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NONADIABATIC GEOMETRIC PHASES OF MULTIPHOTON TRANSITIONS IN DISSIPATIVE SYSTEMS AND SPIN-J SYSTEMS

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ABSTRACT:

We present new developments in nonadiabatic geometric phases along two lines for systems undergoing changes of quantum state in intense fields. We first present a geometric representation of the non-Hermitian Schrodinger equation and introduce the notion of a complex multiphoton Aharonov-Anandan (AA) phase associated with dissipative two-level systems driven by periodic fields. The concept is further extended to include field modulation effects. We then develop the AA phase for spin-j systems in periodic fields and find conditions for cyclic evolution for general multi-level systems. In both cases, generalizations of the Floquet formalism lead to general analytical expressions for geometric phases that can be tested by experiments.

I. INTRODUCTION

The study of the geometric factor accompanied by an adiabatic or cyclic change in classical or quantum systems has received considerable attention in recent times[1-6]. The adiabatic quantum phase discovered by Berry [1] is associated with the adiabatic evolution of a Hamiltonian $\hat{H}(R)$ along a closed curve Γ in the parameter (R) space. When the quantum system remains in an eigenstate of $\hat{H}(R)$ during this cyclic evolution, a geometric phase, the Berry phase, is resultant. This phase depends only on Γ. More recently, Aharonov and Anandan [3] have introduced a new geometric phase, the AA phase, which is the nonadiabatic generalization of Berry's phase and is associated with the cyclic evolution of a quantum system, i.e., a system with state described by $|\psi(t)\rangle$ which returns to itself, apart from a phase factor Φ: $|\psi(T)\rangle = \exp(i\Phi)|\psi(0)\rangle$. Both Berry and AA phases have been detected experimentally [7].

The AA phase is of fundamental interest for systems being driven by strong fields and so are undergoing changes of quantum state. The cyclic evolution associated with the AA phase may be interest for recurring system behavior as well as general phenomenology. Here we present new concepts and methods in AA geometric phases in two directions. We present first in section II a generalized non-Hermitian density matrix formalism for the treatment of dissipative quantum systems in periodic fields in which we introduce the notion of a complex AA geometric phase. This concept is further extended to embrace multiphoton transitions, the *complex multiphoton AA phase*. In section III, we generalize the theory to include the effects of field modulation. The second direction is a Floquet formalism for the AA phase in <u>any</u> spin-j system subject to intense periodic fields which is presented in section IV.

II. COMPLEX GEOMETRIC PHASE IN DISSIPATIVE TWO-LEVEL SYSTEMS

A. Generalized Density Matrix Formulation for Complex Geometric Phase

The AA phase is associated with the cyclic evolution of the quantum system in which the initial state recurs, apart from a phase factor. The associated density matrix therefore is cyclic: $\hat{\rho}'(T)=\hat{\rho}'(0)$ as is the Bloch vector of a two-level system: $\vec{S}(T) = \vec{S}(0)$, $S_i(t) = Tr(\hat{\sigma}_i\hat{\rho}(t))$, $\hat{\sigma}_i$ a Pauli matrix. Consider a two-level system with Hamiltonian operator which includes T_1-type damping.

$$\hat{H}(t) = \hat{H}_o + \hat{V}(t) - i\hat{G} \tag{1}$$

$\hat{H}_o$ represents the unperturbed two-level Hamiltonian with diagonal energies E_α and E_β, $\hat{V}(t)$ the electric dipole coupling to the time-dependent field and $\hat{G}$ is the diagonal damping g_α & g_β representing, for example, the spontaneous decay of these levels. Due to the dissipation, $Tr(\hat{\rho}'(t))$ and $\langle\psi(t)|\psi(t)\rangle$ are decreasing functions. This leads to difficulties in meeting the conditions of cyclic evolution and the geometric representation of the AA phase. To overcome these difficulties, we consider the following generalized density matrix:

$$\hat{\rho}(t) = |\psi(t)\rangle\langle\chi(t)|, \tag{2}$$

defined in the biorthonormal Hilbert space [8] with $|\chi(t)\rangle$ the solution of the Schrodinger equation with the <u>adjoint</u> Hamiltonian $\hat{H}^+(t)$ (in atomic units)

$$i\frac{d}{dt}|\chi(t)\rangle = \hat{H}^+(t)\ |\chi(t)\rangle. \tag{3}$$

The *generalized* Liouville and Bloch equations based upon (2) have the respective *symplectic* forms which are identical to their non-dissipative cases

$$i \frac{d\hat{\rho}(t)}{dt} = [\hat{H}(t),\hat{\rho}(t)] \tag{4a}$$

$$\frac{d\vec{S}(t)}{dt} = \vec{\Omega} \times \vec{S}(t). \tag{4b}$$

Therefore, the trace of $\hat{\rho}(t)$, the norm of $\vec{S}(t)$ and $\langle\chi(t)|\psi(t)\rangle$ remain constant throughout.

Under a cyclic quantum evolution, $\hat{\rho}(t+T) = \hat{\rho}(t)$, $\vec{S}(t+T) = \vec{S}(t)$ and

$$|\psi(t+T)\rangle = \exp(i\Phi)\ |\psi(t)\rangle\ ,\ |\chi(t+T)\rangle = \exp(i\Phi^*)\ |\chi(t)\rangle. \tag{5}$$

where Φ is the now <u>complex</u> total phase and is the sum of the dynamical phase (α_D) and AA geometric phase (β_G): $\Phi=\alpha_D + \beta_G$, where

$$\alpha_D = - \int_0^T \langle\chi|\ \hat{H}\ |\psi\rangle\ dt \quad , \quad \beta_G = \int_0^T \langle\chi'|\ i\partial/\partial t\ |\psi'\rangle\ dt. \tag{6}$$

We have shown the following theorem to be true concerning the geometric representation of this *complex* AA phase [9]:

Theorem: The complex geometric phase β_G is equal to one-half the complex solid angle $\Omega(C)$ enclosed by the complex trajectory C of the vector $\vec{S}(t)$.

In the case the dissipative terms vanish, $|\psi(t)\rangle = |\chi(t)\rangle$ and Φ, α_D and β_G are real quantities.

B. Complex Multiphoton Geometric Phases

Consider a dissipative two-level quantum system undergoing *multiphoton* Rabi floppings in an intense periodic field. The perturbation is given by

$$\hat{V}(t) = -\vec{\mu} \cdot \vec{\varepsilon}_o \cos(\omega t+\phi), \tag{7}$$

where $\vec{\mu}$ is the electric dipole moment of the system, $\vec{\varepsilon}_o$, ω & ϕ are the electric field amplitude, frequency and phase, respectively. In terms of the unperturbed ($|\alpha>$, $|\beta>$) basis, the total Hamiltonian is

$$\hat{H}^{+}(t) = \begin{bmatrix} E_\beta - ig_\beta & V_{\beta\alpha}(t) \\ V_{\alpha\beta}(t) & E_\alpha - ig_\alpha \end{bmatrix} \tag{8}$$

with $V_{\alpha\beta}(t) = <\alpha|\hat{V}(t)|\beta>$. The non-Hermitian time-dependent Schrodinger equation with periodic Hamiltonian (8) may be transformed into an equivalent infinite-dimensional non-Hermitian Floquet matrix ($\hat{A}_F$) eigenvalue problem [8,10]

$$\hat{A}_F\, |\lambda_{\gamma n}> = \lambda_{\gamma n}\, |\lambda_{\gamma n}>, \tag{9}$$

where $\lambda_{\gamma n}$ and $|\lambda_{\gamma n}>$ are the complex quasi-energy eigenvalues and eigenvectors, with $\gamma = \alpha$ or β and $n = -\infty$ to ∞ .

For nearly resonant multiphoton processes, $E_\beta - E_\alpha \equiv \omega_o \cong (2n+1)\omega$, $\hat{A}_F$ can be reduced to a two-dimensional effective Floquet Hamiltonian ($\hat{A}_{eff}$) by extension of appropriate nearly-degenerate perturbation theory [11].

$$\hat{A}_{eff} = \begin{bmatrix} \varepsilon_\beta - (2n+1)\omega + \delta_\beta^{(C)} & u_{\beta\alpha} \\ u_{\alpha\beta} & E_\alpha + \delta_\alpha^{(C)} \end{bmatrix} \tag{10}$$

here $\delta_\alpha^{(C)}$and $\delta_\beta^{(C)}$are the complex AC Stark shifts for states α and (β). $u_{\alpha\beta}$ and $u_{\beta\alpha}$ represent the effective (2n+1) photon coupling.

The biorthogonal eigenvalues and eigenvectors are found following the method described by Faisal [12]. $\hat{A}_{eff}$ is diagonal in the basis $|\lambda_\pm>$ and its adjoint the basis $|\varepsilon_\pm>$

$$\hat{A}_{eff}|\lambda_\pm> = \lambda_\pm|\lambda_\pm> \quad , \quad \hat{A}^{+}_{eff}\, |\varepsilon_\pm> = \varepsilon_\pm|\varepsilon_\pm>. \tag{11}$$

The eigenvalues $\lambda_\pm$ are found to be

$$\lambda_\pm = \kappa \pm q \tag{12}$$

where $\kappa = 1/2\ Tr(\hat{A}_{eff})$, $Tr(\hat{A}_{eff})$ indicating the trace of the matrix, and $q = 1/2\sqrt{(\Delta^2+4u_{\alpha\beta}u_{\beta\alpha})}$ with Δ the detuning parameter, $\Delta = Tr(\hat{A}_{eff}\hat{\sigma}_z)$. Note biorthonormality and closure are satisfied

$$<\epsilon_\pm|\lambda_\pm> = 1,\ <\epsilon_\pm|\lambda_\pm> = 0 \quad , \quad |\epsilon_+><\lambda_+| + |\epsilon_-><\lambda_-| = 1\ . \tag{13}$$

These properties are useful in computing the time evolution operator

$$\hat{U}(t,t_0) \cong \exp[-i\hat{A}_{eff}(t-t_0)]. \tag{14}$$

Therefore, given an initial state of the system at t_0, the wavefunction at time t is approximated, using the eigenvalue properties and biorthonormal closure from above. Note that elements of the complex Bloch vector $\vec{S}(t)=(u,v,w)$ may be computed by substitution of the wavefunction into the component expressions .

The AA phase may now be calculated using the wavefunction method as previously discussed. After an interval of time $T = \pi/q$, the wavefunction completes a cycle

$$|\psi(T)> = \exp[-(\pi+KT)]|\psi(0)>. \tag{15}$$

The total phase Φ and dynamical phase α_D are, respectively

$$\Phi = -\pi - \kappa T \tag{16.a}$$

$$\alpha_D = -\{\kappa T+\pi[<\chi(0)|\lambda_+><\epsilon_+|\psi(0)>-<\chi(0)|\lambda_-><\epsilon_-|\psi(0)>]\}. \tag{16.b}$$

The general formula for the complex AA phase for <u>multiphoton</u> Rabi floppings and period $T = \pi/q$ is

$$\beta_G^{(C)} = -\pi(1-[<\chi(0)|\lambda_+><\epsilon_+|\psi(0)>-<\chi(0)|\lambda_-><\epsilon_-|\psi(0)>]). \tag{17}$$

The corresponding formula for non-dissipative systems is

$$\beta_G = -\pi\{1-[|<\psi(0)|\lambda_+>|^2-|<\psi(0)|\lambda_->|^2]\}. \tag{18}$$

From inspection of the effective Hamiltonian $\hat{A}_{eff}$ it is evident that the AA phase is calculable in terms of important experimental parameters such as initial state, detuning, field strength and phase and so on. This method is thus of great practical importance as well as theoretical importance; the remarkable simplicity of (17) and (18) underscores their generality.

C. Examples

The purpose of this section is to demonstrate graphically the behavior of the Aharonov-Anandan geometric phase as a function of various parameters which may be of interest for experimental study. In what follows, it is assumed the difference in energies of the two-level system, $\omega_o = E_\beta - E_\alpha = 1$ in arbitrary units. The ω and couplings b in these diagrams are reported in units of ω_o.

In figure 1 we show the Aharonov-Anandan geometric phase β_G as a function of the coupling parameter b, where $b = -1/2 \langle\alpha|\vec{\mu}|\beta\rangle \cdot \vec{\varepsilon}_o$. This is a one photon transition. Three frequencies are shown, a resonant and detuned ($\pm$ 0.05 ω_o) cases. The detuned cases are nearly, but not quite, symmetrical , on account of the complex Stark shifting which effects both real and imaginary parts of the geometric phase .

In figure 2 we show oscillations in the Aharonov-Anandan phase as a function of laser phase ϕ for a <u>three-photon</u> transition. The interference effect is sensitive to the initial conditions, requiring a superposition of both states. An interesting feature is that the number of oscillations is characteristic of the particular order of photon process.

III. THE COMPLEX AA GEOMETRIC PHASE IN MODULATED FIELDS

The method described in the previous sections works well provided the parameters of the external field such as amplitude, frequency and phase remain constant. If parameters are modulated, however the formulation involving a time-independent effective Hamiltonian fails to be adequate for the construction of the time evolution operator.

The Hamiltonian operator is written in a more general form

$$\hat{H}(t) = \hat{H}_0 - i\hat{G} - \vec{\mu} \cdot \vec{\varepsilon}_o(t) \cos(\omega(t)t+\phi(t)) . \tag{19}$$

Although this form is similar to that previously studied, and while $\hat{H}_0$ and $\hat{G}$ have their original meaning , the external field interaction may involve time-dependent field amplitude, frequency, phase or any combination thereof. Let $\vec{X}(t) =(\omega(t),\phi(t), \vec{\varepsilon}_o(t))$. In the nearly diabatic basis [13], the matrix form of the Schrodinger equation is

$$i \frac{d}{dt} \begin{bmatrix} c_\beta(t) \\ c_\alpha(t) \end{bmatrix} = \hat{A}_{eff}(\vec{X}(t)) \begin{bmatrix} c_\beta(t) \\ c_\alpha(t) \end{bmatrix} \tag{20}$$

which is similar to the differential equation equation in the original $\{ |\alpha\rangle,|\beta\rangle\}$ basis but, using the Floquet-nearly degenerate perturbation method, involves an operator varying slowly in time.

A very general method for solving (20) is the Magnus approximation [14] which, as implemented here, is similar to the previous method involving a time-independent effective Floquet Hamiltonian, and in fact the eigenvalues and eigenvectors have a similar form as before. Let $\hat{M}$ be defined as

$$\hat{M} = \int_{t_o}^{t_o+\tau} \hat{A}_{eff}(t)dt.$$

There is a basis in which $\hat{M}$ is diagonal.

$$\hat{M}|\lambda_\pm\rangle = \lambda_\pm|\lambda_\pm\rangle, \quad \langle\varepsilon_\pm|\hat{M}=\langle\varepsilon_\pm|\varepsilon_\pm$$

The eigenvalues are:

$$\lambda_\pm(\tau) = \kappa(\tau)\pm q(\tau), \quad \varepsilon_\pm = \lambda_\pm^* \tag{21}$$

where

$$\kappa(\tau) = 1/2 \int_{t_o}^{\tau+t_o} tr(\hat{A}_{eff}(t))\, dt \tag{22}$$

and

$$q(\tau) = 1/2 \int_{t_o}^{\tau+t_o} ([tr(\hat{A}_{eff}(t)\hat{\sigma}_z]^2 + 4\mu_{\alpha\beta}(t)\mu_{\beta\alpha}(t))^{1/2} dt. \tag{23}$$

If $q(\tau)$ is found such that

$$q(\tau) = \pi \tag{24}$$

then the condition for cyclic evolution is realized. The total phase is

$$\phi = -\pi - \kappa(\tau) \tag{25}$$

the dynamical phase

$$\alpha_D = - \{ \kappa(\tau) + \pi [\langle\chi(0)|\lambda_+(\tau)\rangle\langle\varepsilon_+(\tau)|\psi(0)\rangle - \langle\chi(0)|\lambda_-(\tau)\rangle\langle\varepsilon_-(\tau)|\psi(0)\rangle] \}. \tag{26}$$

Note that these expressions revert to the previous expressions for ϕ and α_D if $\vec{X}$ is independent of the time. This is also true of the AA phase, now generalized to the expression

$$\beta_G = -\pi\{ 1 - [\langle\chi(0)|\lambda_+(\tau)\rangle\langle\varepsilon_+(\tau)|\psi(0)\rangle-\langle\chi(0)|\lambda_-(\tau)\rangle\langle\varepsilon_-(\tau)|\psi(0)\rangle]\}. \tag{27}$$

This is a very general expression which should be of use given that the components of $\vec{X}$ are varied sufficiently slow. It is possible to extend the range of validity further by the use of higher-order Magnus approximations. The result is sufficient to show that analytic methods can be used to determine the conditions for cyclic evolution and the AA phase even if the parameters describing the field are modulated.

IV. THE AA GEOMETRIC PHASE OF SPIN-j SYSTEMS IN PERIODIC FIELDS

Another case of signicance is the multi-level system driven by a strong field. In general, cyclic evolution is not common for such a case and so the AA phase is not defined. However, in the instance of a spin-j system, such evolution is possible and this is further instructive in considering the general conditions for cyclic system behavior.

Consider a spin-j system subjected to a static magnetic field along the $\hat{z}$-axis and a linearly polarized, time-dependent magnetic field along the $\hat{x}$-axis. This situation is typical of many magnetic resonance experiments [15]. The Hamiltonian for this system is (in atomic units)

$$\hat{H}(t) = -(\mu/j)\,[\,\hat{j}_x\, B_x^o \cos\omega t + \hat{j}_z\, B_z^o\,] \tag{28}$$

which is expressed in terms of the magnetic field $\vec{B}(t)$ and the angular momentum operators $\hat{j}$. Here μ is the magnetic dipole moment of the spin-j system and j is the angular momentum . This expression may be further developed by expanding $\hat{j}_x$ in terms of the ladder operator, thus realizing the form

$$\hat{H}(t) = \omega_o\, \hat{j}_z + 2\omega_\perp\,(\,\hat{j}_+ + \hat{j}_-\,)\cos\omega t \tag{29}$$

ω_o is the level splitting and $2\omega_\perp$ represents the coupling between the dipole moment and the field.

$$\omega_o = -\mu B_z^o\,/j \quad , \quad \omega_\perp = -\,\mu B_x^o\,/j \tag{30}$$

and where $|\omega_o| >> |\omega_\perp|$.

The Hamiltonian is periodic in time: $\hat{H}(t+2/\omega) = \hat{H}(t)$, so that it is advantageous to use Floquet theory to transform the time-dependent Schrodinger equation into an equivalent time-independent eigenvalue problem as previously discussed . Application of the rotating wave approximation (RWA) truncates this infinite Floquet matrix to dimension (2j+1) x (2j+1). The resulting time-independent RWA Hamiltonian in operator form is

$$\hat{H}_{RWA} = -j\omega \hat{I} + \Delta \hat{J}_z + 2 \omega_\perp \hat{J}_x \tag{31}$$

where $\Delta = \omega_0 - \omega$ is the detuning or degree of off-resonance and $\hat{I}$ is the identity operation. $\hat{H}_{RWA}$ satisfies

$$\hat{H}_{RWA} \mid \varepsilon_m > = \varepsilon_m \mid \varepsilon_m >. \tag{32}$$

The eigenvalues and eigenvectors are determined by a rotation in the xz plane by an angle β

$$\tan\beta = \frac{2\omega_\perp}{\Delta} .$$

The Rabi frequency Ω is defined as

$$\Omega = [\Delta^2 + 4\omega_\perp^2]^{1/2} \tag{33}$$

$$\cos\beta = \Delta/\Omega \quad , \sin\beta = 2\omega_\perp/\Omega .$$

Using the properties developed ,the eigenspectrum of $\hat{H}_{RWA}$ is

$$\varepsilon_m = -j\omega + m\Omega . \tag{34}$$

The system evolves as

$$\mid \Psi(t) > = e^{ij\omega t} \sum_{m=-j}^{j} \mid \varepsilon_m >< \varepsilon_m \mid \psi(0) > e^{-im\Omega t} \tag{35}$$

where $\mid \varepsilon_m >$ are expressed in terms of the rotation matrices $d^{(j)}$. Now the condition for cyclic evolution may be deduced by considering general quasienergies ε_m in (35). It is evident that the quasienergy spectrum must spaced in <u>integer</u> multiples of some common parameter, though not necessarily equally spaced, as is the present case. Such a spectrum of quasienergies guarantees cyclic evolution.

Equation (35) shows that after a period $T = 2\pi/\Omega$ a cyclic evolution is executed by the spin-j system in the magnetic field. The total phase associated with the cyclic evolution is

$$\Phi = j\,(\omega T - 2\pi) \tag{36}$$

and the dynamical phase is given by the expression

$$\alpha_D = j\omega T - 2\pi \sum_{m=-j}^{j} m|<\psi(0)\,|\,\varepsilon_m>|^2 . \tag{37}$$

The gauge-invariant AA phase of the driven spin-j system is

$$\beta_G = -\,2\pi\,(\,j - \sum_{m=-j}^{j} m\,|<\psi(0)\,|\,\varepsilon_m>|^2 . \tag{38}$$

This equation is the main result of this section. It is the extension to 2j+1 levels of the result found previously for two-level systems (18). A particularly interesting case to study is that where the initial state corresponds to $m = -j$, $|\psi(0)> = |j,-j>$.

$$\beta_G = -2\,\pi\,j\,(\,1 - \cos\beta)\,. \tag{39}$$

These expressions are in agreement with results we have obtained with Youhong Huang by the method of SU(2) spin-coherent states [16], which result states β_G is equivalent to the solid angle enclosed by the *generalized* Bloch vector's closed circuit times j (Figure 3 shows the generalized Bloch sphere of the spin-j system). Note this result is the generalization of the previous theorem for the two-level system.

V. CONCLUSIONS

We have presented new concepts and methods in geometric phases along two lines. First, we have introduced the notion of complex AA phases in a dissipative two-level systems and have further extended the concept to include multiphoton processes and the effects of field modulation. The second development involved the AA phase in the spin-j system for arbitrary j. Here we found insight into cyclic evolution of multi-level systems. In both areas, we have found the Floquet formalism to be most useful in formulating the theory and subsequent extensions of this formalism provided general expressions for the AA phase in both the multiphoton dissipative and spin-j cases. We are presently considering extensions of this work to more complex dissipative systems.

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FIGURES

Figure 1

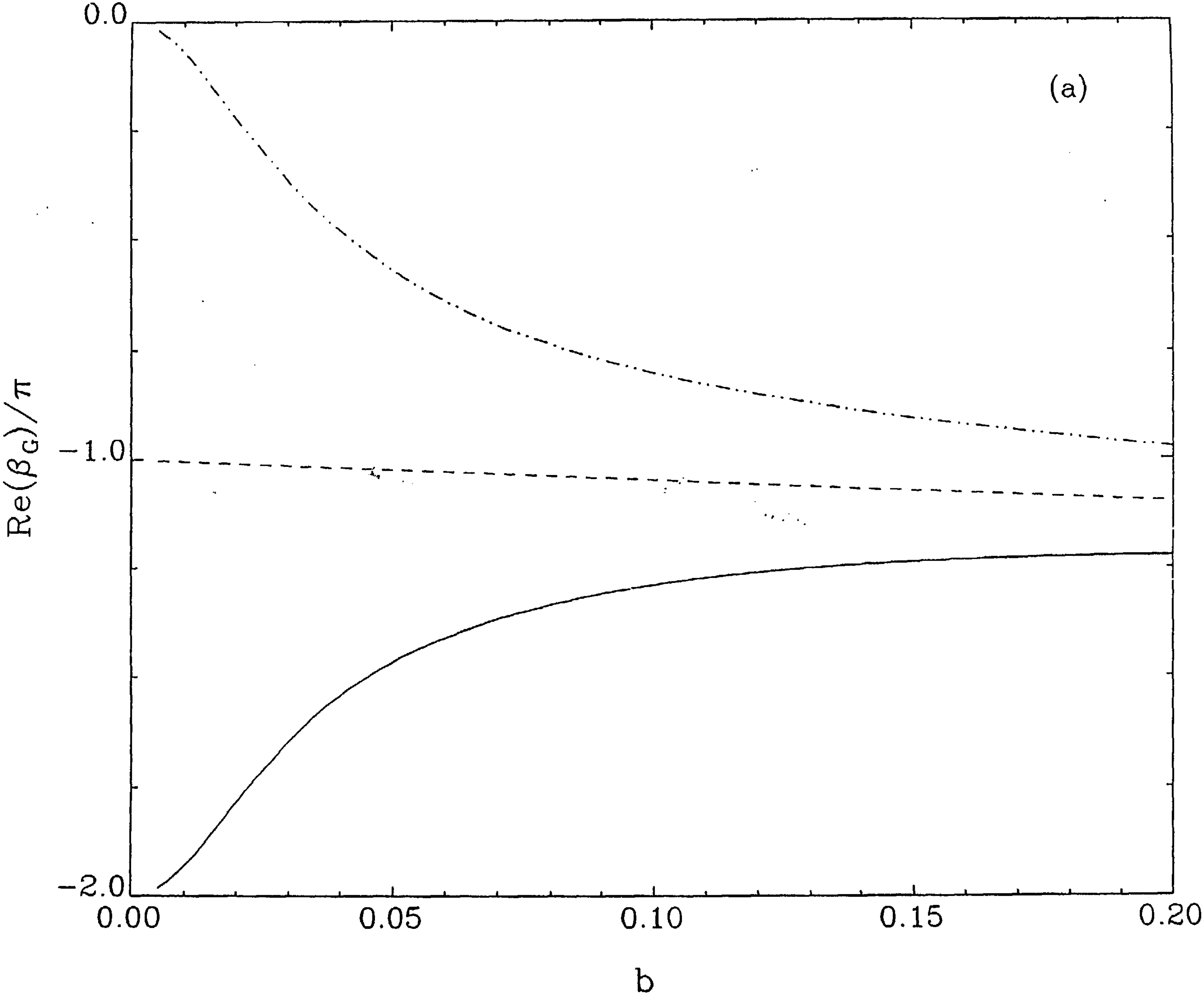
Aharonov-Anandan geometric phase β_G as a function of the coupling b. (a) Re(β_G) and (b)Im(β_G) for field frequencies ω = 0.95 (solid), 1.00 (dashed) and 1.05 (dash-double dot). Laser phase ϕ = 0, damping constants are: g_α = 0.001 and g_β = 0.004. Initial state $|\alpha>$.

Figure 2

Oscillations in Aharonov-Anandan phase. Real (a) and imaginary (b) components are shown. Coupling b =0.1, ω = 0.3333 and damping constants the same as in figure 1. Two initial states are shown: $|\alpha>$ (dashed) and $(|\alpha> + |\beta>)/\sqrt{2}$ (solid).

Figure 3

Generalized Bloch sphere model for any spin j . Every point $\hat{n}$ = $(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ on the unit radius Bloch sphere represents a a spin-coherent state. The unit vector $\hat{n}_o$ (south pole) corresponds to the fundamental vector $|\psi_o> = |j,-j>$. Also shown is a sterographic projection of the vector $\hat{n}$ to a point ξ on the complex plane.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−2.0
0.00
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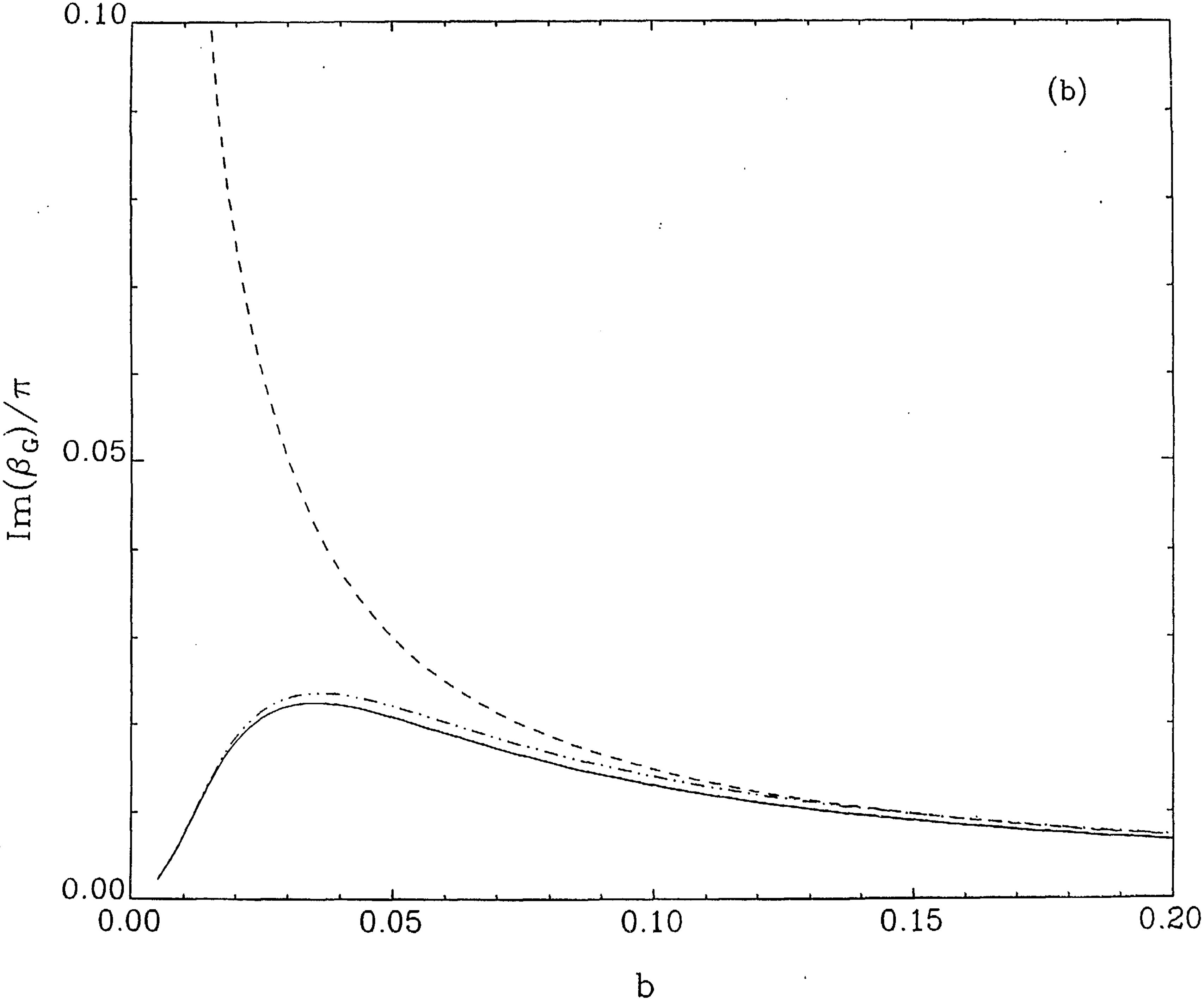
0.15
0.20
b


(b)
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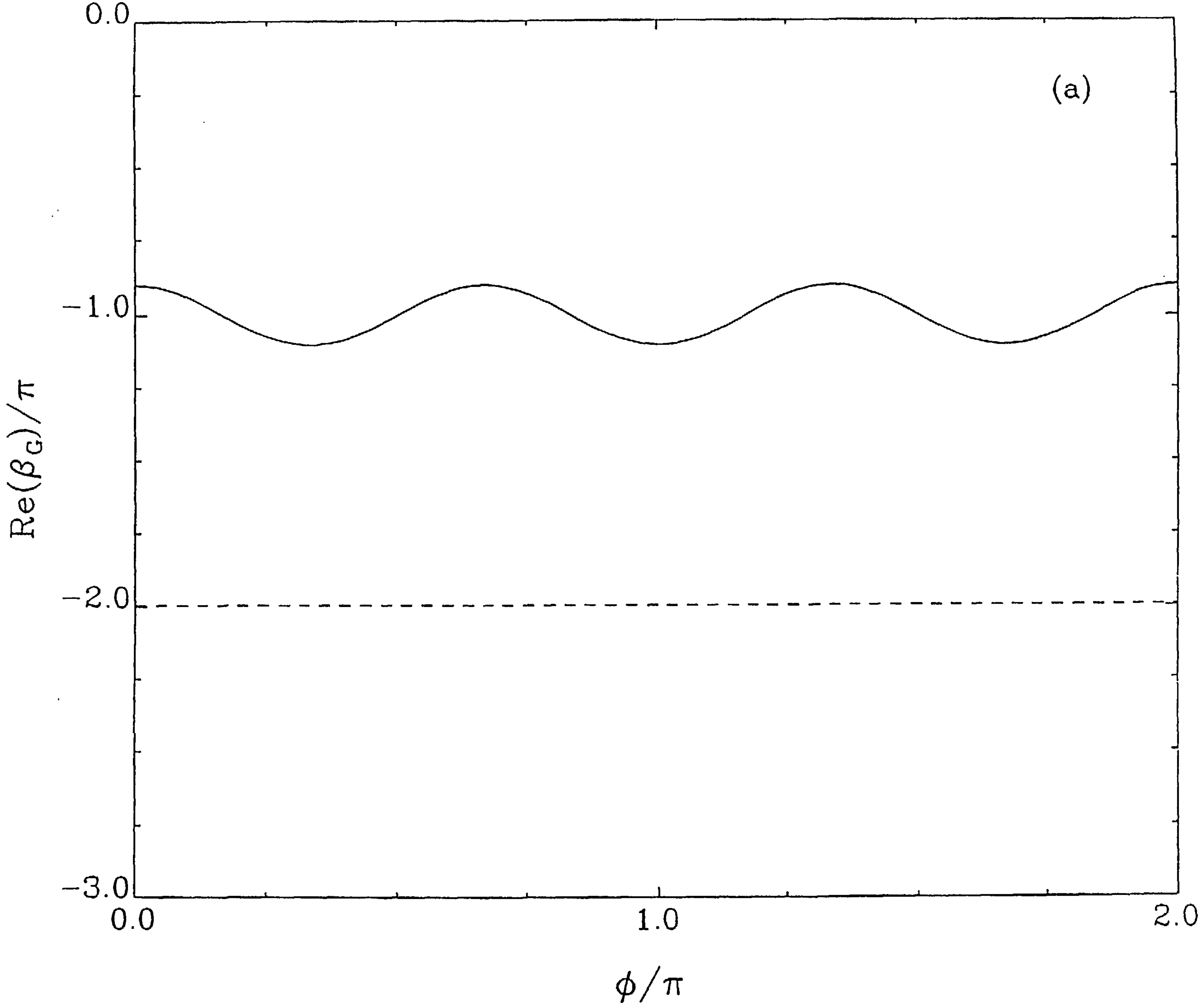
Im(β_G)/π
b



(a)
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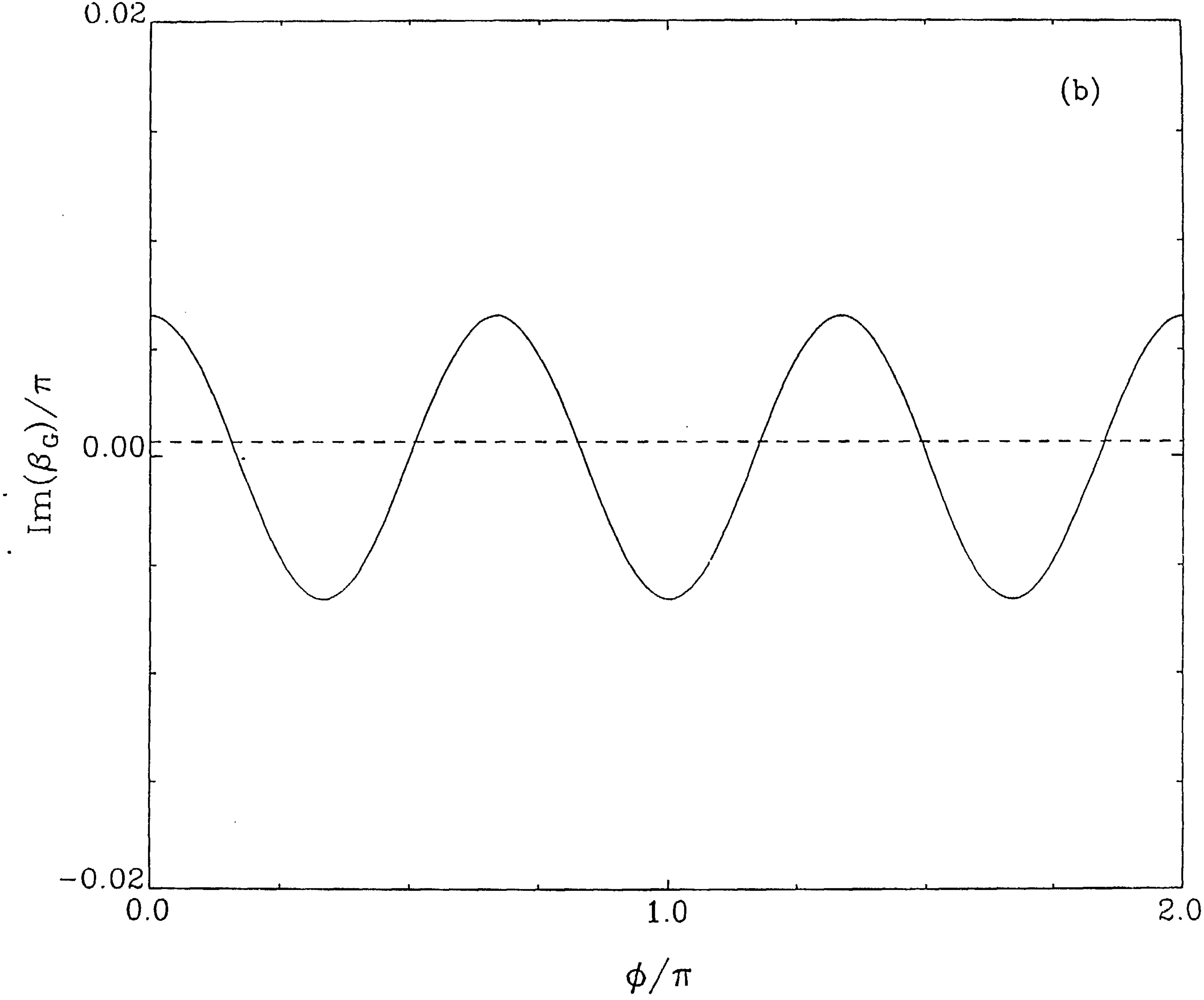
−3.0
Re(β_G)/π
0.0
1.0
2.0
ϕ/π



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0.00
−0.02
Im(β_G)/π
(b)
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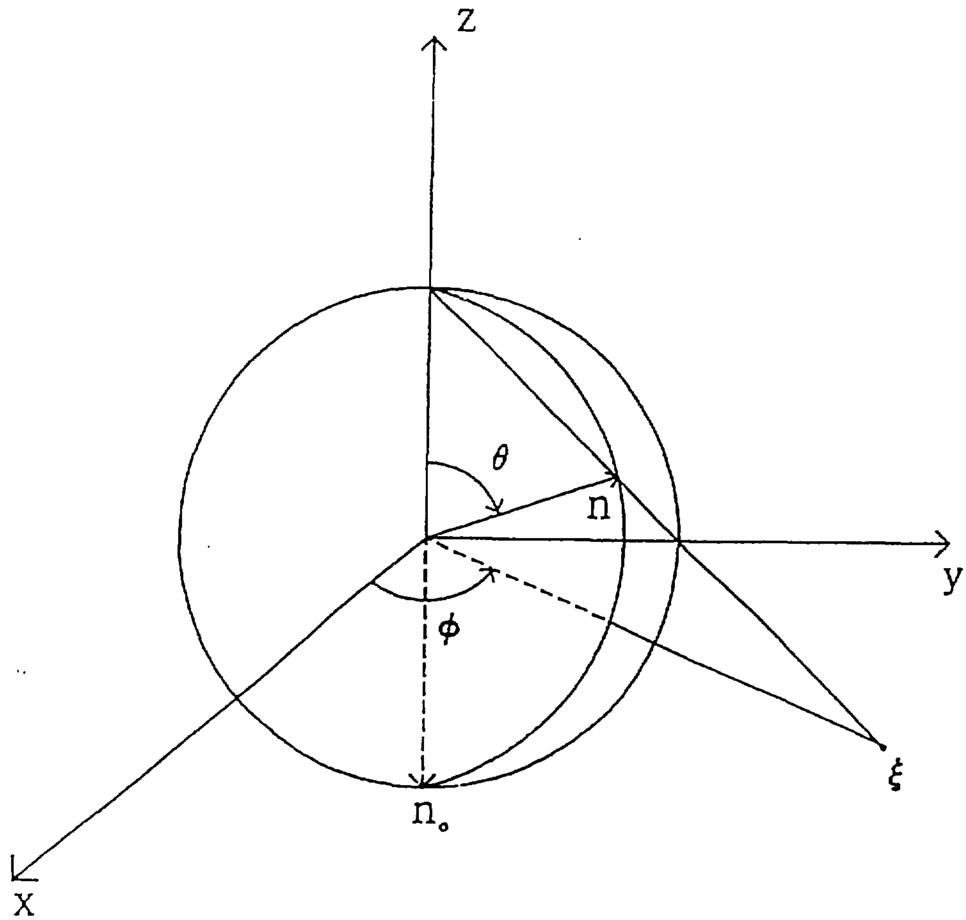
1.0
2.0
φ/π


z
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