Optimizing replay intensity and resolution in aberration-compensated off-axis holograms by ambient humidity control

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Abstract: In hologrammetry it is desirable to reconstruct the real image rather than the virtual image as the latter must be viewed at a distance through the window of the holographic plate itself. When a scene is located in water but the image is replayed into air, it is necessary to correct for the refractive index difference by reconstructing the image with shorter wavelength illumination and changing the beam angle to satisfy the grating equation. However, this means that the Bragg condition may no longer be satisfied during replay, reducing the diffraction efficiency and decreasing the signal-to-noise ratio of the reconstructed images. Changing the replay beam angle to satisfy better the Bragg condition makes the images brighter but also renders them unusable by increasing the optical aberrations. Our solution is to alter the Bragg properties of the hologram by altering the humidity of the surrounding atmosphere. This approach has been experimentally demonstrated for Agfa 8E56HD emulsions by measuring the brightness and resolution of a reconstructed real image from an off-axis hologram over a humidity range from 6 to 93 per cent. The emulsion swelling and its effect on the Bragg properties of the hologram were modelled using the Flory–Huggins theory of polymer swelling.

Keywords: holographic emulsions, photographic emulsions, emulsion swelling, emulsion shrinkage, humidity

1 INTRODUCTION

When using holography for dimensional measurement (hologrammetry) it is often much more convenient to reconstruct the real image, with which it is possible to interact directly, rather than the virtual image which must be viewed at a distance through the window of the holographic plate itself. In applications where the recorded scene is in water, replay is usually into air instead, making it necessary to correct for the refractive index difference [**1**]. This can be done by reconstructing the image with a shorter-wavelength illumination than used for recording, accompanied, for off-axis holograms, by a corresponding change in the replay beam angle to satisfy the grating equation. However, these changes mean that the Bragg condition may no longer be satisfied within the emulsion. This reduces the diffraction efficiency, making the reconstructed images faint and resulting in a poor signal-to-noise ratio (SNR).

For example, the HoloMar collaboration has built both HoloCam, an underwater holocamera for the *in situ* recording of plankton species and distributions, and HoloScan, an associated hologram reconstruction and analysis instrument. HoloCam uniquely incorporates simultaneous in-line and off-axis holography of overlapping sample volumes (Fig. 1a), allowing the recording of organisms over a wide range of sizes and concentrations [**2**]. Holograms are recorded on glass plates using a *Q*-switched frequency-doubled Nd:YAG (neodymium-doped

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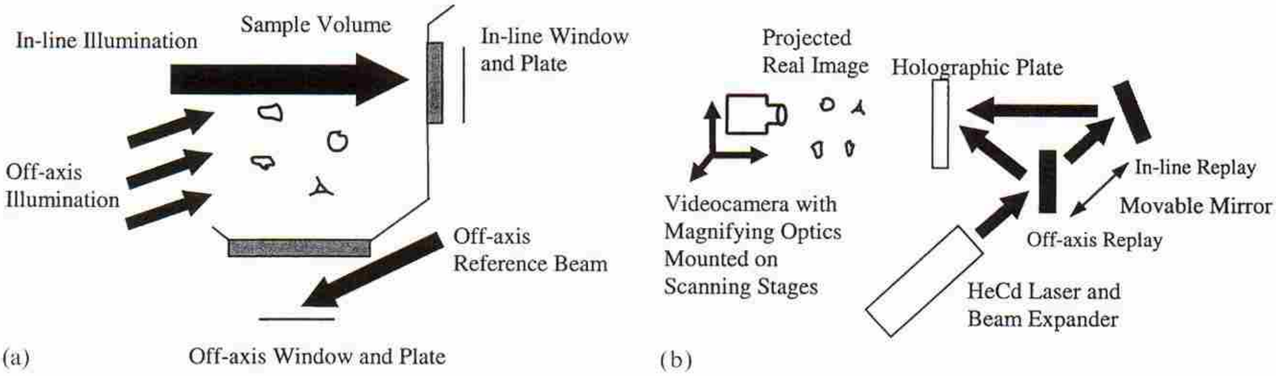

(a) (b)

Fig. 1 Schematics of (a) HoloCam sample volume and (b) HoloScan replay system

yttrium aluminium garnet) laser, which operates at a wavelength of 532 nm and a pulse duration of less than 10 ns to freeze any motion. Up to 20 in-line and 25 off-axis holograms can be exposed during a dive. Manual analysis of large volumes containing thousands of particles is, however, an enormous and time-consuming task, with operator fatigue an unpredictable source of errors. The overall purpose of the data extraction system [**3**] is to locate and identify automatically the various microscopic organisms within the sample volume, allowing for the first time a quantitative analysis of the spatial relationships between both individuals and species and thus improving our understanding of fundamental biological and chemical processes in the upper layers of the oceans.

The presence of the thick window (and 120 mm air space for the off-axis reference beam), combined with the desire to capture large (up to 100 l) volumes makes it impossible to study microscopic objects throughout the sample volume from the virtual image. Thus, although the objects are located in water, image replay is carried out in the laboratory in air using the projected (real) image mode of reconstruction. This change in refractive index can introduce significant aberrations, particularly with off-axis holography, which may be partially corrected by replay at a shorter wavelength [**4**]. The holocamera's window thickness and the air gap to the plate have therefore been optimized for reconstruction with 442 nm illumination from a He–Cd laser.

A schematic diagram of the HoloScan replay machine is shown in Fig. 1b. The holographic plate is illuminated by the collimated beam from a 180 mW He–Cd laser. A movable mirror allows the replay system to be easily switched between in-line and off-axis modes. A charge-coupled device (CCD)-based video camera is traversed through the projected real image on computer-controlled micropositioners, which cover a sample volume up to 200 mm across and 1000 mm deep in 10 μm steps, allowing individual organisms to be isolated and located [**3**]. The video output is captured by a frame grabber and then processed to clean up the image and to identify the true focal plane of each object within the three-dimensional sample volume.

Although the replay laser used here is among the most powerful commercially available He–Cd lasers and the video camera was chosen because of its sensitivity in the blue, many of the off-axis holograms examined during testing produced images too dark to see clearly. This is because the changes in the illumination wavelength and direction mean that the Bragg condition is no longer satisfied during replay, reducing the diffraction efficiency. Changing the replay beam angle to satisfy better the Bragg condition makes the images brighter but also renders them unrecognizable by introducing severe optical aberrations. A possible solution is to alter the Bragg properties of the hologram; in particular, the emulsion thickness can be conveniently controlled by altering the humidity of the atmosphere surrounding the hologram without causing any long-term changes or damage to the holographic plate.

2 REPLAY OF UNDERWATER HOLOGRAMS

2.1 Background

Although the scene recorded is in water, the real image is reconstructed in the laboratory in air. The significant aberrations associated with the refractive index change may then be corrected for by replaying with a shorter wavelength [4]. However, the incidence angle of the replay beam must also be adjusted, so that the illuminating beam gives the same phase distribution across the holographic plate; otherwise additional aberrations will be introduced. This condition is derived from the grating equation and can be expressed as

$$\frac{\lambda}{\sin\theta_{\mathrm{E}}} = \text{constant} \qquad (1)$$

where λ is the wavelength in air and θ_{E} the angle of incidence of the replay beam on the holographic plate. In the case of HoloCam, off-axis holograms of objects in water are recorded with a 532 nm reference beam incident at 60°. These are replayed in air at 442 nm implying a replay beam angle of 46°. Although equation (1) is sufficient to describe a thin hologram, in many cases Bragg effects can be important even if they do not dominate the diffraction process. For example, off-axis holograms from HoloCam can neither be treated as thin holograms according to the formalism of Solymar and Cooke [5] nor do they satisfy the criteria of true volume holograms. The holograms are encoded within the photographic emulsion as a set of fringes that take the form of many parallel planes, similar to a Venetian (louvre) blind. The change in replay beam angle accompanying the wavelength shift leaves the incident illumination unable to pass through the fringe structure. In the intermediate regime between thin and thick holograms, equation (1) may be thought of as governing the reconstructed image fidelity or aberrations while the Bragg properties (grating spacing and slant angle) control only the image intensity.

Off-Bragg replay is possible but, to obtain useful images, a cooled extra-sensitive (expensive) camera would be required, and the SNR would remain poor, as effects such as emulsion scatter are much less sensitive to the illumination direction than the Bragg selectivity of the grating, so that off-Bragg images tend to suffer from relatively high levels of background noise.

2.2 Controlling emulsion swelling

Since most replay parameters of holograms are predetermined at recording [the replay wavelength by the recording wavelength and layout, and the beam angle by equation (1)], it is desirable to find a mechanism for tuning the Bragg properties of the holographic plates to improve their diffraction efficiency. As they are a property of the emulsion itself, the spacing and orientation of the fringes can be altered by swelling or shrinking the emulsion layer. The swelling mechanism should not distort the reconstructed image or increase noise, and the swollen state must be either archivally stable or easily reproducible.

Bjelkhagen [6] discussed a number of techniques for the controlled swelling of holographic plates. Probably the best known is the use of triethanolamine (TEA) for colour selection in reflection holograms. Unfortunately this tends to cause extra scattering and distortion of the holographic image and, since the TEA is gradually lost from the emulsion, the swelling is not fixed over the possible working life of the hologram (possibly decades in applications such as coral reef surveys). Walker and Benton [7] successfully used a series of solutions of propan-2-ol in water in different concentrations to swell emulsions by various degrees, but this approach introduces the complications of working with a liquid-filled gate. Angell [8] suggested adding an organosilane to the fixer. This binds to the gelatin, cutting down hydrogen bonding between gelatin molecules and thus permanently swells the emulsion slightly, but little is known about the effects on the reconstructed image and the scattering properties of the emulsion, or its long-term stability.

Several alternative approaches are based on impregnating the swollen emulsion with an agent that then solidifies *in situ*, permanently distending the emulsion. For example, the method due to Young [9] is to swell the developed emulsion with an aqueous solution of *n*-vinyl 2-pyrrolidone (NVP) and to expose the holographic plate to ultraviolet light, causing the NVP to polymerize within the emulsion. Rallison [10] described a similar method using instead a solution of an optical epoxy. Wreede and Knobbe [11] suggested a more involved technique wherein an emulsion on a porous substrate is exposed to two species (such as water vapour and silane) that chemically react with each other within the microstructure of the gelatin, filling the interstitial voids and making the emulsion resistant to swelling or shrinkage due to moisture variations. While these approaches might allow the permanent fixed swelling of holograms without the need for a sealing step, again too little is known about the effects of such processes on the replayed image and the scattering properties of the emulsion for their immediate use in our application.

The approach described here is to alter the ambient humidity around the emulsion during replay. This produces a very strong swelling and shrinking effect that is easily reversible and repeatable, which should not affect or damage the emulsion (which must of course survive the swelling associated with the development process). Unlike most of the methods mentioned above, altering the ambient humidity also allows rapid selection of *any* arbitrary degree of swelling.

3 HUMIDITY AND HOLOGRAPHIC EMULSIONS

Although a number of workers have studied the effects of humidity on emulsions [**12**], very few data appear to have been published that quantify the relationship between the swelling of an emulsion and the relative humidity (RH) of its surroundings. Wuest and Lakes [**13**] investigated this for colour control of bleached white-light holograms recorded on Agfa 8E75HD plates. They found that the replay wavelength increased approximately linearly at normal laboratory humidities (20–70 per cent RH) and then changed very rapidly above 80 per cent RH.

Spooncer *et al.* [**14**] evaluated the possibility of using the swelling of Agfa 8E75HD plates as a humidity sensor. They also found that the peak reconstruction wavelength of their holograms varied approximately linearly from 2 to 70 per cent RH and then started to increase rapidly.

Spooncer *et al.* [**14**] and Wuest and Lakes [**13**] both looked at the optical effects of emulsion swelling, rather than measuring the water uptake directly; hence in order to obtain a better understanding of the effects of humidity on emulsions, data for hardened gelatin have also been considered.

Data exist for gelatin bonded to a substrate and thus constrained in two dimensions [**15**], and for lumps of material free to expand in all directions [**16, 17**]. In all three cases the quantity actually measured was the mass of water absorbed into the gel, which is here converted into a mechanical swelling by assuming volume additivity.

One complication described by Sheppard *et al.* [**16**] and Spooncer *et al.* [**14**] is that of hysteresis. Hardened gelatin was found to stay in a more swollen state during the drying process than during wetting. The same dimensional state is reached at a 10 per cent RH difference around 50 per cent RH. Sheppard *et al.* found the swelling of plain gelatin and of hardened types to have similar variations with humidity.

Owing to the lack of published data relating the swelling of photographic emulsions to atmospheric humidity, we modelled this process using a simplified form of the Flory–Huggins equation. According to Huggins [**18**] it is possible to relate the activity a_S of a solvent to the volume fraction ϕ_S of solvent and volume fraction ϕ_P of polymer in a solvent–polymer mixture by

$$\ln a_S = \ln \phi_S + \left(1 - \frac{\tilde{V}_S}{\tilde{V}_P}\right)\phi_P + \chi\phi_P^2 \qquad (2)$$

$\tilde{V}_S$ and $\tilde{V}_P$ are the molar volumes under ambient conditions of the solvent and polymer respectively, and for long-chain polymers and simple molecular solvents their ratio is negligible. The Flory–Huggins parameter χ represents the interaction of a particular polymer and solvent combination.

If the solvent vapour can be treated as an ideal gas, then its activity a_S can be expressed as the ratio of the partial pressure of the solvent over the solution to the saturated vapour pressure of the pure solvent:

$$a_S = \frac{p_S}{p_{S,0}} \qquad (3)$$

For water this ratio is known as the relative humidity H_r. Since $\phi_S + \phi_P = 1$, equation (2) can be rewritten in terms of the solvent volume fraction only to give

$$\ln H_r = \ln \phi_S + (1 - \phi_S) + \chi(1 - \phi_S)^2 \qquad (4)$$

Equation (4) relates the emulsion swelling to the ambient humidity H_r using only a single experimentally determined parameter χ. Given that, when washing a photographic plate in water the emulsion generally swells to around five times its original thickness [**6**], equation (4) implies that χ is about 0.6 for holographic emulsions. An important consideration with regard to the validity of equation (4) is that the theoretical basis of the Flory–Huggins equation assumes free (unconstrained) *non-polar* polymers and solvents, and so our expectations should not be too high when using it to model gelatin bonded to glass plates and soaked with water.

Figure 2 shows the swelling of gelatin by ambient humidity, with both experimental data and the predictions of equation (4) using a Flory–Huggins parameter of 0.62 (a value based on the 92 per cent RH data point of Gehrmann and Kast [**15**]). The general trend of the data is similar to the findings of Wuest and Lakes [**13**] described above. The Flory–Huggins

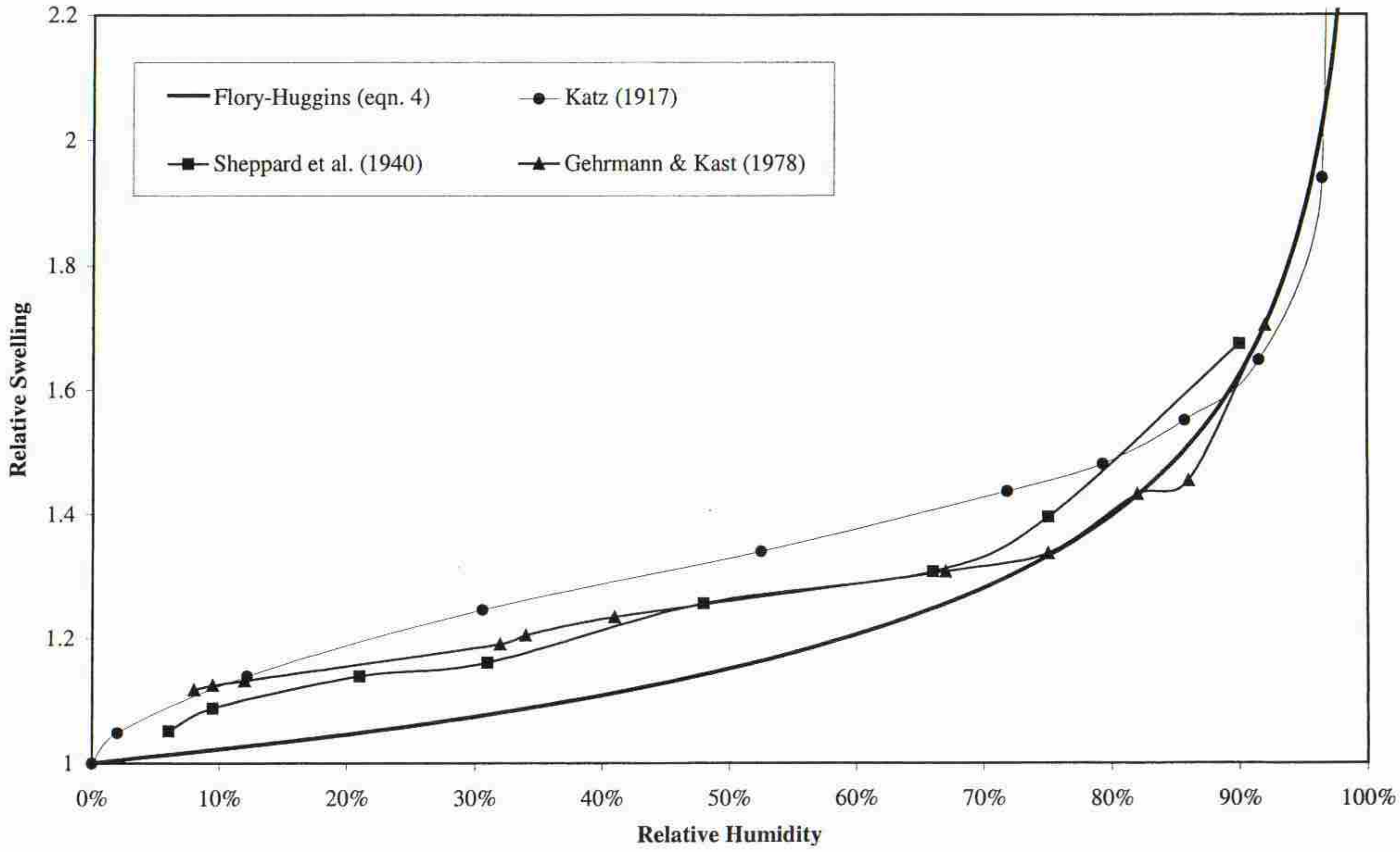

Fig. 2 Gelatin swelling by humidity. A comparison of the Flory–Huggins model [equation (4) with $\chi = 0.62$] with three sets of experimental data

equation can be seen to provide a reasonable fit to that data at higher humidities (and thus the majority of the swelling range) but noticeably underestimates the swelling below about 70 per cent RH.

The Flory–Huggins equation was used to estimate the ambient humidity needed during replay in order to give the 10 per cent swelling required by HoloMar holograms, as discussed in section 2.2. Since HoloCam is sealed and purged with dry nitrogen during deployment to prevent condensation, exposure is assumed to occur at 0 per cent humidity and, from the data shown in Fig. 2, it is expected that optimum replay should require about 30 per cent RH. Thus, replay at normal laboratory humidities of 40–60 per cent should naturally correct for the wavelength shift associated with our final system (indeed, the underestimation of the measured values by the Flory–Huggins equation indicates a risk of overcompensation). Other situations are more complex; some of our holograms of collected plankton in laboratory tanks have been recorded at ambient humidities of over 70 per cent and thus require playback at humidities in excess of 90 per cent.

The dry atmosphere to be used in the HoloCam has other implications. Excessive shrinkage may cause the emulsion to peel from the substrate, or microcracking leading to increased light scattering within the emulsion. There may also be extra fogging of the plates due to static discharge. On the other hand, oxygen and water are known to have desensitizing effects on emulsions [**19**] so that a dry nitrogen atmosphere may even improve the plate sensitivity.

4 EXPERIMENTAL VALIDATION OF APPROACH

To demonstrate the validity of using humidity changes to tune emulsion properties during replay, the variation in the brightness and perceived resolution of a reconstructed real image from a laboratory-recorded hologram with replay beam angle have been measured over a range of humidities (Fig. 3). The image of a USAF 1951 resolution target has been projected from holograms recorded on Agfa 8E56HD 4 in by 5 in glass plates and mounted in a glass-sided enclosure fitted with a humidity sensor (Hycal/Honeywell humidity sensor HIH 3602-C, with an accuracy of ±2 per cent). The plates were mounted on a rotary stage to allow the replay angle to be changed. The air supply was fed from a cylinder of compressed dry low-hydrocarbon air (BOC BTCA74 ‘laser’ air; less than 3 ppm by volume of hydrocarbons), via a bubbler filled with distilled water with a fine-bore needle valve connected in parallel with it. The moisture content of the air flow could thus be set to any chosen value by a suitable adjustment of the valve and held steady for a sufficiently long period of time that the emulsion could reach equilibrium. To avoid hysteresis effects the humidity was generally increased between successive runs. The hologram was illuminated with a 442 nm He–Cd laser over a range of replay angles. The real image was projected directly on to the detector of a CCD camera (JAI CV-M300, 752 × 582 11 μm pixels, automatic gain control disabled) and the perceived resolution found directly from a live monitor. Image brightness was measured by capturing the image and finding the average pixel intensity over a fixed region of the bright background of the resolution target. The camera system was checked for linearity, allowing the pixel values to be scaled to compensate for changes in laser power. The air flow was stopped while taking measurements, to avoid any bowing of the holographic plate with pressure.

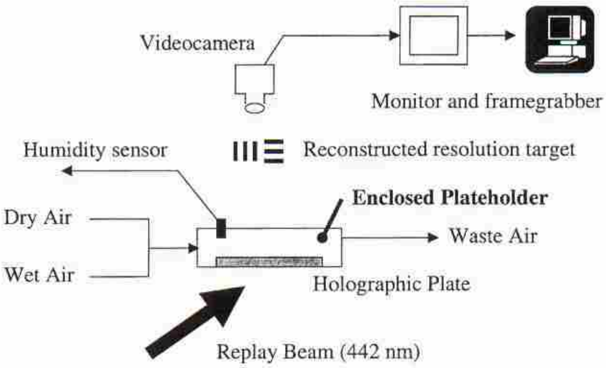

Fig. 3 Replay system with a controlled humidity plate holder

Two amplitude holograms were recorded in the HoloCam laboratory mock-up (i.e. with 532 nm illumination at 60°) with the resolution target 300 mm from the plate, developed with a metol–ascorbic acid developer and fixed normally. Plate 1 was recorded at an unknown humidity (estimated 60–70 per cent RH) and with no water in the sample volume. The change in wavelength on replay will in this case tend to cause rather than minimize aberrations. Plate 2 was recorded at 66 per cent RH with the sample volume filled with water. Although the latter is a more appropriate choice, a flaw in the plate holder design (the outer edge of emulsion face of the plate was the sealing surface clamped tightly against an O-ring) caused plate 2 to crack at high swelling.

For both plates, equation (1) predicts that aberrations will be minimized at a replay angle of 46°. However, interest lies in maximizing the detail that can be studied using our existing video equipment. The effective resolution of the image recorded by the camera depends on both the intrinsic resolution of the reconstructed image and its brightness; the most perfectly reproduced image is useless if it is too faint to be seen.

Figure 4 shows that, at low humidities, only mediocre resolution can be obtained from plate 1, and that the best occurs at a beam angle of nearly 47°. As the humidity is increased, the resolution at larger beam angles remains the same but there is a steady improvement in the best resolution attainable with smaller angles of incidence. The reasons for this can be seen in Fig. 5; the drier emulsions reach their peak diffraction efficiency with beam angles of about 60° (albeit with excessive astigmatism) while not producing any discernible image when illuminated at 46°. As the humidity increases, the emulsion swells, the Bragg selectivity improves and the Bragg angle decreases towards the expected optimum replay angle. The improving visibility of the target increases the discernible resolution. When the humidity has reached 90 per cent the brightest replay occurs at the same beam angle as the least aberrated replay, and the camera can therefore pick out the finer detail and thus make use of the intrinsically higher-resolution image so that it is possible to see detail twice as fine as could be made out with the driest emulsion.

Two data sets for 49 per cent RH are presented; these represent data obtained at the same ambient humidity but separated by several swelling–drying cycles. It is almost impossible to distinguish between these two curves in Fig. 5, which confirms both the repeatability of the apparatus and that the process does not cause lasting changes to the emulsion. Furthermore, several runs were made with the emulsion being dried [labelled (d)], and it may be noted that the two 49 per cent RH rising and the 43 per cent RH falling curves (and also the 64 per cent RH rising and the 54 per cent RH falling curves) are again almost identical. This indicates that the emulsion was swollen to a similar degree at about an 8 per cent RH difference between the wetting and drying cycles, which is consistent with the amount of hysteresis reported by Sheppard *et al.* [**16**].

The intensity measurement procedure discussed above has the shortcoming that significant optical aberrations will cause an apparent reduction in optical efficiency, due to the spreading of light from the bright regions of the reconstructed image into the dark regions. It is believed that this is the explanation for the maximum image brightness falling with decreasing ambient humidity.

The data from plate 2 show the same trends as those from plate 1. As the emulsion swells with increasing humidity, the resolution of the images improves. In Fig. 6 the image brightnesses at two humidities are compared with the corresponding values from plate 1. From this it can be seen that the two separate measurements at 49 per cent RH for plate 1 demonstrate excellent reproducibility. The optimum replay angles are the same for equal plate humidities. This demonstrates that the same humidity optimizes image brightness for more than one hologram.

As the shrinkage of the emulsion during hologram processing has not been measured precisely in these experiments, not all the data that the model requires to predict the effects of humidity were obtained; the fringe spacing and slant angle in the emulsion during replay are not known. However, as the emulsion is constrained to swell or shrink only in a direction normal to the plane of the holographic plate, the fringe spacing along the plane of the plate is known from the recording conditions. As the changes in the fringe spacing and slant angle with swelling are related geometrically, there is only one unknown parameter: the slant angle at zero humidity. This has been predicted by assuming that the swelling of the emulsion is described by the data of Gehrmann and Kast [**15**] shown in Fig. 2 and fitting the predicted replay angle giving the brightest image as a function of humidity with our measured values (Fig. 7). This estimated slant angle could then be used with the Flory–Huggins swelling model, using $\chi = 0.62$. The refractive index of the moist emulsion, n_{emul}, can be estimated using Gladstone's relation for mixtures:

$$n_{\text{emul}} - 1 = \phi_1(n_1 - 1) + (1 - \phi_1)(n_2 - 1) \qquad (5)$$

It can be seen that the observed and predicted replay angles based on the data of Gehrmann and Kast are in close agreement for both our sets of experimental data. This suggests that the results obtained by Gehrmann and Kast are a reasonable representation of the swelling of the 8E56 holographic emulsion.

Although the predictions of the Flory–Huggins

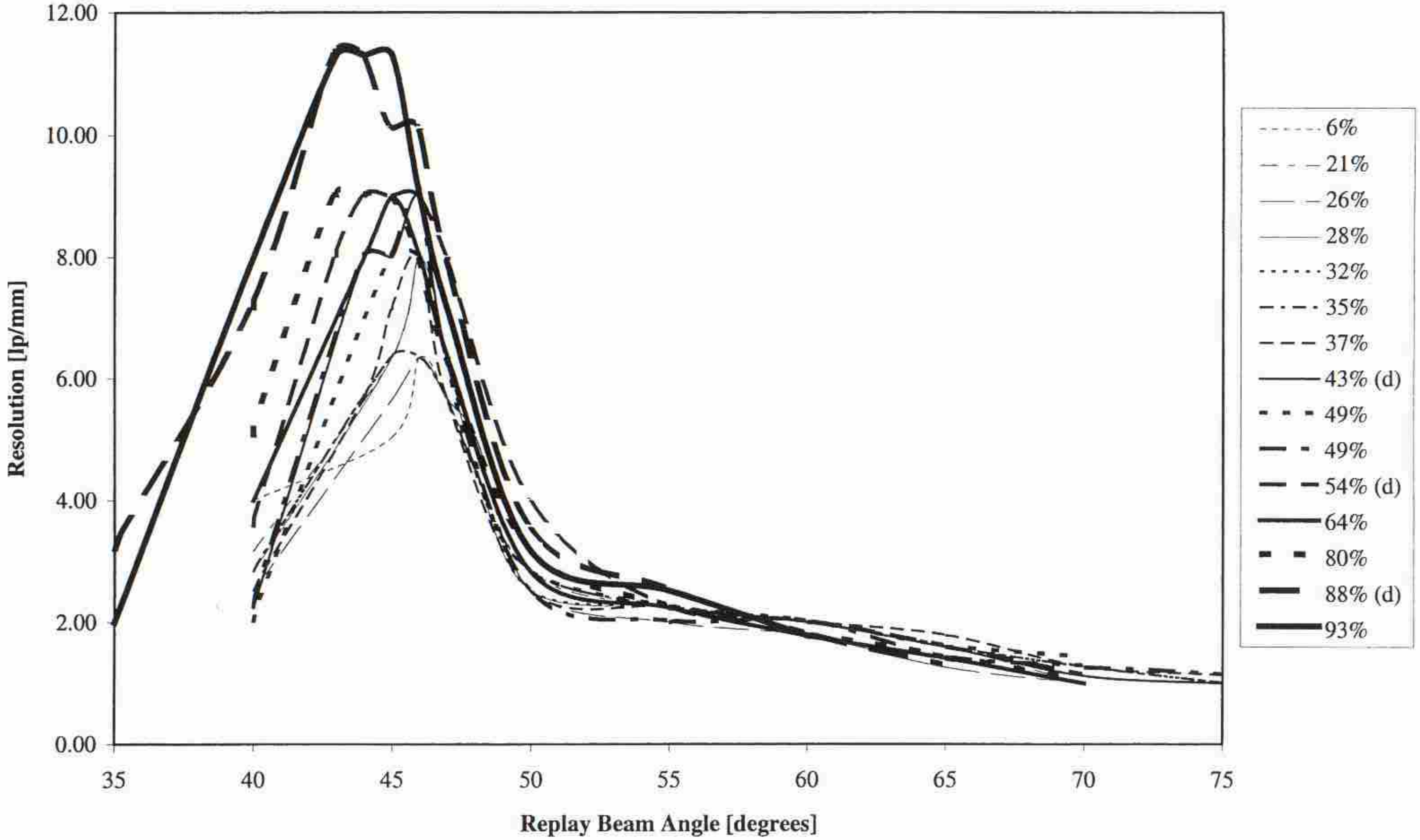

Fig. 4 Variation in reconstructed image resolution (plate 1) with replay beam angle, as a function of plate-holder relative humidity [with the humidity increasing to the quoted value, except for runs labelled (d) when it decreased to that value]

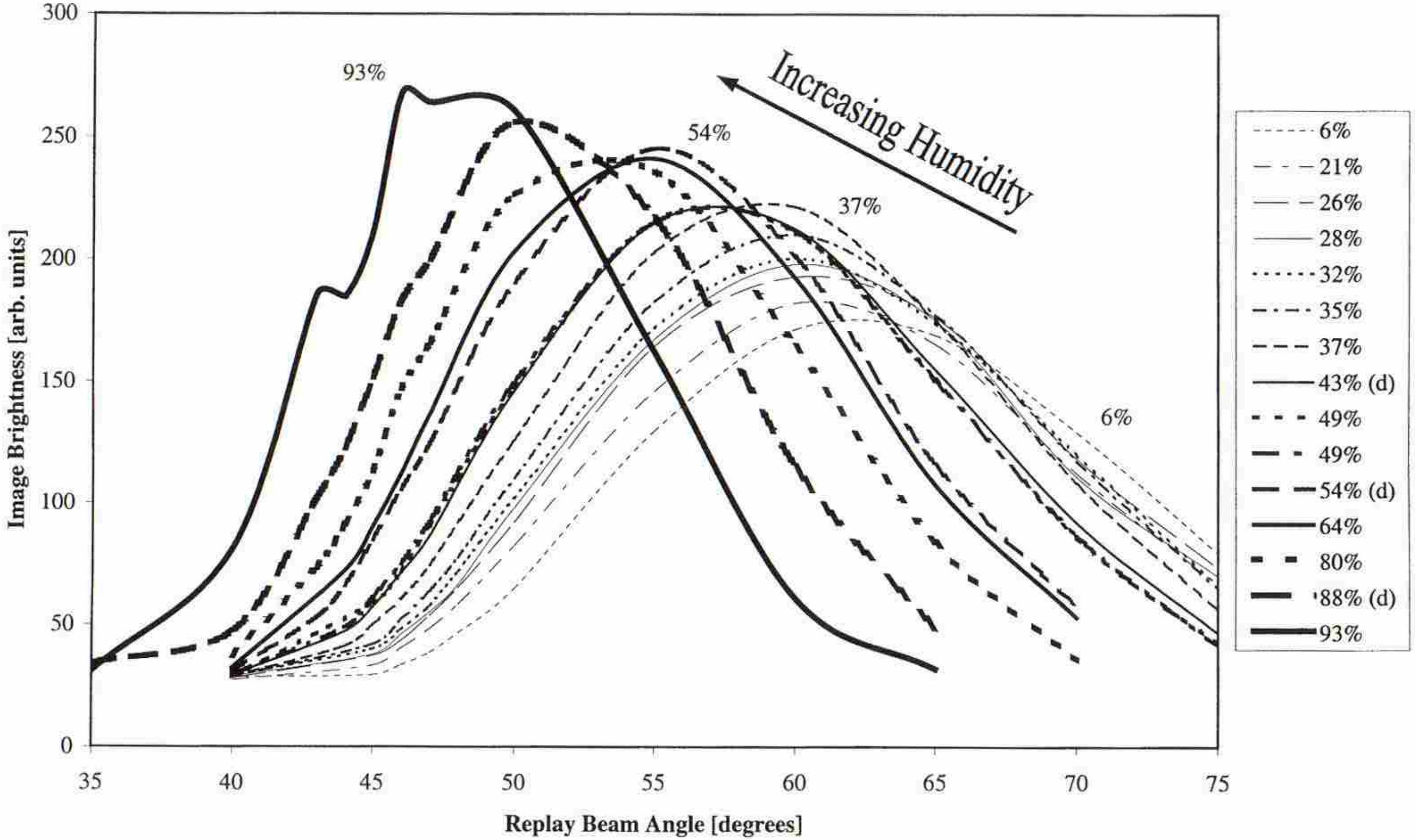

Fig. 5 Variation in reconstructed image brightness (plate 1) with replay beam angle, as a function of plate-holder relative humidity [with the humidity increasing to the quoted value, except for runs labelled (d) when it decreased to that value]

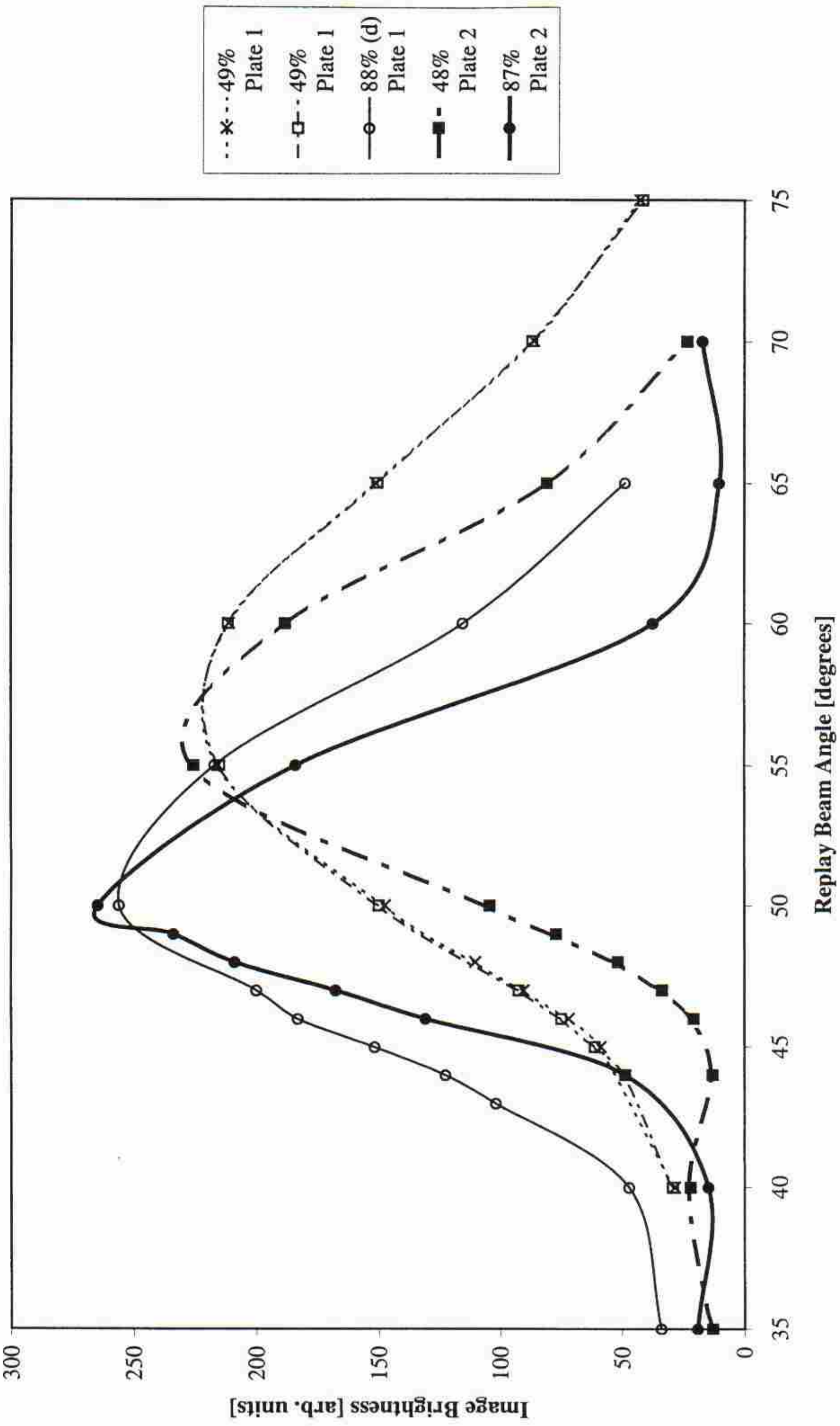

Fig. 6 Variation in image brightness with replay beam angle at two relative humidities, for plates 1 and 2

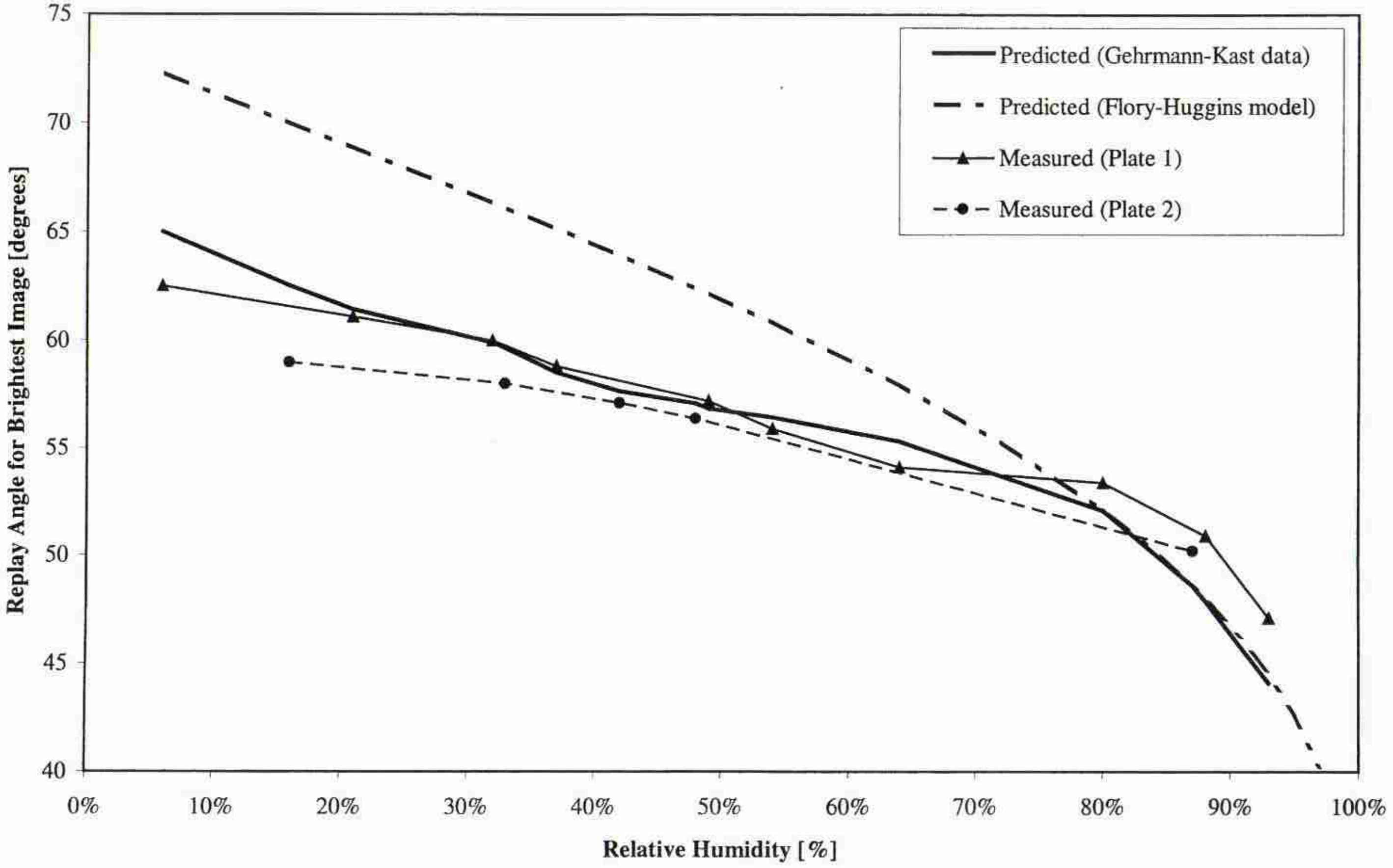

Fig. 7 Comparison of measured replay beam angles giving the brightest image as a function of humidity with predicted values. The predictions are based on the swelling data of Gehrmann and Kast and on a pure Flory–Huggins model with $\chi = 0.62$

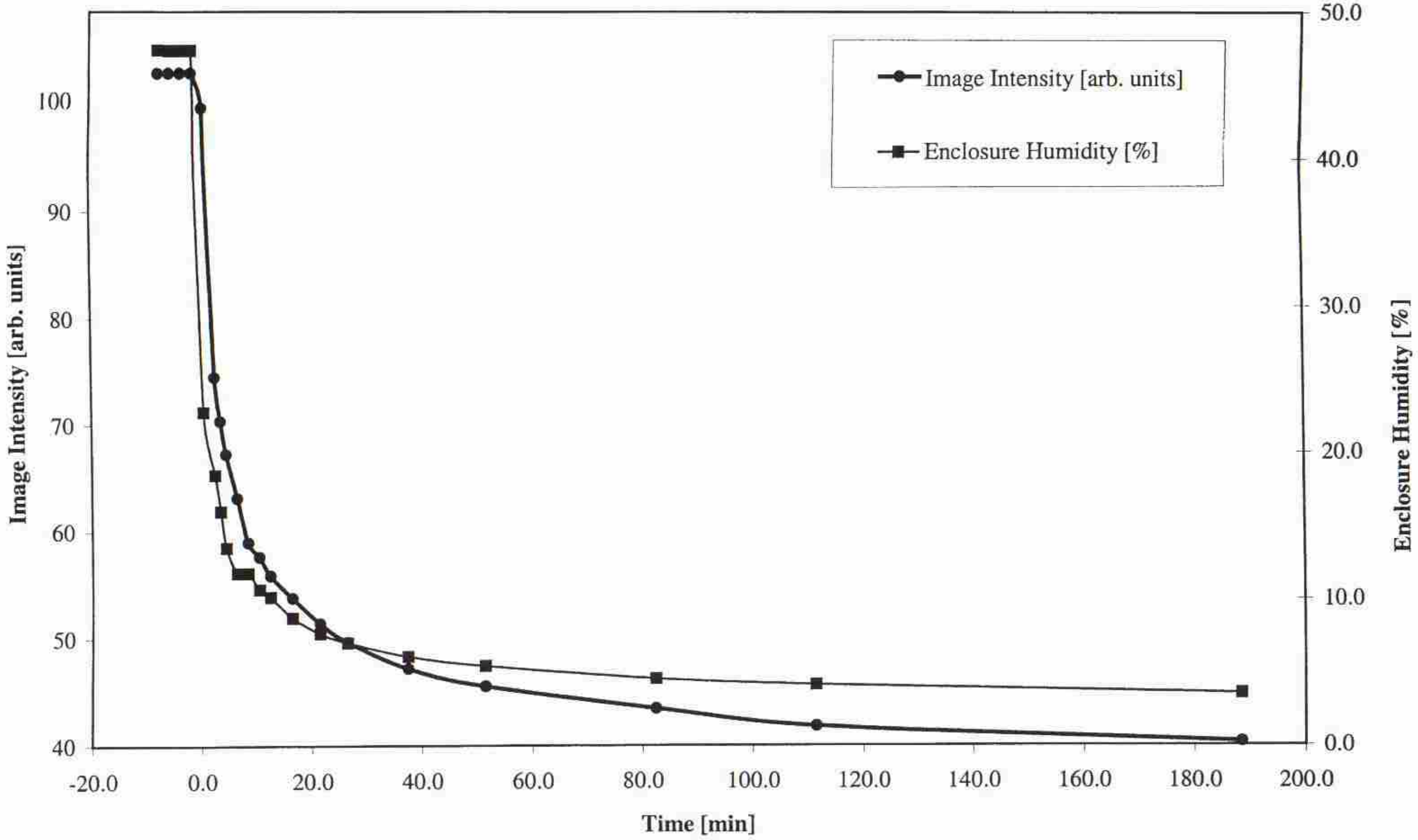

Fig. 8 Change in image intensity and ambient humidity inside the plate-holder enclosure with time. Dry air was passed continuously through the enclosure from time zero

model look significantly worse, the discrepancy primarily exists over the humidity range where swelling effects are small and their consequences for hologram replay least significant. At high humidities (greater than 80 per cent) its predictions agree closely with those based on the swelling considered by Gehrmann and Kast. The Flory–Huggins model is also conveniently simple, requiring only a single parameter χ to be known, whether from the physicochemical properties of the emulsion and solvent or, if reliable data on fringe spacing and slant angle were available, from a fit to the observed optical properties.

5 DYNAMIC RESPONSE

In order to gain an idea of the speed with which the emulsion can react to changes in environment, the humidity inside the plate enclosure and the image brightness were measured while the plate holder was suddenly flushed with dry air. Unfortunately the apparatus was not optimized for dynamic measurements; therefore in practice the emulsion's response here is limited by the rate at which moisture can be removed from the plate holder; however, as can be seen from Fig. 8, the emulsion undergoes significant changes in much less than a minute. Most of the shrinkage occurs within 20 min, which is in accord with the experience of Wuest and Lakes [**13**] and noticeably more rapid than the time-scale of hours encountered with dichromated gelatin emulsions 30 μm thick by Naik *et al.* [**20**].

6 CONCLUSIONS

A practical prototype controlled-humidity plate holder has been built and fitted to an existing hologram replay and analysis system. Using this it has been shown that controlling the ambient humidity is a viable technique for optimizing the emulsion thickness of off-axis transmission holograms on silver halide materials. This allows simultaneous optimization of replay for both minimum aberrations and maximum diffraction efficiency, which substantially improves the quality of extracted images. The measured responses of the holographic emulsion match the observations of previous workers on both holograms and bulk gelatin, as well as a simple model. This has been used to demonstrate the active tuning of hologram properties to provide optimized replay, and the emulsion has been shown to respond to humidity changes within a conveniently short period of time.

A simple model based on the Flory–Huggins equation has been introduced to model emulsion–humidity interactions. This uses only a single experimentally determined parameter. Although it is not useful at common laboratory humidities, it has been demonstrated that it describes emulsion swelling at high humidities, for which there is currently a lack of experimental data. It may be possible to improve this model by rederiving it incorporating more appropriate (polar solvent and polymer) thermodynamics. During a significant number of severe swelling–shrinking cycles, no damage or degradation of the emulsion has been observed.

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