

## Energy Distribution of Secondary Electrons from MgO Single Crystals

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(Received June 6, 1957)

The secondary-electron energy distribution from single crystals of MgO has been measured. A dependence of energy distribution on primary electron energy is established. The dependence is consistent with a qualitative interpretation based on the path length of the secondaries inside the crystal. The single-pulse measurements method has been modified to permit greater sensitivity, so that  $10^4$  electrons per pulse are sufficient for a 30:1 signal-to-noise ratio. Two procedures for establishing the surface potential are compared. Vacua of the order of  $10^{-10}$  mm are employed.

### INTRODUCTION

PRESENT knowledge of the energy distribution of secondary electrons from bulk insulators is very limited.<sup>1</sup> This investigation was undertaken to obtain information on the energy of secondaries from bulk single crystals of MgO in order to assist in elucidating the mechanism of secondary emission in insulators.<sup>2</sup> MgO was selected because of its refractoriness and inertness, its relative stability under small amounts of electron bombardment, and its availability in single-crystal form.

### APPARATUS

Charging of the insulator surface has seriously limited studies of the energy of secondary electrons from bulk insulators. The resolution obtainable in energy-distribution measurements on insulators is dependent on the amount of charging of the target during the measurement and the uniformity of the target surface potential. A single-pulse method has been employed recently<sup>3</sup> to obtain energy-distribution measurements. The number of electrons in a single primary pulse is kept sufficiently small that charging of the surface is held to a tolerable level. The surface potential is then re-established before each measurement.

In the present experiment the sensitivity of the single-pulse method has been increased considerably over that used by other workers.<sup>3,4</sup> Consequently a brief description of the method of making measurements is presented.

A single pulse from a pulse generator results in a burst of primary current to the target. Signals generated in the target circuit are amplified, displayed on an oscilloscope, and photographed. The procedure for determining the yield is the same as that described by Johnson and McKay.<sup>3</sup> However, a large preamplifier input resistor ( $10^8$  ohms) is used so that the input  $RC$  time constant is much longer than the primary-current

pulse duration of several microseconds. The input signal then corresponds to the accumulation of charge during the period of the current pulse. In using the large input resistance a considerable improvement in sensitivity is achieved, and the amount of charge necessary per measurement is consequently reduced. Since the determination of yield involves the ratio of the emitted to incident charge, linear circuit elements are used to limit the band width and further reduce the noise. The system sensitivity in the present experiment is such that  $10^4$  electrons result in a 1-cm deflection on the oscilloscope with a 30:1 signal-to-noise ratio. The primary current pulses were of the order of a milli-microampere and of several microseconds duration.

The energy of the secondaries is measured by the electrostatic retarding-potential method, with the target located at the center of a spherical collector. The collector consists of a spherical glass bulb of 5-inch diameter coated with colloidal graphite to assist in the suppression of secondary electrons formed at the collector surface.

The MgO single crystals,<sup>5</sup> 1 cm $\times$ 1 cm in area, were cleaved in air along the (100) plane to a thickness of  $\frac{1}{2}$  mm immediately before being mounted in the vacuum tube. Consequently the measurements must be taken as typical of gas-covered surfaces of MgO single crystals. Pressures obtained in the vacuum system were of the order of  $10^{-10}$  mm Hg on a continuously pumped system after a bakeout at 250°C. Using the present pulse method and a  $\frac{1}{4}$ -inch beam diameter, the charging of an MgO crystal of these dimensions can be held to 0.05 volt per pulse.

### EXPERIMENT

#### A. Stabilization of Surface Potential

In order to measure the energy of the secondary electrons by the single-pulse method, the target surface must be returned reliably to the same potential before each measurement. Two procedures for stabilizing the surface potential have been compared to determine their applicability for energy-distribution measurements. Retarding-potential curves using each of the

<sup>5</sup> The MgO crystals were obtained from Infra Red Development Company, Welwyn Garden City, Hertfordshire, England.

<sup>1</sup> For a review of this field see K. G. McKay, in *Advances in Electronics* (Academic Press, Inc., New York, 1948), Vol. 1, p. 65.

<sup>2</sup> A preliminary account of this work was presented at the New Haven meeting of the American Physical Society [N. R. Whetten and A. B. Laponsky, *Bull. Am. Phys. Soc. Ser. II*, **1**, 279 (1956)].

<sup>3</sup> J. B. Johnson and K. G. McKay, *Phys. Rev.* **91**, 582 (1953).

<sup>4</sup> H. L. Heydt, *Rev. Sci. Instr.* **21**, 639 (1950).

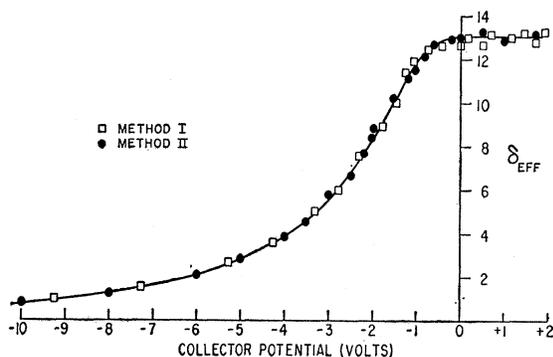


FIG. 1. Retarding-potential curves comparing stabilization methods. The open squares refer to stabilization of the surface potential by thermionic electrons from a nearby filament. The closed circles refer to stabilization by heating the crystal so that conductivity is sufficient to relax the surface charge. The curves have been shifted to correct for contact potential.

surface-potential stabilization methods have been obtained on an MgO crystal. Since the energy distribution of the secondaries is very sensitive to changes in surface potential, the retarding-potential curves provide a sensitive indicator of differences in stabilization.

Method I utilizes a thermionic emitting filament located near the collector surface and concentric with the aperture. A small solid angle is subtended at the target to minimize the distortion of the retarding-potential field. Electrons from the filament lower the target surface potential to the lowest potential of the filament. A tantalum filament was used in these measurements, and was operated at  $1400^{\circ}\text{C}$ . In method II the target is heated until conductivity is sufficient to relax the charges. The target is then cooled before each measurement. The tungsten substrate for the crystal was heated to  $750^{\circ}\text{C}$ .

Retarding potential curves are presented in Fig. 1, and have been shifted to correct for contact potential. There is seen to be good agreement between the curves of methods I and II. Similar agreement was found by using a combination of methods I and II. The data in

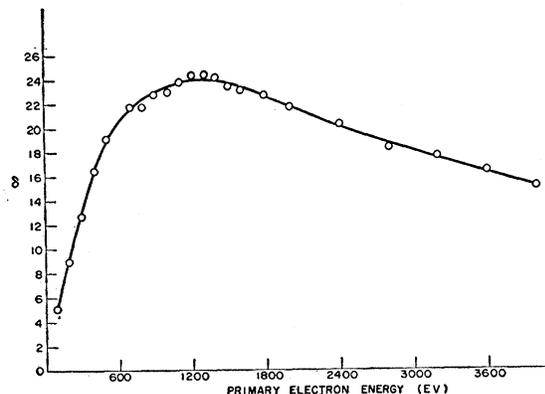


FIG. 2. The secondary-emission yield  $\delta$  is given as a function of primary electron energy for an MgO single crystal.

Fig. 1 were taken at 800-eV primary energy. Corresponding agreement was found at other primary energies.<sup>6</sup>

We have used method I to obtain the energy-distribution curves presented in the following section. The time required for stabilization is much shorter than when the heat-discharge scheme is employed, and the disadvantage of possible changes in the crystal due to heating is eliminated. An internal space charge may remain after stabilization by method I. However, we have found no indication that the secondary energy distribution from single crystals of MgO is affected by such a space charge.

### B. Change in Energy Distribution with Primary Energy

Measurements of energy distribution of the secondary electrons have been made on six different single crystals of MgO. The data for the different crystals are simi-

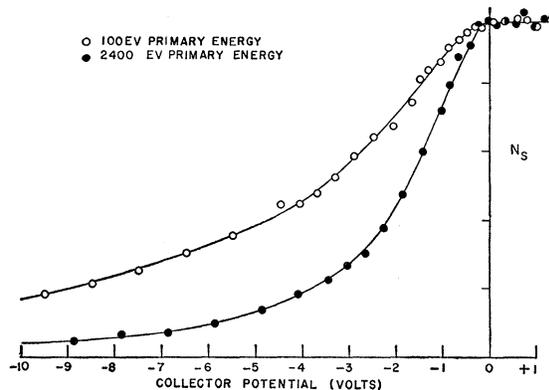


FIG. 3. Retarding-potential curves for different primary energies. The open circles refer to 100-eV primary energy, the closed circles to 2400-eV primary energy. The curves have been scaled to match in the positive collecting-field region.

lar; therefore typical energy-distribution curves are presented.

Figure 2 presents the total yield,  $\delta$ , as a function of the primary electron energy for one of the crystals. Maximum yields in the range 20 to 25 have been observed for a number of crystals prepared in this manner.<sup>7,8</sup>

Retarding-potential curves are presented in Fig. 3. The abscissa corresponds to the retarding potential applied to the collector surrounding the MgO target. Measurements were taken on the same crystal at dif-

<sup>6</sup> Johnson and McKay (reference 4) have compared method II with stabilization by electron bombardment to the collector potential. They find that the latter method results in a significantly different retarding-potential curve which they ascribe to patch-field effects. The writers have obtained similar data confirming the conclusion that the electron-bombardment stabilization method is not suitable for energy-distribution measurements.

<sup>7</sup> R. G. Lye, Phys. Rev. **99**, 1647 (1955).

<sup>8</sup> N. R. Whetten and A. B. Laponsky, J. Appl. Phys. **28**, 515 (1957).

ferent primary electron energies as noted in the caption. The curves have been scaled to be the same magnitude in the positive collecting-field region. The retarding-potential curves corresponding to the higher primary energies have a steeper initial slope, and consequently a relatively greater number of very low-energy electrons. This is also shown in Fig. 4, where the curves have been differentiated to give the energy distribution of the secondaries. The distributions have been normalized so that the same number of secondaries is considered for each curve. It is apparent that the relative number of low-energy electrons in the distribution increases with increasing primary energy. This change in distribution with primary energy is very reproducible with all of the crystals that have been examined. The shift becomes less pronounced at primary energies well above the maximum in the yield curve.

The most probable energy of emission of the second-

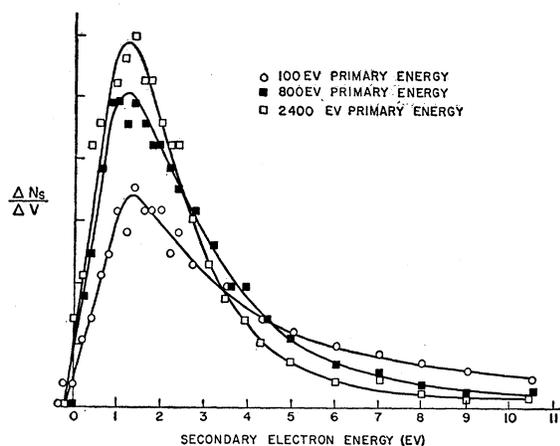


FIG. 4. Secondary-electron energy-distribution curves. The curves correspond to 100-, 800-, and 2400-eV primary energy and have been normalized to include the same number of secondaries in each group.

aries from MgO single crystals is about 1 eV in agreement with results of Johnson and McKay.<sup>3</sup>

#### DISCUSSION

The observed increase in the relative number of low-energy secondary electrons with increasing primary energy for MgO single crystals appears to be inconsistent with results of Vudinsky<sup>9</sup> on NaCl. Vudinsky reports that the average energy of the secondaries increased as the primary energy was increased. It is probable that Vudinsky's work was affected by insufficient stabilization of the surface potential since, as Vudinsky points out, a change in the primary current altered the shape of the energy distribution. The present results are consistent with recent observations of Shul'man and Dement'ev<sup>10</sup> on NaCl and KBr. Shul'man and Dement'ev stabilize the surface potential

<sup>9</sup> M. M. Vudinsky, *J. Tech. Phys. U.S.S.R.* **9**, 1583 (1939).

<sup>10</sup> A. R. Shul'man and B. P. Dement'ev, *J. Tech. Phys. U.S.S.R.* **25**, 2256 (1955).

by heating the target while the measurements are being made as does Vudinsky, but probably have less difficulty with charging because of their multiple-pulse technique. The present results obtained with the single-pulse technique and stabilizing the surface potential before each measurement are taken to be free from extraneous effects due to charging. The single pulse method eliminates the necessity of using thin films or of heating the insulator until conductivity becomes sufficient to prevent the buildup of surface charge. Both of these latter methods are subject to the possibility that the results may not be characteristic of bulk insulators, but may derive from the properties of thin films or from the elevated temperature of the insulator.

The increase in the relative number of low-energy secondaries with increasing primary electron energy may be qualitatively explained by the change in average path length of the secondaries with primary energy. This interpretation is similar to that presented by Stern-glass and Wachtel<sup>11</sup> to explain their data on secondaries from the exit side of evaporated KCl films. We assume that the energy distribution of the secondaries as they are produced inside the crystal does not vary with primary energy in the range investigated. There is little direct experimental evidence available regarding this assumption. However, such a dependence need not be invoked to qualitatively interpret the data. When the primary energy is low, the secondaries are formed close to the surface. Many of them escape without losing energy through interactions while in the crystal. At high primary energies some of the secondaries traverse a considerable distance before arriving at the surface. They lose energy by interactions with lattice vibrations and consequently are more degraded in energy upon arrival at the surface than are the secondaries formed by very low-energy primary electrons. Consequently, relatively more low-energy secondaries emerge as the primary energy is increased, in agreement with the experimental data.

#### CONCLUSIONS

Some specific results of this investigation can be summarized as follows:

1. A dependence of energy distribution of secondary electrons on primary electron energy for MgO single crystals is established. The relative number of low-energy secondaries increases as the primary energy is increased. These results are consistent with a qualitative interpretation based on the average path length of the secondaries inside the crystal.

2. The most probable energy of emission of the secondaries from MgO single crystals is of the order of 1 eV in agreement with conclusions of Johnson and McKay.

3. The sensitivity of the single-pulse method for

<sup>11</sup> E. J. Stern-glass and M. M. Wachtel, *Bull. Am. Phys. Soc. Ser. II*, **1**, 38 (1956).

obtaining secondary-emission measurements has been considerably increased, so that  $10^4$  electrons are necessary for a 1-cm deflection on an oscilloscope with a 30:1 signal-to-noise ratio.

4. No difference in surface-potential stabilization is found between the heat-discharge scheme and stabilization by thermionic electrons from a heated filament.

#### ACKNOWLEDGMENTS

The authors wish to express their thanks to P. E. Pashler, Professor W. G. Shepherd, and J. R. Young for many stimulating discussions of this work, to P. J. van Heerden for suggestions regarding the pulse techniques employed, and to C. R. Bunting for assistance in making the measurements.

## Effects of X-Ray Irradiation on the Frequency-Temperature Behavior of AT-Cut Quartz Resonators

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(Received April 12, 1957)

Results of an investigation of the effects of x-ray irradiation on AT-cut resonators fabricated from natural quartz, synthetic quartz grown on several different cuts of seed plates, and synthetic quartz containing aluminum or germanium as an impurity are reported. The differences in the frequency-temperature characteristics of AT-cut resonators fabricated from natural quartz and all synthetic quartz investigated are known. These differences are nullified by x-ray irradiation of all the synthetic quartz except one which contains germanium. The effect of x-ray irradiation can be removed by thermal bleaching at temperatures between 200 and 500°C for a few hours. The thermal-bleaching temperature is different for different types of quartz.

### 1. INTRODUCTION

IT has been reported<sup>1</sup> that the frequency-temperature characteristics of AT-cut resonators fabricated from synthetic quartz grown on different cuts of seed plates and synthetic quartz containing aluminum, germanium, or other impurities are different from those of resonators fabricated from natural quartz. These differences are measured in terms of inflection temperatures and optimum angles.

Effects of x-ray irradiation of quartz crystals have been observed in discoloration, in frequency change<sup>2</sup> of BT- and CT-cut oscillator plates, in the measurement of dielectric losses,<sup>3</sup> in optical absorptions,<sup>4</sup> etc. The change of the frequency of an oscillator plate due to x-ray irradiation cannot yet be explained by a simple theory. Possible parameters which would change the frequency are the changes of elastic constants, dielectric constants, piezoelectric constants, and the density of quartz. The change of any of the physical constants would have to be interpreted by the change in the

crystal structure of quartz. The change in the crystal structure may be the result of imperfections or the incorporation of impurities by substitution or in the interstices of the crystal structure. For instance, it is not unreasonable to consider the presence of sodium in synthetic quartz since the substance is grown in a sodium carbonate solution by a hydrothermal method. In the synthesis of aluminum-doped quartz,<sup>5</sup> sodium aluminate ( $\text{NaAlO}_2$ ) was added to the sodium carbonate growth solution. Different concentrations of sodium aluminate in the growth solution resulted in different concentrations of aluminum as impurities in the quartz.<sup>6</sup>

The crystal structure of quartz is also distorted by irradiation with x-rays or bombardment with electrons. The nature of lattice distortion due to neutron irradiation has been subject to considerable theoretical study. Seitz<sup>7</sup> described the development and present status of the theory concerning such radiation-induced lattice imperfections in solids.

Bömmel, Mason, and Warner<sup>8</sup> observed two relaxation peaks in quartz crystals at 20° and 50°K. According

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<sup>1</sup> A. R. Chi, Institute of Radio Engineers Convention Record, Part 9, 70-75 (1956).

<sup>2</sup> V. E. Bottom and V. Nowicki, "Engineering Memorandum No. 1," Long Branch Signal Laboratory, September, 1945 (unpublished).

<sup>3</sup> Volger, Stevels, and Van Amerongen, Philips Research Repts. 10, 260-280 (1955).

<sup>4</sup> G. Arnold, Proceedings of the Tenth Annual Frequency Control Symposium, Signal Corps Engineering Laboratory, 61, 1956 (unpublished).

<sup>5</sup> J. M. Stanley and S. Theokritoff, Am. Mineralogist 41, 527-529 (1956).

<sup>6</sup> Spectrochemical analysis of impurities in a quartz crystal was made by J. Mellichamp, Chemical Physics Branch, Signal Corps Engineering Laboratory. The elements in the quartz crystal are measured in parts per million of the total weight of the sample.

<sup>7</sup> F. Seitz, *Imperfections in Nearly Perfect Crystals* (John Wiley and Sons, Inc., New York, 1952), Sec. 1; J. M. Koehler and F. Seitz, *Report of the Bristol Conference on Defects in Crystalline Solids, July, 1954* (The Physical Society, London, 1955).

<sup>8</sup> Bömmel, Mason, and Warner, Phys. Rev. 102, 64 (1956).