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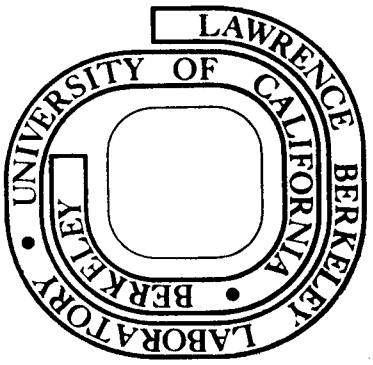

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THE ENERGY DEPENDENCE OF ELECTRON RADIATION DAMAGE IN l-VALINE

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ABSTRACT

Experimental electron diffraction and microscopy results are described regarding the energy dependence of electron radiation damage in the aliphatic amino acid l-valine. The critical exposure, defined as the electron flux necessary to destroy the crystallinity of a material, has been determined over a range of electron energy from 50keV to 3MeV. In terms of the electron velocity the results indicate a β^3 dependence rather than β^2 as predicted from stopping power theory. The results are used to estimate the applicability, especially with respect to potential resolution, of very high voltage electron microscopy in the study of beam sensitive organic materials, and indicate that a factor of 2 times improvement in resolution can be expected at 1MeV compared to 100keV.

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INTRODUCTION

The application of electron microscopy to the study of crystalline organic materials is severely limited by the damage induced in the specimen by the electron beam. This effect is observed in the diffraction mode as the fading of the sharp diffraction spectra, which are normally replaced by a diffuse ring pattern (7,8,5). The general application of high resolution imaging techniques requires that electron radiation damage must be substantially reduced, otherwise the specimen is destroyed before images can be recorded.

The destruction of crystallinity as determined by electron diffraction provides a convenient technique for monitoring the effects of electron irradiation. In addition the electron exposure at which crystalline diffraction spectra are no longer detectable has been found to be a reproducable end point (5). This end point is commonly referred to as the critical exposure (N_{cr}) of the material and is experimentally determined by measuring the time to destroy crystalline diffraction effects for a measured electron current density at the specimen. Using this technique the energy dependence of radiation damage due to high energy electrons has been established in the present work for the amino acid l-valine over a large range of dose rates.

EXPERIMENTAL METHOD

Crystals of l-valine, suitable for electron microscopy, were grown on formvar coated copper grids by the dropwise evaporation of a dilute aqueous solution. The single crystals of material thus obtained are in the form of dendrites which exhibit a characteristic diffraction pattern

(figure 1). The crystals grow with preferred habit and all measurements were done on crystals of the same orientation.

Values of the critical exposure were measured from crystals that had been uniformly irradiated at various constant electron beam currents. At very high electron dose rates, where the loss of crystallinity is very rapid, the end point was determined visually from the flourescent screen. At lower dose rates visual measurements were supplemented by photographic recordings taken at close intervals about the end point.

The determination of the beam current densities were made at the final image plane of all the electron microscopes used. The electron current measurements in the range 50keV to 650keV were made using a Faraday cup previously described (6) whilst those at 1MeV and above employed the Faraday cup of the Toulouse 3MeV microscope (3). The four measurements at 600keV, 700keV, 800keV and 900keV were made using a lithium drifted silicon detector on the U.S. Steel 1MeV microscope.

RESULTS

The magnitude of the critical exposures at all the electron energies were found to be constant over a high proportion of the incident beam current range between 10^{-5}A/cm^2 and 10^{-2}A/cm^2. The values assigned to the critical exposure at each energy were obtained by averaging the measurements from such regions (Fig. 2) and therefore represent values which are indicative of the damage rate in the absence of beam heating or saturation effects due to high incident currents. The value for the critical exposure at 100kV was determined to be $(3.0 \pm 0.3) \times 10^{-3}$ coulombs/cm^2 which is about 50% larger than the value expected from a comparison with previously published data (4).

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Over the full range of voltages the critical exposure was found to increase from a value of $1.1 \pm 0.2 \times 10^{-3}$ coulombs/cm^2 to an average value of $12.8 \pm 2 \times 10^{-3}$ coulombs/cm^2 at the high voltage limit. These results (Table I) are shown graphically in Fig. 3.

Consistent with the findings of Grubb and Groves (5) good correspondence between the visual and photographic determinations of the end point was found even at very low values of electron current density where the exposure times of the photographic emulsions were of the order of 20 seconds.

TABLE I

Values of the experimental critical exposures as a function of electron energy compared to theoretical values derived from stopping power theory.

Electron Energy in MeV	Mean Values of Measured Critical Exposure, N_{cr}, $x10^{-2}$ coulombs/cm^2	The value of N_{cr} expected from stopping power theory normalized to 0.3×10^{-2} coulombs/cm^2 at 0.1MeV
0.05	0.11	.19
0.075	0.17	.25
0.08	0.20	.26
0.1	0.30	.30
0.2	0.50	.44
0.35	0.8	.55
0.5	1.2	.61
0.65	1.0	.64
0.7	1.2	.65
0.8	1.3	.66
0.9	1.4	.67
1.0	1.5	.68
1.5	1.3	.69
2.0	1.5	.68
2.5	1.1	.68
3.0	1.3	.68

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DISCUSSION

In organic materials, since the induction of radiation damage results from electronic excitation and ionization processes, it is pertinent to consider the voltage dependence of the energy loss from the electron beam. The expected energy loss per unit path length is predicted from the relativistic stopping power formula for electrons due to Bethe (1), where radiation losses are ignored, i.e.,

$$\frac{-dE}{dx} = \frac{N.2\pi e^4.Z}{mv^2}\left[\ln\frac{mv^2T}{2I^2(1-\beta^2)} - (2\sqrt{1-\beta^2} - 1 + \beta^2)\ln 2 + 1-\beta^2 + \frac{1}{8}(1 - \sqrt{1-\beta^2})^2\right]$$

where

N = the number of atoms/unit volume in the absorbing material

T = Kinetic energy of the electron

Z = The averaged atomic number of the sample as a whole

e = The electronic charge

m = The electron rest mass

v = The electron velocity

β = The electron velocity ÷ the velocity of light and

I is the mean excitation energy, a parameter which is characteristic of the medium. This parameter has been calculated for a few simple atoms but is usually experimentally derived.

In the case of thin specimens (<1μm) the energy loss rate from high energy electrons (>50keV) is effectively constant through the sample. Hence the average energy deposited per incident electron to such a sample per unit thickness is to a good approximation the energy loss rate at

the incident energy. The variation of the calculated energy loss with primary energy follows a β^2 dependence at low energy. This theoretical curve is shown in Fig. 3, normalized to the 100kV datum point.

The experimental data indicate that no exact correspondence exists between the energy lost from the primary electrons due to ionization processes in the specimen and the radiation damage sustained by the specimen. If the behaviour of the radiation damage process in l-valine between 0.05 and 3MeV is described in terms of a functional dependence upon β, the best data fit is to a β^3 relation, shown also in Fig. 3. It is pertinent to note that this β^3 relationship of the energy dependence of radiation damage has also been observed in polyethylene but over a less extensive energy range (8).

Thus with increasing electron energy a lower proportion of the energy lost by the electron beam is dissipated as radiation damage. From Table I it is apparent that the consistency in the comparison of these data with stopping power theory is best maintained at higher energies.

In terms of the advantages of increased image resolution from the use of very high voltage electron microscopy, in l-valine the most significant advantage is already attained by 0.5MeV. Compared to the value at 100kV this corresponds to an improvement in resolution by a factor of approximately two. The resolution values shown in Fig. 3 are derived from the signal to noise requirements of a statistically noisy image (9,4), with assumed values of a collection efficiency of 25%, a contrast of 10%, and a visibility factor of 5. Such a calculation assumes that the specimen thickness is adjusted as necessary to give constant contrast and that with increasing electron energy any

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loss in sensitivity of the recording photographic emulsion is compensated by an increase in its resolutuion. The effect of resolution compensation has been shown to be characteristic of certain commercial emulsions for electrons of energies less than 700keV (2). These results indicate therefore, that an improvement in resolution of a factor of 2 can be expected by working at 0.5MeV compared to 100kV.

ACKNOWLEDGEMENTS

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0 0 0 0 4 3 0 5 9 9 9

FIGURE CAPTIONS

Fig. 1. An electron micrograph and diffraction pattern of l-valine taken at 650kV. The image, although recorded after the diffraction pattern, still retains some crystallinity evidenced by the black extinction contours.

Fig. 2. The data contributing to the determination of the critical exposure of l-valine at 200kV showing the lifetime of the lowest order crystalline spectra for various values of the electron dose rate to the specimen. The slope indicates that the critical exposure is independent of the dose rate.

Fig. 3. The variation of the critical exposure N_{cr} with increasing electron energy. The relationship of β to the expected resolution attainable from an image is also shown. A variation of β^3 in N_{cr} vs electron energy is obtained.

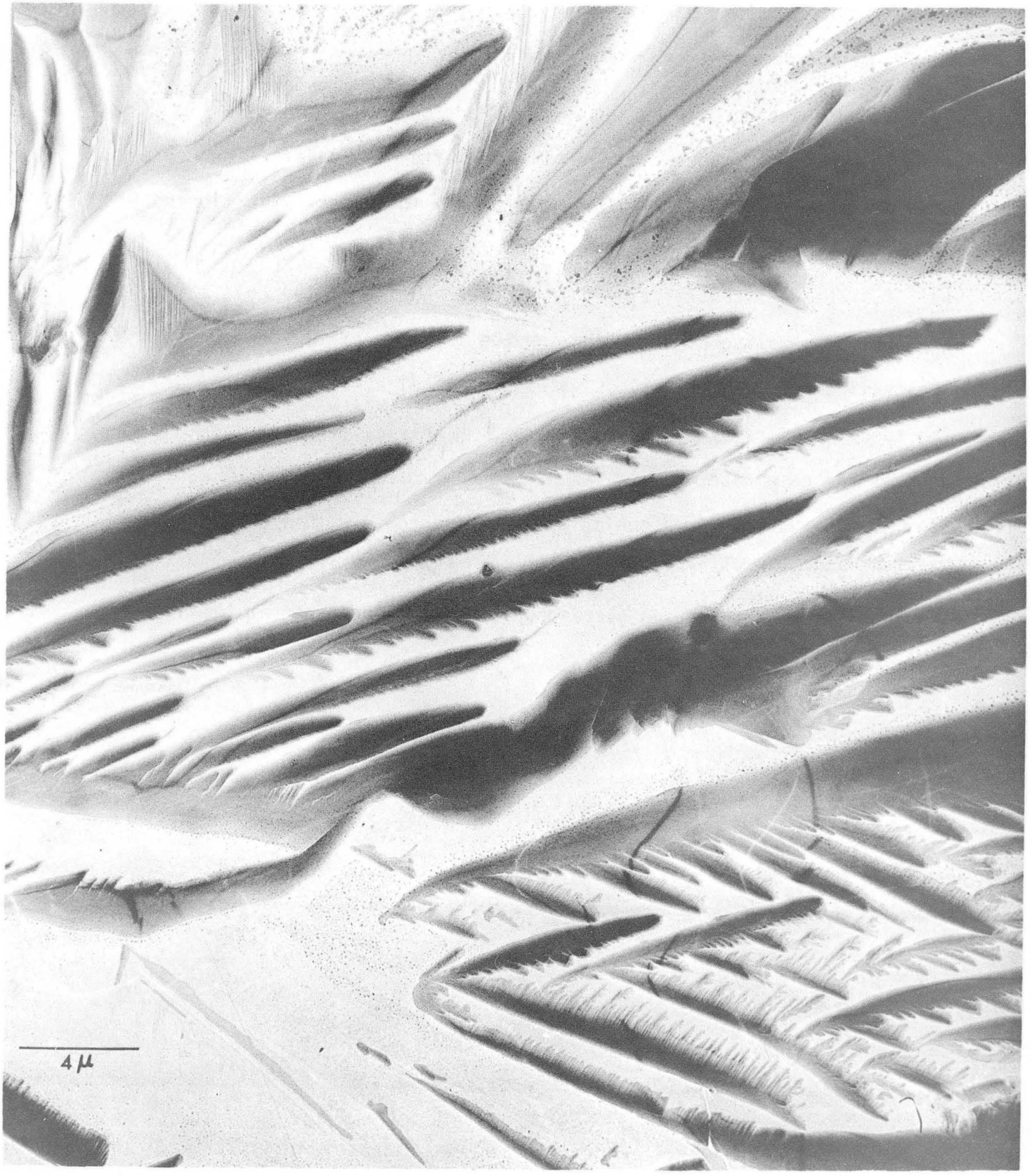

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Fig. 1

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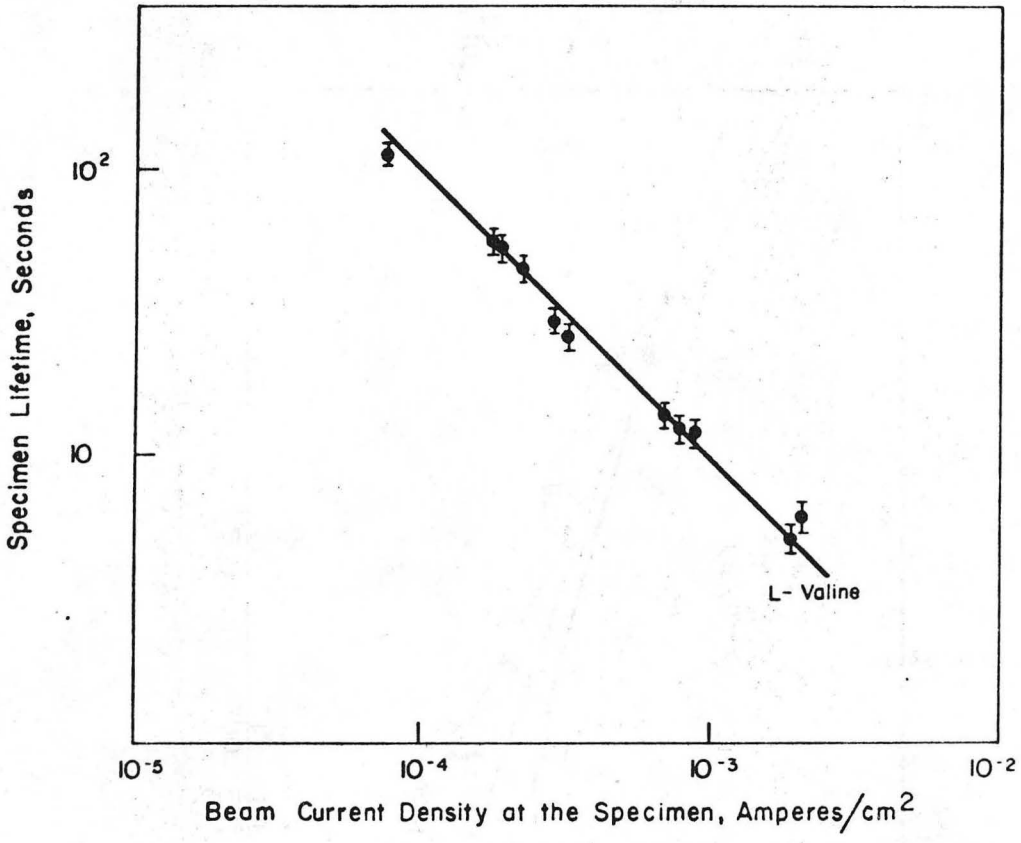

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Fig. 2

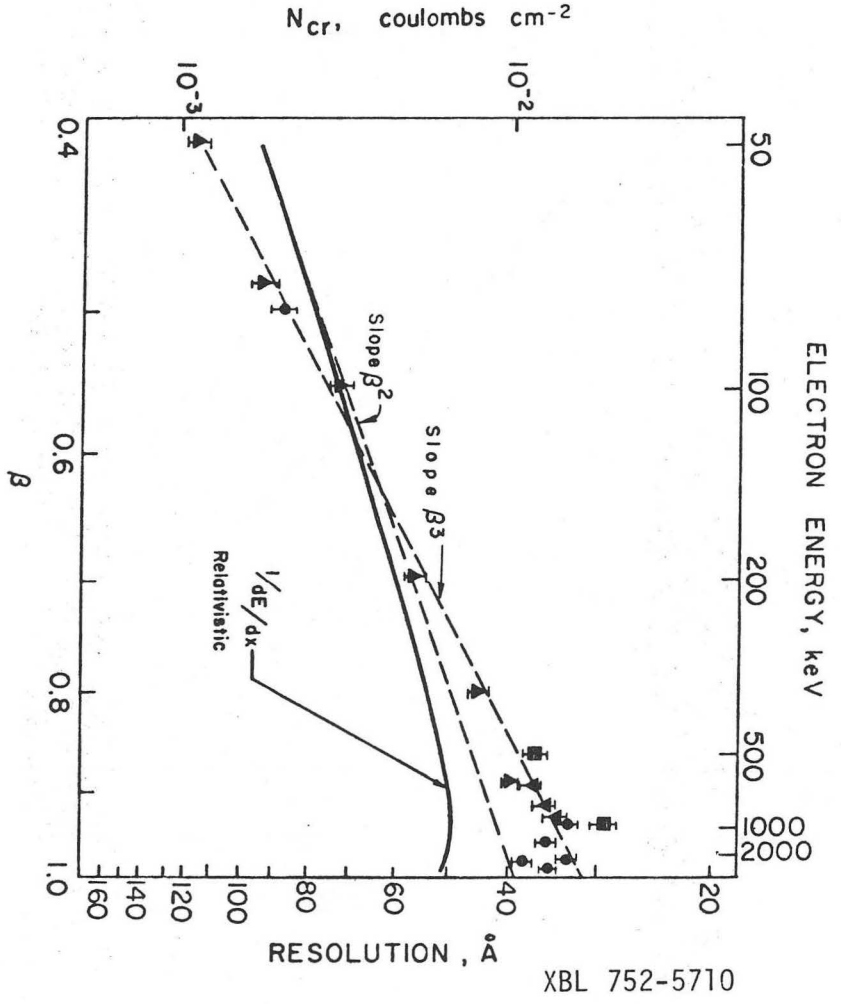
N_{cr}, coulombs cm^{-2}
10^{-3}
10^{-2}
0.4
0.6
0.8
1.0
β
Slope β^2
Slope β^3
Relativistic
$1/dE/dx$
50
100
200
500
1000
2000
ELECTRON ENERGY, keV
160
140
120
100
80
60
40
20
RESOLUTION, Å
XBL 752-5710

Fig. 3

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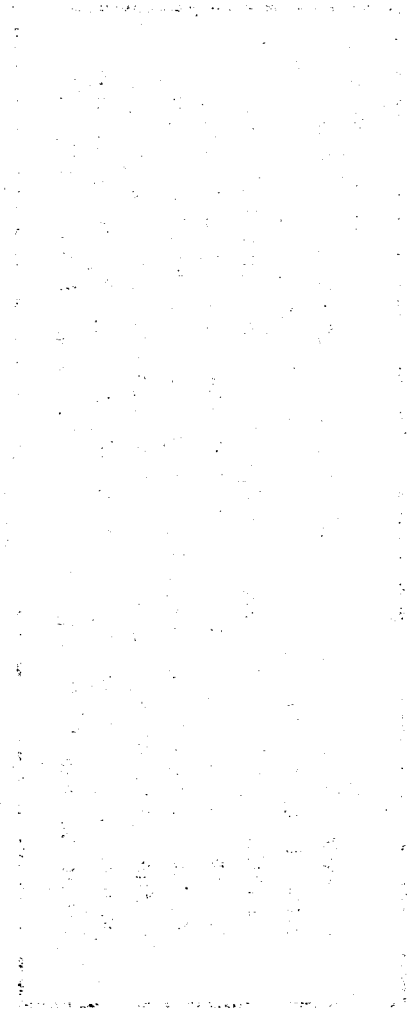
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