Driven Morse oscillator: Classical chaos and quantum theory for two-frequency excitation

M. E. Goggin* and P. W. Milonni
Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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We extend our previous comparison of classical and quantum theories for the excitation and dissociation of a sinusoidally driven Morse oscillator [Phys. Rev. A **37**, 796 (1988)] to the case of two-frequency driving. In both the classical and quantum theories the total threshold intensity for dissociation can be considerably smaller in the two-frequency case. This is consistent with results obtained with more artificial models of laser-driven molecular systems, and we provide an approximate resonance-overlap analysis to explain this trend. We also compare results obtained with adiabatic and sudden turn-on of the applied field, and comment on the use of absorbing boundaries for the computational identification of dissociation or ionization.

I. INTRODUCTION

Recently we compared the classical and quantum theories for a Morse oscillator driven by a monochromatic electric field.[1] Using Morse parameters appropriate to the HF molecule, we focused attention on the classical and quantum predictions for the photodissociation rate and threshold field strength. One of the main conclusions from that study was that the classical and quantum theories are in surprisingly good agreement except for driving frequencies near multiphoton resonances[2] or nonlinear classical resonances of order $N>1$. The disagreement at the higher-order nonlinear resonances was explained in terms of classical resonance overlap. In particular, the disagreement between the classical and quantum theories is most pronounced when the difference between the appropriate classical and quantum resonance frequencies exceeds the width of the classical resonance zone.

Classically, dissociation is associated with resonance overlap and the onset of chaos. We argued in Ref. 1 that in some ways a quantum analogue for classical resonance overlap and chaos is the coupling of a large number of energy levels by the applied field. For very strong fields the number of levels mixed by the field is large enough for substantial continuum excitation (dissociation), in analogy to the diffusive excitation and dissociation found classically when resonance overlap occurs. Semiclassical quantization suggests that the number of levels with significant occupation probability should be proportional to the square root of the applied electric-field amplitude, and this surmise was confirmed in numerical experiments on the driven Morse oscillator.[1]

The fact that the classical and quantum theories come into better agreement as the width of the resonance zones (and therefore the applied field strength) increases may be understood intuitively by comparing the width (ΔJ_N) of a classical resonance with the fundamental quantum unit of action $\hbar$. In the notation of Ref. 1 we have

$$\Delta J_N/\hbar=(K\mid A_N\mid)^{1/2}\ , \tag{1.1}$$

where K is proportional to the amplitude of the applied electric field. For a Morse oscillator initially in the ground state we calculate $\Delta J_1/\hbar=0.32K^{1/2}$ for an $N=1$ resonance. Thus $\Delta J_1/\hbar<1$ for $K<10$, and so we expect that K should be large compared with 10 in order to have reasonably good agreement between the classical and quantum theories for an initially unexcited Morse oscillator. This was indeed the case in Ref. 1, where we focused our attention on the large field amplitudes necessary for photodissociation. In addition, we showed a case for $K=10$ (Figs. 7 and 8 of Ref. 1) for which the classical and quantum theories were not in good qualitative agreement even for an $N=1$ classical resonance. This is consistent with the fact that $\Delta J_1/\hbar$ in this case is not large. In fact, Eq. (1.1) indicates that the classical and quantum theories come into better agreement at larger field amplitudes, a result noted in Ref. 1 and in an earlier paper by Shirts and Davis.[3]

Equation (1.1) may also be invoked to understand why the classical and quantum theories for the driven Morse oscillator differ most significantly at the higher-order classical resonance frequencies: $\mid A_N\mid$, and therefore $\Delta J_N/\hbar$, decreases with increasing N, and so larger pump amplitudes K are required to realize the "classical regime" $\Delta J_N/\hbar\gg1$. For an initially unexcited Morse oscillator and a driving frequency corresponding to a classical $N=4$ resonance, for instance, we require $K=3.4\times10^4$ in order to have $\Delta J_4/\hbar=1$. This is much larger than the critical K value necessary for dissociation given in Fig. 9 of Ref. 1, and so the large discrepancy between the classical and quantum predictions in this case is not surprising.

In the case of the monochromatically driven surface-state electron, or one-dimensional hydrogen atom,[4] the condition $\Delta J_N/\hbar\gg1$ may be shown to take the form

$$\mid 4e^3E_0NJ_N'(N)/3\mid^{1/2}\gg\hbar\omega\ , \tag{1.2}$$

where E_0 is the amplitude of the applied field of angular frequency ω and J_N' is the derivative of the Bessel function of order N. Alternatively, (1.2) may be written in terms of the Keldysh adiabatic tunneling parameter γ: $\gamma\ll\frac{1}{3}J_N'(N)n_0^2$, where n_0 is the principal quantum number corresponding to the classical orbit. Numerical esti-

mates indicate that the condition $\Delta J_N/\hbar \gg 1$ is well satisfied in recent experiments on the microwave ionization of highly excited ($n_0 \approx 66$) hydrogen, for which classical theory has been found to be a remarkably good predictor of the field strength necessary for 10% ionization.[5]

Of course the simple condition $\Delta J_N/\hbar \gg 1$ cannot be regarded in general as sufficient for the accuracy of classical theory and provides only a rough first estimate for when classical theory might be expected to make reasonable predictions.[6] In the case of the kicked pendulum, for instance, $\Delta J/\hbar = 2\sqrt{K}/\tau$, but a purely quantum-localization effect occurs regardless of how large K is made.[7]

Previously we suggested that *quasiperiodically* driven quantum systems, although not chaotic in the strict sense of exponential sensitivity of the state vector to initial conditions, can nevertheless exhibit properties that mimic those of chaotic systems, especially when a large number of incommensurate frequencies are involved in the dynamics.[1,8] We argued, therefore, that "statistical" features like ergodicity or effectively diffusive energy growth do not necessarily require an underlying chaos. In a quasiperiodically kicked two-state quantum system, for instance, there is "ergodicity" on the Bloch sphere, although of course the dynamics of the state vector is not chaotic.[8] And in the quasiperiodically kicked pendulum there is diffusive energy growth, at least over very long times, in contrast to the quantum localization that suppresses the energy growth in the case of the periodically kicked pendulum.[8,9] The fact that statistical properties like ergodicity can be exhibited when there are many incommensurate frequencies in a nonchaotic system has been recognized for a long time. Two important examples we noted earlier are the work of Montroll and Mazur on the ergodic properties of coupled harmonic oscillators[10] and Slater's theory of unimolecular reactions.[11]

Recent experiments of Moorman *et al.*[12] indicate that highly excited hydrogen atoms are more unstable (easier to ionize) when the microwave field is bichromatic rather than monochromatic. We are unaware of any theoretical work on the two-frequency microwave ionization of hydrogen.[13] Numerical experiments of Noid and Stine,[14] however, indicated that a Morse oscillator is more easily dissociated when two laser frequencies are used rather than one. Their treatment assumed the validity of classical dynamics. Later quantum calculations for two-frequency driving using a simpler model[15] showed agreement with the classical theory in that two-frequency driving produced greater excitation. That model, however, could not adequately describe a dissociation process.

It is the purpose of this paper to report the results of rather extensive quantum computations on the two-frequency excitation and dissociation of the Morse oscillator. We also discuss, in the following section, two aspects of this kind of calculation that should be relevant more generally to other systems, namely the effects of sudden field turn-on and artificial absorbing boundaries used to define ionization (or dissociation). Our main results for two-frequency excitation are summarized in Sec. III. Generally we have found, in agreement with the classical results of Noid and Stine, that two-frequency driving lowers the threshold for dissociation. In Sec. IV we provide a heuristic explanation, based on classical resonance overlap, for the tendency of two-frequency driving to lower the dissociation threshold. Our results are summarized in Sec. V.

II. REMARKS ON FIELD TURN-ON AND ABSORBING BOUNDARIES

In Ref. 1 we used the interaction Hamiltonian

$$H_I(t) = -d_1 x\, E_0 \cos(\omega t) \tag{2.1}$$

and assumed that this interaction was effectively switched on suddenly at $t=0$. From an experimental perspective a more realistic interaction would take the form

$$H_I(t) = -d_1 x E(t) \cos(\omega t)\ , \tag{2.2}$$

where the envelope function $E(t)$ takes into account the turning on and off of the field. It is easy to account for such effects in our numerical solution of the Schrödinger partial differential equation as in Ref. 1. We have found, generally speaking, that the artificial sudden turn-on of the field leads to greater excitation of the Morse oscillator compared with the case of an adiabatic turn-on of the field.

For this study we used the interaction Hamiltonian

$$\begin{aligned} H_I(t) &= -KX \sin^2(\mu\tau/4a)\cos(\mu\tau), \quad 0 < \tau < 2\pi a/\mu \\ &= -KX \cos(\mu\tau), \quad 2\pi a/\mu < \tau \end{aligned} \tag{2.3}$$

where the notation is that of Ref. 1 [cf. Eq. (4.9)]. The parameter a is the number of cycles of the field required to "turn on" the field. Both quantum and classical computations, as described previously,[1] were carried out for the interaction (2.3). Typical results are shown in Fig. 1.

It is seen in Fig. 1 that the amount of energy absorbed by the "molecule" depends in a significant way on the field turn-on time. Both the classical and quantum theories predict that a gradual turn-on of the field reduces the excitation energy compared with what is obtained assuming a sudden turn-on. In light of the two-frequency results in the following section, this is not surprising, as a sudden turn-on introduces additional frequency components in the driving field. These results indicate that a detailed comparison of theory and experiment in such computations will generally require one to take into account the details of the field turn-on.

It has sometimes been suggested that the replacement of $\cos(\mu\tau)$ by $\sin(\mu\tau)$ in the interaction (2.1) will mitigate the effects of the artificial sudden turn-on, since then the field is initially zero. We have found, in both the classical and quantum theories of the driven Morse oscillator, that the difference arising from this replacement is quite small compared with that found by assuming an adiabatic turn-on requiring, say, ten cycles or more.

In our previous work[1] we determined the dissociation probability by computing the probability at a given time that the system is in any of the bound states. The dissociation probability was then defined by subtracting this bound-state projection from unity. This approach,

though physically transparent, in practice limits one to the regime of small dissociation probabilities, since the part of ψ with a nonvanishing projection onto the continuum will propagate to the edge of the finite spatial grid assumed in the numerical computations. This gives rise to spurious reflections from the boundaries once the wave function acquires a substantial continuum component. In a similar (ionization) problem Kulander[16] used an absorbing boundary to effectively capture that part of ψ extending to the edge of the grid and so to identify the ionization rate. This technique allows one to go beyond the regime of small dissociation (or ionization) probability. More importantly, this approach may be used in cases where a projection onto all bound states is impractical, as in the case of the hydrogen atom.

We have examined the accuracy of the absorbing-boundary approach for the example of the Morse oscillator, where a comparison can be made with the "exact" results obtained by projection onto the bound states. An example is shown in Fig. 2, which compares the dissocia-

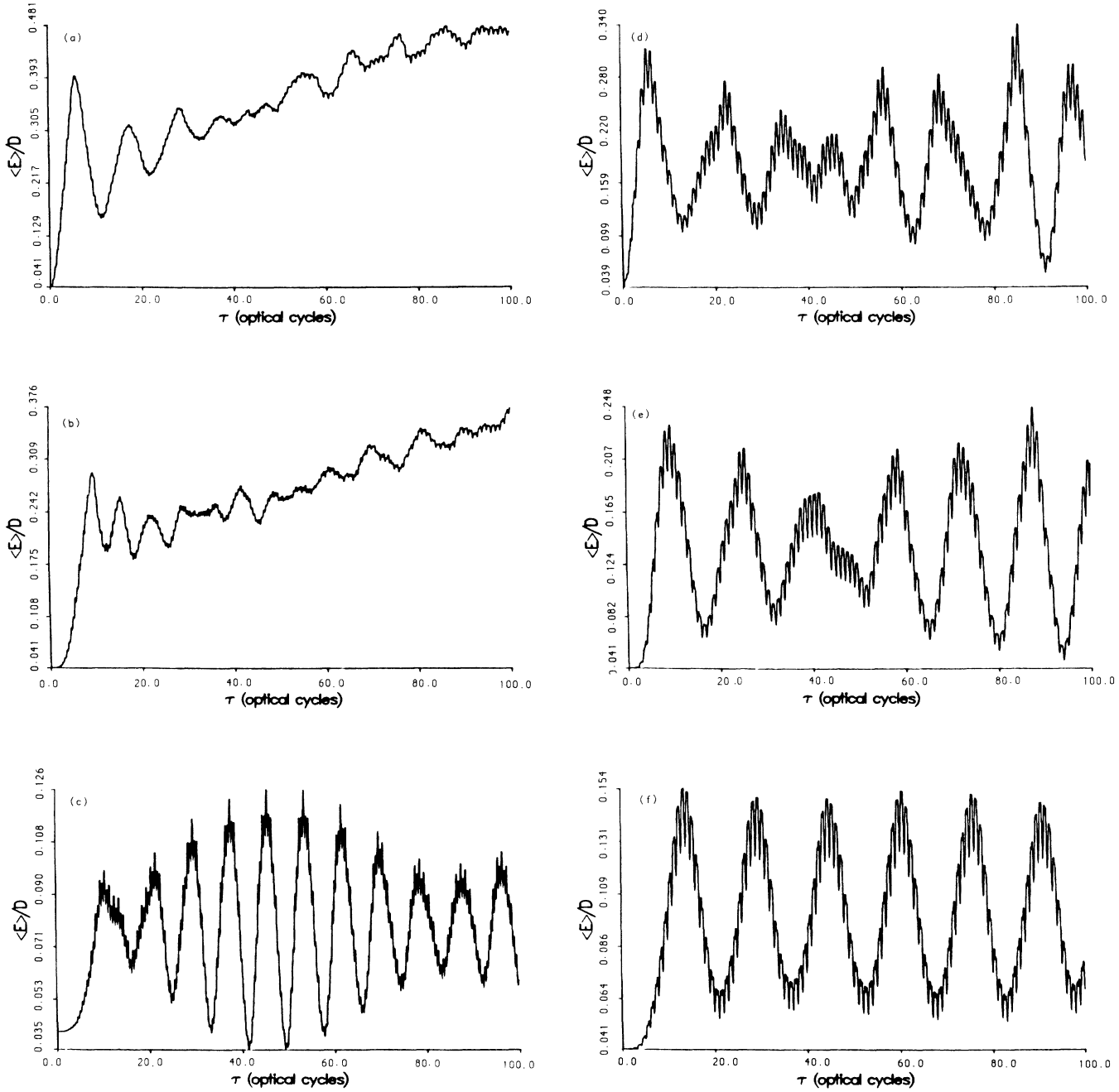

FIG. 1. Energy absorbed by a Morse oscillator with initial energy equal to the quantum-mechanical ground-state energy, $\mu=40$ and $K=45$ for (a),(d) sudden turn-on of the field; (b),(e) turn-on over five optical cycles; and (c),(f) turn-on over ten optical cycles. Plots (a)–(c) are classical calculations and plots (d)–(f) are quantum calculations.

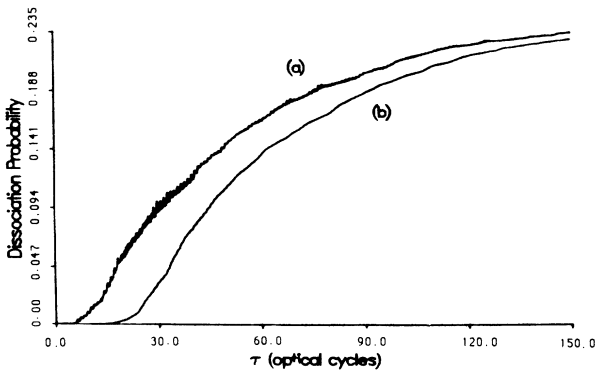

FIG. 2. Dissociation probability calculated using (*a*) projection onto the bound states and (*b*) an absorbing boundary.

tion probability obtained by (*a*) projection onto the bound states and (b) the removal of ψ from the grid. The wiggles in curve (*a*) correspond to bound-continuum-bound transitions, and suggest that not all of the continuum part of ψ "dissociates" immediately. We will refer to that part of ψ that does dissociate immediately as the "dissociative part" of ψ. The time delay between the two curves shown in Fig. 2 is due to the finite time required for the dissociative part of ψ to propagate to the absorbing boundary and register "dissociation." Both computations were done using an absorbing boundary on the grid so that large dissociation probabilities could be compared. The use of an absorbing boundary in the calculation of curve (*a*) has only a small effect on the result. We note, for instance, the significant excitation of the continuum compared with the magnitude of the bound-continuum-bound transitions before the dissociated part of ψ reaches the boundary [when (*b*) becomes nonzero], and the fact that the bound-continuum-bound transitions remain after the dissociated part of ψ reaches the boundary. Apart from the small oscillations and the time delay the two curves are practically the same and give the same dissociation rate. We have confirmed in such examples that a sufficiently distant absorbing boundary provides an accurate measure of dissociation (or ionization) in numerical solutions of the time-dependent Schrödinger equation.

III. TWO-FREQUENCY EXCITATION

In the case of two driving frequencies we use the interaction Hamiltonian

$$H_I(t) = -d_1 x [E_1 \cos(\omega_1 t) + E_2 \cos(\omega_2 t)] . \tag{3.1}$$

The classical Newton equation for the Morse oscillator may be conveniently scaled as follows:

$$\frac{d^2X}{d\tau^2} = -(4/B^2)(e^{-X} - e^{-2X}) + 2K_1 \cos(\mu_1 \tau) + 2K_2 \cos(\mu_2 \tau) \tag{3.2}$$

in the notation of Ref. 1. Similarly, the Schrödinger equation may be scaled to the form

$$\frac{i\partial\psi}{\partial\tau} = -\frac{\partial^2\psi}{\partial X^2} + B^{-2}(1-e^{-X})^2\psi - X[K_1 \cos(\mu_1 \tau) + K_2 \cos(\mu_2 \tau)]\psi . \tag{3.3}$$

As in Ref. 1 we compare the classical theory to the quantum by averaging over an ensemble of classical trajectories with different initial conditions.

Figure 3 shows typical results for one- and two-frequency driving. In this figure one of the driving frequencies is held fixed at $\mu_1 = 42.0$, red-shifted from the value 45.637 for an $N=1$ classical resonance,[1] and the amplitude for this frequency is fixed at $K_1 = 30$. As the second driving frequency μ_2 is scanned, the corresponding amplitude K_2 necessary for dissociation is determined both classically and by numerical integration of the Schrödinger equation, as described earlier. The scaled intensity plotted versus μ_2 in Fig. 3 is proportional to the total intensity associated with both frequencies. As in Ref. 1 we note that the classical and quantum predictions for the threshold intensity are in fairly good agreement. Both theories predict that two-frequency driving lowers the threshold intensity needed for dissociation, i.e., dissociation is more easily accomplished with two-frequency driving.

Similar conclusions apply for excitation of the Morse oscillator below the dissociation threshold. In Fig. 4 we show the maximum energy absorbed as a function of μ_1 and μ_2 for an intensity for which there is no dissociation probability; for simplicity both frequency components have the same amplitude. Generally speaking we can say that a Morse oscillator tends to absorb more energy when it is driven bichromatically than when it is driven monochromatically. As discussed earlier, this trend has been observed in other driven nonlinear oscillator systems. We note, however, that, just as there are optimal frequencies for monochromatic excitation, there are optimal frequency combinations for bichromatic excitation. In particular, if one of the frequencies, say, μ_1, is near the optimal frequency for monochromatic excitation, then the

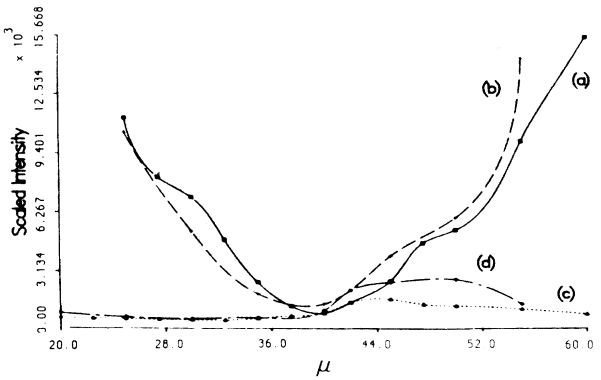

FIG. 3. Threshold intensity for 5% dissociation for (*a*),(*b*) monochromatic driving and (*c*),(*d*) bichromatic driving. Curves (*a*) and (*c*) are the result of classical calculations and curves (*b*) and (*d*) are the result of quantum calculations. $I(\mathrm{TW/cm^2}) = 0.1578 \times$(scaled intensity).

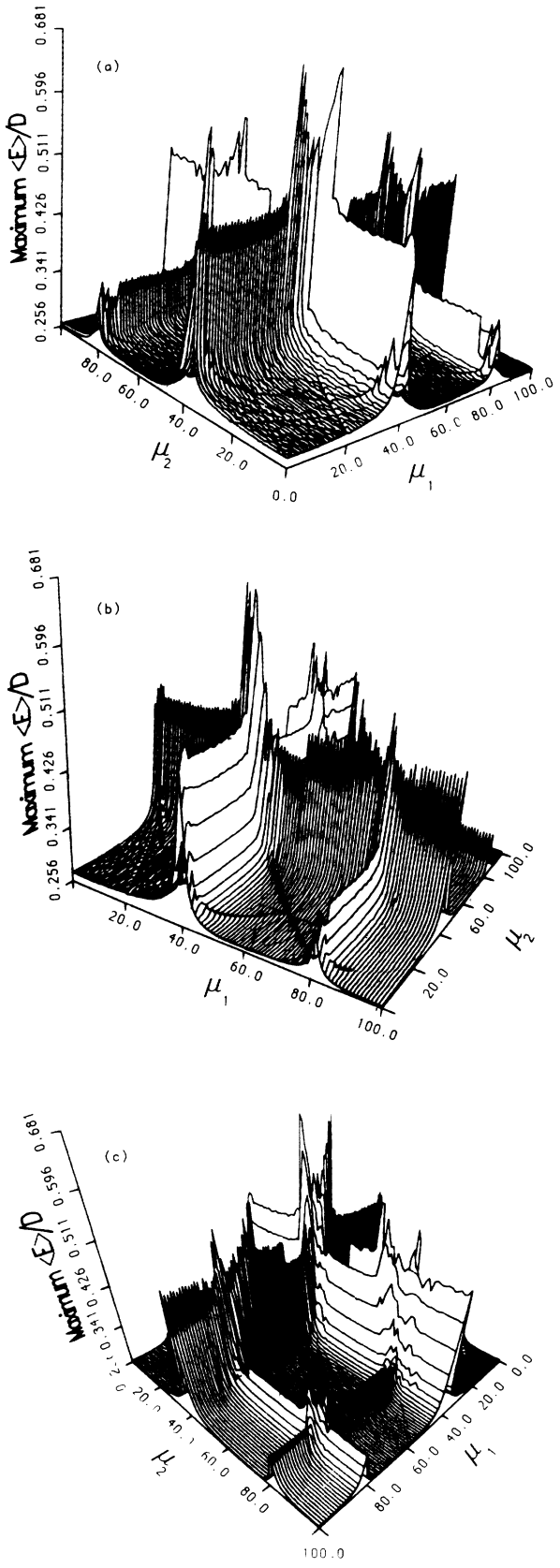

FIG. 4. Maximum energy absorbed by a classical Morse oscillator as a function of μ_1 and μ_2 with $K=5$ and initial energy equal to $\frac{1}{4}$ the dissociation energy.

addition of a second frequency slightly red-shifted from μ_1 will greatly enhance excitation. In Sec. IV we propose an explanation based on an extension of lowest-order classical resonance-overlap analysis.

A somewhat surprising feature of Fig. 4 is the existence of deep "valleys" or holes in the spectrum at $\mu_1 \approx 38$ and $\mu_2 \approx 42-53$ and vice versa. Thus there exist frequency combinations which reduce the excitation of the oscillator. We have found, however, that these holes disappear for sufficiently high values of the driving field K. Since the values of K for which the holes disappear are well below the threshold for dissociation, we conclude that the holes only affect excitation and not dissociation.

IV. RESONANCE ZONES FOR ONE- AND TWO-FREQUENCY DRIVING

According to the classical resonance-overlap criterion for Hamiltonian systems, the onset of chaos is associated with the overlap of neighboring resonance zones in action-angle space.[17] For the driven Morse oscillator a resonance zone may be associated with a driving frequency $\approx\omega_N = N\omega(J)$, where ω is the oscillator frequency, a function of the action J. This defines what may be called a classical N resonance. The resonance-overlap criterion may be cast in the form[1]

$$\Delta\omega_{N+1}+\Delta\omega_N > \omega_{N+1}-\omega_N \ , \tag{4.1}$$

where $\Delta\omega_N$ is the width in the resonance frequency ω_N corresponding to the width ΔJ_N of the action J_N defined by $N\omega(J_N)=\omega$: $\Delta\omega_N \approx \omega_N'(J_N)\Delta J_N$. That is, $\Delta\omega_N$ is the spread in frequency about ω_N for which an N-resonance may be realized. [An expression for ΔJ_N is given in Eq. (2.16) of Ref. 1.] Loosely speaking, then, the resonance-overlap criterion is just the condition that two neighboring resonance zones can be strongly coupled by a single driving frequency $\approx\omega_N$.

We found previously that the resonance-overlap condition for the onset of chaos provides a reasonably accurate value of the driving amplitude needed for dissociation. Typically the critical K value predicted by the resonance-overlap condition is about two to three times larger than what is found in "exact" numerical experiments. The association of resonance overlap, and therefore chaos, with dissociation is reasonable, in that a chaotically meandering trajectory eventually finds its way to the continuum.

It seems worthwhile for our present purposes to elaborate somewhat on the resonance-overlap analysis presented earlier for single-frequency driving.[1] The corrections to the lowest-order perturbation theory discussed below will then be generalized to the more complicated case of two-frequency driving. We begin, therefore, with the Hamiltonian

$$H(J)=H_0(J) - \sum_{N=0}^{\infty} V_N(J)[\cos(N\theta-\omega t)+\cos(N\theta+\omega t)] \ . \tag{4.2}$$

$H_0(J)$ is the unperturbed Hamiltonian and $V_N(J)$ is proportional to the driving amplitude and to the coefficient of $\cos(N\theta)$ in the Fourier expansion of the unperturbed coordinate $x(J,\theta)$. (The unperturbed motion is periodic and as such may always be represented by a Fourier series.) From this form of the Hamiltonian we obtain the action-angle equations

$$\dot{J} = -\sum_{N=0}^{\infty} NV_N(J)[\sin(N\theta-\omega t)+\sin(N\theta+\omega t)] , \tag{4.3}$$

$$\dot{\theta} = \omega_0(J) - \sum_{N=0}^{\infty} V_N'(J)[\cos(N\theta-\omega t)+\cos(N\theta+\omega t)] . \tag{4.4}$$

In the absence of the driving force J is, of course, constant, and we denote this constant initial value of J by $\bar{J}$. Let us assume that the driving force is a weak enough perturbation that

$$J = \bar{J} + \Delta J, \quad |\Delta J/\bar{J}| \ll 1 . \tag{4.5}$$

Then,

$$H \approx H_0(J) - \sum_{N=0}^{\infty} V_N(\bar{J})[\cos(N\theta-\omega t)+\cos(N\theta+\omega t)] - \Delta J \sum_{N=0}^{\infty} V_N'(\bar{J})[\cos(N\theta-\omega t)+\cos(N\theta+\omega t)] \tag{4.6}$$

and

$$\Delta\dot{J} \approx -\sum_{N=0}^{\infty} NV_N'(\bar{J})[\sin(N\theta-\omega t)+\sin(N\theta+\omega t)] . \tag{4.7}$$

Under the further approximation that $\dot{\theta} \approx \omega_0(\bar{J})$ we have

$$\Delta J \approx \sum_{N=0}^{\infty} NV_N'(\bar{J})\{[N\omega_0(\bar{J})-\omega]^{-1}\cos(N\theta-\omega t) + [N\omega_0(\bar{J})+\omega]^{-1}\cos(N\theta+\omega t)\} \tag{4.8}$$

in a lowest-order classical perturbation theory.

The approximation (4.8) indicates that the perturbation becomes strong when $\omega \approx N\omega_0(\bar{J})$, i.e., near an N resonance. Figure 5(a) shows the peak value of the (oscillatory) energy of a driven Morse oscillator as a function of the scaled driving frequency μ for a fixed driving amplitude $K=1$. This plot is obtained by averaging over 20 trajectories, but the basic resonance structure shown is approximately the same for each individual trajectory. In particular, the resonance structure predicted by (4.8) is clearly evident. The width of each N resonance increases with K as $\sqrt{K}$, so that an overlapping of resonances occurs when K is increased. Figure 5(b) shows the broadening of the resonances when K is raised to 20. This figure reveals also that there are additional resonances, at $\omega \approx \omega_0(\bar{J})/2, 3\omega_0(\bar{J})/2, \ldots$, that are not predicted by the lowest-order perturbation theory leading to (4.8).

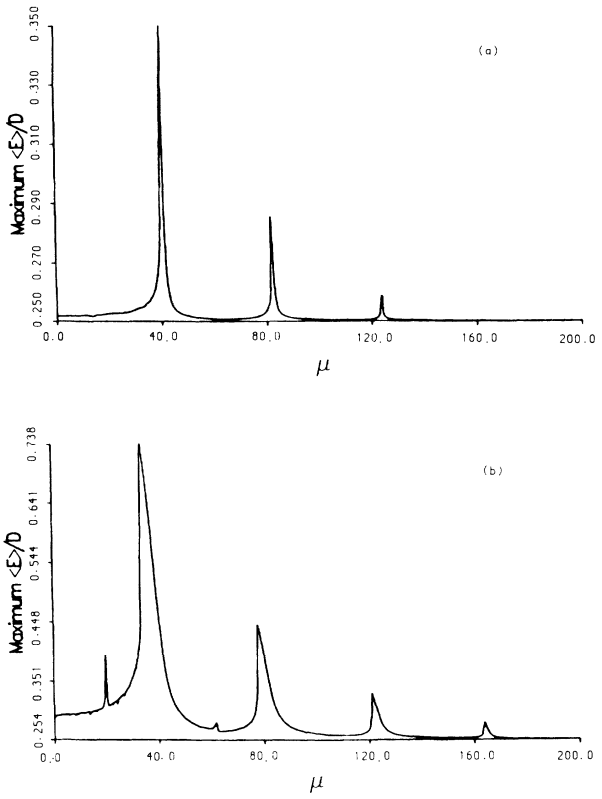

FIG. 5. Maximum energy absorbed by a classical Morse oscillator as a function of μ with an initial energy equal to $\frac{1}{4}$ the dissociation energy and (a) $K=1$, (b) $K=20$.

These additional resonances are brought out by carrying the simple analysis above one step further. Using (4.8) in (4.6) and ignoring terms with $N\omega_0(\bar{J})+\omega$ in the denominator, we obtain

$$H \approx H_0(J) - \sum_{N=0}^{\infty} V_N(\bar{J})[\cos(N\theta-\omega t)+\cos(N\theta+\omega t)] - \tfrac{1}{2}\sum_{N=0}^{\infty}\sum_{M=0}^{\infty} MV_M'(\bar{J})V_N'(\bar{J})[M\omega_0(\bar{J})-\omega]^{-1} \times\{\cos[(M+N)\theta-2\omega t]+\cos(M-N)\theta+\cos(M+N)\theta+\cos[(M-N)\theta-2\omega t]\} . \tag{4.9}$$

Thus at this level of approximation we find resonances not only at $\omega \approx N\omega_0(\bar{J})$, but also at $\omega \approx \frac{1}{2}(M\pm N)\omega_0(\bar{J})$, $M,N=0,1,2,\ldots$. In particular, we have the subharmonic resonances at $\omega_0(\bar{J}), 3\omega_0(\bar{J})/2, \ldots$ seen in Fig. 5(b).

These additional resonances make it easier for resonance overlap to occur at large driving amplitudes. This accounts in part for the fact that in numerical experiments we find critical K values for dissociation smaller than those predicted based on the lowest-order resonance-overlap analysis without the subharmonics. Recently Gu and Yuan[18] have found a series of secondary resonances for the Morse oscillator corresponding to $\omega = n/(n-1)\omega_0(\bar{J})$, $n = 2,3,4,\ldots$. Our results imply, however, that the resonances $\omega = (M \pm N)\omega_0(\bar{J})/2$ contribute the most to excitation and dissociation.

We now turn our attention to the case of two-frequency driving, starting in this case from the Hamiltonian

$$H = H_0(J) - \sum_{N=0}^{\infty} V_N(J)[\cos(N\theta - \omega_1 t) + \cos(N\theta + \omega_1 t) + \cos(N\theta - \omega_2 t) + \cos(N\theta + \omega_2 t)] \tag{4.10}$$

and assuming for the sake of the present discussion that the two fields have the same amplitude. Proceeding as above we obtain

$$\begin{aligned} H \approx{}& H_0(J) - \sum_N V_N(\bar{J})[\cos(N\theta - \omega_1 t) + \cos(N\theta + \omega_1 t) + \cos(N\theta - \omega_2 t) + \cos(N\theta + \omega_2 t)] \\ & - \tfrac{1}{2} \sum_N \sum_M N V_N'(\bar{J}) V_M'(\bar{J}) \\ & \times ([M\omega_0(\bar{J}) - \omega_1]^{-1} \{\cos[(M+N)\theta - 2\omega_1 t] + \cos(M-N)\theta + \cos(M+N)\theta \\ & \quad + \cos[(M-N)\theta - 2\omega_1 t] + \cos[(M+N)\theta - (\omega_1+\omega_2)t] \\ & \quad + \cos[(M-N)\theta - (\omega_1-\omega_2)t] + \cos[(M+N)\theta - (\omega_1-\omega_2)t] \\ & \quad + \cos[(M-N)\theta - (\omega_1+\omega_2)t]\} \\ & + [M\omega_0(\bar{J}) - \omega_2]^{-1} \{\cos[(M+N)\theta - 2\omega_2 t] + \cos(M-N)\theta + \cos(M+N)\theta + \cos[(M-N)\theta - 2\omega_2 t] \\ & \quad + \cos[(M+N)\theta - (\omega_1+\omega_2)t] + \cos[(M-N)\theta + (\omega_1-\omega_2)t] \\ & \quad + \cos[(M+N)\theta + (\omega_1-\omega_2)t] + \cos[(M-N)\theta - (\omega_1+\omega_2)t]\}) . \end{aligned} \tag{4.11}$$

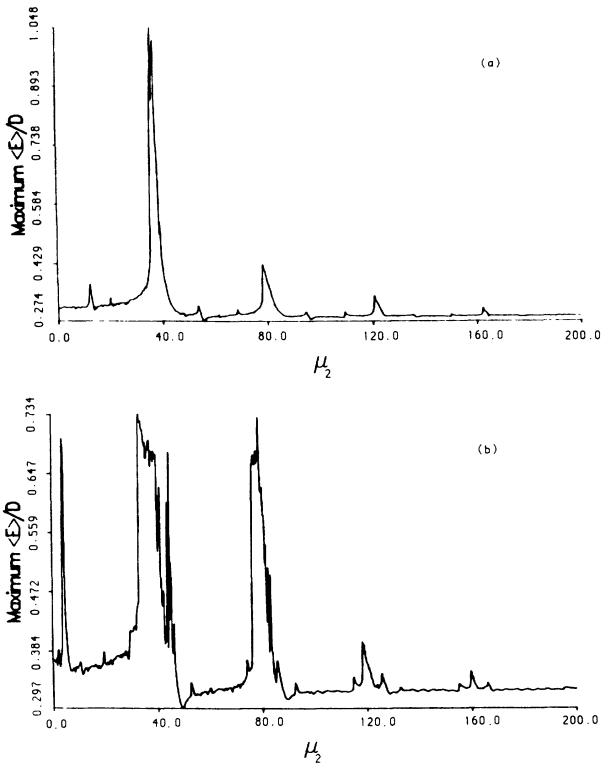

FIG. 6. Maximum energy absorbed by a bichromatically driven classical Morse oscillator starting with energy equal to $\frac{1}{4}$ the dissociation energy as a function of μ_2 with $K = 10$ and (a) $\mu_1 = 27.5$ and (b) $\mu_1 = 34.25$.

In this case we find "additional" resonances at

$$|\omega_1 \pm \omega_2| \approx |M \pm N|\,\omega_0(\bar{J}) \tag{4.12a}$$

and

$$|\omega_1 \mp \omega_2| \approx |M \pm N|\,\omega_0(\bar{J}) . \tag{4.12b}$$

The frequency denominators in (4.11), however, indicate that these two-frequency resonances are only strong when at least one of the frequencies is close to an N-resonance.

In Fig. 6 we show typical results for the maximum energy absorbed as a function of the second frequency μ_2 for two different, fixed, values of μ_1 and the same value of K. We clearly see the additional resonances of Eq. (4.12). Note the shift in the secondary peaks when μ_1 is changed. These secondary resonances can also be seen as small ridges in the "flat" regions of Fig. 4.

V. SUMMARY

In this paper we have extended previous work on the driven Morse oscillator[1] to include bichromatic driving. We have found that, in general, bichromatic driving enhances excitation and dissociation when compared to monochromatic driving, and that there are optimal frequency combinations for the bichromatic driving. An explanation has been presented based on an extension of lowest-order classical resonance overlap.

In addition, we have compared the results of classical and quantum calculations of the threshold field intensity for 5% dissociation for both monochromatic and bi-

chromatic driving and found good agreement over most of the frequency range studied. There are, however, frequencies for which the classical and quantum calculations clearly differ. As dissociation of classical trajectories is a consequence of resonance overlap resulting in diffusive energy growth, it would appear that this system offers an alternative to the microwave ionization of hydrogen for testing the existence of quantum chaos. Unfortunately, the intensities required for dissociation are so great (> 100 TW/cm^2) that a more sophisticated model, which takes into account the ionization as well as dissociation of the diatomic molecule, is required.

Of interest in comparing theoretical predictions with experimental results is the effect of adiabatic versus sudden turn-on of the driving field. We find that sudden turn-on significantly increases the excitation of a Morse oscillator when compared to the excitation produced by a field which turns on over ten or more optical cycles.

Handling dissociation (or ionization) adequately is a problem when solving the Schrödinger equation numerically. One way of treating dissociation (or ionization) is to use an absorbing boundary on the spatial grid and measure the disappearance of the wave function. We have tested the accuracy of this method and found that it provides an excellent means of handling dissociation (or ionization).

We have also given a simple criterion for when we expect reasonably good agreement between classical and quantum mechanics [Eq. (1.1)]. Not surprisingly the parameters used in our calculations of the threshold field intensity for dissociation satisfy this criterion. We believe that the good agreement between classical and quantum mechanics when describing a classically chaotic system bodes well for the coexistence of quantum mechanics and chaos, even though quantum chaos may not exist in any rigorous sense.

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*Present address: Physics Dept., Box 3D, New Mexico State University, Las Cruces, NM 88003.

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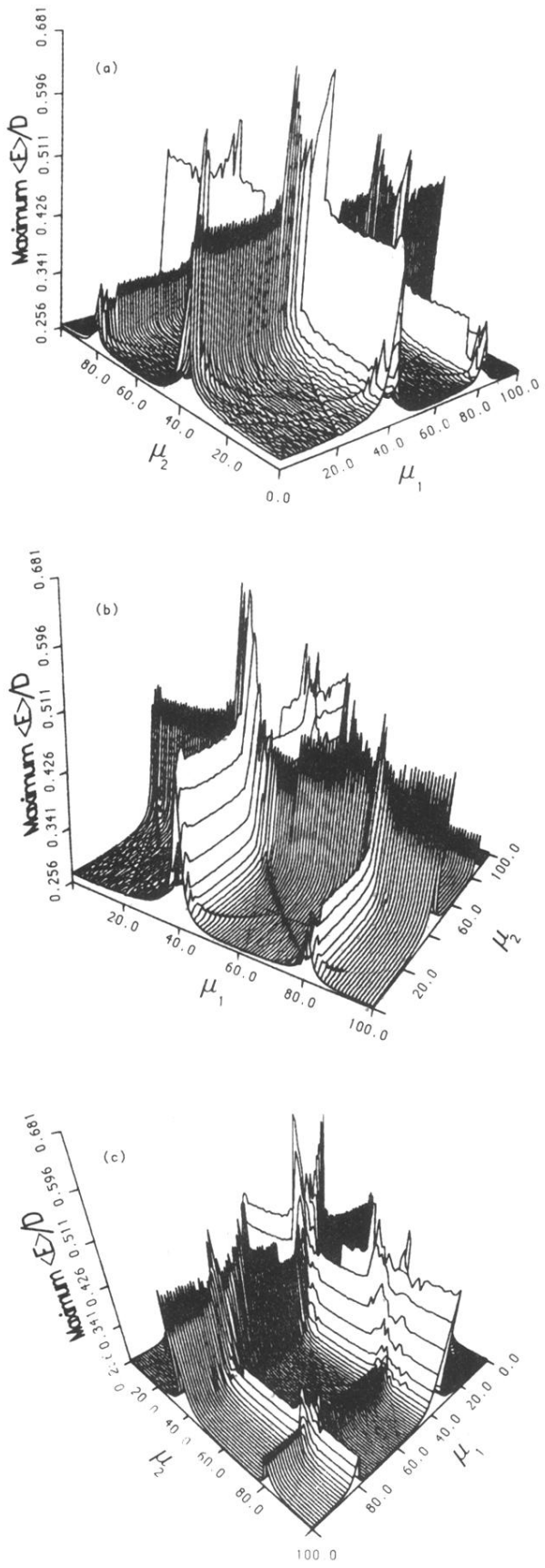

FIG. 4. Maximum energy absorbed by a classical Morse oscillator as a function of μ_1 and μ_2 with $K=5$ and initial energy equal to $\frac{1}{4}$ the dissociation energy.