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EJECTION OF ATOMS FROM METALLIC SINGLE CRYSTALS

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Reprinted from American Journal of Physics,
Vol. 33, No. 12, 1064-1069, December 1965

Reprint Series RS 10

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Price subject to change without notice

ROGER DUHAMEL, F.R.S.C.
Queen's Printer and Controller of Stationery
Ottawa, Canada
1966

Reprinted from AMERICAN JOURNAL OF PHYSICS, Vol. 33, No.12, 1064–1069, December 1965
Printed in U. S. A.

Ejection of Atoms from Metallic Single Crystals

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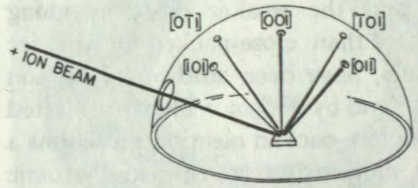
(Received 21 June 1965)

The unique characteristics of experimental data obtained from the directional ejection of atoms from metallic single crystals subjected to ion bombardment are discussed. The advantages, for instructional purposes, are outlined. Simple experiments are described that are designed to give the beginner in crystallography a working familiarity with (1) crystal directions, (2) techniques for orientation determinations, and (3) anisotropic characteristics of crystals. All experiments described may be performed with simple and inexpensive equipment.

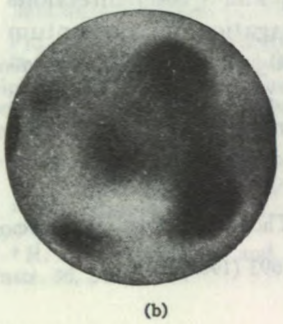
INTRODUCTION

THE purpose of this article is to outline several simple experiments, based on the phenomenon of the ejection of atoms from metallic single crystals during ion bombardment, which provide not only a familiarity with crystal structures but yield information on surface atom arrangements as well. The fundamental advantage of the method for instructional purposes, over x-ray and electron diffraction and optical methods, is that a limited number of crystallographic directions, rather than positions of reflections from atom planes, are experimentally recorded. This unique characteristic of the experimental data greatly simplifies interpretation for students being introduced to crystallography. An added advantage for the cubic system is that directions are also normals, or poles, of planes of the same indices, and thus it becomes possible to directly measure the angle between certain planes. Equipment suitable for the experiments to be described is very inexpensive and results are rapidly recorded without the use of photographic processes.

In 1955 Wehner[1] bombarded single crystals of silver, copper, tungsten, and alpha iron with mercury ions of low energy and reported the ejection of atoms along their close-packed directions, viz. [110] and [111] for face-centered-cubic (fcc) and body-centered-cubic (bcc) metals, respectively. He also found that ejection was stronger, all else being equal, in those close-packed directions pointing away from the ion source [Fig. 1(a)]. From these definitive experiments he concluded that directional ejection from bombarded surfaces depends on (a) penetration of high-speed particles into the specimen, (b) subsequent transfer of momentum throughout the structure by impact between atoms along close-packed rows, some of which necessarily intercept any surface, and (c) the restriction that the energy imparted outwardly to the surface atom be above a threshold value characteristic of the crystal. The directions of these close-packed rows are clearly recorded on collectors by a visible pattern of spots composed of atoms ejected from the crystal during bombardment. An example of such a pattern, on a hemispherical collector cut from a ping-pong ball, is shown in Fig. 1(b). The very limited number of ejection directions recorded results in simple patterns and contributes towards ease of interpretation. The literature indicates that



(a)



(b)

FIG. 1. (a) Schematic representation of an arrangement of specimen, collector and ion beam which illustrates by the widths of the lines that the relative intensity of equivalent ejection directions is a function of angle between ion beam and ejection directions. (b) An ejection pattern, obtained on a hemispherical collector, after bombarding a near (100) aluminum surface. Ejection predominates in the [110] directions pointing away from the ion source.

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[1] G. K. Wehner, J. Appl. Phys. 26, 1056 (1955).

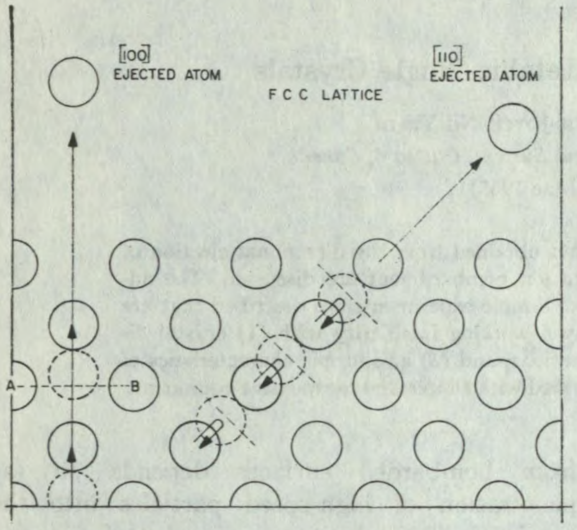

FIG. 2. A face-centered-cubic structure sectioned to reveal [110] and [100] directions. Propagation of momentum required for ejection along [110], involves momentum transfer only. However, for [100] ejection, propagation of momentum is accompanied by a transfer of atoms along the row by one atomic spacing.

good ejection patterns have been obtained under a wide variety of bombarding conditions, usually with inert gas ions, from single crystals of fcc aluminum, beta cobalt, copper, gold, lead, nickel, palladium, and rhodium; from bcc alpha iron, alpha chromium, beta brass and tungsten; and from hexagonal cadmium, magnesium, and zinc. No doubt this list of metals will grow.

Wehner's original suggestion that ejection along close-packed directions was due to momentum transfer alone has withstood the test of time. These, and the weaker ejections along directions other than close-packed in the fcc and bcc crystals, have been studied by Nelson and Thompson[2] and by Nelson.[3] They postulated that in the nonclose-packed ejection directions a mechanism of momentum transfer and atomic replacement sequence occurred. These two types of mechanisms are illustrated in Fig. 2 for ejection along [110] and [100] directions in fcc crystals. For propagation of momentum along the close-packed [110] directions in fcc crystals, no replacement of atoms occurs because impacts take place before the atoms have been displaced half their inter-atomic spacing in that direction and they then return to their original positions, as is indicated by the folded arrows. Silsbee[4] has suggested a mechanism by which a large proportion of the momentum available would be accurately focused along the close-packed rows after a series of collisions. Momentum transfer along [100] directions within the crystal is accompanied by a series of replacements because a displaced atom, having passed the center point before impact, cannot return to its original site, and moves into the vacated site of the atom it has just displaced. It should be noted that at the midpoint the displaced atom is surrounded by a symmetrical ring of four atoms, only two of which, A and B, can be shown in the diagram. This symmetrical ring helps to focus the displaced atom along the [100] direction. Similarly, symmetrical conditions are present in the case of [111] propagation in fcc crystals where the displaced atoms must pass through two three-atom rings to reach the next atom site. The postulated mechanisms are summarized in Table I.

TABLE I. Postulated mechanisms within the crystal for ejection from face-centered- and body-centered-cubic crystals.

Face-centered-cubic	Body-centered-cubic
[110] Momentum transfer only.	[111] and [100] Momentum transfer only.
[100] and [111] Momentum transfer and replacement sequence.	[110] Momentum transfer and replacement sequence.

Patterns from fcc metals, after short bombarding times, usually contain only the family of [110] direction ejection spots. In some instances longer bombarding times bring out an additional family of spots close to [116] directions. Anderson[5] has shown that an atom sitting in a twin position on a (111) plane could receive an impact from a [110] collision sequence which would eject it in a [114] direction, provided that the effective atomic diameter was equal to the nearest-neighbor distance [Fig. 3(a)]. Deviations from [114] result from smaller effective diameters associated with the velocities involved in the collisions and ejection close to [116] has been

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[2] R. S. Nelson and M. W. Thompson, Proc. Roy. Soc. (London) A **259**, 458 (1961).

[3] R. S. Nelson, Phil. Mag. **8**, 693 (1963).

[4] R. H. Silsbee, J. Appl. Phys. **28**, 1246 (1957).

[5] G. S. Anderson, J. Appl. Phys. **33**, 2017 (1962).

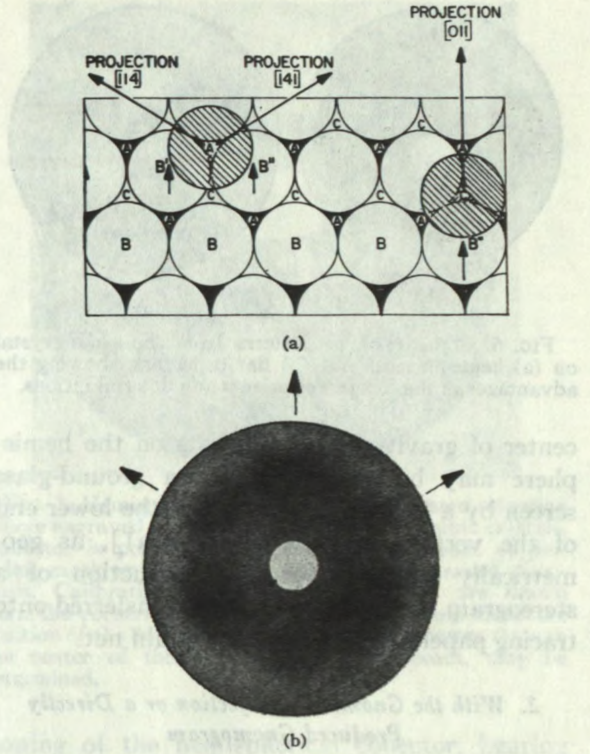

FIG. 3 (a) Arrangement of atoms in a (111) face-centered-cubic surface upon which rest shaded atoms in the two possible positions. Shaded atom, C, occupying a normal position will eject in the [011] direction after receiving impact from atom B°. Shaded atom, A, in nonnormal or twin position will eject in either the [141] or [114] direction depending on whether impact is received from atom B′ or B″. Normal stacking arrangement in fcc lattice is A, B, C, . . . C, A, B, C . . . . (b) Ejection pattern from a silver (111) surface, bombarded at normal incidence, showing three strong ejection spots of the set of [110] directions and three weaker spots due to atoms in nonnormal positions on the surface.

recorded from prepared (111) surfaces [Fig. 3(b)] and from other surfaces where (111) facets could develop as a result of ion bombardment.

In addition to [110] and [116], ejection may also occur weakly along [100] and [111]. The clearest patterns are obtained from fcc single crystals.

Patterns from bcc metals usually contain [111] and [100] spots. However, the former are sometimes severely weakened or even replaced by the occurrence of satellite spots, the positions of which seem to vary with different metals.[6] These extra spots have been variously attributed to last-impact deflections from the close-packed [111] directions by atoms in nonnormal positions on (110) surfaces,[7] or on (130) surfaces which have been observed to develop during bombardment of a (110) surface,[8] and to relaxation around vacancies in (111) surfaces.[3] In general, bcc patterns are inferior to fcc patterns. In the experience of the authors, tungsten single crystals give the best bcc patterns.

I. EXPERIMENTS

A. Identification of Ejection Directions

Since directions are recorded, it is possible, by using hemispherical collectors[9] and by bombarding crystal surfaces placed at the center of curvature of the collector, to obtain patterns from which the angle between pairs of directions may be directly determined. Using a circular protractor (Fig. 4), the angles may be measured to within 4° after the centers of the deposits have been marked with pinholes. Direction indices may be determined by referring to the approximate angles between pairs of directions tabulated in Table II. Spots separated by more than 120° are very rarely encountered, as intensities are greatly reduced in ejection directions at glancing angles to the surface because of the

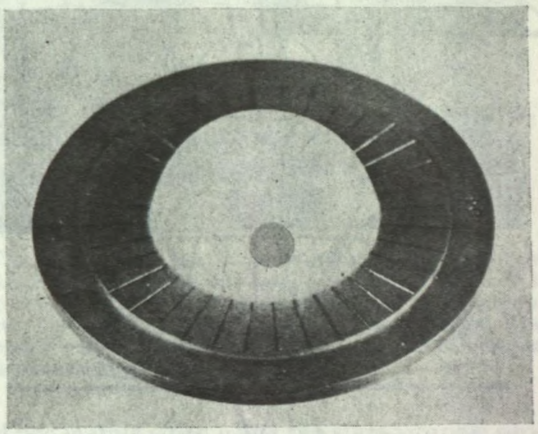

FIG. 4. Circular protractor with white hemispherical collector positioned for measurement of angle between two pinholes which mark positions of centers of ejection deposits on inner surface of collector. By reference to Table II indices of ejecting directions may be determined.

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[6] R. L. Cunningham and Joyce Ng-Yelim, J. Appl. Phys. 35, 2185 (1964).

[7] G. S. Anderson, J. Appl. Phys. 33, 659 (1962).

[8] J. E. Boggio and H. E. Farnsworth, Surface Sci. 1, 399 (1964).

[9] R. L. Cunningham and Joyce Ng-Yelim, Rev. Sci. Instr. 36, 54 (1965).

TABLE II. Directions of interest in patterns from the cubic system and approximate angles between pairs.

Face-centered-cubic pattern			Body-centered-cubic pattern		
Measured Angles ±1°		Direction Pairs	Measured Angles ±1°		Direction Pairs
36°	144°	[110]−[111]	36°	144°	[110]−[111]
		or [110]−[116]	45°	135°	[100]−[110]
45°	135°	[100]−[110]	55°	125°	[100]−[111]
60°	120°	[110]−[110]	60°	120°	[110]−[110]
70°	110°	[116]−[116]	70°	110°	[111]−[111]
77°	103°	[110]−[116]	90°		[100]−[100]
90°		[110]−[110]			or [110]−[111]
		or [110]−[111]			

longer momentum transfer paths within the crystal.

B. The Determination of Crystal Orientation

1. With the Stereographic Projection and a Wulff Net

In addition to making the angles between crystal directions measurable with a protractor, hemispherical collectors also make possible the direct production of stereographic projections for orientation determinations by a simple optical device.[6] Images of the pinholes used to mark the center of gravity of the deposits on the hemisphere may be projected onto a ground-glass screen by a light source placed at the lower end of the vertical diameter [Fig. 5(a)], as geometrically required for the production of a stereogram. The pattern may be transferred onto tracing paper and placed over a Wulff net.

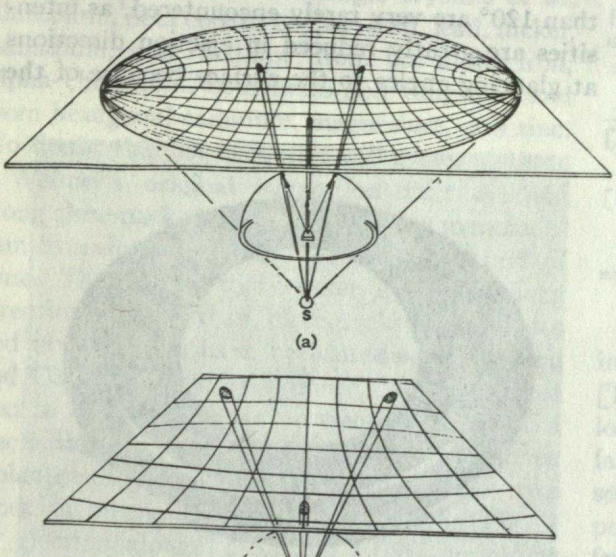

FIG. 5. (a) Schematic representation of the production of a stereographic projection directly from the collector by placing light source S, at bottom of diameter. (b) Schematic representation of the production of a gnomonic projection by raising light source to equatorial plane. Dashed lines in (b) indicate that the hemispherical collector may be removed and a gnomogram directly produced on a flat collector in the bombardment chamber.

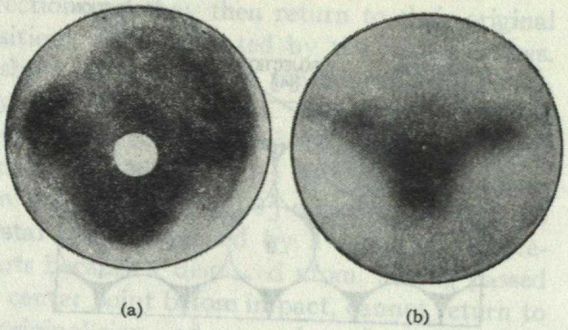

FIG. 6. Comparison of patterns from the same crystal on (a) hemispherical and (b) flat collectors, showing the advantages of the former for orientation determinations.

2. With the Gnomonic Projection or a Directly Produced Gnomogram

It is also possible to make a gnomonic projection by raising the light source to the center of curvature of the hemisphere, i.e., to the position previously occupied by the surface being bombarded, as in Fig. 5(b). In this case the projected beams and the original ejection directions coincide. An alternative but less desirable technique is to eliminate the hemispherical collectors and replace them by flat collectors for the direct production of gnomonic patterns in vacuum, as again indicated in Fig. 5(b). Flat collectors are not recommended, since ejection directions at large angles to the normal produce deposits which are spread out over large areas. In addition to requiring longer times to become visible, it is difficult to determine the center of gravity of these elliptical spots and the symmetry is often not evident. This can be seen in Fig. 6, which is a comparison of the patterns obtained on hemispherical and flat collectors from a crystal which was found to have its surface approximately parallel to the (013) plane.

3. With the Orienting Sphere

There is a more direct method[9] for crystal orientation determinations, which involves posi-

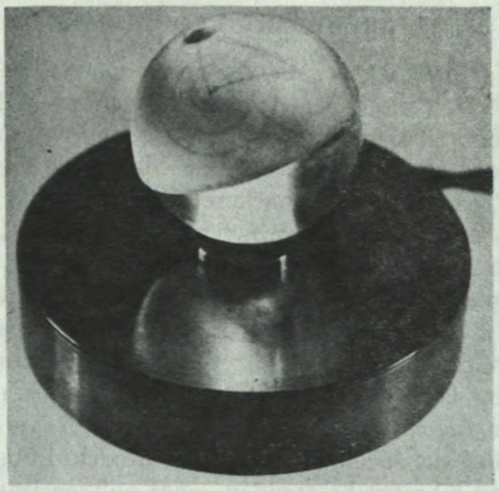

FIG. 7. Hemispherical collector on illuminated orienting sphere engraved with ejection directions for cubic crystals. Collector is positioned for best correspondence of pinholes, marking ejection directions, with engraved directions. Calibrating circles at 10° intervals are drawn from the corners of the unit triangle and from these the position of the normal to the surface, which passes through the center of the hole for the input beam, may be determined.

tioning of the hemispherical collector, bearing the ejection pattern, over a close fitting sphere illuminated from within and having engraved on it the directions of interest. The collector is shifted over the surface of the sphere until the best correspondence of its spots with the engraved directions is obtained. Calibrating circles, also engraved on the illuminated sphere, then permit a rapid determination of the orientation. An orientating sphere with a collector in place is shown in Fig. 7. A single engraved sphere may serve for orientation determinations of all cubic crystals; however, for hexagonal metals where the axial ratios vary from metal to metal, it would be theoretically necessary to engrave a sphere for each metal.

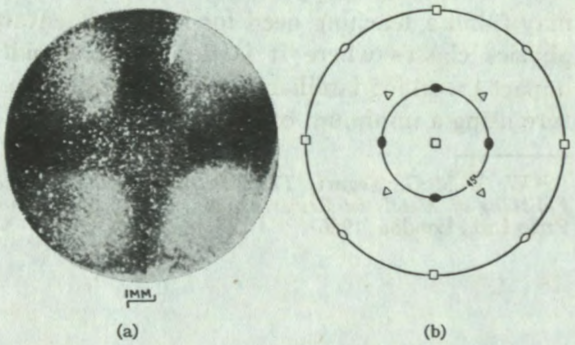

FIG. 8. (a) Optical micrograph of an ion bombarded aluminum (100) surface. More deeply etched areas appear light and are segments of an annular area swept out by the ion beam striking the surface of the rotating sample at 45°. (b) Stereographic projection showing the four [110] directions, indicated by the blackened ellipses, which became parallel to the ion beam during each rotation of the sample.

These orienting techniques have been recently published[6,9] wherein it was shown that orientation could be determined within 3° for many fcc and bcc metals when using the stereographic projections or the orienting sphere.

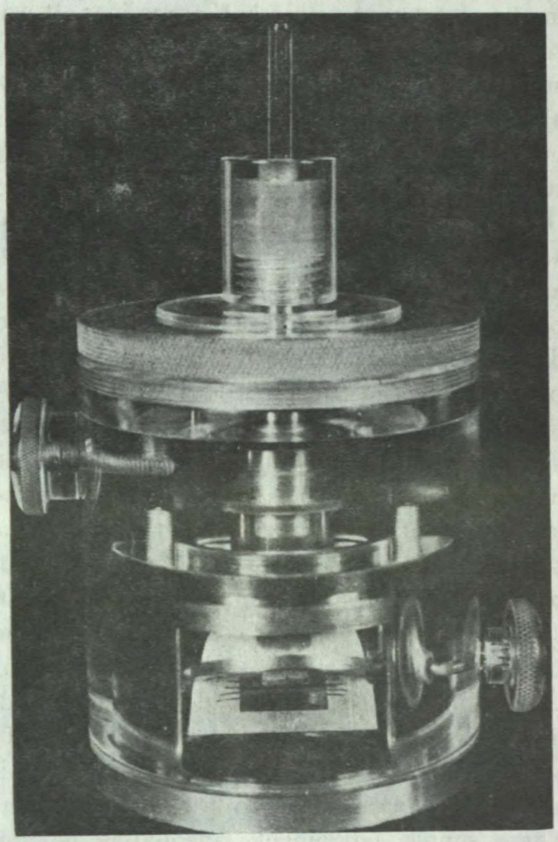

FIG. 9. A simple ion bombardment unit containing a hemispherical collector, cut from a ping-pong ball, and a single crystal specimen mounted centrally on its equatorial plane. The specimen is usually vertically bombarded. The assembly may be tilted so that inspection of the ejection pattern may be made at intervals.

C. The Production of Etch Patterns on Single Crystal Surfaces

The rate of atom ejection is strongly dependent upon which crystal direction is parallel to the ion beam. It has been found, by considering only the geometry of the first few atom planes, that the directions of low yield are also those at which the crystal presents a low cross-section to the incoming ions. Thus a greater average depth of penetration would be expected in those directions. An example of this effect, as measured by etching rates,[10] is given in Fig. 8(a), which shows the (100) surface of an originally brightly electropolished aluminum single crystal etched by ion bombardment with 8 keV argon ions. It was rotated while being bombarded at 45° to its surface. The beam, which had a divergence of about 2°, was directed to a point on the surface not on the axis of rotation. Hence, during one revolution an annular area was bombarded by the beam and all crystal directions inclined at 45° to the surface were brought parallel to it, as indicated in Fig. 8(b). Whenever the beam became directed close to one of the four [110] directions cutting the surface at 45°, indicated by the blackened ellipses in Fig. 8(b), relatively little etching occurred, because of the deeper average penetration of the primary beam and hence the relatively large loss of energy along the long momentum transfer paths back to the surface. Hence, these parts of the surface did not become matte during the limited bombardment. The same point may be conveniently demonstrated, without a rotating sample, by comparing the visible etching effect of identical bombardments along a [110] direction at 45° to a (100) surface and along a [223] direction, which is also approximately 45° to the (100) surface. These simple experiments adequately demonstrate an anisotropic characteristic of crystals which is readily interpretable.

II. EXPERIMENTAL REQUIREMENTS

For these experiments simplest ion sources of the glow-discharge-type are adequate. The authors have recently described[9] a simple apparatus (Fig. 9) consisting of an ion source with an integral bombardment chamber which is very inexpensive and which can give orientations with an accuracy of 3° or better. It should be coupled to a vacuum system capable of maintaining a pressure of 10^{-4} Torr or better, since at these pressures the mean free path of the ejected atoms is considerably greater than the distance from the specimen to the collector. Contamination of the specimen surface with back-diffusing pump oil must be avoided, since polymerization at the bombarded surface readily occurs. Thus, in many vacuum systems a cold trap is required.

Single crystals may be purchased from a number of scientific organizations or they may be grown in the laboratory. Single crystal ingots, from which crystals of desired orientation may be cut, may be grown in vacuum by slowly withdrawing from a furnace a molten charge contained in a long boat. Lead, having a very low melting point, is perhaps the easiest to prepare, is readily available, and gives a good pattern.

Since directional ejection depends on the orderly arrangement of atoms up to the surface, it is necessary that all cold-worked material be removed from cut surfaces. This may be accomplished by electrolytic or chemical polishing.[11]

It is suggested that the experiments described and the simplified apparatus referred to above may fulfill a teaching need for large elementary physics classes where it is desired to rapidly impart a working familiarity with a crystal structure using a minimum of equipment.

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[10] R. L. Cunningham, K. V. Gow, and Joyce Ng-Yelim, J. Appl. Phys. **34**, 984 (1963).

[11] W. J. McG. Tegart, *The Electrolytic and Chemical Polishing of Metals in Research and Industry* (Pergamon Press Ltd., London, 1956).