shifts at r represent the asymptotic phase shifts for a potential which is set to zero for all values of the interatomic separation greater than r. Thus the influence of the potential is reflected in a change in the phase shift. The results for the Bruch and McGee[4] potential FDD-1 with and without retardation effects are shown in Fig. 1. Only the phase shift $\delta_2(r)$ is shown since the contribution to the cross section from the other phase shifts is less than 1% of that due to δ_2. The potential is formed using the Bruch-McGee short-range potential up to 3.51 Å and long-range forms given by

$$V_{LR} = \begin{cases} -0.878(2/\pi)\tan^{-1}(248.7/r)r^{-6} - 2.375r^{-8}, & \text{(1a)} \\ -0.878r^{-6} - 2.375r^{-8}, & \text{(1b)} \end{cases}$$

with r in Å and V in eV. The expression in Eq. (1a) is with retardation and that in Eq. (1b) is without retardation. Calculations for the MDD-2 potential[4] with a joining radius of 3.683 Å have also been done with similar results.

In Fig. 2, the difference in the potential due to the retardation term and the corresponding differences in $\delta_2(r)$ are shown with an expanded scale. The significant change in the phase shift occurs between 3.5 and 8 Å with almost no effect due to the potential at larger radii. The change in the $l = 2$ asymptotic phase shift δ_2 accounts for about 3% changes in the total and differential cross sections since both are nearly proportional to $\sin^2\delta_2$ in this velocity range.

Although the calculations show little or no effect due to long-range retardation, they do show that the low-energy helium scattering results are extremely sensitive to very small differences in the intermolecular forces. Experiments are currently underway to measure these cross sections.

*Research sponsored by the U. S. Air Force Office of Scientific Research, Office of Aerospace Research, under Grant No. AFOSR-71-2070.

[1]J. Konrady and J. Sucher, Phys. Rev. Lett. 28, 74 (1972).

[2]J. P. Aldridge and R. H. Davis, Phys. Rev. Lett. 19, 1001 (1967).

[3]P. Bertoncini and A. C. Wahl, Phys. Rev. Lett. 25, 991 (1970).

[4]L. Bruch and I. McGee, J. Chem. Phys. 52, 5884 (1970).

[5]F. Calogero, *Variable Phase Approach to Potential Scattering* (Academic, New York, 1967).

Observation of Interference Effects in the Mössbauer Absorption Spectra of $E2$ and $E2/M1$ Transitions*

F. E. Wagner,† B. D. Dunlap, and G. M. Kalvius†
Argonne National Laboratory, Argonne, Illinois 60439

and

H. Schaller, R. Felscher, and H. Spieler
Physik-Department, Technische Universität München, 8046 Garching, Germany

(Received 4 January 1972)

In a transmission geometry, dispersion terms were observed in the shapes of the Mössbauer resonance lines of pure $E2$ and mixed $E2/M1$ transitions. For the pure $E2$ transitions studied, the relative magnitude of these terms, which are attributable to interference effects, is typically 1.5% of the depth of the absorption line. We discuss possible consequences of the resulting asymmetric line shape for isomer-shift measurements.

The interference of Mössbauer and Rayleigh scattering has long[1] been established. More recently, interference effects between conversion electrons emitted after nuclear resonance absorption and photoelectrons have been observed in Mössbauer studies[2-4] of the outgoing electrons. These phenomena can[5-8] give rise to a dispersion term in the Mössbauer absorption cross section σ_0, which may be obtained by properly summing over all final states and integrating over all di-

rections of the outgoing photons and electrons. For a thin absorber, the shape of a single-line transmission Mössbauer spectrum can then be written as[6,7,9]

$$N(v)/N(\infty) = 1 - \epsilon(1 - 2\xi X)/(1 + X^2), \quad (1)$$

with $X = 2(v - S)/W$. Here $N(v)$ is the intensity transmitted at a Doppler velocity v, S is the isomer shift between the source and the absorber, W is the full linewidth at half-maximum, and ϵ is the depth of the absorption dip at resonance. The parameter ξ determines the relative magnitude of the dispersion term and is defined as in Ref. 7. This definition differs in the sign from that of Ref. 9.

Since the interaction of low-energy γ rays with atomic electrons has predominantly electric dipole character, one expects[6,7] large dispersion effects in the absorption spectra of Mössbauer transitions of $E1$ multipolarity. Indeed the first observation of a dispersion term in a Mössbauer absorption spectrum was reported[10] for the 6.2-keV $E1$ transition in ^{181}Ta, for which the dispersion effects are particularly large. Subsequently similar, but smaller, dispersion effects were shown[9,11] to exist for other $E1$ resonances. For nuclear transitions with multipolarities other than $E1$, the integration over all directions of the outgoing radiations was expected[12] to result in a virtual cancelation of the dispersion term in σ_0.

In this Letter we show, by a careful analysis of the line shapes for fourteen different Mössbauer transitions, that dispersion terms are clearly present in *absorption* spectra of γ rays with $E2$ or mixed $E2/M1$ character. This fact has to be taken into account in cases where the position of such absorption lines has to be measured with great accuracy.

The results of our experiments are summarized in Table I. All measurements were performed with single-line resonances. Sources and absorbers were cooled to 4.2 K, except for the case of ^{166}Er and ^{236}U, where a temperature of 45 K was used in order to avoid magnetic hyperfine splittings. Nevertheless the ^{166}Er Mössbauer line is strongly broadened, conceivably due

TABLE I. Summary of experiments performed to study the shapes of Mössbauer absorption lines of various isotopes. d is the amount of resonant isotope per unit absorber area, $W_0 = 2\hbar/\tau$ is the minimum observable linewidth (Refs. 13 and 14), W is the experimental linewidth, and ξ is the dispersion parameter.

Isotope	E_γ [keV]	Multipolarity	Source	Absorber	d [mg/cm^2]	W_0 [mm/s]	W [mm/s]	$\xi \cdot 100$
^{166}Er	80.6	E2	^{166}Ho in $HoAl_3$[c]	$ErAl_3$[c]	10	1.87	6.85±0.07	-1.60±0.19
^{170}Yb	84.3	E2	^{170}Tm in $TmAl_3$[c]	$YbAl_3$[c]	15	2.08	2.45±0.03	-1.70±0.38
^{180}Hf	93.3	E2	^{180}Ta in Ta metal	$HfZn_2$	22	1.95	2.93±0.07	-1.82±0.48
^{182}W	100.1	E2	^{182}Ta in Ta metal	W metal	14	2.00	2.78±0.03	-1.71±0.14
^{183}W	99.1	E2	^{183}Ta in Ta metal	^{183}W metal[b]	20	3.32	4.68±0.05	-1.25±0.17
^{184}W	111.1	E2	^{184}Re in Pt metal	^{184}W metal[b]	20	1.95	2.60±0.04	-1.53±0.29
^{186}W	122.5	E2	^{186}Re in Pt metal	^{186}W metal[b]	40	2.21	3.19±0.04	-2.09±0.36
^{186}Os	137.2	E2	^{186}Re[a]	Os metal	10	2.37	2.93±0.04	-1.02±0.25
^{188}Os	155.0	E2	^{188}Re[a]	Os metal	27	2.49	3.34±0.12	-1.51±0.49
^{236}U	45.3	E2	^{240}Pu in PuO_2	$^{236}UO_2$[b])	60	26.3	46.7±1.1	+0.25±0.75
^{99}Ru	90.0	E2+37%M1	^{99}Rh in Ru metal	Ru metal	20	0.147	0.253±0.004	-0.33±0.32
^{171}Yb	66.7	M1+49%E2	^{171}Tm in $ErAl_3$[c]	$^{171}YbAl_3$[b] [c]	15	5.12	5.99±0.06	-1.00±0.14
^{191}Ir	129.5	M1+14%E2	^{191}Os in Os metal	Ir metal	75	22.5	25.7±0.3	-0.50±0.12
^{183}W	46.5	M1+0.6%E2	^{183}Ta in Ta metal	W metal	4	31.8	41.6±0.4	-0.05±0.06

[a]The results for ξ are weighted averages of data taken with sources of ^{186,188}Re in Re, Rh, V, and W. The given linewidths are those obtained with rolled foils of Re metal as source matrices.

[b]These absorbers were prepared from enriched isotopes.

[c]The materials specified as (rare earth)Al_3 were produced by alloying 10 wt% of the rare earth with Al. The samples obtained consisted essentially of a mixture of (rare earth)Al_3 and Al.

to spin-relaxation effects. In most cases, cubic source and absorber matrices could be chosen in order to avoid line asymmetries due to unresolved quadrupole splittings. The quadrupole splittings in the hexagonal metals Ru, Os, and Re are too small to cause a serious line broadening except, perhaps, in the case of the narrow ^{99}Ru line. Here, however, the quadrupole splitting will not give rise to a line asymmetry since the quadrupole hyperfine pattern consists of a virtually symmetric doublet[15] and, moreover, since the splitting is identical in the source and the absorber. We may mention that Mössbauer measurements[10,16] with the 6.2-keV resonance of ^{181}Ta very sensitively demonstrate the absence of quadrupole effects which could influence the line shapes in our measurements with ^{182,183}W. The effective absorber thicknesses $t = fn\sigma_0$ (with f the Debye-Waller factor and n the number of resonant nuclei per unit absorber area) were made no larger than about $t = 2$. Hence our results for the line asymmetries are but very little affected by the enhancement that is expected[9,11] for thick absorbers.

Various Mössbauer spectrometers have been used in our investigation. Either the source or the absorber was moved sinusoidally by an electromechanical velocity drive mounted on top of the helium cryostat. The spectra were recorded in a multichannel analyzer operated in the multiscaler mode. The maximum velocity was usually chosen at least 5 times larger than the full experimental linewidth W at half-maximum to make sure that the observed line asymmetries were not caused by small slopes of the nonresonant baseline. The ξ values listed in Table I are weighted averages of several independent measurements and were obtained by fitting line shapes, according to Eq. (1), to the experimental data.

The nine pure $E2$ transitions studied in Er, Yb, Hf, W, and Os cover the energy range from 80 to 155 keV. The results obtained for them reveal no systematic dependence of ξ on the γ-ray energy or the nuclear charge. With values near $\xi = -0.015$, these dispersion terms are nearly as large as those reported[9] for $E1$ transitions of comparable energy in Gd and Dy. For the 45.3-keV $E2$ transition in ^{236}U, however, our measurements indicate a much smaller ξ value. Small asymmetries are also observed for the mixed $E2/M1$ transitions studied.

The dispersion term in the Mössbauer absorption cross section can be understood[7] as a consequence of, and for pure multipole transitions is proportional to, the phase shift 2ξ, which resonantly scattered γ rays undergo as a result of induced electronic currents. Hannon and Trammell[5,8] have applied their general formalism for calculating such phase shifts to the case of the 90-keV transition in ^{99}Ru, and obtain[5,8,17] $\xi_{E2} = -0.0067$ and $\xi_{M1} = -0.0002$. These results show that smaller dispersion terms are to be expected for $M1$ transitions than for $E2$ transitions. For the asymmetry parameter ξ of a mixed transition, one can write $\xi = (\xi_{M1} + \delta^2\xi_{E2})/(1+\delta^2)$, where δ^2 is the $E2/M1$ mixing ratio. From the calculated ξ values, and with $\delta^2 = 2.7 \pm 0.6$ (Ref. 13), we thus obtain $\xi = -0.0049$, in good agreement with our experimental value of $\xi(^{99}\text{Ru}, 90\text{ keV}) = -0.0033 \pm 0.0032$.

The only other case for which a theoretical result has been published[5,8] is that of the 73-keV γ rays of ^{193}Ir. The energy of this transition falls just below the K edge of Ir. The calculated phase shifts are much smaller[5,8,17] than for ^{99}Ru. This indicates that the main contribution to ξ arises from the K shell and may explain the smallness of the asymmetry for ^{236}U and also for the 46-keV line of ^{183}W. For the latter, however, the fact that it is of virtually pure $M1$ multipolarity alone could have explained the experimental result.

The isomer shifts S observed for the studied transitions were in no case larger than a few percent of the experimental linewidths. Their absolute values are irrelevant in the present context. It should, however, be pointed out that the shifts $S(\xi)$ resulting from least-squares fits of the line shape of Eq. (1) are different from the values $S(0)$ obtained by fitting merely a Lorentzian line. For small ξ, one can write $S(\xi) - S(0) = \lambda\xi W$, where λ is a proportionality factor. The fits of our experimental data yielded empirical values of λ between 1.0 and 1.2. Hence, for rotational transitions like those in ^{170}Yb or ^{182,184,186}W, the assumption of Lorentzian line shapes leads to apparent shifts between the source and the absorber which are comparable with the isomer shifts observed[18,19] for such transitions. In measurements of the relative isomer shifts between different single-line absorbers these apparent shifts should, however, cancel as long as the compared absorbers are thin or have at least equal effective thickness t. A cancelation of the influence of the interference term is also expected in cases where the relative isomer shifts between different sources are measured with the same absorber.

Interesting discussions with Professor Yu. M.

Kagan, Professor G. T. Trammell, Professor J. P. Hannon, and Dr. E. Zech are gratefully acknowledged.

*Work performed under the auspices of the U. S. Atomic Energy Commission and the Bundesministerium für Bildung und Wissenschaft of the Federal Republic of Germany.

†Permanent address: Physik-Department, Technische Universität München, 8046 Garching, Germany.

[1]P. J. Black, D. E. Evans, and D. A. O'Connor, Proc. Roy. Soc., Ser. A 270, 168 (1962).

[2]K. P. Mitrofanov, M. V. Plotnikova, N. I. Rokhlov, and V. S. Shpinel', Pis'ma Zh. Eksp. Teor. Fiz. 12, 85 (1970) [JETP Lett. 12, 60 (1970)].

[3]I. I. Lukashevich, V. D. Gorobchenko, V. V. Skylarevskii, and N. I. Filippov, Phys. Lett. 31A, 112 (1970).

[4]P. Steiner and G. Weyer, Phys. Lett. 36A, 201 (1971), and Z. Phys. 248, 362 (1971).

[5]J. P. Hannon and G. T. Trammell, Phys. Rev. Lett. 21, 726 (1968).

[6]Yu. M. Kagan, A. M. Afanas'ev, and V. K. Voitovetskii, Pis'ma Zh. Eksp. Teor. Fiz. 9, 155 (1969) [JETP Lett. 9, 91 (1969)].

[7]G. T. Trammell and J. P. Hannon, Phys. Rev. 180, 337 (1969) (see Sect. II, C).

[8]J. P. Hannon and G. T. Trammell, Phys. Rev. 186, 306 (1969).

[9]W. Henning, G. Baehre, and P. Kienle, Phys. Lett. 31B, 203 (1970).

[10]C. Sauer, E. Matthias, and R. L. Mössbauer, Phys. Rev. Lett. 21, 961 (1968).

[11]D. V. Gorobchenko, I. I. Lukashevich, V. V. Skylarevskii, and N. I. Filippov, Pis'ma Zh. Eksp. Teor. Fiz. 9, 237 (1969) [JETP Lett. 9, 139 (1969)].

[12]A. M. Afanas'ev and Yu. Kagan, Phys. Lett. 31A, 38 (1970).

[13]*Mössbauer Effect Data Index*, edited by J. G. Stevens and V. E. Stevens (IFI/Plenum Data Corp., New York, 1970).

[14]C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (Wiley, New York, 1967), 6th ed.

[15]O. C. Kistner, Phys. Rev. 144, 1022 (1966); G. Kaindl, W. Potzel, F. E. Wagner, U. Zahn, and R. L. Mössbauer, Z. Phys. 226, 103 (1969).

[16]G. Kaindl and D. Salomon, Phys. Lett. 32B, 364 (1970).

[17]G. T. Trammell and J. P. Hannon, private communication.

[18]W. Henning, Z. Phys. 217, 438 (1968).

[19]F. E. Wagner, H. Schaller, R. Felscher, G. Kaindl, and P. Kienle, in *Hyperfine Interactions in Excited Nuclei*, edited by G. Goldring and R. Kalish (Gordon and Breach, New York, 1971), Vol. 2, p. 603.

Observations of Gain by Stimulated Emission in the Werner Band of Molecular Hydrogen

Ronald W. Waynant
Naval Research Laboratory, Washington, D. C. 20390
(Received 29 November 1971)

Stimulated emission of vacuum ultraviolet radiation has been observed on two $Q1$ lines in the Werner band ($C^1\Pi_u \rightarrow X^1\Sigma_g^+$) of molecular hydrogen. These lines, 1161 and 1230 Å, contain 5 kW peak power and were produced by a traveling-wave discharge system. Using this excitation principle, evidence of gain has been obtained by observations of optimum velocity matching at pulse velocities less than c.

Observation and verification of amplification by the stimulated emission of radiation at photon energies above 10 eV have been made for the first time. The availability of such short-wavelength photons in an intense, directional beam is of immediate importance particularly in such fields as photochemistry, photofragment spectroscopy, and photoionization. Stimulated emission of these highly energetic photons has been produced in molecular hydrogen by rapid inversion of the vibrational levels of the $C^1\Pi_u$ excited electronic state with respect to the upper vibrational levels of the $X^1\Sigma_g^+$ ground state.

Speculation on the possibility of laser emission from the Werner band ($C^1\Pi_u$-$X^1\Sigma_g^+$ was first published by Bazhulin, Knyazev, and Petrash.[1] Figure 1 shows a greatly simplified energy-level diagram for the hydrogen molecule. A fast-rising current can produce an electron energy distribution which will preferentially populate the upper electronic states, inverting the vibrational levels with respect to the upper vibrational levels of the $X^1\Sigma_g^+$ ground state. A detailed rate-equation analysis of the Werner band for such a discharge was carried out by Ali and Kolb.[2] This analysis predicted lasing on several vibrational transitions with peak power density of about 20 kW/cm^3 for the strongest vibrational band. It