Supporting Information for:

Quantum Interstate Phase Differences and Multiphoton Processes:

Quantum Jumps or Dynamic Beats?

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Section 1. Morse Oscillator as an Illustrative Model for Vibrational Excitation

The Morse oscillator [P. M. Morse, "Diatomic Molecules According to the Wave Mechanics. II. Vibrational Levels," *Phys. Rev.* **34**, 57–64 (1929); also R. B. Shirts, "Use of Classical Fourier Amplitudes as Quantum Matrix Elements: A Comparison of Morse Oscillator Fourier Coefficients with Quantum Matrix Elements," *J. Phys. Chem*. **91**, 2258–2267 (1987)] is a useful approximate description of the stretching motion of a diatomic molecule. The Hamiltonian that describes the molecular kinetic and potential energy is

$$H_0 = \frac{-\hbar^2}{2m}\frac{\partial^2}{\partial q^2} + D\left(1 - e^{-\alpha q}\right)^2, \tag{S1}$$

where the parameters are the dissociation energy, D, the reduced mass, m, and the length

parameter, α. The coordinate, q, is the deviation of the bond length from its equilibrium value, q

$= R - R_e$. When an example is needed here, we will choose parameters appropriate for the

hydrogen fluoride molecule, HF: $m = 1.589229 \times 10^{-24}$ g, $D = 6.123$ eV, and $\alpha = 2.2188 \times 10^8$

cm^{-1} [B. G. Dibble and R. B. Shirts, Theoretical Prediction of Ultrahigh Vibration Excitation

Using Picosecond IR Pulse Trains: Coherent Absorption of Several Photons Each of Different

Frequency, *J. Chem. Phys.* **94**, 3451–3467 (1991)]. The energy levels and zero-order

wavefunctions of the Morse oscillator are well known [Morse, (1929) and Dibble/Shirts (1991)].

The energy levels are given by:

$$E_n^0 = \hbar\left(n + 1/2\right)\omega_0 - \frac{\hbar^2\left(n + 1/2\right)^2\omega_0^2}{4D}, \tag{S2}$$

for $n = 0, 1, 2,\ldots n_{max} - 1$, where $\omega_0 = \alpha(2D/m)^{1/2}$ is the classical frequency of vibration in the

low energy limit. For the HF parameters above, $n_{max} = 24$ and there are 24 bound states.

Section 2. Electric Dipole Coupling

When a diatomic molecule interacts with an electromagnetic field, a suitable time-

dependent Hamiltonian for vibration is [L. I. Schiff, *Quantum Mechanics*, 3rd ed. (McGraw-Hill,

New York, 1968), p. 177:

$$H(\vec{x}, t) = \sum_j \frac{1}{2m_j}\left[-i\hbar\nabla_j - \frac{e_j}{c}A(\vec{x}, t)\right]^2 + V(\vec{x}) + e\,\Phi(\vec{x}), \tag{S3}$$

where e_j is the charge of particle j, c is the speed of light, and A and Φ are the vector and scalar

potentials which describe the field. Because of the gauge flexibility in the field, we can choose $\Phi(x) = 0$ and $\nabla \cdot \mathbf{A} = 0$ (Coulomb gauge) without loss of generality [Schiff (1968), p. 177)]. Thus:

$$H(\vec{x},t) = -\sum_j \frac{\hbar^2}{2m_j} \nabla_j^2 + V(\vec{x}) + \sum_j \left[\frac{i\hbar e_j}{m_j c} A(\vec{x},t) + \frac{e_j^2}{2m_j c^2} A \cdot A \right]. \tag{S4}$$

This Hamiltonian can now be separated into time-independent and time-dependent parts. The first two terms are the time-independent Hamiltonian, $H_0(q)$, and the latter two are the time dependent Hamiltonian, $H_1(q, t)$. Since the first term of H_1 dominates the second in most situations, we will neglect the A^2 term. Ignoring uniform translational motion and overall rotation of the molecule, the time-independent terms can be rewritten in terms of the relative motion of the atoms in a diatomic molecule to obtain Eq. (S1). Using a gauge transformation, the time-dependent term can be written as the dipole moment dotted into the electric field vector of the electromagnetic field [R. B. Shirts and T. F. Davis, "Classical Resonance Analysis of Conservative Models of Infrared Multiphoton Absorption," *J. Phys. Chem.* **88**, 4665–4671 (1984)]:

$$H = \frac{-\hbar^2}{2m} \frac{\partial^2}{\partial q^2} + D\left(1 - e^{-\alpha q}\right)^2 - \mu(q) \cdot E(t), \tag{S5}$$

where m is the reduced mass of the diatomic molecule, and the Morse potential from Eq. (S1) is an excellent approximation for $V(q)$; $\mu(q)$ parameterizes the dipole moment of the molecule as it vibrates (for HF, we will use the parameterization of Ogilvie, *et al.* [J. F. Ogilvie, W. R. Rodwell and R. H. Tipping, "Dipole moment functions of the hydrogen halides," *J. Chem. Phys.* **73**, 5221–5229 (1980) and Dibble/Shirts (1991)], and $E(t)$ is the projection of the electric field on the internuclear axis of the molecule. Numerically calculated matrix elements of the dipole moment operator for HF are tabulated in Section 5 below.

Section 3. Perturbation Theory Beyond the Rotating Wave Approximation

The inclusion of the counter-rotating term in Eq. (11) into Eq. (5) through perturbation theory can be done in several ways. Shirley [J. H. Shirley, "Solution of the Schrodinger Equation with a Hamiltonian Periodic in Time," *Phys. Rev.* **138**, B979–B987 (1965)] has given a clear exposition of one method based on Floquet theory. We have investigated another method which gives good results for the cases that we have checked. Our method consists of treating the counter-rotating term in Eq. (11) as a perturbation in ordinary, Raleigh-Schrödinger perturbation theory. We take

$$a_n = \sum_{k=0}^{\infty} \lambda^k a_n^{(k)} \tag{S6}$$

where λ is a dummy perturbation parameter. A factor of λ is likewise placed in the counter-rotating term. Restricting ourselves to two levels and using the same notation at Section IIA, substitution in Eq. (5) and equating terms with equal powers of λ yields the following equations for the amplitudes

$$\dot{a}_1^{(k)} = iV a_2^{(k)} e^{i(\delta t + \gamma)} + iV a_2^{(k-1)} e^{-i(\varepsilon t + \gamma)}$$

$$\dot{a}_2^{(k)} = iV a_1^{(k)} e^{-i(\delta t + \gamma)} + iV a_1^{(k-1)} e^{i(\varepsilon t + \gamma)} \tag{S7}$$

where $\omega - \omega_{21} = \delta$ is the detuning and $\omega + \omega_{21} = \varepsilon$ is the counter-rotating-wave frequency. The zero-order solution to these equations is just the Rabi solution, Eq. (13). The solution for higher orders requires solution of a second-order inhomogeneous differential equation. There can be several inhomogeneous terms, but since the equation is linear and since the inhomogeneous terms are all of the same form, the solution is straightforward though tedious. For small detunings and for small fields (δ, $V \ll \varepsilon$) and retaining only terms of the lowest order in V, the

amplitude of the upper state is given by

$$a_2^{(1)} = \frac{\varepsilon V}{\Omega \left(\varepsilon^2 + V^2 \right)} \left[\Omega \cos\left(\Omega t\right) + i \frac{\delta}{2} \sin\left(\Omega t\right) e^{i\left(\varepsilon t + \delta t/2 + \gamma\right)} \right] \tag{S8}$$

Near resonance, the frequency of this term is $\varepsilon \approx 2\omega$. This term gives, to first order in V, the size and phase of the fast oscillations in Figs. 1 and 2 (neglecting diagonal matrix elements of the dipole moment).

Section 4. Beats Between Coupled Classical Oscillators

This section reviews several features of classical, coupled harmonic oscillator systems which have behavior strictly analogous to that shown by the quantum systems discussed in the accompanying paper. First, consider the simple, familiar harmonic oscillator Hamiltonian

$$H = \frac{p^2}{2m} + \frac{kq^2}{2}, \tag{S9}$$

where q is the coordinate, p is the canonical momentum conjugate to q, m is the mass, k is the force constant, and the angular oscillation frequency is $\omega = (k/m)^{1/2}$. A canonical transformation to action-angle variables (I, θ) can be made (H. Goldstein, *Classical Mechanics*, 2nd ed. (Addison-Wesley, Reading, Mass., 1980), p. 462) where

$$I = \frac{1}{2\pi} \oint p \, dq, \tag{S10}$$

where the integral is over one cycle of the motion and

$$\tan\theta = \frac{-p}{q\sqrt{mk}}. \tag{S11}$$

In these variables the Hamiltonian transforms to

$$H = I\omega, \tag{S12}$$

and the motion is described simply by a time invariant action, I, and a linearly evolving angle

variable:

$$\theta = \omega t + \theta_0, \tag{S13}$$

where θ_0 is the initial phase. Motion can be considered to be in a two-dimensional plane with horizontal q-axis and vertical p-axis, where rotation is in a clockwise direction. The radius of circular motion is constant only when q is scaled by $(mk)^{1/2}$ and is given by $(2I(mk)^{1/2})^{1/2}$.

Now consider two coupled classical oscillators with unit masses and coupling constant k_{12}:

$$H = \frac{1}{2}\left(p_1^2 + p_2^2 + k_1 q_2^2 + k_2 q_2^2\right) + k_{12} q_1 q_2 = \frac{1}{2}\left(p_x^2 + p_y^2 + \omega_x^2 x^2 + \omega_y^2 y^2\right), \tag{S14}$$

where the transformation between the numerically subscripted variables and the x, y subscripted normal modes is given by the F_3-type generating function (Goldstein, p. 384):

$$F_3 = - p_1\left(x\cos\phi + y\sin\phi\right) - p_2\left(- x\sin\phi + y\cos\phi\right) \tag{S15}$$

This transformation is a simple rotation of coordinate system by an angle ϕ, where

$$\tan 2\phi = \frac{2 k_{12}}{k_2 - k_1}, \tag{S16}$$

and

$$\omega_{x,y}^2 = \frac{1}{2}\left(k_1 + k_2\right) \pm \frac{1}{2}\sqrt{\left(k_2 - k_1\right)^2 + 4 k_{12}^2}. \tag{S17}$$

If all of the energy of this system is placed in oscillator #1 at time $t = 0$, this system exhibits classical beats in which the energy flows from oscillator #1 to oscillator #2 and returns with frequency $\omega_x - \omega_y$ and with maximum energy in oscillator #2 given by

$$E_{2max} = \sin^2\left(2\phi\right) = \frac{1}{1 + \dfrac{\left(k_2 - k_1\right)^2}{4 k_{12}^2}} = \frac{1}{1 + \dfrac{\left(\omega_2 - \omega_1\right)^2 \omega_{avg}^2}{k_{12}^2}}, \tag{S18}$$

where the zero-order oscillator angular frequencies are $\omega_i = k_i^{1/2}$ for $i = 1, 2$. The maximum energy in oscillator #2 has a resonant shape (Lorentzian) as a function of frequency difference, with half width at half height k_{12}/ω_{avg}, where $\omega_{avg} = (\omega_1 + \omega_2)/2$ and falling off as the inverse square of the frequency difference.

For near-resonant, weakly-coupled oscillators ($|k_{12}|, |k_2 - k_1| << k_2, k_1$), the period for energy exchange as a function of frequency has a resonance shape with maximum when $k_2 = k_1$, with half-width at half height approximately $(3)^{1/2}k_{12}/\omega_{avg}$ and falling off as the inverse of the absolute frequency difference, $|\omega_1 - \omega_2|$.

If the phase angles of oscillators #1 and #2 are expressed in terms of angle-like variables:

$$\tan\theta_1 = \frac{-p_1}{k_1^{1/2}q_1} \; ; \qquad\qquad \tan\theta_2 = \frac{-p_2}{k_2^{1/2}q_2} \; . \qquad\qquad (S19)$$

The difference in these two phase angles, $d_{21} = \theta_2 - \theta_1 - \pi/2$, shows the same behavior shown in Fig. 3 of the accompanying paper for a quantum two-state system. If a resonant system ($k_2 = k_1$) starts with $p_1 = 1$, $q_1 = q_2 = p_2 = 0$, then d_{21} quickly locks into a value near zero so oscillator #1 drives oscillator #2 and remains in the same phase relationship until all the energy is transferred to oscillator #2. Then d_{21} jumps by $-\pi$, the phase relationship reverses, and oscillator #2 drives oscillator #1 until all the energy is transferred back to oscillator #1. If the system is nonresonant ($k_2 \neq k_1$), the initial phase difference drifts away from the initial favorable relationship, and the extent of energy transfer is limited by the time d_{21} stays near zero (see Fig. 3 and compare with Fig. S1). Similar behavior is also shown by driven oscillator systems as contained in typical first-year physics texts. We propose that any coherent process can be examined in terms of phase differences and understood in terms of periodic phase reversals such as are shown by this classical system.
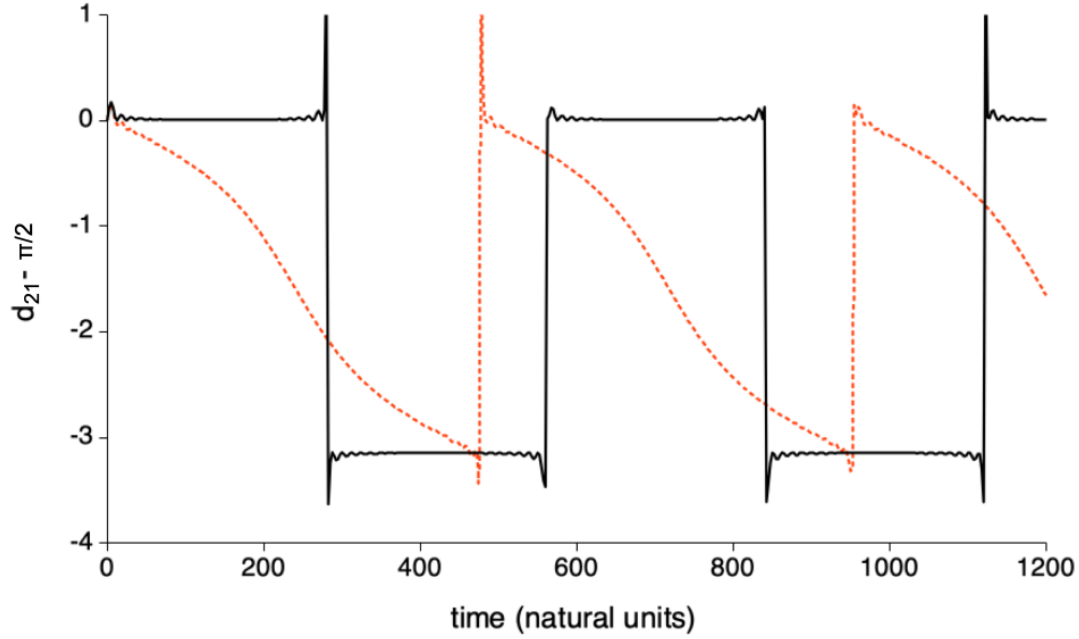
Figure S1. Classical beats for the Hamiltonian given by Eq. (S14). Plots show $d_{21} = \theta_2 - \theta_1 - \pi/2$ versus time for two trajectories. The action-like variables θ_1 and θ_2 are defined by Eq. (S19). The black, on-resonance plot used $k_1 = k_2 = 1$ and $k_{12} = 0.0112$. The red, dashed, plot for negative detuning used $k_1 = 1.014$, $k_2 = 1$, $k_{12} = 0.0112$. For both cases, initial conditions were $p_1 = 1$, $p_2 = q_1 = q_2 = 0$. Classical beat motion is analogous to the quantum dynamics shown in Fig. 3 of the accompanying paper.

Now consider a system of three coupled harmonic oscillators

$$H = \frac{1}{2}\left(p_1^2 + p_2^2 + p_3^2 + k_1 q_2^2 + k_2 q_2^2 + k_3 q_3^2\right) + k_{12} q_1 q_2 + k_{23} q_2 q_3 = \frac{1}{2}\left(p_x^2 + p_y^2 + \omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2\right), \quad (S20)$$

where the transformation from numerically subscripted variables to x, y, z subscripted normal modes can be obtained by a Fourier transform and the solution of a cubic polynomial.

For a suitable choice of parameters, this system displays the same kind of beats seen in quantum two-photon absorption. Fig. S2 shows the energies of three oscillators where initially the energy is placed in oscillator #1. The parameters chosen are $k_1 = k_3 = 1.01$, $k_2 = 1$, and $k_{12} = k_{23} = 0.003$ with initial conditions $p_1 = 2^{1/2}$, $p_2 = p_3 = q_1 = q_2 = q_3 = 0$. The frequency mismatch

between oscillator #1 and oscillator #2 is somewhat greater than the dashed curve in Fig. S1, and

the energy in oscillator #2 only reaches 0.2 before the phase reversal occurs. However, the phase

difference between oscillator #2 and #3 allows most of that energy to flow into oscillator #3, and

after several cycles between #1 and #2, the energy is transferred eventually completely to #3 at

about $t = 4000$. Then the cycle reverses. This transfer only occurs for a narrow frequency range

if ω_1 is varied.   The half-width at half height for energy transfer to #3 is approximately

$k_{12}k_{23}/(\delta\omega_2)$, where δ is the frequency difference between oscillator #1 and oscillator #2.
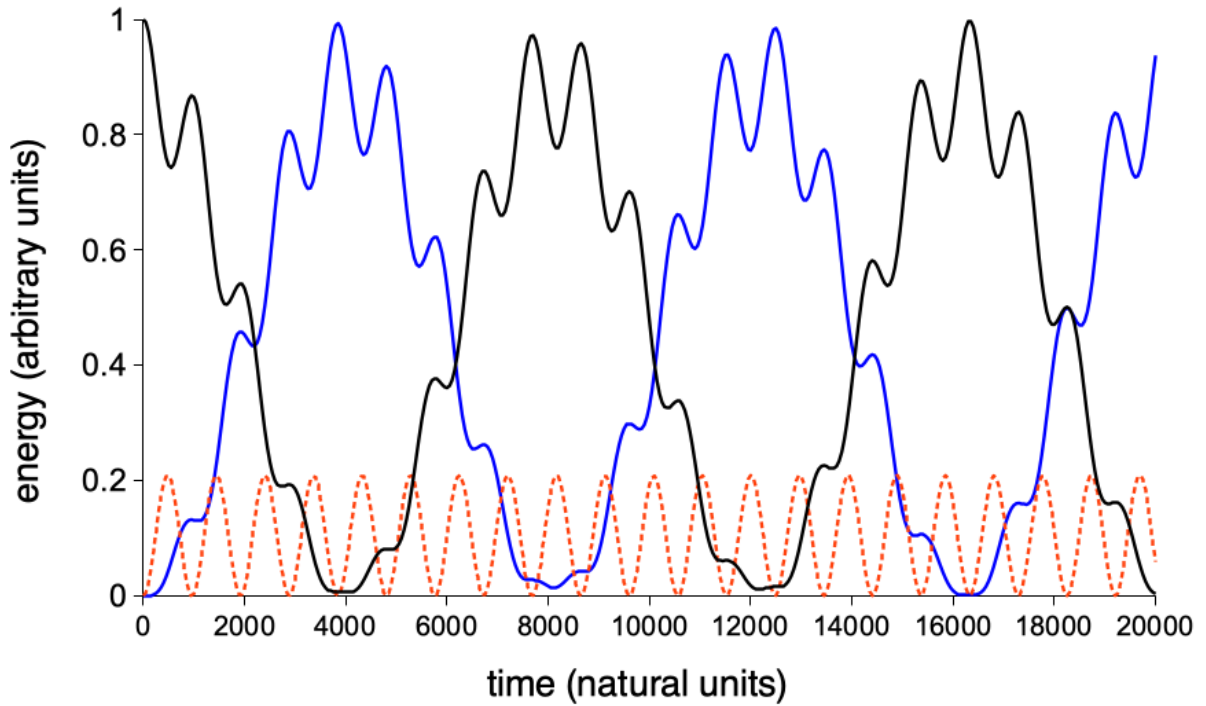
Figure S2. Energy as a function of time for three coupled oscillators defined by Eq. (S20) with $k_1 = k_3 = 1.01$, $k_2 = 1$,

$k_{12} = k_{23} = 0.003$ with $p_1 = 2^{1/2}$ initially and all other variables zero. The black line is for E_1, red dashed line is for E_2

and the blue line is for E_3. Detuning between #1 and #2 limits energy transfer, but resonance between #1 and #3

overcomes the detuning and allows for complete energy transfer in repeating classical beats with a period of about

8000 time units. Compare with quantum dynamics of two-photon absorption in Fig. 6 of the accompanying paper.

     Figure S3 shows, for the same trajectory of Fig. S2, the double angle difference, $d_{21} - d_{23}$,

where $d_{ij} = \theta_i - \theta_j - \pi/2$ as defined before. This variable shows the same periodic phase reversals as the analogous phase difference in quantum mechanical two photon absorption.
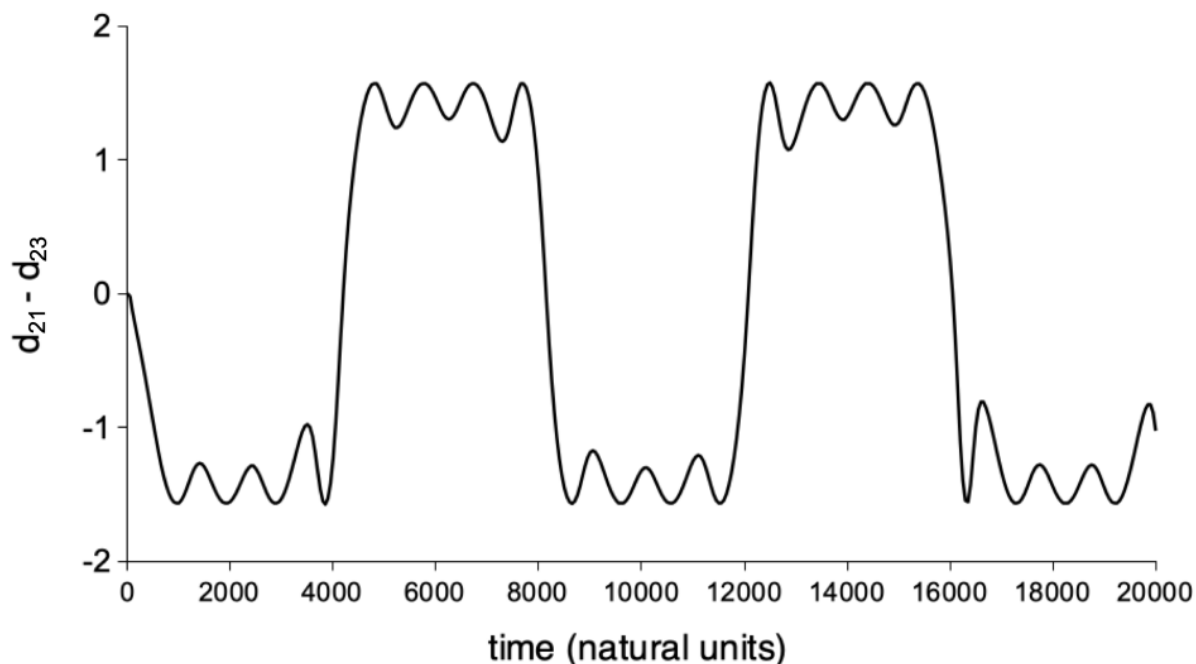
Figure S3. Plot of phase angle difference $d_{21} - d_{23}$ for the same trajectory shown in Fig. S2 and exhibiting periodic phase reversals during energy transfer. The value oscillates between $-\pi/2$ when energy is being transferred to oscillator #3 and $+\pi/2$ when energy is being transferred back to oscillator #1 although the energy in oscillator #2 never gets above 0.2. Compare with quantum dynamics of two-photon absorption in Fig. 10 of the accompanying paper.

Section 5. Hydrogen Fluoride Dipole Moment Matrix Elements

The following dipole moment matrix elements $<n|\mu|m>$ of the HF molecule were integrated numerically using the dipole moment of Ogilvie et al. (1980) and the exact Morse oscillator wave functions with the HF parameters given in Section 1. Only elements for the first three states were used in the calculations included in the journal article.

n	m	$\langle n\|\mu\|m\rangle$
0	0	7.18332680958864E+01
1	0	3.88330920028936E+00
2	0	-4.69038661152100E-01
3	0	4.07901328468283E-02
4	0	-1.28469947096340E-02
5	0	3.16954645622662E-03
6	0	-1.20887867638402E-03
7	0	1.25803279481808E-03
8	0	-1.26874795823292E-03
9	0	1.10063634217087E-03
10	0	-8.80898392410669E-04
11	0	6.79088936028490E-04
12	0	-5.15608325683861E-04
13	0	3.90375044164964E-04
14	0	-2.96897151330197E-04
15	0	2.27802841748126E-04
16	0	-1.76712905599487E-04
17	0	1.38639200994382E-04
18	0	-1.09843284173269E-04
19	0	8.75424123445200E-05
20	0	-6.95969636741100E-05
21	0	5.41767338569602E-05
22	0	-3.92106876114042E-05
23	0	1.98267812315833E-05
1	1	7.34913116736805E+01
2	1	5.40906048520160E+00
3	1	-9.01131372414360E-01
4	1	7.53319061864168E-02
5	1	-3.18190226008979E-02
6	1	1.11750042299042E-02
7	1	-3.61899274801774E-03
8	1	2.78294065388170E-03
9	1	-2.88333004316476E-03
10	1	2.68807555388713E-03
11	1	-2.29577384424221E-03
12	1	1.87412961735973E-03
13	1	-1.49820872935223E-03
14	1	1.18823525464493E-03
15	1	-9.41975340579709E-04
16	1	7.49647559203227E-04
17	1	-6.00073259772329E-04
18	1	4.82998972608909E-04
19	1	-3.89691720066286E-04
20	1	3.12702942475510E-04

21	1	-2.45048467076733E-04
22	1	1.78110952550732E-04
23	1	-9.02371680424806E-05
2	2	7.50730443230427E+01
3	2	6.40819304142863E+00
4	2	-1.44672896257069E+00
5	2	1.06919373697706E-01
6	2	-5.36682902465961E-02
7	2	2.72068128148234E-02
8	2	-9.29975539713747E-03
9	2	5.00793697066806E-03
10	2	-4.81667112805148E-03
11	2	4.74992652373109E-03
12	2	-4.30912302336343E-03
13	2	3.70439451437698E-03
14	2	-3.09986640136503E-03
15	2	2.56229924354493E-03
16	2	-2.10790503369540E-03
17	2	1.73223105811204E-03
18	2	-1.42325961046019E-03
19	2	1.16663881174102E-03
20	2	-9.47295765478085E-04
21	2	7.48583121119961E-04
22	2	-5.46976997040125E-04
23	2	2.77783780812545E-04
3	3	7.64641951272296E+01
4	3	6.94404026677277E+00
5	3	-2.16066286849293E+00
6	3	1.43075845151711E-01
7	3	-6.63931909781412E-02
8	3	5.20929682997977E-02
9	3	-2.17797588886168E-02
10	3	9.20186134320516E-03
11	3	-6.97114250871577E-03
12	3	6.93699346032120E-03
13	3	-6.66871607954348E-03
14	3	6.03056621482825E-03
15	3	-5.25631695134582E-03
16	3	4.49676469670321E-03
17	3	-3.81038137698484E-03
18	3	3.20829440040211E-03
19	3	-2.68106997232101E-03
20	3	2.20924771375367E-03
21	3	-1.76430865530405E-03
22	3	1.29779936343833E-03

23	3	-6.61107382856906E-04
4	4	7.74935309694646E+01
5	4	6.93092048984191E+00
6	4	-3.08685093845048E+00
7	4	2.16751512267760E-01
8	4	-5.23338843771186E-02
9	4	7.94982888441337E-02
10	4	-4.52943678701634E-02
11	4	1.89751476834097E-02
12	4	-1.02492426990056E-02
13	4	8.88769548638462E-03
14	4	-8.82743672207277E-03
15	4	8.44291149574416E-03
16	4	-7.69412815746301E-03
17	4	6.79500297150075E-03
18	4	-5.88604227861969E-03
19	4	5.02106143439422E-03
20	4	-4.20088520945790E-03
21	4	3.39155031660547E-03
22	4	-2.51220825580497E-03
23	4	1.28384310509057E-03
5	5	7.79355478727854E+01
6	5	6.23038275386308E+00
7	5	-4.22910975892226E+00
8	5	3.99367829018695E-01
9	5	2.79276297093417E-04
10	5	8.96765419490746E-02
11	5	-7.94334432681282E-02
12	5	4.07185070485905E-02
13	5	-1.87665245848464E-02
14	5	1.15768419177371E-02
15	5	-1.02333421794534E-02
16	5	1.00609668588080E-02
17	5	-9.65981241244851E-03
18	5	8.89077151968716E-03
19	5	-7.89139993375461E-03
20	5	6.77092300786926E-03
21	5	-5.55265063749037E-03
22	5	4.15027250625105E-03
23	5	-2.12928322252072E-03
6	6	7.75381509959876E+01
7	6	4.71680135046970E+00
8	6	-5.50948064414561E+00
9	6	7.98182122681601E-01
10	6	7.11879031367666E-02

11	6	5.09211989761825E-02
12	6	-1.07766442385408E-01
13	6	7.78451263214559E-02
14	6	-4.12140694432527E-02
15	6	2.09349969533775E-02
16	6	-1.30484627211588E-02
17	6	1.07058232733672E-02
18	6	-1.00076872760179E-02
19	6	9.42989960619736E-03
20	6	-8.55047475571375E-03
21	6	7.29616357214139E-03
22	6	-5.58736654066627E-03
23	6	2.89747269377556E-03
7	7	7.60819864595553E+01
8	7	2.35173031653752E+00
9	7	-6.73810077215305E+00
10	7	1.51898571876853E+00
11	7	7.71581601877454E-02
12	7	-5.59781208829766E-02
13	7	-9.03963893997484E-02
14	7	1.14119701357877E-01
15	7	-8.21427539642789E-02
16	7	4.84944862018611E-02
17	7	-2.73708080352939E-02
18	7	1.66340978720488E-02
19	7	-1.16373311686753E-02
20	7	9.13929351428315E-03
21	7	-7.42269820819857E-03
22	7	5.62791624480817E-03
23	7	-2.91927962340923E-03
8	8	7.34576608005319E+01
9	8	-7.54113346498300E-01
10	8	-7.62897523322876E+00
11	8	2.59237876375461E+00
12	8	-1.30791764569187E-01
13	8	-1.88011105886230E-01
14	8	1.36383743390234E-02
15	8	1.00250736618267E-01
16	8	-1.17085335065358E-01
17	8	9.32601236192716E-02
18	8	-6.44983217412538E-02
19	8	4.25217670323498E-02
20	8	-2.80654464946475E-02
21	8	1.88354479071800E-02
22	8	-1.23803852456786E-02

23	8	5.99372263861836E-03
9	9	6.97214173136274E+01
10	9	-4.33161396019624E+00
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