

## Elastic Versus Inelastic Energy Loss of Recoil Germanium and Silicon Atoms\*

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Experiments of Wikner and van Lint have established the photoneutron method of producing displacement radiation effects by measuring the effect of recoils from  $\text{Si}(\gamma, n)$  reactions on the excess carrier lifetime. These experiments have been extended to Ge together with an additional experiment in which Si and Ge were bombarded by a 1-MeV reactor neutron spectrum and a mono-energetic beam of 14-MeV neutrons. The range of average energies of the recoil atoms from the bremsstrahlung experiment spans the range of the recoil atom energies of the neutron experiment, so that the results can be compared with theoretical calculations which assume varying percentages of elastic and inelastic energy loss of the recoils. The results are in good agreement with the theory given by Lindhard *et al.*, which is based on a broad transition-energy region between elastic and inelastic energy losses.

### I. INTRODUCTION

MEASUREMENTS of the displacement radiation damage induced in Ge by high-energy (15–30 MeV) bremsstrahlung have been made. This experiment, which is identical to the one reported by Wikner and van Lint<sup>1</sup> for Si confirms their interpretation of the radiation damage as being due to displaced recoil atoms which receive energy from a  $(\gamma, n)$  reaction. An additional experiment was also conducted in which Si and Ge were bombarded by a spectrum of reactor neutrons ( $\sim 1$ -MeV average energy) and a monoenergetic beam of 14-MeV neutrons. It turns out that the range of average energies of the recoil atoms from the bremsstrahlung experiment spans the range of the recoil atom energies of the neutron experiment, so that two independent methods are available for determining the radiation damage due to these recoils.

The measurement of the excess-carrier lifetime performed in both experiments is a sensitive way of determining the number of radiation-induced recombination centers which result when the recoiling lattice atom displaces other atoms via *elastic* collisions. Theoretically, it is expected that the recoil atoms in Si, which have an energy  $\sim 100$  keV, will dissipate a significant fraction of their energy via *inelastic* encounters in which other atoms are excited or ionized but in which the “knock-on” atom is not displaced. The same considerations show that the recoiling Ge atoms of energies  $\sim 30$  keV will lose almost all of their energy via elastic collisions. Comparisons between the two experiments are made to shed some light on the relative importance of elastic and inelastic scattering in this critical energy region where both processes are important.

### II. BREMSSTRAHLUNG EXPERIMENT

The bremsstrahlung experiment (see Ref. 1 for details) utilized the known bremsstrahlung spectrum<sup>2</sup>

\* Research sponsored by Harry Diamond Laboratories, U. S. Army Materiel Command.

<sup>1</sup> E. G. Wikner and V. A. J. van Lint, Phys. Rev. **133**, A884 (1964).

<sup>2</sup> N. E. Hansen and S. C. Fultz University of California,

generated by high-energy electrons at 15, 18, 21, 24, and 30 MeV striking a platinum converter to bombard  $1 \times 2.5\text{-mm} \times 10\text{-mm}$  As-doped Ge samples.<sup>3</sup> The absolute bremsstrahlung intensity is measured by counting the number of  $\text{Au}^{197}(\gamma, n)\text{Au}^{196}$  transmutations produced in a monitor wire. The number and energy spectrum of the recoil Ge atoms produced by the photoneutron reaction is then calculated by computer, using the assumption that the nuclear processes are described by the statistical evaporation model<sup>4</sup> with a 15% admixture of resonance direct neutron emission.<sup>5</sup> This calculation is the same as that described in Ref. 1 except that an improved value of the level-density parameter  $\alpha$  has been used for germanium and the silicon data of Ref. 1. The values suggested by Thompson<sup>6</sup> and Erba *et al.*<sup>7</sup> are  $\alpha(\text{Si})=4.5$  and  $\alpha(\text{Ge})=12$ . Finally, the amount of displacement damage is calculated for various assumptions in regard to the energy-loss process.

One of the problems in evaluating the germanium recoil spectrum is that the  $\text{Ge}(\gamma, n)$  cross section has not been measured for the natural element. Accordingly, it was necessary to arrive at a suitable cross section from the known values of the Ge threshold energies<sup>8</sup> for each of its five isotopes and from a knowledge of the systematics of photoneutron reactions given by Montalbetti *et al.*<sup>9</sup> Two possible cross sections were used (see Fig. 1). The cross section (case 1) is the best curve given by systematics in which the half-width, the maximum value, the energy at which the maximum occurs, and the integrated value over energy, are all considered. In addition, the low-energy tail is estimated

Lawrence Radiation Laboratory Rep UCRL-6099, 1960 (unpublished).

<sup>3</sup> Commercially grown samples obtained from Sylvania Corporation.

<sup>4</sup> J. Blatt and V. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), pp. 365–374.

<sup>5</sup> D. H. Wilkinson, Physica **22**, 1039 (1956).

<sup>6</sup> D. Thompson, Phys. Rev. **129**, 1649 (1963).

<sup>7</sup> E. Erba, U. Facchini, and E. Saetta Menichelli, Nuovo Cimento **22**, 1237 (1961).

<sup>8</sup> R. Tobin, J. McElhinney, and L. Cohen, Phys. Rev. **110**, 1388 (1958).

<sup>9</sup> R. Montalbetti, L. Katz, and J. Goldemberg, Phys. Rev. **91**, 659 (1953).

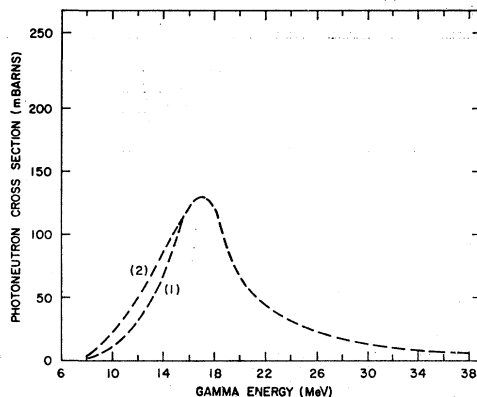


FIG. 1. Two possible  $(\gamma, n)$  cross sections for Ge, the (1) designating the most reasonable from systematics.

from the relative abundances of the isotopes and the measured threshold energies of the  $(\gamma, n)$  reaction. The cross section of case 2 allows for the greatest reasonable deviation from the cross section of case 1 consistent with the threshold data and the extreme skew shapes of a few elements. The main purpose in considering this case is to indicate a maximum error arising from the assumed shape of the cross section. A probable error in the magnitude of the cross section will change the value for the number of recoil atoms but not their distribution of energy. Hence, this error will not affect the relative amounts of energy lost via elastic and inelastic collisions.

The calculated Ge recoil spectra for five different bremsstrahlung energies are presented in Fig. 2. These curves, which are comparable to Fig. 3 of Ref. 1 for Si, are normalized to have an integrated spectrum equal to  $N_d/N_0(\text{Au})$ , where  $N_0(\text{Au})$  is the number of radioactive Au atoms per gram for a given experiment and  $N_d$  is the number of displaced atoms. The value<sup>10</sup> for  $N_d$  as given by Seitz and Koehler<sup>10</sup> is

$$N_d = E/2E_d, \quad (1)$$

where  $E = \int dE_r F(E_r) E_r$ ,  $E_r$  is the total recoil energy,  $F(E_r)$  is the number of recoil atoms per unit energy at energy  $E_r$ , and  $E_d$  is a characteristic displacement energy taken as  $E_d = 30$  eV for Ge.

If the irradiated samples are now subjected to a short pulse of low-intensity gamma photons, the excess-carrier lifetime  $\tau$  of the sample may be measured. From simple considerations of the Shockley-Read<sup>11</sup> theory on the excess-carrier lifetime, it can be shown that the change in the reciprocal lifetime before and after the bremsstrahlung irradiation is proportional to the number of recombination centers  $N$  that have been introduced. If all the recoil atoms underwent elastic collisions, then  $N$  is proportional to  $N_d$  and the ratio  $\Delta(1/\tau)/N_d$  would be constant (case A). If all of the

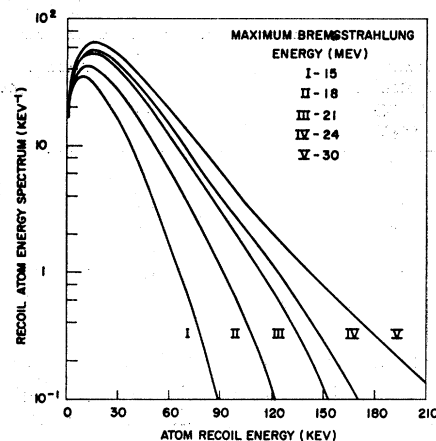


FIG. 2. Calculated germanium recoil spectra for five different bremsstrahlung energies.

recoil atoms initially undergo inelastic collisions, then each recoil will create the same number of displaced atoms as their energy is degraded to the "point" where elastic collisions first occur. In this case (case B),  $N$  is proportional to  $N_r = \int F(E_r) dE_r$ , and  $\Delta(1/\tau)/N_r$  will be constant for the various bremsstrahlung energies.

Physically, both cases represent an extreme situation, so that neither factor need be constant. Lindhard *et al.*<sup>12,13</sup> have examined the problem of how much energy is lost via elastic and inelastic collisions for atoms at various energies. Their theoretical curves, which can be conveniently parameterized for all atom-atom interactions, are the result of numerical calculations based on a continuous representation of successive scattering events. It turns out that both processes are important over a wide range of energies about the critical energy  $E_{LS}$  where the two processes are equal. Lindhard's theory shows that the inelastic losses increase linearly with velocity  $v$  up to velocities  $\leq Z^{2/3}v_0$ , where  $v_0 = e^2/\hbar$  is the velocity of an electron in the hydrogen atom and  $Z$  is the atomic number of the recoil atom. The elastic losses increase from  $v=0$  to some maximum at an energy less than  $E_{LS}$  before gradually decreasing to zero. The value of  $E_{LS} \approx ZA/2$  keV, where  $A$  is the atomic mass of the recoil atoms. This value of the critical energy is of the same order as that given by Seitz and Koehler<sup>10</sup> who first defined the separation between elastic and inelastic processes in terms of the atom's velocity relative to its electron's binding energy.

The fraction of the atom's energy lost in elastic (displacement) events is given in Table I for Si and Ge for their various average recoil energies  $\bar{E}_r$ . It is seen that the Ge atoms are expected to lose almost all of their energy via displacement collisions in the experi-

<sup>10</sup> F. Seitz and J. S. Kohler, in *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 2, p. 305.

<sup>11</sup> W. Shockley and W. T. Read, Jr., *Phys. Rev.* **87**, 835 (1952).

<sup>12</sup> J. Lindhard and M. Scharff, *Phys. Rev.* **124**, 128 (1961).

<sup>13</sup> J. Lindhard, V. Nielsen, *et al.*, *Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd.*, **33**, No. 10 (1963); J. Lindhard, M. Scharff, and H. E. Schiøtt, *ibid.* **33**, No. 14 (1963).

TABLE I. Average recoil energies (keV).

Maximum bremsstrahlung energy (MeV)	Average recoil energy (keV)			Fraction of energy lost in elastic collisions (Lindhard)	
	Si	Ge		Si	Ge
		Case 1	Case 2		
15	...	20.2	18.8	...	0.90
18	68	26.7	24.4	0.53	0.90
21	104	31.0	28.5	0.47	0.89
24	128	33.1	30.5	0.44	0.89
30	153	36.4	33.8	0.43	0.88
38	157	...	...	0.43	...

mental energy range, whereas the Si recoil atoms will lose varying amounts of energy by displacement collision and ionization. A calculation of the number of displaced atoms,  $N_L$ , based on Lindhard's theory can be readily made if, as assumed in Eq. (1), the total number of displacements is taken proportional to the elastic energy transfer only. For this case (case C) the ratio  $\Delta(1/\tau)/N_L$  should be constant.

### III. NEUTRON EXPERIMENT

A number of experiments have been performed on the change of minority-carrier lifetime of Ge and Si samples exposed to neutrons from two different sources. Experiments with 14-MeV neutrons have been done at the Cockcroft-Walton neutron generator at General Atomic. The total flux was measured by placing Cu foils next to the sample and measuring the characteristic decay of  $\text{Cu}^{64}$  from the  $\text{Cu}^{65}(n,2n)\text{Cu}^{64}$  reaction. A 19% fraction of  $\text{Cu}^{64}$  decays via positron emission, which results in a countable number of 0.51-MeV annihilation gamma photons. The flux determinations by this method were then compared to the results of a  $\text{BF}_3$  neutron counter located at various distances from the sample.

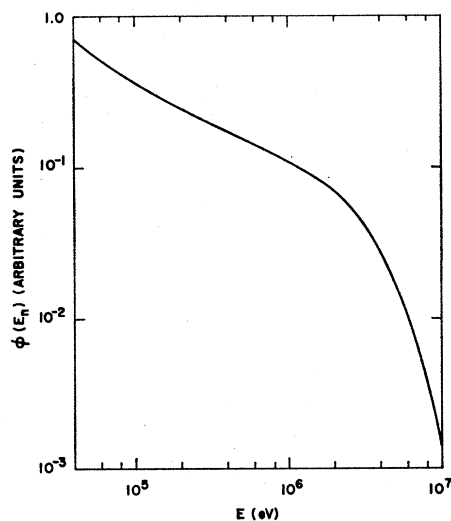


FIG. 3. Fast neutron spectrum behind 3 in. of water from the core of the General Atomic Triga reactor.

The 1-MeV neutron experiments were conducted at the General Atomic Triga reactor at a point behind 3 in. of water from the reactor core. The Triga spectrum,  $\varphi(E_n)$ , of average energy near 1 MeV is shown in Fig. 3. This spectrum was obtained by using a modified DSN method<sup>14</sup> to solve the neutron transport equation using an IBM 7090. It was then compared with experimental measurements using standard fast-neutron activation dosimeters. The calculated and experimental flux determinations agreed well with one another. The total flux for these experiments is given by a measurement of the  $\text{Au}^{197}(n,\gamma)\text{Au}^{198}$  reaction. Two gold wires, one cadmium-shielded, were placed near the sample, giving a measure of the number of thermal neutrons. The fast-neutron flux was then determined from this thermal flux measurement by use of the calculated Triga spectrum.

An exact calculation of the recoil atom spectrum resulting from neutron interactions is not possible because of the lack of experimental data for the elastic, inelastic, and reaction cross sections. However, for 1-MeV neutrons, the large majority of interactions result in elastic scatterings, for which an average recoil energy can be found. For 14-MeV neutrons, about one-half of the interactions are elastic and the other half are inelastic with a small percentage of other reactions. An inelastic reaction is defined as one in

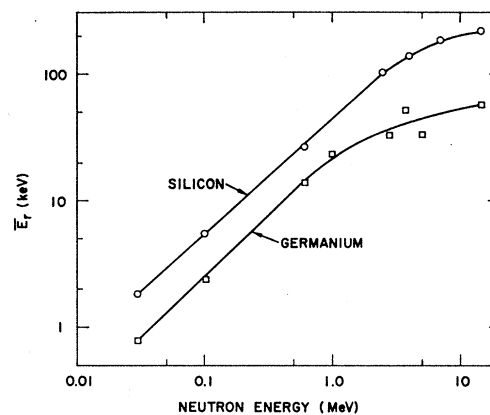


FIG. 4. Average energy of germanium and silicon recoil atoms as a function of neutron energy.

<sup>14</sup> Los Alamos Report, LAMS-2346, October, 1959 (unpublished).

which an incident neutron is absorbed by the nucleus, which then emits a neutron of lesser energy and some gamma radiation. Because the secondary neutron has less energy than the secondary from an elastic process, the maximum energy transfer to the recoil is less for the inelastic process. However, the model of the compound nucleus which applies to the inelastic reaction and some general experimental data<sup>15</sup> indicate that the neutron is emitted isotropically. Hence, a much larger percentage of the maximum transfer energy will be given to the recoil atom. These two considerations tend to cancel, so that the inelastic recoil atoms have been assumed to have roughly the same energy as the elastic recoils.

The average energy of the recoil atoms in a small sample is thus calculated for pure elastic-neutron interactions by the formula

$$\bar{E}_r = [4A/(A+1)^2] \bar{f} E_n, \quad (2)$$

where  $E_n$  is the neutron energy,  $\bar{f} = \frac{1}{2}(1 - \langle \cos\theta \rangle_{av})$  and  $\langle \cos\theta \rangle_{av}$  is the average value of the cosine of the scattering angle as measured in the center-of-mass system. For isotropic scattering,  $\bar{f} = \frac{1}{2}$ . The values for  $\bar{E}_r$  are given in Fig. 4. It is seen that the preference for forward scattering at higher neutron energies leads to a slower rate of increase in the recoil atom energy at  $E_n < 14$  MeV. The value of the average recoil energy for the 14-MeV neutrons is taken from Fig. 4 and the value of  $\langle E_r \rangle = \int \bar{E}_r \varphi(E_n) dE_n / \int \varphi(E_n) dE_n$  is calculated for the reactor spectrum and presented in Table II. The fraction of energy lost by the recoils in elastic collisions is then calculated from Lindhard's theory for the average recoil energy  $\langle E_r \rangle$  and included in this table.

The computed quantities corresponding to cases A, B, and C, respectively, are then given by

$$N_d = k \int \sigma_t \bar{E}_r(E_n) \varphi(E_n) dE_n / \int \varphi(E_n) dE_n, \quad (4)$$

where  $k$  is an arbitrary constant and  $\sigma_t$  is the total

TABLE II. Average silicon and germanium recoil energy resulting from 1- and 14-MeV neutron irradiation and the corresponding fraction of energy lost in elastic collisions by the recoil atoms.

Neutron energy (MeV)	Average recoil energy (keV)		Fraction of energy lost in elastic collisions (Lindhard)	
	Silicon	Germanium	Silicon	Germanium
1	68	25	0.53	0.90
14	220	62	0.35	0.86

<sup>15</sup> M. D. Goldberg, V. M. May, and J. R. Stehn, Brookhaven National Laboratory Report No. BNL-400, 2nd ed., 1962 (unpublished).

TABLE III. Lifetime data for Ge from bremsstrahlung experiment.

Maximum bremsstrahlung energy (MeV)	$\Delta(1/\tau)/N_d$ ( $\times 10^{-6}$ ) (cm <sup>2</sup> /sec)	Ge	
		$N_d/N_0$ ( $\times 10^{-3}$ ) Case 1	Case 2
15	0.55	1.04	1.58
18	0.78	1.65	2.09
21	0.97	2.30	2.70
24	1.19	2.58	2.93
30	1.50	3.02	3.33

neutron cross section,

$$N_r = \int \sigma_t \varphi(E_n) dE_n / \int \varphi(E_n) dE_n \quad (5)$$

and

$$N_L = g_L(\langle E_r \rangle) N_d, \quad (6)$$

where  $g_L(\langle E_r \rangle)$  is the fraction of energy lost in elastic collisions for an average recoil energy  $\langle E_r \rangle$ .

#### IV. EXPERIMENTAL RESULTS AND COMPARISON

The experimental averages of five determinations of the lifetime data for Ge at each bremsstrahlung energy are given in Table III. These data are then expressed in terms of the ratios  $\Delta(1/\tau)/N_d$ ,  $\Delta(1/\tau)/N_r$ , and  $\Delta(1/\tau)/N_L$  as shown in Fig. 5. The constancy of one or more of these ratios is expected according to whether the recoil atoms conform with the assumptions of case A, case B, or case C, respectively. Each ratio is calculated on the basis of the most reasonable Ge( $\gamma, n$ ) cross section of case 1 and the extreme cross section selected for case 2. The corresponding calculations for Si are shown in Fig. 6 for a similar energy range for which the measured value of the Si( $\gamma, n$ ) cross section<sup>16</sup> is assumed to be reliable.

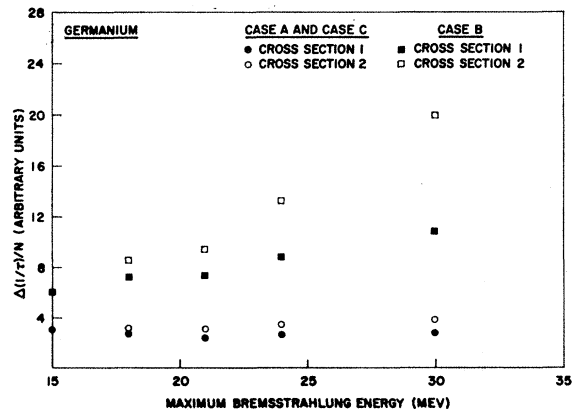


FIG. 5. The values of  $\Delta(1/\tau)/N$  versus maximum bremsstrahlung energy for germanium. The values for each cross section are normalized to be equal at a bremsstrahlung energy of 15 MeV.

<sup>16</sup> L. Katz, R. N. Haslan, *et al.*, Can. J. Phys. 32, 580 (1954).

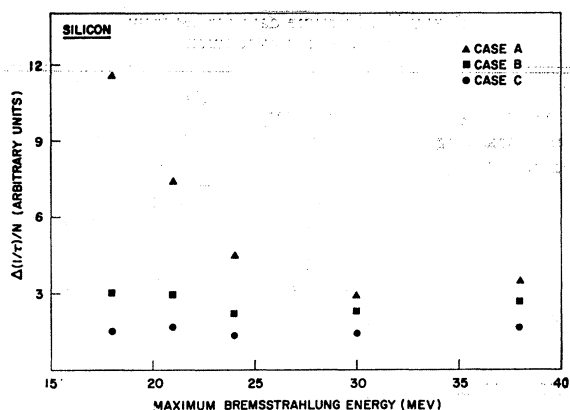


FIG. 6. The value of  $\Delta(1/\tau)/N$  versus maximum bremsstrahlung energy for silicon. The values of  $N$  given in Ref. 1 have been corrected by using the improved value for the level-density parameter of  $\alpha=4.5$ .

The results of the neutron experiment are shown in Table IV. Each measurement of the lifetime change is the average of five determinations with an estimated error of  $\pm 15\%$ . The values of  $\Delta(1/\tau)/N$  for the cases A, B, and C have been calculated as described previously and are expressed in arbitrary units.

A comparison of the results for Ge shows quite clearly that the displaced recoil atoms undergo elastic collisions. Either all of the collisions are elastic, or a constant 90% of them (refer to Table I) are elastic, as predicted by the theory of Lindhard *et al.* The fact that Lindhard's predicted fraction of energy lost to elastic collisions is so nearly constant for the extreme values of the Ge recoil energies makes it impossible to distinguish his model from that of Seitz and Koehler<sup>10</sup> who predict total elastic-energy loss below their critical energy  $E_{SK}(\text{Ge})=90$  keV.

The results for Si demonstrate an appreciable

TABLE IV. Lifetime data for Ge and Si from neutron experiment.

Neutron energy (MeV)	$\Delta(1/\tau)/\Delta\phi$ (Neutron-sec/cm <sup>2</sup> ) <sup>-1</sup>		$\Delta(1/\tau)/N_d$		$\Delta(1/\tau)/N_r$		$\Delta(1/\tau)/N_L$	
	Si	Ge	Si	Ge	Si	Ge	Si	Ge
~1	$13 \times 10^{-7}$	$3.8 \times 10^{-8}$	6.7	3.8	3.6	0.70	13	4.2
14	$28 \times 10^{-7}$	$7.4 \times 10^{-8}$	6.3	3.5	14	2.0	17.5	4.1

amount of recoil-energy loss via ionization. The data of the bremsstrahlung experiment specifically exclude a model in which all of the displacements are due to elastic collisions (proportional to  $N_d$ ), even if considerable allowance is made for error in the  $\text{Si}(\gamma, n)$  cross section at the lower gamma energies ( $\leq 18$  MeV), for it is seen that cases B or C are clearly more constant than case A. In considering the neutron data, we see that the pure ionization model of case B is inadequate in estimating the energy-loss mechanism for silicon atoms in the transition region, although case B is a close approximation to the Seitz-Koehler model for the number of displaced atoms arising from the high-energy silicon recoils. However, the data of both the neutron and bremsstrahlung experiments are in good agreement with Lindhard's theory. These results, which required considerable interpretation (e.g., a statistical model of the nucleus, assumptions about the recoil spectrum from neutron collisions, etc.), cannot be said to specifically confirm the Lindhard model. However, they show that his calculations of a broad transition energy region provide the best available fit with the data and that the critical-energy parameter  $E_{LS}$  is a useful measure of the dominance of elastic and inelastic processes in Ge and Si.

#### ACKNOWLEDGMENT

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