PHYSICAL REVIEW LETTERS

Volume 19

4 SEPTEMBER 1967

NUMBER 10

REVERSILIBILITY OF FAST-CHARGED-PARTICLE TRAJECTORIES IN SINGLE CRYSTALS

E. Bøgh* and J. L. Whitton

Chalk River Nuclear Laboratories, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada (Received 10 July 1967)

Experiments to study the motion of fast charged particles (e.g., 1-MeV protons) in single crystals have hitherto been divided into two types. usually termed "channeling" and "blocking." A channeling experiment involves a measurement of the influence of a crystal lattice on the probability $P(\psi_{ch})$ of hitting a crystal atomic nucleus by the particles of a well-collimated beam. ψ_{ch} is the angle between the beam direction and a crystal axis (or plane). A blocking experiment involves a measurement of the influence of the lattice on the intensity observed outside the crystal $I(\psi_{bl})$ of particles emitted from crystal atomic nuclei. ψ_{bl} is the angle between emission direction and a crystal axis (or plane).

According to Lindhard,¹

$$P(\psi_{ch}) = I(\psi_{bl}) \tag{1}$$

for $\psi_{ch} = \psi_{bl}$ in cases where slowing down of the particles is small enough to be neglected. Equation (1) assumes same particle, particle energy, crystal, and crystal axis in the channeling and the blocking experiments.

Measurements of the Rutherford scattering yield in single crystals offer the possibility of studying channeling and blocking in one experiment.² The present Letter describes how such an experiment has been used to demonstrate Lindhard's "rule of reversibility" [Eq. (1)].

To make clear the possibility of using Rutherford scattering in a demonstration of the valid-

ity of Eq. (1) we point out that in connection with channeling, the term "hitting an atomic nucleus" means coming much closer to an atomic nucleus than the Thomas-Fermi screening distance $a \sim 0.1-0.2$ Å.¹ Similarly, in connection with blocking, the term "emitted from an atomic nucleus" means emitted from a region much closer to an atomic nucleus than $\sim a$. Rutherford scattering takes place at distances ~ $10^{-\$}$ - 10^{-4} Å of an atomic nucleus. Thus $P(\psi_{ch})$ may be identified as the yield $Y(\psi)$ of a process in which a well-collimated particle beam is incident upon a single crystal at an angle ψ with a crystal axis, and the backscattered particles are detected within a small solid angle $d\Omega$ along a direction specified by the polar angles θ and φ with respect to the incident (Z-axis) direction (see Fig. 1). $I(\psi_{\rm bl})$ then is the yield of the inverse process in which the collimated beam is incident along the direction (θ, φ) and the scattered particles are leaving the crystal along the negative Z axis at the angle ψ with the crystal axis.

So far, the validity of Eq. (1) has not been fully established experimentally. In earlier channeling experiments^{2,3} for $\psi = 0$, the scattering yield of protons has been observed to be ~1% of "normal yield" (i.e., the yield from an amorphous target), but in the corresponding blocking experiments the observed intensity reductions have been no more than a factor of ~10.^{4,5} Recent work⁶ has shown that such experiments depend strongly on the degree of



FIG. 1. Principle of the reversibility experiment.

perfection of the crystal, in particular on surface disorder (e.g., amorphous oxide), and the lack of reversibility in the earlier data may very likely be due to differences in the crystals used in different laboratories.

The experiment to be described here was performed with a 1-MeV proton beam in the Van de Graaff accelerator at Chalk River. Canada. A well-annealed and electropolished tungsten crystal was used, and the yield of protons scattered through $\sim 135^{\circ}$ was measured. The scattered protons were energy analyzed to define the width and the depth beneath the crystal surface of the scattering zone.⁷ In the channeling part of the experiment, the yield was measured as a function of the angle ψ between the proton beam and the $\langle 100 \rangle$ axis, by tilting the crystal with respect to the beam. The scattered protons were detected by a solid-state detector which viewed the crystal along a direction (θ, φ) that did not coincide with any prominent crystal axis or plane. In the blocking part, the incoming and the out-going directions were exchanged, viz., the beam was incident along the direction (θ, φ) , and the yield was measured as a function of the angle ψ between the direction of the outgoing protons and the $\langle 100 \rangle$ axis. This was accomplished by moving the detector. The angular resolution in the determination of ψ was, in the channeling case, determined by the beam divergence $(<0.1^{\circ})$ and in the blocking case, by the acceptance angle of the detector $(=0.1^{\circ})$.



FIG. 2. Yield of protons scattered 135° in W. Primary proton energy=1 MeV. Thickness of scattering zone ~1000 Å. Depth beneath surface of scattering zone ~3000 Å. Solid circles denote channeling results; open circles, blocking results.

Figure 2 shows the measured yield of scattering events taking place in a ~ 1000 -Å-thick zone 3000 ± 500 Å below the crystal surface.

The agreement between the experimental results and Eq. (1) is well within the total experimental uncertainty. The deviation of $I(\psi_{bl})$ from the normal yield is slightly less than the deviation of $P(\psi_{ch})$ for ψ values of 2-4°. However, the result of this type of measurement depends on the experimental definition of the scattering zone depth; when one takes into account the reproducibility of this depth on changing from the geometry of the channeling part to the geometry of the blocking part, the small discrepancy with Eq. (1) is not considered significant.

It is beyond the scope of this Letter to discuss the rule of reversibility in detail. In a statistical mechanical treatment of channeling and blocking it can be regarded as a special case of the general reciprocity law for direct and inverse processes which is based on Liouville's theorem, i.e., on the conservation of volume in phase space. In optics the analogous reciprocity law is well known; it states that the irradiance at a point A from a source at B is the same as the irradiance at B when the same source is at A.

A more detailed report of experimental studies of the conditions under which Eq. (1) does or does not apply will be published later.

*Permanent address: University of Aarhus, Aarhus, Denmark.

¹J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 34, No. 14 (1965).

²E. Bøgh and E. Uggerhøj, Phys. Letters <u>17</u>, 116 (1965); Nucl. Instr. Methods <u>38</u>, 216 (1965).

³E. Bøgh, Phys. Rev. Letters (to be published).

⁴D. T. Gemmell and R. E. Holland, Phys. Rev. Letters 14, 945 (1965).

⁵A. F. Tulinov, V. T. Kulinkauskas, and M. M. Malov, Phys. Letters <u>18</u>, 304 (1965).

⁶E. Bøgh, in <u>Proceedings of the Cairo Solid State</u> <u>Conference at the American University at Cairo, Sep-</u> tember, 1966 (Plenum Press, Inc., New York, 1967); E. Bøgh and J. L. Whitton, to be published.

⁷A. B. Brown, C. W. Snyder, W. A. Fowler, and C. C. Lauritsen, Phys. Rev. 82, 159 (1951), Appendix B.

CRITICAL OPALESCENCE OF LIQUID SODIUM-LITHIUM MIXTURES

H. Brumberger, N. G. Alexandropoulos, and W. Claffey Department of Chemistry, Syracuse University, Syracuse, New York (Received 31 July 1967)

The critical opalescence of binary liquid mixtures near the critical demixing temperature T_c has been investigated by means of light or x-ray scattering for various systems (see Brumberger¹ for a review of the experimental literature). The observed increase in scattered intensity as T approaches T_c from the one-phase region is due to the increasing ease with which concentration fluctuations (and consequently fluctuations in electron density or refractive index) occur, and to the rapidly increasing longrange correlation between these fluctuations. Various theoretical treatments have been given.²⁻⁵ Our purpose in this Letter is to report the experimental detection of what we believe to be critical opalescence in a type of system where this phenomenon has not previously been observed, namely a binary liquid-metal system, Na-Li.

The small-angle x-ray scattering of the sodium-lithium system in the critical region, at a composition of 58 with respect to Na, was investigated using a Kratky camera,⁶ MoK α radiation monochromatized by filter and pulseheight discrimination, and a monitor detector to eliminate intensity variations due to any source instabilities. The liquid metals were confined in a high-temperature sample cell described in detail elsewhere.⁷ The cell was filled directly from a mixing crucible where samples were prepared, melted, homogenized, and freed of any undissolved impurities in situ under highpurity argon. Sample thickness in the x-ray path was 3.62 mm. Widely different values of T_c have been reported^{8,9}; for this reason, a series of angular scans were made over a substantial temperature range, 430 to 280°C.

The temperature of the sample was initially allowed to decrease at the rate of 1°C per 30 min; when the desired temperature was reached, the sample was allowed to equilibrate for 30 min. during which interval the intensity at a fixed angle was read every 2 min. In the subsequent interval, a complete angular intensity scan was made, the sample temperature was then lowered to the next desired value, and the procedure was repeated. Temperatures were measured by means of a Wenner potentiometer and iron-iron/Constantan thermocouples calibrated against National Bureau of Standards certified, mercury-in-glass thermometers of the appropriate range. The temperature of the sample could be controlled to ±0.05°C or better, and was uniform over the irradiated region to this precision.

Figure 1 shows a typical plot of scattered intensity (counts min⁻¹) at a fixed scattering angle θ of 10⁻³ rad (an equivalent Bragg spacing of about 700 Å) versus temperature. A substantial increase in intensity in a range of about 10°C above 301°C and a sudden sharp drop below this temperature are observed. From this graph one would estimate that T_C is ~301°C; this is somewhat below the value of 303°C shown for this composition by Kanda, Faxon, and Keller,⁹ but in good agreement in view of the experimental uncertainties and slight differences in sample purity, composition, and technique.

The general features of the effect are reproducible; some variations in T_c , intensity distribution, and temperature range of the effect are observed from sample to sample. These are primarily due to slight variations in purity, sample thickness, and composition. The