The Clash Between Chaos and Symmetry in an Ancient Process:
The Working of Metals

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Abstract: Although the working of metals has been practised since antiquity, modern appreciation that metals are crystalline has revealed the puzzle of how they can be deformed and yet retain their crystalline symmetry. The solution requires acceptance of a dialectic relationship between a defect structure displaying random disorder superimposed upon that of the strictly ordered crystal.

Fig. 1 shows a model of Chinese Emperor Qin Shihuang's chariot. The original is buried near Xi'ang and is on display there with the Terra Cotta Warriors. It dates from about 200 BCE. The chassis and roof shown in grey are made from bronze sheet. The elegant roof is over 3 metres long but only a few millimetres thick. It is extraordinary that the ancient metallurgists were able to make it by casting, then deforming, the sheet: it is malleable bronze. The deformation strengthens it, makes it more self-supporting over the interior of the chariot. For this to be possible, the metal must be free from inclusions and very homogeneous in composition. The Emperor's chariots exemplify ancient skills of metal forming, remarkably comparable to those shown in modern automobiles.

Figure 1. A model of Chinese Emperor Qin Shihuang's chariot

Metals which can be shaped and strengthened without having to be cast in their final form play a central role in civilisation: steel armour, coins of gold or brass, lead roofing, cutlery and cookware, copper engravings, as well as aeroplanes, ships, and wheeled vehicles: all require knowing how to deform metals without causing them to crack or buckle. Useful deformable metals are nearly all of cubic crystalline atomic structure, in which very nearly perfect crystals form grains in random orientations joined together by fundamental atomic forces. How is it possible for crystals to be shaped and moulded like clay?

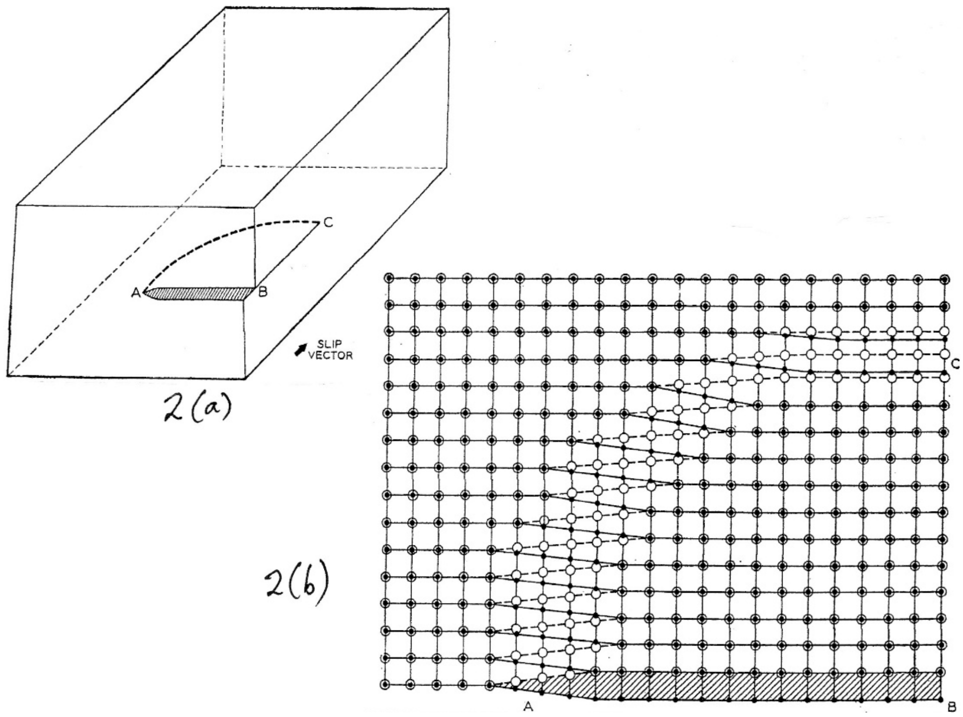

Figure 2. Diagram from *Dislocations in Crystals* by W. T. Read Jr., McGraw-Hill, N. Y., 1953.

The answer to this question is that the crystal planes can slip over one another by the passage of lines of disrupted symmetry called dislocations. Fig. 2 from an early textbook shows how. In fig. 2(a) a thumbnail area of a crystal plane, ABC, slips one atomic spacing over another to produce a ledge as shown cross-hatched. The area is bounded by a curved edge, AC, an arc of dislocation where the crystal structure is destroyed, as shown in Fig. 2(b). The arc can move forward by the local motion of the disrupted atoms, and in so doing spread the slipped plane but leaving behind the crystal, still perfect.

Already one can see the interplay between symmetry, perfect crystalline symmetry, and the line of dislocation, whose motion is required to change the crystal's shape. Dislocations play an essential role in the formation of the crystal, as well as in its utility.

The elastic response of the crystal to applied forces changes the spacings between its atoms, and changes too the shape of their pattern. But it is reversible: on unloading the pattern returns to its original form. The elastic response is independent of the source of the crystal, how it is grown, what previous forces it has experienced, and so on. It is very reproducible, independent of the 'history' of the crystal. But the plastic response is very different. The simplest example is possibly that of the metal paper clip: if you bend it once, it will deform and assume a different shape. But if you bend it back and forth, it will degrade and rather easily break in two. The response of the paper clip depends upon its 'history'. It shows 'fatigue'. When dislocations were first observed, it became clear that the structure of the arrays inside the crystal is very complicated and variable, history dependent, so the problem of understanding plastic behaviour becomes one of classifying the arrays and understanding their response to the applied forces.

The simplest such arrays, and the first to be well understood, are those introduced by backwards-and-forwards deformation, causing plastic fatigue. Fig. 2 shows a typical array. The crystal has been subjected to cycles of backwards and forwards shear deformation. After many such cycles, on the plane of shear, groups of dislocations in a ripple pattern appear, looking rather like the sand under waves near the shore of the sea. The symmetry is what you might expect: the pattern is overall perpendicular to the shearing direction, as if bits of crystal are scraped up and periodically deposited. The pattern of sand dunes swept by winds in the desert also comes to mind. But the pattern is not simple: its variability, the undulations along the ripples, are an essential symmetry-breaking feature. By their movement the undulations allow the ridges to move and to take up a well-defined average spacing.

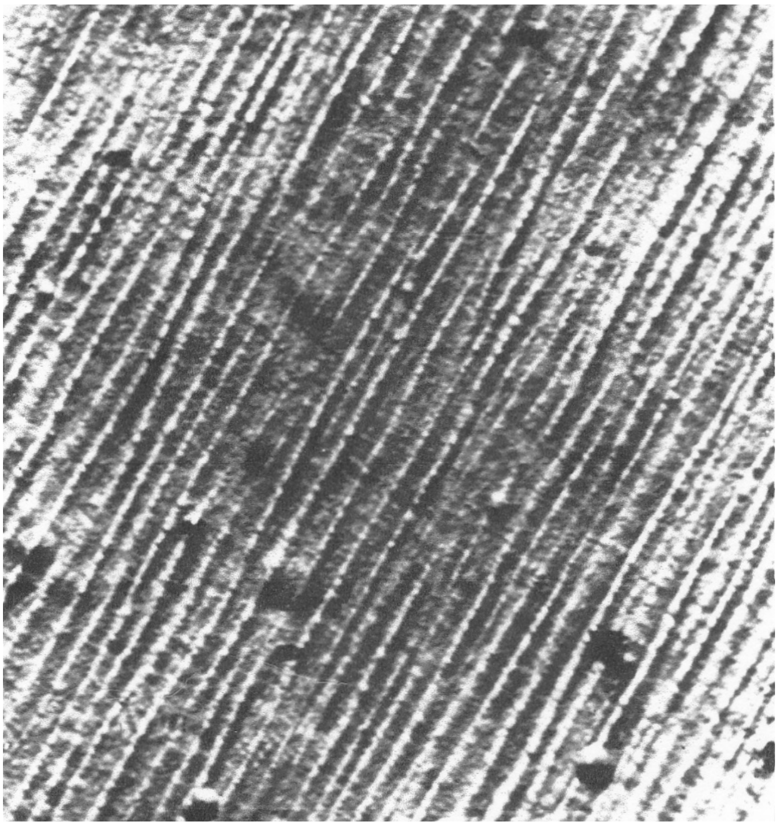

Figure 3. The structure of ‘ripples’ caused by backwards and forwards shear on the slip plane of magnesium. Each ripple is a bundle of dislocations, revealed by etching. The average spacing of the ripples is 8μm. The round pits are an artefact of the etching. Reproduced from ‘Studies of Fatigue Damage in Magnesium single Crystals’, Ph.D. thesis by R. Kwadjo, Cambridge, 1973.

The study of such patterns in ductile metals is of engineering significance: the endurance limit in fatigue, which is the magnitude of the backwards-and-forwards stress amplitude below which no cracking is developed, is just twice the stress required to bow a dislocation between the bundles of dislocations.

The patterns themselves suggest that structures of perfect symmetry cannot build up if they do not possess some irregularity giving them flexibility to allow change.

But what of the apparently more straightforward deformation that the roof of the emperor’s chariot underwent before it was assembled? When a metal is subjected to a steadily increasing load, the plastic strain continuously increases and is permanent: it doesn’t flow back when the load is removed. Is it possible to understand how this happens? The answer here is far more complicated and still subject to controversy among the many ‘plasticians’ in the world: engineers, materials scientists, physicists. Two facts are (mostly) agreed: firstly, on the mesoscale, that is, between atomic size and the size of the macroscopic object, plastic deformation is not homogeneous. It occurs in discrete slips, slip bands, or 'avalanches of slip', each lasting several microseconds. Secondly, attempts to see the structure of dislocations in these bands while they are forming have been frustrated. The transient bands are triggered suddenly and randomly. They have been compared to earthquakes: there seems to be no way of predicting where or when they will occur. One imagines that in the future, perhaps with the next generation of extremely bright sources of synchrotron radiation, we may be able to see them. What can be seen in the electron microscope is the residue of the bands, the most recent leaving their traces over the traces of others which have triggered before them. The apparently continuous curve of stress required to continue macroscopic plastic strain is an 'emergent property' of the system: it is the 'coarse-grained' (that is, 'smoothed') outcome of underlying, unrecorded, jerky events.

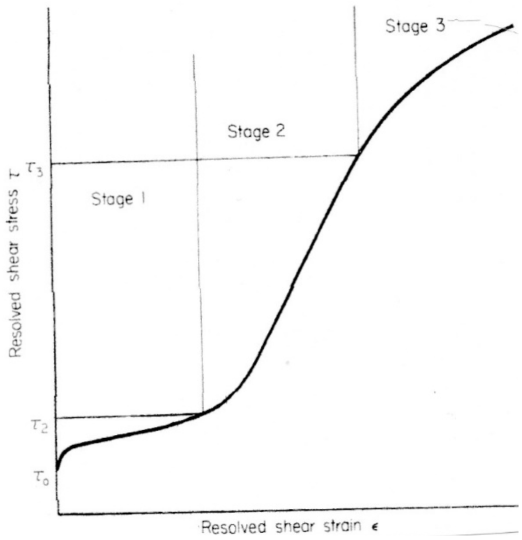

Figure 4. The typical 'stress/strain' curve of a ductile single crystal, reproduced from *The Plastic Deformation of Metals* by R. W. K. Honeycombe, Edward Arnold (Publishers) Ltd., London, 1968. The 'resolved shear stress' is the load per unit area of the shearing plane, and the 'resolved shear strain' is the plastic displacement per unit length measured perpendicular to the plane. The critical stresses for the transitions from one stage to the next are labelled: τ_0 for the onset of flow after the elastic region, τ_2 for the onset of stage 2, leading to failure in stage 3.

The puzzle is that, as has been known for many years, plastic flow of a ductile single crystal displays universal regularities. Fig. 3 shows a typical curve showing the increase of the shearing force required to continue plastic flow as a function of the plastic distortion it has already caused. After an initial very small elastic strain, not shown in the figure, one finds a 'precursor' interval, called Stage 1, where the response is variable, depending upon crystal orientation and its perfection. Then one finds a nearly linear region, Stage 2, which for each available slip system has a slope about equal to the elastic slope divided by 400. The slope is almost the same for all ductile crystals.

It does not depend upon the history of the crystal. It depends very little upon temperature or the rate of straining except at extremely high rates. The linear increase of force required to continue straining is the strengthening caused by the strain. Called 'work hardening', it is an essential characteristic of metal deformation. After Stage 2, one finds an exhaustion region, Stage 3, where the slope progressively decreases. The crystal fails before the curve flattens out.

What is the outline explanation for this universal behaviour? It must be rather like that for the universal gas law, the ideal gas, so beloved of first-year physics.

Figure 5. Each ellipsoidal slip band (smartie) is jammed against its neighbour and makes a variable angle with the plane of the paper (the shear plane)

The answer lies in the structure of the slip bands. If they are pictured as a densely packed array of ellipsoids, like smarties in a package, they present a universal but random structure. That they are ellipsoidal has a deep explanation. In an ellipsoid, all the dislocations in the band experience the same stress, enabling them to drive each other forward co-operatively. Each band almost instantaneously contributes its plastic strain. Once the pattern of interlocking ellipsoids is established in the crystal, more strain can be produced by generating new ellipsoids which overwrite the existing ones, yet not changing the random pattern of ellipsoids. The pattern only shrinks, to produce more and more bands, each new band getting progressively smaller. But as the pattern shrinks, the dislocations are forced to bend ever more sharply, thereby causing the characteristic work-hardening. At a high enough stress, the ellipsoids cannot be contained, they penetrate their boundaries, and the hardening progressively diminishes: the work-hardening saturates, and Stage 3 takes over.

There is a remarkable feature of this picture: the plastic offset produced so suddenly by each band is constant over the whole of Stage 2. The number of atomic spacings in each offset is approximately equal to the inverse of the angle (in radians) between the ellipsoid and the plane of shear. Its constancy is evidence of an underlying pattern, changing only in scale, as the deformation progresses.

So we have found a three-dimensional structure in which variability plays a crucial role, but which possesses the overall symmetry of shear. Each ellipsoid has degrees of freedom, a variable angle of tilt as well as variable size. Computer models show that the volume fraction occupied by the ellipsoids is very nearly 75%. As the straining proceeds, this stays constant. The structure is scale invariant, satisfies the 'renormalisation group', that is, it looks the same at any magnification. In the language of materials science, it follows 'similitude'.

In terms of this postulated structure, all characteristics of the work-hardening curve can be estimated accurately and consistently. It is very frustrating that the structure has not yet been seen directly. The perfect ellipsoids are too short-lived and their location impossible to predict. What is seen is an imperfect record of their existence, not unlike the fossil record left by evolution.

There is an underlying principle satisfied by all these structures, the crystal and its dislocations. They display 'maximum suppleness'. They are 'self-organised critical structures', in which there are a maximum number of elements which can participate in an avalanche. Each element is on the verge of departure from the standing pattern. Bifurcations are incessantly occurring: the ridges in the fatigue structure can split, or move sideways left or right; the ellipsoids can tilt clockwise or anticlockwise. These are the degrees of freedom which allow any new avalanche to overwrite the existing structure, refine it, but maintain its character. The overall degrees of freedom are maximised in number if they are distributed over all the space available, so no element gets 'frozen out'. The concept of maximum suppleness enables one to calculate emergent properties without detailed knowledge of the sequence of events leading to an avalanche, rather the way one can calculate the behaviour of an ideal gas without knowledge of individual molecular trajectories. Just as it's remarkable that all gases of whatever molecular identity behave approximately as an ideal gas, so it is that all ductile crystals behave in the same way. In stage 2 hardening they too obey a universal equation of state. 'Maximum suppleness' applies to the state of the straining crystal. It replaces the principle of 'maximum entropy' which applies to systems in thermal equilibrium. Suppleness in a complex mechanical system under stress does not depend upon temperature. It is the chance that somewhere in it an avalanche will occur. It is maximised when every element in the system is on the edge of participation in one.

Perhaps the aesthetic lesson is the Japanese notion of wabi-sabi: the beauty of austere symmetrical perfection be appreciated only if there is an element of chaotic imperfection. Imperfections are a product of the object's history, to be incorporated and admired. They are the structures which mediate between the real fragile world, subject to change, and an ideal world which cannot ever be realised.

Suppleness characterises the response of systems subject to change under the action of intensive forces. A good example is delta formation at the mouths of rivers opening out into the sea: as the silt carried from the source is deposited, it forms a barrier. Further flow is impeded and the river must push to find a way around the barrier. The river bifurcates, splits into two rivulets. The process repeats. A structure is built up which is narrow at the river and ever widening at the sea: a quasi-triangular delta structure of universal shape, supporting a pattern of successive bifurcations. Each river delta is unique, yet is a recognisable product of an irreversible process which comes to an end only when the source is exhausted. Maximum suppleness implies that at the spreading edge of the delta each rivulet is potentially on the verge of splitting again.

Wabi-sabi encourages us to appreciate the clash between the unique historical record of an object and its ideal form.

Prof Adrian Sutton FRS: Comments on ‘The Clash Between Chaos and Symmetry in an Ancient Process: The Working of Metals’ by L M Brown

It was a pleasure to read this paper. Professor Brown has pioneered the idea that the internal structure of a plastically deformed metal evolves into a self-organised critical state displaying ‘maximum suppleness’. It is not only a beautiful idea but in his hands it has provided a quantitative understanding of the universal features of stage 2 work-hardening.

For readers of this journal it may come as a surprise that there is so much going on inside a crystal of a pure metal when it is plastically deformed. The atomic structure of the crystal is largely unchanged. It is at larger length scales where new structures appear – the ellipsoids of sheared regions, well-illustrated in Fig.5 using smarties. This is the length scale of microstructure, which all metallurgists and materials scientists learn is fundamental to the mechanical properties of all crystalline materials. They also learn that linear defects called dislocations are the agents of plastic deformation: as they glide along planes in the crystal they shear the crystal irreversibly bringing about a permanent change of shape of the crystal. The theoretical challenge has been to understand the many-body problem of billions, even trillions, of dislocations in a cubic centimetre of crystal, where each dislocation is a flexible line that interacts through long-range elastic fields with other dislocations, and where their interaction at short range can result in the generation of new dislocations or their annihilation. In my view Professor Brown’s ideas have provided a framework to understand this extremely complex phenomenon, and to make sense of the universality of stage 2 work hardening.

Suggestions for further reading:

The theory underlying the ellipsoidal shape of slip bands is expounded in a recent book by Adrian P. Sutton: Physics of elasticity and crystal defects, Oxford Series on Materials Modelling, 2nd edition, Oxford University Press, 2024, DOI: 10.1093/oso/97801989081.001.0001.

The initiation of cracks in fatigue is briefly reviewed by Professor Brown: Philosophical Magazine, 2013, vol 93, pp 3809 – 3820, <http://dx.doi.org/10.1080/14786435.2013.798048>

Self-Organized Criticality is expounded in the book by Henrik Jeldtoft Jensen, Cambridge University Press 1998, ISBN 0-521-48371-9.