LBL-29350
Preprint

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Materials & Chemical Sciences Division

National Center for Electron Microscopy

Submitted to Zeitschraft für Kristollographie

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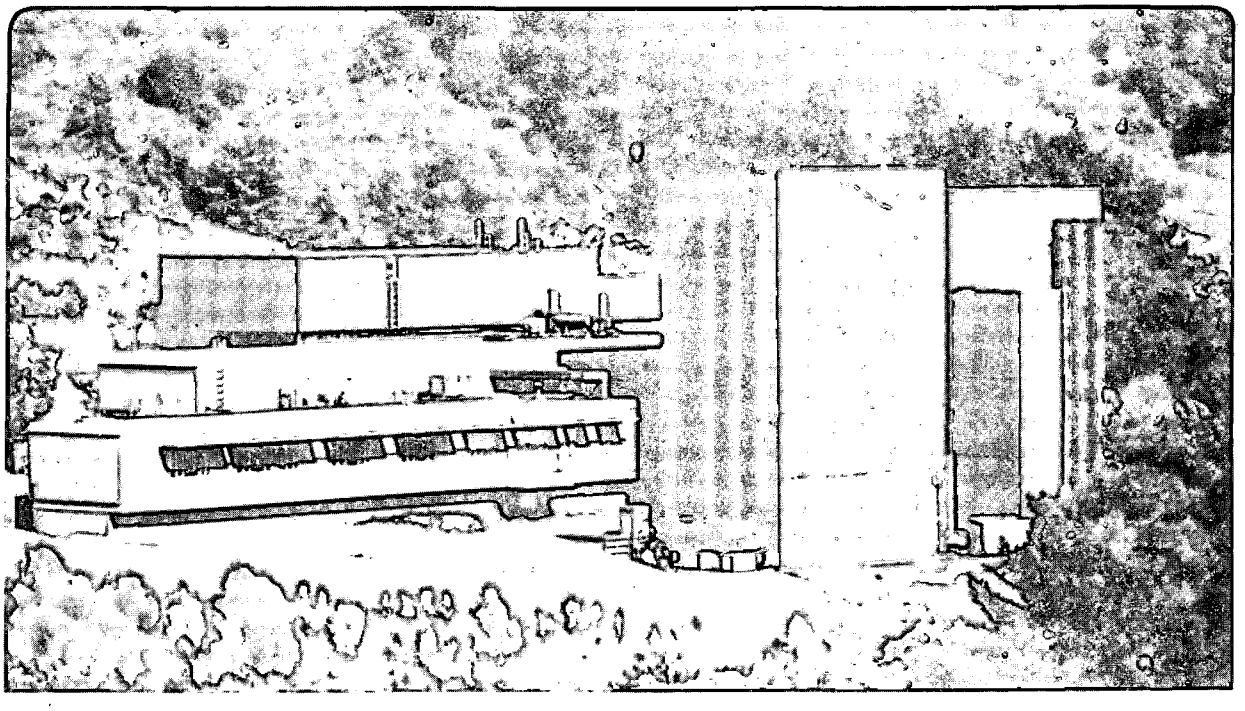
July 1990



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Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

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COMMENTS ON 'HRTEM-BILDKONTRASTSIMULATION VON STRUKTUREN MIT PUNKTDEFEKTEN IN SPEZIELLEN LAGEN'

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This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

Short Communication

Comments on "HRTEM-Bildkontrastsimulation von Strukturen mit Punktdefekten in speziellen Lagen"

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Key Words: HRTEM image simulation, point defects

Abstract

We wish to point out that papers by Rahman (1988) and Rahman & Weichert (1990) contain a basic error in concept. In these papers, the authors seek to shorten the amount of computation necessary for a simulation of a high-resolution transmission electron microscope (HRTEM) image of a structure containing a defect, using as an example an oxygen vacancy in mullite. Unfortunately, the method used leads to errors in the simulated image.

The usual method of computing an image of a non-periodic structure is to construct a "supercell" consisting of a number of perfect unit cells surrounding the cell(s) containing the defect (e.g. Iijima & O'Keefe, 1978). The computation is then carried out using the supercell as the new unit cell for the defect structure. Note that the new defect structure is periodic with the periodicity of the chosen supercell, and thus the number, n, of original unit cells making up the supercell must be chosen large enough not only to sample diffraction space sufficiently finely, but also to keep the images of adjacent defects from interfering. Such interferences can arise if electrons scattered from non-perfect regions near to the defect in one supercell contribute to the image of the defect in an adjacent supercell. In addition, if a smooth "periodic continuation" of the structure at the supercell boundary is not possible, the area immediately surrounding the defect of interest must be far enough away from the unsmooth boundary to avoid interferences between the image of the defect and that of the (probably non-physical) boundary.

Given that computation of an image of a supercell consisting of n perfect unit cells will require between n and n^2 times as much computing time as one unit cell, Rahman (1988,1990) attempts to shorten the process by constructing a type of supercell of the same size as the original cell. This construction is carried out by first computing the crystal potentials for two different cells, one perfect and the other with periodic vacancies, then using a "cut and paste" technique to produce the potential for a cell with a vacancy at one edge and an atom in the corresponding site at the opposite edge. The net result of this procedure is a crystal potential that may be reasonable when considered in isolation, but, when repeated periodically, leads to a non-physical step in crystal potential right at the position of the defect of interest. Since the discrete Fourier transform of an isolated area is effectively the transform of the periodic extension of the area, then when use is made of discrete Fourier transforms in computing the simulated image from the model potential, a non-physical result is obtained.

Figure 1 shows the crystal potential (top), and resultant simulated image (bottom) computed for copper in [001] orientation. Column (a) shows results for perfect Cu, column (b) for Cu containing a periodic vacancy. Column (c) shows the potential and image obtained using a supercell consisting of one perfect cell and one with a vacancy. Column (d) is the result of Rahman's technique applied to the potentials shown in (a) and (b), and shows both the resultant model of the crystal potential, with the atom at 0,1/2,1/2 sliced in half to produce a step in the resulting potential distribution (top), and the image resulting from this non-physical model (bottom). Comparison of the image areas around the vacancy in (b) and (c) shows that the supercell calculation successfully models the aspect of the vacancy image. On the other hand, the image area around the vacancy/atom position in (d) does not match either the vacancy image (above the vacancy/atom position), or the atom image (below the vacancy/atom position).

We have chosen copper as the example in order to provide a clear demonstration of the non-physical nature of a structure that can be produced by careless application of the "cut and paste" technique. Applied to oxygen positions in mullite, the technique produces only slight (but significant) differences under the conditions used by Rahman (1988,1990). It has been shown by Epicier et al. (1990) that a resolution of better than 1.9Å is required to detect the presence of oxygen atoms in mullite. Thus at the 2.4Å resolution of the microscope used in the mullite examples of Rahman (1988,1990), the contrast of the image is largely due to mullite cations, so that the presence or absence of an oxygen vacancy (or half of one) makes very little difference to the image character.

Finally it should be noticed that the mullite images computed by Rahman (1988,1990) appear to be reversed in contrast. Such a reversal can occur if the sign of the wave aberration function does not match the sign chosen for the time-dependent Schrödinger equation in deriving the expressions used in the simulation programs (Saxton et al., 1983). Images simulated with both the well-tested SHRLI (O'Keefe et al., 1978) and NCEMSS (Kilaas, 1987) programs could be made to match those computed by Rahman (1988,1990), but only when the spherical aberration was set to a non-physical *negative* value, suggesting that such a sign problem exists in the programs used by Rahman.

Acknowledgement

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division of the U.S. Department of Energy under contract no. DE-AC03-76SF00098.

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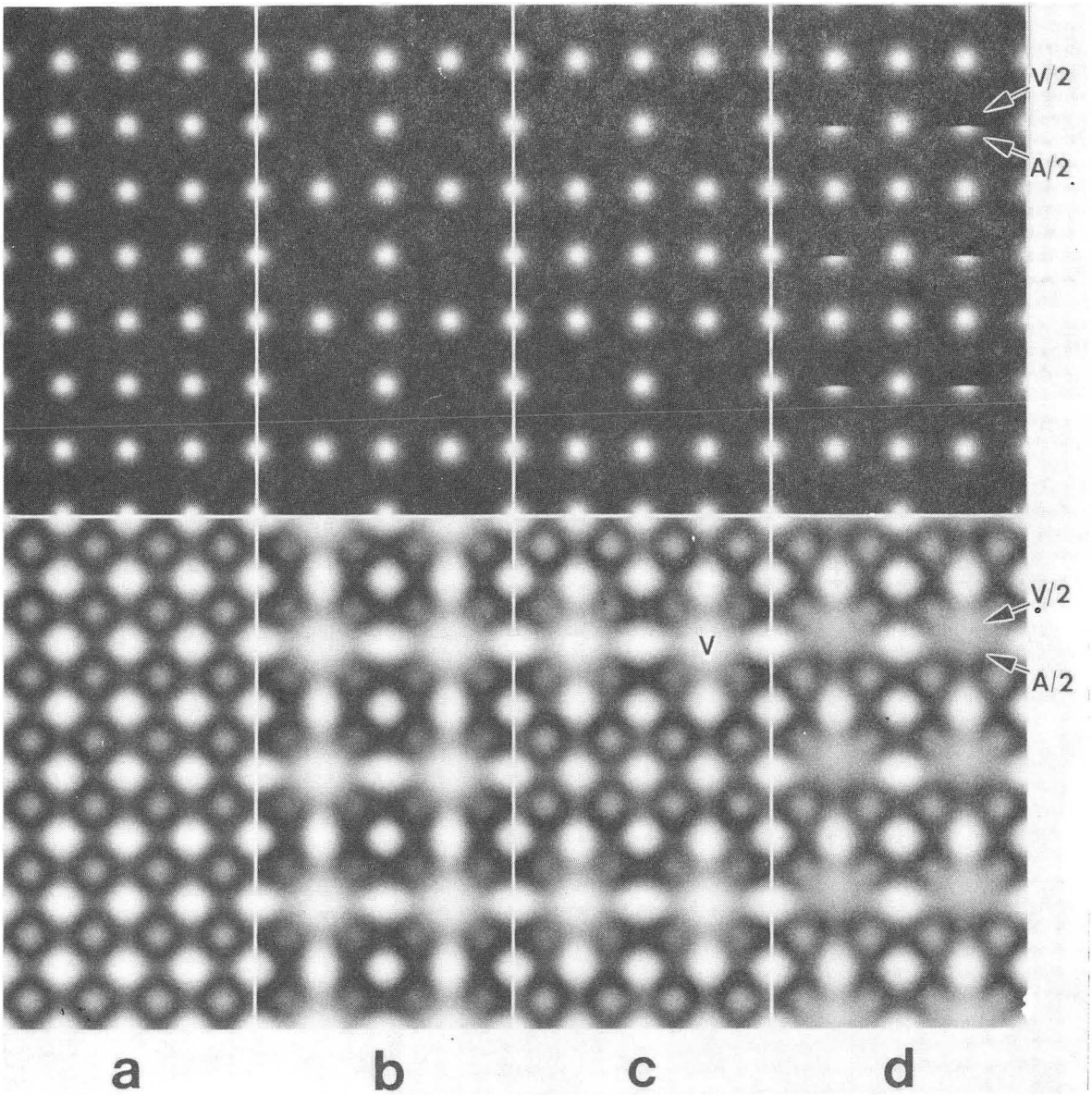

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Crystal potential (top), and image (bottom) computed for copper in [001] orientation. (a) perfect Cu, (b) Cu containing a periodic vacancy, (c) conventional supercell, consisting of one perfect cell and one with a vacancy (V), (d) Rahman-type "supercell" from the potentials shown in (a) and (b); positions of the half vacancy and half atom are marked V/2 and A/2 respectively. Note the non-physical step in potential at the vacancy/atom position in (d), and the resulting non-physical image. Simulation conditions for all images are for 60Å specimen thickness, imaged at 800keV in the JEOL ARM-1000, with C_S of 2mm and underfocus of -500Å.

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