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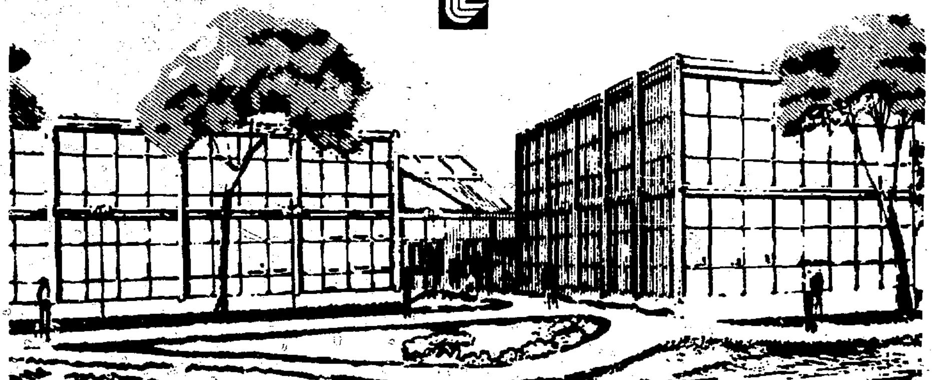

MULTIPHOTON RESONANCES*

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The long-time average of level populations in a coherently-excited anharmonic sequence of energy levels (e.g., an anharmonic oscillator) exhibits sharp resonances as a function of laser frequency. For simple linearly-increasing anharmonicity, each resonance is a superposition of various multiphoton resonances (e.g., a superposition of 3, 5, 7, ... photon resonances), each having its own characteristic width predictable from perturbation theory.

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Multiphoton Resonances

Recent experimental and theoretical studies are rapidly elucidating mechanisms responsible for laser-induced dissociation of molecules. Coherent excitation may, under suitable conditions, be a significant mechanism, and thus there is now emerging a considerable literature dealing with this theoretical model. One of the most striking characteristics of coherent excitation is the possibility of multiphoton resonances. Today I want to discuss some of the properties of these resonances.

To simplify interpretation, I shall examine a very simple model (Figure 1): a succession of linked transitions forming a ladder-like chain, excited by a single monochromatic laser. The parameters which characterize this sequence are the successive Rabi frequencies (I shall consider two specific sequences: either all Rabi frequencies equal or Rabi frequencies increasing in proportion to the square root of the level number, as in a harmonic oscillator), the anharmonicity A, and the detuning of the laser frequency away from the fundamental molecular frequency. I shall assume that all Rabi frequencies are much larger than any dissociation rates or rates of equipartition of energy into other modes, so that we deal with a simple lossless system.

The dynamics of this system are expressed by the Schrödinger equation. After making the conventional rotating-wave approximation (Figure 2), I solve the resulting time-dependent equations by standard techniques. Let us examine the behavior of the solutions to these equations, with the aim of gaining insight into behavior which may occur in more complicated physical situations.

First, let me remind you of the way in which anharmonicity affects excitation (Figure 3). If we tune the laser into resonance with the fundamental molecular frequency, then in the absence of any anharmonicity the excitation diffuses steadily upward until in an N-level system it encounters the final level -- the top of the excitation ladder. The presence of anharmonicity alters this behavior: we only excite part way up the ladder. These successive pictures show how increasing anharmonicity places an effective upper limit on the excitation reached. These curves are for harmonic-oscillator Rabi frequenices; similar behavior occurs for equal Rabi frequency.

Actually, it is the ratio of anharmonicity to Rabi frequency (say, the first one) which dictates the dynamics. Thus, the sequence of increasing anharmonicity is also the sequence of diminishing laser intensity. By increasing the power sufficiently, we can excite as high as we wish. For example, to excite the fifth level of an anharmonic oscillator with appreciable probability, we need to make the first Rabi frequency some five times the anharmonicity.

One might well ask if we cannot improve upon this excitation by retuning the laser away from the fundamental frequency (Figure 4). Here we see how we tune the laser to achieve one-photon (discussed above), two-photon, and three-photon resonances. One expects that by means of such tunings we could reach high excitation levels.

As Elliott and Feldman pointed out, the frequencies of these resonances are regularly spaced at integral numbers of anharmonicity units A away from the fundamental frequency (Figure 5).

We have previously seen that when the first Rabi frequency equaled the anharmonicity then we got little excitation. Let us look at the time-averaged populations over a long time interval for several choices of resonant frequency (Figure 6). As we see here, we can get half the population into any excited state. Not only do we populate the resonant level (levels 2, 5, and 8 for the three curves shown) but we also have population in neighboring levels. The width of the distribution-in-n increases with the Rabi frequency; it becomes very narrow as the laser power diminishes.

I have shown you the results for infinite-time averages. The short-time averages look quite different. Indeed, the shorter the time over which we average, the less pronounced is the high-level excitation. This is because the n-photon transition into the excited state is slow -- it occurs gradually over many Rabi cycles (Figure 7). Here we see how the transition comes about. This first frame shows the first 20 Rabi periods of an anharmonic oscillator having anharmonicity equal to the first Rabi frequency for a laser tuned to the four-photon resonance between levels one and five. We see that population remains in level one. Over a much longer time, however, we see the emergence of level five -- a slow Rabi flopping between levels one and five.

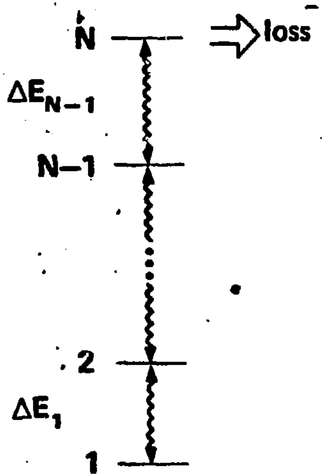
Now let us hold fixed the laser intensity and sweep the frequency, looking again at long-time average populations (Figure 8). Doing so, we scan an excitation spectrum. We see a broad background, upon which are superimposed a succession of sharp, equidistant resonances. The two frames depict two different laser intensities. As we see, a higher intensity laser raises the accessibility of high-lying levels.

The individual resonances have a structure which is rather curious; not only are the levels narrow, but each of the sharp resonances appear as a superposition of two distinct Lorentz profiles of quite different width (Figure 9). Here we see an expanded view of one of the resonances, using a logarithmic scale. The two peaks are clearly visible; they differ in width by nearly three orders of magnitude. That is to say, we have a resonance of width 10^{-5} A on top of a width of 10^{-2} A. The widths increase with laser intensity.

It is possible to give a simple explanation for this structure and for the intensity dependence of the widths. We have previously seen that the multiphoton resonance acts like a two-level atom but with an effective Rabi frequency. Indeed, this idea has been used for many years as a means of modeling multiphoton processes. We use perturbation theory: the effective Rabi frequency for a k-photon resonance is the product of k successive Rabi frequencies divided by (k-2) successive cumulative detunings (Figure 10).

This procedure gives us a simple way to compare the widths of the various resonances; they are in the ratios of their effective Rabi frequency (Figure 11). The table displays these, ranked across according to the detuned frequency of the resonance and down according to the initial level.

N-LEVEL QUANTUM SYSTEM



Atom:

Bohr frequencies $\Delta E_n/\hbar$
Dipole moments d_n

Laser:

Frequencies ω_n
Intensities I_n

Interaction:

Rabi frequencies $\Omega_n = d_n \sqrt{\frac{8\pi I_n}{c}}$

Assume:

Stimulated >> incoherent

N-LEVEL RWA EQUATION

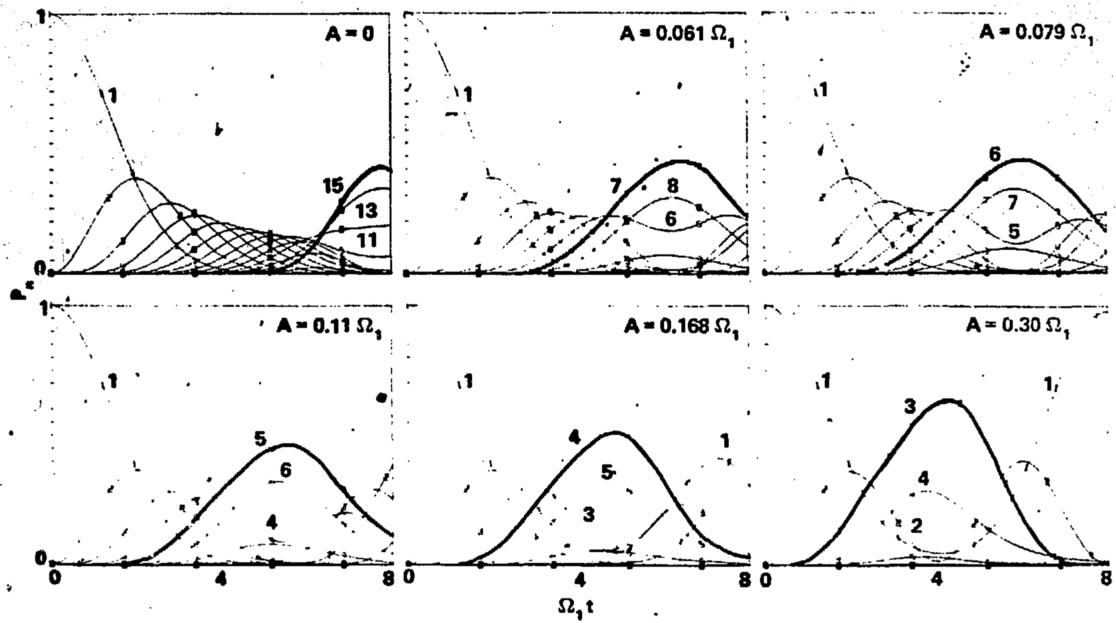
$$
i\frac{d}{dt}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ \vdots \\ C_{n-1} \\ C_n \end{bmatrix} = \begin{bmatrix} D_1 & \frac{1}{2}\Omega_1 & O & & & \\ \frac{1}{2}\Omega_1 & D_2 & \frac{1}{2}\Omega_2 & & & \\ O & \frac{1}{2}\Omega_2 & D_2 & & & \\ & & & \ddots & & \\ & & & & D_{n-1} & \frac{1}{2}\Omega_{n-1} \\ & & & & \frac{1}{2}\Omega_{n-1} & D_{n-2} \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ \vdots \\ C_{n-1} \\ C_n \end{bmatrix}
$$

Oscillator: $\Omega_n = \sqrt{n}\ \Omega_1$

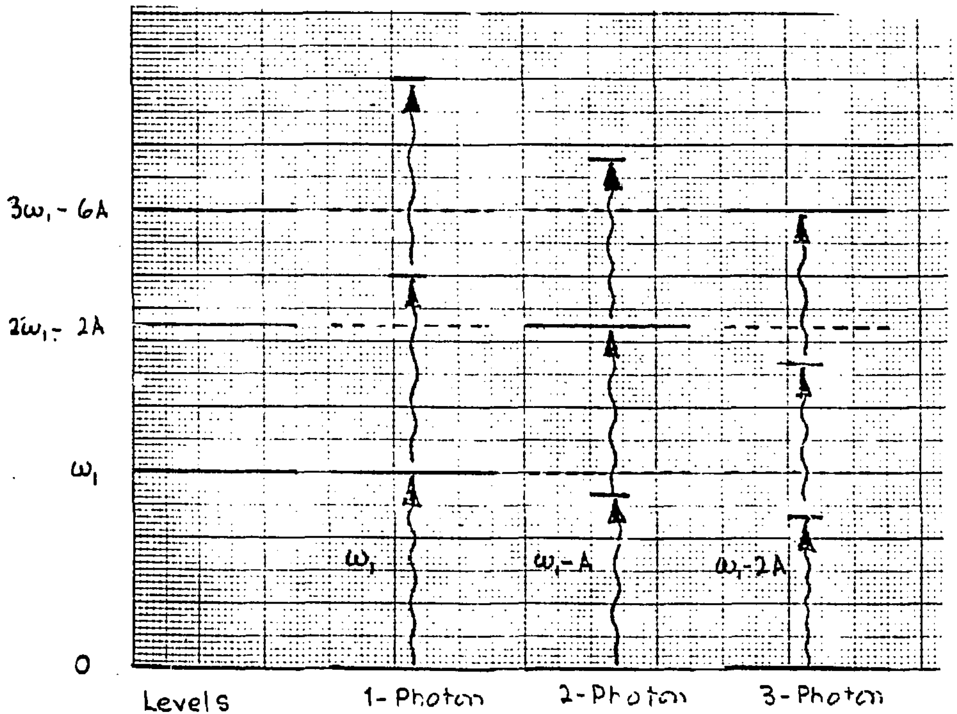
Equal Rabi: $\Omega_n = \Omega_1$

Anharmonic: $D_n = n - 1(\omega - \omega_o) + (n-1)(n-2)\ A$

HARMONIC RABI, $\omega = \omega_1$



MULTIPHOTON RESONANCES

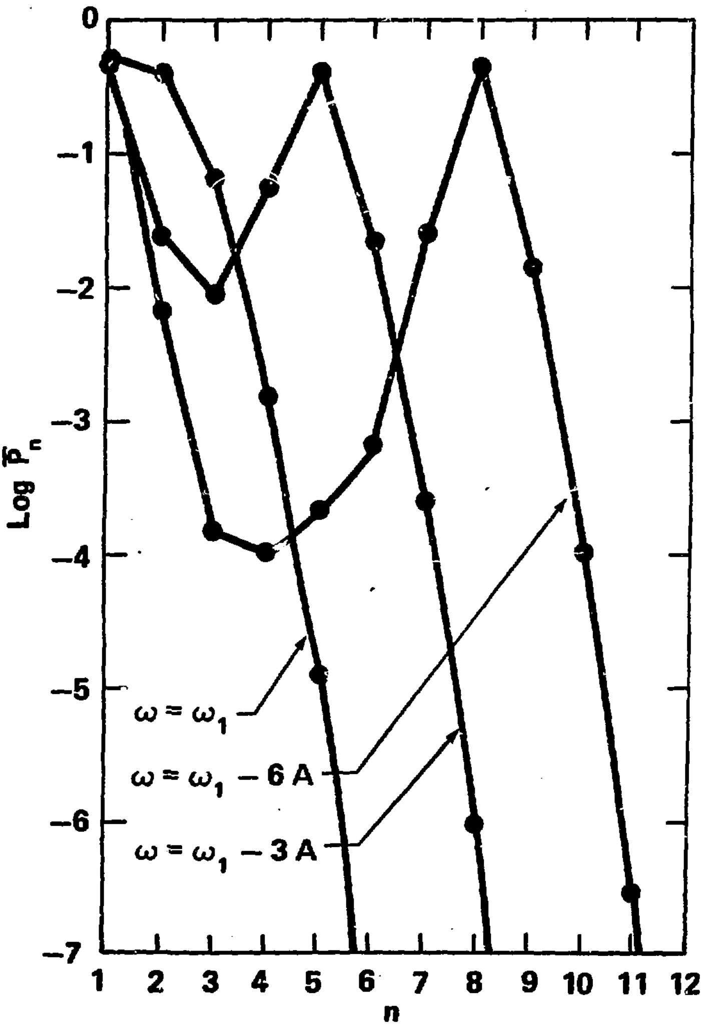


RESONANT VALUES OF $(\omega_1 - \omega)/A$

	n′ = 2	3	4	5	6	7	8	9	10
n = 1	0	1	2	3	4	5	6	7	8
2		2	3	4	5	6	7	8	9
3			4	5	6	7	8	9	10
4				6	7	8	9	10	11

For $n \rightarrow n'$ resonance: $\omega_1 - \omega = (n + n' - 3)\, A$

RESONANT ENHANCEMENT, $\Omega_1 = A$



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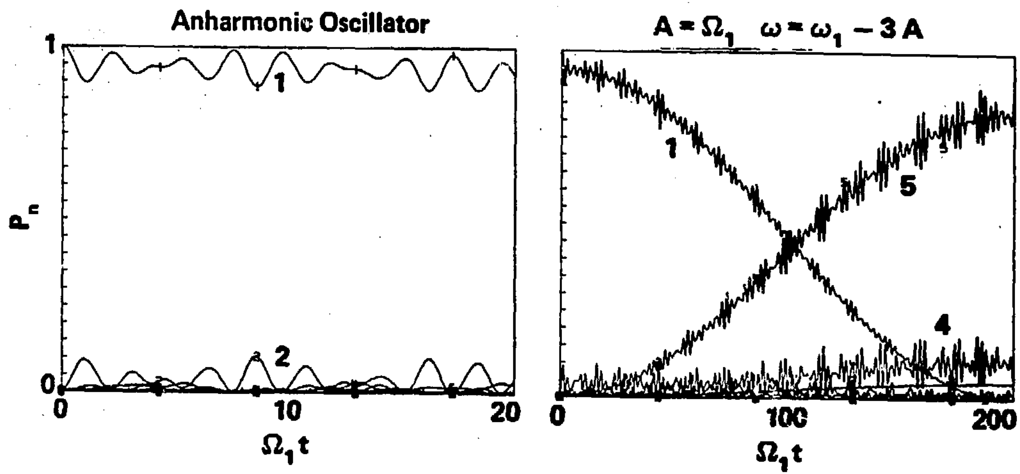
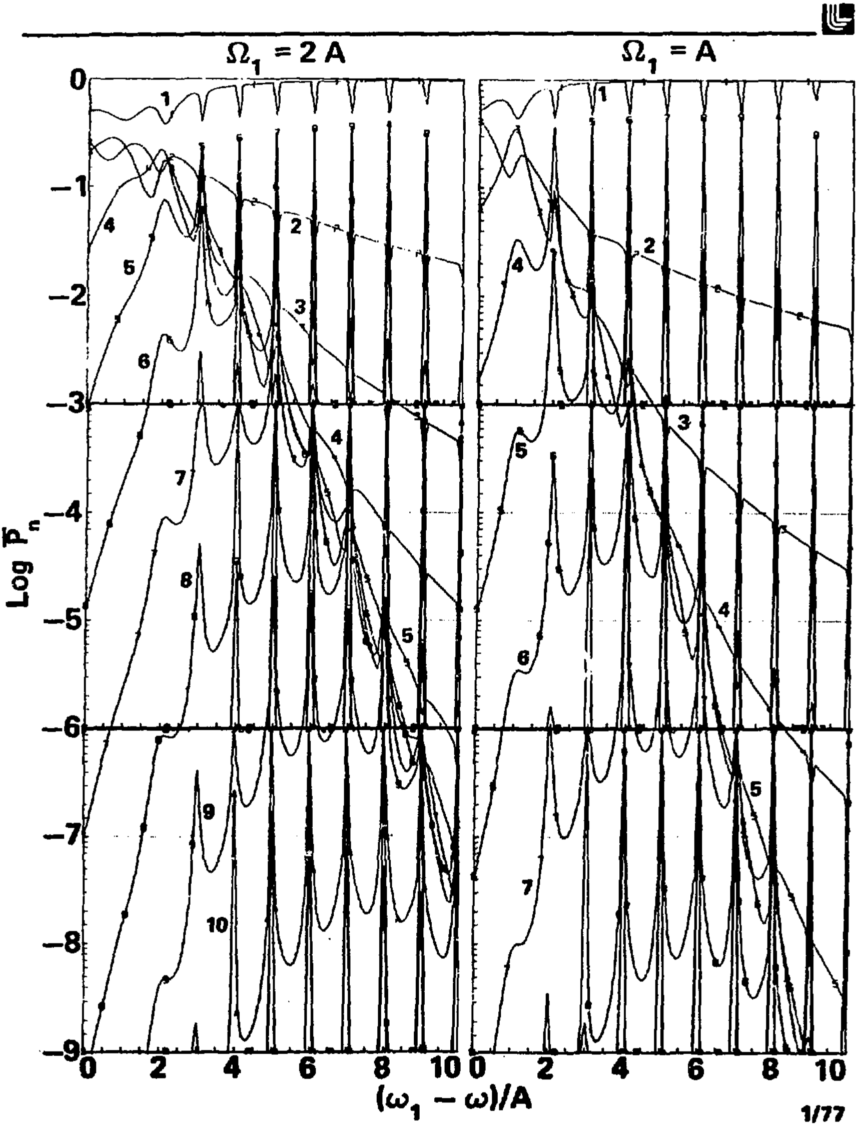


Anharmonic Oscillator
1
2
0
P_n
0
10
20
Ω_1 t
A = Ω_1   ω = ω_1 − 3 A
1
5
4
0
100
200
Ω_1 t



$\Omega_1 = 2A$
$\Omega_1 = A$
Log $\bar{P}_n$
0
−1
−2
−3
−4
−5
−6
−7
−8
−9
0 2 4 6 8 10
$(\omega_1 - \omega)/A$
1 2 3 4 5 6 7 8 9 10

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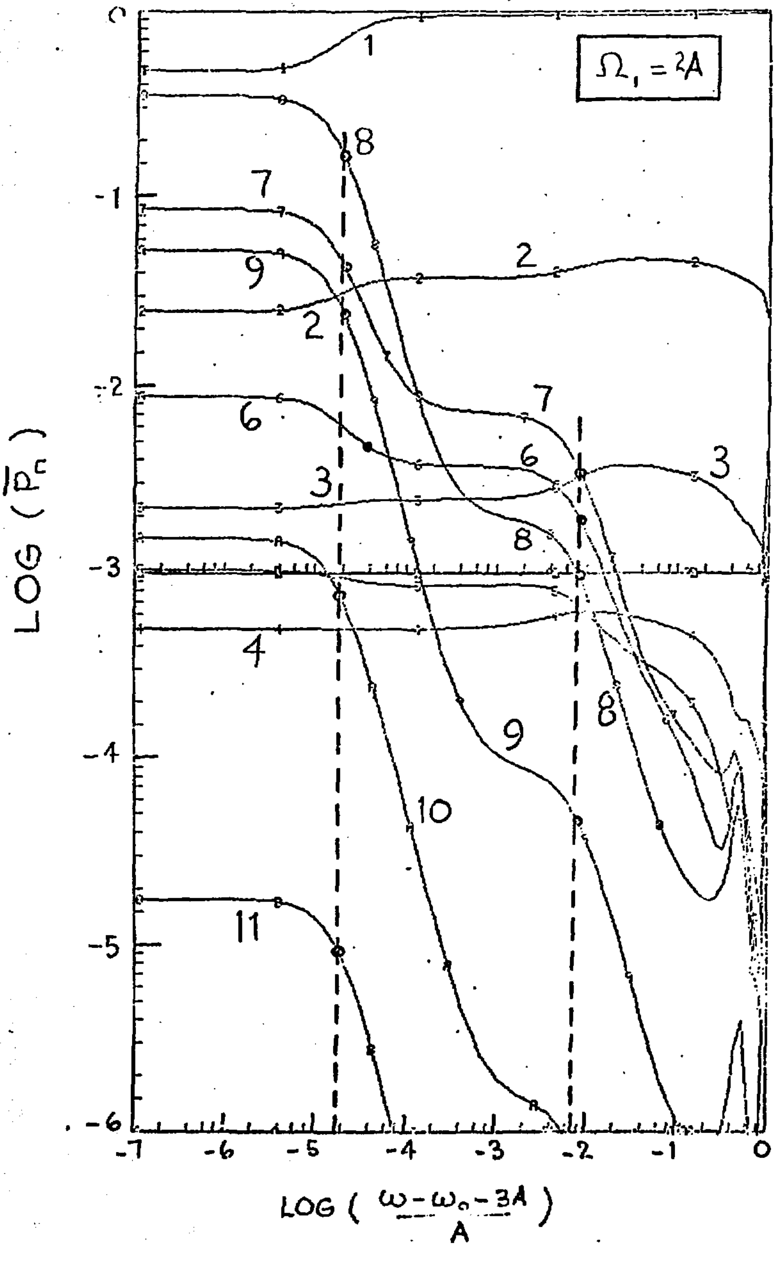
HARMONIC RABI, $\omega \cong \omega_0 + 3A$

Fig. 9



MULTIPHOTON RESONANCE $n \rightarrow m = n+k$

RWA Matrix;

$$\begin{bmatrix} D_n & \frac{1}{2}\Omega_n & & & \\ & D_{n+1} & \frac{1}{2}\Omega_{n+1} & & \\ & & \ddots & & \\ & & & D_{m-1} & \frac{1}{2}\Omega_{m-1} \\ & & & & D_m \end{bmatrix}$$

Effective Rabi Frequency;

$$\frac{1}{2}\tilde{\Omega} = \frac{(\frac{1}{2}\Omega_n)(\frac{1}{2}\Omega_{n+1})\cdots(\frac{1}{2}\Omega_{m-1})}{(D_{n+1})(D_{n+2})\cdots(D_{m-1})}$$

$$= \left(\frac{\Omega_1}{2A}\right)^k \left[\frac{(n-1)!}{(n+k-2)!}\right]^2$$

Equal Rabi

$$= \left(\frac{\Omega_1}{2A}\right)^k \left[\frac{(n-1)!}{(n+k-2)!}\right]^2 \left[\frac{(n+k-1)!}{(n-1)!}\right]^{\frac{1}{2}}$$

Harmonic Rabi

EFFECTIVE RABI FREQUENCY (Uniform Rabi Freq.)

$\left(\frac{\Delta_1}{A}\right) =$	0	1	2	3	4	5
	$1 \rightarrow 2$	$1 \rightarrow 3$	$1 \rightarrow 4$	$1 \rightarrow 5$	$1 \rightarrow 6$	$1 \rightarrow 7$
	x	$x^2/2$	$x^3/16$	$x^4/288$	$x^5/9216$	$x^6/5\times10^5$
			$2 \rightarrow 3$	$2 \rightarrow 4$	$2 \rightarrow 5$	$2 \rightarrow 6$
			x	$x^2/8$	$x^3/144$	$x^4/4608$
					$3 \rightarrow 4$	$3 \rightarrow 5$
					x	$x^2/8$

$x = \Omega_1/A$