Materials Science Forum Vols. 273-275 (1998) pp. 29-40
online at http://www.scientific.net

Future Trends: Texture Analysis for Structure-Sensitive Properties

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Keywords: X-Ray Diffraction, Structure-Sensitive Properties, Grain Boundaries, Microstructure, Residual Strain

Abstract

This paper argues the need to extend modern texture analysis to considerations beyond the conventional associations between the field of lattice orientation and the morphology of the internal structure (such as is presently accomplished in orientation imaging microscopy). What is needed for progress towards understanding the important class of structure-sensitive properties, is a further coupling with the elastic strain field. We argue that an experimental capability for examining this coupling is possible. This paper describes the advances that are required in representation, and some details of a particular instrument for 3-dimensional x-ray diffraction microscopy that could satisfy these needs.

1. INTRODUCTION

The past three decades have produced many advances in that subdiscipline of materials science which is now known as the *modern texture analysis*. On this occasion, as we celebrate the lifetime achievements and service of Professor Hans-Joachim Bunge who has been the father of this subject, it is appropriate to examine what has been accomplished. It is also appropriate to consider what remains to be done by future generations who will build upon the foundations laid by Prof. Bunge and others who felt his influence during this particularly fruitful period. It is this second end which is the main focus of this paper.

Modern texture analysis comprises three main components: (a) the set of experimental methods from which coupled data on crystallographic texture and internal structure (microstructure) are obtained, (b) the methods of systematic analysis of that data to obtain useful quantitative representations based upon that data, and (c) the systematic study of the relationships between such representations and the important properties and technological applications of materials.

During these decades of the emergence of the modern texture analysis, great progress has been achieved by estimates and bounds on that class of material properties which are known as *structure-insensitive*. Here we think of internal structure as comprising all departure of crystalline materials

from idealized, strain-free, defect-free reference crystals, including their polycrystallinity. (We shall return to this concept in section 2.) Of course, the term structure-insensitive is something of a misnomer since all properties of materials are more or less dependent upon internal structure, including the many details described by the texture, the state of dislocation and the presence of grain boundaries and interfaces. Nevertheless, in nature some properties are far less sensitive to the defect aspects of internal structure. For this reason they are termed "structure insensitive".

A familiar example is linear elasticity in polycrystalline materials. Since the atomic disturbance near grain boundaries is only a few atomic distances in width, the fraction of the material volume that is affected by the atomic disruption at interfaces can be very small in materials of ordinary grain size. It is only when the grain size begins to approach the nanometer scale that we see significant departures from the range of linear elastic behavior which is now known to be functionally dependent upon the elementary volume fractions and orientation correlations. These aspects of internal structure are the mainstream focus of modern texture analysis. For the class of macroscopic material properties that are structure-insensitive over some range of internal structure and applied stimulus (examples include linear elasticity, thermal and electrical conductivity, piezoelectric effect, the initial yield surface, some types of magnetic properties, etc.), the mature methods of modern texture analysis provide an essential framework for representation, experimental determination and for analysis.

For the second category of materials properties ... those which are *structure-sensitive* ... a substantial expansion of the modern texture analysis will be required if progress is to be made. Structure-sensitive properties often depend upon the presence of grain boundaries or interfaces, upon the distribution of types of these defects, and upon the details of their connectedness in the semi-continuous network of interfaces. Examples include intergranular fracture and corrosion and various kinds of embrittlement phenomena. Other structure-sensitive properties, such as strain hardening, depend upon the details of the local strain distribution, the dislocation structure and the evolution of these with deformation. Still others, such as intergranular stress corrosion cracking, seem to depend upon all of these aspects of the defect state. It is within the context of this structure-sensitive category of properties that many of the most challenging materials problems of our generation must be addressed; and it is in this direction that we focus the remarks of this paper.

The paper asks the following two questions: Can the modern texture analysis be extended to experiments, representations and analysis that can reveal the defect structure of polycrystalline materials ... knowledge that could help solve the problems associated with structure-sensitive properties? If so, what advances should be pursued to realize the greatest possible impact? We are convinced that the answer to the first of these questions is an emphatic yes! Modern texture analysis can and should maintain a key role in solving the technological problems associated with structure-sensitive properties. The answer to the second question is explored in this paper.

We shall first describe an extended framework for the representation of mesostructure in polycrystalline materials. Then, we describe the concept of the 3-D x-ray microscope, as it is being developed now, and as it may evolve in the next decade. It is illustrated that such an instrument has the potential to more fully realize the advanced representation of mesostructure that is required to address structure-sensitive properties of polycrystalline materials.

2. AN EXTENDED FRAMEWORK FOR THE REPRESENTATION OF MESOSTRUCTURE

So varied is the internal structure of polycrystalline materials, at the wide range of length scales over which it occurs, that it is not possible to describe all aspects of that structure by mathematical representation. In fact, this may not even be desirable since we are often far more interested in the behavior of an ensemble of polycrystalline samples, rather than any particular element of that ensemble.

Here we shall present a particular mathematical representation which makes useful connections with the available methods of localized diffraction which are emphasized later in the paper. This particular representation makes contact with certain aspects of the atomic structure of crystalline materials, but it remains fundamentally detached from atomistics. As such it is distinctly associated with internal structure at intermediate length scales (mesoscales). We shall refer to such as *mesostructure*. The framework is sufficiently advanced that it facilitates consideration of potential future developments in the methods of local diffraction and their analysis.

In that which follows, we shall assume that the material of interest is comprised of a single phase. For this phase it is convenient to define a *strain-free reference crystal*. All departure from this reference crystal is attributed to some aspect of the internal structure. (The extension of these ideas to polyphase materials requires additional reference crystals.) The strain-free reference crystal can be described in a suitable global coordinate frame by fixing a (strain-free) reference lattice, L^o, with its basis vectors $(\vec{a}_1^o, \vec{a}_2^o, \vec{a}_3^o)$. The strain-free reference crystal obtains from L^o by decorating each lattice point with a basis comprising one or more atoms (molecules) arranged in identical fashion (the motif). Six numbers, called the reference lattice parameters, specify the metrics of the reference lattice. Three of these specify the lengths of the basis vectors, and the remaining three fix the angles between them.

Consider the set of all linear transformations of L^o defined by a (microscopic) translation $\vec{p}$ and the action of a second-order tensor, F, on the basis vectors of the reference lattice, according to the relation

$$\vec{a}_i = F\vec{a}_i^o + \vec{p}. \qquad (1)$$

F is called the *deformation gradient tensor*. Both F and $\vec{p}$ are considered to be functions of material position $\vec{x}$ and time t in the body of the polycrystal:

$$F = F(\vec{x},t),\ \vec{p} = \vec{p}(\vec{x},t). \qquad (2)$$

Transformations of the type defined by (1) preserve material lines and planes as lines and planes, while altering the angles between lines and planes, and their lengths and interplanar spacings, respectively.

The new (local) basis vectors $(\vec{a}_1, \vec{a}_2, \vec{a}_3)$, which define the local lattice L, are described by Eq. (1). These are fully fixed by a set of six local lattice parameters and an *orientation* of the local lattice

(three additional parameters) for a total of nine. The separation of the orientation and lattice parameters obtains naturally by the *polar decomposition* of F according to

$$F = RA. \tag{3}$$

Here $R \in O(3)/\Gamma$, the set of all physically-distinct orthogonal tensors of second-order, is also known as the *fundamental zone.* (Γ is the symmetry subgroup of the reference crystal, and $O(3)/\Gamma$ is the set of all left cosets of Γ in the three-dimensional orthogonal group $O(3)$. For the reader unfamiliar with the theory of continuous groups and symmetry effects the work of Bröcker and tom Dieck [1] is quite accessible.) $A \in T_2^{s,pd}$, the set of all symmetric, positive-definite second-order tensors.

The central preoccupation of the modern texture analysis has been with the orientation R, but with a particular restriction which comes naturally in diffraction experiments (Friedel's Law). Orthogonal tensors may have $\det R = \pm 1$. However, conventional diffraction experiments are not able to distinguish centers of symmetry associated with the *handedness* of crystals. Thus, orientations with determinant +1 are indistinguishable from those with determinant -1. For crystals possessing a natural inversion center this is not a problem since both orientations represent the same crystal orientation; but when this is not the case diffraction leaves us with an ambiguity between right-handed and left-handed crystals. Thus, the fundamental zone of central interest to modern texture analysis has been $SO(3)/\Gamma$, where $SO(3)$ represents the group of *special* orthogonal tensors with determinants of +1. Hereafter the word *orientation* will be used in this restricted sense.

The symmetric, positive-definite tensors $A \in T_2^{s,pd}$ establish the changes in the six lattice parameters associated with F. It is known that for polycrystalline materials which deform plastically, A can be written as

$$A = I + \varepsilon, \tag{4}$$

where ε is the well-known symmetric infinitesimal strain tensor [2]. (Each of the individual components of ε is small compared to unity.) For such materials only those strains associated with (Cauchy) stresses, σ, lying within the yield surface, $\sigma \in \mathrm{Y}$, of the crystal, can be considered. Thus, $\varepsilon \in E \subset T_2^{s,pd}$, where

$$E = \left\{ \varepsilon : \sigma = C\varepsilon \in \mathrm{Y} \subset T_2^{s,pd} \right\}. \tag{5}$$

In Eq. (5) C denotes the fourth-order elastic stiffness tensor. For crystalline phases obeying Schmid's law for yielding, the elastic region, Y, can be expressed as

$$\mathrm{Y} = \left\{ \sigma : \hat{\mathrm{m}}^{(s)} \cdot \sigma \hat{\mathrm{n}}^{(s)} < \tau_c^{(s)} \text{ for all slip systems} \right\} \tag{6}$$

Here $\hat{m}^{(s)}$ and $\hat{n}^{(s)}$ are the slip direction and slip plane normal directions, and $\tau_c^{(s)}$ the critical resolved shear stress associated with slip system (s).

The final part of the description expressed by Eq. (1) is the vector $\vec{p}$ which describes the translation of the reference crystal which brings it into coincidence with the local crystal. Clearly, $\vec{p}$ must belong to the set of three-dimensional vector space associated with all possible translations in space:

$$\vec{p} \in V . \tag{7}$$

Thus, in the context of Eq. (1) the specification of the *state* of the local crystal in the mesostructure requires some twelve parameters ... three fixing the orientation, six fixing the strain state, and three fixing the translation. We shall use the symbol $S(\vec{x},t)$ to denote the complete specification of the state of the polycrystal at position $\vec{x}$, and time t in the body of the polycrystal. It should be evident from the description above that

$$S \in SO(3) / \Gamma \times E \times V . \tag{8}$$

The field of local states, $S(\vec{x},t)$, lying in the neighborhood of material points at position $\vec{x}$, and time t, is the advanced representation of mesostructure that is of interest in this paper. Clearly, it does not describe any aspect of atomic scale structure. However, because it carries the local strain and translation, it is more comprehensive than previous mesoscale representations.

3. THE DIFFRACTION RELATION

For our purposes here we briefly recall the basic diffraction relation. Consider that a probing particle (electron, x-ray photon), of wavelength λ and direction of motion $\hat{s}_o$, interacts according to Bragg's law to produce a diffracted particle of unchanged wavelength and direction of motion $\hat{s}$. The Bragg relation requires that diffraction can only occur when these physical parameters associate with elements of the reciprocal lattice $\vec{q} \in reL$ (associated with L) according to the relation

$$\frac{\hat{s} - \hat{s}_o}{\lambda} = \vec{q} . \tag{9}$$

For the reciprocal lattice associated with the strain-free reference crystal we shall denote elements of the reciprocal lattice by vectors $\vec{q}^{\,o} \in reL^o$. The magnitude of the reciprocal lattice vectors is equal to the inverse of the d-spacing associated with planes in the strain-free reference crystal. For the local crystal, of arbitrary state $S(\vec{x},t)$, the local reciprocal lattice vectors are related to those of the reference crystal by the expression

$$\vec{q} = F^{-1}\vec{q}^{\,o} \equiv (I - \varepsilon)R^T\vec{q}^{\,o} . \tag{10}$$

Thus, Eq. (9) can be expressed as

$$\frac{\hat{s}-\hat{s}_o}{\lambda}=(I-\varepsilon)R^T\vec{q}^o. \qquad (11)$$

Since the basic experimental parameters (left side of Eq. (11)) are often restricted in experiments, rotations of the sample may be performed to obtain conditions which satisfy Bragg's law. Let this additional rotation be identified by the symbol R^s, not to be confused with the orientation of the crystal. In order to retain all experimental parameters on the left hand side of Bragg's relation, we shall include the effects of sample rotation by expanding Eq. (11) to become

$$\vec{q}=R^{sT}\left(\frac{\hat{s}-\hat{s}_o}{\lambda}\right)=(I-\varepsilon)R^T\vec{q}^o \qquad (12)$$

where R^{sT} is the transpose of R^s, which is equal to the inverse of R^s for orthogonal tensors.

We remark that the translational component of the local state has not been included in Eq. (12). Translations (e.g. between neighboring grains at a grain boundary) can be measured by the methods of transmission electron microscopy under rather stringent and limited conditions. (These include the necessity of preparing very thin samples.) Further consideration of transmission electron microscopy lies beyond the primary interest of this paper. However, a few remarks about translations are in order.

There is no doubt that variable translations between adjacent lattices at a grain boundary can strongly impact the thermodynamic and kinetic properties of the boundary. In particular we note that the free volume of the boundary is immediately affected by relative translations normal to the dividing plane. What is not clear, however, is just how independent the translation parameters are from the orientation and strain parameters of the participating lattices.

The application of the diffraction Eq. (12) varies from one experiment to another. However, it is clear that the question of resolving $S(x,t)\in SO(3)/\Gamma\times E$ (the three rotation and six strain parameters of the local state) depends upon satisfying Eq. (12) for a minimum of nine reciprocal lattice vectors. In principle, both electron, neutron and x-ray diffraction experiments are capable of producing sufficient resolution in reL. However, the methods of electron diffraction are rather limited since the maximum penetration of electrons into crystalline samples is on the order of 20 nanometers. Thus, the measurement of near-surface strains by electron-diffraction is severely affected by the presence of free surfaces and their concomitant, image forces. This limitation effectively restricts the range of state space that is accessible in bulk polycrystalline materials in the transmission electron microscope to $SO(3)/\Gamma\times V$. (In fact, owing to limiting spatial resolutions in the best instruments which are on the order of 0.06 nanometers, only a small subregion of V is accessible. Further discussion of these limitations is beyond the scope of interest in this paper.) For the case of electron backscattered diffraction using the scanning electron microscope, which is the basis for orientation imaging microscopy (OIM) (cf. Adams, Wright and Kunze [3]), the effective range of state space that is accessible is $SO(3)/\Gamma$. Neutron diffraction, which has excellent

penetration, could access $SO(3)/\Gamma \times E$, but is limited in spatial resolution by the flux densities and focusing technologies that are available.

The penetration power of x-ray photons is intermediate to those of electrons and neutrons. In the 40 - 100 keV energy range, which has become available at several synchrotrons in recent years, penetration depths are typically in the millimeter range. At the same time, the flux at the synchrotron is several orders of magnitude higher than at the neutron reactor. In the next section of this paper we consider the possibility of developing a microscope, based upon x-ray diffraction, which is capable of resolving local states in the space $SO(3)/\Gamma \times E$.

4. TOWARDS A 3-D X-RAY DIFFRACTION MICROSCOPY

The concept for a 3-dimensional x-ray microprobe that would examine the local strain or orientation at interior locations in bulk polycrystals has already been described [4]. An instrument is presently being developed for implementation at the European Synchrotron Radiation Facility, ESRF, in Grenoble. Plans are that the instrument will allow for several ways of defining the local probe volume, based on the use of advanced slits [5], or optical elements [4] or x-ray tracing [6]. However, none of these technical solutions are well suited to the problem at hand here: the fast, high spatial resolution simultaneous mapping of orientation and strain. We here propose an extension of these ideas, which couples the determination of local orientation and strain more efficiently.

To illustrate our proposal, the schematic of the set-up used for local orientation measurements in the work of Poulsen et al. [4] is shown in Fig. 1. The approach is quite simple. The probe volume is defined by the intersection of the narrow incoming beam - preferably focused in two dimensions - with the extension of the aligned cylindrical slit which has conical openings along the Debye Scherrer cones of relevance. The sample is rotated around an ω–axis perpendicular to the beam direction - corresponding to R^s in the diffraction Eq. (12) - in order to sample a sufficient number of reflections associated with diffraction from a particular grain. For each reflection the sample is translated until the recorded intensity is reduced to half of its maximum intensity (presumably the maximum occurs when the probe volume falls entirely inside the grain). At this location it is expected that the probe volume is bisected by the diffracting grain and other grains of the surrounding corona which do not share the same elements of reL. When this condition is achieved, the centroid of the probe volume is taken as an approximation of the location of the grain boundary. In previous work [4] the two-dimensional cross sections of several internal grains were obtained by this method. Extension of this idea to 3-dimensional characterizations is a trivial matter of extending the sample translations to 3 axes.

Thus, the feasibility for a 3-dimensional x-ray diffraction microscopy has been demonstrated. Contrast is defined by lines of misorientation in the orientation field, in the same manner as in OIM. Future development of this instrument must necessarily address, not only improvements in the spatial resolution of the local orientation field, but also advances in the temporal resolution of the instrument. These would be facilitated by robotic control of the sample translations and rotations required for mapping the internal grain structure, and a more sophisticated approach to estimating boundary positions.

A cross section of the proposed new set-up is shown in Fig. 2. The main alteration has to do with the conical slit, which is replaced with a conical array of cylindrical holes (a conical Soller slit), each of which is aligned such that its axis intersects the incoming beam at the same point. Such an arrangement could, in principle, add sufficient additional resolution in reL to enable the full application of Eq. (12) to determine, not only R, but ε. Thus, a microscopy on $SO(3)/\Gamma \times E$ would be achieved.

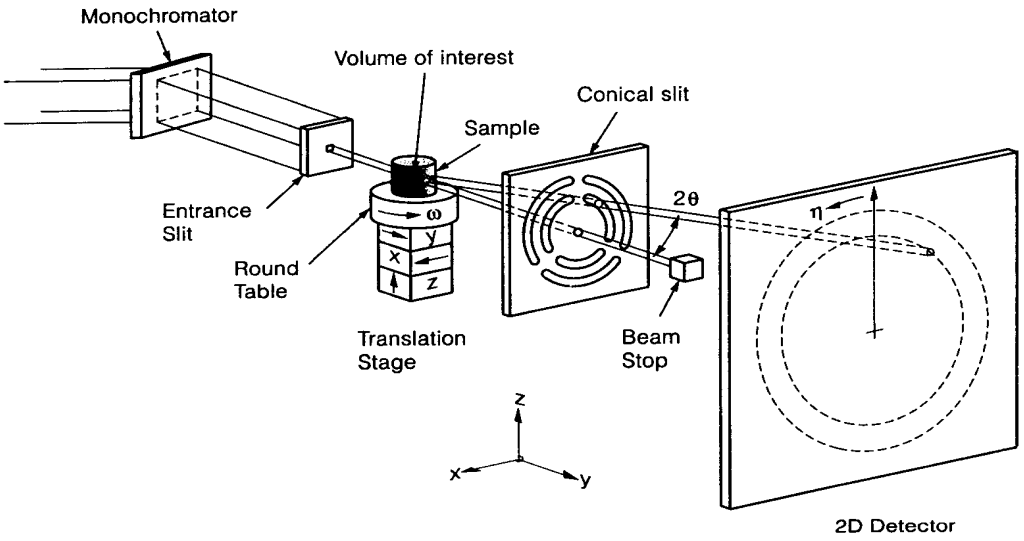

Figure 1. Schematic representation of the 3-D x-ray microscope currently under development

The realization of the conical Soller slit set-up poses several instrumental challenges. Firstly, it will be necessary to develop a conical type of lithography (or other method) capable of producing holes with an aspect ratio of 10^4 or better in materials several millimeters thick. Secondly, to determine strains with an accuracy of $\Delta\varepsilon = 10^{-4}$, while covering the full Debye-Sherrer rings, detectors have to be manufactured with $10^4 \times 10^4$ pixels or more.

Owing to the relatively high energies that are required for adequate penetrations, diffraction occurs at relatively small 2θ values. This gives rise to probe volumes that are elongated relative to the diameter of the incoming beam by aspect ratios of approximately 10 at the lowest 2θ values. Since

the probe volume is somewhat different for each cylindrical slit, it is evident that the spatial resolution of the instrument is dictated by the width of the incoming beam and by the energy of the photons, which fixes the aspect ratios of the probe volume.

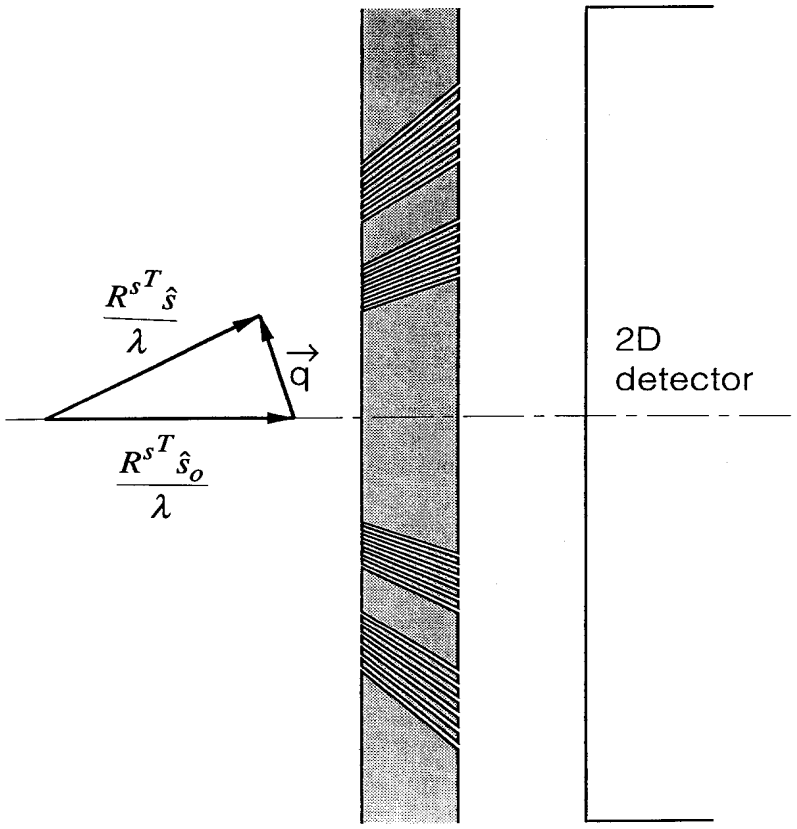

Figure 2. Cross section of the proposed conical Soller slit for resolving both lattice orientation and strain.

5. CONCLUDING REMARKS

It would be difficult to over-state the potential importance of a successful 3-dimensional microscopy which could couple the measurement of local orientation and strain with morphological information on the mesostructure. Such a microscopy could have a profound impact in accelerating the development of our understanding of the evolution of defect structure in polycrystalline materials. The present state of understanding of the coupling between the orientational and morphological aspects of internal structure, and the defect structure which governs structure-sensitive properties, is primarily viewed through the lens of model constitutive relationships. These constitutive models comprise many assumptions, and they are known to fail to adequately describe many aspects of the evolution of the defect structure. Clearly, further progress could be advanced by a microscopy which is capable of directly measuring the coupling between the defect structure and the microstructure. We suspect that without such an experimental capability the development of these vital coupling models will not be sufficiently successful to ameliorate the problems associated with structure-sensitive properties.

As an example of the importance of the coupling between orientation and strain, we mention the relationship between elastic strain, ε, lattice misorientation, ΔR, and the dislocation tensor, α, which can be expressed as:

$$\alpha = curl(\Delta R + \varepsilon). \tag{13}$$

The second-order tensor α describes the net Burger's vector $\vec{B}$ associated with a unit Burger's circuit whose normal orientation is described by $\hat{n}$ according to the expression

$$\vec{B} = \alpha \hat{n}. \tag{14}$$

In the absence of elastic strain fields, Eq. (13) reduces to that one given by J. F. Nye [7] (which ignores the ε term in (13)). Current approaches to the defect structure rely upon Nye's relation which has limited applicability given the absence of the elastic strain term. The important point, illustrated by Eq. (13), is that inference of quantitative information about the defect structure requires a coupling of orientational and (elastic) strain data.

The ultimate success of the instrument we have proposed depends upon three key developments: (I) the production of a highly intense, narrow beam of photons at the entrance of the sample, (II) precision robotic control of the sample motions required for solving relations (12) and for mapping the morphology of the internal structure, and (III) the development and manufacture of the conical Soller slit at the exit of the sample which must provide sufficient spatial and *reL* resolution.

Regarding the first requirement (I), there is considerable reason for optimism. Since 1960 the brilliance (photons/sec/mm^2/mrad^2/0.1%BW) of available sources has increased from 10^7 to approximately 10^{19}., and there is reason to believe that such progress will continue in the near future. Requirement (II) would build upon the existing library of algorithms for motion control based upon vision systems, and is viewed as challenging, but not requiring substantial new breakthroughs. Requirement (III) involves new hardware or advances in existing hardware; and it may pose a very

significant hurdle. (Alternative approaches, which do not require the slit have been envisioned, although we shall not comment upon these possibilities here. These would eliminate the technical difficulties associated with the slit, but substantial technical difficulties are anticipated to produce a high spatial resolution pixel detector.) Given the strong incentives for developing the instrument, we are optimistic that all of the key requirements can be met within the next 5-10 years.

Acknowledgments

This work was supported partially (B. L. Adams) by the MRSEC Program of the National Science Foundation under Award Number DMR-9632556 and partially by the Danish Natural Science Research Council through Dansync. The authors gratefully acknowledge many helpful discussions with S. Garbe, W. Pantleon, and U. Lienert.

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