Continuous electrical in situ contact area measurement during instrumented indentation

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The primary tool for mechanical characterization of surfaces and films is instrumented indentation using the Oliver-Pharr data analysis method. However, this method measures contact area between the indenter and sample indirectly, thus confounding instrumented indentation tests when characterizing dynamic properties, thin films, and materials that "pileup" around the indenter. Here, we demonstrate an electrical technique to continuously measure the in situ contact area by relating nonlinear electrical contact current–voltage (*I–V*) curves to the instantaneous contact area. Using this approach, we can obtain hardness as a continuous function of applied force.

I. INTRODUCTION

Mechanical characterization of films and surfaces is an invaluable tool for ongoing research in a broad range of fields including protective coatings, electronic devices, and functionalized surfaces.[1–4] In a relatively short period of time, instrumented indentation (e.g., nanoindentation) has emerged as a dominant method that is used as frequently as traditional tensile testing—if not more often. With instrumented indentation, the contact areas between the indenter and sample are inferred from force–displacement curves (Fig. 2, inset) via the celebrated Oliver-Pharr method.[4–6] The fact that there is no independent, in situ measurement of the indentation contact area is a fundamental limitation when we recognize that the measured properties are represented in terms of the applied force and the associated contact area (i.e., units of Pa). While the Oliver-Pharr method can infer the contact area from the unloading stiffness of the testing system, the approach breaks down in applications such as thin films,[7–9] materials that pileup,[10,11] and characterization of dynamic properties. Here, we show that simultaneous electrical measurement of the in situ contact area can extend instrumented indentation to overcome these limitations when evaluating conducting samples. Indenting annealed Cu, we find the electrical *I–V* curves to be nonlinear and use them to calculate contact area and hardness continuously. This method can be extended to other areas where the Oliver-Pharr method faces difficulties: characterization of dynamic properties, indentation of thin films,[7–9] and indentation of materials that pileup.[10,11]

The Oliver-Pharr method differs from other indentation test methods. Most conventional indentation testing methods (with the exception of the Rockwell technique) define the hardness of a material such that it can be related to the applied force P, divided by the ex situ post-test measured projected indent area A. During instrumented indentation tests, the applied force and the indenter displacement h are monitored continuously. The Oliver-Pharr method allows the in situ, projected contact area to be inferred from the unloading stiffness of the system.[4–6] Consequently, images of the resulting depression are not required, and the hardness H can be more generally defined in terms of the applied force and in situ projected contact area A_c (i.e., $H = P/A_c$). In addition to defining the hardness of a material even in the absence of permanent (plastic) deformation, the elastic properties of the specimen can be measured. These features make the Oliver-Pharr method both powerful and convenient, but it is based on an elastic contact model by Sneddon[12] that applies to bulk, isotropic elastic materials during

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quasi-static unloading. The technique is applied to elastic–plastic materials by assuming a parabolic indenter tip shape,[6] a method that is justified empirically, but does not apply to materials that pileup.[10,11] Furthermore, the presence of plasticity under the indenter limits the applicability of Oliver-Pharr to the beginning of the elastic unloading of the indenter. The situation becomes even more difficult when the analysis method is extended to inhomogeneous systems such as thin films on substrates.[8,9] The continuous stiffness measurement partially mitigates the restriction on unloading,[13,14] and numerical models can provide important insight into the behavior of layered systems[15] and materials that exhibit pileup.[10,11] However, all of the limitations can be removed if we have access to a continuous measure of the in situ contact area. We find that the in situ contact area can be found via electrical measurements of the contact between a conductive indenter and a more conductive sample, even though the contact *I–V* curve is nonlinear.

During the past two decades, electrical measurements have been made during indentation tests to provide insight into deformation processes and the electrical performance of the contact. For example, stress-induced phase transformations in diamond-cubic semiconductors,[16,17] domain wall motion in piezoelectric materials,[18–20] and the current–voltage (*I–V*) behavior of a conductive diamond-metal contact have been studied.[17] However, the extension of these techniques to measure the in situ contact area has not been made because of empirical and theoretical challenges. Early research correlated the ex situ contact area with the electrical resistance of a metal-coated diamond indenter and a metal specimen.[21] This technique of contact area measurement was made possible by the Holm equation for ohmic electrical contact resistance at an asperity $R \propto \rho/a$, where ρ is the electrical resistivity and a is the contact asperity radius.[22,23] Unfortunately, conductive coatings wear away during repeated use, so harder, less conductive materials must be used for the indenter. Subsequent studies with semiconducting and moderately conductive materials such as silicon carbide[24] and doped diamond[25] revealed that the electrical properties of the indenter-sample system were more complicated than the assumed (ohmic) metal–metal junction. The presence of a surface oxide or contamination on the specimen and/or the nonmetallic nature of the indenter tip rendered the contact non-ohmic. While an electrical arc discharge between a silicon carbide indenter and sample surface improved the reproducibility of the contact resistance measurement[24] by presumably cleaning the surface of organic contamination and the native oxide, this strategy was not effective for doped diamond indenters[25] and is an unacceptable surface modification for shallow (micro- and nanoscale) indentation depths. Recent studies demonstrated that the non-ohmic contact between a doped diamond indenter and noble metal is reproducible,[17] but an interpretation of the nonlinear *I–V* response has remained elusive. Our work demonstrates for the first time that the nonlinear contact *I–V* curves of a doped diamond indenter tip on a metallic substrate can be used to continuously measure the in situ contact area and hardness of materials. Not only does this work clarify previous electrical indentation results,[17,24,25] but it also provides a solution for the limitations of the Oliver-Pharr analysis method.

II. METHODS

Our electrically coupled instrumented indentation apparatus is schematically illustrated in Fig. 1. Initial *I–V* measurements showed that the electrical contact between a conductive diamond tip and annealed Cu (Fig. 2) or Au films (reproducing results in Ref. 17) is non-ohmic. Consequently, the Holm equation cannot be used to measure contact area for conductive diamond on a metallic substrate, and an alternate methodology must be pursued. We chose annealed Cu to develop the new method, because it is well characterized both mechanically and electrically, and is known to be a material to which the Oliver-Pharr method applies[26]; thus, it is a suitable sample for testing the efficacy of continuous in situ contact area measurement. Subsequent indentation tests were then conducted in a force-controlled mode, loading up to a maximum force P_{max} over a 100 s period, followed by a brief (20 s) hold at P_{max}, and an identical unloading rate to zero force. Each second during indentation we performed a linear staircase voltage sweep from −10 V to +10 V via 20 equal steps and recorded the force, displacement, and current. Thus, for each indentation test we recorded a single *P–h* curve and a series of *I–V* curves synchronously.

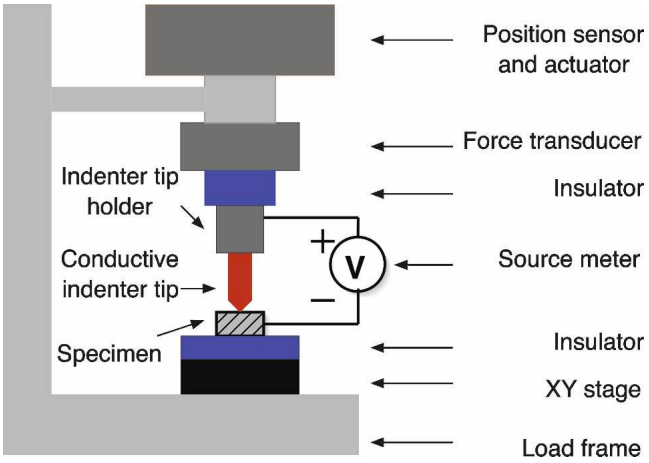

FIG. 1. Schematic illustration of the experimental setup. The actuator, force and displacement sensors, and configuration of the electrical measurement are shown.

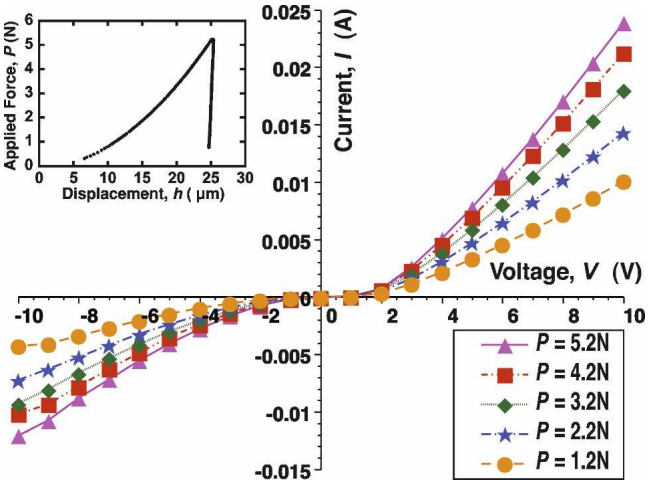

FIG. 2. Typical experimental results from indentation of annealed copper. The inset shows an applied force–displacement (*P–h*) curve for a maximum force of 5.2 N. The main graph shows contact *I–V* curves taken at applied forces ranging from 1.2 N to 5.2 N recorded during the same indent.

The electrical coupled instrumented indentation apparatus (Fig. 1) is a modification of an Instron 5848 microtester (Norwood, MA) that enables simultaneous electrical measurement during instrumented indentation. The microtester has a stiff load frame, a force transducer, and a displacement sensor. To run reliable electrical measurements simultaneously with instrumented indentation, the following changes were made. First, a conductive Berkovich indenter tip (Micro Star Technologies, Huntsville, TX) was used that had a nominal resistivity of 0.04 Ω·m; thus, electrical conduction occurred when applying voltage over the contact between the tip and a conducting sample. Second, a Keithley 2400 source meter (Cleveland, OH) was connected to the system for electrical measurements. The negative test lead of the source meter was connected to the sample, and the positive lead to the top end of the conductive indenter tip through a custom indenter tip holder. The indenter tip holder isolated the tip from the rest of the apparatus, while keeping the electrical connection to the top end of the indenter tip mechanically stable. Two electrical insulators isolated the electrical measurement circuit from the force transducer, the displacement sensor, and the Instron load frame. Finally, a custom sample stage provided accurate positioning and easy handling of samples.

Copper alloy 101 (oxygen-free, 99.99% pure) samples were annealed at 550 °C for 3 h in a vacuum chamber, mechanically polished to 0.05-μm finish, and cleaned with acetone and isopropyl alcohol prior to testing. The experimental control and data acquisition for the tests were automated through a customized LabVIEW program (Austin, TX). The spacing between any two indents was at least 10 times the lateral extent of the indent, and was controlled manually via a two-axis stage that was mounted on the load frame.

III. RESULTS AND DISCUSSION

We performed ten indents on annealed Cu for each of five conditions with maximum forces that ranged from 1.2 N to 5.2 N. Typical *P–h* and *I–V* curves are shown in Fig. 2 (the inset shows a *P–h* curve from an indentation with a maximum force of 5.2 N), with *h* corrected for initial penetration depth and machine compliance.[27] More linear and symmetric *I–V* curves were reported by Ruffell et al.[17] (and reproduced in our laboratory) for indentation of Au using a conductive diamond tip, perhaps because Au does not form an oxide layer on the surface and has a higher work function than Cu. The nonlinearity and asymmetry in the *I–V* curves might be due to nonuniform doping level and defect density distribution in the conductive diamond tip, and/or the oxide layer that forms quickly on the surface of annealed Cu when exposed in air.[28] In contrast to the observations of Goldsmid et al.,[25] the *I–V* curves that we measured during our instrumented indentation tests were nonlinear, asymmetric, and "reproducible."

Regardless of the origins of the nonlinear *I–V* response of the conductive diamond-annealed Cu contact, we have correlated them with the in situ contact areas. As shown in Fig. 2, the current through the contact increased monotonically with the applied force. Intuitively, we expected the area under the *I–V* curves, denoted Γ, to be correlated with indentation contact area A_c. This correlation in turn provided a way to calculate contact area from contact *I–V* measurements. We characterized each contact *I–V* curve by adding the absolute values of the integrated areas under the curve at positive voltages (from 0 to +10 V) and negative voltages (from −10 V to 0 V), and we found that the random variations in Γ at the same applied force were less than 3%. In addition, for *I–V* curves corresponding to the same value of Γ from our 50 indentation tests, the variations in current at the same voltage were averaged to be less than 9% including point-to-point variation. These not only indicated that electrical measurements were reproducible, but also that Γ was a good representation of an *I–V* curve and thus could be used to calibrate the contact area. It should also be noted that other characterizations of the contact *I–V* curve might be equally well suited for this purpose, but the absolute area is simple and effective.

To obtain the in situ contact area from online contact *I–V* measurements during instrumented indentation, we needed to know how Γ and contact area A_c were correlated. To find this relation, we used the Oliver-Pharr method to find A_c at the maximum force for our 50 indentation experiments. We then calculated Γ at the constant, maximum force after the hold period by numerically integrating the corresponding contact *I–V* curve at positive and negative voltages, respectively, using Simpson's rule,[29] and adding the absolute values of

the two integrated areas. Figure 3 shows contact area A_c versus Γ from all 50 experiments. A least square power-law fitting function to the (Γ, A_c) data was found to be

$$A_c = (5.279 \pm 0.354) \times 10^{-7} \times \Gamma^{1.754 \pm 0.028} \quad , \quad (1)$$

where A_c was in m^2 and Γ was in A-V. Equation (1) was only valid for Γ values between 0.0483 and 0.134 (applied forces between 1.2 N and 5.2 N, respectively), because the curve fitting was based on data in this range. The reasons that we chose a power-law function to fit (Γ, A_c) data were twofold. First, Γ has to be zero when the contact area A_c is zero, which is accomplished through a power-law fit. Second, for an ohmic contact, the relation between A_c and Γ obeys the form $A_c \propto \Gamma^2$. We made the reasonable assumption that some similarity exists between an ohmic contact case and a non-ohmic contact case, which leads to a power-law function $A_c \propto \Gamma^n$, for the non-ohmic contact case. Interestingly, the exponent found in our fitting function [Eq. (1)] was comparable to the exponent in an ohmic contact case (1.754 and 2, respectively).

Using the empirical power-law fitting function [Eq. (1)], we continuously determined the in situ contact area during loading up to P_{max} at a given applied force by measuring the contact *I–V* curve, and thereby found hardness as $H = P/A_c$. Figure 4 shows results from the 40 tests (2.2 N $\leqslant P_{max} \leqslant$ 5.2 N) and the hardness from the Oliver-Pharr method for all 50 tests. Experiments with $P_{max} = 1.2$ N have been omitted because of the Γ validity restrictions. From Fig. 4, we found that in situ hardness H decreased from 0.56 GPa to 0.36 GPa with applied force between 1 N and 5.2 N. The values match well with reported Vickers hardness, 50 kg/mm^2.[30] The estimated errors in in situ hardness averaged 2.4%. Error estimates of hardness on the plot were obtained by using

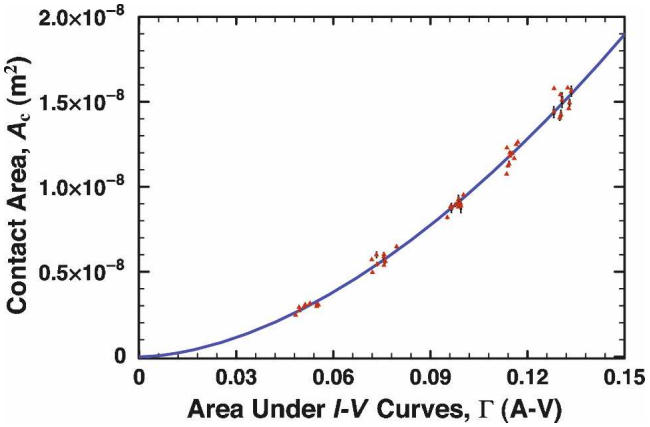

FIG. 3. Contact area (A_c) versus absolute area under *I–V* curve (Γ) with power-law fitting function Eq. (1). A strong correlation between the absolute area under *I–V* curve and the contact area measured from the unloading compliance was observed.

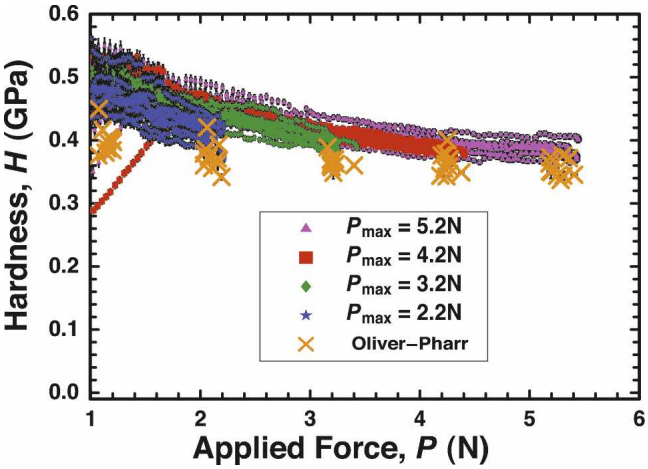

FIG. 4. Hardness versus applied force from loading curves of indents with P_{max} ranging from 2.2 N to 5.2 N. Hardness values from Oliver-Pharr from all 50 indents are shown for comparison.

the Bootstrap method[29] and a standard error propagation technique.[31] Instead of the single hardness value found from a *P–h* unloading curve using the Oliver-Pharr method (denoted by the yellow "×" symbols in Fig. 4), our analysis method of an electrical coupled instrumented indentation test gives hardness as a continuous function of applied force from the loading (or unloading) portion of *P–h* curves. These results are similar to those available from the continuous stiffness measurement method[6] that use the unloading portion of *P–h* curves; however, unlike the continuous stiffness measurement method, our technique can be applied in cases where the Oliver-Pharr method is problematic.

The trend of decreasing hardness with increasing applied force observed in Fig. 4 and the method of characterizing the contact *I–V* curve warrant further discussion. The trend in hardness shown in Fig. 4 was believed to be due to the indentation size effect,[32] which was confirmed by the mean hardness from the Oliver-Pharr method. However, the magnitude of the size effect was enhanced in the hardness calculated from the electrical measurements. This enhanced size effect needs to be further explored to determine whether it is a dynamic effect or an artifact of the analysis method. Additionally, other methods of characterizing *I–V* curve may work equally well as we previously mentioned. We tried three other methods to calculate Γ: (i) integrate *I–V* curve from 0 V to 10 V, (ii) integrate *I–V* curve from −10 V to 0 V, and (iii) integrate *I–V* curve from −10 V to +10 V. The first two methods (i.e., integrating to find the area under the positive and negative portions of the *I–V* curve, respectively) delivered similar results as presented in this article, while the third method resulted in qualitatively different results. Integrating over the complete *I–V* curve from −10 to 10 V led to an unexpected, apparent load-rate-dependent softening. While the third analysis strategy

appears to capture important features of the rate of the contact area change, the results are less robust from a numerical analysis perspective. These different methods of calculating Γ will be further explored and reported in future publications.

IV. CONCLUSIONS

In this work we have demonstrated that the in situ contact area during instrumented indentation of conducting materials can be measured continuously via the nonlinear (non-ohmic) contact *I–V* curve. Our progress followed on previous work where electrical measurements during instrumented indentation were made but not used for contact area measurement,[16–20] and where contact resistance had been assumed to be ohmic and then correlated with ex situ contact area measurements.[24,25] We used our technique to indent annealed Cu and made four important observations.

(1) The contact *I–V* curve for a conductive diamond-annealed Cu system was nonlinear and asymmetric, making analysis of contact area via Holm's equation impossible (Fig. 2).

(2) At the start of unloading, the absolute area under the *I–V* curve correlated well with the in situ contact area, and this correlation was a power-law function within the studied range (Fig. 3).

(3) In situ contact area and hardness *H* were obtained as a continuous function of applied force, and in situ hardness decreased with applied force during loading, reflecting an indentation size effect (Fig. 4).

(4) Other measures of integrated area of the *I–V* curve can be correlated with the in situ contact area and its rate of change.

As diamond-metal contacts are frequently non-ohmic and dynamic response requires in situ contact area measurements, our technique opens doors for many applications of quantitative instrumented indentation, namely materials that pile up, thin films, dynamic mechanical studies, and perhaps others. Moreover, direct access to contact area may expand our notions of mechanical measurement; structure in specimens such as film-substrate interfaces will shift from being a confounding factor in elasticity and plasticity studies to a subject of study itself via instrumented indentation.

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REFERENCES

1. B. Bhushan and X. Li: Nanomechanical characterisation of solid surfaces and thin films. *Int. Mater. Rev.* **48,** 125 (2003).
2. S.J. Bull: Nanoindentation of coatings. *J. Phys. D: Appl. Phys.* **38,** R393 (2005).
3. A. Gouldstone, N. Chollacoop, M. Dao, J. Li, A.M. Minor, and Y.L. Shen: Indentation across size scales and disciplines: Recent developments in experimentation and modeling. *Acta Mater.* **55,** 4015 (2007).
4. G.M. Pharr and W.C. Oliver: Measurement of thin film mechanical properties using nanoindentation. *MRS Bull.* **17,** 28 (1992).
5. W.C. Oliver and G.M. Pharr: An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments. *J. Mater. Res.* **7,** 1564 (1992).
6. W.C. Oliver and G.M. Pharr: Measurement of hardness and elastic modulus by instrumented indentation: Advances in understanding and refinements to methodology. *J. Mater. Res.* **19,** 3 (2004).
7. T.Y. Tsui and G.M. Pharr: Substrate effects on nanoindentation mechanical property measurement of soft films on hard substrates. *J. Mater. Res.* **14,** 292 (1999).
8. S.M. Han, R. Saha, and W.D. Nix: Determining hardness of thin films in elastically mismatched film-on-substrate systems using nanoindentation. *Acta Mater.* **54,** 1571 (2006).
9. R. Saha and W.D. Nix: Effects of the substrate on the determination of thin film mechanical properties by nanoindentation. *Acta Mater.* **50,** 23 (2002).
10. A. Bolshakov and G.M. Pharr: Influences of pileup on the measurement of mechanical properties by load and depth-sensing indentation techniques. *J. Mater. Res.* **13,** 1049 (1998).
11. Y.T. Cheng and C.M. Cheng: Effects of "sinking in" and "piling up" on estimating the contact area under load in indentation. *Philos. Mag. Lett.* **78,** 115 (1998).
12. I.N. Sneddon: The relation between load and penetration in the axisymmetric boussinesq problem for a punch of arbitrary profile. *Int. J. Eng. Sci.* **3,** 47 (1965).
13. W.C. Oliver and J.B. Pethica: Methods for continuous determination of the elastic stiffness of contact between two bodies. U.S. Patent No. 4 848 141, July 18, 1989.
14. J.B. Pethica, R. Hutchings, and W.C. Oliver: Hardness measurement at penetration depths as small as 20 nm. *Philos. Mag. A* **48,** 593 (1983).
15. X. Chen and J. Vlassak: Numerical study on the measurement of thin film mechanical properties by means of nanoindentation. *J. Mater. Res.* **16,** 2974 (2001).
16. A.B. Mann, D. van Heerden, J.B. Pethica, P. Bowes, and T.P. Weihs: Contact resistance and phase transformations during nanoindentation of silicon. *Philos. Mag. A* **82,** 1921 (2002).
17. S. Ruffell, J.E. Bradby, J.S. Williams, and O.L. Warren: An in situ electrical measurement technique via a conducting diamond tip for nanoindentation in silicon. *J. Mater. Res.* **22,** 578 (2007).
18. V. Koval, M.J. Reece, and A.J. Bushby: Ferroelectric/ferroelastic behavior and piezoelectric response of lead zirconate titanate thin films under nanoindentation. *J. Appl. Phys.* **97,** 74301 (2005).
19. A. Rar, G.M. Pharr, W.C. Oliver, E. Karapetian, and S.V. Kalinen: Piezoelectric nanoindentation. *J. Mater. Res.* **21,** 552 (2006).

20. S. Sridhar, A.E. Giannakopoulos, and S. Suresh: Mechanical and electrical responses of piezoelectric solids to conical indentation. *J. Appl. Phys.* **87,** 8451 (2000).
21. V.R. Howes, H.J. Goldsmid, and C.A. Baird: Hardness measurement at constant depth using an indenter partially coated with a conducting film. *J. Phys. E: Sci. Instrum.* **20,** 1507 (1987).
22. R. Holm: *Electric Contacts: Theory and Applications* (Springer-Verlag, New York, 1967), p. 516.
23. R.S. Timsit: Electrical contact resistance: Fundamental principles, in *Electrical Contacts: Principles and Applications*, edited by P.G. Slade (Marcel Dekker, 1999), p. 1073.
24. L. Wieczorek, V.R. Howes, and H.J. Goldsmid: Electrical contact resistance and its relationship to hardness. *J. Mater. Sci.* **21,** 1423 (1986).
25. H.J. Goldsmid, V.R. Howes, and C.A. Baird: Measurement of hardness using a semiconductor diamond indentor. *J. Mater. Sci. Lett.* **6,** 1043 (1987).
26. Y.Y. Lim and M.M. Chaudhri: The effect of the indenter load on the nanohardness of ductile materials: An experimental study on polycrystalline work-hardened and annealed oxygen-free copper. *Philos. Mag. A* **79,** 2979 (1999).
27. A.C. Fischer-Cripps: *Nanoindentation* (Springer-Verlag, New York, 2004), p. 266.
28. M.A. Lampert and P. Mark: *Current Injections in Solids* (Academic Press, New York and London, 1970), p. 351.
29. W.H. Press, S.A. Teukolsky, B.P. Flannery, and W.T. Vetterling: *Numerical Recipes in Fortran: The Art of Scientific Computing*, 3rd ed. (Cambridge University Press, 2007), p. 1356.
30. Properties and selection: Nonferrous alloys and special-purpose materials, in *Metals Handbook*, Vol. 2 (ASM International, 1990), p. 1328.
31. J.R. Taylor: *An Introduction to Error Analysis: The Study of Uncertainties in Physical Measurements* (University Science Books, 1997), p. 327.
32. W.D. Nix and H. Gao: Indentation size effects in crystalline materials: A law for strain gradient plasticity. *J. Mech. Phys. Solids* **46,** 411 (1998).