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Image resolution enhancement by combining information from electron diffraction pattern and micrograph

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An electron micrograph of chlorinated copper phthalocyanine at 2 Å resolution taken on the Kyoto 500 kV electron microscope has been enhanced to 1 Å resolution by incorporating the information from the corresponding electron diffraction pattern. Structure-factor amplitudes up to 1 Å resolution were obtained from the electron diffraction pattern. Phases of the structure factors within 2 Å resolution were derived from the Fourier transform of the electron micrograph. A phase-extension technique introduced from X-ray crystallography was then used to derive the phases between 2 and 1 Å resolution. The final image was obtained by the inverse Fourier transform using the structure factor amplitudes from the electron diffraction pattern and the phases from the electron micrograph and from the phase-extension procedure.

1. Introduction

It is well known that high resolution electron microscopy (HREM) gains an advantage over other methods in determining structures of minute crystals. Structure images taken under the optimum defocus condition reveal directly the structure of the examined sample projected along the direction of incident beam. Images with defocus amounts other than the optimum defocus can be restored by means of image deconvolution. Different methods were developed for image deconvolution in linear and non-linear cases [1–9]. However, the resolution of images, either taken under the optimum defocus condition or obtained by means of image deconvolution, is limited by the resolution of the electron microscope. On the other hand, electron diffraction patterns bear information corresponding to much higher resolution limit. It has been recommended to determine the crystal structure by combining the information from an electron micrograph and that from the corresponding electron diffraction pattern, because this can greatly simplify the process of structure analysis and yield much better results than by using either one of them. A number of procedures have been proposed for this purpose [10–12]. The methods have been proved to be valid for simulated images, and the resolution of the final image can be beyond the limit of the electron microscope. Here we describe a practical application of the image resolution enhancement technique to an experimental electron micrograph of chlorinated copper phthalocyanine. The procedure is based on

the phase extension technique used in direct methods in X-ray crystallography as described in ref. [12].

2. Experimental electron micrograph and diffraction pattern

The sample used in the following test is crystalline chlorinated copper phthalocyanine, $C_{32}N_8Cl_{16}Cu$. The unit cell dimensions are $a = 19.62$ Å, $b = 26.04$ Å, $c = 3.76$ Å and $\beta = 116.5°$. Fig. 1 shows the experimental electron micrograph together with the schematic diagram of the molecule. The micrograph was taken on the Kyoto 500 kV electron microscope with $\lambda = 0.0142$ Å, $C_s = 1.06$ mm, $D = 100$ Å and $\Delta f \simeq 500$ Å. This micrograph was digitized using the Perkin Elmer PDS microdensitometer data acquisition system with 50×50 μm^2 aperture. The two-dimensional unit cell ($a' = a \sin \beta = 17.56$ Å, $b = 26.04$ Å) was divided into 110×124 pixels. Ten such unit cells were measured and then averaged. The resulting digitized image is shown in fig. 2a as a half-tonegraph. The electron diffraction pattern, taken from the same sample, is shown in fig. 2b. The whole pattern (4.5×4.5 cm) was divided into 2251×2251 pixels and digitized using the above-mentioned microdensitometer with a 20×20 μm^2 aperture. The integrated intensity of each reflection was obtained by summing up the density, $\ln(I_0/I)$, of the pixels within the diffraction spot and then subtracting the background.

3. Results of phase extension

Structure-factor amplitudes up to 1 Å resolution were obtained from the square root of the electron diffraction intensities. Phases within 2 Å resolution were derived from the electron micrograph. A multisolution phase extension procedure similar to that of Yao [13] was used to derive the unknown phases within 1 Å resolution. A large starting set was used, which consists of the reflections with known phases within 2 Å resolution and ten additional reflections beyond 2 Å resolution selected by the convergence mapping procedure [14]. Random phases were assigned to the ten additional reflections. Fifty trials were calculated. The best result was picked up by using the residual figure of merit:

$$R_\alpha = \sum_H |\alpha_{\text{est}} - \alpha| / \sum_H \alpha_{\text{est}},$$

where

$$\alpha^2 = \left[\sum_{H'} \chi \cos \Phi\right]^2 + \left[\sum_{H'} \chi \sin \Phi\right]^2,$$

$$\alpha_{\text{est}} \simeq \sum \chi \frac{I_1(\chi)}{I_2(\chi)},$$

$$\chi = 2\sigma_3\sigma_2^{-3/2} |E(H)E(H')E(H-H')|,$$

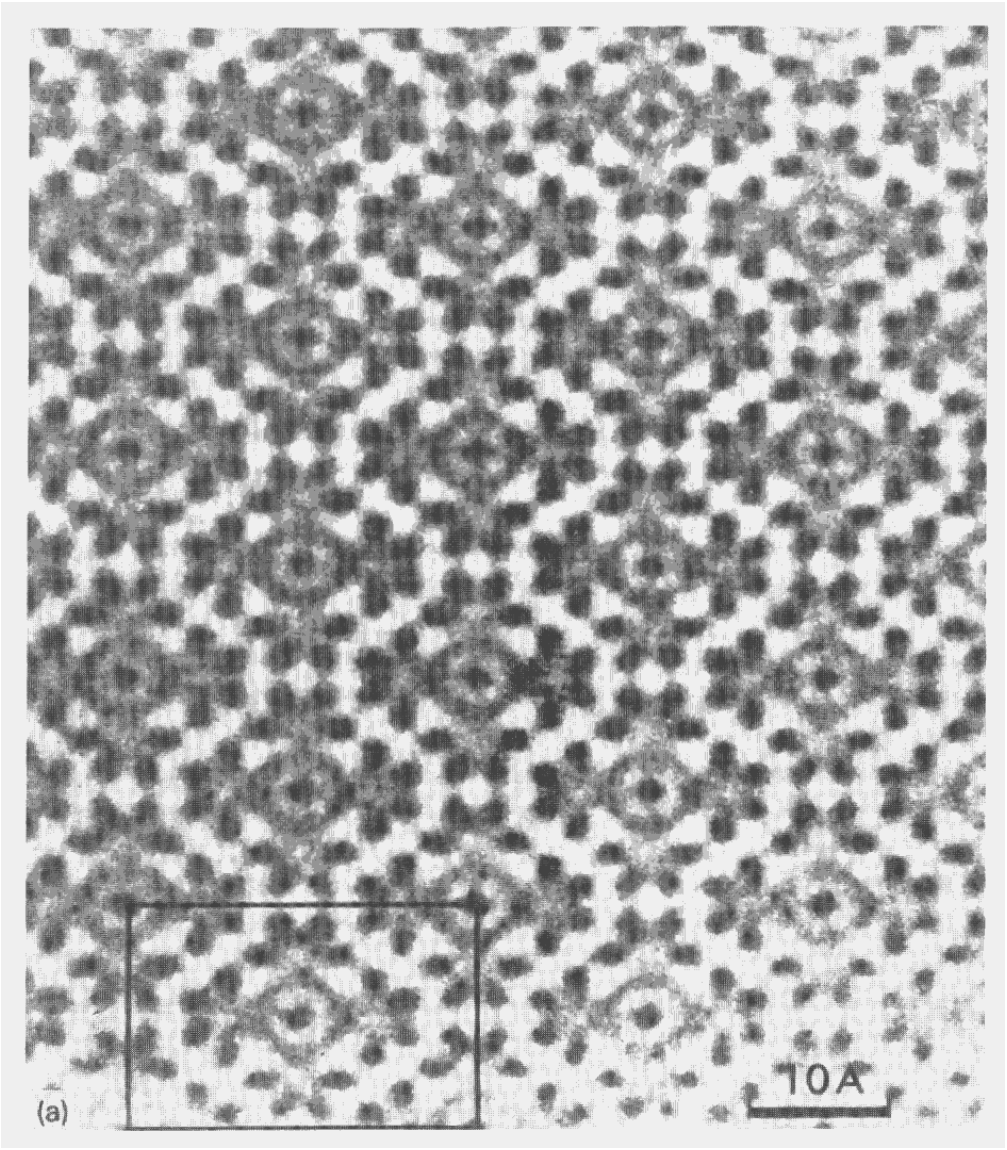
$$\Phi = \varphi_{H'} + \varphi_{H-H'}.$$

The above definition of the residual figure of merit is the same as that used in the program MULTAN [15]. The result of phase extension is shown in fig. 2c in comparison with the theoretical image of 1 Å resolution shown in fig. 2d.

4. Discussion and conclusion

In the above procedure the dynamical scattering was not taken into consideration. The image shown in fig. 2c implies that this does not have much influence on the result of image resolution enhancement.

In principle, direct methods used in X-ray crystallography can also solve the phase problem of electron diffraction, and the crystal structure can be determined by using electron diffraction data alone. In order to compare the present result with the result obtained by electron diffraction analysis, a test was carried out with the above electron diffraction data. Random starting phases were assigned to all the available reflections. Fifty trials were calculated. The best five resulting phase sets were examined. They give similar E-maps. The one corresponding to a minimum residual figure of merit is shown in fig. 2e. Obviously it is difficult to solve the true structure from this E-map. This means that the preliminary phases obtained from the Fourier transform of the image are essential to the success of phase extension.



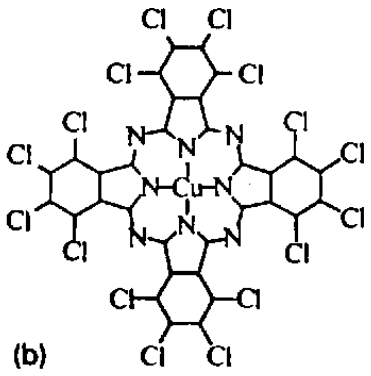


Fig. 1. (a) 2 Å resolution electron micrograph of crystalline chlorinated copper phthalocyanine taken on the Kyoto 500 kV electron microscope with $\lambda = 0.0142$ Å, $C_s = 1.06$ mm, $D = 100$ Å and $\Delta f = -500$ Å. (b) Schematic diagram of a molecule of chlorinated copper phthalocyanine.

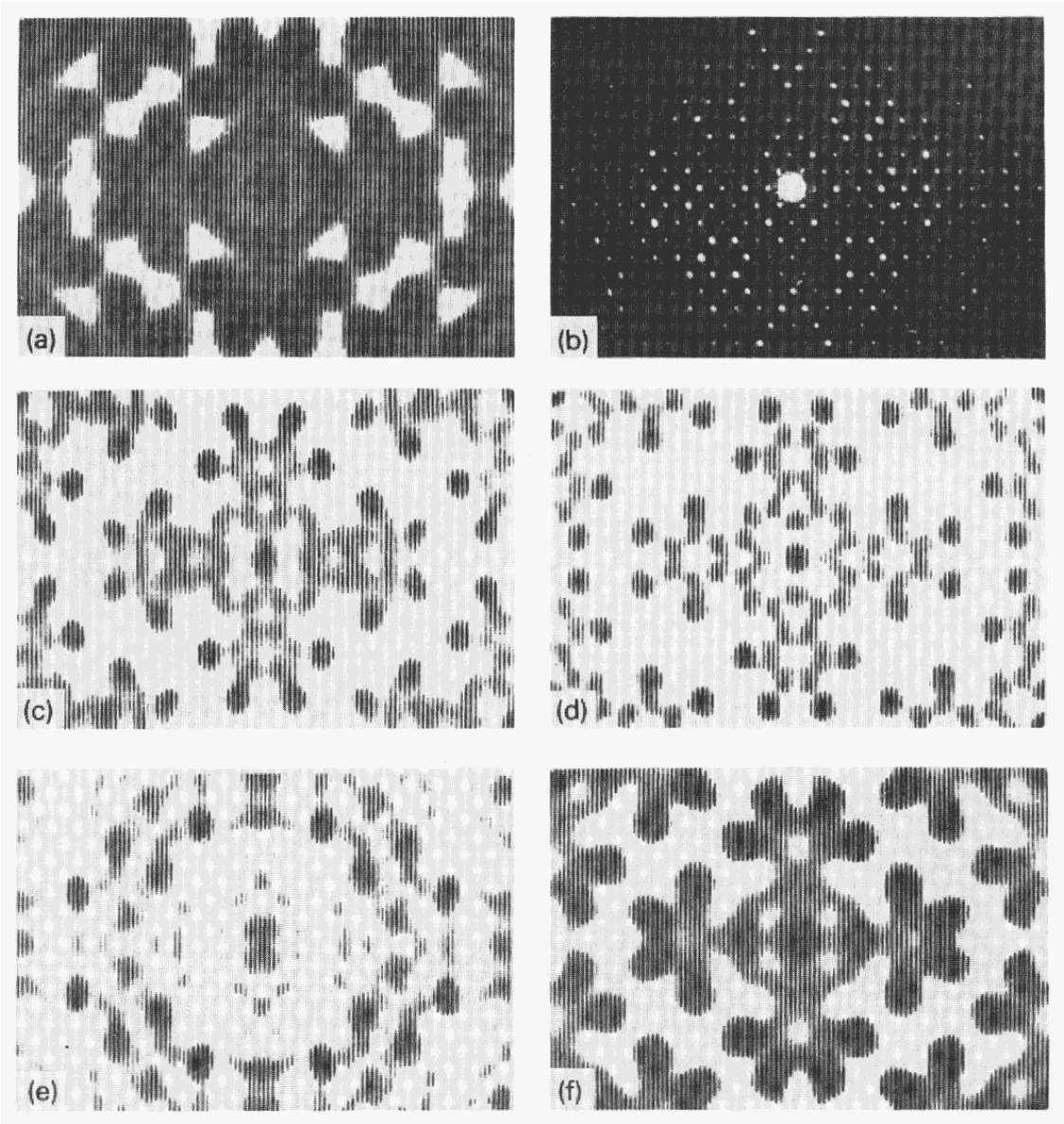


Fig. 2. (a) Half-tone-graph representation of the digitized 2 Å resolution electron micrograph of chlorinated copper phthalocyanine. (b) Electron diffraction pattern of chlorinated copper phthalocyanine at 1 Å resolution. (c) Fourier map of the crystalline chlorinated copper phthalocyanine obtained by the phase extension technique combining the information from (a) and (b). (d) Theoretical image of the crystalline chlorinated copper phthalocyanine at 1 Å resolution. (e) The best E-map obtained from the direct-method phasing of the reflections on (b). (f) Result of image deconvolution from (a).

Although the electron micrograph shown in fig. 1 was taken near the Scherzer defocus condition [16], the image quality can be improved by means of image deconvolution. Fig. 2f shows the deconvoluted image which was described in ref. [17]. As is seen, although the technique is efficient the resulting image cannot reveal individual atoms, because the original resolution of the micrograph is about 2 Å and the deconvolution technique, in principle, does not enhance image resolution.

It is concluded that the combination of electron diffraction pattern and electron micrograph yields results much better than from either of them alone. This will play an important role in the image processing of high resolution electron micrographs of crystalline samples.

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