, the hardness H_f of the film will be known.

To reduce error, multi-point measurement should be followed and make the mean value as the final result. Assume N times of measurement are done. According to Eq. (3), there is

$$H_f = H_s + \frac{1}{N}\sum_{n=1}^{N}(H_n - H_s)\exp(2.2h_n/t) \qquad (4)$$

where h_n is the depth of nth indentation, and H_n is the corresponding hardness. In order to improve prediction precision, more than 5 points within the scope of $0.2t < h_n < 1.2t$ should be selected.

4 Experimental Verification

For film/substrate composites, as discussed above, Eq. (4) can give the hardness of the film when the film thickness t, substrate hardness H_s are known and the nanoindentation curve is acquired from experiment. In the present work, experiments are done to verify Eq. (4). In the experiments, typical Ti/Si materials are selected as the specimens, and the thickness t of Ti film is 200nm. According to indentation, the hardness H_s of the substrate is 10.5GPa. Fig. 11 gives the hardness-displacement curve obtained with experiment.

As shown in Fig. 11, the hardness of the specimen gradually falls along with the reduction of the indentation depth, which indicates that the specimen is a soft film on a hard substrate. The hardness of the film cannot be determined by experiment because the thickness of the film is only 200nm. In fact, the hardness obtained from experiments shows the combined action of the thin film and substrate.

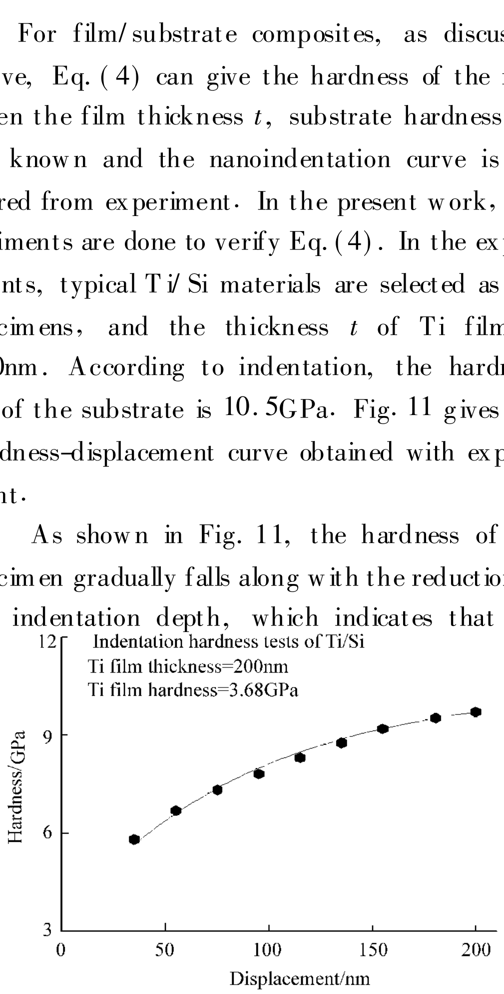

Fig. 11 Hardness-displacement curve of Ti/Si

According to Eq. (4), the hardness H_f of thin film Ti is 3.68GPa. In order to directly obtain the hardness of film Ti with indentation, a Ti/Si composite with a thick film is then selected as another specimen. The hardness of the thick film Ti directly given by nanoindentation is 4.10GPa. The relative error of the hardness calculated by Eq. (4) to the experimental result is less than 10%, which indicates that the present method is effective for film/substrate composites.

5 Conclusions

(1) For film/substrate composites, the hardness-displacement curve of nanoindentation can be simulated by FEM, which overcomes the precision limitations of nanoindentation when the indentation depth is less than 100nm.

(2) Based on the nanoindentation, the hardness of a thin film on the substrate can be effectively obtained from Eq. (4).

References

[1] Oliver W C, Pharr G M. An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments[J]. J Mater Res, 1992, 7(6): 1546- 1583.

[2] Bhattacharya A K, Nix W D. Analysis of elastic and plastic deformation associated with indentation testing of thin films on substrates[J]. Int J Solids Structures, 1988, 24(12): 1287- 1298.

[3] Sun Y, Bell T, Zheng S. Finite element analysis of the critical ratio of coating thickness to indentation depth for coating property measurements by nanoindentation[J]. Thin Solid Films, 1995, 258: 198- 204.

[4] Tsui T Y, Joost Vlassak, William D. Nix, indentation plastic displacement field: Part I. The case of soft films on hard substrates[J]. J Mater Res 1999, 14(6): 2196- 2203.

[5] Tsui T Y, Joost Vlassak, William D. Nix, indentation plastic displacement field: Part II. The case of hard films on soft substrates[J]. J Mater Res, 1999, 14(6): 2204- 2209.

[6] Takeshi Sawa, Yasushi Akiyama, Atsushi Shimamoto, and Kohichi Tanaka. Nanoindentation of a 10 nm thick thin film [J]. J Mater Res, 1999, 14(6): 2228- 2232.

[7] Kramer D E, Volinsky A A, Moody N R, Gerberich W G. Substrate effects on indentation plastic zone development in thin soft films[J]. J Mater Res, 2001, 16(11): 3150-3157.

[8] 李敏, 梁乃刚, 张泰华, 王林栋. 纳米压痕实验的三维有限元数值模拟研究, 力学学报(已接收).
Li M, Liang N G, Zhang T H, Wang L D. 3D finite element simulation of the nanoindentation process. (To be published) (in Chinese)

[9] Cheng Y T, Cheng C M. Scaling approach to conical indentation in elastic-plastic solids with work hardening[J]. J Appl Phys, 1998, 84(3): 1284-1291.

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