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Kinetics of Stimulated Mössbauer Emission in Neutron-Pumped Krypton-83

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OFFICE MEMORANDUM

TO : Holders of LA-7004-MS

DATE: November 14, 1977

FROM : ISD-6

SUBJECT : Erratum

SYMBOL :

Page 2, Line 7.

Change (0.56 MeV) to (0.56 meV).

KINETICS OF STIMULATED MOSSBAUER EMISSION IN NEUTRON-PUMPED KRYPTON-83

by

G. C. Baldwin and Laurie E. McNeill

ABSTRACT

Using an idealized kinetic model for a gamma-ray laser system pumped by a spatially uniform delta-function burst of fast neutrons, a computer study has been made of the growth, decay, and attenuation of resonant 9.3-keV recoilless gamma radiation from ^{83}Kr , as a function of neutron-burst intensity, gamma-ray linebreadth, temperature, dilution of krypton in a beryllium host, and nonresonant absorption coefficient of the host. The isomer is formed by neutron capture in a 40-eV resonance, and the 144-ns transition lifetime is short in comparison with the time for neutrons to moderate. The kinetic behavior of this system is therefore determined largely by the time dependence of the neutron spectrum and only slightly by the reciprocal linebreadth of the graser transition. Because the lower state is stable, inversion is rapidly lost, so that, for observable gain, an unrealistically high source intensity is needed. Use of a beryllium host, which increases the Debye temperature, is negated by its parasitic absorption. Although this transition is unsuitable for a graser, these findings help to illustrate useful properties of nuclear isomers and solid hosts for which stimulated emission might be observable.

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I. INTRODUCTION

A. Krypton-83 as a Graser Candidate

Possibility in principle of stimulated emission of gamma radiation has been recognized for many years,¹ but possibility in practice has yet to be demonstrated. In particular, no specific transitions have yet been identified for which an experimental demonstration of the phenomenon appears probable. Given the incomplete state of our knowledge of nuclear properties, however, one cannot yet exclude the possibility of identifying transitions which can be stimulated.

With the purpose of illustrating the problems involved and of identifying the relative importances of various parameters which characterize a graser transition, we report here a detailed study of one particular isotope, ^{83}Kr , which has been suggested² as a promising candidate for graser action because of the following properties.

- 1) ^{83}Kr is a well-known Mössbauer isotope, emitting a 9.3-keV gamma ray with low recoil energy (0.56 MeV) in its 144-ns isomeric transition to the ground state.³
- 2) Its resonance cross section³ is approximately 140 times the nonresonant absorption cross section of krypton and 100 000 times that of Be.⁴
- 3) ^{83}Kr can be generated by neutron capture in ^{82}Kr . Its thermal neutron capture cross section is 45 b, and there is a 40-eV capture resonance with resonance integral of 200 b.⁵
- 4) In the neutron capture reaction, the probability of forming the 9.3-keV state ($I = 7/2+$) is high relative to that of forming the ground state ($I = 9/2+$), because of the large spin difference between the compound state formed in the capture reaction ($I = 1/2+$) and the ground state of the isotope.⁶

Offsetting these advantageous properties somewhat are the facts that the 1.86-h isomeric level at 41.8 keV is populated, instead of the 9.3-keV level, in two-thirds of all neutron capture events, and the internal conversion coefficient of the latter (18.9) is moderately high, reducing the probability of gamma-ray emission. Moreover, krypton is a noble gas.

Bowman's proposal² includes two specific suggestions: 1) to emphasize the 40-eV capture resonance in a neutron burst-pumped graser system, by arranging for the fast neutrons to thermalize in a block of moderator material, which has been set into motion by their momentum, Doppler-upshifting the average energy of the thermalized neutrons to the capture resonance; 2) to incorporate the krypton in a beryllium host.

B. Time Dependence of Stimulated Emission

The present analysis is not directly addressed to the specific system described in Bowman's proposal, as that would introduce considerable complications. Rather, in the present study, we treat the kinetics of Mössbauer radiation in an idealized, hypothetical case;⁷ namely, solid material containing ^{82}Kr , immersed in an infinite hydrogenous moderator at rest, and exposed to a spatially uniform delta-function time burst of S fast neutrons cm^{-3} . The neutron flux per unit velocity, as a function of time and velocity, is given by the analytical expression⁸

$$\phi(v,t) = S (v\Sigma_s t)^2 \exp(-v\Sigma_s t) , \quad (1)$$

on the assumption that capture in the specimen has negligible effect on the neutron flux. The quantity Σ_s is the macroscopic scattering cross section; throughout the calculations, the value 1.40 cm^{-1} has been used.⁹

Finite time is required for neutrons to moderate to the resonance region, and still longer, before they thermalize. Thus, there is a time lag in the appearance of the excited states. According to Eq. (1), the time for maximum neutron flux at any particular velocity v is $2/v\Sigma_s$; at the 40-eV resonance in ^{82}Kr , this amounts to 162 ns, which exceeds the mean lifetime of the excited state. To thermalize requires 6.5 μs . This time lag reduces the effective source strength.

A further time lag is expected for the system of resonant nuclei to respond to spontaneous emission after the excited states have formed.^{7,10} When the gamma-ray line is of near-natural width, this is comparable with the mean lifetime of the transition. It has been suggested by Vorontsov and Vysotskii¹¹ and by Hannon and Trammel¹² that the decay during this lag time increases the critical inversion density necessary for stimulation gain to exceed nonresonant absorption in a pulsed graser. However, their analyses assume instantaneous formation of the inversion and overlook the time spread due to neutron moderation.⁷ This moderation time ensures that pumping be sustained over a time interval, which can be comparable with or even exceed the transition lifetime, according to the energy dependence of the capture cross section. To take this complexity into account requires a numerical analysis of each proposed system.

In the absence of resonant interactions, the spatial dependence of the radiation which emerges from the graser body is controlled by photoelectric absorption, so that one observes lower intensity in the axial direction than in directions transverse to a thin filament. There is also an anomalous time dependence and further reduction of the emergent intensity if resonant absorption by ground states occurs.¹³ Stimulated emission, if appreciable, would be manifested by apparent reduction of the time-dependent self-absorption. In addition, if the probability of stimulated emission were high enough for net gain, greater intensity would be observed in the axial than in the transverse direction.

II. NATURE OF THIS STUDY

A. Theoretical Model

A computer code (described in Ref. 14) has been developed for numerical analyses of conceptual, neutron burst-pumped graser systems. We report here its application to the particular case of ^{83}Kr .

We have assumed that the isomer is formed by neutron capture in a thin filament of solid material containing ^{82}Kr , which is immersed in a large hydrogenous moderator and subjected to a burst of S fast neutrons cm^{-3} at time $t = 0$. These subsequently moderate according to Eq. (1). With the code, we first calculate population densities of the parent isotope and of the 9.3-keV and ground states of ^{83}Kr , as functions of time after the source burst, by solving differential rate equations in which changes of population are produced by neutron capture and radioactive decay. Inelastic scattering, which may reexcite the ground state or deexcite the isomer, was not considered in this approximation because cross sections were not available for the excited state.

Given the population densities, Mössbauer parameters, linebreadth, and nonresonant removal cross sections of the graser material, the code next calculates the amplitude of a plane electromagnetic wave originating in spontaneous emission from an isomeric nucleus at $x = 0$. The theoretical basis for this code, obtained by solving Maxwell's equations for an excited medium in the small signal approximation, is given in Ref. 7.

Appropriate parameters were varied in order to ascertain the effects of line broadening, krypton dilution in a beryllium host, temperature, and non-resonant absorption coefficient. The neutron source strength S was given a sequence of values in order to ascertain the pump intensity for which stimulated emission from an elongated crystal would be manifested by increased, rather than reduced intensity in the axial direction, relative to the intensity in directions transverse to the graser axis. Intensities were computed by integrating squares of amplitudes of incoherent waves initiated from spontaneous sources uniformly distributed along the length of the filament.⁷

B. Computer Code

The computer codes GRASER and GRAZER¹⁵ used in this study calculate the following quantities.

$N_p(t)$ = the number density of the parent nuclide as a function of time.
 $N_2(t)$ = the number density of the upper graser level as a function of time.
 $N_1(t)$ = the number density of the lower graser level as a function of time.
 $Q(t)$ = the product of the quantity $\sigma_r \Gamma/4$ and the time integral of the inversion density N^* , where

$$N^*(t) = N_2(t) - G N_1(t) , \quad (2)$$

Γ is the linewidth, σ_r is the resonant cross section, and
 G is the statistical weight ratio of the two states.

$A(x,t)$ = the vector potential of the plane wave as a function of axial distance x from the spontaneous source and time t after the source burst.

The wave vector potential is that initiated by a single spontaneously emitting nucleus located at $x = 0$. Code GRAZER calculates also the space integral of the intensity resulting from a uniform distribution of sources along the length of the graser body, and normalizes it to the case of no recoilless interaction (resonant absorption, stimulated emission) in which the intensity is affected only by nonresonant self-absorption.

The input parameters used in these calculations are defined and listed in Appendix A; the equations which are solved are given in Appendix B.

III. RESULTS

A. Format

Figures 1-17 and Table I display the results of the parameter study.

For each case, the nuclear populations and the vector potential are displayed in two subplots on a single page. Each lower plot displays N_p , N_2 , N_1 , and A vs time, scaled in units of the mean lifetime λ^{-1} of the upper state, with depth x as parameter. Each upper plot shows the common logarithm of the vector potential vs λt for five values of x (GRASER, Figs. 1-16), or vs x for five values of t (GRAZER, Fig. 17); distances are scaled in units of μ^{-1} , the nonresonant absorption length of the system. The values of all relevant parameters are also displayed along the right of each plot. Note that the isomer ratio of ^{83}Kr was assumed to be unity, for reasons given in the introduction.

Definitions and units for all quantities can be found in Appendix A.

B. Parameters Varied

Five parameters were varied in the course of this study: 1) S , the number density of fast neutrons injected at $t = 0$, to determine the minimum source strength required to produce an observable effect; 2) N_p/N_h , the ratio of the concentrations of krypton and beryllium, in order to study the effect of using a beryllium host crystal; 3) the linebreadth Γ of the transition, to study the effect of inhomogeneous line broadening; 4) the absolute temperature T of the system, in selected cases, to illustrate the effect of heating by the pump, which reduces the recoilless fraction; 5) the nonresonant absorption coefficient, to study the effect of anomalous transmission (photon channeling or the Borrmann effect^{15,16}). Results of the first four parameter variations are displayed in Figs 1-17; of the fifth, in Table I.

C) Effect of Varied Neutron Burst Intensity

Figures 1-6 display the effect of varied source strength in a crystal of solid krypton (Debye temperature 72 K). The source strength S was varied between $5.0(20) \text{ cm}^{-3}$ and $1.0(24) \text{ cm}^{-3}$, with four intermediate values. The linebreadth was held constant at 10 times the natural breadth, and the temperature was assumed to be essentially absolute zero ($T \ll \theta$).

As the source strength was increased, the parent became depleted and, therefore, the populations of the upper and lower states developed more and more rapidly. At $S = 5.0(22)$, the parent was essentially depleted at 1.4 mean lifetimes after the burst; at $S = 1.0(24)$, this occurred after 0.4 mean lifetimes. With increasing source strength, equalization of the upper and lower state densities occurred at earlier times because the upper state was being generated more rapidly and, by its decay, feeding the lower state at a faster rate. This caused the integral of the inversion density, and thus the function Q , first to rise faster and then to fall off sooner as S was increased. At source strengths of $5.0(22)$ and above, Q became negative after fewer than 2.4 mean lifetimes and it is expected that the vector potential would thereafter be oscillatory, owing to predominance of resonant absorption,^{7,13} but its values are beyond the range of the logarithmic plot.

At low source strengths, the vector potential displayed normal exponential decay and attenuation, but, as S increased, departures from exponential behavior became increasingly evident at depths of four or five absorption lengths into the crystal. With $S = 1.0(22)$, $A(x,t)$ was nearly independent of depth x at $\lambda t = 1.4$; and, with $S = 5.0(22)$, actual gain was achieved, the vector potential

increasing with increasing depth. For the case $S = 3.0(21)$, the space integral of the vector potential was found to crest after 1.44 mean lifetimes at a value of 1.704 (Fig. 7).

D. Effect of a Beryllium Host

Use of a rigid, low-absorption host material has been suggested by Gol'danskii and Kagan¹⁷ as well as by Bowman.² In order to evaluate the merits of this suggestion, four different concentrations of krypton in beryllium were studied with atom ratios of beryllium to krypton ranging from 10 to 1000. These changes, shown in Figs. 7-9, of course, affect only the ordinate scale of the pumping curves.

When the concentration was changed from that of solid krypton (Fig. 4) to a beryllium to krypton ratio of 10 (Fig. 7), the vector potential displayed a noticeable increase. This was mainly due to an increase in the Debye temperature from 72 to 474 K, which raised the recoilless fraction from 0.87 to 0.98. The value of 474 K for the Debye temperature of the beryllium-krypton system was estimated using the equation¹⁸

$$\theta(\text{Be-Kr}) = \theta(\text{Be}) \left(\frac{M_{\text{Be}}}{M_{\text{Kr}}} \right)^{1/2}, \quad (3)$$

where M_{Be} and M_{Kr} are the atomic masses of beryllium and krypton, respectively.

As the concentration of ^{82}Kr in beryllium was reduced, amplification fell, owing to the smaller density of emitting nuclei. Although the total nonresonant absorption coefficient also declined, from 231 cm^{-1} for pure solid krypton to 2.56 cm^{-1} in the 1:1000 mixture, this was not sufficient to offset the reduction in concentration of the emitting nuclei; in other words, the parasitic absorption of the host material is too high. At 1000:1 dilution, gain was eliminated even at a source strength of 5.0(22).

Note that as the krypton/beryllium ratio is changed the absolute depth corresponding to a given value of μx in Figs. 7-9 changes because of the change in μ .

E. Effect of Inhomogeneous Line Broadening

For one selected source strength, the inhomogeneous broadening factor of the gamma-ray line was varied between 20 and 1 (natural line) in pure krypton (Figs. 10-14). As expected, the pumping curves were not affected by this change. We did not introduce the more realistic, but complicating, feature of a time-dependent linebreadth associated with accumulating lattice damage.¹⁹

The vector potential rose steadily above the exponential decay curve as the gamma-ray line was narrowed, and, for a source strength of 3.0(21), net gain was achieved with a twofold broadened line. At this linebreadth, the space integral of the intensity crested at 9.5 after 1.92 mean lifetimes.

As the broadening was progressively reduced, the maximum departure of the vector potential from pure exponential decay occurred at increasingly late times (namely, at $\lambda t = 1.2$ for $\Gamma = 20\lambda$, vs $\lambda t = 2.2$ for $\Gamma = \lambda$). This illustrates the principle, noted in the Introduction, that a resonant system requires time, of the order of the reciprocal linebreadth, to respond.^{7,10-12} However, the shift due to this effect is not as great as the time lag due to neutron moderation.⁷

If these calculations are repeated, first with the resonance integral set equal to zero and, then, with the thermal neutron capture cross section set equal to zero, one finds that essentially all of the excitation in this isotope is due, not to thermal, but to resonance neutron capture. The major part of the observed time lag in wave development is for neutron moderation to the velocity of the capture resonance. When the source intensity is sufficient for significant excitation, the parent becomes depleted and the states excited by resonance capture decay before the neutrons thermalize; moreover, the maximum neutron flux is higher at resonance than at thermal energies.

F. Effect of Increased Temperature

In preceding sections, a temperature of 0 K was assumed for ease of computation, even though this is not a reasonable experimental condition since the pumping process inevitably heats the solid.^{17,19} Therefore, the case of pure krypton with a source strength of 3.0(21) and tenfold broadened line was re-examined, assuming now a temperature of 100 K, below the 116-K melting point of krypton. Figures 15 and 16 show the results. The Mössbauer fraction was reduced from 0.87 to 0.07, thereby lowering the asymptotic cross section from 1.20(-19) cm² to 9.12(-21) cm². This caused $Q(t)$ to drop, in turn restoring $A(x,t)$ essentially to normal exponential decay. The space integral of the intensity was therefore reduced to a maximum value of only 1.035.

Elevated temperature would, of course, have less devastating effect on the beryllium-krypton system because of the higher Debye temperature.

G. Effect of Reduced Nonresonant Absorption

It has long been known that photoelectric absorption is greatly reduced for one of the wave modes which form in a single crystal as a result of Bragg diffraction,²⁰ and that this effect would greatly reduce the inversion density required for amplification by stimulated emission^{15,16,21} of multipole orders higher than electric dipole. The possibility that the Borrmann effect could persist in a solid subjected to fast neutron bombardment seems very remote. However, reduction of the neutron source strength would reduce the amount of damage, so it is interesting to consider this possibility.

If one assumes that the Borrmann effect can occur in pure solid krypton, then the neutron source intensity S is reduced, for a given set of the other parameters, in the same ratio as the gamma-ray absorption coefficient.

If, in view of the results of the dilution study, we attempt to reduce the parasitic absorption of a beryllium host by invoking the Borrmann effect (ignoring the fact that krypton will probably occupy interstitial sites in the beryllium host, tending to destroy the pure modes essential to the effect) we find only a slight improvement: in the case 1:1000 krypton:beryllium with $S = 3(21)$, the integrated intensity from a filament 5 nonresonant absorption lengths long was approximately doubled when the host absorption cross section was reduced by a factor of 100; note, however, that the total absorption coefficient of the system was reduced only by a factor of two. We ascribe the twofold increase in integrated intensity mainly to the doubled amount of graser material.

II. DISCUSSION

A. Limitations of the Analysis

This study has ignored several processes by which the populations of graser states may be affected. Neutron capture and inelastic scattering by nuclei in the graser states and in the 1.86-h state could redistribute the populations. It was unfortunate that we lacked sufficient data to include their effects; however, we doubt that the qualitative conclusions relative to ^{83}Kr would be altered by including additional neutron interaction terms.

Gamma radiation from the neutron source (which, although unspecified, must obviously accompany a nuclear explosion) was assumed to be without influence on the pumping. If the parent material included ^{84}Kr and if the pumping radiation were sufficiently intense, the (γ, n) reaction would contribute to

the pumping with a different time dependence, but this would be at the expense of the neutron-excited component. In general, photon pumping is not promising because of the narrow bandwidths of nuclear absorption resonances at low energies and of the relatively low cross sections for photonuclear reactions in comparison with neutron capture cross sections.

The model we have employed for calculating the neutron flux is, of course, highly idealized. It is probably satisfactory in predicting the time dependence if not the magnitude of the flux. Neglecting depletion of the neutron gas by capture in the parent, and assuming negligible neutron leakage from the necessarily finite system both tend to overestimate the flux and, therefore, to underestimate the source strength. The fact that, in hydrogen, neutrons moderate in the vicinity of their first points of collision has been taken to justify the assumption of uniformly distributed density of injected neutrons, but no account was taken of the spreading in time of neutrons which must enter a finite system from a necessarily external source of finite size.

We have used the numerical value of the scattering probability for water⁹ in a formula which was derived for unbound hydrogen.⁸ For other moderators of different hydrogen density the value should, of course, differ, and the time scale of wave development would be modified. Extremely high compression of the moderator would reduce the moderation time scale, so that population inversion could develop more rapidly, reducing the mismatch to the transition lifetime. This consideration deserves further study, in view of current attempts to achieve thermonuclear fusion by laser-driven compression.

B. Limitations of ^{83}Kr

In order to produce a detectable effect of stimulated emission in ^{83}Kr , we find that the fast neutron source strength is far higher than that which was estimated for ^{181}Ta by Gol'danskii and Kagan¹⁷ on a much simpler (and more optimistic!) model. With a tenfold-broadened line, source strengths above $1.0(22) \text{ cm}^{-3}$ are needed in pure krypton and still greater in dilute systems; the enormity of this number is realized when one considers that it represents 16 milligrams of neutron gas per gram of moderator!

Narrower linebreadth and lower photoelectric absorption would both operate to reduce this requirement. But it is unrealistic to hope for a much narrower line in the use of this isotope. Fast neutron damage is observed to give appreciable line broadening at much lower fluences than are involved in this problem in studies of materials with much higher Debye temperature and,

therefore, greater lattice stability.¹⁹ It is clearly unrealistic to hope for a Borrmann effect in pure solid krypton, because the effect is so extremely sensitive to lattice perfection. Raising the Debye temperature with a beryllium host is defeated by parasitic absorption, although it ameliorates somewhat the effects of heating.

The facts that the ^{83}Kr transition is to a stable ground state, that its lifetime is short in comparison with the neutron moderation time, and that the value of the resonance integral is not extremely high, all mitigate against a high value for the argument of the function upon which amplification depends.⁷

Therefore, unless the assumptions of this study are drastically altered, it appears that ^{83}Kr is not a likely candidate for neutron-pumped lasers. Its lifetime, production cross sections, and Debye temperature are too low, and its nonresonant absorption cross section is too high to permit one to benefit from the other attractive properties enumerated in the Introduction when it is used in a system excited by capture of neutrons moderating from a short burst at any reasonable level of intensity.

C. Indicated Directions to Search for Graser Candidates

Although this somewhat restricted analysis does not completely rule out the use of ^{83}Kr , it does indicate that more promising candidates are needed and it points out characteristics which should be considered in the search. They include, in addition to the obvious ones contributing to high resonance cross section;

- 1) unstable lower state, although not much shorter lived than the upper isomer;⁷
- 2) strong neutron capture resonance at high neutron energy, such that the neutron moderation time is less than the lifetime of the transition;
- 3) capture state spins much nearer to upper than to lower isomeric state;
- 4) low nonresonant absorption coefficient at the transition energy (either low atomic number or favorable location near an absorption edge); and
- 5) high Debye temperature in the pure element.

If the Borrmann effect is to be invoked, the neutrons should be well moderated before they reach the graser body, in order to avoid lattice damage from the fast neutrons. Thus one is forced to use thermal neutron capture and the transition must then be of much longer lifetime than any hitherto considered; inhomogeneous linebreadth will then be the limiting factor. Hopefully,

a crystal which manifests the Borrmann effect may exhibit relatively little inhomogeneous broadening from defects.²²

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FIGURES 1-6

EFFECT OF SOURCE STRENGTH

Fig. 1. $S = 5.0(20)$, pure krypton, tenfold-broadened line, $T = 0$.

Fig. 2. $S = 1.0(21)$, pure krypton, tenfold-broadened line, $T = 0$.

Fig. 3. S = 3.0(21), pure krypton, tenfold-broadened line, T = 0.

Fig. 4. $S = 1.0(22)$, pure krypton, tenfold-broadened line, $T = 0$.

Fig. 5. $S = 5.0(22)$, pure krypton, tenfold-broadened line, $T = 0$.

Fig. 6. $S = 1.0(24)$, pure krypton, tenfold-broadened line, $T = 0$.

FIGURES 7-9

EFFECT OF DILUTION

Fig. 7. $S = 1.0(22)$, 1:10 Kr:Be, tenfold-broadened line, $T = 0$.

Fig. 8. $S = 1.0(22)$, 1:100 Kr:Be, tenfold-broadened line, $T = 0$.

Fig. 9. $S = 1.0(22)$, 1:1000 Kr:Be, tenfold-broadened line, $T = 0$.

FIGURES 10-14

EFFECT OF LINE BROADENING

Fig. 10. $S = 3.0(21)$, pure krypton, twentyfold-broadened line, $T = 0$.

Fig. 11. $S = 3.0(21)$, pure krypton, tenfold-broadened line, $T = 0$.

Fig. 12. $S = 3.0(21)$, pure krypton, fivefold-broadened line, $T = 0$.

Fig. 13. $S = 3.0(21)$, pure krypton, twofold-broadened line, $T = 0$.

Fig. 14. $S = 3.0(21)$, pure krypton, natural line, $T = 0$.

FIGURES 15-16

EFFECT OF TEMPERATURE

Fig. 15. $S = 3.0(21)$, pure krypton, tenfold-broadened line, $T = 0$.

Fig. 16. $S = 3.0(21)$, pure krypton, tenfold-broadened line, $T = 100$.

FIGURE 17

SPATIAL VARIATION

Fig. 17. $S = 1.0(22)$, pure krypton, tenfold-broadened line, $T = 0$. Upper plot displays the variation of vector potential with distance.

TABLE I
NORMALIZED SPACE INTEGRAL OF AXIAL INTENSITY FROM A LONG FILAMENT

<u>LAMBDA $\rightarrow$ T</u>	<u>SPACE INTEGRAL OF INTENSITY (NORMALIZED)</u>
0.000	1.0000
0.096	1.0020
0.192	1.0147
0.288	1.0526
0.384	1.1312
0.480	1.2664
0.576	1.4761
0.672	1.7791
0.768	2.1898
0.864	2.7058
0.960	3.2921
1.056	3.8697
1.152	4.3239
1.248	4.5384
1.344	4.4444
1.440	4.0554
1.536	3.4636
1.632	2.7981
1.728	2.1741
1.824	1.6600
1.920	1.2746
2.016	1.0026
2.112	0.8161
2.208	0.6878
2.304	0.5973
2.400	0.5310

S = 1.0(22), pure krypton, tenfold-broadened line, T = 0,
corresponding to the plot shown in Fig. 17.

APPENDIX A

TABLE A-1 - INPUT PARAMETERS USED

Parameter	Meaning	Units	Value
S	Number density of fast neutrons injected at $t = 0$	cm^{-3}	5.0(20)-1.0(24)
N_p	Number density of ^{82}Kr nuclei at $t = 0$	cm^{-3}	1.2(20)-2.5(22)
N_h	Number density of the host	cm^{-3}	0-1.2(23)
t_2	Lifetime of upper state	s	1.4(-7)
t_1	Lifetime of lower state	s	1.0
μ_p	Nonresonant absorption cross section of Kr	b	9241.5
μ_h	Nonresonant absorption cross section of Be	b	12.1-0.121
E_γ	Energy of the gamma transition	keV	9.3
I_2	Upper state angular momentum $\div \hbar$		7/2
I_1	Lower state angular momentum $\div \hbar$		9/2
η	Isomer ratio (ratio of number of upper states formed by neutron capture to number of lower states so formed)		1.0
ξ	Fraction of all captures that form either of the graser levels		0.33
E_o	Effective energy of single neutron capture resonance in ^{82}Kr	eV	40
I_γ	Resonance integral of the capture resonance in ^{82}Kr	b	200
σ_{th}	Thermal neutron capture cross section in ^{82}Kr	b	45
θ	Effective Debye temperature of the system	K	72-474
α	Internal conversion coefficient		19.6
A	Atomic mass number		83
B	Factor by which the gamma line is inhomogeneously broadened		1-20
Σ_s	Macroscopic neutron elastic scattering cross section of the moderator	cm^{-1}	1.4
t_n	Neutron lifetime in the moderator	s	1.0(-5)
T	Absolute temperature of the system	K	0-100
β	Branching ratio of the isomeric transition (0-1) (identically 1 for Mossbauer isomers)	-	1

TABLE A-2 - PARAMETERS CALCULATED BY CODES

Parameter	Meaning	Formula	Units	Value
μ	Nonresonant absorption coefficient of system	For Kr-Be: $\mu = (N_h - N_p) \mu_h + N_p \mu_p$ For pure Kr: $\mu = N_p \mu_p$	cm^{-1}	2.56 - 231
R	Recoil energy	$5.37(-4) E_\gamma^2 / A$	eV	5.596(2)
f	Mössbauer fraction	$\exp \left\{ \left[-1.5 R / k\theta \right] \left[1 + (\pi T / \theta)^2 \right] \right\}$		0.066-0.979
σ_γ	Asymptotic cross section for stimulated emission or resonant absorption	$\frac{2.44(-16) E_\gamma^{-2} f \beta t_1}{(1+\alpha)(t_1+t_2) B}$	cm^2	9.12(-21) - 1.20(-18)

APPENDIX B Basic Equations

The pumping equations solved by GRASER and GRAZER are:

$$\dot{N}_p(t) = -R(t)N_p(t)$$

$$\dot{N}_2(t) = \xi\eta R(t)N_p(t) - \lambda_2 N_2(t)$$

$$\dot{N}_1(t) = \xi(1-\eta)R(t)N_p(t) + \lambda_2 N_2(t) - \lambda_1 N_1(t)$$

$$\dot{Q}(t) = [N_2(t) - G N_1(t)] \sigma_Y \Gamma/4 ,$$

where

$N_2(t)$ is the number density of the upper state as a function of time;

$N_1(t)$ is the number density of the lower state as a function of time;

$Q(t)$ is the time integral of the inversion density multiplied by $\sigma_Y \Gamma/4$;

Γ is the total linebreadth of the gamma line;

$\lambda_2 = 1/\tau_2$ is the decay constant of the upper state;

$\lambda_1 = 1/\tau_1$ is the decay constant of the lower state;

G is the ratio of statistical weights of the two states.

The rate function $R(t)$ is defined as

$$R(t) = \int_0^\infty \sigma_{\text{cap}}(v) \phi(v,t) dv \exp(-t/t_n) ,$$

where $\phi(v,t)$ is given in Eq. (1). The neutron capture cross section is described by a single effective neutron capture resonance at energy E_0 with resonance integral I_Y and a $1/v$ contribution whose thermal capture cross section is σ_{th} .

Thus we have

$$R(t) = \exp(-t/t_n) \left[\sigma_{\text{th}} v_{\text{th}} + I_Y v_r (v_r \Sigma_s t)^2 \exp(-v_r \Sigma_s t) \right] S$$

The expression used for the vector potential is that due to Baldwin and Suydam:⁷

$$A(x,t) = \exp \left[-(\mu x + \Gamma_{\text{tot}} t)/2 \right] \left[\text{BESSL} + \Delta \Gamma \int_0^t \exp(\Delta \Gamma t') \text{BESSLI} dt' \right]$$

$$\text{where } \text{BESSL} = I_0 \left[2\sqrt{X Q(t)} \right] \text{ for } Q(t) \geq 0$$

$$= J_0 \left[2\sqrt{-X Q(t)} \right] \text{ for } Q(t) < 0 ,$$

$$\text{and } \text{BESSLI} = I_0 \left\{ 2\sqrt{X [Q(t) - Q(t')]} \right\} \text{ for } Q(t) \geq Q(t')$$

$$= J_0 \left\{ 2\sqrt{X [Q(t') - Q(t)]} \right\} \text{ for } Q(t) < Q(t') .$$

$\Delta\Gamma$ is the excess halfbreadth of the gamma line given by $\Delta\Gamma = (\lambda_2 + \lambda_1)(B - 1)/2$.

The space integral of the intensity (normalized) is given by

$$\left[\mu \exp(\lambda_2 t) / (1 - \exp(-X_{\max})) \right] \int_0^{X_{\max}} |A(x, t)|^2 dx .$$

t_n , the neutron die-away time in the moderator, was taken to be so long, in this work, that the factor $\exp(-t/t_n)$ could be taken as unity.