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This project concerns the demonstration of the feasibility of the tuning and perhaps even stimulation of nuclear radiation. Theory has indicated that anti-Stokes Raman upconversion of intense but conventional long wavelength sources of radiation produced by scattering from isomeric states of nuclear excitation could lead to significant sources of tunable γ-radiation characterized by the natural Mossbauer widths of the lines. This would result in lines with sub-Angstrom wavelengths and   CONTINUED OVER

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20. ABSTRACT Continued

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threshold depends upon the resolution of basic issues lying in
an interdisciplinary region between quantum electronics and nuclear
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ANNUAL SUMMARY REPORT

for the period

1 January 1985 through 31 December 1985

for

Office of Naval Research
Contract N00014-81-K-0653
Task No. NR 395-072

THE DEMONSTRATION OF THE FEASIBILITY OF THE
TUNING AND STIMULATION OF NUCLEAR RADIATION

Short Title: GAMMA RAY LASER

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PROJECT DESCRIPTION

This project concerns the demonstration of the feasibility of the tuning and stimulation of nuclear radiation. It repre-sents a critical line of investigation in our overall program concerned with the feasibility of a gamma ray laser.

Theory, supported by our experiments conducted under this contract, has indicated that anti-Stokes Raman upconversion of intense but conventional laser radiation produced by scattering from isomeric states of nuclear excitation could lead to signifi-cant sources of tunable γ-radiation characterized by the natural Mossbauer widths of the lines. Further computations have sug-gested that this type of coherent, as well as a type of incohe-rent, optical pumping could even lead to appreciable levels of inversion of the populations of nuclear levels that would be capable of supporting the growth of stimulated γ-ray intensities. Whether or not these processes can reach threshold depends upon the resolution of basic issues lying in an interdisciplinary region between quantum electronics and nuclear physics that have not been previously addressed. It is the purpose of this con-tract work to study these issues experimentally in order to guide the development of the technology and methods needed to exploit the enormous potential of this effect.

SCIENTIFIC PROBLEM

The viability of the concept for the tuning of γ-radiation by adding the variable energy of an optical photon produced by a tunable laser depends upon the existence in the nucleus of nearly

resonant intermediate states which would make it possible to dress the nuclear states with the laser photons. This produces the energy shifts. Transitions between the dressed states would then occur at the sum and difference frequencies characteristic of the nuclear transition plus or minus the energies of integral numbers of laser photons.

Whether the necessary arrangements of nuclear states do exist is the central issue being addressed in this contracted work. Surprisingly, such information is currently unknown because such potentially useful states would lie in the "blind spots" of the conventional techniques of nuclear spectroscopy. Normal Mossbauer spectroscopy provides enormous resolution but a tuning range that is inadequate, by order of magnitude, to support any possible study of transitions to the intermediate states of a multiphoton process. Conversely, crystal spectrometers provide broad tuning ranges but levels of resolution that miss by two orders of magnitude the threshold that would be necessary to separate the transitions to the initial and intermediate states. As a consequence, the ideal arrangement of nuclear energy levels needed for the Raman upconversion process could be a common occurrence that has gone unnoticed because of the inadequacies of conventional nuclear spectroscopy.

The critical problem in this research has two facets: 1) the development of an appropriate spectroscopic technique and 2) the search for a suitable medium for a large scale effect. The dressing of the nuclear states not only affects their energies but also changes their transition properties. Forbidden nuclear transitions should become allowed so that the metastability of

isomeric states would be "switched off" as the states were dressed. This would greatly enhance the prospects for stimulating the gamma ray transition in addition to rendering it tunable. It is the development of the investigative instrumentation and the verification of these predicted effects that comprise the scientific problem addressed by this contract research.

TECHNICAL APPROACH

For the resolution of the central issue of the existence of potentially useful intermediate states in a multiphoton upconversion of optical photons to γ-ray energies, it was first intended to demonstrate sum frequency generation in one case in which non-resonant intermediate states were known to exist. This was the case in which both initial and intermediate states were magnetic sublevels of the same nucleonic state and in which the transitions were mediated by a magnetic dipole, M1 operator. Experimental data reproduced in the literature suggested that such a process had already been, unknowingly, demonstrated for the generation of radiofrequency sidebands to Mossbauer transitions at the sum and difference frequencies. This has suggested the development of a new instrument, a Nuclear Raman Spectrometer designed to support the needed studies of nuclear structure with the precision of Mossbauer spectroscopy applied over a tuning range of energies lying considerably beyond the state-of-the-art at the time our work began. Pursuant to this goal we are conducting a conventional single-photon Mossbauer experiment in the presence of an intense radiofrequency field with measurement and parameterization of the conversion efficiency into the sum fre-

quency line to determine the practical limits on the ultimate linewidths and tuning ranges that can be achieved. This technique will then be used in a bootstrap approach to support a search for accidentally resonant intermediate states. By replacing the radiofrequency excitation with tunable higher frequencies, it is expected that the tuning range of Mossbauer spectroscopy can be extended by further orders-of-magnitude.

PROGRESS DURING THIS REPORTING PERIOD

During the past year a prototype Nuclear Raman Spectrometer was designed, constructed and has now begun to operate. Built from an Apple computer and a Wavetek frequency synthesizer it provides for the conduct of swept frequency spectroscopy of nuclear levels without the need to employ any mechanical effects for tuning that are required in conventional implementations of Mossbauer spectroscopy. The successes of the new NRS technique for nuclear spectroscopy indicate that a much higher resolution, by perhaps six orders of magnitude, can be achieved through a reasonable upgrade of the apparatus. If the range of tunability does extend to the ferromagnetic spin resonance (FSR) frequency, then it will be possible to construct a swept frequency device capable of continuously tuning over a range of 10^{11} linewidths, an enormous improvement in the state-of-the-art of nuclear spectroscopy.

Since development of the NRS device required most of the year, the considerable progress made in understanding the basic effects through which nuclear states are dressed by photons of the radiofrequency field had to be accomplished with tedious

manual adjustments of data acquisition parameters. Nevertheless, our database was substantially expanded this past year and we obtained the first NRS spectrum actually showing a better resolution than has been obtained with conventional Mossbauer spectroscopy. The particular system being examined was that of Fe-57 whose energy levels are reproduced here in Fig. 1 for convenience.

The concept behind the NRS demonstration completed this past year is shown in Fig. 2. The unsplit source energy corresponded to the wavelength shown in Fig. 2 by the heavy vertical line. At the lower mixing frequency shown in the upper data, the particular feature 5+ lies at longer wavelength than the source, while at higher frequency it lies at a lower value. It can be imagined that with a smooth change of frequencies between the two values, a point would be reached at which the transmission of γ-photons from the source would be suddenly reduced. The realization of this supposition is seen in Fig. 3. In addition to the absorption resonance resulting from the expected, 5+ line, another smaller feature appeared. It corresponded to 3++, the second order sideband to transition #3. It hints at the greater sensitivity of the NRS technique.

In any case, the point of the experiment was not to characterize the energy levels of ^{57}Fe but to demonstrate the effectiveness of this new NRS technique for nuclear spectroscopy in general, based on dressing the nuclear states. In practice, the method would probably be most useful with the rf field being replaced by a microwave or infrared photon field.
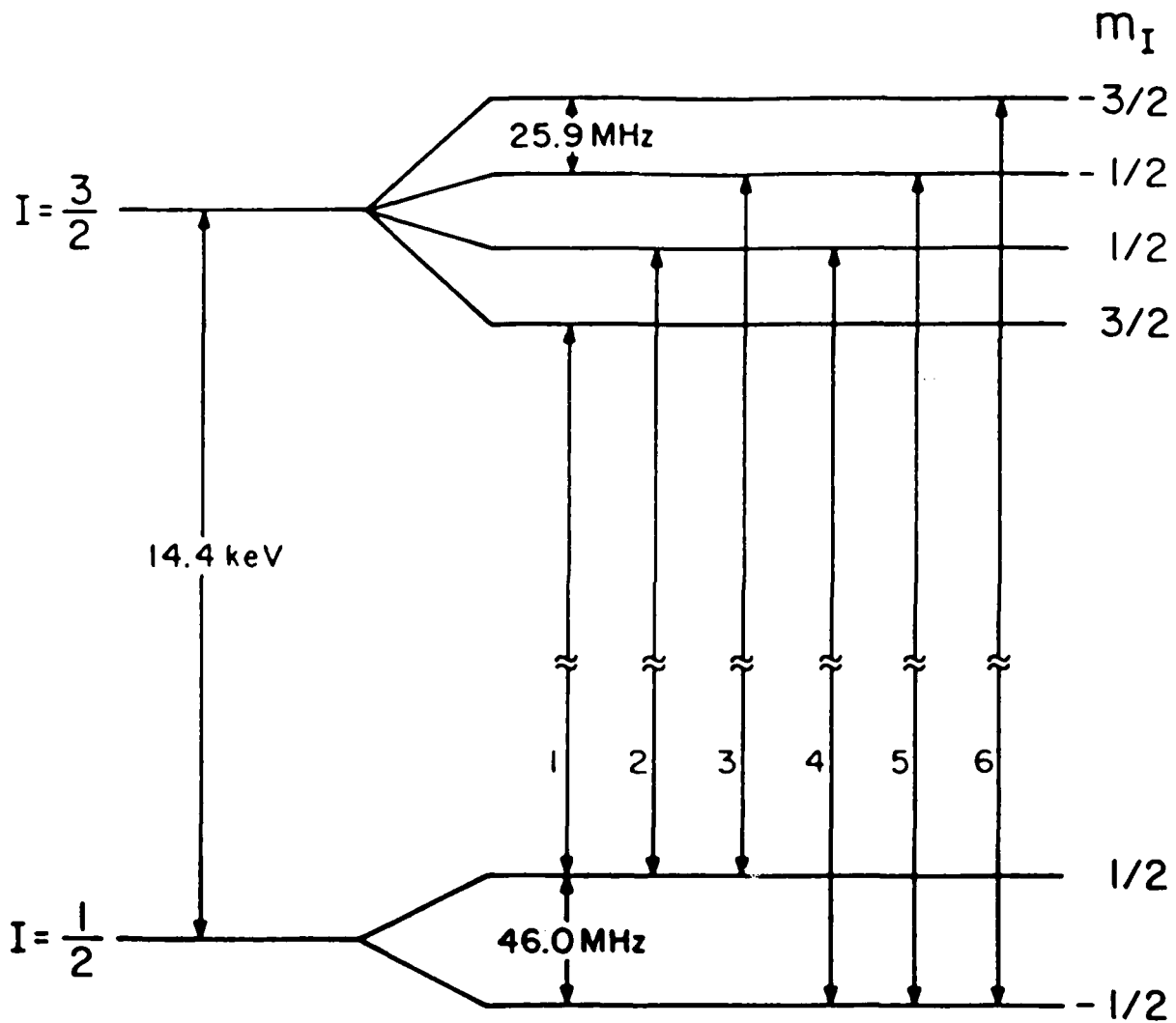
Figure 1

Energy level diagram of the Fe-57 demonstration nuclei used in
these experiments.
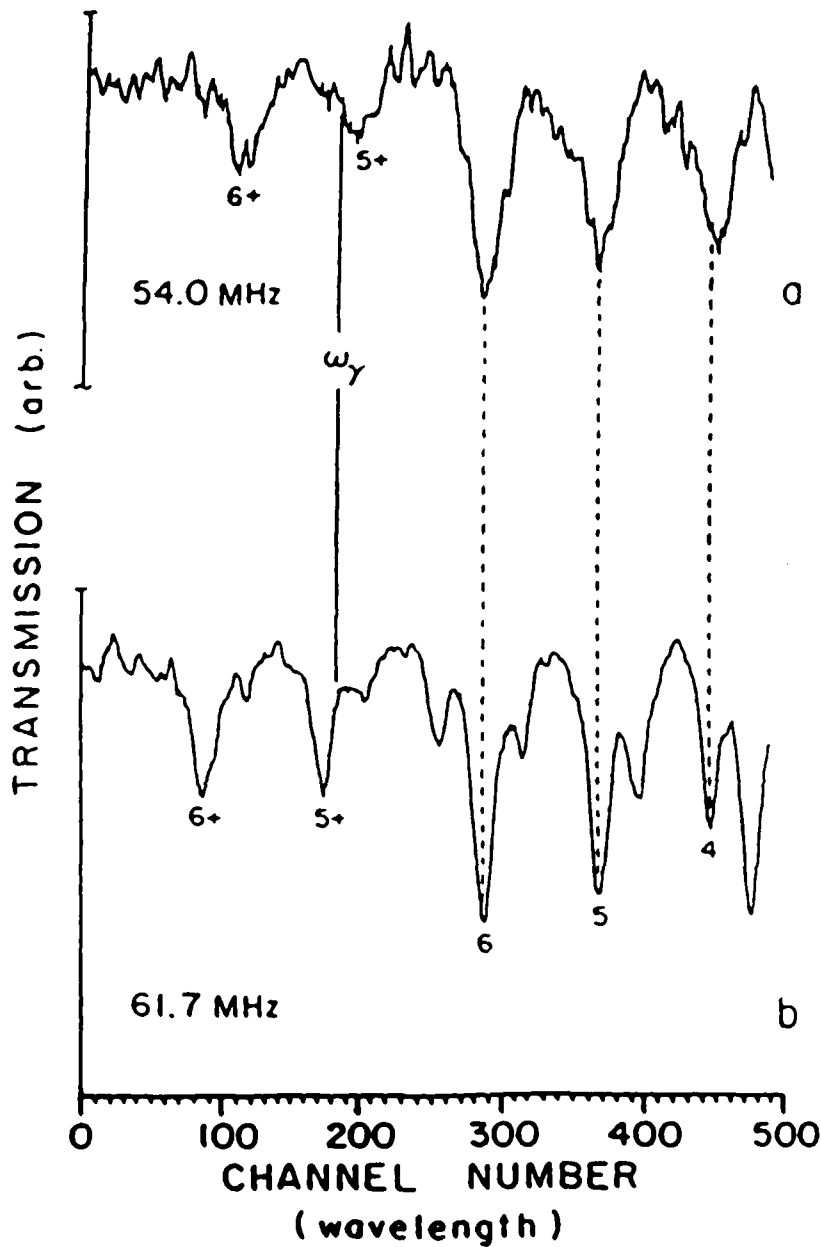
Figure 2

Two spectra that show that the result of increasing ω_{rf} is to push first-order sidebands away from the parents. Source frequency lies at ω_γ. The sidebands are identified by the digit corresponding to the parent line followed by one plus sign for each of photon added into the transition energy for that sideband.
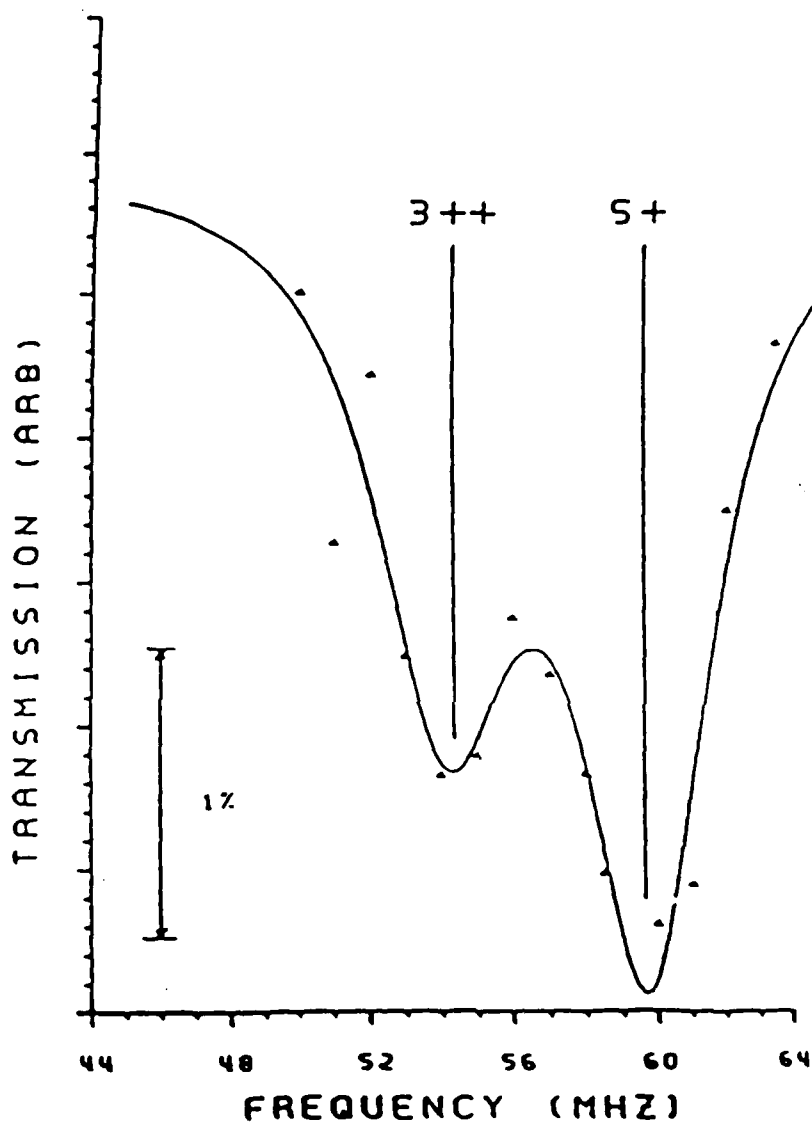
Figure 3

Gamma transmission intensity plotted as a function of rf photon frequency. The larger absorption line corresponds to the first order, sum-frequency sideband of the $|1/2,-1/2\rangle \rightarrow |3/2,-1/2\rangle$ nuclear transition of ^{57}Fe, while the smaller line corresponds to the second-order, sum-frequency sideband of the $|1/2,1/2\rangle \rightarrow |3/2, -1/2\rangle$ transition.

The details of this demonstration of the efficiency of our NRS technique are found in the reprint of the article, by B. D. DePaola, S. S. Wagal and C. B. Collins, "Nuclear Raman Spectroscopy," J. Opt. Soc. Am B2, 541 (1985), reproduced in the Appendices. Its utilization in a particularly demanding application is discussed in the preprint of the manuscript reproduced in the second position in the Appendices, by B. D. DePaola, S. S. Wagal and C. B. Collins, "Nuclear Raman Spectroscopy with No Mechanical Tuning," (pending).

Some of our most recent experiments along these lines have concerned the necessity of the dressed state model to the explanation of our phenomena. Our first experiments[1,2] demonstrating the ferromagnetic enhancement of coherent pump powers raised a considerable controversy in the traditional Mossbauer community as to whether the multiphoton model was necessary or merely sufficient to explain the accumulated results of some of their past experiments,[3,4,5] as well as ours. In those early works the nuclei were almost always arranged to be within or near ferromagnetic materials in order to enhance the fields being applied. Probably because of an initial similarity of the appearance of the sidebands to spectra that had been produced even earlier by introducing sinusoidally varying Doppler shifts with a mechanical device,[6] the rf. sidebands were attributed to periodic Doppler shifts caused by drumhead vibrations of the absorber foil that were driven by magnetostriction in the ferromagnetic parts and fortuitously amplified by mechanical resonances. Quantitative agreement between theory and experiment

was not obtained with that model, and ingenious refinements were introduced into the explanations. Vibration amplitudes were assumed to be microscopic and localized, but to be at least partially coherent across a sample. Broadband acoustic reson- ances in the sample and mounting were assumed and sometimes supported by experiments designed to enhance or damp them. De- spite the accretion of a large body of phenomenology supposed to prove a magnetostrictive-acoustic origin, quantitative agreement was never achieved with any computation of the amplitudes of the sidebands as either functions of applied power or of frequency. In contrast to the limitations of the traditional explanation, the dressed state model we developed in 1984 was reported[1] to give a good agreement with the measured dependence of sideband ampli- tude upon applied power in our experiments which were generically similar.

We recently completed a further consideration of the neces- sity of the dressed state explanation. In particular we ad- dressed two questions not previously considered: 1) whether the amplitude of the effect correlated with permeability as opposed to magnetostrictive constant of the samples involved and 2) whether quantum structure could be found in the dependence of the sideband amplitude upon frequency. Both questions were answered in the affirmative, results seemingly incompatible with any mag- netostrictive contribution to the origin of rf. sidebands on nuclear transitions.

The apparatus used in this experiment is shown in Fig. 4 where it can be seen to conform to the arrangement described in our previous work[2] and to the traditional pattern[4] for this type

of experiment, with one exception. The absorber arrangement used
in the first of the experiments just completed consisted of a
foil of non-magnetic stainless steel of 2.0 μm thickness that had
been enriched in ^{57}Fe. It was wrapped in a single foil of natu-
ral iron having 6.0 μm thickness, sandwiched in the graphite
plates and mounted in the foil carrier as shown in the inset of
Fig. 4. Data shown in Fig. 5a were obtained when this sample was
driven by 16.0 W of radio-frequency power applied to the resonant
circuit containing the coil, with the Q-factor arranged to be
37.5. Sidebands out to the fifth order can be clearly seen. For
comparison the spectrum obtained for the stainless foil in the
fields but without the driver is shown in Fig. 5c and the spec-
trum of the natural iron wrapper in the rf fields is shown in
Fig. 5d. In the latter case the overlapping and blending of line
and sideband components at this frequency can be seen to produce
no appreciable structure in agreement with classical observations.[4]

Nickel has a much larger magnetostrictive constant than
iron, the comparison being -30.0 to 5.0 in customary units.[7]
Data shown in Fig. 5b were obtained by replacing the iron foil
driver with a nickel foil of 2.0 μm thickness, wrapped around the
same absorbing core in the same manner. The literature[4] is
unanimous in attributing larger sidebands to thinner drivers so
the more substantial development of sidebands with the iron foil
driver seen in Figs. 5a and 5b is completely inconsistent with a
magnetostrictive origin of the effect. On the other hand, the
dressed state model of Mossbauer sidebands provides a mechanism
for transferring a large component of time-varying magnetization
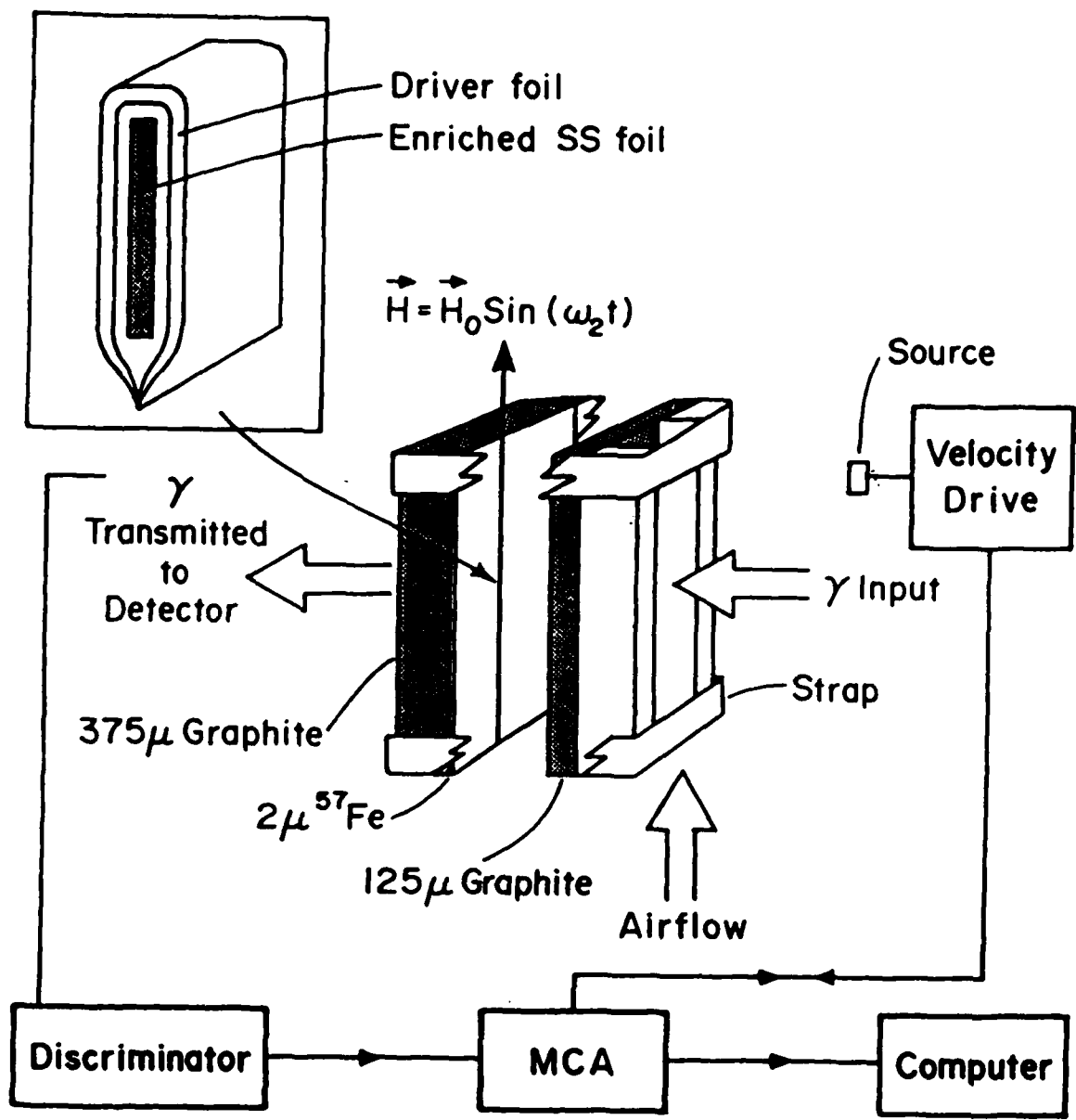
Figure 4

Schematic representation of the experimental apparatus, together
with detailed views of the foil carrier and layered composition
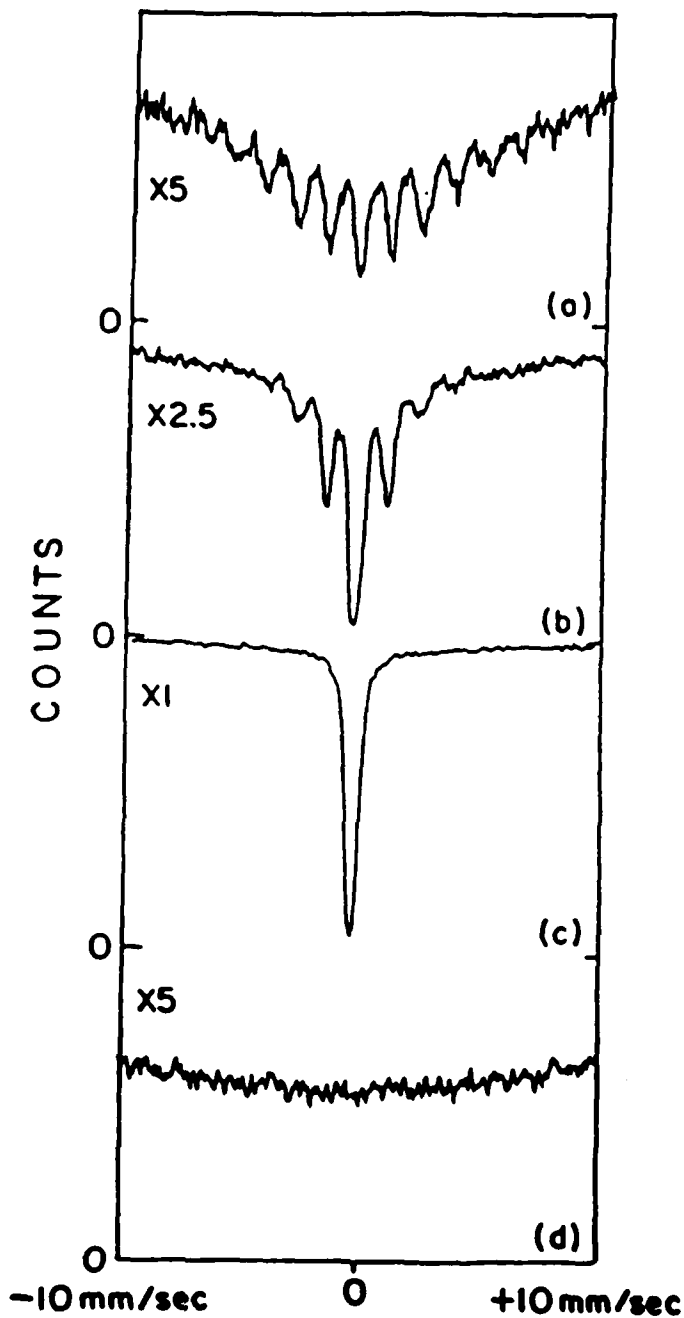of the absorbing foil.

Figure 5

Typical data obtained for several foil arrangements at 16.0 W of radiofrequency power at 15.0 MHz. Shown for each spectrum is a numerical multiplier recording the relative factor by which the data was multiplied for convenience in presentation.

(a) Nonmagnetic, enriched stainless steel foil of 2.0 μ thickness wrapped in a natural Fe foil driver of 6μ thickness.

(b) The stainless foil wrapped in a Ni foil driver of 2μ thickness.

(c) The stainless foil only.

(d) The natural Fe foil driver only.

from a ferromagnetic to a non-ferromagnetic foil along an inter-
face.[1]

As we described earlier,[1] the physical mechanism for the
dressing of the states is a precession of the nuclear spins about
a depolarizing field, $\vec{H}_d$ developed from time-varying magnetic
poles induced on the surface of a magnetic foil by the concurrent
precession of the internal saturation magnetization, $\vec{M}_s$ of the
medium. As shown in studies of ferromagnetodynamics[8,9] such
depolarizing fields can reach intensities whose magnitudes repre-
-ent a substantial fraction of the large value of $|M_s|$. Thus, by
forcing the precession of $\vec{M}_s$ with the modest fields from the
external coil, very large surface polarizations (and $\vec{H}_d$) are
driven in response. In the geometry of Fig. 4 opposite poles
will be induced on the faces of the driving foil in contact with
the nonmagnetic core foil and some part of $\vec{H}_d$ should be continued
through the core. Subsequent precession of the ^{57}Fe nuclei in
the core foil about this $\vec{H}_d$ which continued into it should mix the
nuclear states there as quantitatively described earlier.[1] The
critical distinction from the magnetostrictive picture is that
now the effect should scale with $\vec{H}_d$ which in turn scales with $\vec{M}_s$.
Such behavior is seen in Figs. 5a and 5b in the comparison of the
effect of the iron driver foil with M_s = 21600 to that of the
nickel driver with[7] M_s = 6100.

The principal parameter determining the amplitude associated
with a dressed state and, hence, with the intensity found in a
sideband is the angle, ϕ_g through which a ground state nucleus
precesses before the fields reverse. From a semiclassical de-
velopment[1] it was shown that

$$\phi_g \sim \frac{|H_0|\omega_g}{\omega_2{}^2} \qquad\qquad (1)$$

where $\vec{H}_0$ was the externally applied field, ω_g was the resonant
NMR frequency of the ground state, 45.49 MHz in pure iron, and ω_2
was the frequency of the applied fields. Magnetic resonance
studies use similar geometries and there the applied fields in
samples are estimated[10] to be

$$|H_0| \simeq 6(PQ/V\nu)^{1/2} \qquad , \qquad\qquad (2)$$

where P is the rf. power in Watts, Q is the quality factor of the
resonator containing the foil, V is the effective volume in the
coil containing the fields and ν is the frequency in MHz. Substitu-
ting Eq. (2) into (1) and collecting conversion factors suggests
that the principle scaling parameters, Z for the sideband ampli-
tude is,

$$Z = (PQ)^{1/2}\omega_2{}^{-2.5} \qquad . \qquad\qquad (3)$$

The dependence of sideband amplitude upon Z predicted by
the dressed state model was shown[1] to agree with that actually
measured when Z was varied by changing the power, P at a single
frequency, ω_2 = 61.7 MHz. In the work just completed we
continued such a comparison to include variations of the scaling
parameter, Z caused by changing frequencies. For these measure-
ments the source was simplified to a single foil of ^{57}Fe of 2.0
μm thickness held between the same graphite plates so that the
arrangement used previously[1] to determine the dependence upon

power would be reproduced. As the frequency was changed the
Q-factor of the resonant circuit was measured and the power was
adjusted to maintain the product PQ = 600 W. The resulting data
are shown in Fig. 6, together with the predictions of the dressed
state model. The curve for the latter was the same one used to
display[1] the dependence upon P but now replotted to accommodate
the variation of Z caused by changes of frequency from the pre-
viously fixed value of 61.7 MHz.

Several critical points emerge from consideration of Fig. 6.
For the frequencies reasonably far from resonance at 45 MHz, the
dressed state model gives good agreement with measurements when
the same scaling is maintained that had produced[1] a similar level
of agreement with variations caused by changing power. Since the
model treats the actual precession of the nucleus as a semiclas-
sical event, it is not surprising that the range of validities is
limited to small angles of precession. At the higher frequencies
and lower powers the nuclei cannot precess far before the $\vec{H}_d$
field reverses. Conversely, it is extremely significant that a
strong resonance is observed in the conversion efficiency into
the sidebands, just at the NMR frequency for resonantly flipping
the nuclei. This seems clearly a consequence of the mixing of
states mechanism that is inherent in the dressed state model and
completely inconsistent with magnetostrictive phenomenology.

Because of the complexity of the overlapping of higher order
sidebands, it was necessary to examine the sidebands at the
lowest frequenices at a reduced value of power. Shown in the
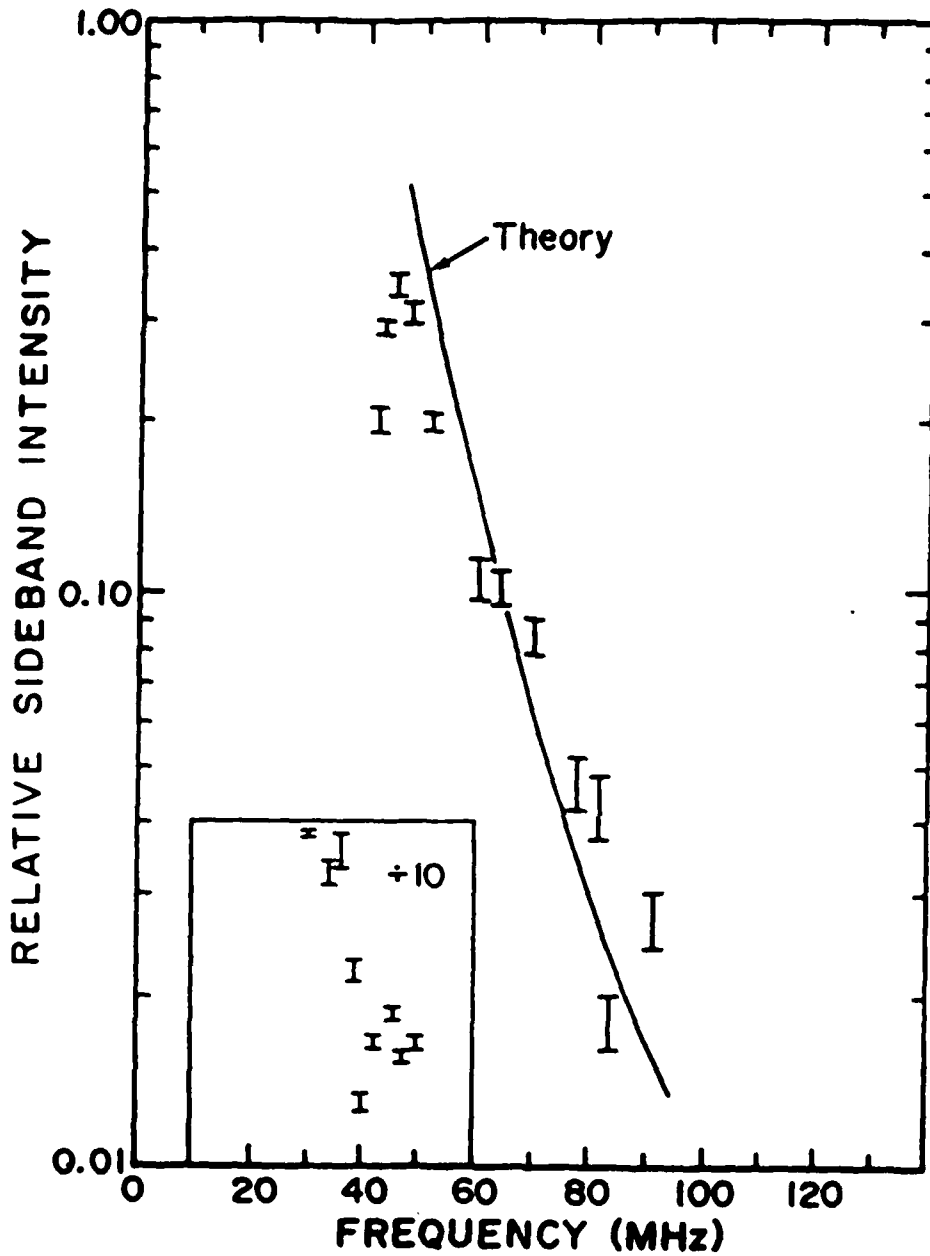inset to Fig. 6 are additional data obtained with PQ = 300.

Figure 6

Plot of the relative intensity of the first order sideband to the parent intensity remaining when pumped by a product of radiofrequency power, P and quality factor Q of PQ=600 W at the frequencies shown. Within the inset are results obtained with PQ=300 W and reduced by a factor of 10 for convenience in displaying. For comparison the predictions of a semiclassical, dressed state model of nuclear excitation are shown by the solid curve.

Both data sets cover the range of frequencies containing the resonance and both concur in the existence and shape of the structure.

While many curious effects remain in the literature to suggest magnetostrictive contributions to the develoment of rf. sidebands to Mossbauer transition in some circumstances we believe it is now completely clear that: 1) the strong correlation of sideband amplitude with permeability rather than magnetostrictive constant, 2) the agreement of the dependences of sideband amplitudes upon power and frequency with the dressed state model, and 3) the strong resonance in conversion efficiency into the sideband intensity at 45 MHz argue overwhelmingly for a multiphoton process in this experiment. The observed efficiency with which the nuclear states can be dressed tends to support an optimistic perspective on many of the other exciting analogs of the processes of quantum electronics that may be realized at the nuclear level and in particular the efficiency of the creation of dressed nuclear states that facilitate the coherent pump scheme for a gamma ray laser.

SIGNIFICANCE

The critical significance of our most recent work reviewed above is that dressed states have been demonstrated with just the properties expected. Now the problem for coherently pumping a gamma laser returns to the spectroscopic. To dress an isomeric state requires a certain arrangement of nuclear levels that would make them indetectable to conventional techniques of nuclear spectroscopy. Our method of NRS is the only means found to date

that can be used to search for this combination among the 29 best candidates.

The successes of the new NRS apparatus for nuclear spectroscopy indicate that a much higher resolution, by perhaps six orders of magnitude, can be achieved through a reasonable upgrade of the apparatus. If the range of tunability does extend to the ferromagnetic spin resonance (FSR) frequency, then it will be possible to construct a swept frequency device capable of continuously tuning over a range of 10^{11} linewidths, an enormous improvement in the state-of-the-art of nuclear spectroscopy.

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APPENDICES

"Nuclear Raman Spectroscopy," by B. DePaola, S. Wagal and C. B. Collins, J. Opt. Soc. Am. B2, 541-543 (1985).

"Nuclear Raman Spectroscopy with No Mechanical Tuning," by B. D. DePaola, S S. Wagal and C. B. Collins, J. Opt. Soc. Am. B (pending).

Nuclear Raman spectroscopy

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An experiment is described in which the nuclear analog to Raman spectroscopy has been applied to ^{57}Fe. The results of the experiment are given, and future applications of the technique are proposed.

In this paper we report major progress toward our overall goals of demonstrating the feasibility of tuning, and ultimately of stimulating, the emission of gamma radiation. Theory[1-5] developed over the past five years has shown the inherent viability of many of the analogs for nuclear excitation of the familiar concepts of quantum electronics. Model studies[5,6] have shown that hardware requirements to achieve threshold for stimulated emission in the subangstrom region would resemble lasers currently used in inertial fusion studies but that threshold should be accessed at substantially lower pump energies than will be needed for breakeven. It appears that existing devices might prove adequate *if suitable isotopes actually exist.* This is the critical point. Despite the many applications of beautiful and involved techniques of nuclear spectroscopy, the current data base is inadequate in both coverage and resolution either to answer the question of whether an acceptable isotope exists or to guide in the selection of a possible candidate medium for a gamma-ray laser.

To evaluate experimentally the feasibility of tuning and stimulating gamma radiation has required an iterative approach. As was recently reported,[7] the absolute scale of the matrix elements important to the realization of the nuclear analogs to the various possible frequency-mixing processes was confirmed to agree with theory. Incidentally, this implied that the techniques of nuclear spectroscopy could be extended in the direction needed to support a search for gamma-ray laser media by exploiting a type of multiphoton Mössbauer spectroscopy suggested by the same theories. Subsequently, the experimental feasibility of such a technique was reported.[6] Described here are the most recent results known to us confirming the practicality of the resulting methodology denoted nuclear Raman spectroscopy in this work.

Nuclear Raman spectroscopy (NRS) is a type of Mössbauer absorption spectroscopy in which the intensity of transmitted, single-frequency gamma photons is measured as a function of the frequency of a second, long-wavelength, photon field in which the nuclei are immersed. There are several advantages of NRS over more conventional Mössbauer techniques. One of these is the potential for an increased frequency resolution in the spectrum. This would be possible because the frequency of the long-wavelength photons can be controlled to a much higher degree than that of the gamma photons.

Another advantage of NRS is the potential for extending the range of nuclear spectroscopy. Using current techniques, one is typically limited in either resolution ($\sim$100 eV for gamma-fluorescence-type experiments) or range ($\sim$10^{-4} eV for conventional Mössbauer studies). If the NRS technique

were extended to use coherent microwave or infrared sources of variable-frequency photons, a great deal of new information could be learned about the nucleus.

As mentioned above, in a previous paper[6] some early efforts in NRS were reported. In those experiments, the feasibility of that technique was experimentally demonstrated, using a rf field as the source of long-wavelength photons. A problem associated with those experiments was a relatively low signal-to-noise ratio, resulting both from short data accumulation times and slight fluctuations in rf power levels for different frequencies. Those difficulties have been overcome in this work.

The energy-level diagram relevant to the absorber foil used in this work is given in Fig. 1. The splitting in the upper and lower manifolds is caused by the presence of strong static magnetic fields in the material. The selection rule $\Delta m = 0$, ± 1 allows six component transitions between the lower and upper manifolds. For convenience, these are designated in the figure by the numbers 1–6, as has been traditional in nuclear physics. Sidebands to these single-photon transitions (often referred to as parent lines) are given the same number labeling their parents, followed by one or more pluses or minuses, indicating positive (high-energy) or negative (low-energy) sidebands, respectively, from states dressed by the number of rf photons corresponding to the number of pluses or minuses.

Figure 2 represents the results of a series of experiments[7] designed to determine the dependence of sideband intensity on applied power. The measured sideband intensities were first normalized by dividing them by the intensity of their parents. The upper curve gives the dependence of the first-order sideband intensity on rf power, whereas the lower curve gives the dependence of the second-order sideband intensity on rf power. As will be shown, these curves proved useful in the analysis of the NRS data.

The new experiment was carried out in the following way: An ^{57}Fe foil was irradiated by a beam of essentially monoenergetic gamma photons and by a strong field of tunable rf radiation. The intensity of the transmitted gamma photons was then measured as a function of the rf ω_{rf}, which was varied while the gamma frequency ω_γ was held constant. The gamma-photon frequency was held fixed by using a conventional Mössbauer-spectrometer motor in constant-velocity mode. The rf-field strength in the sample foil was held as constant as possible, and ω_{rf} was varied at the oscillator. Because a resonant LC tank circuit, which was used to enhance the rf field, had to be retuned, and its Q had to be
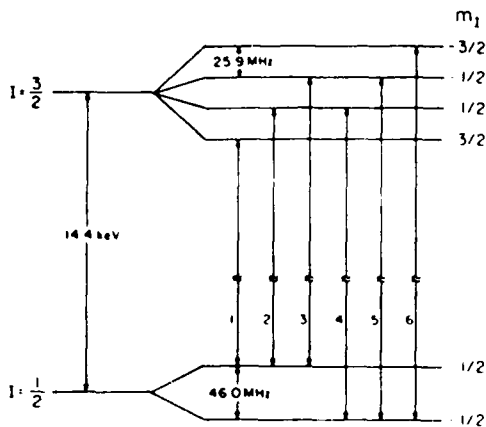
Fig. 1.   Energy-level diagram for ^{57}Fe, showing the nuclear hyperfine splitting and the six magnetic-dipole-allowed transitions.
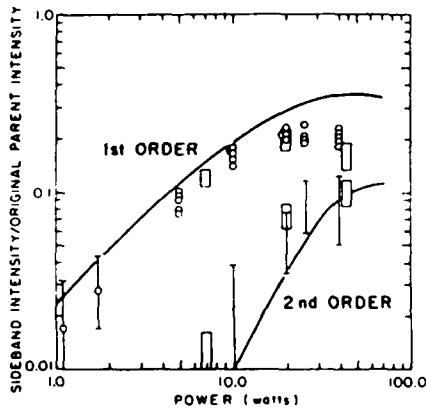

Fig. 2.   Comparison of experimental and theoretical computations of sideband intensities. Differing intensities at the same power reflect the measurement of sidebands from different parent transitions. (See Ref. 7 for a more detailed explanation).

measured for each frequency, only one data point, the average of 1024 channels in a multichannel analyzer, was taken per run.   Further details of the experimental arrangement are given in Ref. 8.

Figure 3 helps to explain the experiment further.   Figure 3a is part of a conventional Mössbauer spectrum in which the tuning resulted from progressively increasing Doppler shifts produced mechanically.   Clearly displayed in Fig. 3a is a typical sideband spectrum with $\omega_{rf} = 2\pi \times 54$ MHz, while in Fig. 3b $\omega_{rf} = 2\pi \times 61.7$ MHz.   With a constant source velocity of 7.78 mm/sec, the frequency of the gamma photons, ω_γ, would lie at the position indicated, regardless of ω_{rf}.   The effect of increasing ω_{rf} is to push the sidebands away from the parents, through the two-photon resonance at ω_γ.

Each data point was corrected for input power fluctuations and the Q of the LC tank for each frequency, as well as for the weakening of the gamma source with time, by the formula

$$S_n' = \frac{\left[B - S_n \exp\left(\frac{+t}{\tau}\right)\right] f(P_s)}{f\left(\frac{P_n Q_n}{Q_s}\right)}. \qquad (1)$$

where S_n is the nth unprocessed data point, P_n is the power read with a directional power meter, Q_n is the quality factor of the LC tank circuit for the nth data point, t is the relative age of the source (in days), and B is the baseline of the spectrum.   The symbol τ in this expression is equal to the half-life of ^{57}Co ($\sim$271 days), divided by the natural logarithm of 2.   The function $f(P_n)$ is given by the upper curve in Fig. 2 and was useful in normalizing all the data to a standard power and Q factor (i.e., for $n = s$) of 12.2 W and 50.8, respectively.   The resulting spectrum is shown in Fig. 4.   The triangles represent the normalized data points, while the solid curve in the figure is the best fit of these points to the sum of two Lorentzians subtracted from a constant baseline.   The Simplex algorithm[9] was used to fit this function to the data.

Theoretically, the function to be fitted to the data should be of the following form:

$$S = B - (L_1 + L_2). \qquad (2)$$

where

$$L_n = \frac{h_n \sigma_n^2}{\sigma_n^2 + (\omega_{0n} - \omega_{rf})^2}. \qquad (3)$$

Here, h_n is the maximum height, σ_n is the half-width at half-maximum (HWHM), and ω_{0n} is the center of the nth Lorentzian; B is the baseline.   This means that, for the two absorption lines in Fig. 4, seven parameters had to be fitted to the data.

The estimated standard deviation of the resulting curve is <5%.   The minima in the solid curve are within $\sim$1 MHz of their theoretical positions.   This is not unreasonable agreement considering there are only 14 data points spread over a range of 15 MHz.

In addition to the expected 5+ first-order sideband to transition #5 appearing at 59.7 MHz, an additional absorp-
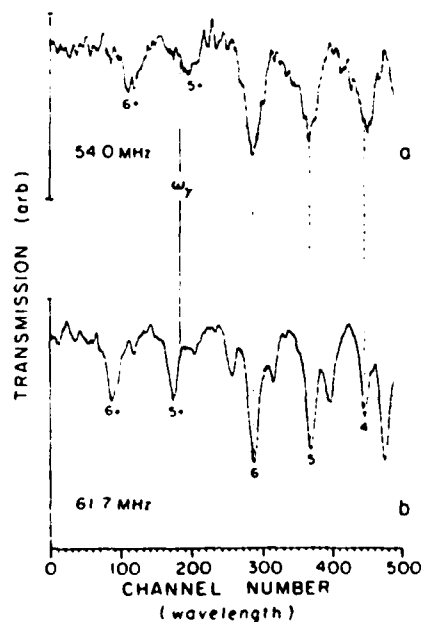

Fig. 3.   Two spectra that show that the result of increasing ω_{rf} is to push first-order sidebands away from the parents, through a two-photon resonance at ω_γ.
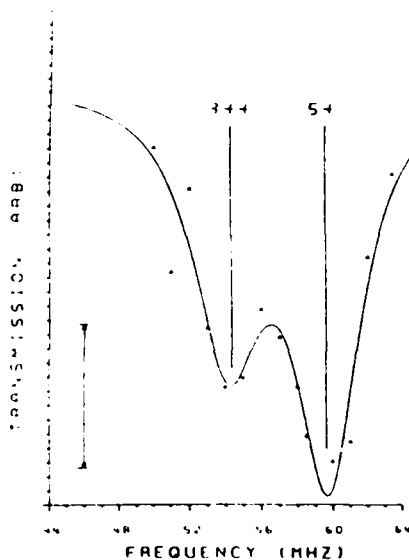
Fig. 4. Gamma transmission intensity plotted as a function of rf photon frequency. The larger absorption line corresponds to the first-order, sum-frequency sideband of the $|1/2, -1/2\rangle \rightarrow |3/2, -1/2\rangle$ nuclear transition of ^{57}Fe, while the smaller line corresponds to the second-order, sum-frequency sideband of the $|1/2, 1/2\rangle \rightarrow |3/2, -1/2\rangle$ transition

tion line appears at 54.1 MHz. This corresponds to 3++, the second-order sideband to transition #3. The separation between these two lines is also within ~1 MHz of the theoretical value.

There were several potential sources of error in this experiment, and they will now be discussed. First, with only 14 data points to span a 15-MHz interval, better resolution along the frequency axis could not be expected. Obviously, the small number of points precluded the use of a running averge in order to reduce the statistical fluctuations in the signal. In addition, because there were only twice as many data points as there were parameters to fit, the best fit of the data to the expected function must be viewed with caution.

A final source of error resulted from the method used to normalize the data. As mentioned in the explanation of Eq. (1), $f(P_n)$ is given by a plot of observed first-order sideband intensity as a function of applied power (measured at the directional power meter). In fact, this procedure is not valid for points that are affected by a second- or higher-order sideband. Unfortunately, there was no satisfactory way to deal with this problem: The spectra could not be deconvolved until the points were first normalized, and the points could not be properly normalized until the spectra were sufficiently deconvolved to weight the normalization process. In the case of the data shown in Fig. 4, the input powers and Q_s were close enough in value that the correction factors were all <2 (with most <1.3).

One way that this last error could have been eliminated is by tuning the gamma-photon frequency to be less than that required to make transition #1 by an amount greater than the sum of the total widths of the two manifolds. Then, by applying an rf field with a high enough frequency, that is, for ω_{rf} ranging from $2\pi \times 124$ to $2\pi \times 202$ MHz, the entire upper manifold could be seen as first-order sidebands with no second-order lines entering the spectrum. Unfortunately, because of the limitations of the LC tank circuit used in this work, the highest frequency that could be used was well below this amount.

In any case, the point of the experiment was not to characterize the energy levels of ^{57}Fe but to demonstrate the effectiveness of a new technique for nuclear spectroscopy in general, based on multiphoton effects. In practice, the method would probably be most useful with the rf field being replaced by a microwave or infrared photon field. The fact that a two-photon nuclear resonance can be detected by varying the longer-wavelength photon has significant implications. For example, the technique could be used to conduct Mössbauer spectroscopy with much higher resolution than ever before. This would be possible because no mechanical tuning need be used. In addition, if the technique were extended from rf to microwave or infrared photons, the range of Mössbauer spectroscopy would be dramatically increased.

ACKNOWLEDGMENT

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Nuclear Raman Spectroscopy with No Mechanical Tuning

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ABSTRACT

An experiment is described in which the nuclear analog to Raman Spectroscopy has been applied to ^{57}Fe. Unlike previous experiments, no mechanical movements of the source relative to the absorber were employed, allowing extremely high-resolution measurements. Future applications of the technique are discussed.

In previous theoretical papers [1-5], it had been concluded that many of the concepts of quantum electronics should be applicable to the nucleus. In recent experiments [6-8] these arguments have been shown to be fully justified. In our last communication [7] we reported the newly developed technique of Nuclear Raman Spectroscopy (NRS) that represents a realization of one of these possibilities. As discussed there [7], this technique promises to give higher resolution and a better tuning range than conventional Mossbauer methods that depend upon controlled Doppler shifts to vary transition energies. Due to its versatility NRS should prove to be very useful in building the spectroscopic database that will ultimately guide the search for a suitable material for a gamma ray laser.

The purpose of the present experiment was to demonstrate just one aspect of the newly developed Nuclear Raman Spectroscopy, namely its extremely high resolution. In particular, the technique was used to directly measure the isomeric shift between a ^{57}Co source and an isotopically enriched (97%) ^{57}Fe absorber. Such shifts occur because of the non-zero density of s-state electrons within the finite volume of the nucleus. The precise degree to which the emission lines from one material are displaced from the absorption lines in another depends upon the states of nuclear excitation and upon the electron distributions which in turn are affected by the differing bulk fields of the hosts.

Using conventional Mossbauer techniques one source of error in the measurement of isomer shifts comes from an uncertainty in the drive motor velocity, and therefore in the gamma photon

frequency at the very low velocities needed to compensate the relatively small isomeric shifts usually encountered. Using NRS, however, there need not be any relative motion between source and absorber. As in the analogous process occurring at optical frequencies, the nuclear Raman lines appear at energies equal to the sums or differences of the energy of the parent nuclear transition and that of one or more of the photons of the driving fields, in this case, magnetic fields oscillating at radiofrequencies (RF). In the NRS technique the "tuning" is accomplished by changing the frequency of the RF field. For the "typical" Mossbauer source, ^{57}Co the Doppler shift produced by a drive velocity of 1 mm/sec corresponds to a frequency change of 11 MHz. Because current state-of-the-art Mossbauer drive motors cannot be controlled to better than 0.04 mm/sec (corresponding to a frequency shift of 440 kHz), NRS provides much higher resolution since modern electronics oscillators are stable to better than 1 Hz in 1 MHz. Therefore, the only significant errors remaining in the measurement should be those due to inhomogenous line broadening and electronic absorption.

Another limitation of the resolution that is inherent in the velocity tuning technique is the "cosine smearing effect." That is, the effective energy, E_{eff}, of photon E_0 emitted by the source when the velocity v is

$$E_{eff} = E_0 \left[1 + \frac{v\cos\theta}{c} \right] . \tag{1}$$

where $v\cos\theta$ is the effective velocity component in the direction of the observer. This can contribute significantly to the observed

linewidth to an extent depending upon the geometry of the experi-
ment and upon the angles subtended by the source, the absorber and the
detector. Again, in NRS with no mechanical movement this source
of error does not exist.

Figure 1 is the energy level diagram relevant to ^{57}Fe as
used in Mossbauer work. Figure 2 shows the gamma photon trans-
mission as a function of gamma photon wavelength. To obtain this
data, the transition energy of the gamma source was swept through
the range of wavelengths in the conventional fashion, that is, by
accelerating it relative to the absorber so that decreasing
velocities, and hence, decreasing energies are linearly displayed
along the abscissa. A 26.6 Watt, 61.85 MHz field was applied to
the absorbing sample, thereby creating the sidebands to the
parent lines as described in previous reports [6-8].

In the absence of any isomeric shift, the application of a
61.85 MHz field should cause the first negative sideband of
transition #6 to coincide with the first positive sideband to
transition #1 as shown in Fig. 2. If, however, there is an
isomeric shift of magnitude Δ between the source and absorber,
then the two sidebands will be separated by 2Δ and will be cen-
tered about 61.85 MHz. The sidebands important to this experi-
ment are labeled by giving them the same number as their parents
and then adding a plus (for the first positive sideband) or a
minus (for the first negative sideband), depending upon whether a
radiofrequency photon was being added or substracted, respec-
tively, to the total transition energy. A "running average"
technique was applied to the data to improve the signal-to-noise

ratio. No further processing was done in order to insure that data would not be biased.

Notice that in the center of the spectrum shown in Fig. 2 midway between the third and fourth parent lines, appears a large absorption line that corresponds to an overlap of the 1+ and 6- sidebands. With the resolution afforded by conventional methods, these two lines appear as one. With the newly developed NRS technique, however, the two are easily separated.

The experimental arrangement was the same as that discussed previously [8], with the exception that in this work the ^{57}Co source was not attached to a drive motor, but instead it was held fixed with respect to the ^{57}Fe absorber. The intensity of the transmitted gamma radiation was measured while the frequency was varied between 51 MHz and 62.5 MHz in order to scan the sideband through the parent line. It was not possible to tune beyond 62.5 MHz because of the limitations on the RF oscillator.

Figure 3 explains the positions of the sidebands for different radiofrequencies and how that data is used to determine the isomeric shift. In Fig. 3, the line marked 6- is the first negative sideband of transition 6 and 1+ is the first positive sideband of transition 1 . As shown, the source line is displaced from the transition center by an energy Δ, the isomer shift. The parent lines 1 and 6 are each 61.85 MHz away from the transition center. When the applied radiofrequency, ν_2, is less than 61.85 MHz the positions of the sidebands are as shown in Fig. 3a. When $\nu_2 = \nu_{R_1}$, as shown in Fig. 3b, where $\nu_{R_1} = (61.85 - \Delta)$ MHz, the first negative sidebands of transition 6 will coincide with the source line and we get a strong absorption. If

$\nu_2 = \nu_{R_2} = (61.85 + \Delta)$ MHz as in Fig. 3c, then the first positive sideband of transition 1 coincides with source and we get another absorption peak. Thus one can determine the isomeric shift from the measured values of ν_{R_1} and ν_{R_2}; that is $\Delta = (\nu_{R_2} - \nu_{R_1})/2$.

In actual practice, data were taken for a set of discrete frequencies that could be spanned by the available RF generators. The number of gamma photons transmitted by the absorber were counted in a multichannel scalar which was swept through 1024 addresses at which data could be accumulated for 20 second periods of time in each, in order to preserve a record of any long term drift of the counting rates or power levels. None were observed and counts from all 1024 channels were averaged together for a "run" at a single frequency. Each resulting data point then had to be corrected for frequency dependent variation of various parameters, in particular for changes in the fraction of the input power reflected from the absorber sample and for changes in the quality, Q-factor of the resonant circuit containing the coil in which the absorber foil was placed. These corrections were made explicitly as shown in the following expression:

$$S_n' = \frac{[B - S_n \exp(t/\tau)] f(P_s)}{f\left[\dfrac{P_n Q_n}{Q_s}\right]} , \qquad (2)$$

where S_n represents the accumulated signal for the nth data point, P_n is the applied power as measured by a directional power meter and Q_n is the quality factor of the resonator in which the

absorber is mounted. The factor $\exp(t/\tau)$ takes into account the aging of the source. This correction is required because data were collected over a period of weeks which is a non-negligible fraction of the half life of the source. The term, B is the baseline for the spectrum. The function $f(P_n)$ is given by the upper curve in Fig. 4 and was useful in normalizing all the data points to a "standard" power and Q factor (i.e., for n=s) of 12.2 Watts and 50.8 respectively.

For this experiment we took the data at two different times, the two experimental series being represented in Fig. 5 by "Δ" and "*". We observed that in both sets the shape and position of the absorption spectrum was the same but the absolute percentage of absorption was different. It was later observed that two localized discolorations had formed on the foil (a result of RF heating), which suggests that between the two runs the foil had either slipped from its original position or the field pattern inside the coil had changed, due to some instability. In the future we plan to overcome this problem by using a more homogeneous field from a longer and larger diameter coil. The resulting data were normalized to each other by multiplying one set by a constant factor. The normalized points are shown by a "*" in Fig. 5. The solid line is the best fit of a Lorentzian to the data. The function used for fitting was

$$L = \frac{y\sigma}{\sigma^2 + (\omega_0 - \omega_{rf})^2} \quad , \tag{3}$$

where y is the maximum height, σ is the halfwidth at half maximum, ω_0 is the center frequency and ω_{rf} is radiofrequency. The

estimated standard deviation of the fit was better than 5%.

Even though the intention was to measure the isomeric shift by scanning both the 6- and 1+ peaks, the RF oscillator currently installed in the system was uanble to reach the higher frequencies necessary. Still, the present experiment gives a good idea of the isomeric shift between our source and absorber pair, namely 3 MHz. Obviously, a larger number of data points would serve to increase both the accuracy and confidence level of the data. Likewise, a more stable rf source with a broader tuning range would help reduce errors, and allow an even more precise measurement.

In conclusion, an experiment was described in which the nuclear analog to Raman Spectroscopy was performed. The system involved no mechanical tuning, a feature which allows unprecedented resolution, in principle. In this work the technique was used to measure the isomeric shift between a ^{57}Co source and a pure ^{57}Fe absorber. The accuracy of the measurement was limited only by source and absorber linewidths, and restrictions imposed by inexpensive electronics (the RF oscillator). It is thought that with state-of-the-art electronics, the resolution of the technique could be increased by several orders of magnitude.
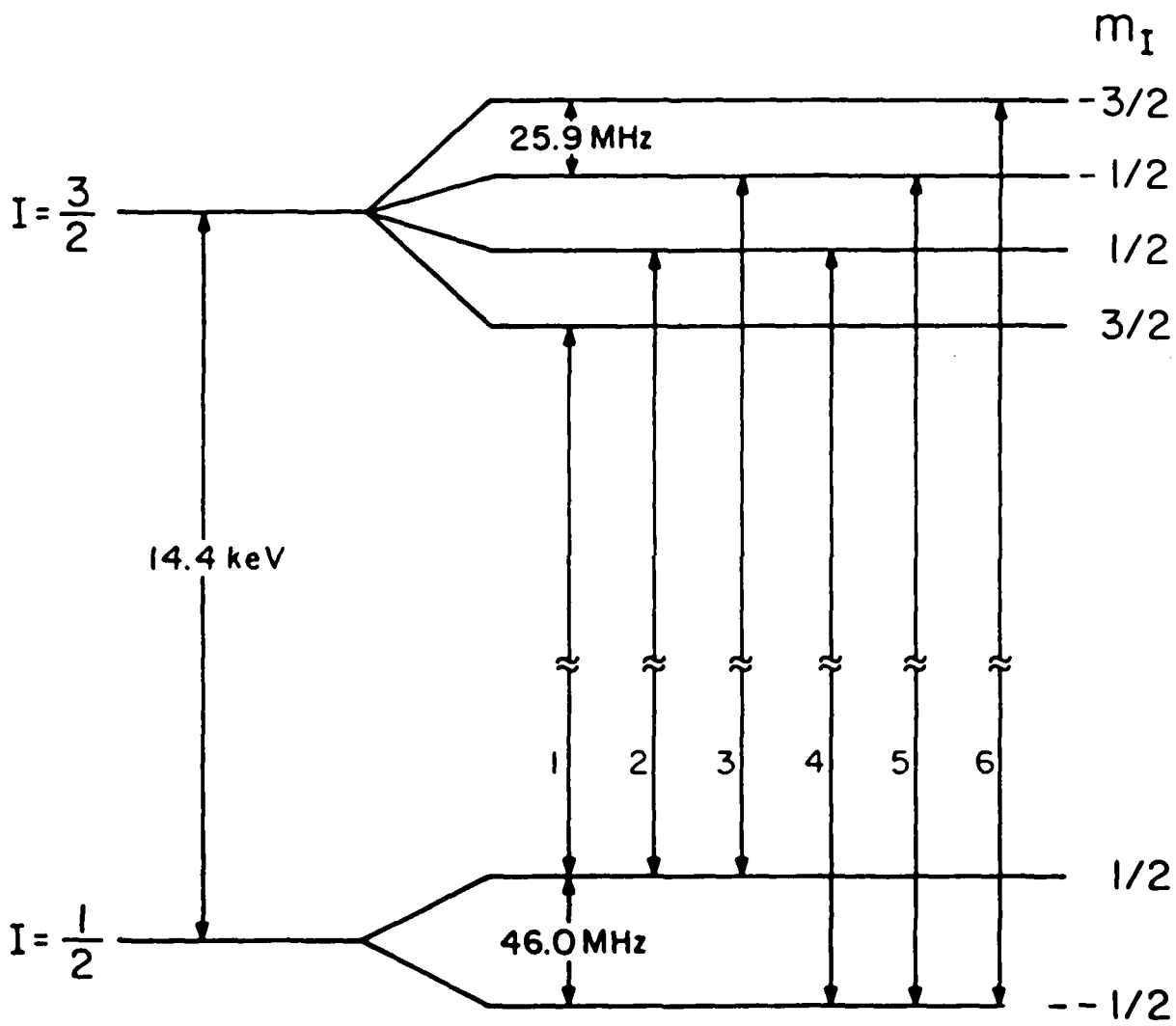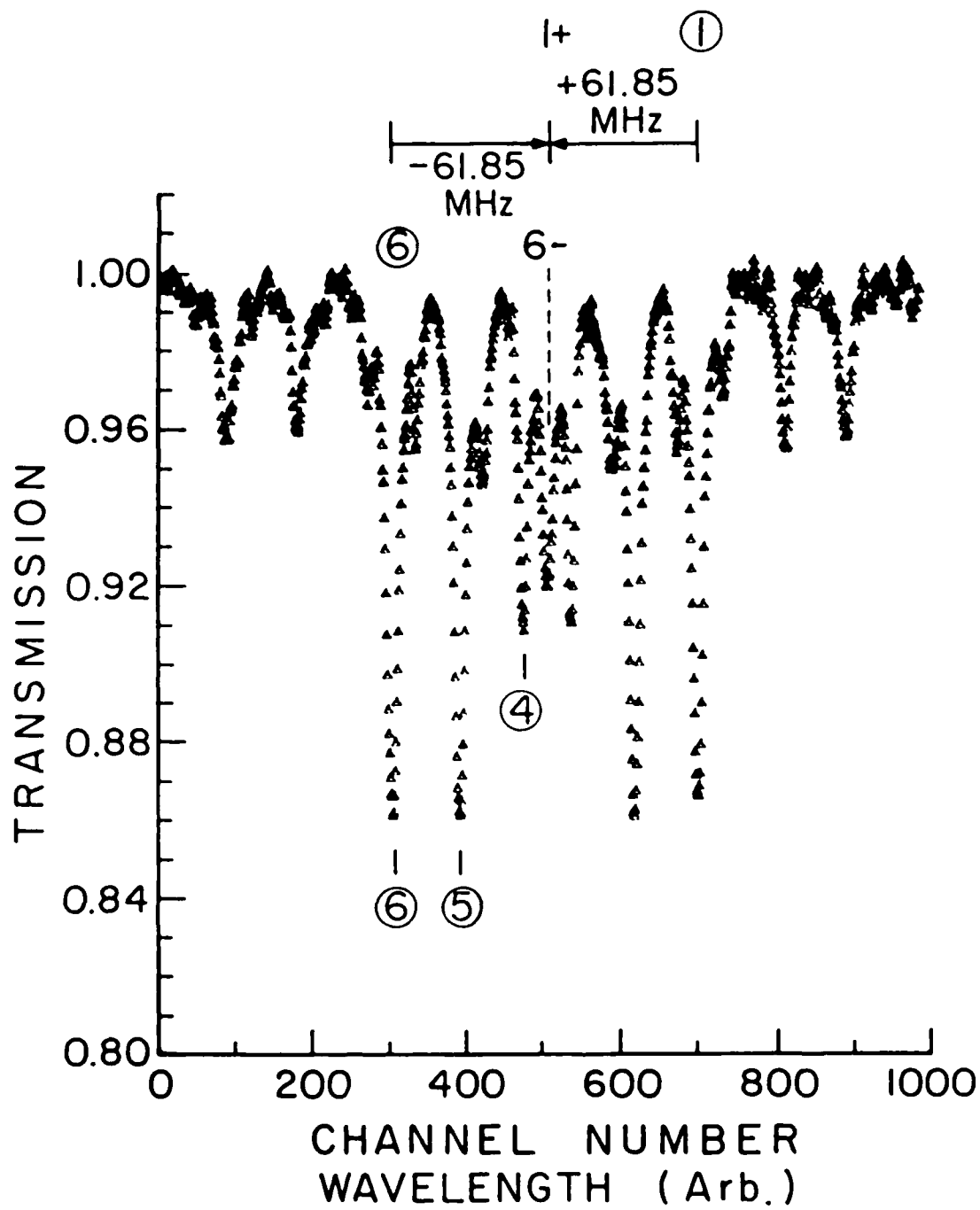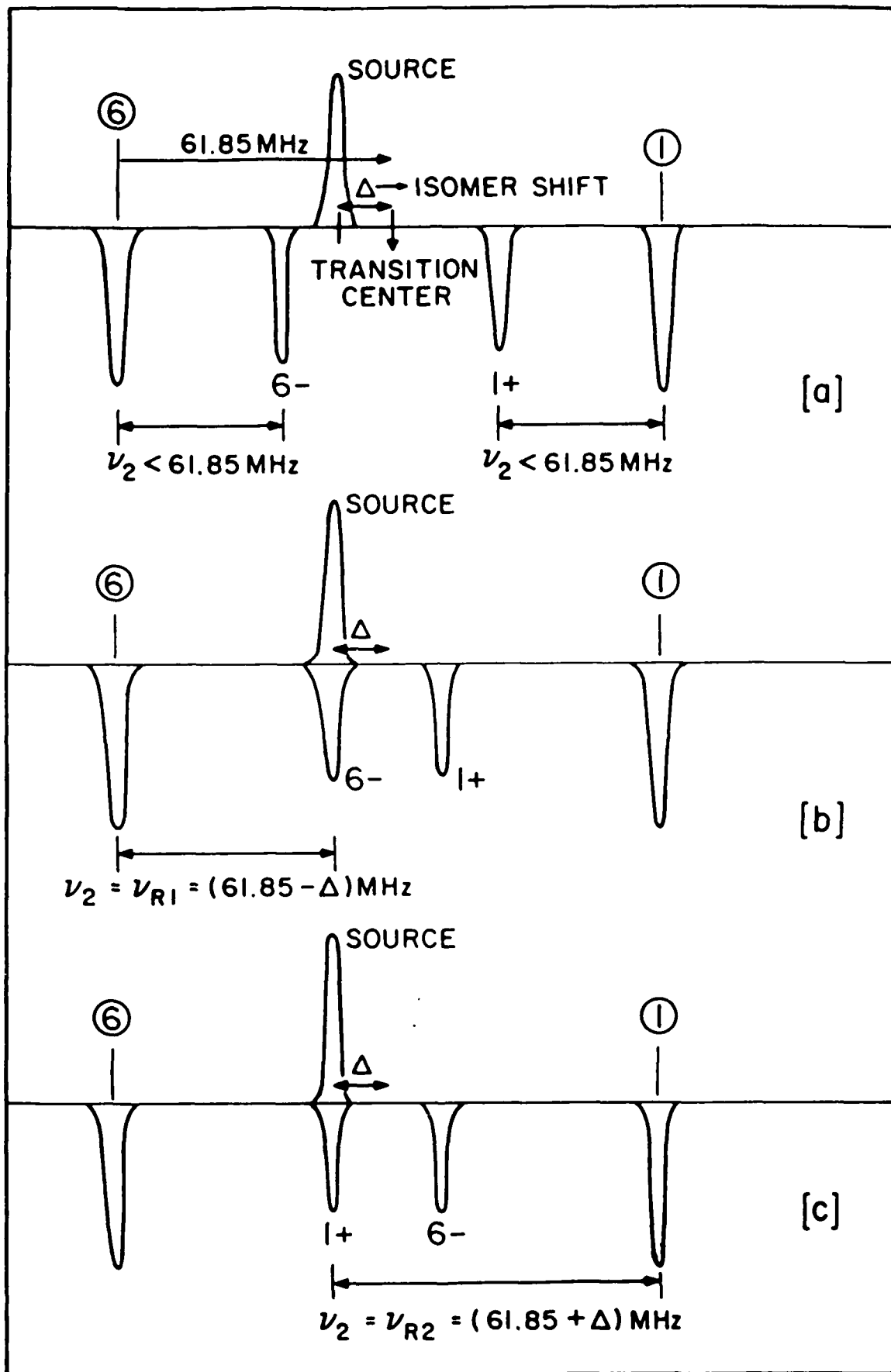
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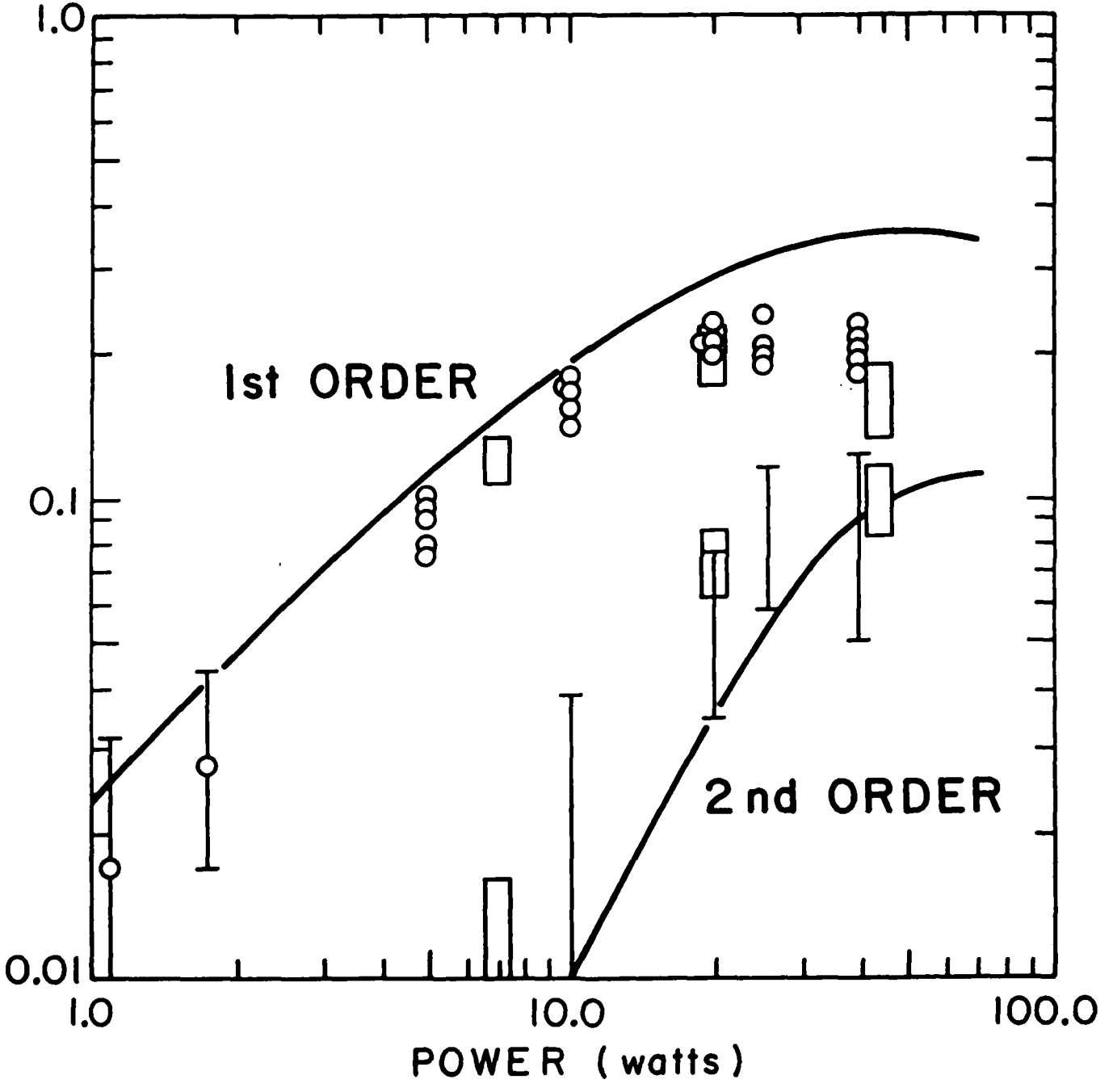
FIGURE CAPTIONS

Fig. 1: Energy level diagram for ^{57}Fe showing the nuclear hyperfine splitting and the six magentic dipole-allowed transitions.

Fig. 2: Typical data obtained for pure ^{57}Fe at 10.1 Watts of radiofrequency power at 61.85 MHz. Only five points running average have been applied.

Fig. 3: Schematic representation of the way the isomeric shift is measured. ν_2 = radiofrequency, Δ - isomeric shift, ν_{R_1} = (61.85 - Δ) MHz, ν_{R_2} = (61.85 + Δ) MHz, 6- is the first negative sideband of parent transition #6, and 1+ is the first positive sideband of parent transition #1.

Fig. 4: Comparison of experimental and theoretical computations of sideband intensities. Differing intensities at the same power reflect the measurement of sidebands from different parent transitions.

Fig. 5: Gamma transmission intensity plotted as a function of RF photon frequency. This case corresponds to ν_2 = ν_R = (61.85 - Δ) MHz as in Fig. 3.
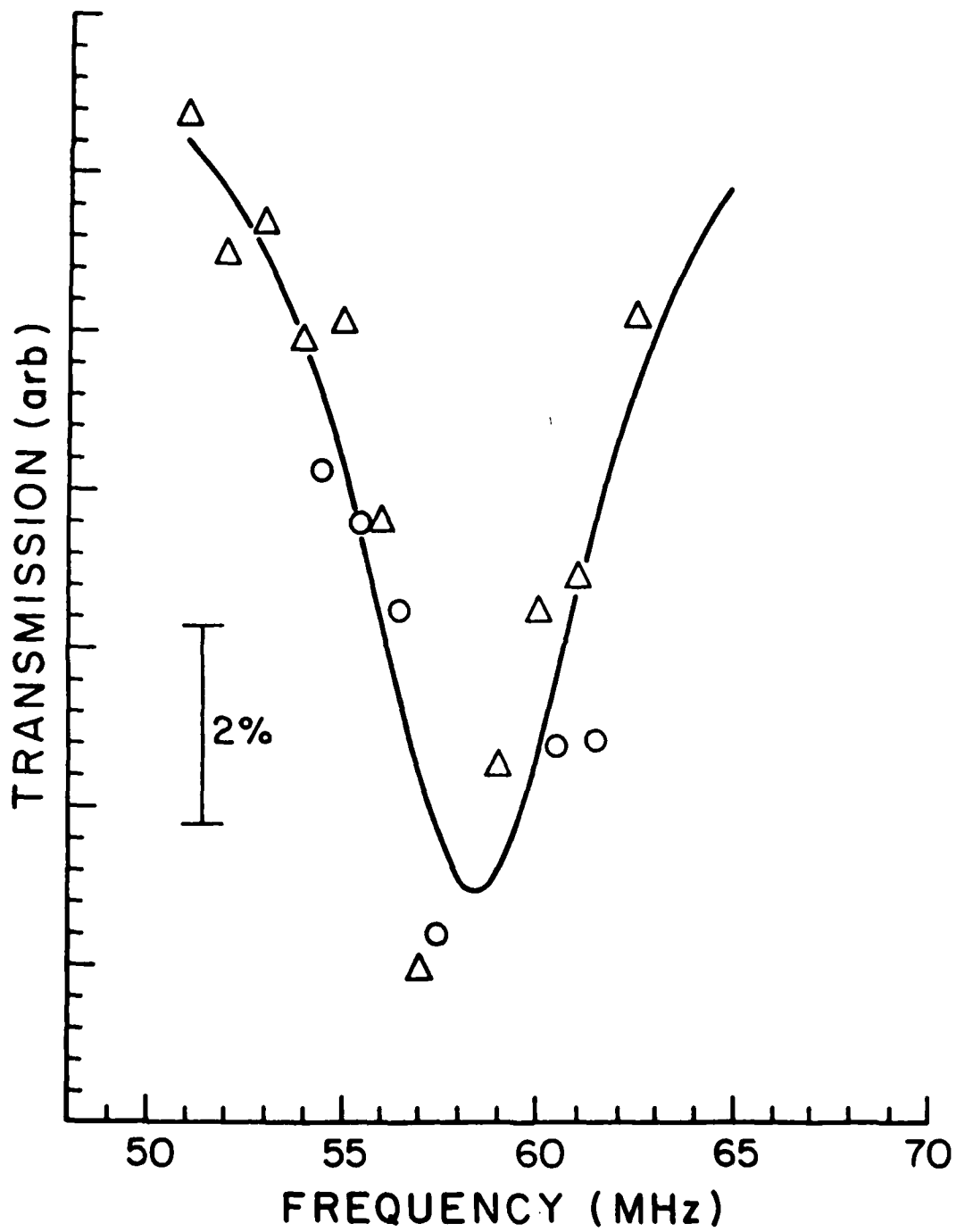
SOURCE

⑥   61.85 MHz   ①

Δ → ISOMER SHIFT

TRANSITION CENTER

6−   1+   [a]

$\nu_2 < 61.85\,\text{MHz}$ $\nu_2 < 61.85\,\text{MHz}$

SOURCE

⑥   ①

Δ

6−   1+   [b]

$\nu_2 = \nu_{R1} = (61.85 - \Delta)\,\text{MHz}$

SOURCE

⑥   ①

Δ

1+   6−   [c]

$\nu_2 = \nu_{R2} = (61.85 + \Delta)\,\text{MHz}$

END

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