Mechanical stress in a dielectric solid from a uniform electric field

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Mechanical stress in a dielectric solid from application of a uniform electric field is usually assumed to be described by "Maxwell stress," proportional to the first power of the relative dielectric constant, κ. Significant corrections are found from energy minimization when the dependence of permittivity on strain is included. Electrostriction coefficients are evaluated by the use of a model dielectric consisting of a simple-cubic lattice of linearly polarizable point dipoles. Compressive stress in the applied-field direction is larger than expected by more than a factor of κ. The force density exerted on internal space charge needs to be corrected by the same factor. Stress components also have been calculated, with identical results, through direct summation of microscopic forces. This method permits identification of the origins of electrically induced stress. The dominant contribution is a compressive stress in the field direction, proportional to κ^2, from attraction between free charge at the electrodes. This component can attain tens of MPa at fields approaching the intrinsic dielectric strength. A lateral tensile stress independent of κ also is present, which may assist electrical breakdown in some crystalline dielectrics. These stress components are augmented by short-range, dipolar forces throughout the bulk of the dielectric. Deformations accompanying poling of poly(vinylidene fluoride) are considered and found to be influenced by electrically induced stress.

I. INTRODUCTION

Although the topic of electrically induced, mechanical stress in dielectric solids is over a century old, its complexity is not widely appreciated. References 1 through 8 are a few of the many publications which point out details that must be considered; no convenient formula is available for evaluating the forces. Regardless of this, almost invariably when a determination of the electrically induced stress is required, the so-called "Maxwell stress" $\kappa\epsilon_0 E^2/2$, where E is the field magnitude, ϵ_0 is the permittivity of free space, and κ is the relative dielectric constant, is either presumed to be applicable or obtained through a superficial virtual-work derivation. Similarly, the force density exerted on a dielectric containing a distribution of free charge, ρ_f, is taken to be $\rho_f \mathbf{E}$. We find in both cases that the actual stress can be more than a factor of κ larger.

A standard, thermodynamically derived, textbook expression for the volume force density is[3]

$$\mathbf{F}=\rho_f\mathbf{E}-\tfrac{1}{2}\epsilon_0 E^2\nabla\kappa+\tfrac{1}{2}\epsilon_0\nabla\left[E^2\eta\frac{\partial\kappa}{\partial\eta}\right]. \quad (1)$$

The force density depends not only on the more familiar quantities but also on the variation of the dielectric constant with strain, here represented by the derivative with respect to η, the material density. Correct values of stress can be obtained from this formula, to the extent that the strain dependence of κ is approximated by a density dependence. Although Sec. II B shows such an approximation to be inaccurate, Eq. (1) serves to illustrate an important point: When the variability of κ is ignored a major source of electrostrictive effects, the third term in the expression, is discarded. Aside from forces due to space charge, only the electrostatic-energy density, or Maxwell stress remains.

The tendency for expressions such as Eq. (1) to be overlooked in stress calculations may be practical in nature. Force contributions described by the gradient terms are difficult to evaluate fully since major portions reside in regions of complicated fringing fields even with simple geometries. In addition, the obscurity of the underlying physics may present an impediment to comprehension or belief.

Electrically induced forces usually have little consequence, but there are several areas of study in which they cannot be ignored. One example is the electrooptic response of a dielectric material displaying strain birefringence. Perhaps of greater importance are two areas of current interest. (1) Zeller and Schneider[9] have argued that electrofracture mechanics are intimately involved in dielectric aging and breakdown; we show that considerable stress arises even with uniform fields. (2) The mechanism of poling in the ferroelectric polymer poly(vinylidene fluoride) remains controversial; consideration of electrically induced forces may help to resolve the discrepancies in measured lattice constants[10–12] and account for deformations which accompany poling.

The goal of this article is to demonstrate, through straightforward calculations, what forces are present under the simplest of conditions, a parallel-plate capacitor containing a homogeneous dielectric. Two methods of calculation leading to identical results will be pursued. In Sec. II A the energy of the system is minimized to obtain Eqs. (7) and (8) for the stress induced by the applied field. These expressions include strain derivatives of the dielectric constant, which are evaluated in Sec. II B through the adoption of a microscopic model for the dielectric. Finally, a direct computation of the field-induced stress is carried out in Sec. II C by summation of microscopic forces. These exercises allow the origins of the stress components

to be identified, from which useful, generally applicable principles are found.

II. CALCULATION OF STRESS COMPONENTS

A. Energy minimization

In what follows we restrict our attention to an ideally simple situation, a macroscopically homogeneous sheet of dielectric material of uniform thickness, d, with electrodes deposited on its faces. A principal electric axis is assumed to parallel the thickness direction $\hat{\mathbf{z}}$, so that an electric field, $\mathbf{E}=(V/d)\hat{\mathbf{z}}$, results from the application of a potential difference V to the electrodes. Likewise, the polarization vector will lie in the thickness direction parallel to $\mathbf{E}$. Lateral dimensions of the sheet are required to be much larger than its thickness and can be considered infinite, allowing the effects of fringing fields at the edges of the sheet to be ignored. Although beyond the scope of this treatment, generalization to more complex situations should be possible following the procedures outlined below.

The first method of stress calculation is based on the minimization of free energy. Including the energy stored in the voltage source, the electrical free energy is given by

$$F_e=-CV^2/2\ , \tag{2}$$

where the capacitance is[13]

$$C=\kappa_3\epsilon_0 A/d \tag{3}$$

and A is the lateral area of the sheet. A dependence on the tensile-strain components, s_i,[14] is introduced into the dielectric constant through the uniaxial electrostriction coefficients γ_{3i},

$$\kappa_3=\kappa(1-\gamma_{31}s_1-\gamma_{32}s_2-\gamma_{33}s_3)\ , \tag{4}$$

where $\gamma_{3i}=-\kappa^{-1}\partial\kappa_3/\partial s_i$. The change in capacitance due to changes in area and thickness must also be included. The electrical free energy is then

$$F_e=-\frac{\kappa\epsilon_0 AV^2}{2d}(1-\gamma_{31}s_1-\gamma_{32}s_2-\gamma_{33}s_3+s_1+s_2-s_3)\ . \tag{5}$$

The mechanical contribution to the free energy, F_m, is also needed. At this point a set of elastic constants could be assigned and an expression for F_m obtained. After finding the strain which minimizes the sum of F_e and F_m, the corresponding stress components, σ_i, could be recovered with the aid of the elastic constants that had been adopted. The stress, however, is independent of elastic constants and a much simpler approach is to expand the mechanical free energy about the strain at equilibrium. The differential is given by

$$dF_m=Ad\ (\sigma_1 ds_1+\sigma_2 ds_2+\sigma_3 ds_3)\ . \tag{6}$$

We now solve the equations, $\partial(F_e+F_m)/\partial s_i=0$, for the stress components, from which

$$\sigma_1=\tfrac{1}{2}\kappa\epsilon_0 E^2(1-\gamma_{31})\ , \tag{7}$$

$$-\sigma_3=\tfrac{1}{2}\kappa\epsilon_0 E^2(1+\gamma_{33})\ . \tag{8}$$

For simplicity, σ_2 is assumed to be equal to σ_1. The quantities in the parentheses represent corrections to the Maxwell-stress value which arise from the variation of κ with strain. Provided these quantities are both positive the stress in the thickness direction is compressive, as would be expected, but is tensile in the plane of the sheet.

B. Model dielectric

In order to complete the stress calculation a model dielectric is needed from which the electrostriction coefficients may be evaluated. A simple-cubic lattice of linearly polarizable point dipoles is an uncomplicated conceptual dielectric which resembles actual materials, for example, alkali halides.[15] This model will be adopted because the summations needed to find the field or field gradient at a dipole site are relatively easy to perform on a cubic lattice.

A Lorentz-cavity approach is used to find the field at dipole sites;[16] however, the lattice constants are no longer equal in all three directions but altered according to the uniaxial strain components. It is convenient to allow the Lorentz cavity to be deformed along with the array of dipoles; in an undeformed lattice the cavity would be a perfect cube, $2N+1$ lattice constants wide (with N an arbitrary integer), centered on a dipole site. The field at the central dipole site is the superposition of fields from three sources: the macroscopic applied field, the fictitious charge on the cavity wall which accounts for distant dipoles, and nearby dipoles within the cavity. The latter two contributions both depend on strain.

The field $\mathbf{E}_d$ due to nearby dipoles is considered first. A single dipole of moment m (proportional to the local field at dipole sites) oriented in the $\hat{\mathbf{z}}$ direction and located at a point $\mathbf{r}$ produces an electric field at the origin given by

$$\mathbf{E}_m=\frac{m}{4\pi\epsilon_0}\left[\frac{3(\mathbf{r}\cdot\hat{\mathbf{z}})\mathbf{r}-r^2\hat{\mathbf{z}}}{r^5}\right]\ . \tag{9}$$

Due to the symmetrical arrangement of nearby dipoles, only the z component will survive in the field sum. Furthermore, the strain can be factored out to first order, resulting in

$$\mathbf{E}_d=\frac{m\hat{\mathbf{z}}}{4\pi\epsilon_0 a^3}(s_1+s_2-2s_3)\times\sum_{i,j,k=-N}^{N}\frac{i^4+j^4+k^4-3(i^2j^2+j^2k^2+i^2k^2)}{(i^2+j^2+k^2)^{7/2}}\ , \tag{10}$$

where a is the undeformed lattice constant; i, j, and k take integer values; and $\mathbf{r}=0$ (i, j, and k simultaneously zero) is excluded from the sum. If the summation is carried out one shell of dipoles at a time, successive contributions rapidly approach a N^{-3} power law, and an extrapolation can be made to an infinitely large value of N. To seven digits, the sum has the value 4.035 766.

The remaining strain-dependent field, $\mathbf{E}_w$, comes from the Lorentz-cavity wall. We note that the macroscopic polarization is the dipole moment per unit volume, m/a^3, and obtain

$$\mathbf{E}_w=\frac{m\hat{\mathbf{z}}}{3\epsilon_0 a^3}[(1-s_1-s_2-s_3)+(\sqrt{3}/\pi)(s_1+s_2-2s_3)] . \tag{11}$$

This field contains two strain-dependent terms, the first of which responds to changes in the density of dipoles while the second, like $\mathbf{E}_d$, is insensitive to purely volumetric deformations.

Now that the field at a dipole site has been determined, the electrostriction coefficients may be obtained by solving for the macroscopic polarizability and differentiating with respect to strain. The results are

$$\gamma_{31}=\frac{(\kappa-1)(\kappa+2)}{3\kappa}-0.504\,932\frac{(\kappa-1)^2}{\kappa} , \tag{12}$$

$$\gamma_{33}=\frac{(\kappa-1)(\kappa+2)}{3\kappa}+1.009\,864\frac{(\kappa-1)^2}{\kappa} . \tag{13}$$

The first term in each of these is identical to the familiar result from differentiating the Clausius-Mossotti relation with respect to density; the second terms contain numerical constants specific to the cubic lattice. These constants agree with those found by Purvis and Taylor[17] using a somewhat different method, from strain derivatives of the Lorentz factor, but are expressed to greater accuracy.

Quantities needed to evaluate the tensile stress in the plane of the sheet [Eq. (7)], and compressive stress in the thickness direction [Eq. (8)], are listed in Table I for various values of the dielectric constant. The stress component σ_3 is larger, by more than a factor of κ, than the Maxwell-stress value, while the correction to σ_1 passes through a minimum near $\kappa=3$.

It is of interest to compare our results from the cubic lattice with those from a hypothetical dielectric described by Eq. (1), in which the dielectric constant depends only on density. Evaluating the stress in the latter dielectric is readily accomplished by dropping the second terms in Eqs. (12) and (13). Results are listed in Table II. For small values of κ, σ_1 is tensile and σ_3 compressive, as with the cubic lattice. At κ values beyond 2.73, however, σ_1 changes sign and becomes compressive, approaching σ_3 at large κ.

C. Summation of microscopic forces

Our model dielectric is ideally suited to a direct calculation of stress components by summation of microscopic forces, a method which helps to elucidate the underlying physics. Results will be compared with those obtained in the preceding section. The stress in the thickness direction is considered first.

TABLE I. Stress components σ_1 and $-\sigma_3$ in a cubic lattice of point dipoles, normalized to $\kappa\epsilon_0E^2/2$.

κ	σ_1 $(1-\gamma_{31})$	$-\sigma_3$ $(1+\gamma_{33})$
1	1.00	1.00
2	0.59	2.17
3	0.56	3.46
5	0.75	6.10
10	1.49	12.78
20	3.15	26.19

TABLE II. Stress components σ_1 and $-\sigma_3$ in a hypothetical dielectric having $\gamma_{31}=\gamma_{33}=(\kappa-1)(\kappa+2)/3\kappa$, normalized to $\kappa\epsilon_0E^2/2$.

κ	σ_1 $(1-\gamma_{31})$	$-\sigma_3$ $(1+\gamma_{33})$
1	1.00	1.00
2	0.33	1.67
3	−0.11	2.11
5	−0.87	2.87
10	−2.60	4.60
20	−5.97	7.97

A cross section of the sheet of the dielectric is depicted in Fig. 1. As a conceptual aid the dielectric is partitioned into two regions by a plane perpendicular to the z axis. This boundary is indicated by the horizontal line across the center of the illustration between rows of dipole sites. The stress component σ_3 is simply the force, per unit area, exerted on all dipoles and free charge above the dividing plane by all dipoles and free charge in the lower region. By symmetry, only the z component of force is present.

Included in σ_3 is the force acting between dipoles. Our procedure is to find the field derivative $\partial E_z/\partial z$ at representative dipole sites in the upper region arising from dipoles in the lower region. The stress is accumulated layer by layer through multiplication of the force, $m\,\partial E_z/\partial z$, on single dipoles lying along the upper z axis by the density a^{-2} of dipole sites in a layer. With the aid of Eq. (9) these operations are expressed by

$$\sigma_3(\text{dipolar})=\frac{3m^2}{4\pi\epsilon_0 a^6}\sum_{k=1}^{n}\sum_{i,j=-N}^{N}\frac{k^2(3i^2+3j^2-2k^2)}{(i^2+j^2+k^2)^{7/2}} . \tag{14}$$

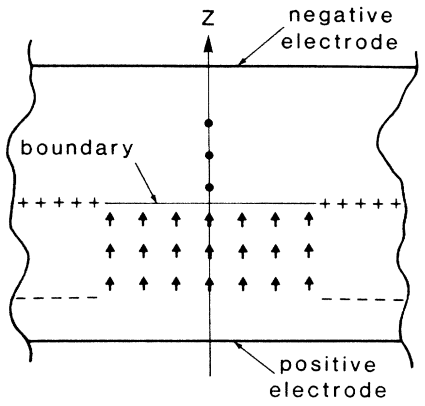

FIG. 1. Cross section of the model dielectric, with the spacing between dipoles enlarged for clarity. The boundary is a conceptual aid in the calculation of σ_3; dipoles, and free charge at the electrode, in the lower region are the sources of the field in the upper region. Dots indicate the dipole sites at which $\partial E_z/\partial z$ is evaluated. The layers of charge are used to approximate the field gradient from laterally distant dipoles.

The indices i and j apply to the x and y coordinates, in units of the lattice constant, of dipoles in the lower region. Appropriately placed layers of fictitious charge, indicated in Fig. 1, are used to approximate the field gradient from laterally distant dipoles and avoid summation with an infinitely large value of N. The index k corresponds to the distance between a layer of dipoles in the lower region and a single dipole in the upper region on which force is exerted.

Dipolar contributions to σ_3 are extremely short range in nature. 99.7% of the total is accounted for by adjacent dipole layers, and $n=3$ is sufficient for seven-digit accuracy. The sum in Eq. (14) has the value $-0.718\,792\,3$, the minus sign indicating a compressive stress.

The only additional contribution to σ_3 is the attractive force, proportional to κ^2, between the layers of free charge at the two electrodes. The total stress component in the thickness direction is

$$-\sigma_3=0.343\,198(\kappa-1)^2\frac{\epsilon_0E^2}{2}+\kappa^2\frac{\epsilon_0E^2}{2}\ . \tag{15}$$

Both terms are compressive; the first is the short-range, dipolar contribution which originates throughout the volume of the dielectric and is proportional to the square of the polarization, while the second and larger term arises from pressure exerted by the electrodes.

When one compares Eq. (15) with the previous determination of σ_3, contained in Eqs. (8) and (13), the two expressions seem to be nonidentical because of the different numerical constants and arrangement of terms. These expressions, however, are equivalent within the seven-digit accuracy of the numerical constants. It may be of interest that the triple sums, $\Sigma^{(10)}$ and $\Sigma^{(14)}$, in Eqs. (10) and (14) evidently are related according to $3\Sigma^{(10)}+9\Sigma^{(14)}=4(\pi-\sqrt{3})$, in the limit $N,n\to\infty$, $n\ll N$.

The stress in the plane of the sheet is obtained by a procedure similar to that employed above. The dividing boundary is now oriented perpendicular to the x axis as shown in Fig. 2, and the x component of force on a single dipole depends on the field derivative $\partial E_x/\partial z$. In order to find the short-range, dipolar contribution to σ_1 a summation analogous to that in Eq. (14) is needed, where the indices j and k, rather than i and j, range over an effectively infinite set of integer values. However, it is not necessary to perform this summation; derivatives of the dipolar field, Eq. (9), are related such that

$$\sigma_1(\text{dipolar})=-\sigma_3(\text{dipolar})/2\ .$$

The long-range contribution to σ_1 is less readily obtained. We consider first the source of remote electric fields. Both free charge and dipoles in the left-hand region produce fields which extend beyond the dividing boundary. The latter field arises when the width of the layer of dipoles depicted in Fig. 2 is allowed to become comparable to the thickness of the dielectric sheet. The remote field is, therefore, due to the sum of free and polarization charge at the electrodes in the left-hand region.

Force is exerted on both free charge and dipoles by the field extending into the right-hand region. Since this field is smoothly varying, summation over individual dipoles is replaced by integration over the volume of the dielectric. Integration of $\partial E_x/\partial z$ with respect to the z coordinate simplifies the volume integral to an area integral of the force on the polarization charge at the electrodes. Accordingly, free and polarization charge can be combined into a "total" electrode charge, and the long-range contribution to σ_1 is effectively the repulsion between the total charge on the two halves of each electrode, $\epsilon_0E^2/2$.

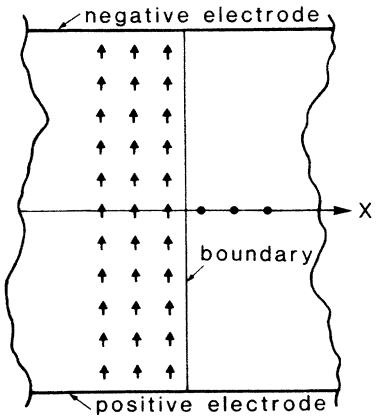

FIG. 2. Cross section of the model dielectric, arranged for calculation of σ_1. Dipoles and free charge are now divided into left and right regions. Dots indicate the dipole sites at which $\partial E_x/\partial z$ is evaluated.

From the paragraphs above, the stress in the plane of the sheet is the sum of two tensile terms,

$$\sigma_1=0.171\,599(\kappa-1)^2\tfrac{1}{2}\epsilon_0E^2+\tfrac{1}{2}\epsilon_0E^2\ . \tag{16}$$

The first term originates with the lateral repulsion between neighboring dipoles and dominates at κ greater than 3.41, while the second is long range in nature and independent of the dielectric constant. As was found with σ_3, the expression for σ_1 in Eq. (16) is a numerical identity with the result from energy minimization.

III. DISCUSSION

The mechanical stress can become quite large at fields approaching the intrinsic dielectric strength. For example, according to Table I, the compressive stress $-\sigma_3$ ranges from 40 to 450 atm (4 to 46 MPa) with $\kappa=3$ and electric fields from $(0.3-1.0)\times10^9$ V m^{-1}. Of particular interest is the presence of the tensile stress σ_1 perpendicular to the electric field in our model dielectric. Although calculated to be severalfold smaller in magnitude than σ_3, tensile components may play a role in the electrical breakdown of some crystalline dielectrics by helping to enlarge microfractures in cleavage planes lying parallel to the field direction.

It is also of interest that charge may be injected from the electrodes at high fields,[9,18] altering the internal-field and stress distributions. The force exerted on the dielectric as a result of a thin layer of space charge is readily determined in a one-dimensional situation by considering the field discontinuity at the layer of charge. The effective pressure is obtained from the discontinuity in stress at the layer, evaluated according to either Eq. (8) or (15). A

surprising result is found; the force density is larger by a factor of $(1+\gamma_{33})$ than the value usually assumed, $\rho_f \mathbf{E}$. The same conclusion is reached when one recognizes, guided by Eq. (15), that force arises from two distinct sources: the gradient in the short-range attraction between planes of dipoles within the region of space charge and the long-range interaction, $\kappa\rho_f \mathbf{E}$, between the space charge and free charge on the electrodes.

A paradox is suggested by the foregoing discussion. The force density exerted directly on the space charge is actually $\kappa\rho_f \mathbf{E}$ rather than $\rho_f \mathbf{E}$, but the latter is consistent with the transport of carriers in an applied field. This conflict is resolved upon consideration of the balance of forces. No net, time-averaged, force is experienced by a drifting carrier; the three separate contributions due to the field from free charge, fields from dipoles, and dissipative interactions sum to zero. The first two of these impel transport and are accounted for by the macroscopic field. With respect to stress, the first is the source of the force density exerted on the space charge while the remaining two serve to transfer this force to the lattice.

Knowing that a major stress contribution is simply the force between layers of free charge can be very useful. We consider, for example, the stress in polarized poly (vinylidene fluoride) at zero applied potential. The saturation remanent polarization of this inhomogeneous, semicrystalline polymer is about 6×10^{-2} C m^{-2},[19] from which the pressure $\sigma_f^2/2\epsilon_0$ exerted by the electrodes is $\approx$200 MPa if all the induced free charge resides on the electrodes. With a Young's modulus of 2.5×10^9 Pa,[20] an 8% thickness deformation is predicted from the free-charge attraction alone. This is more than adequate to explain the few-percent dimensional changes we observe upon poling the material. Of course, the actual situation is considerably more complex, with charge injected during the poling process and varying amounts of compensating space charge[21] at the faces of polarized crystalline domains. This will affect the distribution of stress, applying some directly to the less compliant[22] crystallites, as well as altering the internal field, which governs the dipolar forces through induced polarization. In the extreme case, polarization would be completely compensated by space charge at domain faces, resulting in zero fields (averaged over a molecular scale) in both crystalline and amorphous regions. Clearly the large electrically induced stress, in addition to affecting the macroscopic dimensions, influences the lattice constants in poly(vinylidene fluoride). Such stress could be present in the ferroelectric domains even without poling. Variations in the distribution of stress may well account for the half-percent discrepancies between published lattice-constant measurements.[10–12]

IV. CONCLUSIONS

The electrically induced stress has been derived for an ideally simple situation, a uniform field applied in the thickness direction of a dielectric sheet, parallel to a principal electric axis. Significant corrections to the Maxwell-stress value are found from energy minimization when the dependence of the permittivity on strain is included. A model dielectric consisting of a simple-cubic lattice of linearly polarizable point dipoles is employed to evaluate the electrostriction coefficients, and the correction to σ_3 is found to be greater than a factor of the relative dielectric constant, κ. Likewise, the force density exerted on internal space charge is larger than expected by the same factor.

Stress components also have been calculated through direct summation of microscopic forces, with results identical to those from energy minimization. The direct-summation method permits the origins of the electrically induced stress to be identified. The dominant contribution is a compressive stress in the applied-field direction, proportional to κ^2, from the attraction between free charge at the electrodes. A lateral tensile stress independent of κ also is present. These stress components are augmented by short-range, dipolar forces throughout the bulk of the dielectric.

The methods we have outlined can be generalized to more complex situations, or can be extended to lattices of dipoles other than simple cubic where different short-range forces will be found. Basic concepts, however, are immediately applicable to certain high-field phenomena of current interest, including dielectric breakdown and poling of ferroelectrics.

ACKNOWLEDGMENT

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is used to distinguish the dielectric constant applicable to an electric field along the z axis when the material is under strain (and not in general electrically isotropic) from κ, the dielectric constant in the absence of strain.

[14]In the notation used s_i has the value of the tensor tensile-strain component ε_{ii}. We do not consider any dependence of the dielectric constant on shear-strain components.

[15]The relationship between the local field and polarization derived with a simple-cubic lattice, $\mathbf{E}_{\text{loc}}=\mathbf{E}+\mathbf{P}/3\epsilon_0$, is valid for most alkali halide crystals. See J. R. Tessman, A. H. Kahn, and W. Shockley, Phys. Rev. **92**, 890 (1953).

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