Electron Deformation Density at Temperatures around 20 K *

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A 20 K diffractometer for X-ray measurement is described. Electron density experiments at 15 K for oxalic acid and at 23 K for acetamide were carried through, leading to high-resolution experimental and multipole static density maps.

Key words: 20 K single crystal X-ray diffractometer; Deformation density; Oxalic acid dihydrate; Acetamide.

With a closed-cycle He-cryostat, which we recently integrated into a four-circle diffractometer with offset χ-circle diffractometer, routine X-ray measurements down to 15 K are now possible with the aim of precise electron deformation-density maps. Based on experiments and some promising results with our 50 K diffractometer [1], this device is built up using a Huber type 5012 Eulerian cradle, controlled by a newly developed motor-driver interface using the MC 68008 microprocessor and a 3-channel programmable temperature controller (type ITC4, Oxford Instruments). A special vacuum chamber made of beryllium was used.

The quality of the measurements was checked with α-oxalic acid dihydrate [2], which we chose as the standard in the field of charge density. Figure 1 shows the experimental deformation density map at 15 K with a cut-off value of $\sin\theta/\lambda = 0.71$ Å^{-1}.

The second compound is acetamide, as its structure has also been examined several times and very precise neutron data at 23 K are available [3]. Comparison of X-ray intensity data at various temperatures shows an enormous increase of intensity, especially for high-order reflections, when the sample temperature is reduced from 100 K to about 20 K. This is illustrated for the (h 0 0) reflection series in Fig. 2, which gives the relative intensity increase with respect to room temperature. The experimental electron density map ($X_{lo}-X_{hi}$) at 23 K with cut-off limit at $\sin\theta/\lambda = 0.66$ Å^{-1} (Figure 3) shows the expected strong maxima in the centre of the covalent bonds and the well-resolved maxima of the oxygen lone-pair region. Only the hydrogen atoms were taken from neutron data, since X–X maps showed unsatisfying results. Multipole refinements and *ab initio* molecular-orbital calculations have been made taking hydrogen bonded partner molecules into account. The obtained maps were in good agreement with the experimental ones [4].

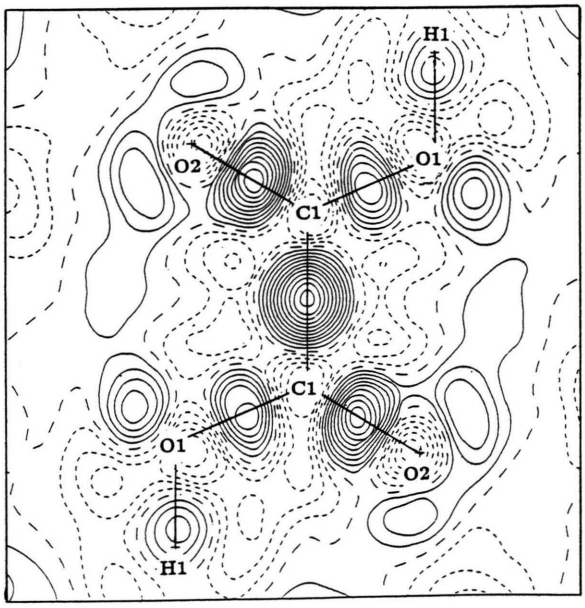

Fig. 1. $X_{lo}-X_{hi}$ electron density map of the oxalic acid subunit of α-oxalic acid dihydrate at 15 K in the plane of the molecule. Contours are at 0.05 eÅ^{-3}.

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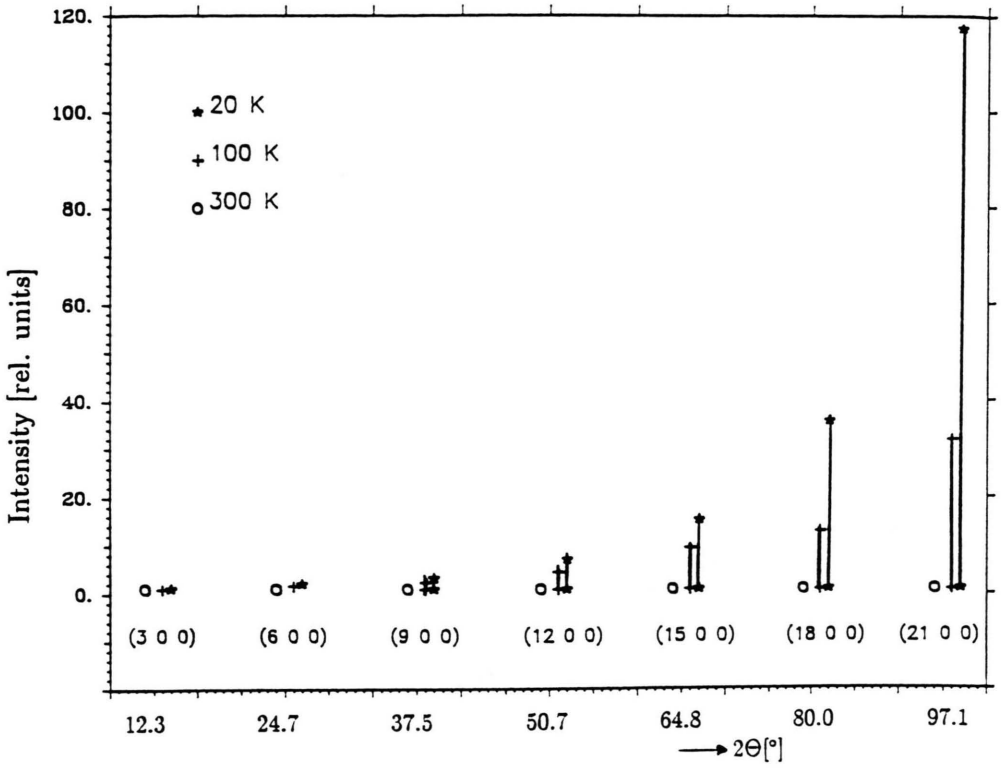

Fig. 2. Relative reflection-intensity gain for the (h 0 0) reflection series upon decreasing the temperature for acetamide, normalized to 300 K-values each.

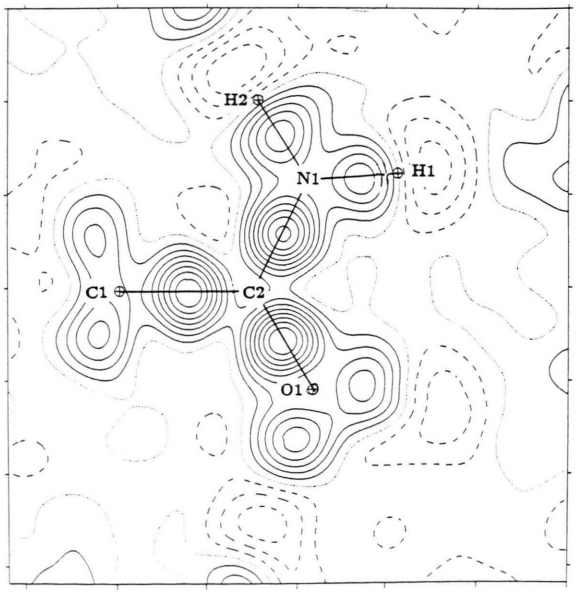

Fig. 3. $X_{lo}-X_{hi}$ electron density map of acetamide at 23 K in the plane of the molecule. Contours are at 0.05 eÅ^{-3}.

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