approached the single peak broadens considerably and at room temperature three separate peaks are observed. The peak-height-to-background ratio is too unfavorable for a polarization analysis at this time. Further studies on the torsional mode are underway.

Even with its inherent limitations, the measurement of coherently scattered phonons in undeuterated hydrogenous substances should be extremely useful for studying the effects of isotopic substitution by comparing similar phonons measured in the deuterated substances. This technique is also invaluable for those materials where large deuterated crystals are not readily attainable.

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Effect of Thermal-Neutron Irradiation on the Elastic Moduli of LiF†

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The three elastic-constant changes, and the dilation, of LiF have been measured as a function of neutron flux. The elastic constants all decrease with flux, the change being linear for fluxes less than $10^{16} n/\mathrm{cm}^2$. This is contrary to expectations based on considerations of Dienes and Nabarro, and consequently, it makes measurements of elastic constants less useful as a tool for distinguishing between interstitials and vacancies than had previously been expected. The ratio of the relative bulk modulus to the volume change is -1.8, which is much smaller than some corresponding values previously reported for copper. A comparison of the experimental results with various available theories shows that a nonlinear elastic sphere-in-hole calculation disagrees the least, the expectations from Zener's considerations come next, and the rest of the theoretical estimates differ in order of magnitude and even in sign from the experimental results.

I. INTRODUCTION

THE measurements described here were stimulated by an unresolved question in the field of radiation damage. In 1952, Dienes[1–3] gave a theoretical estimate of the effect of radiation on the elastic constants of simple metals. For copper he predicted that interstitials would increase and vacancies would decrease the elastic constants by amounts of the order of 10 and 1% per at.% of interstitials and vacancies, respectively. He thus concluded that the effects should be easily observable in copper or similar metals provided thermal annealing is prevented, and that changes in elastic constants may serve as a useful tool for distinguishing between interstitial atoms and lattice vacancies.

Because of the important need of a measurement which distinguishes between vacancies and interstitials in the interpretation of radiation damage, this prediction was followed by a number of attempts to measure the elastic constants of irradiated materials. Although these calculations stimulated much work in radiation damage, the hopes held for such a measurement have by and large not been realized. The measurements have proved to be very difficult ones. It was early found that dislocation effects often overshadowed the bulk effects of point defects, and extensive studies of dislocation effects in irradiated materials have since been made.

In those cases where the dislocation effects have been isolated, there remains an enormous disagreement (two to three order of magnitude) between different investigators making measurements of the bulk effect on the same materials. At liquid-helium temperature in copper irradiated with α particles, König *et al.*[4] found a Young's modulus change of $\Delta E/E=-130\%$ per at. % of Frenkel defects, while Thompson *et al.*[5] found that the change, if any, for reactor irradiated copper at liquid-helium temperature was less than 1% per at. % of Frenkel defects. In earlier work at −195°C, Dieckamp and Sosin[6] reported a change of $\Delta E/E=-(7\pm3)\%$ per at. % of defects. This figure has since been revised[4] to -140% per at. % of defects since het number of defects formed by irradiation was assumed to be too high in the original work. In the König *et al.* and the Dieckamp and Sosin measurements, the Young's modulus of a polycrystalline copper foil fixed at one end to perform transverse oscillations was measured, whereas the Young's modulus of a single crystalline copper rod oscillating longitudinally was measured by Thompson *et al.* A review of the results of other measurements at different temperatures and in different materials is given by König *et al.* In no case have all the elastic constants been measured.

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The theoretical picture is also confused. Numerous other theoretical estimates have been given, and these also disagree over a range of two to three orders of magnitude (spanning the same range as the measurements) and even as to the sign of the effect. An argument has been given by Zener[7] which suggests that the effect of point defects should be similar to that of phonons. Zener points out that for both point defects and for thermal waves, the strain-energy content of the solid is mostly shear strain energy and he uses this to give an approximate calculation of the shear elastic-constant change of dilute alloys. On this basis, one might expect all the elastic constants to decrease with Frenkel defect content, in contrast to the prediction of an increase by Dienes. Also, one might expect the ratio of the bulk-modulus charge to the volume change to be similar to that for thermally induced changes, or about -5% per percent volume change for most materials. If the volume change per Frenkel pair is supposed to lie between 1 and 1.5 atomic volumes, then a bulk-modulus change of about -4 to -8% per % Frenkel pairs would be expected on this basis. An estimate by Nabarro[2] using a linear elasticity theory predicts 3.8% and -2.3% per % of interstitials and vacancies, respectively. On the other hand, a recent linear elastic calculation by Melngailis[8] yields values of the order of that found by König *et al.* and Dieckamp and Sosin. A different approach, based on finite anisotropic elasticity, will be described later which predicts magnitudes smaller than any of the above, in disagreement with the results of König *et al.* and Dieckamp and Sosin but not with the results of Thompson *et al.* and of the present work.

Faced with this complexity, the potential importance of this type of measurement for the field of radiation damage, the unsettled nature of both the experimental and theoretical situation at present, and the possibility that different elastic constants might behave quite differently from one another making comparisons between experiments difficult, it was decided that it might be useful to measure the three independent elastic-constant changes in neutron irradiated LiF. This system offers many advantages. It is relatively easy to obtain large effects at room temperature because of the large cross section of the capture reaction $\mathrm{Li}^6(n,\alpha)\mathrm{He}^3$ for thermal neutrons (950 b) with an energy release of 4.8 MeV per fission. As natural lithium contains 7.5% of Li^6, a large amount of damage is caused by thermal neutrons.

The properties of neutron-irradiated LiF have been extensively investigated. The main results have been reviewed and summarized by Gilman and Johnston[9] and Van den Bosch.[10] Measurements of optical absorption, lattice parameter, density and magnetic susceptibility show that the damage is stable at room temperature, annealing out in the 300–500°C range. Binder and Sturm[11] find that x-ray and density measurements are equivalent for exposures of 6×10^{16} *nvt*, giving evidence that vacancies and interstitials are introduced in pairs. For exposures greater than 3×10^{17} *nvt*, decreases in lattice expansion given by the x-ray measurements are found by Mayer and co-workers[12] and by Smallman and Willis,[13] but no decrease in the density was found by Senio and Tucker.[14] This has been interpreted to mean that the strains caused by interstitials have disappeared, with the interstitials going into defect clusters. There is some discrepancy between measurements by different investigators, most likely caused by differences of irradiation temperatures, which leads to some uncertainty in the interpretation. Annealing studies of lattice expansion by Binder and Sturm[15] support a vacancy-interstitial pair annihilation mechanism, but other workers find more complicated annealing kinetics which depend upon the neutron dose. Magnetic susceptibility measurements by Van den Bosch[10] show that isolated *F* centers are produced for low doses, but that these tend to agglomerate into *M* centers at doses of the order of 0.7×10^{15} n/cm^2. For doses greater than about 10^{18} n/cm^2, Li metal precipitates and density changes occur which are likely due to escaping fluorine gas.

The principal questions addressed in this investigation are then the following. Do any of the elastic constants ever increase with radiation damage, as might be expected from the work of Dienes? That calculation was specifically for copper, but it might be expected that similar considerations would apply as well to LiF since Born-Mayer terms in the energy have a major impact on the elastic constants in both cases. Are the relative effects on the different elastic constants and the volume similar to those found by temperature changes, as might be expected from the viewpoint of Zener? Do the different elastic constants behave differently enough to allow for the wide differences observed by different measurement techniques? Do the results favor any of the existing theoretical predictions?

II. EXPERIMENTAL METHOD

Single crystals of LiF were purchased from the Harshaw Chemical Co. They were in the form of cubes with cube faces corresponding to (110), $(1\bar{1}0)$, and (001)

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crystalline planes. The orientation of the faces was checked by means of x-ray Laüe back reflection, and found to be within 1° of the specified crystalline plane. One pair of faces on a 12-mm cube was hand-lapped flat and parallel, and all velocity measurements were carried out on this sample. The length changes were carried out on a cube having 9.6-mm edges.

The three independent elastic moduli, c_{11}, c_{12}, and c_{44}, were determined by measuring the sound velocity of the three propagation modes in the [110] direction, e.g., the longitudinal and the two shear waves, the latter polarized in the [1$\bar{1}$0] and [001] directions, respectively. The longitudinal elastic constant, c_L, is given by $c_L=(c_{11}+c_{12}+2c_{44})/2$; the [1$\bar{1}$0] polarized shear, c', is given by $c'=(c_{11}-c_{12})/2$, and the [001] polarized shear, c, is c_{44}. The 10 MHz sound waves were generated by $\frac{1}{4}$-in.-diam crystalline quartz transducers, X and Y cut for the longitudinal and shear waves, respectively, with phenyl salycilate (salol) serving as the bonding agent.

The sound velocities were measured by the McSkimin pulse-superposition method.[16,17] Since we were only interested in the changes in the sound velocity, the resonant frequency of the composite structure (bonded transducer crystal) closest to the resonant frequency of the transducer was monitored as a function of the integrated neutron flux. The frequency changes are the negative of the change in time of travel of a sound wave through the sample.[16,17] From these transit time changes, the variation in the sound velocity as a function of the integrated flux could be determined.

The volume dilation under irradiation was determined from the length changes in an irradiated sample. These changes were measured with a highly sensitive comparator gauge (Johansson Mikrokator), which was standardized prior to each measurement with gauge blocks. Simultaneously with the length changes, the weight of the sample was also checked to make sure there was no mass loss during irradiation. No mass changes greater than one part in 10^4 were detected.

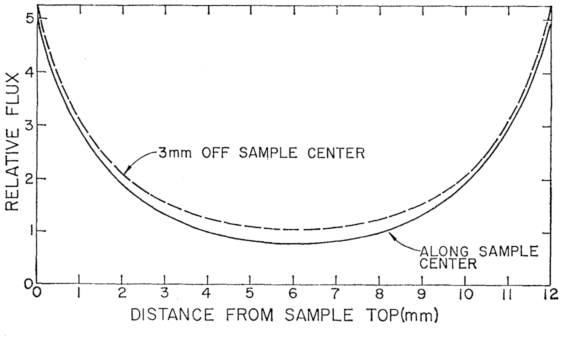

FIG. 1. Calculated variation of the flux inside a 12-mm cube of LiF relative to the flux outside the sample.

The neutron irradiations were carried out in the University of Illinois Mark II Triga graphite reactor. The lower-flux irradiations were carried out at the periphery of the core, while the final high-flux irradiations were done inside the core. The rise in temperature of the sample during irradiation is estimated never to have exceeded 60–70°C, so that no appreciable annealing of the damage should have occurred. The γ rays which are also present in the reactor cause a negligible amount of damage, and the same applies to fast neutrons.[9,10]

When evaluating the experimental data, the nonuniformity of the neutron distribution inside the sample, due to the self-shielding of the material, should be taken into account. As this is a substantial correction, the method used to make the correction will be described in detail. This correction is unavoidable in the present work as the specimen had to have dimensions large compared to the neutron penetration depth for the sound velocity measurements. The neutron spectrum in the Illinois reactor is approximately Lorentzian, centered about a neutron temperature of 190°C. This means the penetration depth in the LiF samples is 2.84 mm,[11] which is four to five times smaller than the over-all sample dimensions.

Because of the geometry of the sample, no analytic treatment can be given for this correction, so a numerical treatment is necessary. The flux at any point in the interior of the sample was found by numerically integrating the flux over all the faces of the sample. The resulting flux variation found in this manner for the 12-mm sample is shown in Fig. 1, both along a line through the center of the sample, and along a line parallel to the first but located midway between the sample center and one of the sample sides. Two important features can be noted from this figure. First, there will be at most a 10% variation in the flux in the region where the sound waves travel, since they are generated by the quartz transducers which have a diameter of about 6 mm. Therefore, the raw data for the elastic constants (as well as that for the length changes) were analyzed using the flux variation along the center and off-center lines through the sample and an average taken between the two. In no case did the two results differ by more than 10%.

The second feature of the flux variation noted from the figure is that the local flux throughout a large region of the sample is four to five times less than the flux at the surface. Therefore, the elastic-constant and length variations of the bulk specimen measured in the present work should yield information as to the local variations up to only a small fraction of the flux on the exterior of the specimen.

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In order to find these local variations the quantities were first expanded in a power series in the local flux:

$$\frac{\Delta l}{l}=\sum a_N\phi^N,\qquad \frac{\Delta c}{c}=\sum b_N\phi^N. \tag{1}$$

In the above, $\Delta l/l$ is the local length expansion, ϕ is the local flux, and c represents any of the elastic constants. These quantities were integrated (numerically) along the length of the specimen to give an expression for the total change in the bulk specimen:

$$\frac{\Delta L}{L}=\int_0^L \frac{\Delta l}{l}(z)\frac{dz}{L},$$

$$\frac{\Delta\tau}{\tau}=\int_0^L\left(\frac{\Delta l}{l}-\frac{\Delta v}{v}\right)\frac{dz}{L}=-\frac{1}{2}\left[\frac{\Delta L}{L}+\int_0^L\frac{\Delta c}{c}(z)\frac{dz}{L}\right], \tag{2}$$

where $\Delta L/L$ and $\Delta\tau/\tau$ are the measured length and transit time change in the bulk specimen, respectively, and $\Delta v/v$ is the local velocity change in the specimen.

The length change appearing in the elastic-constant expression is that appropriate for the 12-mm sample on which the transit time measurements were taken. Since the length of the specimen on which the length-change measurements were made was only 9.6 cm, it was necessary to correct these measurements before using them in the expression for $\Delta\tau/\tau$. This was accomplished by first analyzing the length-change measurements to find the local length-change-flux relationship and then (numerically) integrating this relationship over the 12-mm sample, using the flux variation calculated for that specimen.

Combining Eqs. (1) and (2),

$$\frac{\Delta L}{L}=\sum a_N\phi_0\left[\int_0^L\frac{R(z)dz}{L}\right]^N,$$

$$\frac{\Delta\tau}{\tau}=-\frac{1}{2}\left\{\frac{\Delta L}{L}+\sum_N b_N{\phi_0}^N\left[\int_0^L\frac{R(z)dz}{L}\right]^N\right\}. \tag{3}$$

Therefore, the expansions for the bulk crystal changes are similar to those for the local change, Eq. (1). However, the flux has been weighted by the factor involving the integral of $R(z)$, the ratio of the local flux to the exterior flux plotted in Fig. 4, over the length of the specimen. The weighting factor is in the range of 0.2–0.4, depending on the exponent N.

The coefficients in Eq. (1) were found by using a least-squares fit of the expressions in Eq. (3) to the smoothed experimental curve for each of the different parameters measured, where the integrals were again computed numerically. These fits were carried out for third-, fifth-, and seventh-order polynomial expansions in each case, but in all cases, it was found that the third-order fit gave the smoothest variation of the local expressions in Eq. (1). Also, in all cases the local expressions given in Eq. (1) began making enormous fluctuations at values of the flux higher than about $10^{17}n/\text{cm}^2$ or about six times less than the maximum surface flux of $6\times10^{17}n/\text{cm}^2$. This is undoubtedly the result of the large reduction of the flux inside the sample, and in keeping with the conclusions noted earlier in regard to the range of validity of the local expressions.

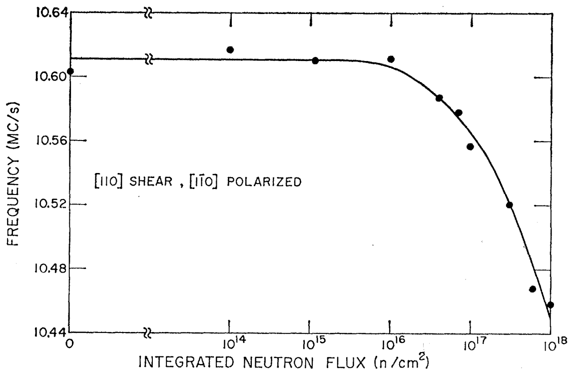

FIG. 2. Variation of the resonant frequency corresponding to the elastic constant C', as a function of integrated neutron flux on the surface of the 12-mm LiF sample.

III. RESULTS

All measurements were made at room temperature. Figures 2–4 show the experimental data of the change in the resonant frequency as a function of ϕ_0 for the three different sound-propagation modes. The integrated length-change result is shown in Fig. 5. From the smoothed curves of Figs. 2–5, the relative change in length, the longitudinal elastic modulus C_L, and the fast and slow shear moduli C and C' as a function of ϕ were determined by the method outlined above.

These quantities are shown in Figs. 6–9, respectively. The results are given only to $10^{17}n/\text{cm}^2$ for the reasons

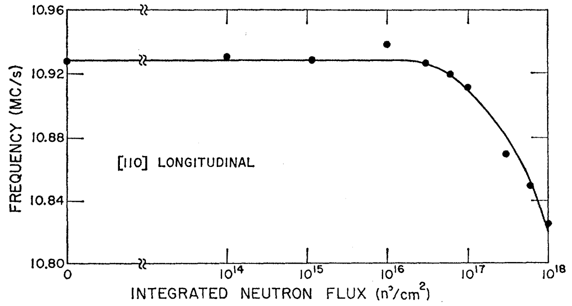

FIG. 3. Variation of the resonant frequency corresponding to the elastic constant C_L, as a function of integrated neutron flux on the surface of the 12-mm LiF sample.

already mentioned. It has also been noted above that each quantity calculated from the data was calculated using the flux variation along a central and an off-center line through the specimen, so the curves shown in Figs. 6–9 represent averages of these two calculations (differing by no more than 10%). Because there is a good deal of scatter in the measurements at smaller fluxes, an analysis of the variation of one of the elastic constants (c') was carried out for two different smoothed curves of the bulk changes of the frequency of the c' mode. No significant change (less than 1%) in the final result for the local change in c' was found. Further analysis of the data and calculation revealed that the local flux dependence of all the quantities—in particular the slope of the initial region—is determined primarily

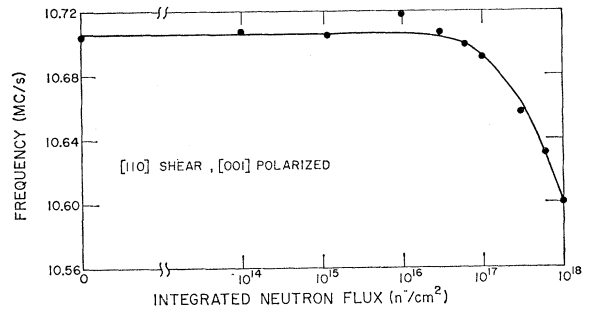

FIG. 4. Variation of the resonant frequency corresponding to the elastic constant C_{44}, as a function of integrated neutron flux on the surface of the 12-mm LiF sample.

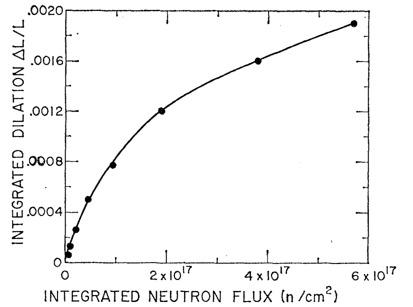

FIG. 5. Total expansion of a 9.6-mm LiF cube as a function of external neutron flux.

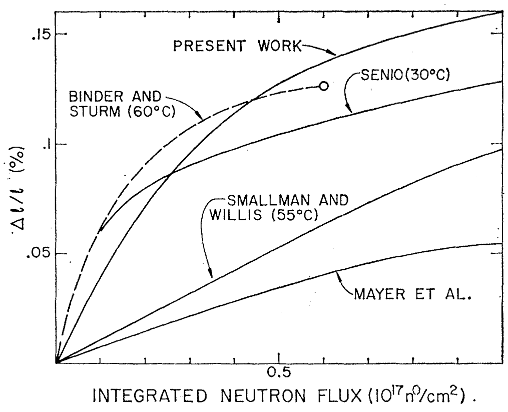

FIG. 6. Local expansion of LiF as a function of local integrated neutron flux, as determined by a number of different experiments.

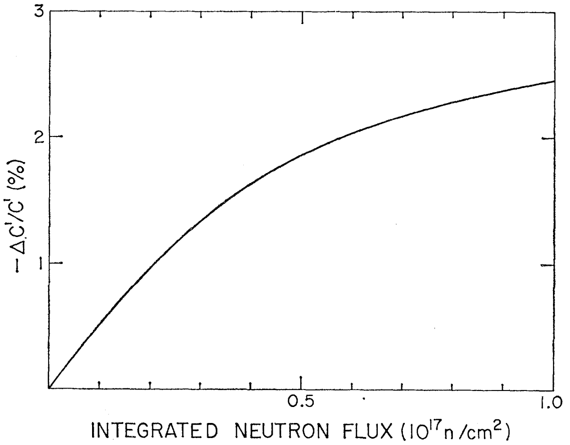

FIG. 7. Local variation of the elastic constant C' as a function of local integrated neutron flux.

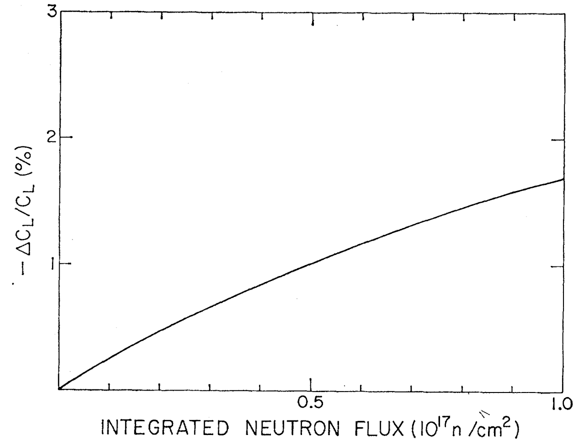

FIG. 8. Local variation of the elastic constant C_L as a function of local integrated neutron flux.

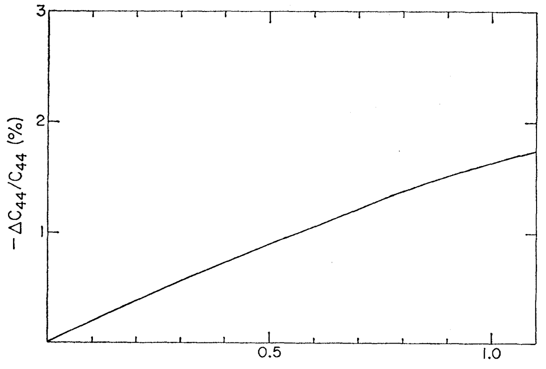

FIG. 9. Local variation of the elastic constant C_{44} as a function of local integrated neutron flux.

by the shape of the bulk measurements in the high-flux region, where the relative error in the measurements is least. It is felt that the final results shown in Figs. 6–9 are reliable to within 20–30%, the lower-flux values being the most reliable portion of the results.

Also included in Fig. 6 are several other measurements of the expansion produced by slow neutron bombardment of LiF.[11–15,18] The dashed portion of the Binder and Sturm curve represents an interpolation between the linear curve obtained by them at very low fluxes[18] and the single point at $6\times10^{16}n/\text{cm}^2$ obtained in a later work.[11] There is a large amount of variation between the results of the various experiments, and this has been interpreted[19] to be due in part to varying temperatures of the sample during irradiation. This does not prove to be entirely satisfactory. For example, the result obtained by Senio at 30°C lies between those obtained by Binder and Sturm at 60°C and by Smallman and Willis at 55°C. The earlier measurements of defect-produced expansion are of either x-ray lattice-parameter change[11–15] or local strain[18] so the rather large flux correction used in the present work was not necessary in those cases. The low-flux Binder and Sturm measurements[18] exhibited a very linear flux dependence, while other measurements,[11–15] at higher fluxes, are all characterized by a leveling off with flux, and, in some cases, pass through a maximum at high fluxes.[12,13] The present results are consistent with this trend, being linear at fluxes less than $10^{16}n/\text{cm}^2$ and leveling off at higher fluxes. They are taken over just the range where the transition from linear flux dependence to the leveling off takes place. The slope of the variation in the linear region is 0.43% per $10^{17}n/\text{cm}^2$, or about one-half that found by Binder and Sturm.[18]

This same general behavior is found for all the elastic constants, as shown in Figs. 7–9. The c' variation, which is the largest of the three, is very similar to the linear expansion. The other two constants, which have a smaller variation, show much less tendency to level off. The difference between the variation in c and c_L is almost within the limit of error in the present work, but the shape of the c' curve is definitely different from c_L and c. The slopes of the linear regions of these three elastic-constant variations are -5.3%, -2.4%, and -1.9% per $10^{17}n/\text{cm}^2$ for c', c_L, and c, respectively.

IV. DISCUSSION

The length-change measurements made in the present work can be compared with previously existing data. The values found, shown in Fig. 6, are of the same order of magnitude as previous values, but differ by amounts which are typical of the differences already existing between different investigators. This difference has already been commented upon in the previous section. For fluxes $\phi>5\times10^{16}n/\text{cm}^2$, the present values are higher than previous values while the results of Binder and Sturm and Senio are larger for low flux. The results show that there is a linear region for $\phi<10^{16}n/\text{cm}^2$, followed by an approach to saturation for $\phi>10^{17}n/\text{cm}^2$.

There exist no other data with which to compare the elastic-constant changes in LiF. Indeed, this is the only complete set of all three elastic-constant changes with irradiation for any material. These data again show a linear region for low flux followed by a less than linear rise for high fluxes. The elastic constants decrease with flux in all cases. The fact that the changes are of the same order of magnitude for different elastic constants means that one could not account for large discrepancies in measurements, such as have been found in copper, made by different techniques as measurements of different linear combinations of elastic constants. It should also be noted that the magnitude of the elastic-constant changes (particularly, the ratio of the elastic-constant change to the volume change which will be discussed further later) is much smaller than that found for copper by König *et al.* and by Dieckamp and Sosen.

The nonlinearity of the curves for dilatation and elastic constants with flux implies that the concentration of defects increases less than linearly with flux or the property measured increases less than linearly. Evidence for the latter case is given by the fact that the different curves do not saturate at the same value of flux. As mentioned earlier, previous optical measurements also provide evidence that the nature or dispersal of the damage changes with dose.

In what follows, we quantitatively consider further only the low-flux region where the curves are linear. For this region, we take as a working assumption that the defects are isolated so that the slopes of the curves represent a property per defect. Further, relying on the measurements of Binder and Sturm[11,15] showing that lattice parameter and length change are equal for the damage-production-rate curve at low fluxes and also during annealing, we suppose that interstitials and vacancies are present in equal numbers as Frenkel pairs.

The results for the elastic-constant changes with flux are tabulated in the second line of Table I.[20–26] Temperature and pressure coefficients are also listed in Table I, which will be used in the further discussion. The changes in c' are the largest, and in c the smallest. In Table II are listed the ratios of the change in the bulk modulus to the change in volume produced by point defects, temperature, and pressure. The first line of Table II is computed from the initial slopes of the curves in Figs. 6–9. The pressure coefficients are taken

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TABLE I. Elastic constants of LiF and their changes with flux, temperature, and pressure.

	C'	C_L	C	Reference
$C\alpha\beta\,(10^{11}\text{ dyn/cm}^2)$	3.46	13.94	6.28	Reference 20 and 21
$(1/C\alpha\beta)(dC\alpha\beta/d\phi)[10^{-19}(n/\text{cm}^2)]$	-5.3	-2.4	-1.9	Present work
$(1/C\alpha\beta)(dC\alpha\beta/dT)\,(10^{-4}\text{ deg}^{-1})$	-10.1	-7.0	-3.3	Leibfried and Ludwig[a] analysis of data of Hassuhl,[b] Briscoe and Squires,[c] and Susse[d] for high-temperature linear region
$(1/C\alpha\beta)(dC\alpha\beta/dT)\,(10^{-4}\text{ deg}^{-1})$	-10.7	-3.9	-2.8	Hassuhl,[b] Briscoe and Squires's[c] data at room temperature
$(1/C\alpha\beta)(dC\alpha\beta/dp)\,(10^{-12}\text{ dyn/cm}^2)$	-10.0	-5.6	-2.2	Barsch and Chang[e] isothermal correction of data by Miller and Smith[f]

[a] Reference 22.
[b] Reference 23.
[c] References 20 and 21.
[d] Reference 24.
[e] Reference 25.
[f] Reference 26.

from the compilation of Barsch and Chang.[25] The temperature coefficients of the elastic constants are further discussed later. The temperature coefficient of the volume, $d\ln V/dT$ (the volumetric thermal expansion), is a value estimated for the Debye temperature (1.5×10^{-4} deg^{-1}) using the empirical formula given by Barsch and Chang.[25] The coefficient $(d\ln B/dp)/(d\ln V/dp)$ is simply dB/dp, also given and corrected to its isothermal value by Barsch and Chang. The theoretical values of $d\ln B/d\ln V$ for the sphere-in-hole model are described later in this section. The results in both tables are later compared with various available theoretical estimates.

Since all of the elastic constants decrease with flux, the increase of elastic constants with concentration of Frenkel pairs expected on the basis of Dienes' considerations is not found. Also, the linear elastic considerations of Nabarro predict the wrong sign. Further, the linear elastic calculation of Melngailis, which supports the large value of $(\Delta B/B)/(\Delta V/V)=-100$ found by König *et al.* and Dieckamp and Sosin, is much too large for the experimental value of $(\Delta B/B)/(\Delta V/V)=-1.8$ found here. The latter number is an experimental ratio of measurements made on the same specimen, and is independent of any assumptions about the number of Frenkel defects per unit flux. If the volume change per Frenkel pair is supposed to lie between one and two atomic volumes, then the bulk-modulus change found here is a decrease of only 2–4% per percent of Frenkel pairs instead of 140% found by König *et al.* and Dieckamp and Sosin and the upper limit of 1% by Thompson *et al.* for copper.

TABLE II. Ratio of bulk modulus to volume changes in LiF for changes induced by point defects, temperature, and pressure.

$\dfrac{d\ln B/d\phi}{d\ln V/d\phi}$	-1.8	Experimental present measurements
$\dfrac{d\ln B/dT}{d\ln V/dT}$	-6.5	Experimental
$\dfrac{d\ln B/dp}{d\ln V/dp}$	-5.2	Experimental
$\dfrac{d\ln B}{d\ln V}$		Calculated for sphere-in-hold model
	-1.0	Nonlinear isotropic model
	-3.3	Nonlinear anisotropic model

If we suppose, with Zener, that the effects of the point defects will be like that of thermal waves, then we should expect to find similar ratios for the elastic-constant changes with flux and temperature. In Table I, the temperature coefficients of the elastic constants are listed. Two sets are shown. One set (line 4) corresponds to room temperature and the other set (line 3) corresponds to the high-temperature limit (linear region) of the elastic-constant changes with temperature. There is a qualitative correspondence between the flux and temperature coefficients. The order is correct; c' changes the fastest and c the slowest in both cases. However, a quantitative correspondence is lacking. The room-temperature coefficients correspond somewhat more closely to the flux coefficients but one should expect the high-temperature linear-temperature data to be of more physical significance in representing anharmonic effects. The temperature coefficient of the ratio of the bulk-modulus change to the volume change is also not in good quantitative agreement (-1.8 for Frenkel-pair–induced changes versus -6.5 for thermally induced changes).

As the damage in the specimen is not uniform and the specimen is consequently internally stressed by the inhomogeneous stress field, one might obtain some volume and elastic-constant changes from this source. In a simple model, we suppose that the effect of the more heavily damaged skin of the specimen exerts approximately a hydrostatic stress on the bulk of the specimen. In this case, one would expect the measured quantities to correlate with the corresponding pressure-induced changes. These pressure coefficients are also listed in Tables I and II and again one finds that, although a qualitative correspondence is found, a quantitative correspondence is lacking.

Another comparison of the experimental results can be made with a nonlinear elastic calculation of the bulk-modulus–to–volume-change ratio for a sphere-in-hole model of a point defect. This calculation[27] predicts in

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[27] J. Holder and A. V. Granato, Phys. Rev. (to be published).

the isotropic, nonlinear elastic model,

$$\frac{\Delta B/B}{\Delta V/V}=\frac{3B(GB'-BG')[3B+4G-B(B'G-BG')]}{(3B+4G)[(3B+4G)(BG'-G)+4G(GB'-BG')]}, \tag{4}$$

where G is an average shear elastic constant and the primes denote differentiation with respect to pressure. In the anisotropic nonlinear model, the calculation gives[27]

$$\frac{\Delta B/B}{\Delta V/V}=\frac{\{B'+S'(S'-1)-S''\}}{(S'-1)}, \tag{5}$$

where

$$S'=\tfrac{1}{2}B\left(9\frac{\partial \ln C}{\partial p}+2\frac{\partial \ln(2B+0.667C')}{\partial p}+\frac{1}{2}\frac{\partial \ln C'}{\partial p}-\frac{\partial \ln(3B+4C')}{\partial p}-\frac{\partial \ln(3B+C'+3C)}{\partial p}\right)$$

$$S''=\tfrac{1}{2}B^2\left(\left(81\frac{\partial \ln C}{\partial p}\right)^2+2\left[\frac{\partial \ln(2B+0.667C')}{\partial p}\right]^2+\tfrac{1}{4}\left(\frac{\partial \ln C'}{\partial p}\right)^2-\left[\frac{\partial \ln(3B+4C')}{\partial p}\right]^2-\left[\frac{\partial \ln(3B+C'+3C)}{\partial p}\right]^2\right).$$

For LiF, using the tabulated pressure derivatives of Barsch and Chang, the predicted values are $(\Delta B/B)/(\Delta V/V)=-1.0$ and -3.3 for the isotropic and anisotropic calculations, respectively, compared with the experimental value of -1.8. Also, the isotropic model predicts -0.03 for the ratio in copper, in agreement with the results of Thompson *et al.* but in gross disagreement with the results of König *et al.* and of Dieckamp and Sosin. In summary, the nonlinear elastic-constant calculations disagree the least, the expectations from Zener's considerations come next, and the rest of the theoretical estimates differ in order of magnitude and even in sign with the experimental results.

V. SUMMARY

(1) The elastic constants of LiF all decrease with neutron flux. This is contrary to expectations based on considerations of Dienes and of Nabarro and, consequently, makes measurements of elastic constants less useful as a tool for distinguishing between interstitials and vacancies than had previously been expected.

(2) The elastic constants and dilation are linear with neutron flux up to about $10^{16}n/\text{cm}^2$ and then tend to saturation values above $10^{17}n/\text{cm}^2$. The ratios of elastic-constants change with flux at higher doses indicating that the nature or dispersal of the damage changes with dose.

(3) All of the elastic-constant changes as well as the volume changes were measured. As the changes are all of the same order of magnitude, one would not expect different measurements by different observers of different linear combinations of the elastic constants to be different in order of magnitude.

(4) The measured value of the bulk-modulus change to volume change in LiF is almost two orders of magnitude smaller than that found in copper by König *et al.* and by Dieckamp and Sosin, but is not inconsistent with the value for copper found by Thompson *et al.*

(5) A comparison of the experimental results with various available theories shows that a nonlinear elastic sphere-in-hole calculation disagrees the least, the expectations from Zener's considerations come next, and the rest of the theoretical estimates differ in order of magnitude (Melngailis) and even in sign (Dienes and Nabarro) with the experimental results.

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