PHYSICAL REVIEW B VOLUME 12, NUMBER 11 1 DECEMBER 1975

Mössbauer studies on ^{83}Kr aftereffects in solid krypton

B. Kolk*
Laboratorium voor Algemene Natuurkunde Rijksuniversiteit Groningen, Westersingel 34, Groningen, Netherlands
(Received 10 July 1975)

Mössbauer investigations on ^{83}Kr in solid krypton at 4.2 K show that the recoilless fraction of a solid-krypton source is about 40% lower than that of a solid-krypton absorber. This effect is attributed to aftereffects caused by the highly converted 32-keV γ-ray decay which precedes the recoilless γ-ray transition.

I. INTRODUCTION

Nuclear transitions that involve the creation of holes in atomic electron shells, such as internal conversion (IC) or electron capture (EC), may be accompanied by one or more Auger processes. As a result of the emission of atomic electrons, the atom may become highly ionized. For instance, in xenon gas containing $^{131}Xe^m$, which decays via internal conversion to its ground state, xenon atoms with charges up to +22 have been observed.[1]

In solids, therefore, highly charged atoms will pick up electrons from their surroundings. Wertheim[2] suggested that in insulators the recombination time may be long enough to observe these highly ionized states when the IC or EC process is followed immediately by a recoilless γ-ray decay. Delayed-coincidence Mössbauer measurements of Trifthäuser and collaborators[3] on ^{57}Co in various insulators show that this atom is already in its final state at the moment the resonant γ ray is emitted, an indication that the ion is neutralized within 10^{-9} sec. However, the final state after EC or IC can still be different from the initial ionic state of the atom, as has been observed in various cases,[4] because of the lattice defects caused by the Auger processes. The consequences of the preceding decay on the recoiless γ-ray emission are called aftereffects.

In this paper such effects on the recoilless γ-ray emission of ^{83}Kr in solid krypton will be studied. Because of the weak van der Waals forces between the noble-gas atoms, the lattice may easily be damaged by the processes accompanying the highly converted γ-ray decay in ^{83}Kr (Fig. 1). Lattice defects formed in the nearby surroundings of an atom emitting a recoilless γ ray will affect the recoilless fraction $f(T)$. In order to observe such effects the $f(T)$ values of solid-krypton sources at 4.2 K were determined, and compared with those of a solid-krypton absorber.

Since in the solid-krypton source only the local surroundings of the γ-ray-*emitting* atom can be damaged by the preceding decay, the recoilless fraction of a γ-ray-*emitting* atom, $f_{se}(T)$, will be distinguished from that of a γ-ray-*absorbing* atom, $f_{sa}(T)$. The latter determines the resonant self-absorption which is appreciable and gives rise to an apparently lower recoilless fraction and to a considerable line broadening. The recoilless fraction of the absorber will be denoted by $f_a(T)$.

Aftereffects were not taken into consideration in previous work[5,6] on ^{83}Kr in solid krypton. That work will, therefore, be discussed first.

II. PREVIOUS WORK ON ^{83}Kr IN SOLID KRYPTON

A. Recoilless-fraction measurements

Mössbauer experiments on ^{83}Kr in solid krypton have been performed by Gilbert and Violet (GV).[5] They used solid-krypton sources as well as absorbers. The measurements were carried out at various temperatures and for various source and absorber thicknesses. Surprisingly the experiments yielded $f(T)$ values considerably smaller than theoretically expected. Various attempts to remove this discrepancy were made without success.[7]

As was pointed out before,[8,9] the maximum resonant cross section $\sigma_0 = (1.89 \pm 0.35) \times 10^{-18}$ cm^2 used by GV was calculated with a wrong value for the

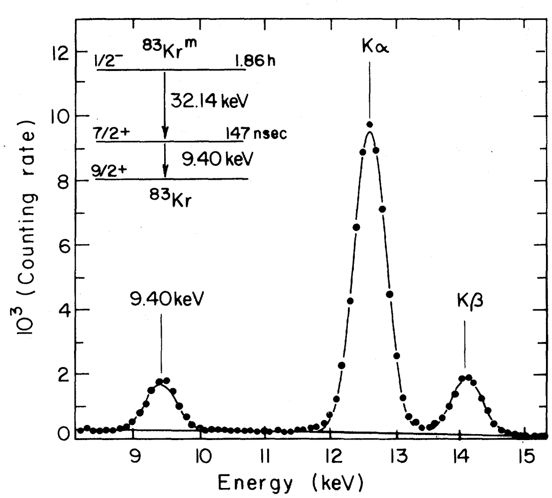

FIG. 1. γ-ray spectrum of a solid $^{83}Kr^m$ source taken with a Si(Li) detector in the experimental arrangement described in Sec. III. The highly converted 32.14 keV transition causes the $K\alpha$ and $K\beta$ x rays.

total IC coefficient ($\alpha_T = 11 \pm 2$) of the resonant γ-ray transition. A remeasurement[10,11] of this quantity yielded $\alpha_T = 19.6 \pm 0.7$, which led to the appreciably smaller value $\sigma_0 = (1.08 \pm 0.04) \times 10^{-18}$ cm^2 used here. Furthermore, possible after effects, which could have reduced the recoilless fraction of the source, were ignored in the GV analysis. Therefore, a reevaluation of the GV data was necessary.

Since GV carried out a sufficient number of measurements for different absorber thicknesses at 7 K, it was possible to determine the recoilless fraction of the absorber, f_a(7 K), from their experimentally observed linewidth Γ_{expt}.[12] For an effective absorber thickness $t_a = af_a\sigma_0 n_a < 10$, Γ_{expt} is within 0.5% given by[13]

$$\Gamma_{\text{expt}}(n_s, n_a) = \Gamma_n(1 + 0.27\, af_a\sigma_0 n_a) + \Gamma_s(n_s) + \Delta\Gamma, \quad (1)$$

where $\Delta\Gamma$ incorporates possible instrumental line broadening, Γ_n is the natural linewidth, Γ_s the full width at half-maximum of the recoilless γ-ray energy distribution emitted by the source, a the abundance of ^{83}Kr in natural krypton, and n_s and n_a are the number of krypton atoms per cm^2 in the source and absorber. Hence, for each source thickness n_s the corresponding values of Γ_{expt} as a function of n_a should lie on a straight line whose slope is determined by f_a. Least-squares fits of the GV linewidth data with Eq. (1) resulted in $f_a(7\text{ K}) = 0.87 \pm 0.08$. As will be noticed, the value of f_a thus derived is independent of the instrumental line broadening $\Delta\Gamma$ and of the recoilless fraction of the source.

According to GV, their data were corrected for instrumental line broadening, thus $\Delta\Gamma = 0$ in Eq. (1). An extrapolation of the $\Gamma_{\text{expt}}(n_s, n_a)$ data to $n_a = 0$ then yielded the linewidth $\Gamma_s(n_s)$ for each of their sources. These values are plotted in Fig. 2. We also calculated the line broadening due to resonant self-absorption as function of the source

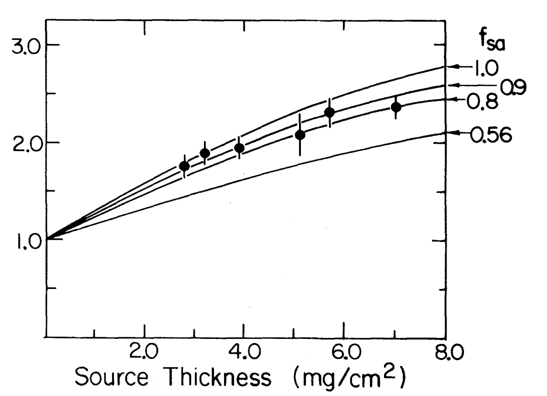

FIG. 2. Half-width Γ_s of γ-ray distribution emitted by a solid ^{83}Kr^m source derived from the GV data.

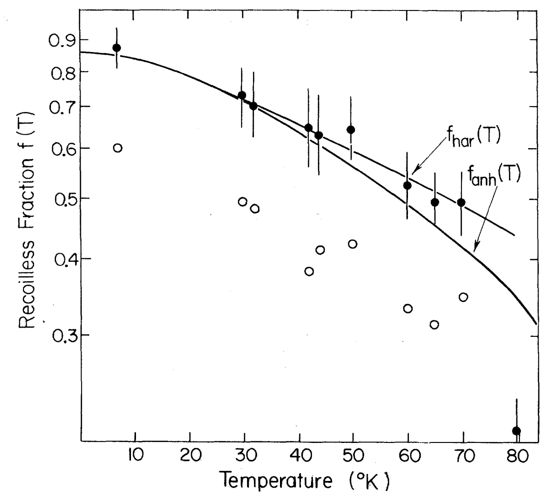

FIG. 3. Corrected f_a values (●) and original values (○) of GV. The solid curves represent f values for a harmonic and an anharmonic solid-krypton lattice as calculated in Refs. 8 and 9.

thickness n_s for various values of f_{sa}(7 K). The best agreement with the $\Gamma_s(n_s)$ data was obtained for $f_{sa}(7\text{ K}) = 0.9 \pm 0.1$, in good agreement with the value found above for the solid-krypton absorber.

The resonant absorption areas $A_n(T)$ measured by GV can be expressed as[9]

$$A_n(T) = f_{\text{eff}}(7\text{ K})\, F(f_a(T)), \quad (2)$$

with

$$f_{\text{eff}}(7\text{ K}) = N_r/(N_r + N_{nr}),$$

where N_r and N_{nr} are the recoilless and nonrecoilless intensities of the 9.4-keV γ rays after self-absorption in the solid-krypton source. The absorber function $F(f_a(T))$ for a single line absorber is given by modified Bessel functions.[14] The values of f_{eff}(7 K), derived from the absorption area A_n(7 K), are represented in Table II, and will be compared with our results in Sec. III.

Next, the recoilless fractions $f_a(T)$ of the absorber for $T > 7$ K were evaluated from the normalized absorption areas $A_n(T)$ using the effective recoilless fractions of the sources determined above. The results are shown in Fig. 3 together with the values of GV. In this figure the f values calculated for an anharmonic and a harmonic solid-krypton crystal with the phonon-frequency spectrum of Brown and Horton[15] are also given. A more extended discussion of anharmonic effects has been given in Refs. 8 and 9.

The reevaluated $f_a(T)$ values show a fair agreement with the theory, in contrast to the original results of GV. Hence, the recoilless fraction of ^{83}Kr in solid krypton shows no anomalies as far as resonant absorption in the absorber is concerned.

Furthermore, it is important to notice that in our evaluation of the $f_a(T)$ values the recoilless fraction of the source is canceled because the evaluation for $T > 7$ K is, in principle, based on the following equation:

$$F(f_a(T)) = \frac{A_n(T)}{A_n(7\text{ K})} F(f_a(7\text{ K})). \qquad (3)$$

Hence, a possible reduction of the source recoilless fraction f_{se}, due to aftereffects, will not influence the $f_a(T)$ results derived above.

B. Resonant self-absorption measurements

Brown[6] evaluated the $f(4\text{ K})$ value of ^{83}Kr in solid krypton from the resonant *self*-absorption by measuring the intensity ratio $R = I(12.6)/I(9.4)$ for various source thicknesses n_s. The resonant self-absorption will affect the intensity of the 9.4-keV γ rays, $I(9.4)$, but not that of the 12.6-keV x rays, $I(12.6)$. In Brown's analysis, possible aftereffects were not taken into account. The data were fitted for various values of σ_0, yielding f values ranging from 0.70 to 0.85. An extrapolation of the intensity ratio R to zero source thickness yields $R_0 = 2.5$, in contrast to our result[9,11] $R_0 = 3.16 \pm 0.05$ measured with a Si(Li) detector and a Xe proportional counter using an ^{83}Kr(Al) source.[16] This last value was confirmed by Ruby *et al.*[10]

Brown attributed the spread in his R values to deposition of the krypton gas on the walls of the source cell, instead of on the beryllium window, where the source thickness n_s was measured. An underestimate of the effective source thickness gives the false impression of higher resonant self-absorption and thus yields too high an f value. The source thickness was determined with a ^{57}Co source always present in the solid-krypton cell. The photoelectric absorption of the 14.4-keV ^{57}Co γ rays whose energy lies just above the K edge of krypton (14.2 keV), gives rise to an increase in the krypton x-ray intensity. This effect leads also to an overestimate of the f value.

Hence, from the resonant self-absorption measurements of Brown only an upper limit of the recoilless fraction can be determined. Using $f_{sa}(4\text{ K}) = 0.87$ and $R_0 = 3.16$ we evaluated from Brown's data $f_{se}(4\text{ K}) \leqq 0.70$. Thus, the resonant self-absorption data seem to indicate that the recoilless fraction of a γ-ray-*emitting* atom, f_{se}, is smaller than f_{sa} or f_a.

III. MÖSSBAUER MEASUREMENTS WITH SOLID KRYPTON SOURCES

A. Measurements

To establish whether a difference between f_{se} and f_a exists, measurements with solid-krypton sources were carried out. The ^{83}Kr activity was produced by the reaction $^{82}\text{Kr}(n,\gamma)\,^{83}\text{Kr}^m$. For this purpose, 10-ml Kr gas, enclosed in a quartz capsule, was irradiated in the high-flux reactor ($\phi = 5 \times 10^{13}$ n/cm^2 sec) of the Reactor Centrum Nederland at Petten, Netherlands. After a $\frac{1}{2}$-h cooling period, the quartz capsule was broken and the krypton gas was frozen on a Mylar foil, coated with aluminum and kept at liquid-nitrogen temperature. The source was then slowly cooled down in the cryostat from liquid-nitrogen to liquid-helium temperature. To determine the source thickness the intensity ratio of the 12.6-keV x rays to the 9.4-keV γ rays was measured with a Si(Li) detector, Fig. 1, and compared with that observed for an infinitely thin $^{83}\text{Kr}^m$(Al) source[16] with the same effective area and in the same geometry. A correction for resonant self-absorption in the source was made. The effective thicknesses of both solid krypton sources, thus derived, are shown in Table I.

Krypton in β-hydroquinone (β-HQ) was chosen as resonant absorber for the following reasons: This absorber is easy to handle and the weight of the krypton per cm^2, w_a, can easily be determined by methods described in Refs. 9 and 17. For the absorber used here, $w_a = 4.0 \pm 0.6$ mg/cm^2. Further, the recoilless fraction of ^{83}Kr in β-HQ is reasonably well known[9,17]; $f_a(4\text{ K}) = 0.89 \pm 0.07$.

The Mössbauer measurements were carried out at 4.2 K in a cryostat with thin Mylar windows, to minimize the nonresonant absorption of the 9.4-keV γ rays. The resonant-absorption spectra shown in Fig. 4 were taken with a conventional electromechanical feedback system in combination with a multichannel analyzer in the constant-acceleration mode. The velocity region of the spectra was chosen sufficiently large to allow the observation of a possible quadrupole splitting which could arise from $[\text{Kr}_2]^+$ centers, as will be discussed in Sec. IV. However, within the statistics only a central absorption line due to neutral krypton atoms was observed. In spectrum (b) of Fig. 4 the curved background is caused by solid-angle effects.

B. Determination of $f_{se}(4\text{ K})$

Both spectra in Fig. 4 were fitted with a Lorentzian line. Actually, the absorber line shape is not Lorentzian, because of a small quadrupole splitting.[17] However, owing to the considerable line

TABLE I. Experimental data of Mössbauer-effect measurements carried out with solid-krypton sources using a β-hydroquinone absorber. Source thickness ω_s, background-correction factor γ, and normalized absorption area A_n.

ω_s (mg/cm^2)	γ%	A_n (units of $\frac{1}{2}\Gamma_n$)
6.35 ± 1.0	75 ± 3	4.64 ± 0.5
4.0 ± 0.65	78 ± 3	5.5 ± 1.0

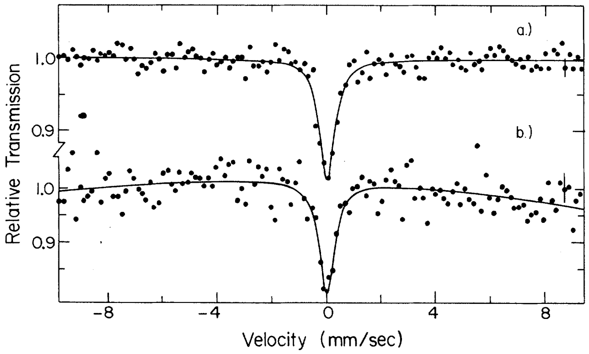

FIG. 4. Mössbauer-effect spectrum of solid $^{83}Kr^m$ sources versus a Kr hydroquinone absorber taken at 4.2 K. Spectrum (a) contains 7.3×10^3 counts in the first channel; spectrum (b) 2.5×10^3 counts.

broadening in the source arising from resonant self-absorption this asymmetry is hard to observe. The error made by fitting the absorption line with a single Lorentzian lies within the accuracy of the measurements. From the least-squares fit the normalized resonant absorption area A_n was determined,

$$A_n = \frac{2}{\Gamma_n}\int_{-\infty}^{+\infty} \frac{N(\infty)-N(v)}{\gamma N(\infty)}\,dv, \qquad (4)$$

where $N(v)$ is the measured counting rate as function of the source velocity v relative to the absorber, $N(\infty)$ the counting rate extrapolated for $v\to\infty$, and γ the background correction factor, which is the fraction of $N(\infty)$ arising from the 9.4-keV γ rays. The factor $2/\Gamma_n$ in Eq. (4) makes A_n dimensionless. Values of γ and A_n are displayed in Table I.

The effective recoilless fraction $f_{\rm eff}$ of the solid-krypton source can be derived from the normalized resonant-absorption area $A_n = f_{\rm eff}\,F(t_a)$, with

$$F(t_a) = \frac{2}{\Gamma_n}\int_{-\infty}^{+\infty} \left(1 - e^{-t_a\sigma_r(v')/\sigma_0}\right)dv', \qquad (5)$$

where $\sigma_r(v')$ is the resonant-absorption cross section of the absorber and $t_a = a\sigma_0 f_a n_a = 7.1 \pm 0.7$ for the KrHQ absorber used. Because of the quadrupole splitting of ^{83}Kr in HQ, $\sigma_r(v')$ is split into 11 lines, so that Eq. (5) cannot be expressed by modified Bessel functions as in the case of a single line absorber. The integral $F(t_a)$ was calculated using the values $Q^*/Q = 1.99\pm0.05$ and $e^2qQ(c/E_\gamma) = 1.00 \pm 0.12$ mm/sec found in Ref. 17, where Q^* and Q are the quadrupole moments of the 9.4-keV level and ground state of ^{83}Kr, respectively, and q is the maximum electric field gradient at the ^{83}Kr nucleus. With the value $F(7.1\pm0.7) = 12.7\pm1.3$ the $f_{\rm eff}$ results given in the upper half of Table II are obtained.

It can readily be derived from the definition of $f_{\rm eff}$ given in Sec. II A that

$$f_{\rm eff} = \frac{N_r}{N_{nr}+N_r} = \frac{f_{se}\tilde{B}}{(1-f_{se})+f_{se}\tilde{B}}, \qquad (6)$$

with the self-absorption correction factor

$$\tilde{B} = \frac{\sigma_{nr} n_s}{1-e^{-\sigma_{nr}n_s}}\int_{-\infty}^{+\infty} B(v';\Gamma_s)\,dv', \qquad (7)$$

where $B(v';\Gamma_s)$ is the resonant γ-ray distribution emitted by the source, given by Eq. (A1) in the Appendix. The calculated $\tilde{B}$ values together with the f_{se} results derived from the $f_{\rm eff}$ values with Eq. (6) are shown in Table II. A weighted average yields $f_{se} = 0.53\pm0.06$.

Similarly, f_{se} can be determined from the effective recoilless fractions of solid-krypton sources which were obtained from the GV data in Sec. II A. The results, shown in the lower part of Table II, give a weighted average of $f_{se} = 0.56\pm0.04$ in very good agreement with the f_{se} value derived above. Both results, however, deviate considerably from the value $f_a(7\text{ K}) = 0.87\pm0.08$ found for a krypton absorber. This discrepancy cannot be explained by resonant self-absorption even if the sources were infinitely thick, since in that case one derives readily from Eq. (7) and Eq. (A6) in the Appendix that

$$\tilde{B} = (1 + af_{sa}\sigma_0/\sigma_{nr})^{-1/2}. \qquad (8)$$

Hence, for $f_{sa} = 0.87$, $\tilde{B}$ amounts to 0.27 which, if $f_{se} = f_a$, would yield an $f_{\rm eff} = 0.65$, a value much higher than those obtained experimentally. (See Table II.)

An explanation might be the poor quality of the krypton crystals used as sources, or an increase in source temperature due to dissipation of decay energy. One would then expect the recoilless fraction of an *absorbing* atom in the source, f_{sa}, to be also lowered by about 40%. However, the analysis of the linewidth broadening due to resonant self-absorption in Sec. II A showed that $f_{sa}\simeq f_a$, which

TABLE II. Recoilless fraction f_{se} of a γ-ray-emitting $^{83}Kr^m$ atom in solid krypton measured by us (upper half of the table) and evaluated from $f_{\rm eff}$ derived in Sec. III from GV measurements.

ω_s (mg/cm^2)	$\tilde{B}$	$f_{\rm eff}$	f_{se}
6.35 ± 1.0	0.52 ± 0.05	0.365 ± 0.057	0.52 ± 0.07
4.0 ± 0.65	0.61 ± 0.05	0.431 ± 0.095	0.55 ± 0.10
		average	0.53 ± 0.06
7.0	0.50	0.33 ± 0.03	0.50
5.7	0.54	0.37 ± 0.03	0.52
5.1	0.56	0.40 ± 0.04	0.54
3.8	0.62	0.49 ± 0.05	0.49
3.2	0.65	0.61 ± 0.06	0.71
2.8	0.68	0.49 ± 0.05	0.58
		average	0.56 ± 0.04

TABLE III. Results obtained for the recoilless fraction of a solid-krypton absorber (f_a) and sources (f_{sa} and f_{se}) at 4 K.

Recoilless fraction		Derived from
f_a	0.87 ± 0.08	GV data
f_{sa}	0.9 ± 0.1	GV data
f_{se}	0.53 ± 0.06	present work
f_{se}	0.56 ± 0.04	GV data
f_{se}	$\leqq 0.70$	Brown's data

excludes these possible explanations of the reduction of f_{se}. More probably, the deviation of f_{se} from f_{sa} and f_a arises from aftereffects, caused by the preceding γ-ray transition, as will be discussed next.

IV. AFTEREFFECTS

More than 99.9% of the ^{83}Kr^m nuclei decay to the 9.4-keV Mössbauer state by IC. In this process the released nuclear energy (32 keV) is carried away by a core electron which is ejected from the atom. Because of the electric-octupole ($E3$) character of the 32-keV transition (Fig. 1) the conversion electron will most probably be an L or M electron. The hole in these inner shells will be filled very quickly by an electron from a higher shell, with emission of either an Auger electron or an x ray. The last process has a very low probability when the initial hole is in an L shell or higher.

Hence, while the ^{83}Kr nucleus is in the 9.4-keV state (hereafter denoted by ^{83}Kr*) the preceding IC process and the accompanying Auger processes have produced various holes in the $4p$ valence band of solid krypton. These holes will be trapped close to the ^{83}Kr* atom since the mobility of holes in the rare-gas solids is very low.[18] Druger and Knox[19] showed that a hole can be trapped by two rare-gas atoms R, forming an $[R_2]^+$ center. In a simple model such an $[R_2]^+$ center can be regarded as two-rare-gas atoms sharing a $4p_z$ hole.

It might be that the ^{83}Kr* atom itself is a part of a $[Kr_2]^+$ center. In that case the recoilless γ rays should show an isomer shift $\delta = -0.80$ mm/sec and an electric-quadrupole splitting with $B = e^2qQ(c/E_\gamma) = -69$ mm/sec. The values of δ and B are obtained from an interpolation of the $^{83}KrF_2$ Mössbauer data,[17,20] in which case the krypton atom is missing 0.94 ($4p_z$) electrons. Owing to the large quadrupole splitting these recoilless γ rays will not contribute to the central absorption line arising from neutral ^{83}Kr* atoms. This effect may cause an apparently lower recoilless fraction f_{se}. A fit of the data shown in Fig. 4 to a quadrupole-split spectrum characterized by the values of δ and B given above did not indicate the existence of such a $[Kr_2]^+$ component, but did not exclude this possibility either, because of the statistics of the data and the uncertainty in the estimated values of δ and B.

Because of the high mobility of electrons in rare-gas solids,[18] most holes, however, probably recombine within 10^{-9} sec with Auger electrons or with electrons produced by inelastic collisions of the Auger electrons with the krypton atoms. When a recombination occurs, roughly 10 eV is released. Since the interatomic forces in solid krypton are very weak, this energy is sufficient to displace one or both Kr atoms of the $[Kr_2]^+$ center from their lattice sites. The remaining part of the released energy will excite lattice vibrations which can yield a local temperature increase far above the melting point (115 K). Another contribution to the local temperature increase arises from the energy of the Auger electrons dissipated in the surroundings of the ^{83}Kr* atom. As a result of thermal expansion the lattice will be locally deformed. Because of the very weak restoring forces in solid rare gases we assume that the inelastic deformation exists longer that 10^{-7} sec. The lattice constant a_0 in the deformed region is then increased on the average with Δa, where $\Delta a/a_0$ will be approximately of the order of magnitude of the relative expansion of a Kr lattice brought from 4 K to melting temperature, i.e., $\Delta a/a_0 \simeq 0.15$. In this local region the potential in which the Kr atom moves will have a flat center of about Δa, similar to the potentials for light van der Waals solids like Ne, D_2, H_2, and He, and for dense van der Waals gases. For such anharmonic potentials the theory used to calculate the recoilless fraction of the solid-krypton absorber in Refs. 8 and 9 is no longer valid. Therefore the following model was used: The atom in question is considered to move in a cage formed by the nearest-neighbor atoms. The mean-square displacement $\langle x^2\rangle$ of the atom in the direction of the emitted γ ray arises then from the motion of the atom in the cage and from the motion of the cage itself, so that the recoilless fraction can be written as

$$f = e^{-k^2(\langle x^2\rangle_{loc} + \langle x^2\rangle_{cage})} = f_{loc} \times f_{cage},$$

where k is the wave number of the recoilless γ ray. To simplify the calculations considerably we assumed $f_{cage} \simeq f_{loc}$ and evaluated f_{loc} for a hard-wall potential[21] using $\Delta a/a_0 = 0.15$. The result yields an upper limit of f because, in fact, the actual potential is described better by a soft-wall potential. The calculations yielded $f_{se} \leqq 0.6$, in agreement with the observed result $f_{se} = 0.53$.

V. CONCLUSIONS

The recoilless fraction $f_a(T)$ of ^{83}Kr in a solid krypton *absorber* as function of temperature shows fair agreement with theory (Fig. 3). However, from three independent measurements the remarkable result has been evaluated that at 4 K the recoilless fraction of the solid-krypton source, f_{se}, is about 40% lower than that of the solid-krypton absorber (see Table III). As pointed out in Sec. III B this effect cannot be attributed to resonant self-absorption or to poor quality of the krypton crystals used as source.

As discussed in Sec IV, the 32-keV IC decay preceding the resonant γ-ray transition generates holes in the $4p$ band of solid krypton which will be trapped close to the ^{83}Kr* atom. A hole trapped by the ^{83}Kr* itself gives rise to a large quadrupole splitting and an apparently lower recoilless fraction. Within our statistics no such quadrupole splitting was observed.

It is likely that most of the holes, because of the very high mobility of electrons in rare-gas solids, recombine very quickly with the Auger or other electrons, whereby in each case an energy of about 10 eV is released. This energy is sufficient to disorder the lattice locally, since the van der Waals forces between the atoms are very weak. The ^{83}Kr* atom in such a disordered region moves in an anharmonic potential. A simple model yields for that case $f_{se} \leqq 0.60$, in agreement with the observed result $f_{se} = 0.53 \pm 0.06$.

ACKNOWLEDGMENTS

The measurements at the Reactor Centrum Nederland at Petten were carried out with kind permission of the staff, whom the author also wants to thank for their assistance during his work at Petten. Further he wishes to thank H. de Waard, N. Benczer-Koller, and G. Collins for their helpful discussions.

APPENDIX: RESONANT SELF-ABSORPTION EFFECTS

For an ^{83}Kr^m source of solid krypton one readily derives[9] that if the ^{83}Kr atoms are homogeneously distributed in the source, the resonant γ-ray distribution can be expressed by

$$B(v';\Gamma_s) = (\Gamma_n/\Gamma_1)\; L(v';\Gamma_1)\, C(v';n_s)\;, \qquad \text{(A1)}$$

with

$$\Gamma_1/\Gamma_n = (1 + a f_{sa}\sigma_0/\sigma_{nr})^{1/2}, \qquad \text{(A2)}$$

$$L(v';\Gamma) = \frac{2/\pi\Gamma_n}{1+(2v'/\Gamma)^2}\;, \qquad \text{(A3)}$$

and

$$C(v';n_s) = (1 - e^{-[\sigma_{nr} + a f_{sa}\sigma_r(v';\Gamma_n)]x_s})/\sigma_{nr}x_s\,, \qquad \text{(A4)}$$

where

$$\sigma_r(v';\Gamma_n) = \frac{\sigma_0}{1+(2v'/\Gamma_n)^2}\;. \qquad \text{(A5)}$$

From Eqs. (A1)–(A4) it follows that for thin sources the resonant γ-ray distribution can be represented by a Lorentz distribution $L(v;\Gamma_n)$ with natural linewidth Γ_n as expected. In the case of thick sources, where $C(v';n_s) = (\sigma_{nr}n_s)^{-1}$, the resonant γ-ray distribution is given by

$$B(v';\Gamma_s) \simeq \frac{(\Gamma_n/\Gamma_1)\, L(v';\Gamma_1)}{\sigma_{nr}a_s}\,. \qquad \text{(A6)}$$

This is a Lorentzian distribution with linewidth Γ_1. Hence, the source linewidth Γ_s varies from the natural linewidth Γ_n to a maximum Γ_1. Γ_s as a function of the source thickness n_s was determined from Eq. (A1) numerically. The results are shown in Fig. 2 for various values of the recoilless fraction f_{sa}.

---

*Present address: Rutgers University, Department of Physics, New Brunswick, N. J. 08903

[1]A. H. Snell, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1968), p. 1545.

[2]G. K. Wertheim, Phys. Rev. 124, 764 (1961); H. N. Ok and J. G. Mullen, *ibid* 168, 550 (1968); G. K. Wertheim and H. J. Guggenheim, J. Chem. Phys. 42, 3873 (1965).

[3]W. Triftshäuser and P. P. Craig, Phys. Rev. 162, 274 (1967); P. Jung and W. Triftshäuser, *ibid.* 175, 512 (1968); W. Triftshäuser and D. Schroeer, *ibid.* 187, 491 (1969).

[4]R. Ingalls, C. J. Coston, G. DePasquali, and H. G. Drickamer, J. Chem. Phys. 45, 1057 (1966); J. Chappert, R. B. Frankel, and A. Misetich, Phys. Rev. 179, 578 (1969); J. M. Friedt and G. K. Shenoy, J. Chem. Phys. 59, 3831 (1973); F. Ambe, S. Ambe, H. Shofi, and N. Saito, *ibid.* 60, 3773 (1974).

[5]K. G. Gilbert and C. E. Violet, Phys. Lett. A 28, 285 (1968); K. G. Gilbert, thesis (University of California, 1968) (unpublished), UCRL No. 50474.

[6]J. B. Brown, Jr., in *Mössbauer Effect Methods*, edited by I. J. Gruverman *et al.* (New England Nuclear Corp., New York, 1974), Vol. 9, p. 23.

[7]P. Vashishta and K. N. Pathak, Physica 48, 474 (1970); J. S. Brown, Phys. Rev. B 3, 21 (1971); K. Mahesh and S. V. Kapil, Phys. Status Solidi B 47, 397 (1971); S. S. Nandwani, D. Raj, and S. P. Puri, J. Phys. C 4, 1929 (1971).

[8]B. Kolk, Phys. Lett. A 35, 83 (1971).

[9]B. Kolk, thesis (RU Groningen, Netherlands, 1974) (unpublished).

[10]S. L. Ruby, R. G. Clark, and L. E. Glendenin, Phys. Lett. A 36, 321 (1971).

[11]B. Kolk, F. Pleiter, and W. Heeringa, Nucl. Phys. A 194, 614 (1972).

[12]A few measurements were carried out by GV at 5 K. However, the relative difference between f(5 K) and f(7 K) is less than 0.5%, which is much smaller than the experimental error limits, so that henceforth f(4.2 K) = f(5 K) = f(7 K).

[13]O. D. A. O'Conner, Nucl. Instrum. 21, 318 (1963).

[14]D. P. Johnson and J. G. Dash, Phys. Rev. 172, 983 (1968).

[15]J. S. Brown and G. K. Horton, Phys. Rev. Lett. 18,

647 (1967).

[16]The $^{83}Kr^m$ (Al) source was made (Refs. 9 and 11) by implantation of ^{82}Kr in a very thin Al foil and by irradiating that foil with thermal neutrons to produce $^{83}Kr^m$. The resonant self-absorption in these sources was zero and the nonresonant self-absorption negligible.

[17]B. Kolk, Phys. Rev. B 12, 1620 (1975).

[18]L. S. Miller, O. Howe, and W. E. Spear, Phys. Rev. 166, 871 (1968); H. D. Pruett and H. P. Broida, *ibid.* 164, 1138 (1967).

[19]S. D. Druger and R. S. Knox, J. Chem. Phys. 50, 3143 (1969).

[20]S. L. Ruby and H. Selig, Phys. Rev. 147, 348 (1966).

[21]J. G. Dash, D. P. Johnson, and W. M. Vischer, Phys. Rev. 168, 1087 (1968).