

Algebraic Dynamics Study a Single Molecule Driven by a Time Dependent Laser Radiation Field

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Abstract The dynamical properties of a single molecule driven by a time dependent laser radiation field are researched. Based on the $su(1,1) \oplus h(3)$ ($h(3)$ is Heisenberg algebra) dynamical symmetry structure of the system, the exact solutions of the system is obtained by using the algebraic dynamics method. The shift between the frequency Ω of the laser radiation field and the molecule vibration frequency ω under resonance phenomenon is studied.

Key words adiabatic energy levels; algebraic dynamics; geometric phase; resonance

代数动力学研究含时激光辐射场驱动下的单分子

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【摘要】 该文研究了含时激光辐射场驱动下的单分子系统的动力学性质。基于系统的 $su(1,1) \oplus h(3)$ 代数结构 ($h(3)$ 满足 Heisenberg 代数) 和代数动力学方法, 不仅获得了系统的解析解, 而且还研究了系统的非绝热能级和几何相位。最后研究了非绝热能级和几何相位与激光辐射场频率的函数关系, 展示了系统存在的共振现象以及分子共振吸收时激光辐射场频率和分子振动频率之间的漂移现象。

关键词 非绝热能级; 代数动力学; 几何相位; 共振

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Algebraic dynamics is a theory which studies the quantum system by the algebraic method^[1-7]. It always use the group relationship of the system operators or group structure of the system to discuss the properties of the system. In the past years, it has been extensively used in nuclear physics for many autonomous systems. However, many systems are non-autonomous which depend on time in many experiments. So, It's necessary to extend the theory of algebraic dynamics to resolve the non-autonomous problems because it can help us control the systems. Several typical non-autonomous quantum systems have been researched by the method of dynamical algebras, e.g. the $su(1,1)$ dynamic structure for the particle moving in time dependent Paul trap^[8], the polarization of spin particle in accelerator forms a $su(2)$ dynamic system^[9],

the spin particle in a rotating magnetic field, and the Berry phase of a laser in helical optical fiber forming a $su(2)$ dynamic system^[10-16]. As an important controlling and measurement method for particles and substance, the laser radiation field is always chosen by people^[17-19]. When the laser radiation field interacts with particles or substance, an important physical phenomenon, resonance excitation effect will occur. By this physical phenomenon, human can understand many important properties of the particles and substance, such as molecular structure and vibration frequency of the substance. Since the laser radiation fields always depend on time, these quantum system become the non-autonomous quantum system and are accompanied with the resonance excitation effects. So, it is interesting to research the diabatic energy and

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geometric phase^[20-22] under the resonance excitation phenomena.

In this work, we research the properties of the single molecule which is driven by a time dependent laser radiation field. Firstly, we analyze the $su(1,1) \oplus h(3)$ ($h(3)$ is Heisenberg algebra) Lie algebraic structure of this system. Secondly, the exact solution of the system has been obtained by the use of algebraic dynamics method. Finally, based on the exact solutions, we study the changing properties of the diabatic energy levels and the geometric phase. Simultaneously, we show that there exists a shift between the laser frequency and the vibration frequency of the molecule when the resonance excitation occurs.

1 The Model Hamiltonian and Algebraic Structure

A single molecule is driven by a time dependent laser radiation field, and the system Hamiltonian is described by the following model which represents a charged harmonic oscillator in a laser radiation field:

$$\hat{H} = \frac{1}{2m} \left(\hat{\mathbf{P}} - \frac{q}{c} \hat{\mathbf{A}} \right)^2 + \frac{1}{2} m \omega^2 \hat{x}^2 \quad (1)$$

Considering the potentials $\mathbf{A} = \left(-\frac{c\varepsilon_0}{2\Omega} \sin(\Omega t), B_0 x, 0 \right)$ and $\phi = -\frac{1}{2} \varepsilon_0 \cos(\Omega t) \hat{x}$, (1) can be written as:

$$\begin{aligned} \hat{H}(t) = & \frac{\hat{p}_x^2 + \hat{p}_y^2 + \hat{p}_z^2}{2m} + \frac{q\varepsilon_0 \sin(\Omega t)}{2m\Omega} \hat{p}_x + \\ & \left(B_0 \hat{p}_y - \frac{1}{2} q\varepsilon_0 \cos(\Omega t) \right) \hat{x} + \\ & \left(\frac{1}{2} m \omega^2 + B_0^2 \right) \hat{x}^2 + \frac{(q\varepsilon_0)^2}{8m\Omega^2} \sin^2(\Omega t) \end{aligned} \quad (2)$$

where q is the molecule charge; c is velocity of light, the oscillator potential of the molecule is $\frac{1}{2} m \omega^2 \hat{x}^2$; B_0 is a constant which describes the magnetic field amplitude of the laser electromagnetic field and directs the z axis; ε_0 and Ω are electric field amplitude and frequency of the laser electromagnetic field respectively.

It is easy to prove $[\hat{p}_y, \hat{H}] = [\hat{p}_z, \hat{H}] = 0$. Define

the new operators $\hat{k}_+ = \frac{1}{2} \hat{p}_x^2; \hat{k}_0 = -\frac{i}{4} [\hat{p}_x \hat{x} + \hat{x} \hat{p}_x];$

$\hat{k}_- = \frac{1}{2} \hat{x}^2; k_1 = \hat{p}_x; \hat{k}_2 = \hat{x}$. For certain eigenvalues

$p_y, (p_z)$ of operators $\hat{p}_y, (\hat{p}_z)$, the Hamiltonian $\hat{H}(t)$ can be rewritten as:

$$\hat{H}(t) = \frac{p_y^2 + p_z^2}{2m} + X_+ \hat{k}_+ + X_- \hat{k}_- + X_1 \hat{k}_1 + X_2 \hat{k}_2 + X \quad (3)$$

Where the parameters are:

$$\begin{cases} X_+ = \frac{1}{m}; X_- = m\omega^2 + 2B_0^2; X_1 = \frac{q\varepsilon_0 \sin(\Omega t)}{2m\Omega} \\ X_2 = B_0 p_y - \frac{1}{2} q\varepsilon_0 \cos(\Omega t); X = \frac{(q\varepsilon_0)^2}{8m\Omega^2} \sin^2(\Omega t) \end{cases} \quad (4)$$

Through (3) we find that the system has the dynamical algebraic structure $su(1,1) \oplus h(3)$ ^[23]. It can be checked that: 1) \hat{k}_+, \hat{k}_0 and \hat{k}_- span $su(1,1)$ Lie algebra. 2) \hat{k}_1, \hat{k}_2 and 1 span $h(3)$ ($h(3)$ is Heisenberg algebra) Lie algebra. 3) It satisfy the following communication relations ($\hbar = 1$):

$$[\hat{k}_+, \hat{k}_-] = 2\hat{k}_0 \quad [\hat{k}_0, \hat{k}_\pm] = \pm \hat{k}_\pm \quad [\hat{k}_1, \hat{k}_2] = -i \quad (5)$$

Simultaneously, the generators of $su(1,1)$ and $h(3)$ also satisfy the following communication relations:

$$\begin{cases} [\hat{k}_+, \hat{k}_1] = 0 \quad [\hat{k}_0, \hat{k}_1] = \frac{1}{2} \hat{k}_1 \quad [\hat{k}_-, \hat{k}_1] = i\hat{k}_2 \\ [\hat{k}_+, \hat{k}_2] = -i\hat{k}_1 \quad [\hat{k}_0, \hat{k}_2] = -\frac{1}{2} \hat{k}_2 \quad [\hat{k}_-, \hat{k}_2] = 0 \end{cases} \quad (6)$$

From (4) we know that the system has the dynamical algebraic structure $h\omega(4)$ ^[24] for the parameters $X_1 = X_2 = X(t) = 0 (B_0 = 0)$.

2 The Exact Solutions of the System Using the Algebraic Dynamic Method

The time-evolution of the system satisfy the time-dependent Schrödinger equation:

$$i \frac{\partial}{\partial t} |\psi(t)\rangle = \hat{H}(t) |\psi(t)\rangle \quad (7)$$

Adopting the solving steps of algebraic dynamics^[12-16], firstly, introduce the gauge transformation:

$$U_g(t) = \exp(iv(t)) \exp(iv_2(t)\hat{k}_2) \exp(-iv_1(t)\hat{k}_1) \times \exp(iv_-(t)\hat{k}_-) \exp(iv_0(t)\hat{k}_0) \quad (8)$$

where $v(t), v_2(t), v_1(t), v_-(t)$ and $v_0(t)$ are all time-dependent parameters.

The Schrödinger equation under the gauge transformation (7) ($\hbar = 1$) becomes:

$$i\partial|\bar{\psi}_n(t)\rangle/\partial t = \hat{H}(t)|\bar{\psi}_n(t)\rangle \quad (9)$$

Here the gauged Hamiltonian is given by:

$$\begin{cases} \hat{H}(t) = U_g^{-1}(t)\hat{H}(t)U_g(t) - iU_g^{-1}(t)(\partial U_g(t)/\partial t) \\ |\psi_n(t)\rangle = U_g(t)|\bar{\psi}_n(t)\rangle \end{cases} \quad (10)$$

Substituting (3) and (7) into (9), after some complex calculations, one has:

$$\hat{H}(t) = Z_+ \hat{k}_+ + iZ_0 \hat{k}_0 + Z_- \hat{k}_- + Z_1 \hat{k}_1 + Z_2 \hat{k}_2 + Z \quad (11)$$

Here the coefficients of (11) are:

$$\begin{cases} Z_+ = \exp[-v_0(t)]X_+ \quad Z_0 = -\dot{v}_0(t) + 2X_+v_-(t) \\ Z_0 = -\dot{v}_0(t) + 2X_+v_-(t) \\ Z_- = \exp[v_0(t)][\dot{v}_-(t) + X_- + X_+v_-^2(t)] \end{cases} \quad (12)$$

$$Z_1 = \exp\left[-\frac{v_0(t)}{2}\right][-\dot{v}_1(t) + X_1 + X_+v_2(t)]$$

$$Z_2 = \exp\left[\frac{v_0(t)}{2}\right]\{\dot{v}_2(t) + X_2 + X_-v_1(t) +$$

$$[-\dot{v}_1(t) + X_1 + X_+v_2(t)]v_-(t)\}$$

$$Z = \dot{v}(t) + v_1(t)\dot{v}_2(t) + X + X_2v_1(t) + X_1v_2(t) +$$

$$\frac{1}{2}X_-v_1^2(t) + \frac{1}{2}X_+v_2^2(t)$$

Because we can choose the appropriate transformation which is one of the advantages of algebraic dynamics^[9-15] to simplify the calculation, which is also easy to find the Cartan operators, the best choice of the gauge transformation satisfies the following conditions:

$$\frac{Z_-}{Z_+} = \frac{\exp[2v_0(t)]}{X_+}[\dot{v}_-(t) + X_- + X_+v_-^2(t)] = \text{const} = k \quad (13a)$$

$$Z_0 = -\dot{v}_0(t) + 2X_+v_-(t) = 0 \quad (13b)$$

$$Z_1 = \exp\left[-\frac{v_0(t)}{2}\right][-\dot{v}_1(t) + X_1 + X_+v_2(t)] = 0 \quad (13c)$$

$$Z_2 = \exp\left[\frac{v_0(t)}{2}\right]\{\dot{v}_2(t) + X_2 + X_-v_1(t) + [-\dot{v}_1(t) + X_1 + X_+v_2(t)]v_-(t)\} = 0 \quad (13d)$$

$$Z = \dot{v}(t) + v_1(t)\dot{v}_2(t) + X + X_2v_1(t) + X_1v_2(t) + \frac{1}{2}X_-v_1^2(t) + \frac{1}{2}X_+v_2^2(t) = 0 \quad (13e)$$

After some calculation, the solutions of the parameters of (13) can be obtained as follows:

$$v_-(t) = 0, \quad v_0(t) = 0, \quad k = \frac{X_-}{X_+} = [(m\omega)^2 + 2mB_0^2]$$

$$v_1(t) = A[\cos(\Omega t) - \cos(k't)] - \frac{B_0 p_y}{m\omega^2 + 2B_0^2}$$

$$A = \frac{q\epsilon_0}{[2(B_0)^2 + m(\omega^2 - \Omega^2)]} \quad k' = \sqrt{\frac{m\omega^2 + 2B_0^2}{m}}$$

$$v_2(t) = mA[k'\sin(k't) - \Omega\sin(\Omega t)] - \frac{q\epsilon_0}{2\Omega}\sin(\Omega t)$$

$$v(t) = -\int_0^t [v_1(t')\dot{v}_2(t') + X + X_2v_1(t') + X_1v_2(t') + \frac{1}{2}X_-v_1^2(t') + \frac{1}{2}X_+v_2^2(t')]dt' \quad (14)$$

Here, consider the initial conditions $v(0) = 0$, $v_2(0) = 0$, $v_1(0) = \text{const}$, $v_-(0) = 0$ and $v_0(0) = 0$.

Putting (13) to (10), we obtain the covariant Hamiltonian:

$$\hat{H}(t) = \frac{p_y^2 + p_z^2}{2m} + f(t)\hat{I}(0) \quad (15)$$

Where:

$$\hat{I}(0) = \hat{k}_+ + k\hat{k}_- = \frac{1}{2}\hat{p}^2 + \frac{1}{2}k\hat{x}^2, \quad f(t) = \frac{1}{m} \quad (16)$$

The Cartan operator $\hat{I}(0)$ does not depend on time explicitly and has the standard form of harmonic oscillator. So, the time-dependent dynamical symmetry can be covered into the stationary dynamical symmetry by choosing a proper gauge transformation.

The eigen problems of the Cartan invariant operator $\hat{I}(t) = U_g(t)\hat{I}(0)U_g^{-1}(t)$ can be obtained. Let $|n\rangle$ be the eigenstate of $\hat{I}(0)$ (here n is the quantum number of the standard form of harmonic oscillator), namely:

$$\hat{I}(0)|n\rangle = \left(n + \frac{1}{2}\right)\omega_0|n\rangle \quad n = 0, 1, 2, \dots, \omega_0^2 = k \quad (17)$$

so the eigenvalues and eigenstates are:

$$\hat{I}(t)U_g(t)|n\rangle = \left(n + \frac{1}{2}\right)\omega_0U_g(t)|n\rangle = \left(n + \frac{1}{2}\right)\omega_0|\phi_n(t)\rangle \quad (18)$$

Where $|\phi_n(t)\rangle = U_g(t)|n\rangle$ is the eigenstate of $\hat{I}(t)$ with eigenvalue $\left(n + \frac{1}{2}\right)\omega_0$.

So the covariant Schrödinger equation (9) has the following solutions:

$$|\bar{\psi}_{p_y, p_z, n}(t)\rangle = \exp[-i\Theta_{p_y, p_z, n}(t)]|\phi_{p_y, p_z, n}\rangle \quad (19)$$

where:

$$\Theta_{p_y, p_z, n}(t) = \int_0^t \left[\frac{p_y^2 + p_z^2}{2m} + \left(n + \frac{1}{2}\right)\omega_0 f(\tau) \right] d\tau$$

$$|\varphi_{p_y,p_z,n}\rangle = \frac{e^{i(p_y y + p_z z)}}{2\pi} |n\rangle \tag{20}$$

In order to obtain the solutions of (7), firstly, we need to rewrite $|\bar{\psi}_n(t)\rangle$ under the coordinate x representation as follows:

$$|\bar{\psi}_{p_y,p_z,n}(t)\rangle = \frac{e^{i(p_y y + p_z z)}}{2\pi} N_n(\alpha) \times \exp[-i\Theta_n(t)] \exp[-(\alpha x)^2 / 2] H_n(\alpha x) \tag{21}$$

here $N_n(\alpha) = [\alpha / \sqrt{\pi} 2^n n!]^{\frac{1}{2}}$, $\alpha^2 = \omega_0 = \sqrt{k}$, and $H_n(\alpha x)$ is the Hermite polynomial.

Secondly, using the following relations:

$$\exp[-iv_1(t)\hat{k}_1] F(x) = \exp[-iv_1(t)\hat{p}] F(x) = F(x - v_1(t)) \tag{22}$$

The orthonormal nonadiabatic basis can be directly obtained and given as follows:

$$|\psi_{p_y,p_z,n}(t)\rangle = U_g(t) |\bar{\psi}_{p_y,p_z,n}(t)\rangle = \frac{e^{i(p_y y + p_z z)}}{2\pi} \exp[-i\Theta_{p_y,p_z,n}(t)] |\phi_n(t)\rangle = \frac{e^{i(p_y y + p_z z)}}{2\pi} N_n(\alpha) \exp[-i[\Theta_n(t) - v(t)]] \exp[iv_2(t)x] \times \exp\left\{-\frac{1}{2}[\alpha(x - v_1(t))]^2\right\} H_n(\alpha(x - v_1(t))) \tag{23}$$

The equation describes a motion of quasi-harmonic oscillator in coordinate space and the origin of this coordinate space constantly moves and stretches. At the same time, there are a collective velocity potential $v_2(t)x$ and a time-dependent phase factor for this system.

The general solutions of the time-dependent Schrödinger equation (7) can be expanded by the nonadiabatic basis:

$$|\psi(t)\rangle = \sum_n C_{p_y,p_z,n} |\psi_{p_y,p_z,n}(t)\rangle = \sum_n C_{p_y,p_z,n} \frac{e^{i(p_y y + p_z z)}}{2\pi} \exp[-i\Theta_{p_y,p_z,n}(t)] |\phi_n(t)\rangle \tag{24}$$

where $C_{p_y,p_z,n}$ is an expansion coefficient that is not dependent on time. All the dynamical information is included in the nonadiabatic basis.

3 The Diabatic Energy Levels of the System

Using (10), (14), (15), the diabatic energy levels

of the system can be obtain by:

$$E_{p_y,p_z,n}(t) = \langle \psi_{p_y,p_z,n}(t) | \hat{H}(t) | \psi_{p_y,p_z,n}(t) \rangle = \langle \bar{\psi}_{p_y,p_z,n}(t) | \hat{H}(t) | \bar{\psi}_{p_y,p_z,n}(t) \rangle + \langle \bar{\psi}_{p_y,p_z,n}(t) | iU_g^{-1}(t) \frac{\partial U_g(t)}{\partial t} | \bar{\psi}_{p_y,p_z,n}(t) \rangle = \frac{p_y^2 + p_z^2}{2m} + \left(n + \frac{1}{2}\right) \omega_0 f(t) - \{v_1(t)\dot{v}_2(t) + v(t)\} \tag{25}$$

From(24), it is easily found: 1) the change of the diabatic energy levels comes from the factor $\{v_1(t)\dot{v}_2(t) + v(t)\}$. 2) the changing approach of the diabatic energy levels are quasi-periodic due to the periodic changing parameters parameters $v_1(t), v_2(t)$ and $v(t)$.

To better understand the properties of the diabatic energy levels, the changing behaviors are shown in Fig. 1 (Here, the parameters $n=0$, $\omega = 2\ 100\ \text{cm}^{-1}$, $p_x=1, p_y=0, m=1 \times 10^{-27}$, $q=1$ and $\epsilon_0=1$). The diabatic energy level shows an abrupt increase when the laser frequency Ω arrives about $2\ 100\ \text{cm}^{-1}$, corresponding to “the resonance absorption effect of molecular”, and the different peaks display the different magnetic field amplitudes B_0 . Simultaneously, with the increasing of the magnetic field amplitude B_0 , the laser frequency Ω diverges from the molecule vibration frequency ω , which is characterized by the peak-shifting of the resonance absorption. The above results may be helpful for the explanation of laser-induced effect of biological genetic variation^[25], e.g. the bond of biological genetic may be broken and recombined which is aroused by the resonance absorption effect.

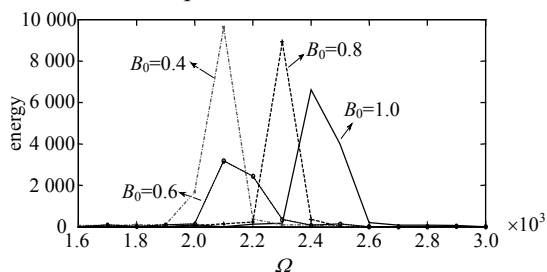


Fig. 1 The change of diabatic energy levels

It is surprising why the resonance phenomenon occurs. The results stem from the parameters $v_1(t), v_2(t)$ and $v(t)$ (see (26)). From (15) we find that the changing period of the parameters $v_1(t), v_2(t)$ and

$v(t)$ not only depends on the Ω , but also on k' which depends on the ω and B_0 . So the above reasons lead to resonance phenomenon and a divergence between Ω and ω . Moreover, from the parameters k' and A in (15), we know that: 1) the shift of the resonance frequency increases with the decrease of the ω and the increase of the B_0 ; 2) The peak value displays different for different B_0 .

4 Geometric Phase of the System

The system states will acquire a total phase which composes of the dynamical phase and the geometric phase when the parameters of the system go through a time-dependent evolution. The dynamical phase depends both on the path and on the rate of the path, while the geometric phase depends on the influence of the external environment or the interaction with the background.

The dynamical phase γ_d [2-3,24] is:

$$\gamma_d = -\int_0^t \langle \psi_{p,p_2,n}(\tau) | \hat{H}(\tau) | \psi_{p,p_2,n}(\tau) \rangle d\tau \quad (26)$$

The geometric phase β [2-3,24] is:

$$\beta = \gamma_{0-t} - \gamma_d = \gamma_{U_g} + \gamma_I \quad (27)$$

where the total phase γ_{0-t} is:

$$\begin{aligned} \gamma_{0-t} = & \text{Im} \left(\ln \langle \psi_{p,p_2,n}(0) | \psi_{p,p_2,n}(t) \rangle \right) = \\ & -\Theta_{p,p_2,n}(t) + \text{Im} \left(\ln \langle \bar{\psi}_{p,p_2,n}(t) | U_g(t) | \bar{\psi}_{p,p_2,n}(t) \rangle \right) = \\ & -\Theta_{p,p_2,n}(t) + \gamma_{U_g} \end{aligned} \quad (28)$$

where $\Theta_{p,p_2,n}(t)$ is the total phase of $|\bar{\psi}_{p,p_2,n}(t)\rangle$, $\gamma_{U_g} = \text{Im} \left(\ln \langle \bar{\psi}_{p,p_2,n}(t) | U_g(t) | \bar{\psi}_{p,p_2,n}(t) \rangle \right)$ is the phase induced by the gauged transformation, and $\gamma_I = -(\nu_1(t)\dot{\nu}_2(t) + \dot{\nu}(t))$.

In order to further study the changing properties of the geometric phase, we calculate the geometric phase for $n=0,1$ respectively.

For $n=0$, the geometric phase is:

$$\beta = \nu + \frac{\nu_1\nu_2}{\alpha^2 + 1} - (\nu_1(t)\dot{\nu}_2(t) + \dot{\nu}(t)) \quad (29)$$

For $n=1$, the geometric phase is:

$$\beta = \nu + \frac{\nu_1\nu_2}{\alpha^2 + 1} - (\nu_1(t)\dot{\nu}_2(t) + \dot{\nu}(t)) - \theta \quad (30)$$

Where θ is the phase angle of the complex number $(\nu_2^2 + \alpha^2\nu_1^2 - \alpha^2 - 1) + i(\nu_1\nu_2(1 - \alpha^2))$.

Contrasting (30) and (31), it is easy to know that the difference of the Berry phase is aroused by the θ for $n=0$ and $n=1$. Because the phase angle θ depends on the parameters $\nu_1(t), \nu_2(t)$ and α , it shows the quasi-periodic changing behavior and depend on the value of ω and B_0 .

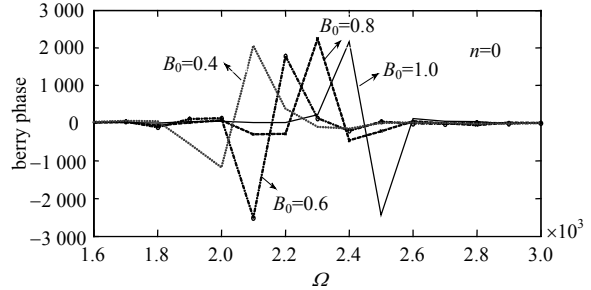


Fig. 2 The change of geometric phase

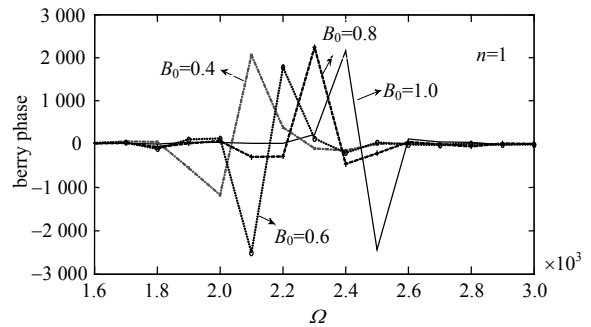


Fig. 3 The change of the geometric phase

The changing of the geometric phase β are shown in the Fig. 2 and Fig. 3 for $n=0$ and $n=1$ respectively (The related parameters are same as Fig.1). We found that: 1) The geometric phase β also presents resonance phenomenon when the laser frequency Ω gets about 2100 cm^{-1} . 2) Similar to the diabatic energy level, there also exists a shift of the peak of the resonance for the difference between the laser frequency Ω and the molecule vibration frequency ω . 3) With the increase of the magnetic field amplitude B_0 , the shift of the peak of the resonance increases. 4) The influence of the phase angle θ is small for $n=0$ and $n=1$. The results stem from the changing period of the parameters $\nu_1(t), \nu_2(t)$ and $\nu(t)$ are dependent on the k' and Ω . Simultaneously, the parameters k' and A depend on the ω and B_0 . It arouses that the shift of the resonance frequency increases with the decrease of the ω and the increase of the B_0 .

5 Conclusion

Based on $su(1,1) \oplus h(3)$ dynamical symmetry of the molecule which is driven by a time dependent laser radiation field, we obtain the exact solutions of the system by use of algebraic dynamics and further discuss the changing properties of the diabatic energy levels and geometric phase. It is found that there exists the resonance phenomenon for the diabatic energy levels and geometric phase when the resonance frequency Ω is close to the molecular vibration frequency ω , and a divergence which depends on the magnetic field amplitude B_0 exists between Ω and ω . The present work may be helpful for the explanation of laser-induced effect of biological genetic variation, and it shows that the method of dynamical algebras is useful for the study non-autonomous quantum system which has some algebraic structures.

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