Effect of a Sinusoidal Excitation Amplitude on the Performance of an Atomic-Beam Spectrometer

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(Received 19 January 1967)

A theoretical analysis has been made of the transition probabilities for a Rabi-type atomic-beam spectrometer in which the exciting field amplitude seen by the atoms has a sinusoidal rather than a rectangular envelope. The time-dependent Schrödinger equation was integrated numerically and a velocity average of the transition probabilities performed. The results indicate that the linewidth increases as the fourth root of the excitation power and that frequency shifts due to coupling of the exciting field with other atomic states are reduced by an order of magnitude at excitation powers much above optimum.

1. INTRODUCTION

IN some recent experiments by Harrach[1] hyperfine transitions in thallium were observed with an atomic-beam spectrometer. An attempt was made to observe a shift in the frequency of the $F=0$, $M=0$, to $F=1$, $M=1$ transition due to virtual, nonresonant transitions to the $F=1$, $M=-1$ state. Such a shift has been predicted theoretically[2,3] to be proportional to the power in the microwave field inducing the transitions and inversely proportional to the static magnetic field (C field) which separates the $F=1$ sublevels. However, the observed power-dependent shifts were an order of magnitude smaller than predicted and had a dependence on the magnetic field suggesting they were perhaps due to overlap with the $\Delta M=0$ transition, which was also observed. Also, the resonance line width was found to be proportional to the fourth root of the microwave input power, whereas the usual theories of atomic-beam spectrometers give a width proportional to the square root of the input power.[4]

In Harrach's experiment the cavity mode used for introducing microwave radiation to the atomic beam was such that the magnetic field had a sinusoidal variation in the longitudinal direction. Thus, the field amplitude seen by atoms passing through the cavity had a time dependence corresponding to half a sine wave, rather than a rectangular wave, as is assumed in the usual theoretical treatments (see Fig. 1). The present work was undertaken to see if this difference could account for Harrach's observations. Section 2 discusses the mathematics for finding the resonance transition probability appropriate to the experiments. Section 3 describes the numerical techniques employed to evaluate the mathematics. Section 4 describes the results and Sec. 5 summarizes the situation.

[1] R. J. Harrach, thesis, University of Colorado, 1965, Chap. V (unpublished); Natl. Bur. Std. (U.S.) Tech. News Bull. 346, 1966.
[2] H. Salwen, Phys. Rev. **99**, 1274 (1955).
[3] M. Mizushima, Phys. Rev. **133**, A414 (1964).
[4] N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956), Chap. V. Although an increase in linewidth with increasing excitation power is well-known experimentally, no quantitative measurements have been published [N. F. Ramsey (private communication)].

2. MATHEMATICAL FORMULATION

In the very low C fields used for the experiments, the hyperfine spectrum of Tl^{205} exhibits four states: the $F=0$ ground state, which we take to have zero energy, and three equally spaced $F=1$ levels, whose energies

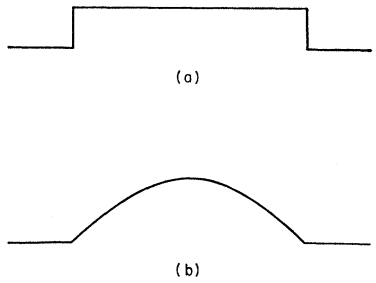

FIG. 1. Amplitude of the radiation field seen by atoms in an atomic-beam spectrometer: (a) usual Rabi configuration; (b) configuration used by Harrach.

(in frequency units) we call Ω, $\Omega+a$, and $\Omega-a$. The microwave excitation field at frequency ω connects the $F=0$ state to each of the $F=1$ states with matrix elements $2b\cos\omega t$ or $2c\cos\omega t$. The Hamiltonian describing the thallium atoms in the radiation field can then be written

$$\begin{pmatrix} \Omega+a & 0 & 0 & 2c\cos\omega t \\ 0 & \Omega & 0 & 2b\cos\omega t \\ 0 & 0 & \Omega-a & -2c\cos\omega t \\ 2c\cos\omega t & 2b\cos\omega t & -2c\cos\omega t & 0 \end{pmatrix},$$

where b and c are assumed real. If the radiation field is parallel to the static field, c is zero, if perpendicular, b is zero. Both are proportional to the amplitude of the microwave magnetic field.

It is possible to make part of the interaction terms in the Hamiltonian time-independent by a "phase-factor-

ing" transformation[5] which redefines the Schrödinger probability amplitude by $e^{i\mu t}$. Choosing $\mu=\frac{1}{2}(\omega+\Omega+a)$ for each of the $F=1$ levels and $\mu=\frac{1}{2}(-\omega+\Omega+a)$ for the $F=0$ level, the new probability amplitudes obey a Schrödinger equation with the Hamiltonian

$$\begin{pmatrix} -\Delta & 0 & 0 & c \\ 0 & -\Delta-a & 0 & b \\ 0 & 0 & -\Delta-2a & -c \\ c & b & -c & \Delta \end{pmatrix}$$

+terms oscillating with freq. 2ω,

where $\Delta=\frac{1}{2}(\omega-\Omega-a)$. The time-independent part includes the resonant interactions with the microwave field. For b, $c\ll\omega$ the oscillating part can be neglected (rotating field approximation).[6] In the experiments c/ω was at most 4×10^{-7}.

Assuming Δ, b, and c are all small compared to a, we further reduce the Hamiltonian to a two-by-two matrix

$$\begin{pmatrix} -\Delta & c \\ c & \Delta+c^2/2a+b^2/a \end{pmatrix}. \tag{1}$$

Here we have retained the submatrix involving the two resonant states which are nearly degenerate after the phase-factoring transformation. The terms with a in the denominator are second-order perturbation corrections which approximately account for the effects of the other two states.[2,7] If an atom initially in the upper state experiences the radiation field for a time t, the probability that a transition will occur to the lower state is

$$P(t)=(c^2/q^2)\,\sin^2qt, \tag{2}$$

with

$$q^2=c^2+(\Delta+c^2/4a+b^2/2a)^2.$$

This is the usual Rabi transition probability. To correspond to an atomic-beam experiment, it must be averaged over the different times t that different atoms spend in the radiation field region due to their distribution in velocity:

$$\langle P\rangle=(c^2/q^2)[\tfrac{1}{2}-I(2ql/\alpha)], \tag{3}$$

where l is the length of the radiation field region, $\alpha=(2kT/m)^{1/2}$ is related to the average velocity of atoms in the beam, and the function I is tabulated in Ref. 4. The resonance in $\langle P\rangle$ is peaked at $\Delta+c^2/4a+b^2/2a=0$ or $\omega=\Omega+a-c^2/2a-b^2/a$. The last two terms represent the power-dependent shift which the experiments had hoped to observe.

[5] J. H. Shirley, thesis, California Institute of Technology, 1963, Chap. IV (unpublished). Compare also K. Freed, J. Chem. Phys. **43**, 1113 (1965).

[6] J. H. Shirley, J. Appl. Phys. **34**, 783 (1963); F. Bloch and A. Siegert, Phys. Rev. **57**, 522 (1940).

[7] J. H. Shirley, Ref. 5, Appendix A.

In the preceding analysis it was assumed that the amplitude of the radiation field (b, c) was constant during the time the atom was exposed to it. In the actual experiment the amplitude had a sinusoidal variation. If we replace b by $(\frac{1}{2}\pi)\,b\,\sin\nu t$, and c by $(\frac{1}{2}\pi)\,c\,\sin\nu t$, then redefine phases to make the Hamiltonian traceless, we obtain the Hamiltonian

$$\begin{pmatrix} -\Delta+\delta\omega\,\sin^2\nu t & (\frac{1}{2}\pi)\,c\,\sin\nu t \\ (\frac{1}{2}\pi)\,c\,\sin\nu t & \Delta-\delta\omega\,\sin^2\nu t \end{pmatrix}. \tag{4}$$

Here $\delta\omega=-(\frac{1}{4}\pi^2)\,(c^2/4a+b^2/2a)$, $\nu=\pi v/l$, v is the velocity of an atom, and the Schrödinger equation is to be integrated from $\nu t=0$ to $\nu t=\pi$.

The factor $\frac{1}{2}\pi$ is introduced to keep the area under the curves in Fig. 1 the same. Intuitively we would then expect to get about the same transition probability. In fact, if $\Delta=\delta\omega=0$ (on resonance, no shift) the Schrödinger equation with the Hamiltonian (4) can be solved exactly, giving a transition probability at $\nu t=\pi$:

$$P=\sin^2(\pi c/\nu)=\sin^2(cl/v),$$

which is the same as (2) under the same conditions ($q=c$, $t=l/v$). Hence, at the peak of the resonance the Hamiltonians (1) and (4) both give the same transition probability and the same optimum excitation condition $2cl/\alpha=1.200\pi$.[4]

Except for the special case just mentioned, the Schrödinger equation with the Hamiltonian (4) cannot be solved analytically. It is of a type periodic in time for which an elegant formalism has been developed.[8] However, this formalism does not lead to any shortcuts or useful approximations in the present case. The only approximation of any value is the adiabatic approximation. At high powers ($ct\gg\frac{1}{2}\pi$), Eq. (2) shows that $P(t)$ oscillates several times while the atom traverses the radiation field region. If this oscillation is rapid compared to the rate of change of the $\sin\nu t$ modulating c ($q\gg\nu$), then (2) should give an approximate solution at each instantaneous value of $\sin\nu t$:

$$P(t)\approx\frac{(\pi^2/4)\,c^2\,\sin^2\nu t}{q^2(t)}\,\sin^2\int_0^t q(t')\,dt', \tag{5}$$

with $q^2(t)=(\pi^2/4)\,c^2\,\sin^2\nu t+(\Delta-\delta\omega\,\sin^2\nu t)^2$. At best (5) is valid only in the wings of the line $\Delta\gg\nu$. At $\nu t=\pi$, (5) says the wings of the line have no shift and no amplitude. This suggests that the line shape resulting from (4) is narrower and less shifted than that given by (2).

3. THE NUMERICAL METHOD

To obtain more definitive results the Schrödinger equation with the Hamiltonian (4) was integrated numerically with the aid of a digital computer. The fourth-order Runge–Kutta method was used. A trial run using the Hamiltonian (1) and then comparing the results of

[8] J. H. Shirley, Phys. Rev. **138**, B979 (1965).

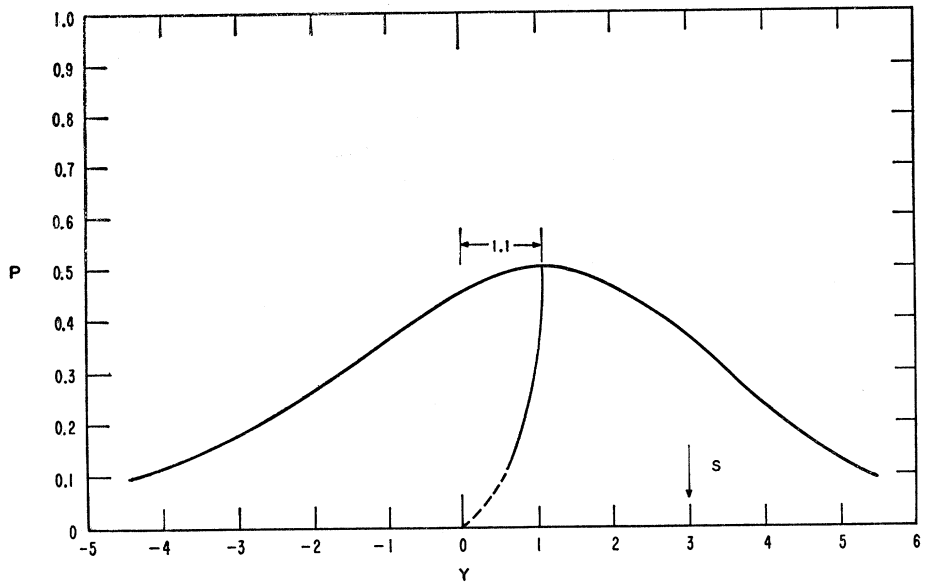

FIG. 2. Line shape for $X=5$, $S=3$. The locus of the midpoint between the two sides is also shown.

the numerical integration with the analytic solution (2) indicated that the accumulated error in the normalization of the wave function gave a crude but useful bound on the error in the transition probability. Accordingly, the number of steps used in the numerical integration was automatically controlled to maintain the normalization within 0.005 of 1.

For a quantitative comparison with experiment, it is necessary to average the transition probability P over the distribution of velocities in the atomic beam. In Ramsey's work[4] this average is expressible in terms of the function

$$I(X)=\int_0^\infty y^3 \exp(-y^2)\cos(X/y)\,dy$$

[e.g. Eq. (3)]. Since we have only numerical values instead of the cosine function for the integrand, we must do the integral numerically. The method was first checked by computing $I(X)$ numerically and comparing the results with Ramsey's table. To avoid the rapid oscillations near $y=0$, the variable of integration was changed to $Z=y^{-1}$:

$$I(X)=\int_0^\infty Z^{-5}\exp(-Z^{-2})\cos XZ\,dZ.$$

This integral was evaluated by Simpson's rule. The step size must be reduced as X is increased to maintain accuracy. The experience gained with this integral was utilized in the final program which computed the velocity averaged transition probability according to the formula

$$\langle P\rangle=2\int_{0.3}^{2} Z^{-5}\exp(-Z^{-2})\,P(XZ,\,YZ,\,SZ)\,dZ. \quad (6)$$

Here

$$X=2cl/\pi\alpha, \qquad Y=2\Delta l/\pi\alpha, \quad \text{and} \quad S=\delta\omega l/\pi\alpha \quad (7)$$

are dimensionless measures of the microwave field amplitude, deviation from resonance, and expected frequency shift, respectively, and $Z=\alpha/v$. The arguments of P are the parameters required for the integration of the Schrödinger equation in dimensionless form. The 2 in front is for normalization. The lower limit of 0.3 was used instead of zero since the weighting function is negligible for smaller values of Z. The upper limit corresponds to excluding from the velocity distribution all atoms whose velocities are less than $\frac{1}{2}\alpha$. The geometry of the spectrometer used in the experiments was such that these slower atoms actually do not contribute to the observed signal.[9]

To obtain one value of $\langle P\rangle$ for given parameters X, Y, S, it is necessary to integrate the Schrödinger equation numerically for each point needed to evaluate (6) by Simpson's rule. The number of steps required for either integration is modest at $X=1$, but increases rapidly to 50 or 100 for $X=10$, the actual number used being determined by the computer. For a check on the error accumulated in such lengthy calculations, they were carried out using the time-independent Hamiltonian (1) and comparing the results with (3). Out of about 40 points compared, the largest discrepancy was 2%, with most values agreeing to less than 1%.

To study the linewidth, the shift S was set equal to zero. Then $\langle P\rangle$ was calculated for three values of Y in the vicinity of $Y=(\pi X/2)^{1/2}$. Parabolic interpolation was used to find $Y_{1/2}$, the value of Y such that $\langle P(X,\,Y)\rangle=\frac{1}{2}\langle P(X,\,0)\rangle$.

When $S\neq 0$, the line shapes are somewhat asymmetric (see Fig. 2). The frequency shift found depends upon the height at which the line is split. Note that the wings of the line appear to approach no shift, in

[9] R. J. Harrach (private communication).

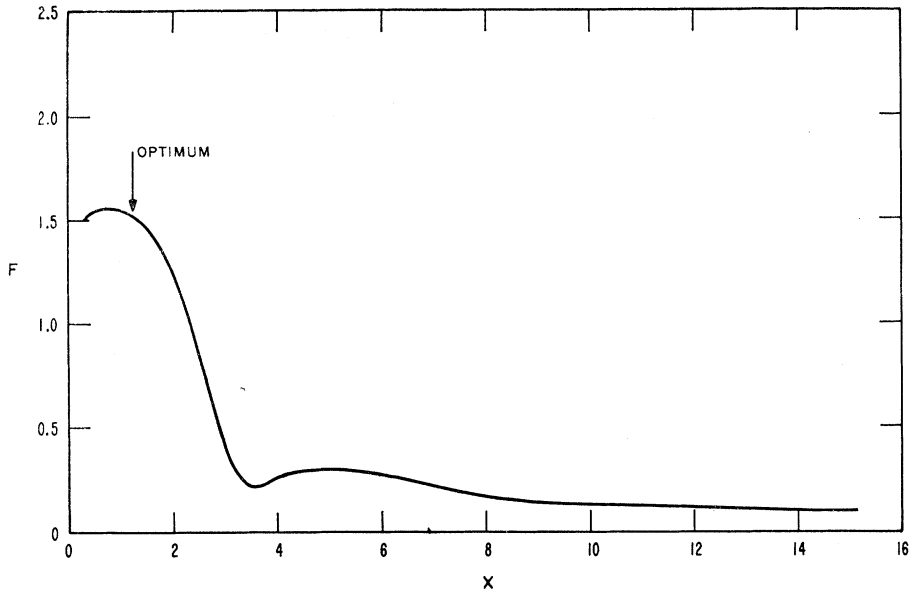

FIG. 3. The ratio $F(X)$ of calculated shift to expected shift as a function of exciting field amplitude.

agreement with the adiabatic approximation. For a consistent procedure analogous to the experimental method, the line center was defined as the midpoint of the line at 70% of its maximum height. Using the linewidths previously determined, the computer selected Y values which gave values of $\langle P \rangle$ approximately 60 and 80% of the height on each side. Linear interpolation then provided the Y values at 70% of peak height, which were averaged to yield the line center Y_c.

4. RESULTS

The half-widths $Y_{1/2}$ found by the computer by using the method described are given in Table I and compared with the empirical formula. The X values were chosen to correspond to the experimental data.[1] For large X the half-width becomes $Y_{1/2}=1.07(\pi X/2)^{1/2}$. Using $\alpha=2.7\times10^4$ cm/sec and $l=16$ cm, the full width becomes $w=(\alpha/l)Y_{1/2}=2.5(P_i/P_0)^{1/4}$kHz, where P_i is a measure of the microwave power input to the spectrom-

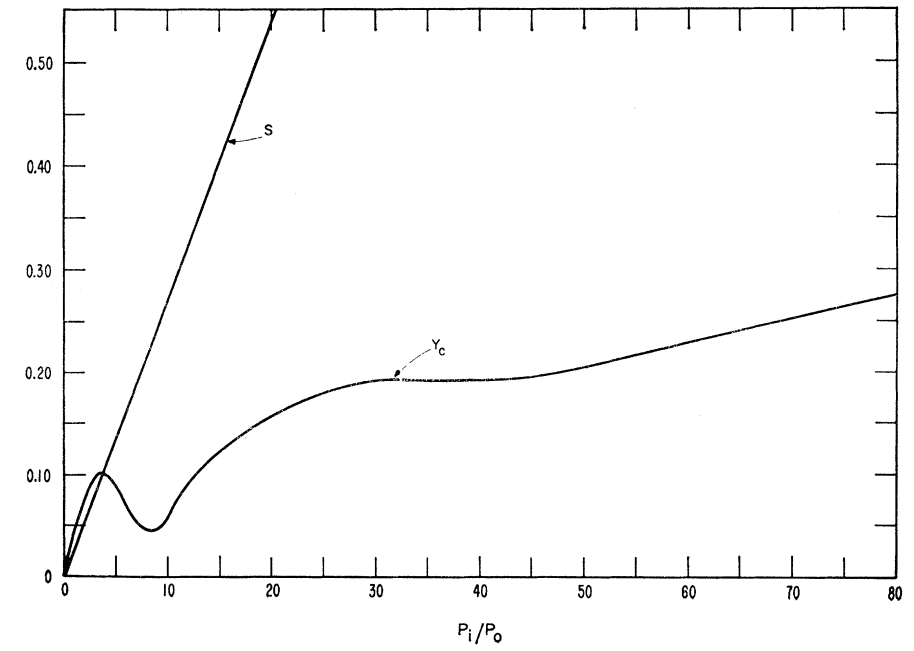

FIG. 4. Theoretical power dependence of S and Y_c for experimental conditions.

eter and P_0 is the corresponding value at optimum power: $P_i/P_0=(X/1.2)^2$. The experimental result[1] was $w=2.2(P_i/P_0)^{1/4}$kHz.

For a given X and S, let the Y value at the line center defined in the preceding section be represented by $Y_c=F(X)S$. Adiabatically the line center occurs at $Y=2S\sin^2\nu t$. Hence $F(X)$ represents some resultant average of $2\sin^2\nu t$. We might guess that $F(X)=1$, but any value of F between 0 and 2 would be possible. Conceivably F could depend on S as well as X, but limited data indicate that it does not, at least for shifts small compared to the linewidth. The function $F(X)$ as revealed by the numerical calculations is shown in Fig. 3. The value 1.5 near optimum power may arise because most of the transition takes place while the sine function is large. The small values of F for large X imply that the shifts are much smaller than anticipated, in agreement with the experiments.

Experimentally, the shifts were determined for various values of the input power keeping other variables fixed. Recalling the definition of S [see Eq. (7)] and of $\delta\omega$ [see Eq. (4)], we find that S should be increased as X^2 or linearly with the input power. The matrix elements in (1) were evaluated for a C field of 15 mG ($a=0.44\times10^3$ sec^{-1}). With $b=0$ and α and l as given previously we arrived at $S=0.0186X^2$. With this value for S, Y_c was computed at X values corresponding to the experimental data. The results are found in Table II along with the theoretical [i.e., $(\alpha/2l)Y_c$] and experimental shifts in hertz. The former are uncertain by 15 to 30 Hz and the latter by about 50 Hz. Clearly the order of magnitude of the shifts is in agreement.

The computed frequency shifts Y_c are plotted as a function of excitation power in Fig. 4. The straight line disappearing out of the top of the graph is S, the expected shift if $F(X)=1$. Harrach fitted a straight line to the corresponding experimental data, since the scatter was too great to show such features as the dip in Fig. 4. The frequency corresponding to no shift was determined experimentally by extrapolating to zero power. A straight line fitted to the right-hand portion of Fig. 4 will not extrapolate to zero shift; hence, the experimental shifts in Table II should probably be increased. The only significant number that can be extracted from Harrach's data is the slope of his fitted line at high powers. At the three higher C field values for which Harrach plotted his data these slopes are consistent with a slope obtained from the right-hand side of Fig. 4. For the three lower C field values the experimental slopes are higher, probably because of the presence of overlap shifts.

TABLE I. Empirical linewidth $(\frac{1}{2}\pi X)^{1/2}$, computed linewidth $Y_{1/2}$, and their ratio as a function of excitation power P_i or dimensionless amplitude X.

P_i/P_0	X	$(\frac{1}{2}\pi X)^{1/2}$	$Y_{1/2}$	Ratio
1	1.20	1.37	1.43	1.04
5	2.68	2.05	2.11	1.03
20	5.36	2.90	3.10	1.07
80	10.72	4.10	4.38	1.07
160	15.18	4.89	5.23	1.07
250	18.96	5.46	5.84	1.07

TABLE II. Dimensionless frequency shift S expected for rectangular envelope, dimensionless shift Y_c computed for sinusoidal envelope, computed shift in hertz, and experimental shift in hertz as a function of excitation power P_i or dimensionless amplitude X.

				Shifts in hertz	
P_i/P_0	X	S	Y_c	Theor.	Expt.
3	2.08	0.08	0.097	80	0
10	3.79	0.27	0.055	45	35
20	5.36	0.53	0.159	135	−5
30	6.57	0.80	0.193	165	5
50	8.48	1.34	0.206	175	135
80	10.73	2.14	0.276	235	175
100	12.00	2.68	0.304	255	245
150	14.70	4.02	0.371	315	370

5. SUMMARY

A theoretical analysis of the transition probability in the atomic-beam spectrometer used by Harrach indicates that the linewidth increases as the fourth root of the microwave excitation power and that natural frequency shifts are reduced by an order of magnitude at input powers of the order of 100 times optimum. These results adequately explain the experimental observations made by Harrach. Further confirmation of the theory could be achieved by observing the dip in frequency shift at about 8 times optimum power, although better experimental reproducibility would be required. It is probable that the natural shifts sought in the experiment were observed in their reduced form, but the scatter in the data and the presence of overlap shifts make this conclusion subject to question. The experiment should be redone using a rectangular microwave field amplitude, which can be achieved, for example, by sending the beam longitudinally down the center of a rectangular cavity operated in the TM_{210} mode. The natural shift should then be large enough to dominate all other frequency shifts and permit a quantitative test of its form as given in Eq. (2).

ACKNOWLEDGMENT

The author wishes to thank Dr. Harrach for several discussions concerning the experiment.