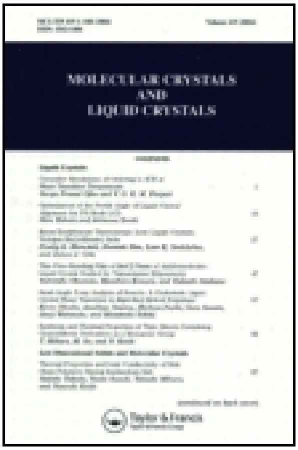

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Characterizing the Scale of Liquid Crystalline Textures: Rheinberg Differential Color Contrast

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Characterizing the Scale of Liquid Crystalline Textures: Rheinberg Differential Color Contrast

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The limitations of referring to domain size as a unit of microstructural scale are discussed. A more practically useful measure of scale is given by the defect density. For this purpose, the distribution of defects in a texture can be highlighted with maximum contrast and resolution by using Rheinberg differential color illumination.

Keywords: domain, texture, scale, defects, Rheinberg, liquid crystal

INTRODUCTION

Characterizing and controlling microstructural scale, to optimize physical properties, is central to Materials Science. For example, a fine grain size in a metal translates to increased yield strength at room temperature, decreased high temperature creep resistance and increased electrical resistivity. The thermomechanical history required in order to obtain a particular grain (crystal) size and dislocation density in metals has been investigated exhaustively. Analogous studies have been performed on ceramics and polymers. However, although the physical properties of liquid crystalline materials must similarly be dependent on microstructural scale, the factors that determine domain size and defect (disclination) densities have received scant attention. It has been noted that some liquid crystalline polyesters have textures that are an order of magnitude finer than the textures of their oligomers[1-2]—but not even this observation has been explained. Theoretical studies of the liquid crystalline state have concentrated on the molecular parameters or environmental conditions that promote formation of liquid crystalline phases; they have not attempted to quantify the scale over which long range orientational order develops.

The impact of microstructural scale on physical properties is illustrated by the following examples:

- In conducting polymers and non-linear optical polymers, microstructures that promote extensive electronic or optical scattering must be avoided.
- Ways of increasing the compressive strength of liquid crystalline polymers are being sought actively. While several attempts have been made to enhance this property by tailoring the type of molecule and the type of order, the *dependence of strength on scale has been neglected.* If analogies are to be drawn with more traditional materials, one would expect a fine microstructure (i.e., a high density of orientational defects) to be beneficial.

DESCRIPTIONS OF MICROSTRUCTURAL SCALE

Attempts to identify a simple microstructural unit in liquid crystalline materials have led to the concept of a "domain". However, there is a lack of consensus about the physical nature of a domain. According to one view (Marrucci, quoted in Reference 3), a domain is a region in which the *net director orientation is zero.* While the director field surrounding a single disclination could meet this description in principle (Figure 1), the existence of such an idealized topology is unlikely. One would typically expect several line disclinations or even walls to be associated with each domain, to meet the requirement of no net director orientation. There is no *a priori* simple relationship between the size of a domain and the number of orientational defects that it contains. Yet, as we have seen, *it is the defect density that affects physical properties*, so the "domain size" as defined here does not necessarily help with property prediction.

An alternative definition[5-6] invokes the opposite extreme of director orientation in describing a domain: a region in which the *director orientation is essentially constant*, thus distinguishing it from a domain boundary where there is a rapid reorientation of the director over a relatively short distance. This description appeals to our familiarity with single crystals as the basis of metal and ceramic microstructures. A domain defined in this way need not contain any disclinations (Figure 2 and Reference 7), though they certainly can be accommodated (Figure 3 and Reference 7). Thus, as with Marrucci's definition, it again is not possible to

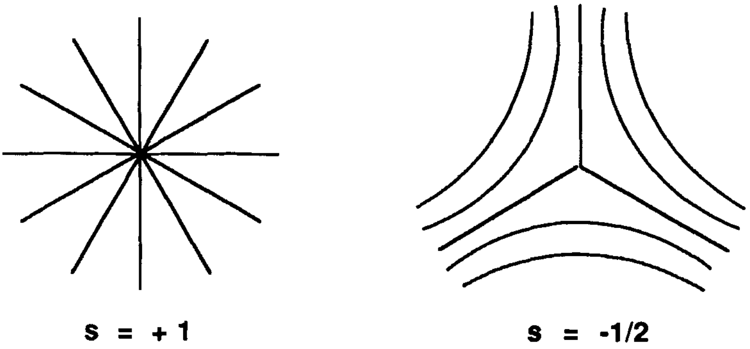

FIGURE 1 Schematic examples of undistorted director fields around an isolated disclination in a nematic.[4] The net director orientation in the vicinity of the disclination is zero in each case.

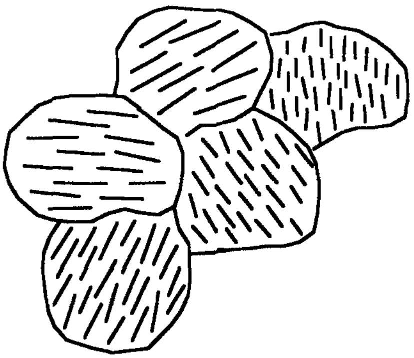

FIGURE 2 Schematic representation of a liquid crystalline microstructure in which regions of approximately constant director orientation are separated by abrupt boundaries. As a reminder that the directors have an out-of-plane component of orientation, their projections on the specimen plane have different lengths.

anticipate the number of disclinations that must be associated with any given domain. Also, there is an implication that a domain is a *discrete* area in the microstructure, clearly separated from its neighbors by a definite boundary or wall. This distinction can indeed be made—but only in some special cases:

- Nematic textures sometimes contain closed walls that may[7–8] or may not[7] contain disclinations. When a wall does contain disclinations, their distribution is analogous to the arrangement of dislocations in a low angle grain boundary in a metal.
- After liquid crystalline fluids are sheared, the molecular order typically relaxes to a pattern in which the director orientation varies periodically with position measured parallel to the shear direction.[9] A "divergence function" can be plotted,[10] showing how the angle between the director and the shear direction changes as a function of position along the shear direction. If the divergence function has a sawtooth profile, as in Kevlar,[11] the regions of approximately constant director orientation can be regarded as domains under the present definition.

More generally, it is possible to identify regions in which there is a rapid local change in director orientation, *without* these regions forming continuous (closed) boundaries around a domain. They can even exist within an otherwise constant director field, as already illustrated in Figure 3. The absence of experimental evidence for definitive domain walls in liquid crystalline polymers has been commented on previously.[12–13] This is taken into account by Noel's description of a domain[14] as a highly ordered region embedded in a matrix of less order.

An attempt has been made to define domains so that each is centered on a single disclination line.[15] While this is possible in the case of nematic schlieren textures, it is not satisfactory if other defects such as inversion walls are present. Also, the positioning of domain boundaries is arbitrary.

The difficulty of referring to domain size as the generalized unit of microstructural scale in liquid crystalline materials is apparent: there is some leeway in how we choose to define a domain, and there is no simple correlation between the domain

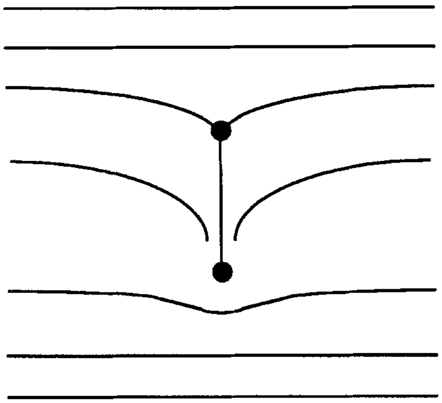

FIGURE 3 Schematic representation of an isolated orientational discontinuity that can exist entirely within an otherwise aligned director field.

size (however defined) and the concentration of orientational defects. It becomes expedient to dispense with dissecting the microstructure into domains, and, instead, to consider the distribution of orientational defects directly. We may think of the number density of these defects as being analogous to the dislocation density in a crystalline material. Practically, it is the *distribution of defects*, and not domain size, that most directly affects physical properties. We will therefore focus our attention on characterizing the distribution of orientationl discontinuities in liquid crystalline microstructures.

CHARACTERIZING ORIENTATIONAL DEFECT DENSITY BY LIGHT MICROSCOPY

Because orientational discontinuities are associated with abrupt changes in optical properties, they usually can be characterized by light microscopy. It is useful to distinguish between the following two approaches:

- The average separation of discontinuities can be quantified in terms of the average size of regions over which optical orientation is comparatively constant.
- The discontinuities themselves can be highlighted.

The first type of analysis can be achieved via the established microscopical method of observing thin specimens between crossed polars. The specimens may be in the fluid liquid crystalline state. Alternatively, in the case of polymeric liquid crystals, the mesophase in a small bulk sample may be preserved to room temperature by quenching, and a thin specimen is obtained by sectioning. (The latter preparation technique avoids the defect distribution being affected by interactions with the glass confining surfaces.) Specimens appear dark locally where the vibration directions (not necessarily the molecules)[16] are parallel to the transmission directions of the polars. The average size of ordered regions then corresponds to the average size of light regions as measured for a variety of crossed polar orientations. However, the value of this approach is limited by the fact that, unless images are digitized, or are otherwise compared with calibrated reference values of optical density, determining the size of a "light" area involves subjective judgement. Furthermore, observations made with the specimen between crossed polars may not distinguish between regions that differ only in their out-of-plane molecular orientation (Figure 4).

Observations of texture between crossed polars do not highlight orientational defects explicitly. Instead, textures provide the preferred means of characterizing director orientations *between* the defects. Even a feature as "simple" as an inversion wall gives rise to a complex pattern of extinction (Figures 5a and 5b), which adds to the difficulty of interpreting fine microstructures. Orientational discontinuities in a schlieren texture are usually identified by looking at how the texture changes in a series of micrographs as the crossed polars are rotated:

- At point disclinations, and at disclination lines normal to the specimen surface, dark bands are seen to rotate about fixed points in the microstructure.
- Other abrupt discontinuities of in-plane molecular orientation are characterized by bands of extinction that move laterally over a very narrow range of positions during a 360° rotation of the crossed polars.

We have investigated a number of alternative contrast mechanisms available to the light microscopist, to specifically emphasize optical discontinuities. In conventional bright field illumination (both polars withdrawn, Figure 5c), optical discontinuities show up as a result of light scattering. The discontinuities cause light to deviate from its original path so that it cannot subsequently be collected by the objective; thus, the discontinuities are seen as locally darker features in the image. (The same scattering process is responsible for the turbid appearance of bulk liquid crystalline specimens.) Resolution is good, because the most effective scattering is achieved by discontinuities that have a scale comparable to the wavelength of light, and it is therefore the cores of disclinations that are darkest. Unfortunately, because only a small percentage of the light incident on the specimen is scattered, the *contrast* (visibility of detail) is poor: the fine detail is difficult to discern against the light that has *not* been affected by passage through the sample.

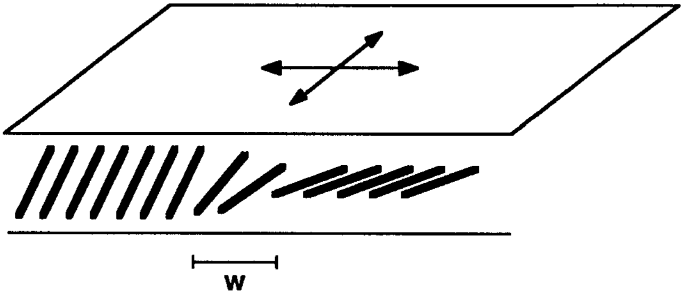

FIGURE 4 While molecular orientation varies abruptly over the distance w, observations of transmitted light intensity between crossed polars may not reveal the presence of two "domains", if all the molecules lie in the plane of the diagram. The crossed polar orientations that would make the entire specimen appear dark are marked on the top surface.

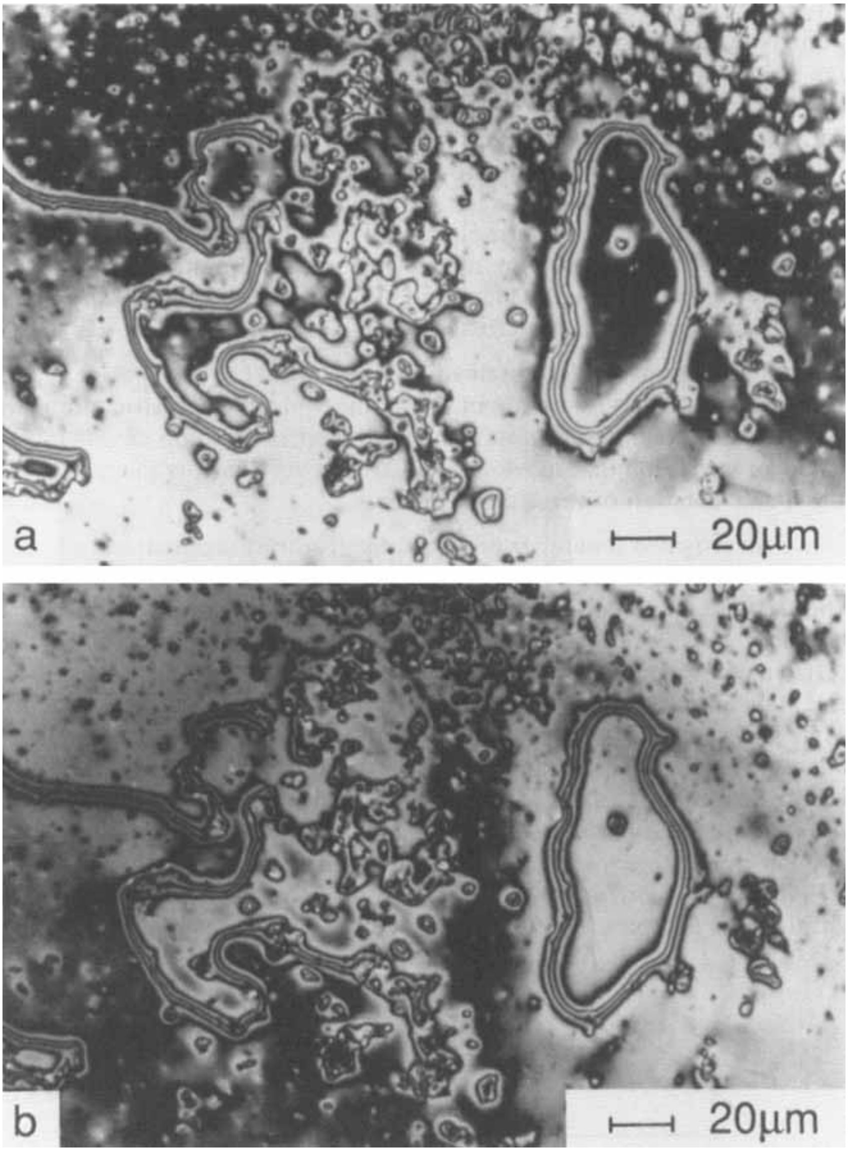

FIGURE 5 Nematic microstructure, including inversion walls, photographed under a variety of illumination conditions. The specimen was poly(p-hydroxybenzoic acid), quenched from 480°C; number average degree of polymerization = 15. (a) Crossed polars; polarizer vibration direction E–W; (b) Crossed polars rotated 45° from orientation in (a); (c) Bright field (no polars); (d) Phase contrast; (e) Dark field; (f) Rheinberg differential color illumination; blue central stop and green outer annulus; fine detail more visible than in the other cases. The phase contrast image was obtained with a ×40 objective; a ×32 objective was used for the others. See Color Plate V.

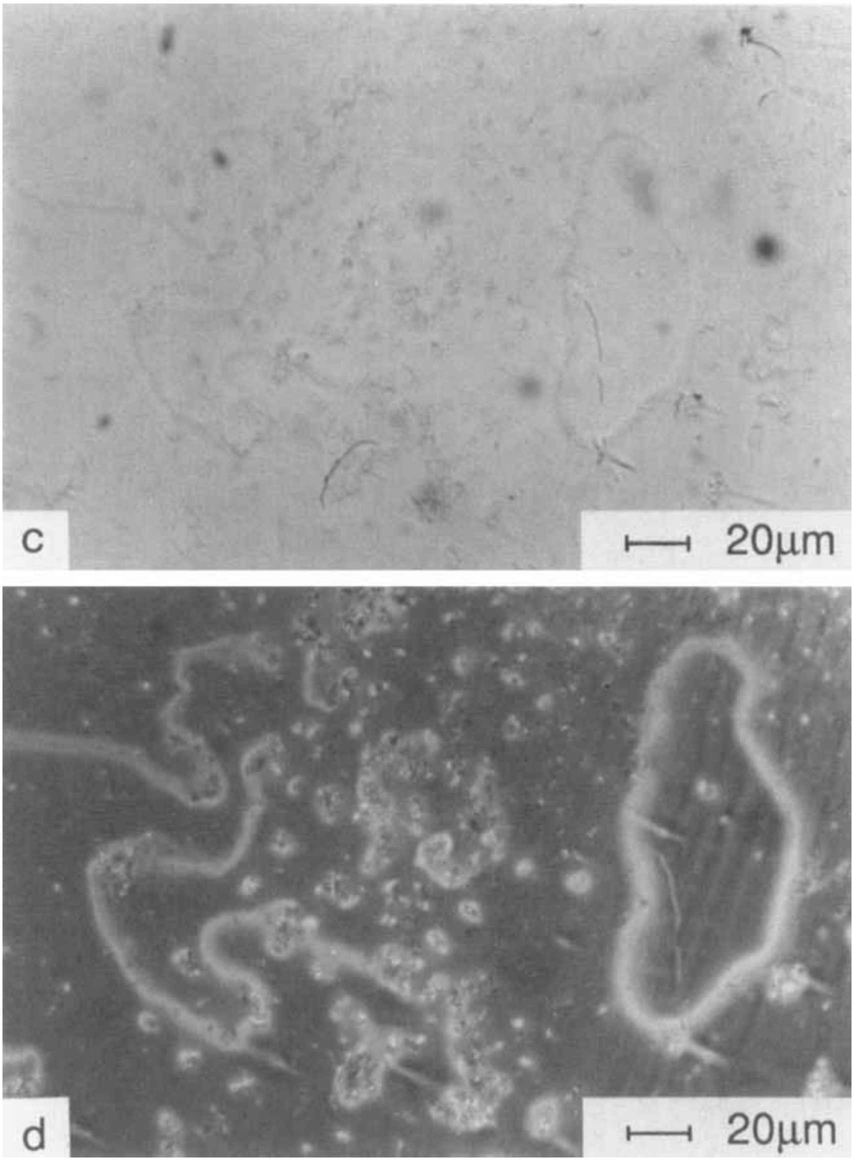

FIGURE 5 (*continued*) See Color Plate VI.

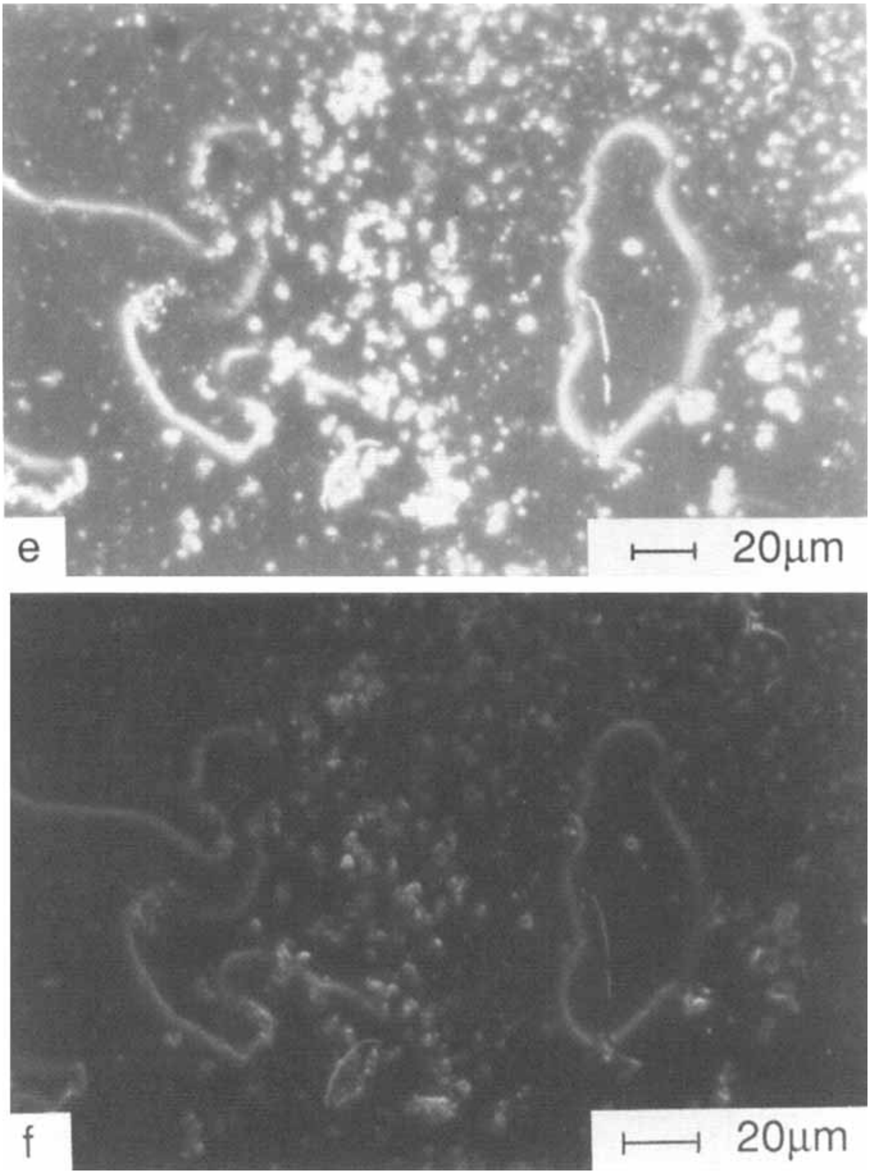

FIGURE 5 (*continued*) See Color Plate VII.

There are several techniques in light microscopy that enhance contrast relative to what can be achieved in bright field. Phase contrast[17-18] can be used to image point-to-point changes in refractive index. However, the gain in contrast is accompanied by a loss of resolution, because the refractive index change even at some distance from the core of abrupt discontinuities contributes to contrast. The discontinuities consequently are easier to see than under bright field conditions, but they also appear to be wider and more blurred (Figure 5d). It therefore is difficult to distinguish detail in regions that contain a large number of closely spaced disclinations.

Dark field microscopy (Figure 5e) essentially inverts the contrast observed in bright field—optical discontinuities appear as bright features on a dark background.[17] The contrast is excellent, because only the scattered light contributes to the image. The principal shortcoming of this technique is the loss in resolution that occurs if an objective with a high numerical aperture is needed to form images of the microstructure. It can be demonstrated theoretically[19] that the maximum resolution obtainable in bright field will always exceed that obtainable in dark field if the objective numerical aperature is greater than 0.43. In the case of liquid crystalline polymers, where microstructures can be an order of magnitude finer than in low molecular weight liquid crystals,[1-2] high objective numerical apertures are essential to resolving the microstructure adequately.

We have found that *Rheinberg differential color illumination* (Figure 5f) provides optimum contrast for imaging disclinations, without compromising the maximum resolution obtainable with a given objective. While the technique was invented almost a hundred years ago,[20] its use has been limited to biological specimens[21-26] and to textiles.[25,27] To obtain Rheinberg illumination, concentric color filters are placed below the substage condenser so that the axial and peripheral illuminating rays incident on the specimen have different colors (Figure 6). Because the peripheral illumination is responsible for resolving and reconstructing the finest detail in the image, this detail appears in the color of the outer filter, against a background color that is due to the inner filter. The resulting contrast is superior to that obtained by any mechanism that requires the viewer to differentiate between grey levels.[28-32] In addition to ensuring that both resolution and contrast are maximized, Rheinberg illumination offers control over the image in the following ways:

- Both coarse and fine microstructural detail can be included in the image.
- Both the color and intensity of the fine detail can be selected relative to that of the background.
- By varying the relative diameters of the two filters, one can control the scale of detail that appears as background or highlight respectively.

CONCLUSIONS

The microstructural scale of liquid crystalline materials can be characterized unambiguously in terms of the concentration of orientational defects. Attempts to quantify the scale in terms of "domain size" are limited by the difficulty of defining domains in most microstructures. Also, the defect concentration is associated di-

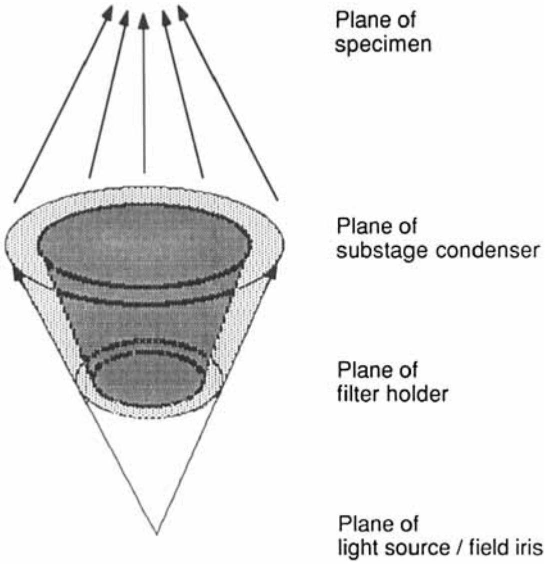

FIGURE 6 Schematic representation of Rheinberg illuminator.

rectly with physical properties, but is not a simple function of the size of arbitrarily described domains. While defects can be revealed by many light microscopical techniques, they are most usefully highlighted relative to the rest of the microstructure by using Rheinberg differential color contrast. This technique maximizes both contrast and resolution simultaneously.

Acknowledgments

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