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X-Ray Studies of Crystals Vibrating Piezoelectrically

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The original investigation has been extended by using both Laue patterns and regular reflection from the crystal surfaces. Much of the previous work was repeated and confirmed. Quartz was found to be more satisfactory for this work than Rochelle salt. A large increase of intensity in the Laue pattern was found for the fundamental of the low frequency mode of vibration with a Y-cut quartz crystal. The large increases found in previous work were due to the high frequency or "thickness" modes of vibration. The modes of vibration determined by the longer dimensions of the crystals gave an intensity increase for the case of regular reflection. It was shown that the negligible increase for the modes of vibration determined by the thickness of the crystals was not caused by an imperfect surface layer. In a complete survey made of a long X-cut crystal, a definite distinction was found between the regions of nodes and antinodes of motion. By direct polishing experiments, it was found that the intensity increases for the cases of regular reflection were due mainly to a reduction of secondary extinction. It is pointed out that in all cases these x-ray methods show a difference in the effect between the X-cut and Y-cut crystals for similar modes of vibration.

INTRODUCTION

It was first shown by Fox and Carr[1] that a marked increase of the intensity in the Laue pattern of thin quartz crystals occurred when the crystals were vibrating piezoelectrically along the direction of the thickness of the plates. Similar results for thick crystals have been reported by other investigators.[2–6] They observed not only an increase of intensity in the Laue pattern, but also a filling in of the space between the double spots.

Fox and Cork[7] studied this effect further by using the regular x-ray reflection from the major surfaces of thin Y-cut[8] crystals that were vibrating along the direction of their Y axis. They found that this mode of vibration gave no increase of intensity in the reflected line. Colby and Harris[9] in a similar experiment examined the effect upon a line reflected from the middle of a long X-cut quartz bar that was vibrating at the fundamental and second harmonic of its low frequency mode in the direction of its Y axis. They found that the vibration produced a definite increase of intensity for the fundamental but little or no increase for the second harmonic. The middle of their crystal represented a node of motion for the fundamental and an antinode for the second harmonic.

In the case of Laue patterns obtained from quartz crystals, when vibrating at their low frequency modes, several observers[3, 5, 10] have reported little or no intensity increase for the vibration along the Y axis of X-cut crystals. Similar effects have also been reported for the vibration along the Z axis of X-cut crystals.[5]

Some of the work in this investigation has been similar to that of the other observers. The results of these experiments are in agreement with those which have been previously reported. For the vibration determined by the thickness of the crystals, however, more emphasis should be given to the difference in the effect upon the Laue pattern between the thick X-cut and Y-cut crystals. The double spots in the pattern obtained with the thick X-cut crystal were bridged over by the vibration, while this effect was slight for the pattern obtained with the Y-cut crystal. It seems that the vibration made it possible for the interior planes of the X-cut crystal to contribute to the intensity of the spot.

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* Now at Western College, Oxford, Ohio.

[1] G. W. Fox and P. H. Carr, Phys. Rev. **37**, 1622 (1931).

[2] S. Nishikawa, Y. Sakisaka and I. Sumoto, Phys. Rev. **38**, 1078 (1931).

[3] C. S. Barrett and C. E. Howe, Phys. Rev. **39**, 889 (1932).

[4] J. M. Cork, Phys. Rev. **42**, 749 (1932).

[5] S. Nishikawa, Y. Sakisaka and I. Sumoto, Sci. Pap. Inst. Phys. and Chem. Res. **25**, 20 (1934); Phys. Rev. **43**, 363 (1933).

[6] G. W. Fox and W. A. Fraser, Phys. Rev. **47**, 899 (1935).

[7] G. W. Fox and J. M. Cork, Phys. Rev. **38**, 1420 (1931).

[8] The terminology used here is in accordance with that suggested by W. G. Cady, Proc. I. R. E. **18**, 2136 (1930). Diagrams of these cuts are also shown in reference 1, in which the X and Y-cuts are referred to as the Curie and 30-degree cuts, respectively.

[9] M. Y. Colby and S. Harris, Phys. Rev. **46**, 445 (1934).

[10] M. Y. Colby and S. Harris, Phys. Rev. **43**, 562 (1933).

On the other hand, the large increase of intensity found for the Y-cut crystal seemed to come from an increased efficiency of the planes which had been reflecting in the nonvibrating case, instead of contributions from additional planes.

The work which is to be reported here represents an extension of this investigation. This intensity effect upon the Laue pattern and a reflected line was studied by using additional modes of vibration. Also, the dependence of the intensity effect upon different parts of the crystals and upon different surface preparations of the crystals was investigated. This additional information gives a better understanding of the relation between this intensity effect and x-ray extinction and the influence upon these effects by certain modes of piezoelectric vibration.

EXPERIMENTAL

Both quartz and Rochelle salt crystals were used in this series of experiments. The quartz crystals were of the conventional X- and Y-cuts and were of various dimensions. Table I will give a brief description of the principal crystals and their fundamental frequencies, which were used in this investigation.

TABLE I. *Description of crystals.*

CRYSTAL PLATE	CUT	x (mm)	y (mm)	z (mm)	DIRECTION OF ELECTRIC FIELD	nX (kc)	nY (kc)	nZ† (kc)
A	Y	19.0	6.0	19.7	Y	138.2	371.3	——
B	X	4.2	25.5	27.5	X	638.3	92.3	124.0
C	X	4.0	43.0	16.0	X	——	63.2	——
*D**	X	10.0	100.0	20.0	X	——	26.3	——

* This crystal was kindly loaned to the writer by Professor N. H. Williams.

† The terminology suggested by Cady, reference 8, is also used here. The expression nX means, simply, the fundamental frequency of the wave propagated approximately in the direction of the X axis, etc.

The crystals were maintained in vibration at one of their resonance frequencies by an adaptation of the circuit described by Wright and Stuart.[11] The plate voltages were varied from 220 to 500 volts.

The Laue pictures were taken by using the radiation from the tungsten target of a standard 200-kv Coolidge tube, operating at 100 kv and 2 milliamperes. The exposure times were varied from 7 to 15 hours, depending upon the thickness of the crystal. Two cameras were arranged to receive the radiation from the tube, so that it was possible to conduct two experiments simultaneously. One of these cameras has been described elsewhere.[4] The other one was similar, except that it was possible to take the pictures at a crystal-to-plate distance of either 5 cm or 10 cm.

The reflection pictures were taken from an x-ray tube equipped with a tungsten target, with the $L\alpha_1$ and the $L\beta_1$ lines in the first and second orders. The Bragg spectrometer was the same as used by Fox and Cork.[7] Several comparisons between the lines taken with the crystal vibrating and at rest were made on the same plate by moving the plateholder. In order to facilitate the measurement of the intensity change, a line was usually taken at double the exposure time of the comparison lines with the crystal at rest. The exposure times were relatively short, being only 5, 10, and 20 minutes for each line on the plate. These lines were reflected from the planes parallel to the major surfaces of the crystals with but one exception, when the planes parallel to the end of a crystal were used. These planes are the $(2\bar{1}\bar{1}0)$ planes for the X-cut crystals, the $(10\bar{1}0)$ planes for the Y-cut crystals, and also the $(10\bar{1}0)$ planes for the planes parallel to the end of the large X-cut crystal.

DISCUSSION OF RESULTS

A study was made to determine the effect upon the Laue pattern obtained from a Y-cut quartz crystal when vibrating at the fundamental of its low frequency mode of vibration along its X axis. This mode of vibration gave an intensity increase comparable to that found for the vibration determined by the thickness of the crystal. This was the only mode of vibration, besides that determined by the crystal thickness, which gave such a pronounced increase of intensity in its Laue pattern. Also, as will be seen later, it is the only case in which a marked effect was found both for the Laue pattern and a reflected line.

The fundamentals of the low frequency modes of vibration gave a marked increase of intensity for the case of regular reflection with all of the crystals used. Two such frequencies were available with the X-cut crystal, B, one of which was presumably determined by its dimension along the Y axis and the other by its dimension along

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[11] R. B. Wright and D. M. Stuart, Bur. Standards J. Research **7**, 519 (RP356) (1931).

the Z axis. Typical microphotograms of this effect for the low frequency fundamental determined by the X axis with the Y-cut crystal and the Z axis with the X-cut crystal are shown in Fig. 1 A and B. This definite increase of intensity was unchanged by turning the crystals through 90 degrees for each of the three low frequency fundamentals used with the two crystals. It was also present at various positions on the crystal surfaces. This demonstrated that a "disturbed" area occupied most of the face of the crystal. This observation is in qualitative agreement with the many experiments of Osterberg.[12]

The removal of this intensity effect, due to the piezoelectric vibration, by polishing is illustrated in Fig. 1C. Not only was this effect removed; but it was again made to reappear by appropriately etching the surface. This process of destroying and again creating the intensity effect was done a number of times. It has been well established that polishing removes the extinction effects at the surface of a crystal. Since this polishing experiment was more correctly a fine grinding rather than a high polish, it is evident that the effect found in a reflected line is due to the reduction of the secondary extinction by the vibration. These observations are in accordance with the general agreement that this increase of intensity during vibration is due to a reduction of extinction effects within the crystal. It may be that a reduction of both the primary and secondary extinction occurs in the case of the Laue patterns.

The microphotometer curve in Fig. 1D shows the absence of an intensity effect in the reflected line from the face of the Y-cut crystal while vibrating at its high frequency fundamental in the direction of its thickness. The curve in Fig. 1E shows a small effect for the similar mode of vibration with the X-cut crystal; but it is not comparable to that shown for the low frequency fundamentals, as can be seen by again referring to Fig. 1B.

It has been suggested[3] that the presence of an imperfect surface layer was responsible for the null results of Fox and Cork. This hypothesis has been refuted by this experiment, in which both large and negligible results were obtained from the same crystal by changing the mode of vibration. Both of the crystals, B and C, show a definite effect for the vibrations determined by their longer dimensions, while they show little effect when the motion is determined by the thickness dimension. This is especially convincing, as it was possible to take such a set of

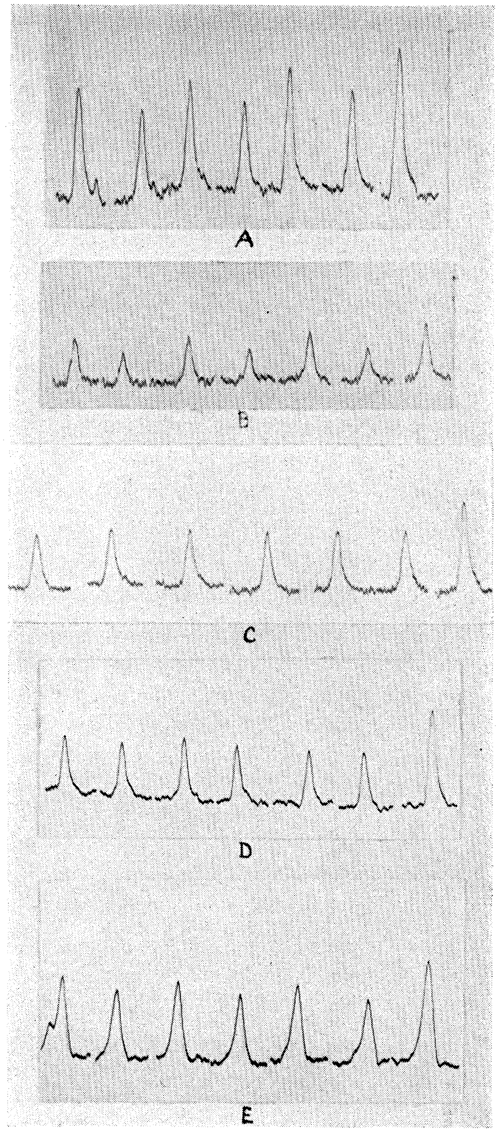

FIG. 1. Typical microphotograms of reflection pictures taken from rectangular quartz plates. Exposures: 10 min., 1st, 3rd, and 5th lines, vibrating; 2nd, 4th, and 6th lines, at rest; 20 min., 7th line, at rest. (A) X vibration of Y-cut crystal. (B) Z vibration of X-cut crystal. (C) Same as (B), except that the crystal surface was polished. (D) Y vibration of Y-cut crystal. (E) X vibration of X-cut crystal.

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[12] H. Osterberg, Phys. Rev. **43**, 819 (1933); Rev. Sci. Inst. **5**, 183 (1933).

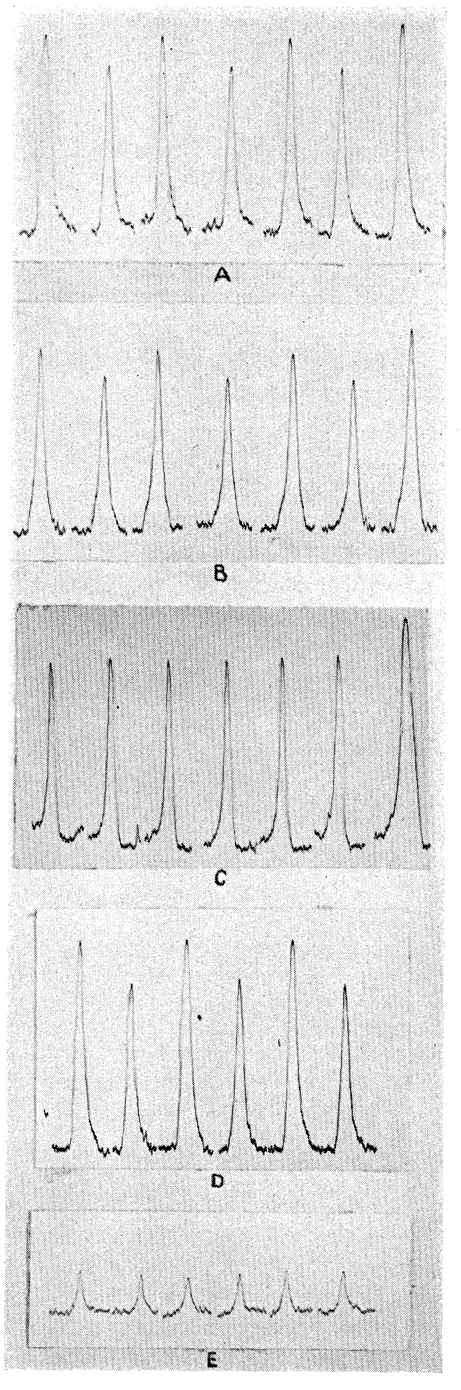

FIG. 2. Typical microphotograms of reflection pictures with the Y vibration of the 10 cm X-cut quartz bar. Exposures: 5 min., 1st, 3rd, and 5th lines, vibrating; 2nd, 4th, and 6th lines, at rest. 10 min., 7th line, at rest. (A) Middle position of bar, plate voltage 320. (B) 3 cm from middle of bar, plate voltage 320. (C) 4.5 cm from middle of bar, plate voltage 320. (D) Same as (B), except plate voltage 500. (E) An end of the bar, plate voltage 500.

pictures by making minor adjustments in the oscillatory circuit, without disturbing the crystal.

The negligible effect for the "thickness" mode of vibration in the reflection pictures is explained by the absence of change in lattice constant at the surface.[7, 9] The motion must also be of such a nature that no breaking up of the "mosaic blocks" occurs in a manner as to produce a change in the secondary extinction. The small effect in the Laue patterns for the low frequency fundamentals of the X-cut crystals may be due to the fact that a similar configuration exists within the region through which the x-ray beam is passing. Thus, there would be no changes in the crystal lattice or "mosaic blocks" which in turn would give a change in the extinction effects.

A complete survey of one face of the quartz bar, D, was made while it was vibrating at the fundamental of its low frequency mode of vibration in the direction of its length. By using a narrow electrode with a slit in its center for maintaining the vibration, nearly the entire face of the crystal was available for reflection. Comparison pictures using the second order $L\beta_1$ line of tungsten were taken at intervals of 5 mm along the Y axis. The entire crystal holder was moved for obtaining the different positions, so that the crystal was not changed relative to its vibrating system.

Assuming that the crystal motion can be represented as a standing wave with a node of motion at the center and antinodes at the ends, the maximum effect would be expected to occur at the node and a zero effect at the antinodes. Although this was found to be true, there was not a sharp nodal point. Instead, there was a nodal region and an antinodal region in which the effect was a maximum and a minimum. The nodal region extended over a space of about 6 cm in which the effect was a maximum. This nodal region was not quite symmetrical with the center of the crystal. There was only a space of about one cm in length at each end in which the effect could be said to be absent. These regions were studied further by using on the crystal a larger electrode which had only three windows, one at the center, two at a distance of 3 cm from the center, and a space at each of the ends. The microphotometer curves in Fig. 2 A, B, C, and D show a repetition of the former results.

A similar series of reflection pictures was taken with the same crystal by using the third harmonic of the same mode of vibration. For the third harmonic there was even less distinction between the expected nodal and antinodal regions than existed for the fundamental. Indeed, a "disturbed" area seemed to occupy the major portion of the reflecting surface of the crystal. On the basis of a large number of pictures, the effect was greater for the nodal than for the antinodal regions.

The microphotogram in Fig. 2E shows that no increase of intensity was present in the reflection pictures taken from the end of this long crystal, while it was vibrating at the fundamental of its low frequency mode of vibration. This result was expected since for this mode of vibration the planes parallel to the end of the crystal are in a configuration similar to that of the planes parallel to the face of the crystal for the mode of vibration determined by the thickness of the crystal.

An extensive study was made of a number of Rochelle salt crystals. The increases of intensity were small and could not be consistently repeated. This may be attributed to the difficulty of maintaining the equivalent of an etched surface for the reflection pictures; and for the Laue pictures, to the fact that most of the modes of vibration used were similar to those of quartz which showed little effect in their Laue pattern. Fox and Fraser[6] report an effect for Rochelle salt, but much smaller than for quartz.

The writer wishes to acknowledge his appreciation to Professor J. M. Cork of this department for suggesting the problem and for aid in interpreting the results. He is especially indebted to Professor N. H. Williams also of this department for the loans of equipment from his laboratory, and for valuable criticism and encouragement during this investigation.

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Excitation Potential of $K\alpha_{3,4}$ Satellite Lines

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With a two-crystal vacuum spectrometer the curve of the intensity of $K\alpha$ satellite lines *vs.* voltage of the x-ray tube has been determined for the $K\alpha_{3,4}$ lines of titanium. For voltages greater than 11 kv the satellite intensity (total area of the $K\alpha_{3,4}$ satellite structure) relative to the intensity of the Ti $K\alpha_1$ lines is 2.21 percent; the ratio of peak intensities α_4/α_1 is 0.69 percent. The measured excitation potential of the Ti $K\alpha_{3,4}$ lines is 5450±100 volts, 500 volts in excess of the excitation potential of the $K\alpha_{1,2}$ lines. Assuming 0.85 as the screening constant of a missing K electron on an L electron, one calculates that the voltage required to produce a state of KL_{III} ionization in the titanium atom is 5455 volts. This value is in excellent agreement with the measured excitation potential and the conclusions of the experiments are in support of the Wentzel-Druyvesteyn theory of the origin of the $K\alpha_{3,4}$ satellite lines.

I. INTRODUCTION

THE phenomena associated with nondiagram or satellite lines represent an enigma of many years standing in x-ray spectroscopic theory. For some time we have been able to account satisfactorily for the essential features of the production of diagram lines as either dipole or quadrupole radiations, but, perhaps because of a paucity of accurate, unambiguous experimental information, we are confronted with various contradictory theories which attempt to "explain" satellite lines. The voltage required for the production of satellites is of crucial importance in testing the validity of these various theories and in the present paper we shall be concerned with a determination of this excitation voltage.

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* National Research Fellow.

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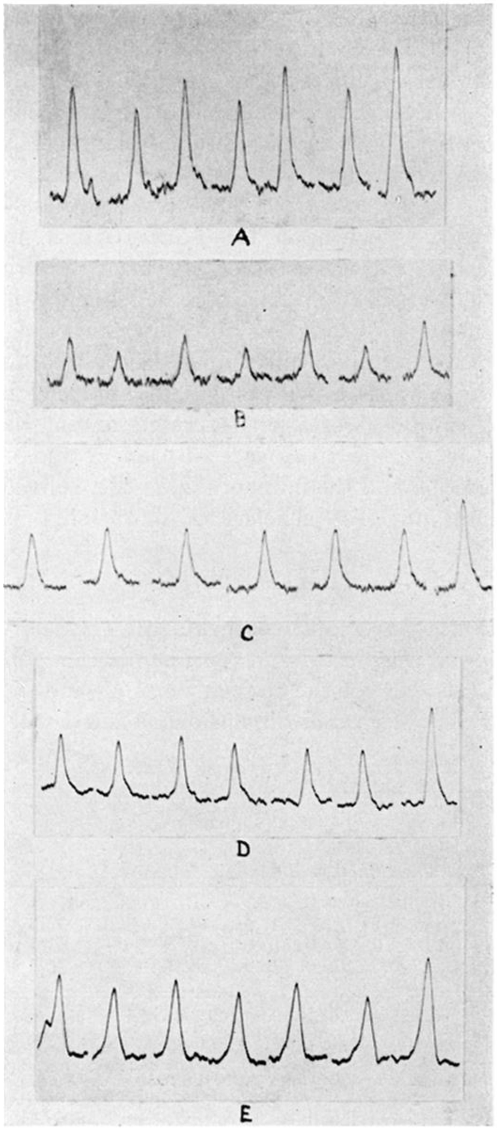

FIG. 1. Typical microphotograms of reflection pictures taken from rectangular quartz plates. Exposures: 10 min., 1st, 3rd, and 5th lines, vibrating; 2nd, 4th, and 6th lines, at rest; 20 min., 7th line, at rest. (A) X vibration of Y-cut crystal. (B) Z vibration of X-cut crystal. (C) Same as (B), except that the crystal surface was polished. (D) Y vibration of Y-cut crystal. (E) X vibration of X-cut crystal.

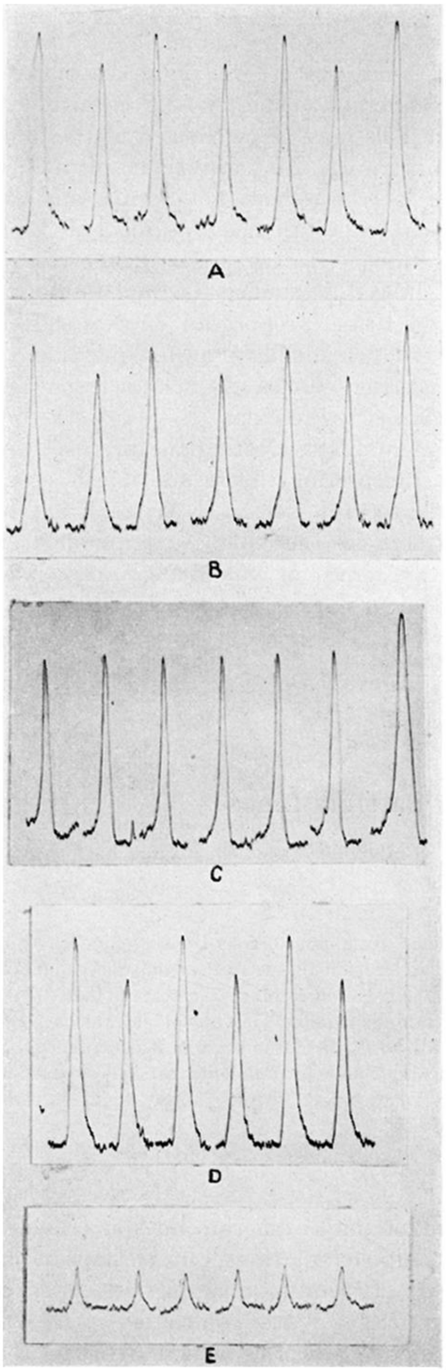

FIG. 2. Typical microphotograms of reflection pictures with the Y vibration of the 10 cm X-cut quartz bar. Exposures: 5 min., 1st, 3rd, and 5th lines, vibrating; 2nd, 4th, and 6th lines, at rest. 10 min., 7th line, at rest. (A) Middle position of bar, plate voltage 320. (B) 3 cm from middle of bar, plate voltage 320. (C) 4.5 cm from middle of bar, plate voltage 320. (D) Same as (B), except plate voltage 500. (E) An end of the bar, plate voltage 500.