

Wir führen folgende Bezeichnungen ein:

$$\begin{aligned} \kappa_0(-) &= \kappa_0^{(1)}(\cdot), \\ \kappa_1(-) &= [1/\varrho(t)] \sum_{k=1}^3 (\bar{F}^1)_{1k}(t) \kappa_{2,k1}^{(1)}(\cdot) \dots \\ &\quad \text{usw. bis} \\ \kappa_9(-) &= [1/\varrho(t)] \sum_{k=1}^3 (\bar{F}^1)_{3k}(t) \kappa_{2,k3}^{(1)}(\cdot), \quad (\text{A.6}) \\ \kappa_{10}(-) &= [1/\varrho(t)] \kappa_{1,1}^{(1)}(\cdot) \dots \kappa_{12}(-) \\ &= [1/\varrho(t)] \kappa_{1,3}^{(1)}(\cdot); \\ \lambda_0(\dots) &= \lambda_0^{(1)}(|), \\ \lambda_1(\dots) &= (1/\varrho) \sum_{k=1}^3 (\bar{F}^1)_{1k} \lambda_{2,k1}^{(1)}(|) \dots \text{usw. bis} \\ \lambda_9(\dots) &= (1/\varrho) \sum_{k=1}^3 (\bar{F}^1)_{3k} \lambda_{2,k2}^{(1)}(|), \quad (\text{A.7}) \\ \lambda_{10}(\dots) &= (1/\varrho) \lambda_{1,1}^{(1)}(|) \dots \lambda_{12}(\dots) \\ &= (1/\varrho) \lambda_{1,3}^{(1)}(|). \end{aligned}$$

**Satz 3:** Wir setzen in Satz II  $n=12$ , für  $x$  (A.3), für  $\kappa_0 \dots \kappa_{12}$  (A.6) mit  $r=1$ ,  $(-)= (x(t), x(t'), \dot{x}(t'); t-t')$  und  $(\cdot)$  nach (24), für  $\lambda_0 \dots \lambda_{12}$  (A.7) mit  $(\dots) = (x(t), \dot{x}(t))$  und  $(|)$  nach (18a). Dann folgt aus Satz II mit (A.6), daß die Funktionen  $\kappa_\alpha^{(1)}$  ( $\alpha=0, 1, 2$ ) überall verschwinden müssen. Daher gilt a fortiori

$$\int_{-\infty}^t \kappa_\alpha^{(1)}(\cdot) dt' = 0$$

für alle  $t$  und alle Vorgeschichten  $(\cdot)$  (24) mit  $r=1$ ; q. e. d.

## The Diffusion of $^3\text{He}$ and $^4\text{He}$ in LiF

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Helium isotopes were produced in single crystals of LiF by means of the nuclear reaction  $^6\text{Li}(n, ^3\text{H})^4\text{He}$  and by the subsequent decay of  $^3\text{H}$  into  $^3\text{He}$ . At a constant temperature of 620 °C the isotopic ratios of the outgassed helium isotopes were measured as a function of time respectively as a function of the released fraction. The ratio of the diffusion constants  $D_3/D_4$  was found to be at most 1.02 which is considerably smaller than the maximum classical value of 1.155.

Information on atomic jump mechanisms in solids can be obtained from the isotope effect in diffusion. Deviations from the simple  $m^{1/2}$ -law are frequently observed. According to theory<sup>1-3</sup>, in interstitial diffusion no correlation of subsequent jumps exists, as it usually does in vacancy type processes. In this case the measured effect is directly related to the kinetic energy factor  $\Delta K$ , the magnitude of which tells to what extent the diffusive motion of the jumping atom is accompanied by lattice relaxations. If

this factor is close to unity, then the diffusing particle is essentially decoupled in its motion from the lattice and the  $m^{1/2}$ -relation would be obeyed by classical systems.

A number of experiments concerning interstitial solute diffusion are known (see Ref. <sup>4-12</sup>). The mass effect was found to be nearly equal to the maximum classical value for the diffusion of interstitials such as  $\text{Li}^+$  in metals and semiconductors and of rare gases in quartz. For hydrogen metal systems the

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<sup>1</sup> G. H. VINEYARD, J. Phys. Chem. Solids **3**, 121 [1957].

<sup>2</sup> I. G. MULLEN, Phys. Rev. **121**, 1649 [1961].

<sup>3</sup> A. D. LECLAIRE, Phil. Mag. **14**, 1271 [1966].

<sup>4</sup> E. M. PELL, Phys. Rev. **119**, 1014 [1960].

<sup>5</sup> A. J. BOSMAN, P. E. BROMMER, and G. W. RATHENAU, J. Phys. Radium **20**, 241 [1959].

<sup>6</sup> W. JOST and A. WIDMANN, Z. Phys. Chem. **45 B**, 285 [1940].

<sup>7</sup> G. M. MCCracken and H. M. LOVE, Phys. Rev. Letters **5**, 201 [1960].

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<sup>9</sup> W. M. JAMES, J. Amer. Chem. Soc. **75**, 3093 [1955].

<sup>10</sup> R. C. FRANK, W. L. LEE, and R. L. WILLIAMS, J. Appl. Phys. **29**, 898 [1958].

<sup>11</sup> W. EICHENAUER, W. LÖSER, and H. WITTE, Z. Metallk. **56**, 287 [1965].

<sup>12</sup> Y. EBISUZAKI, W. J. KASS, and M. O'KEEFE, J. Chem. Phys. **46**, 1373 [1966]; **48**, 1867 [1968]; **49**, 3329 [1968]; and Phil. Mag. **15**, 1071 [1967].



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mass effect showed a temperature dependence and ranged from values close to 1.4 down to values  $<1.0$ . It was shown that quantum mechanical effects caused this behaviour<sup>3, 12</sup>.

So far, the mass effect has not been studied for interstitially diffusing impurities in ionic crystals. The reason seems to be simply that vacancy type diffusion mechanisms dominate for charged impurities in most ionic systems such as the alkali halides. The situation is different for neutral impurities.

There is experimental and theoretical support for the assumption that rare gases diffuse interstitially in alkali halides<sup>13-17</sup>. Depending on the particular system, on temperature and other parameters, the rare gases also undergo trapping and detrapping processes which give rise to more or less complicated temperature patterns<sup>18-20</sup>. These traps are presumably defects such as single vacancies, vacancy pairs and clusters, which are present either due to intentional or unintentional doping or are introduced simultaneously with the rare gas, e. g. by neutron irradiation. By this latter technique typical rare gas concentrations in the range of about  $10^{13}$ – $10^{17}$  atoms/cm<sup>3</sup> are obtained. With increasing concentration, and damage, decreasing mobilities were observed. Different rare gases, introduced simultaneously into the same specimen, were found to diffuse independently<sup>16, 20a</sup>.

Rare gas concentrations of  $10^{19}$  atoms/cm<sup>3</sup> and more, localized in a surface layer several 100 Å thick, are obtained by ion bombardment techniques<sup>21-26</sup>. The results show some similarities with neutron activated samples. Interstitial diffusion and/or bubble migration is thought to be operative.

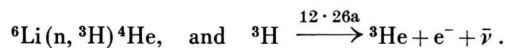
It seemed worthwhile to perform a mass effect experiment on one of the rare gas/alkali halide systems in order to learn more about the diffusion process. For experimental reasons the He/LiF system

is most suited for this purpose. The relative mass difference of  $^3\text{He}$  and  $^4\text{He}$  is high, and reasonable amounts of isotopes can be introduced. The He/LiF system is characterized by a single activation energy of about 1.8 eV in the temperature range above 400 °C<sup>13</sup>. Similar results were obtained for other rare gas/alkali fluoride systems.

Considerable interest in the isotope effect of rare gas diffusion also exists in the field of cosmochemistry. Anomalous isotopic compositions of helium and neon were observed in meteorites<sup>27</sup>. It is thought that in some early stage of meteorite history a diffusion process caused this anomaly.

## Experimental

Single crystals of LiF were irradiated in the thermal column of the Brookhaven National Laboratory graphite reactor with a total dose of about  $2 \times 10^{17}$  n/cm<sup>2</sup> in May 1963. The samples were small cubes with edge lengths ranging from 1 to 3 mm. The helium isotopes were introduced by the nuclear reactions



Concentrations of approximately  $10^{-2}$  cm<sup>3</sup> (STP)  $^4\text{He/g}(\text{LiF})$  were measured. If mass spectrometers of high sensitivity and low background on the masses 3 and 4 are used, enough  $^3\text{He}$  is obtained one year after the neutron irradiation. The results reported here were obtained with samples four years old.

A very important point is the concentration profile of the rare gases introduced. The ratio of the concentrations of  $^3\text{He}$  and  $^4\text{He}$  should be very constant all over the bulk of the sample because both  $^3\text{He}$  and  $^4\text{He}$  are produced by the same nuclear event. Also, the recoil ranges are small (in the order of 10 μm). Only a surface layer of this size has a disturbed distribution. It was chemically removed in all experiments.

Due to the high cross section of the nuclear reaction ( $\sigma_{n,\alpha} = 950$  barn for  $^6\text{Li}$  with thermal neutrons) the neutron flux is considerably reduced for samples of several mm thickness. Therefore a concentration gra-

<sup>13</sup> S. KALBITZER, Z. Naturforsch. **17 a**, 1071 [1962].

<sup>14</sup> H. P. MUNDT and A. K. H. RICHTER, Z. Naturforsch. **20 a**, 267 [1965].

<sup>15</sup> M. I. NORGETT and A. B. LIDIARD, Phil. Mag. **18**, 1193 [1968].

<sup>16</sup> F. W. FELIX, Phys. Status Solidi **27**, 529 [1968].

<sup>17</sup> H. GAUS, Z. Naturforsch. **20 a**, 1298 [1965]; **23 a**, 985 [1968].

<sup>18</sup> F. FELIX, P. SCHMELING, and K. E. ZIMEN, EUR **259.d** [1963].

<sup>19</sup> A. K. H. RICHTER and K. E. ZIMEN, Z. Naturforsch. **20 a**, 666 [1965].

<sup>20</sup> P. SCHMELING, Phys. Status Solidi **11**, 175 [1965]; J. Phys. Chem. Solids **28**, 1185 [1967].

<sup>20a</sup> Similar results were obtained by one of the authors (S. K.) for heavily irradiated specimens of KBr and KI. The following activation energies and preexponential factors were obtained:

<sup>39</sup>A, <sup>82</sup>Kr/KBr: 34.7, 37.1 kcal/mol;  $2 \times 10^2$ ,  $3 \times 10^2$  cm<sup>2</sup>/sec;

<sup>39</sup>A, <sup>128</sup>Xe/KI: 34.9, 40.3 kcal/mol;  $6 \times 10^3$ ,  $3 \times 10^3$  cm<sup>2</sup>/sec.

<sup>21</sup> R. KELLY, C. JECH, and H. J. MATZKE, Phys. Status Solidi **25**, 641 [1968].

<sup>22</sup> H. J. MATZKE, Can. J. Phys. **46**, 621 [1968].

<sup>23</sup> H. J. MATZKE, Z. Naturforsch. **22 a**, 507 [1967].

<sup>24</sup> R. KELLY and H. J. MATZKE, J. Nucl. Mater. **17**, 179 [1965].

<sup>25</sup> H. J. MATZKE and J. A. DAVIES, J. Appl. Phys. **38**, 805 [1967].

<sup>26</sup> H. J. MATZKE, Phys. Status Solidi **18**, 317 [1966].

<sup>27</sup> J. ZÄHRINGER, Z. Naturforsch. **17 a**, 460 [1962].

dient will be present. However, by proper choice of the sample thickness and by taking the center portion of the cubes it was estimated that the deviations in concentrations could be restricted to about  $\pm 3\%$ .

Furthermore it is assumed that the  $^3\text{He}$  profile is identical with the  $^3\text{H}$  distribution, because the recoil energy of the tritium decay is so small and both He and T, when trapped, are practically immobile at room temperature.

The samples were wrapped into gold foil and put into a stainless steel furnace with a gold wire gasket. The gases released are purified from all chemically active components with Ti- and Cu-CuO-furnaces and a charcoal trap cooled to liquid nitrogen temperature. Both the extraction system and the spectrometer were made of stainless steel. After approximately 10 minutes the inlet valve of the purification system was opened and after further 40 minutes, required for complete purification and cooling the furnaces to about  $100^\circ\text{C}$ , the mass spectrometric measurements were started. Usually, approximately 20 times both mass 3 and mass 4 were measured alternatively and a mean value for the ratio  $^3\text{He}/^4\text{He}$  was derived therefrom. At the end of each run a known  $^3\text{He}/^4\text{He}$  standard was added to the measured gas sample in order to correct for the mass discrimination of the spectrometer and for absolute calibration. To determine the mass effect factor  $\gamma = D_3/D_4$  the ratios and the amounts of the He-isotopes were measured as a function of time respectively of the fraction of gas released. If  $\gamma$  differs from unity, a monotonously decreasing  $^3\text{He}/^4\text{He}$ -ratio should be observed.

## Results

Figure 1 shows the results obtained with a sample of  $2.05 \times 1.73 \times 1.70$  mm at about  $620^\circ\text{C}$ . The

sample was prepared from an irradiated crystal of about 2.5 mm cube edge length by dissolving the outer layers in hydrochloric acid of low concentration. The vertical dashed lines indicate that the run was interrupted in order to weigh the sample, or because of vacuum or spectrometer troubles. The weighing was necessary since it was observed that sublimation of the sample took place. The last weighing showed that the initial cube edge length had decreased by about 3%. It was estimated that this loss could not cause measurable changes in the  $^3\text{He}/^4\text{He}$  ratio. After a total gas release of about 80% the sample was melted and the residual gas analyzed. The result is indicated by the symbols at 100% release. The kinetics of the rare gas release were found to represent volume diffusion. It is seen from the data of the standard in Fig. 1 that variations of about 10% in the isotopic ratio can occur when changes in the instrument were necessary. Usually these variations were observed as a consequence of opening the system and after replacing the multiplier and baking the spectrometer. On the other hand it is seen that under unchanged conditions the isotopic ratio remains fairly constant to approximately 1% or better.

These discontinuities in the isotopic ratio occurred for both the helium released from the sample (Fig. 1, upper curve) and for the standard (Fig. 1, middle curve). The lower curve in Fig. 1 was obtained by correcting the measured values with the

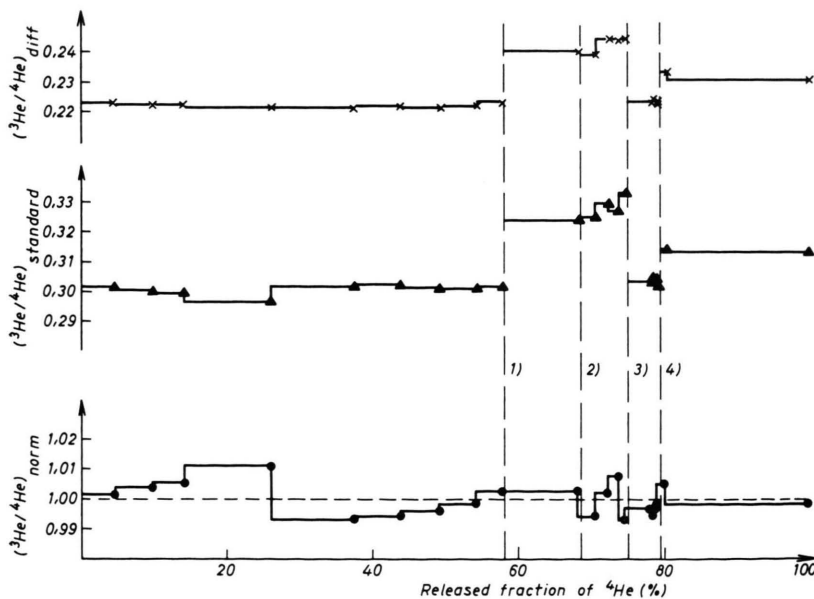


Fig. 1. The upper curve shows the ratios of the released amounts of  $^3\text{He}$  and  $^4\text{He}$  labeled as  $^3\text{He}/^4\text{He}$  diff. The corresponding ratios taken from a  $^3\text{He}/^4\text{He}$ -standard are shown below. The diffusion ratios normalized with the standard values are given by bottom curve labeled as  $^3\text{He}/^4\text{He}$  norm. It is seen that the systematic changes in sensitivity, due to opening the system, are largely removed. The numbers at the vertical dashed lines mean: 1) and 4) weighing the sample, 2) replacing a leaky valve, 3) replacing the multiplier.

standard and normalizing to one. It is obvious that the mass effect is rather small. This is seen more clearly from Fig. 2. The curves were computed from Eqs. (8) and (9) of the appendix for three assumed values of the mass effect factor  $\gamma$ . They represent

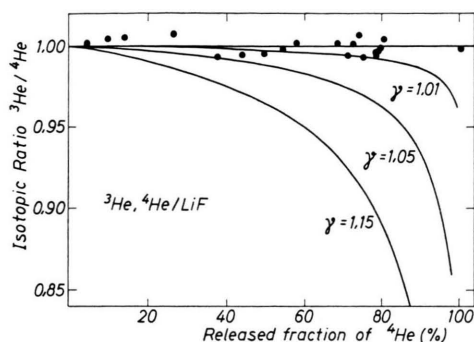


Fig. 2. Differential isotope ratios were calculated as a function of the fractional release of  $^4\text{He}$  for different values of  $\gamma = D_3/D_4$ . The computed curves were shifted by  $1 - \gamma^{1/2}$  in order to have them pass all through point (0,1). The experimental values are indicated by dots.

differential isotopic ratios as a function of the released fraction of  $^4\text{He}$ . Each curve was shifted by the corresponding value of  $1 - \gamma^{1/2}$  in order to obtain a common origin. The curvature, which is characteristic for the magnitude of the isotope effect, is left unchanged by this procedure. Again it is seen that the mass effect is rather small. The upper limit is found to be approximately 1.02 by taking three times the standard deviation.

### Discussion

The mass effect factor determined here is considerably smaller than the values usually found in diffusion systems with interstitial solutes. The systems which have been studied so far were characterized by either rather small interstitials in close packed lattices, e. g.  $\text{Li}^+$  in W,  $\text{H}^{(+)}$  in Fe, Ni, Pt and Pd,  $\text{C}^{(+++)}$  in Fe, or by rather large interstitials in open lattice structures, e. g. He and Ne in quartz, or even by small interstitials in open lattices, e. g.  $\text{Li}^+$  in Si. The He/LiF system is defined by a rather large interstitial (radius: about 1 Å) and a close packed fcc lattice (LiF spacing: 2 Å). Very likely, the equilibrium position of the interstitial He atom is body centered inside the elementary cube consisting of each four  $\text{Li}^+$  and  $\text{F}^-$  ions, while the saddle point configuration corresponds to a face centered

He atom inside of each two  $\text{Li}^+$  and  $\text{F}^-$  ions<sup>13, 15</sup>. Considering the atoms as hard spheres one sees from simple geometrical considerations that considerable lattice relaxation must occur for both configurations of the He atom. Calculations of the activation energies of neutral interstitials in alkali halides take into account for lattice relaxations up to the seventh neighbors<sup>15</sup>.

If we make the reasonable assumptions that He diffuses interstitially in LiF and that trapping and detrapping by certain vacancy structures does not introduce correlation effects, then the mass effect is determined by the kinetic energy factor. Because of the high temperature applied in this experiment (620 °C) it is also rather unlikely that quantum mechanical effects play a dominant role, as they do in hydrogen metal systems at intermediate temperatures. However, this could be checked by degassing at different temperatures. A collective type of diffusion process, such as bubble migration, which would not distinguish between different isotopes, can be ruled out, since the activation energy observed for this system corresponds closely to those of the other rare gas/alkali halide systems, where trapping, but no collective process seems to take place. The bubbles observed in n-irradiated LiF<sup>28</sup> seem to be empty or predominantly filled with fluorine. At higher temperatures there are changes in distribution and size of these cavities. However, they would have to migrate to the surface, if they would constitute the transporting entity for the helium diffusion.

It would be easier to account for the small isotope effect by a vacancy type mechanism, since both correlation and kinetic effects would reduce its dependence on mass. Although this possibility cannot be ruled out at the present state of knowledge, it seems not very likely.

This would mean, in absence of any plausible alternative, that the kinetic energy factor is unusually small for the He/LiF system.

We want to thank the Brookhaven National Laboratory for the reactor irradiation.

### Appendix: Mathematical Forms

The fractional release  $F$  of a species diffusing out of a sphere is given by the approximations:

<sup>28</sup> J. J. GILMAN and W. G. JOHNSTON, J. Appl. Phys. **29**, 877 [1958].

$$F = (6/\pi^{1/2}) \sqrt{Bt}, \quad (F < 10\%) \quad (1)$$

$$F = (6/\pi^{1/2}) \sqrt{Bt} - (3\pi^2) Bt, \quad (F < 85\%) \quad (2)$$

$$F = 1 - (6/\pi^2) \exp(-Bt) \quad (F > 85\%) \quad (3)$$

where  $B = \pi^2 D/R^2$ ;  $t$  = time,  $D$  = diffusion constant,  $R$  = radius of the sphere.

For two isotopes with indices 3 and 4 we obtain after elimination of time and with  $D_3/D_4 = \gamma$ :

$$F_3/F_4 = \sqrt{\gamma}, \quad (4)$$

$$F_3/F_4 = (6\pi/F_4) \left\{ \sqrt{2\gamma} \sqrt{1 - \pi F_4/6} - \sqrt{1 - \pi F_4/3} - \gamma(1 - \pi F_4/6 - \sqrt{1 - \pi F_4/3}) \right\}, \quad (5)$$

$$F_3/F_4 = \{1 - (\pi^2/6)^{\gamma-1}(1 - F_4)^\gamma\}/F_4. \quad (6)$$

The differential ratios of the outgassed isotopes are found to be:

$$dF_3/dF_4 = \sqrt{\gamma}, \quad (7)$$

$$dF_3/dF_4 = \gamma \left\{ \frac{1}{\sqrt{1 - \pi F_4/3}} - 1 \right\} \cdot \left\{ \frac{1}{\sqrt{2\gamma}} \cdot \sqrt{1 - \pi F_4/6 - \sqrt{1 - \pi F_4/3}} - 1 \right\}, \quad (8)$$

$$dF_3/dF_4 = \gamma \left\{ (\pi^2/6)(1 - F_4) \right\}^{\gamma-1}. \quad (9)$$

From the Eqs. (7) – (9) the curves shown in Fig. 2 were computed. The error introduced by regarding a sphere instead of a cube is rather small; the term  $\pi^2/6$  in Eq. (9) has to be replaced by  $\pi^6/512$ .

## A Unified Interpretation of Mass and Charge Distribution and Prompt Neutron Evaporation in Low-Energy Nuclear Fission

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A simple model for a transition state configuration in low-energy fission is given. In its light the latest experimental data on mass and charge distribution and on prompt neutron evaporation are discussed.

The three important characteristics of low-energy nuclear fission, viz, the asymmetry, manifested in the mass distribution, the constant shape and position retained by the heavy mass peak in different fission reactions and the “sawtooth” form of the prompt neutron evaporation curve, have been ascribed to a dumb-bell shape of the compound nucleus immediately before scission<sup>1–3</sup>. The possible effects of the existence of closed shells with 82 neutrons<sup>2</sup>, 50 protons or neutrons<sup>4</sup>, and 82 neutrons plus 50 protons (= 132 nucleons)<sup>3, 5</sup> in the transition state configuration have also been discussed. Based on these concepts FAISSNER and WILDERMUTH<sup>5</sup> proposed their cluster theory and attempted to give a quantitative description of mass<sup>5</sup> and charge distribution<sup>6</sup> in the fission process. This model, however, has two drawbacks: a) It assumes

“equal scission probability anywhere in the neck”, an assumption that cannot be confirmed experimentally nor be expected theoretically. b) It postulates subshells at the mass numbers  $A = 84, 90, 96$ , and 100 in order to reproduce the mass yield curve. These subshells do not have convincing support outside the model.

We believe that the assumption of equal scission probability cannot be maintained and that, in consequence, the mass yield curve cannot be predicted in full from such a simple dumb-bell model. Certain characteristics of the mass yield curve, however, can serve as a check on the validity of the overall picture. A better issue for comparing predictions and experimental findings are the prompt neutron evaporation data and, especially, the charge distribution curve.

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<sup>5</sup> H. FAISSNER and K. WILDERMUTH, Nucl. Phys. **58**, 177 [1964].

<sup>6</sup> H. FAISSNER, Z. Naturforsch. **21 a**, 1021 [1966].