Geometric phases in multiphoton ionization

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(Received 17 January 1992)

We discuss the geometric phase accumulated by the wave function of an atom ionizing in the presence of a bichromatic field as physical parameters are varied adiabatically around a closed circuit. As an illustration we calculate the geometric phase for a hydrogen atom in the presence of 355-nm light and its third harmonic when the phase and intensity of the two components are varied. The wave function need not be single valued after one complete circuit—two circuits may be necessary to map the original eigenray onto itself. Furthermore, the geometric phase may be complex, and may therefore modify the ionization yield calculated from the width of the instantaneous quasienergy.

PACS number(s): 32.80.Rm, 32.90.+a

I. INTRODUCTION

The seminal article of Berry [1] has generated enormous interest in the geometric phase that is accumulated by a wave function, in addition to the normal dynamical phase, when the system is adiabatically transported along a closed circuit in some physical parameter space. For an excellent, but by no means exhaustive, collection of papers dealing with different aspects of this phase, we refer the reader to the book *Geometric Phases in Physics* [2]. The focus of that book is on nondissipative systems, but as pointed out by Garrison and Wright [3], the geometric phase may be complex when the system is dissipative [4].

In the present paper we discuss the geometric phase within the context of an atom undergoing multiphoton ionization in the presence of bichromatic light that is a coherent superposition of two waves with commensurable frequencies $p\omega$ and $q\omega$ (where p and q are positive integers). As an illustration, we calculate the geometric phase of the wave function of the hydrogen atom for a closed circuit in a three-parameter space where the three physical parameters are the individual intensities of the two components of the bichromatic light beam and the relative phase of these components. Since the nonadiabatic variation of the interaction is periodic in time, we can cast the problem into a (nearly) time-independent one by making the Floquet ansatz, whereby the state vector, with the overall dynamical phase factor $\exp(-i\int^t dt'\, E/\hbar)$ removed, is expanded in a Fourier (harmonic) series $\sum_N \exp(-iN\int^t dt'\,\omega)\mid \mathcal{F}_N\rangle$. The harmonic components $\mid \mathcal{F}_N\rangle$ satisfy a set of coupled, almost time-independent, equations which take the form of the familiar eigenvalue problem posed by the time-independent Schrödinger equation containing the Floquet Hamiltonian. The eigenvalue of the Floquet Hamiltonian is the quasienergy E, and since the system is dissipative (the atom ionizes) E is complex, with $-2\,\mathrm{Im}E$ being the ionization width. The geometric phase may be defined as the phase accumulated by the column vector whose components are the $\mid \mathcal{F}_N\rangle$, when the system is transported around a closed circuit in some parameter space [5]. It turns out that the geometric phase is in general complex (although it may be nearly real, approximately equal to an integer multiple of π for some circuits) and hence the geometric phase modifies the ionization yield calculated by integrating, over the circuit, the width of the instantaneous quasienergy.

In discussing ionization by bichromatic light, we generalize the analysis of Garrison and Wright, who calculated the complex geometric phase in the case of a two-level atom ionizing in the presence of *mono*chromatic light of fixed frequency ω and varying phase Φ. While the calculation carried out by Garrison and Wright is highly instructive, the restriction to a one-parameter space, where the circuit is the line $0 \le \Phi \le 2\pi$, limits the behavior of the system in ways that we now remark on. First, the quasienergy is independent of the overall phase of the light wave, since the quasienergy, being the eigenvalue of a time-independent (Floquet) Hamiltonian, cannot depend on the origin of time, and the overall phase of the light wave can always be transformed away by a shift in the origin of time. Consequently, the variation of Φ, by itself, cannot lead to a degeneracy in the quasienergy; at least two other parameters, upon which the quasienergy

does depend, would have to be varied to give rise to a degeneracy (in a three-parameter space). Therefore, in the one-parameter Φ space, the eigenray is *single valued* — that is, the eigenfunction is the same at the points $\Phi = 0$ and 2π, aside from the overall geometric phase. More significantly, the imaginary part of the geometric phase can be rather easily incorporated into the dynamical phase. This can be seen by first observing that an adiabatic variation in Φ, amounting to 2π over a very long time interval, is equivalent to an infinitesimal variation in the frequency ω. Now, an infinitesimal circuit in the one-parameter ω space cannot produce a nonzero geometric phase. However, since E depends on ω, a very small change in ω does affect the overall dynamical phase factor $\exp(-i\int^t dt'\, E/\hbar)$, noting that the very small change in E is magnified by the long time interval. In fact, since the geometric phase must be independent of the way in which Φ is varied from 0 to 2π, as long as other parameters are not varied, we are free to choose the variation to be linear in t, in which case the temporal variation in Φ may be interpreted simply as a *constant* shift $d\Phi/dt$ in the frequency ω. We have verified that the complex geometric phase obtained by letting Φ vary linearly from 0 to 2π over a long time interval T gives the same modification to the ionization yield as does a shift in the frequency by the small but constant amount $2\pi/T$, holding Φ fixed so that the geometric phase is exactly zero. In other words, the imaginary part of the geometric phase is dynamical in origin. More generally, suppose that the intensity I of the monochromatic light varies, and that the system follows a prescribed closed circuit in the Φ-I plane over a time interval $t_1 \le t \le t_2$. The contribution B_Φ arising from a variation in Φ, to the geometric phase B, may be absorbed into the dynamical phase and evaluated as

$$B_\Phi = -\int_{t_1}^{t_2} dt'\, \Delta E, \tag{1}$$

where, if the quasienergy E is evaluated at the current value of the intensity, ΔE is the change in E at time t due just to the change $d\Phi/dt$ in ω. Since Φ is a function of I along the closed circuit, we can reexpress B_Φ as

$$B_\Phi = -\oint dI\, \frac{d\Phi}{dI}\frac{\partial E}{\partial \omega}. \tag{2}$$

The time t does not appear explicitly in this last expression, so B_Φ is a "geometric" phase, depending only on the circuit in the Φ-I plane, but of course if the time dependence of Φ is altered, so is the functional dependence of Φ on I.

However, when the light is bichromatic, the quasienergy depends on the *relative* phase of the two component fields [6]. By adjusting the relative phase and one other parameter, such as the strength of one of the two component fields, it is possible to arrange that at a particular point in this two-parameter space, the quasienergy is degenerate. In other words, we can choose a circuit in this two-parameter space which encloses a degeneracy in the eigenvalue of the Floquet Hamiltonian. (In a three-parameter space the circuit would enclose a *line* of degeneracies.) As a consequence, the eigenray need not be single valued. (The eigenray is single valued when the Floquet Hamiltonian matrix is real and symmetric, that is, when ionization is negligible, as explained in Appendix B.) In Fig. 1 we show the trajectory of the quasienergy eigenvalue for the hydrogen atom, initially in the $1s$ state, when the atom is irradiated by bichromatic light and the intensities I_L and I_H of the low- and high-frequency components, and the phase Φ_H of the high-frequency component, are varied. In this example, the ratio I_H/I_L is kept constant, and I_L and Φ_H are varied along a rectangle in the I_L-Φ_H plane whose sides correspond to either constant I_L or constant Φ_H; this example is very similar to the one discussed in Ref. [7]. The light is linearly polarized, the low-frequency component has a wavelength of 355 nm, and the other component is the third harmonic. The $1s$ and $2p$ states are connected by a three-photon transition that is nearly on resonance. We label the corners of the rectangle in the I_L-Φ_H plane counterclockwise as A, B, C, and D, starting from the left lower corner. We show trajectories for three cases, corresponding to different values of the ratio I_H/I_L. In case (a) the real part of the $1s$ eigenvalue undergoes an avoided crossing (point α) with the real part of the $2p$ eigenvalue, as I_L is varied from 0.1×10^{13} W/cm^2 (point A) up to 1.3×10^{13} W/cm^2 (point B) along the line $\Phi_H = 0°$; it then undergoes a true crossing (point β) as I_L is varied back to zero along the line $\Phi_H = 160°$ (from points C to D). At an avoided crossing of the real parts of the quasienergies the characters of the adiabatic states interchange; in other words, the character of the

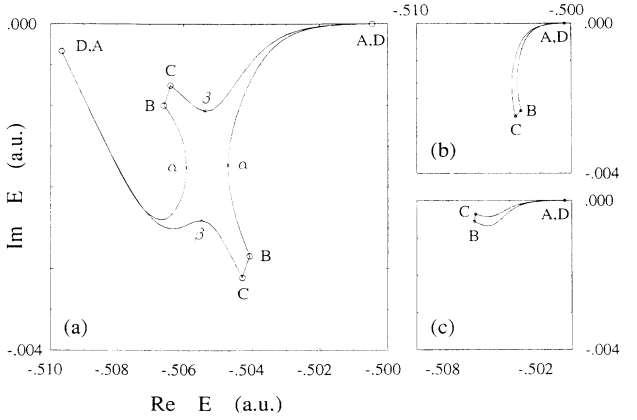

FIG. 1. Trajectories of the complex quasienergy of the $1s$ level of the hydrogen atom when the atom is irradiated by a coherent superposition of 355-nm light and its third harmonic; two parameters I_L and Φ_H are varied, where I_L is the intensity of the low-frequency component and Φ_H is the phase of the high-frequency component ($\Phi_L = 0$). At the points A–D, the values of I_L (in units of 10^{13} W/cm^2) and Φ_H (in degrees) are (A) $I_L = 0.1$, $\Phi_H = 0°$; (B) $I_L = 1.30$, $\Phi_H = 0°$; (C) $I_L = 1.30$, $\Phi_H = 160°$; (D) $I_L = 0.1$, $\Phi_H = 160°$. The intensity of the harmonic field is (a) $0.075I_L$, (b) $0.150I_L$, and (c) $0.040I_L$. In case (a) there is an avoided crossing near the points marked α, and a true crossing near the points marked β.

state corresponding to a particular adiabatically evolving eigenvalue changes from $1s$ to $2p$, or vice versa, as the avoided crossing is passed. On the other hand, the characters of the adiabatic states do not change at a true crossing of the real parts of the quasienergies. Hence, in case (a), the $1s$ state adiabatically evolves into the $2p$ state after one complete circuit, and vice versa. However, if the circuit is traversed again, the $2p$ state adiabatically evolves into the $1s$ state, and vice versa. Therefore, upon *two* circuits, when the avoided crossing is passed twice, the $1s$ state is mapped into itself aside from a geometric phase. As discussed further in Sec. III B and Appendix B, a degeneracy is enclosed by a circuit in case (a). In case (b) the real part of the $1s$ eigenvalue undergoes an avoided crossing as I_L is varied along the line $\Phi_H = 0°$, and another avoided crossing as I_L is varied back to zero along the line $\Phi_H = 160°$. In case (c) the real part of the $1s$ eigenvalue undergoes a *true* crossing as I_L is varied along the line $\Phi_H = 0°$, and another true crossing as I_L is varied back to 0.1×10^{13} W/cm^2 along the line $\Phi_H = 160°$. Therefore, in both cases (b) and (c) the $1s$ state is mapped into itself, aside from a geometric phase, after only *one* circuit. In these cases the circuit normally does not enclose any degeneracies, but it may happen that the circuit encloses two degeneracies.

As noted above, the geometric phase is complex when the relative phase of the two components of the bichromatic field is varied. Once again, however, the imaginary part of the geometric phase may be viewed as dynamical in origin. For a temporal variation in the relative phase is equivalent to a loss of commensurability of the two frequencies, and this may be taken into account by making a *double* Fourier-series expansion of the wave function using two incommensurable frequencies [8]. Nevertheless, it is more elegant to view the modification to the ionization yield as being due to a complex geometric phase rather than a loss of commensurability.

In the following section we set up the Floquet eigenvalue problem, and we briefly discuss the behavior of the Hamiltonian and its eigenvectors under time reversal. In Sec. III we examine the geometric phase, we investigate the behavior of the Floquet eigenvectors and eigenvalues near a degeneracy, and we give an illustration for the hydrogen atom. In Appendix A we discuss the effect of time reversal in more detail, and we construct the appropriate norm for the Floquet eigenvector. In Appendix B we examine the general analytic (branch point) structure of the Floquet eigenvalue.

II. THE EIGENVALUE PROBLEM

A. The interaction

We consider an atom exposed to a bichromatic radiation field having two commensurable frequencies ω_H and ω_L with $q\omega_H = p\omega_L$, where p and q are integers such that $p > q$ (the subscripts L and H stand for low- and high-frequency, respectively). Introducing the "fundamental" frequency

$$\omega \equiv (1/q)\omega_L, \tag{3}$$

we have $\omega_L = q\omega$ and $\omega_H = p\omega$. We assume that the two component fields are polarized in the same plane, the xy plane say, and we write the unit (complex) polarization vectors as

$$\hat{\epsilon}_L = \cos(\xi_L/2)\hat{\mathbf{x}} + i\sin(\xi_L/2)\hat{\mathbf{y}}, \tag{4}$$

$$\hat{\epsilon}_H = \cos(\xi_H/2)\hat{\mathbf{x}} + i\sin(\xi_H/2)\hat{\mathbf{y}}, \tag{5}$$

where ξ_L and ξ_H are the ellipticity parameters. Treating the radiation as a classical field, within the dipole approximation, the vector potential is

$$\mathbf{A}(t,\Phi) = A_L \mathrm{Im}(\hat{\epsilon}_L e^{-i\Phi_L - i\int^t dt'\,\omega_L}) + A_H \mathrm{Im}(\hat{\epsilon}_H e^{-i\Phi_H - i\int^t dt'\,\omega_H}), \tag{6}$$

where A_L and A_H are real, and where the argument Φ of the vector potential *now* denotes the two phases Φ_L and Φ_H *collectively*. Note that we could transform one of these two phases away by a transformation of ω; e.g., to transform Φ_L away, we absorb it into $q\int^t dt'\omega$, so $\omega \to \omega - (1/q)d\Phi_L/dt$. In other words, only the relative phase $\Phi_H - (p/q)\Phi_L$ is significant. The interaction of an electron, of charge e and mass μ, with the field is $-(e/\mu c)\mathbf{A}(t,\Phi)\cdot\mathbf{p}$, where $\mathbf{p}$ is the canonical momentum. Hence the interaction $V(t,\Phi)$ of the atom with the field can be expressed as

$$V(t,\Phi) = V_{L+}(\Phi_L)e^{-i\int^t dt'\,\omega_L} + V_{L-}(\Phi_L)e^{i\int^t dt'\,\omega_L} + V_{H+}(\Phi_H)e^{-i\int^t dt'\,\omega_H} + V_{H-}(\Phi_H)e^{i\int^t dt'\,\omega_H}, \tag{7}$$

where, assuming for simplicity a one-electron atom (we merely sum over electrons in the case of a many-electron atom),

$$V_{L+}(\Phi_L) = i(e/2\mu c)A_L e^{-i\Phi_L}\hat{\epsilon}_L\cdot\mathbf{p}, \tag{8}$$

$$V_{L-}(\Phi_L) = V_{L+}(\Phi_L)^\dagger, \tag{9}$$

$$V_{H+}(\Phi_H) = i(e/2\mu c)A_H e^{-i\Phi_H}\hat{\epsilon}_H\cdot\mathbf{p}, \tag{10}$$

$$V_{H-}(\Phi_H) = V_{H+}(\Phi_H)^\dagger. \tag{11}$$

Note that $V(t,\Phi)$ is locally periodic with period $2\pi/\omega$, provided that ω is nearly constant over one cycle.

We have only indicated the explicit dependence of $V(t,\Phi)$ on the time t. The interaction also depends implicitly on t through the temporal variation of A_L, A_H, and possibly other parameters, e.g., ξ, Φ, and ω (but not ω_L and ω_H independently). However, we assume that the evolution of the system is adiabatic, which requires that the implicit temporal variation of the parameters be slow on the time scale of one cycle $2\pi/\omega$ and on the time scale set by the Rabi flopping frequency if the atom is nearly on resonance with the light.

B. Floquet ansatz

The time-dependent Schrödinger equation is

$$i\hbar\frac{d}{dt}\,|\,\Psi(t,\Phi)\rangle = [H_a + V(t,\Phi)]\,|\,\Psi(t,\Phi)\rangle, \tag{12}$$

where H_a is the atomic Hamiltonian. To pass to the time-independent problem, we first remove a dynamical

phase factor and write the state vector of the atom as

$$|\Psi(t,\Phi)\rangle = e^{-i\int^t dt' E(\Phi)/\hbar}\,|\mathcal{F}(t,\Phi)\rangle, \tag{13}$$

and substitute into Eq. (12) to give

$$i\hbar\frac{d}{dt}\,|\mathcal{F}(t,\Phi)\rangle = [H_a + V(t,\Phi) - E(\Phi)]\,|\mathcal{F}(t,\Phi)\rangle. \tag{14}$$

We now take note of the fact that, at least over an interval of a few cycles, $V(t,\Phi)$ is periodic, and we make the harmonic expansion

$$|\mathcal{F}(t,\Phi)\rangle = \sum_N e^{-iN\int^t dt'\,\omega}\,|\mathcal{F}_N(\Phi)\rangle, \tag{15}$$

which, upon substitution into Eq. (14), using Eq. (7), yields

$$\sum_N H^{(MN)}(\Phi)\,|\mathcal{F}_N(\Phi)\rangle = E(\Phi)\,|\mathcal{F}_M(\Phi)\rangle + i\hbar\frac{d}{dt}\,|\mathcal{F}_M(\Phi)\rangle, \tag{16}$$

where $H^{(MN)}(\Phi)$ is the operator

$$\begin{aligned} H^{(MN)}(\Phi) = (\,&H_a - M\hbar\omega)\delta_{MN} + V_{L+}(\Phi_L)\delta_{M,N+q} \\ &+V_{L-}(\Phi_L)\delta_{M,N-q} \\ &+V_{H+}(\Phi_H)\delta_{M,N+p} + V_{H-}(\Phi_H)\delta_{M,N-p}. \end{aligned} \tag{17}$$

The "Floquet" Hamiltonian, denoted by $H(\Phi)$, is the matrix operator whose elements are $H^{(MN)}(\Phi)$. If we denote by $|\,\mathcal{F}(\Phi)\rangle\rangle$ the column vector whose elements are the harmonic components $|\,\mathcal{F}_M(\Phi)\rangle$, we can reexpress Eq. (16) as

$$H(\Phi)\,|\,\mathcal{F}(\Phi)\rangle\rangle = E(\Phi)\,|\,\mathcal{F}(\Phi)\rangle\rangle + i\hbar\frac{d}{dt}\,|\,\mathcal{F}(\Phi)\rangle\rangle. \tag{18}$$

The Floquet ansatz, which is appropriate when the state of the atom changes slowly on the time scale of one cycle $2\pi/\omega$, amounts to neglecting the term in the time derivative on the right-hand side of Eq. (18). This ansatz results in the eigenvalue problem

$$H(\Phi)\,|\,\mathcal{F}(\Phi)\rangle\rangle = E(\Phi)\,|\,\mathcal{F}(\Phi)\rangle\rangle. \tag{19}$$

The eigenvalue $E(\Phi)$ is complex if the system is dissipative, i.e., if the atom ionizes; the imaginary part of the eigenvalue is, aside from a sign, the half-width for stationary decay. Note that since we can always transform one of the two phases Φ_L and Φ_H away by a shift in the origin of time, without affecting the eigenvalue, $E(\Phi)$ can only depend on the *relative* phase $\Phi_H - (p/q)\Phi_L$. Note also that $E(\Phi)$ may vary implicitly on time through various parameters, and we stress this must be taken into account in evaluating the dynamical phase factor $\exp[-i\int^t dt' E(\Phi)/\hbar]$.

In constructing a convenient norm for the Floquet eigenvector $|\,\mathcal{F}(\Phi)\rangle\rangle$, one which is conserved in time, we exploit the time and reflection symmetries of the Hamiltonian. To this end, we introduce the operator $\mathcal{S}$ which is the product of the time-reversal operator and the operator which effects a reflection in the xz plane. We discuss the properties of $\mathcal{S}$ in more detail in Appendix A. We note here that $\mathcal{S}^\dagger\mathcal{S} = \mathcal{S}\mathcal{S}^\dagger = 1$ and $\mathcal{S}^\dagger = \mathcal{S}$; however, since $\mathcal{S}$ is *anti*linear, and therefore complex conjugates c numbers [9], it is *anti*unitary. We denote the action of $\mathcal{S}$ by a superscript $(*)$, e.g., we write $|\,\psi^{(*)}\rangle = \mathcal{S}\,|\,\psi\rangle$ for any ket $|\,\psi\rangle$. It follows from the antilinearity of $\mathcal{S}$ that if $|\,\psi\rangle$ and $|\,\chi\rangle$ are any two kets [9]

$$\langle\psi^{(*)}\,|\,\chi\rangle = \langle\chi^{(*)}\,|\,\psi\rangle. \tag{20}$$

In Appendix A we show that

$$\mathcal{S}V_{L\pm}(\Phi_L)\mathcal{S}^\dagger = V_{L\pm}(-\Phi_L), \tag{21}$$

$$\mathcal{S}V_{H\pm}(\Phi_H)\mathcal{S}^\dagger = V_{H\pm}(-\Phi_H), \tag{22}$$

and also that [7]

$$E(-\Phi) = E(\Phi). \tag{23}$$

Since $\mathcal{S}$ commutes with H_a it follows that

$$\mathcal{S}H(\Phi)\mathcal{S}^\dagger = H(-\Phi), \tag{24}$$

and hence that

$$H(\Phi)\,|\,\mathcal{F}^{(*)}(-\Phi)\rangle\rangle = E^*(\Phi)\,|\,\mathcal{F}^{(*)}(-\Phi)\rangle\rangle. \tag{25}$$

In other words, $\langle\langle\mathcal{F}^{(*)}(-\Phi)\,|$ is a left-hand eigenvector of $H(\Phi)$ with eigenvalue $E(\Phi)$.

If $|\,\psi(t)\rangle$ and $|\,\chi(t)\rangle$ are any two kets which are periodic in t, with period $2\pi/\omega$, and if their harmonic components are $|\,\psi_N\rangle$ and $|\,\chi_N\rangle$, we define column vectors $|\,\psi\rangle\rangle$ and $|\,\chi\rangle\rangle$ whose elements are the harmonic components. Furthermore, we define the following scalar product:

$$\langle\langle\psi\,|\,\chi\rangle\rangle = \sum_N\langle\psi_N\,|\,\chi_N\rangle. \tag{26}$$

In particular, from Eq. (A15), we have

$$\langle\langle\mathcal{F}^{(*)}(-\Phi)\,|\,\mathcal{F}(\Phi)\rangle\rangle = 1. \tag{27}$$

As we show in Sec. III A, this normalization holds for all times, as long as the atom evolves adiabatically.

C. Floquet matrix

We can represent $H(\Phi)$ as a matrix by expanding the harmonic components on a basis, the set $|\,b_{Nn}\rangle, n = 1,2,\ldots$, say. Thus, writing

$$|\,\mathcal{F}_N(\Phi)\rangle = \sum_n a_{Nn}\,|\,b_{Nn}\rangle, \tag{28}$$

we can construct the Floquet matrix, $\mathbf{H}(\Phi)$, which is a block matrix, with the $(M\text{-}N)$th block a submatrix whose $(m\text{-}n)$th element is $\langle b^{(*)}_{Mm}\,|\,H^{(MN)}(\Phi)\,|\,b_{Nn}\rangle$; only those blocks for which $M = N$, $M = N \pm p$, or $M = N \pm q$ are nonvanishing. Using $\mathcal{S}^\dagger\mathcal{S} = 1$ and Eqs. (22) and (11), and, in the last step below, Eq. (20) with $\mathcal{S}^\dagger = \mathcal{S}$ — in the last step we use $\langle\psi\,|\,[\mathcal{S}^\dagger\chi\rangle] = \langle\psi\,|\,[\mathcal{S}\chi\rangle] = \langle\chi\,|\,[\mathcal{S}\psi\rangle]$ — we have

$$\langle b^{(*)}_{Mm} \mid V_{H\pm}(\Phi_H) \mid b_{Nn}\rangle = \langle b^{(*)}_{Mm} \mid \mathcal{S}^\dagger V_{H\pm}(-\Phi_H)\mathcal{S} \mid b_{Nn}\rangle$$
$$= \langle b^{(*)}_{Mm} \mid \mathcal{S}^\dagger V^\dagger_{H\mp}(-\Phi_H) \mid b^{(*)}_{Nn}\rangle$$
$$= \langle b^{(*)}_{Nn} \mid V_{H\mp}(-\Phi_H) \mid b_{Mm}\rangle, \tag{29}$$

with a similar relation satisfied by the matrix elements of $V_{L\pm}(\Phi_L)$. It follows that, with the superscript t denoting the transpose,

$$\mathbf{H}(\Phi)^t = \mathbf{H}(-\Phi), \tag{30}$$

and therefore if $H(\Phi) = H(-\Phi)$ the matrix $\mathbf{H}(\Phi)$ is complex symmetric. Let $\mathbf{N}$ be the overlap matrix, which is a block *diagonal* matrix; the Nth diagonal block is a submatrix whose elements are $\langle b^{(*)}_{Nm} \mid b_{Nn}\rangle$, and from Eq. (20) we see that $\mathbf{N}$ is complex symmetric, that is, $\mathbf{N}^t = \mathbf{N}$. If $\mathbf{b}$ is a column vector with elements b_{Nn}, the eigenvalue problem has the matrix form

$$\mathbf{H}(\Phi)\mathbf{b} = E(\Phi)\mathbf{N}\mathbf{b}. \tag{31}$$

Incidentally, since a matrix and its transpose have the same eigenvalue spectra, it immediately follows from Eqs. (30) and (31) that $E(-\Phi) = E(\Phi)$, in accordance with Eq. (23).

III. THE GEOMETRIC PHASE

A. Preliminary discusssion

Suppose that the Hamiltonian $H(\Phi)$ depends implicitly on one or more parameters which vary slowly with time. Let $\mathbf{\Lambda}$ collectively denote these parameters. If the temporal variation of $H(\Phi)$ is sufficiently slow, the system will remain in an eigenstate of (the evolving) $H(\Phi)$; this is the adiabatic hypothesis — the Floquet ansatz in the present context — and we have made this hypothesis in neglecting the time derivative on the right-hand side of Eq. (18). Taking the gradient with respect to $\mathbf{\Lambda}$ of both sides of Eq. (19), we obtain

$$[E(\Phi) - H(\Phi)]\nabla_{\mathbf{\Lambda}} \mid \mathcal{F}(\Phi)\rangle\rangle$$
$$= -[\nabla_{\mathbf{\Lambda}}E(\Phi) - \nabla_{\mathbf{\Lambda}}H(\Phi)] \mid \mathcal{F}(\Phi)\rangle\rangle. \tag{32}$$

In view of Eq. (25), projection of $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid$ onto the left-hand side of Eq. (32) must yield zero. Therefore, introducing the projection operator

$$Q \equiv 1 - \mid \mathcal{F}(\Phi)\rangle\rangle\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid, \tag{33}$$

and multiplying the right-hand side of Eq. (32) by Q, we can formally express the gradient of $\mid \mathcal{F}(\Phi)\rangle\rangle$ as

$$\nabla_{\mathbf{\Lambda}} \mid \mathcal{F}(\Phi)\rangle\rangle = C \mid \mathcal{F}(\Phi)\rangle\rangle + [E(\Phi) - H(\Phi)]^{-1}$$
$$\times Q[\nabla_{\mathbf{\Lambda}}H(\Phi)] \mid \mathcal{F}(\Phi)\rangle\rangle, \tag{34}$$

where the term in $\nabla_{\mathbf{\Lambda}}E(\Phi)$ is absent since it vanishes upon multiplication by Q, provided that Eq. (27) holds. The right-hand side of Eq. (34) is nonsingular as long as $E(\Phi)$ is nondegenerate, but if $E(\Phi)$ is degenerate the last term on the right-hand side is singular. At first sight, the constant C appears to be arbitrary, although of course if we wish to preserve the normalization of Eq. (27) our choice of C will effect $\mid \mathcal{F}^{(*)}(-\Phi)\rangle\rangle$. In fact, however, we must choose $C = 0$, for otherwise the neglect of the term in the derivative with respect to t on the right-hand side of Eq. (18) would not be justified. Indeed, noting that this derivative equals $(d\mathbf{\Lambda}/dt)\cdot\nabla_{\mathbf{\Lambda}}$, and taking into account Eq. (25), we must have, from Eq. (18),

$$\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \nabla_{\mathbf{\Lambda}} \mid \mathcal{F}(\Phi)\rangle\rangle = 0, \tag{35}$$

a condition which, in general, forces $\mid \mathcal{F}(\Phi)\rangle\rangle$ to be multivalued. It follows that if $\mid \bar{\mathcal{F}}(\Phi)\rangle\rangle$ is any solution of the eigenvalue problem which is continuous and differentiable in Λ, we have

$$\mid \mathcal{F}(\Phi)\rangle\rangle = e^{i\bar{B}} \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle, \tag{36}$$

where $\bar{B}$ is given by

$$i(\nabla_{\mathbf{\Lambda}}\bar{B})\langle\langle\bar{\mathcal{F}}^{(*)}(-\Phi) \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle$$
$$+\langle\langle\bar{\mathcal{F}}^{(*)}(-\Phi) \mid \nabla_{\mathbf{\Lambda}} \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle = 0. \tag{37}$$

If we follow the eigenvalue $E(\Phi)$ along a closed circuit in parameter space, it must return to its original value upon completion of the circuit unless the eigenvalue is degenerate at some point inside or on the loop. For, barring a degeneracy, $E(\Phi)$ must be single valued everywhere inside and on the loop. Consequently, the eigenvector $\mid \mathcal{F}(\Phi)\rangle\rangle$ must return to its original form, aside from a geometrical phase factor, e^{iB} say. In other words, the eigen*ray* is mapped into itself. The geometric phase B is, using Eq. (37), the closed circuit integral

$$B = i\oint \frac{\langle\langle\bar{\mathcal{F}}^{(*)}(-\Phi) \mid \nabla_{\mathbf{\Lambda}} \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle}{\langle\langle\bar{\mathcal{F}}^{(*)}(-\Phi) \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle}\cdot d\mathbf{\Lambda}, \tag{38}$$

provided that $\mid \bar{\mathcal{F}}(\Phi)\rangle\rangle$ is chosen so that it returns to itself exactly after one circuit [11]. [In contrast to $\mid \bar{\mathcal{F}}(\Phi)\rangle\rangle$, which returns to itself without a phase change, $\mid \mathcal{F}(\Phi)\rangle\rangle$ carries the geometric phase.] If the loop does enclose a degeneracy of $E(\Phi)$, one circuit is normally (but not always — see below) insufficient to return the eigenvalue and eigenray to their original values, for in following the loop a branch cut in the complex plane of one of the parameters is normally traversed (see Sec. III B and Appendix B), and in this case more than one closed circuit must be followed before the eigenvalue and eigenray return to their original values (the branch cut must be crossed again). In the special case where $H(-\Phi) = H(\Phi)$, that is, where $H(\Phi)$ is invariant under the "time reversal" effected by $\mathcal{S}$, we can, assuming $E(\Phi)$ is nondegenerate, choose $\mid \mathcal{F}(-\Phi)\rangle\rangle$ to be equal to $\mid \mathcal{F}(\Phi)\rangle\rangle$. In this case, the geometric phase can only be either 0 or $\pm\pi$, since if $\mid \mathcal{F}(\Phi)\rangle\rangle$ returns to $e^{iB} \mid \mathcal{F}(\Phi)\rangle\rangle$, then $\mid \mathcal{F}^{(*)}(\Phi)\rangle\rangle$ returns to $e^{-iB^*} \mid \mathcal{F}^{(*)}(\Phi)\rangle\rangle$, and it follows from the normalization, Eq. (27), that $e^{2iB} = 1$. In fact, in this case, if the circuit does not enclose a degeneracy, $B = 0$; this follows by shrinking the circuit to zero, noting that B must vanish for an infinitesimally small circuit, and that B cannot abruptly change from $\pm\pi$ to zero

as the circuit shrinks unless the circuit passes through a degeneracy. The condition $H(-\Phi) = H(\Phi)$ is satisfied for $\Phi = 0$, $\pm\pi$, or $\pm 2\pi$. If the field is monochromatic (i.e., one of the two components of the bichromatic field is absent), the eigenvalue $E(\Phi)$ is independent of Φ, and since we can always satisfy $H(-\Phi) = H(\Phi)$ by shifting the origin of time, we must have $B = 0$ or $\pm\pi$ for all Φ — *provided* that Φ is not varied, for we can only shift the origin of time once [12]. The preceding statements on B are true whether or not the system is dissipative. However, if the system is nondissipative, $E(\Phi)$ is real, and therefore, recalling Eq. (25) and assuming $E(\Phi)$ is nondegenerate, $|\mathcal{F}^{(*)}(-\Phi)\rangle\rangle$ is proportional to $|\mathcal{F}(\Phi)\rangle\rangle$; choosing the proportionality constant to be unity, Eq. (27) becomes $\langle\langle\mathcal{F}(\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle = 1$, and it follows that B is real even if Φ is varied. As noted in the Introduction, if the system is dissipative, B may be complex, unless $H(\Phi)$ is invariant under $\mathcal{S}$, in which case, as just shown, B is real, and is an integer multiple of π, even when the system is dissipative.

In Appendix A we prove that the norm $\langle\mathcal{F}^{(*)}(-\tilde{t} + t_0, -\Phi) \mid \mathcal{F}(\tilde{t} + t_0, \Phi)\rangle$ is conserved over time intervals for which $V(t, \Phi)$ is (nearly) periodic, and as long as the adiabatic hypothesis holds, namely, as long as the system evolves as an eigenstate of the slowly varying $H(\Phi)$, we expect this norm to remain nonzero at each point in parameter space — we can thereby impose the normalization of Eq. (27), that is $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle = 1$. However, the evolution of the system ceases to be adiabatic if the trajectory in parameter space passes through a degeneracy of $E(\Phi)$. Hence we may expect that at a degeneracy $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle$ can vanish. We now show, independently of the adiabatic hypothesis, that $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle$ can vanish only at a degeneracy. We are always free to demand that, in the absence of degeneracies, Eq. (35) holds for all Φ, since we are free to choose $C = 0$ in Eq. (34). Since $\mathcal{S}$ commutes with ∇_Λ, it follows, using Eq. (20), that, in the absence of degeneracies

$$\langle\langle\mathcal{F}(\Phi) \mid \nabla_\Lambda \mid \mathcal{F}^{(*)}(-\Phi)\rangle\rangle = 0 \tag{39}$$

and this implies that

$$\nabla_\Lambda\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle = 0. \tag{40}$$

According to Eq. (40), the norm $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle$ can be chosen so as to be constant, and therefore nonzero, in any region of parameter space where $E(\Phi)$ is nondegenerate; in other words, $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle$ can vanish only at those exceptional points where $E(\Phi)$ is degenerate. Incidentally, since Eq. (35), and therefore Eq. (40), must hold if the atom evolves adiabatically, $\langle\langle\mathcal{F}^{(*)}(-\Phi) \mid \mathcal{F}(\Phi)\rangle\rangle$ remains constant for all time if the adiabatic hypothesis holds.

B. Behavior at degeneracies

We now examine the behavior of the eigenvalues and eigenvectors in the neighborhood of a degeneracy. [We remind the reader that the geometric phase may be nonzero even if the circuit does not enclose a degeneracy, unless $\mathbf{H}(\Phi)$ is complex symmetric.] We may reduce the full Floquet matrix to a 2×2 matrix which pertains to the subspace of the two states that have nearly degenerate eigenvalues. Let us denote the diagonal and off-diagonal elements of this matrix by H_{11} and H_{22}, and by H_{12} and H_{21}, respectively. These elements may be, for example, the matrix elements of one-dimensional blocks $H^{(11)}(\Phi)$, $H^{(22)}(\Phi)$, $H^{(12)}(\Phi)$, and $H^{(21)}(\Phi)$. The eigenvalues of this matrix are $E_\pm \equiv \bar{E} \pm \Delta E/2$, where

$$\bar{E} = (H_{11} + H_{22})/2, \tag{41}$$

$$\Delta E = \sqrt{(H_{11} - H_{22})^2 + 4H_{12}H_{21}}. \tag{42}$$

A degeneracy occurs when $\Delta E = 0$, that is, when either

$$H_{11} - H_{22} + 2i\sqrt{H_{12}H_{21}} = 0 \tag{43}$$

or

$$H_{11} - H_{22} - 2i\sqrt{H_{12}H_{21}} = 0. \tag{44}$$

Without loss in generality, we may assume that it is Eq. (43) that holds, although in the case where the matrix is Hermitian Eq. (44) also holds. If the matrix is (real or complex) symmetric, two parameters must in general be adjusted to arrange for the real and imaginary parts of the left-hand side of Eq. (43) to vanish. If the matrix is Hermitian, H_{11} and H_{22} are real and $H_{12} = H_{21}^*$, and therefore we require that $H_{11} - H_{22} = 0$ and $H_{12} = 0$; hence, for a complex Hermitian matrix (H_{12} complex) three parameters must be adjusted to satisfy Eq. (43). However, if the system is dissipative, the matrix is not Hermitian (it may or may not be symmetric) and we may only need to vary two parameters to satisfy Eq. (43). We suppose that we have only two parameters at our disposal, λ and μ say, and that there is a degeneracy when these parameters have the *real* values λ_0 and μ_0. For $(\lambda, \mu) \approx (\lambda_0, \mu_0)$ we have, assuming the matrix elements are analytic and single valued in λ and μ,

$$\Delta E \propto \sqrt{P(\lambda - \lambda_0) + Q(\mu - \mu_0)}, \tag{45}$$

where P and Q are (possibly complex) constants. Writing $z \equiv P\lambda + Q\mu$, we have

$$\Delta E \propto \sqrt{z - z_0}, \tag{46}$$

with $z_0 \equiv P\lambda_0 + Q\mu_0$. In general, E is not an analytic function of z (the Cauchy-Riemann equations are not satisfied in the z plane). However, E is an analytic function of λ (and of μ), and it follows from Eq. (46) that if $\mu = \mu_0$, the energy normally has a branch point on the real axis of the complex λ plane, at λ_0. However, if the matrix is real and symmetric, Eq. (44) also holds, so that ΔE has a double zero at (λ_0, μ_0). For a matrix that is nearly real and symmetric, the double zero separates into two isolated zeros, and there are two nearby degeneracies in the λ-μ plane; in the neighborhood of these degeneracies we have

$$\Delta E \approx \sqrt{(z - z_0)(z' - z_0')}, \tag{47}$$

where the variable z' can be expressed as $P'\lambda + Q'\mu$, and where $z_0' \approx z_0^*$. Evidently, for a matrix that is exactly real and symmetric we have $z' = z^*$ and $z_0' = z_0^*$, so that

the branch point on the real λ axis, at λ_0, disappears when $\mu = \mu_0$. We can parametrize our 2×2 matrix by the Cayley-Klein parameters θ and ϕ, which in general are complex and are defined by

$$e^{2i\theta} = \frac{(H_{11} - H_{22}) + 2i\sqrt{H_{12}H_{21}}}{(H_{11} - H_{22}) - 2i\sqrt{H_{12}H_{21}}}, \tag{48}$$

$$e^{2i\phi} = H_{21}/H_{12}. \tag{49}$$

(Note that $\phi = 0$ if the matrix is symmetric, and that $\theta = \pm i\infty$ at a degeneracy.) The eigenvectors are

$$u_+(\theta,\phi) = \begin{pmatrix} \cos(\theta/2)e^{-i\phi/2} \\ \sin(\theta/2)e^{i\phi/2} \end{pmatrix}, \tag{50}$$

$$u_-(\theta,\phi) = \begin{pmatrix} \sin(\theta/2)e^{-i\phi/2} \\ -\cos(\theta/2)e^{i\phi/2} \end{pmatrix}, \tag{51}$$

and, recalling that the superscript t denotes transpose, we have $u_\pm(\theta,-\phi)^t u_\pm(\theta,\phi) = 1$ and $u_\mp(\theta,-\phi)^t u_\pm(\theta,\phi) = 0$. A loop in the λ-μ plane which encircles the point (λ_0,μ_0) corresponds to a loop in the z plane which encircles z_0. Now, as long as this loop does not enclose both degeneracies, the numerator of the right-hand side of Eq. (48), which is proportional to $z - z_0$, has an argument which changes by $\pm 2\pi$ as the loop is followed (the denominator is proportional to $z' - z_0'$ and its argument would also change by $\pm 2\pi$ if both degeneracies were enclosed). Therefore θ changes by $\pm\pi$, and hence the eigenvectors transform into one another upon a complete circuit; for example, if θ increases by π, we have $u_+(\theta,\phi) \rightarrow -u_-(\theta,\phi)$ and $u_-(\theta,\phi) \rightarrow u_+(\theta,\phi)$. From Eq. (46) we see that E_+ and E_- also interchange. Upon a second circuit, the original eigenrays are mapped into themselves, and the original eigenvectors are mapped into themselves up to a geometrical phase factor of $\pm\pi$. If the loop encloses both of the degeneracies in the λ-μ plane, θ changes by $\pm 2\pi$, and the original eigenrays are mapped into themselves after a single circuit.

Note that $u_+(\theta,\phi)$ does not return to itself exactly after a double loop, rather, $u_+(\theta,\phi)$ carries the geometric phase. A similar remark applies to $u_-(\theta,\phi)$. Thus, if we were to calculate the geometric phase from Eq. (38), around a double loop enclosing a single degeneracy say, and if we were to use one of the eigenvectors $u_+(\theta,\phi)$ and $u_-(\theta,\phi)$, say $u_+(\theta,\phi)$, we would obtain the value $B' \equiv \int u_+(\theta,-\phi)^t du_+(\theta,\phi) = -(i/2)\int \cos(\theta)d\phi$ rather than the correct answer $B = B' \pm \pi$. Note that the geometric phase associated with a double loop around the degeneracy is, in general, complex and does not equal π as it would in the case of a symmetric Hamiltonian (for which $\phi \equiv 0$ and hence $B' \equiv 0$). However, the geometric phase is exactly π, if upon the second turn of the double loop the contour is retraced exactly — at least to the extent that the system can be adequately modeled by a two-state approximation. To see this, we first note that after a single turn around the loop, θ has increased or decreased by π, as explained above. Since $\cos(\theta \pm \pi) = -\cos(\theta)$, we find upon evaluating $B' = -(i/2)\int \cos(\theta)d\phi$ that the contribution from the first loop exactly cancels the contribution from the second loop. However, in the general case where the loop does not retrace itself, no such cancellation occurs and the geometric phase will be complex (and hence differ from π).

C. Illustration

We now discuss the results obtained for the geometric phase for the hydrogen atom irradiated by bichromatic light, corresponding to the three cases (a)–(c) of Fig. 1 that we discussed in the Introduction. We recall that the intensity I_L of the low-frequency component and the phase Φ_H of the high-frequency component are varied along a rectangle in the I_L-Φ_H plane, whose sides correspond to either constant I_L or constant Φ_H; we fix $\Phi_L = 0$, but I_H varies with I_L along the circuit because the ratio I_H/I_L is kept constant. We have labeled the corners of the rectangle in the I_L-Φ_H plane counterclockwise as A, B, C, and D, starting from the left lower corner. To calculate the geometric phase, we used the expression of Eq. (38), which requires the wave function to be single valued. We have enforced this single

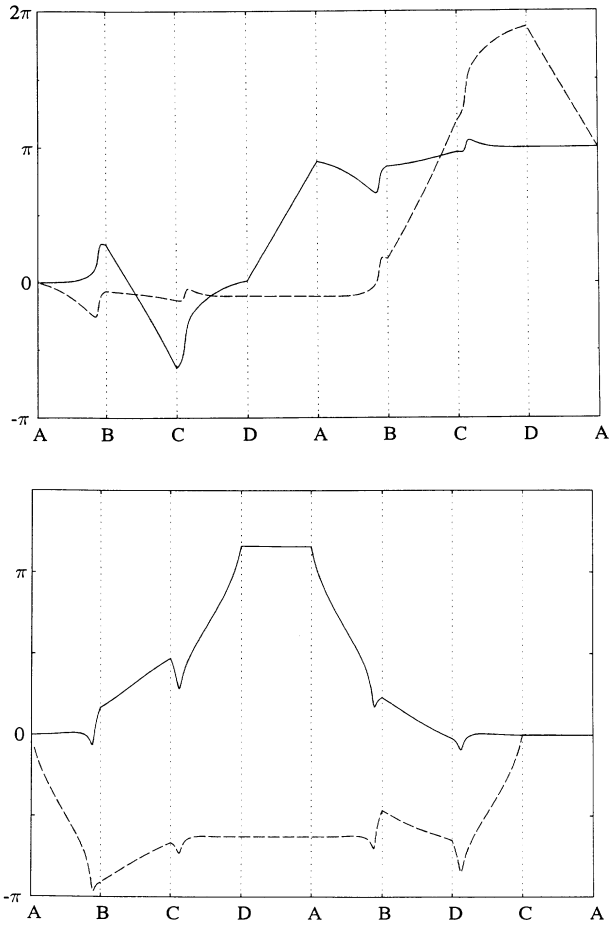

FIG. 2. The geometric phase corresponding to Fig. 1(a). Solid curve, normalization of the wave function such that its projection on the $1s$ state equals unity; dashed curve, normalization with the projection on the $2p$ state set equal to unity (see text). The upper part of the figure shows the real part of the geometric phase. The lower part of the figure shows the imaginary part.

valuedness somewhat arbitrarily, by requiring the projection of the wave function onto the (unperturbed) $1s$ or $2p$ state to be unity everywhere on the circuit, or, more precisely, $\langle 1s \mid \bar{\mathcal{F}}_0(\Phi)\rangle = 1$ and $\langle 1s \mid \bar{\mathcal{F}}_0(-\Phi)\rangle = 1$, or $\langle 2p \mid \bar{\mathcal{F}}_3(\Phi)\rangle = 1$ and $\langle 2p \mid \bar{\mathcal{F}}_3(-\Phi)\rangle = 1$. This also fixes the normalization. In order to illustrate how the geometric phase builds up, we show the value of

$$B(P_0, P) = i \int_{P_0}^{P} \frac{\langle\langle \bar{\mathcal{F}}^{(*)}(-\Phi) \mid \nabla_{\mathbf{\Lambda}} \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle}{\langle\langle \bar{\mathcal{F}}^{(*)}(-\Phi) \mid \bar{\mathcal{F}}(\Phi)\rangle\rangle} \cdot d\mathbf{\Lambda}, \quad (52)$$

as the point P describes the circuit in the parameter space, starting from the point $P_0 \equiv$ A and passing by the points B, C, and D; of course, once P returns to P_0, after a complete circuit, $B(P_0, P)$ is identical to the geometric phase of Eq. (38) (and does not depend on the normalization). In Figs. 2–4, respectively, we display the cases corresponding to Figs. 1(a)–1(c), respectively. In the upper part of each figure the real part of the $B(P_0, P)$ is shown, while in each lower part the imaginary part is shown. Note that, as indicated on the horizontal scale, a double loop was needed to map the $1s$ state back onto itself in case (a) (Fig. 2), whereas in cases (b) and (c) (Fig. 3 and 4, respectively) a single loop was taken. As seen from these figures, the value $B(P_0, P)$ is complex and for $P \neq P_0$ depends markedly on the choice of normalization. However, this is of no physical importance: a given choice of normalization amounts to multiplying the eigenvector $\mid \bar{\mathcal{F}}(\Phi)\rangle\rangle$ by a complex factor, which varies with the point P in different ways for different normalizations, and the differences in the values of $B(P_0, P)$ merely compensate for the variations in this complex factor. In other words, different normalizations of $\mid \bar{\mathcal{F}}(\Phi)\rangle\rangle$ lead to eigenvectors $\mid \mathcal{F}(\Phi)\rangle\rangle$ — defined by Eq. (36) with $\bar{B} \equiv B(P_0, P)$ — that differ only by a constant numerical factor. Hence no conclusions can be drawn, about the modifications to the ionization yield calculated from the width of the instantaneous quasienergy, from the magnitude of the imaginary part of $B(P_0, P)$ plotted in Figs. 2–4(b), as long as P has not reached the end point of the circuit. However, at the end point, a nonvanishing imaginary part of the geometric phase B indicates that the actual ionization yield is different than would have been obtained by assuming that at each instant the ionization rate is simply the instantaneous value of $-2\,\mathrm{Im}E/\hbar$.

In Figs. 3 and 4 we illustrate that, in accordance with the discussion in the preceding sections, the geometric phase is in general complex for a single loop not encircling a degeneracy, provided $H(-\Phi)$ is not equal to $H(\Phi)$. With reference to Fig. 2, the geometric phase is real and equal to π, within the accuracy of the computation. One may have expected the geometric phase for this case to

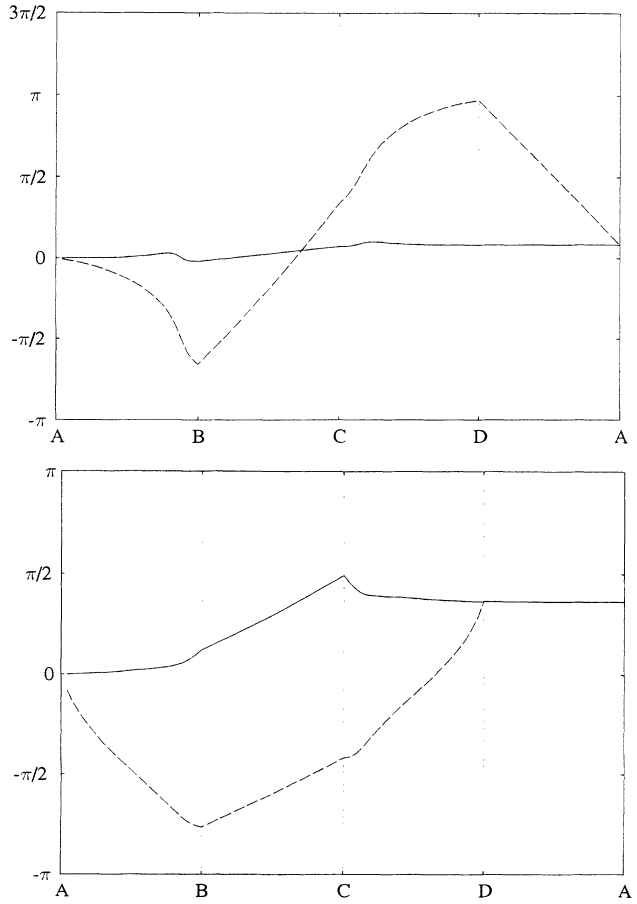

FIG. 3. Same as in Fig. 2, but for the case corresponding to Fig. 1(b).

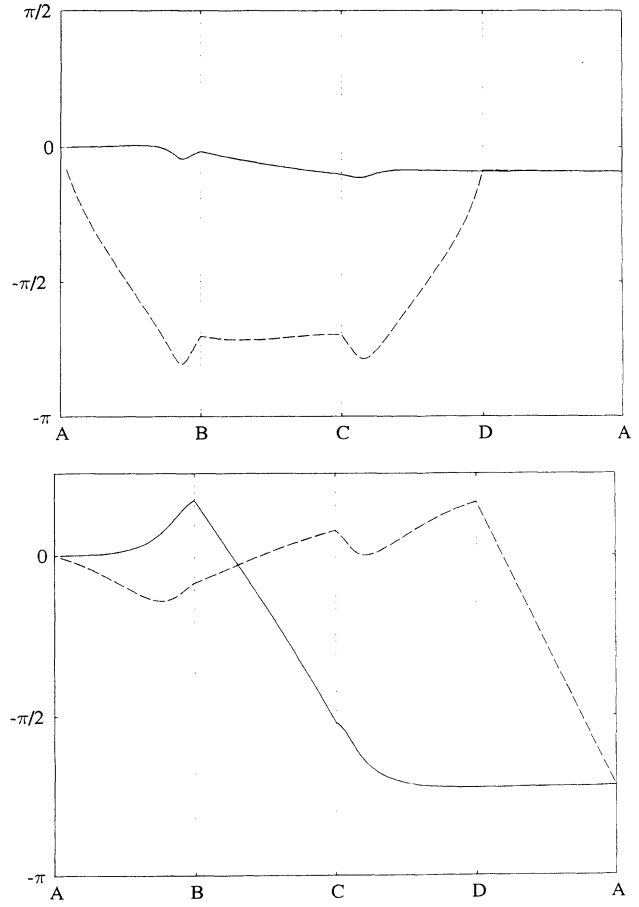

FIG. 4. Same as in Fig. 2, but for the case corresponding to Fig. 1(c).

be complex. However, this result is in accord with the analysis of Sec. III B, where we demonstrated that to the extent that the atom is adequately described by a two-state approximation, the geometric phase is equal to π (to the accuracy of the calculation) provided that the second loop retraces the first loop around the degeneracy. We have checked that the geometric phase is indeed complex if the second loop does not exactly retrace the first one (but still encircles the degeneracy). In another case, not illustrated here, Φ is constant but I_L and I_H are varied independently around the sides of a rectangle in the I_L-I_H plane defined by 1.1×10^{13} W/cm^2 $\leq I_L \leq 1.2\times10^{13}$ W/cm^2, 0.5×10^{12} W/cm^2 $\leq I_H \leq 1.5\times10^{12}$ W/cm^2. This loop encircles a degeneracy. As in Fig. 5, the geometric phase is approximately π after two identical loops [when $\Phi_L = 0$ and $\Phi_H = 10°$ we find $B \approx \pi - 0.14i$, but when $\Phi_L = \Phi_H = 0$, we find $B = \pi$ — not surprisingly since $H(-\Phi) = H(\Phi)$ and B cannot be complex].

Finally, we note that for monochromatic ionization the imaginary part of the geometric phase is zero if only the intensity, but not the phase of the field [14], is changed adiabatically: in this important case, the nonresonant decay rate can be safely evaluated from the imaginary part of the quasienergy.

IV. CONCLUSION

The wave function of an atom undergoing multiphoton ionization in a laser field may acquire a complex geometric phase on top of the usual dynamical phase when the phase of the laser field varies slowly with time. This implies that a naive calculation of the (adiabatic) ionization yield, obtained by integrating the instantaneous Floquet decay rate, may sometimes give an incorrect answer. We illustrated this for the case of a bichromatic field with commensurable frequencies whose relative phase is varied slowly with time. However, if the varying phase were to be incorporated by allowing for a drifting frequency, the geometric phase due to the variation of the phase of the light field would be entirely contained in the dynamical phase; but this would imply a loss of commensurability of the frequencies. In cases where the loop encloses a degeneracy, a *double* loop is in general necessary to map the original eigenray onto itself. We find that, whenever the second loop retraces the first one exactly, the geometric phase may be very close to π, a consequence of the fact that not too far from the degeneracy the atom may be accurately modeled by a two-state approximation. Finally, let us remark that for a frequency-chirped monochromatic laser pulse it may be most convenient to incorporate the imaginary part of the geometric phase in the dynamical phase through the dependence of the quasienergy on the frequency; the residual geometric phase then exactly equals a multiple of π, the multiple depending on the number of degeneracies of the quasienergy enclosed by the loop in the frequency-intensity plane.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation, under Grant No. PHY-9017079, by the United Kingdom Science and Engineering Research Council, and by a RIC grant from the University of Durham.

APPENDIX A: TIME-REVERSAL AND THE NORMALIZATION

In this appendix we examine the time symmetry of the interaction and, utilizing this symmetry, we construct a convenient norm for the state vector, which is conserved in time. (The usual norm is inappropriate, due to the exploding exponential behavior of the Floquet eigenfunction in position space.) The Hamiltonian of the system "atom plus radiation field" is invariant under time reversal. However, we treat the radiation field as an external entity. Thus we define the domain of the time-reversal operator $\mathcal{T}$ to be the space of atomic coordinates only. Note that

$$\mathcal{T}^\dagger\mathcal{T} = \mathcal{T}\mathcal{T}^\dagger = 1, \tag{A1}$$

but that $\mathcal{T}$ is *anti*unitary [9] since it complex conjugates c numbers. Note further that, since the time-reversal operator acting on the time-reversed world produces the original world, $\mathcal{T}^2$ must be the identity operator up to a phase factor, and from the fact that $\mathcal{T}$ is antiunitary the phase factor must be ±1:

$$\mathcal{T}^2 = \pm1. \tag{A2}$$

The interaction of the atom with the field is only locally symmetric in time since, in general, the interaction is only locally periodic. It is convenient to display the local periodicity of the field by shifting the origin of time by t_0, introducing a new time variable $\tilde{t} = t - t_0$, where t_0 is such that over a few cycles $\int^t dt'\,\omega = \omega\tilde{t}$. Thus we can write

$$\begin{aligned} V(\tilde{t}+t_0,\Phi) = {} & V_{L+}(\Phi_L)e^{-i\omega_L\tilde{t}} + V_{L-}(\Phi_L)e^{i\omega_L\tilde{t}} \\ & + V_{H+}(\Phi_H)e^{-i\omega_H\tilde{t}} + V_{H-}(\Phi_H)e^{i\omega_H\tilde{t}}. \end{aligned} \tag{A3}$$

The time-reversed field is obtained by complex conjugating the polarization vectors. Since $\mathcal{T}$ complex conjugates c numbers and anticommutes with $\mathbf{p}$, the interaction of the atom with the time-reversed field is simply $\mathcal{T}^\dagger V(-\tilde{t}+t_0,-\Phi)\mathcal{T}$. We can also complex conjugate the polarization vectors by reversing the sign of $\hat{\mathbf{y}}$. Thus, introducing the operator $\mathcal{I}_y$, which acts only on the atomic coordinates and which effects a reflection in the xz plane, we can write the interaction of the atom with the time-reversed field in the alternative form $\mathcal{I}_y V(\tilde{t}+t_0,\Phi)\mathcal{I}_y^\dagger$. It follows that if we define

$$\mathcal{S} \equiv \mathcal{T}\mathcal{I}_y, \tag{A4}$$

we have

$$\mathcal{S}V(\tilde{t}+t_0,\Phi)\mathcal{S}^\dagger = V(-\tilde{t}+t_0,-\Phi), \tag{A5}$$

which expresses the time symmetry of $V(\tilde{t}+t_0,\Phi)$.

In arriving at this symmetry relation we used

$$\mathcal{S}^\dagger\mathcal{S} = \mathcal{S}\mathcal{S}^\dagger = 1, \tag{A6}$$

which follows from Eq. (A1) and the fact that $\mathcal{I}_y$ is unitary and commutes with $\mathcal{T}$. We note some further properties of $\mathcal{S}$: Since $\mathcal{I}_y^2 = \pm 1$, where [9] the sign correlates with the sign on the right-hand side of Eq. (A2), we have

$$\mathcal{S}^2 = 1, \tag{A7}$$

and hence, from Eq. (A6), we have

$$\mathcal{S}^\dagger = \mathcal{S}. \tag{A8}$$

$\mathcal{S}$ is *anti*linear (actually antiunitary) since $\mathcal{T}$ is.

Letting $\mathcal{S}$ act on both sides of Eq. (14), using Eq. (A5), noting that $dt = d\tilde{t}$ and changing $\tilde{t}$ to $-\tilde{t}$, we find that

$$\int_0^{2\pi/\omega} d\tilde{t}\, \langle \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi) \mid i\hbar \frac{d}{d\tilde{t}} \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle$$

$$= \int_0^{2\pi/\omega} d\tilde{t}\, \langle \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi) \mid [H_a + V(\tilde{t}+t_0,\Phi) - E(\Phi)] \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle \tag{A10}$$

$$= \int_0^{2\pi/\omega} d\tilde{t}\, \langle \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi) \mid [H_a + V(\tilde{t}+t_0,\Phi) - E(-\Phi)] \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle, \tag{A11}$$

where in going from the first to the second step we integrated by parts, noting that, as long as ω is (nearly) constant over one cycle, $\langle \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi) \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle$ is periodic and therefore the surface term vanishes; we also used the Hermiticity [10] of the Hamiltonian $H_a + V(\tilde{t}+t_0,\Phi)$. Comparing the right-hand sides of Eqs. (A10) and (A11) it follows that [7] $E(-\Phi) = E(\Phi)$, as stated in Eq. (23). Using this last result, in conjunction with

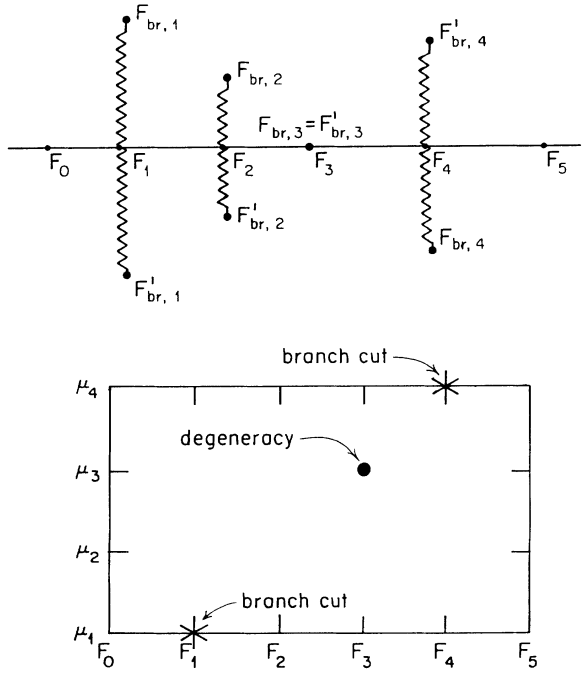

FIG. 5. Upper: branch cuts of the quasienergy in the complex F plane for different values of a parameter μ. The Floquet Hamiltonian is exactly real and symmetric. Lower: possible circuit in the corresponding F-μ plane.

$\mid \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi)\rangle$ is an eigensolution corresponding to eigenvalue $E^*(-\Phi)$ (where here $*$ denotes complex conjugate):

$$i\hbar \frac{d}{d\tilde{t}} \mid \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi)\rangle$$

$$= [H_a + V(\tilde{t}+t_0,\Phi) - E^*(-\Phi)] \mid \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi)\rangle. \tag{A9}$$

We now use Eqs. (14) and (A9) to write

$$i\hbar \frac{d}{d\tilde{t}} \langle \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi) \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle = 0. \tag{A12}$$

Hence the norm $\langle \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi) \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle$ is conserved in time, at least over intervals of a few cycles or so. Consequently, choosing the norm to be unity, and using the harmonic expansion

$$\mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle = \sum_M e^{-iM\omega\tilde{t}} \mid \mathcal{F}_M(\Phi)\rangle, \tag{A13}$$

together with the similar expansion

$$\mid \mathcal{F}^{(*)}(-\tilde{t}+t_0,-\Phi)\rangle = \sum_M e^{-iM\omega\tilde{t}} \mid \mathcal{F}_M^{(*)}(-\Phi)\rangle, \tag{A14}$$

we obtain

$$\sum_N \langle \mathcal{F}_{N-L}^{(*)}(-\Phi) \mid \mathcal{F}_N(\Phi)\rangle = \delta_{L0}. \tag{A15}$$

The scalar product of $\exp[-iE(\Phi)\tilde{t}/\hbar] \mid \mathcal{F}(\tilde{t},\Phi)\rangle$ with itself, while it may only formally exist, represents the probability for finding the electron inside some finite volume $\mathcal{V}$, and this decays in time, as $\exp[2\,\mathrm{Im}E(\Phi)\tilde{t}/\hbar]$, due to the flux passing out of $\mathcal{V}$ as the atom ionizes. On the other hand, the scalar product of $\exp[-iE(\Phi)\tilde{t}/\hbar] \mid \mathcal{F}(\tilde{t}+t_0,\Phi)\rangle$ with $\mathcal{S}\exp[iE(-\Phi)\tilde{t}/\hbar] \mid \mathcal{F}(-\tilde{t}+t_0,-\Phi)\rangle$ is preserved in time because the flux passing out of $\mathcal{V}$ is compensated for by the flux passing into $\mathcal{V}$ in the time-reversed state.

APPENDIX B: BRANCH POINTS IN A COMPLEX PARAMETER PLANE

Let us take, as an example, two parameters, with one of these parameters the field strength F of one of the two components of the bichromatic light field. The other

parameter, μ say, may be either the field strength of the other component, or the relative phase of the two components. To understand the behavior of an eigenvector as these parameters are varied it is useful to briefly comment on the analytic structure of $E(\Phi)$. This appendix is complementary to Sec. III C, where we discussed the behavior near a degeneracy in the eigenvalue, for a two-parameter space.

The quasienergy $E(\Phi)$ has branch points in the complex F plane corresponding either to a resonance or to a multiphoton ionization threshold [13]. Here we consider the passage past a resonance. At a (resonance) branch point, the complex energy eigenvalues for two different levels coincide; normally this can only occur for a complex value of F since both the real and imaginary parts of the two eigenvalues must coincide, and two parameters (the real and imaginary parts of F) must be adjusted to arrange for the levels to intersect. However, since we have another parameter μ at our disposal, we can arrange for the branch point to occur on the real F axis. Note that there is always a second branch point associated with a resonance, which, as long as the decay width is very small, is close to the conjugate of the first branch point. To see this, suppose that both fields are sufficiently weak near the resonance that the decay width is negligible, so that $E(\Phi)$ is very nearly real when F is real. It follows from the Schwarz reflection principle that if there is a branch point at $F_{\rm br}$, there is also one at $F_{\rm br}^*$, at least if the decay width is small. Even if the decay width is not small, these two "conjugate" branch points must exist even though they may not be complex conjugates of each other [15].

We now study what happens as we follow a rectangular circuit in the real F-μ plane, starting from the corner (F_0, μ_1), moving to the corner (F_5, μ_1), then to (F_5, μ_4), and finally back to (F_0, μ_1), where the subscripts on F and μ indicate different values (see the lower parts of Figs. 5–7). Suppose that at the values μ_i, $i = 1 - 4$, of μ there are resonance branch points at $F_{{\rm br},i}$ and $F'_{{\rm br},i}$, where $F_{{\rm br},i}$ and $F'_{{\rm br},i}$ are conjugate branch points. We draw a branch cut between each pair of conjugate branch points, and if the ith cut intersects the real F axis we denote the point of intersection by F_i. At the point F_i there is an avoided crossing of the real parts of the two quasienergy eigenvalues involved in the resonance, and when the branch cut is crossed, the character of one of the states changes to that of the other state, and vice versa.

In Fig. 5 we show a possible behavior in the case where the decay width is negligible and the "conjugate" branch points are truly conjugates. As μ (say the strength of the "other" component of the field) varies, from μ_1 to μ_4, the branch points vary, and one of them may cross the real F axis. This happens in Fig. 5, when $\mu = \mu_3$; since the branch points are exact conjugates, they both merge into a single point on the real F axis, at F_3. Therefore the point (F_3, μ_3) inside the rectangle is a point of degeneracy, but not a branch point. As we follow the rectangular path in the real F-μ plane, we first cross the branch cut at F_1, and subsequently cross the branch cut at F_4; since

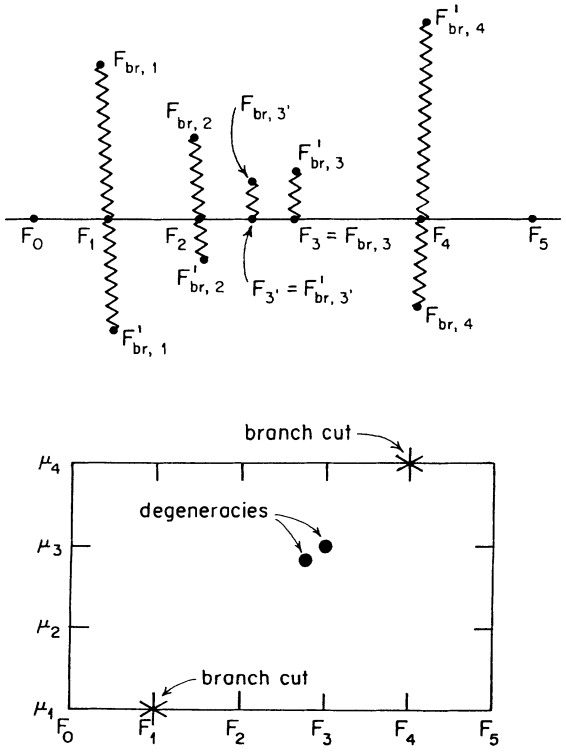

FIG. 6. Upper: branch cuts of the quasienergy in the complex F plane for different values of a parameter μ. The Floquet Hamiltonian is roughly real and symmetric. Lower: possible circuit in the corresponding F-μ plane.

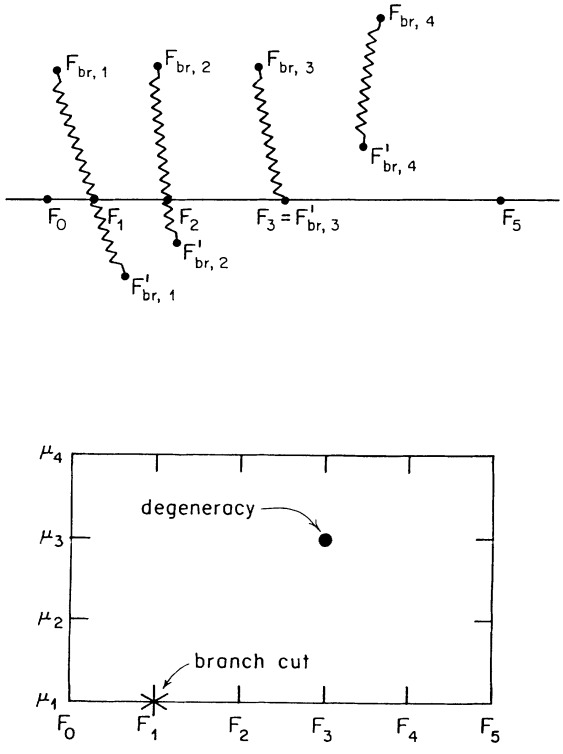

FIG. 7. Upper: branch cuts of the quasienergy in the complex field plane for different values of a parameter μ. The Floquet Hamiltonian is complex and nonHermitian. Lower: possible circuit in the corresponding F-μ plane.

we cross a branch cut twice, the eigenray is mapped into itself upon one complete circuit. When the decay width is small but not negligible, the two conjugate branch points do not merge into a single point on the real F axis, for a certain value of μ; rather, there are two nearby branch points on the real F axis, at F_3 and F_3' say, which occur for nearby values of μ, say μ_3 and μ_3', as shown in Fig. 6. For all practical purposes, unless the circuit passes between the points (F_3, μ_3) and (F_3', μ_3'), the two branch points can be viewed as a single point of degeneracy [16].

In Fig. 7 we show a possible behavior in the case where the decay width is significant, so that the conjugate branch points are far from being conjugates. Again, as μ (say the phase Φ_H of the high-frequency component) varies, a branch point may cross the real F axis. This happens in Fig. 7, when $\mu = \mu_3$; only one branch point sits on the real F axis, at F_3. Thus the point (F_3, μ_3) is not only a point of degeneracy, it is also a branch point. As we follow the rectangular path in the real F-μ plane, we cross the branch cut at F_1, corresponding to an avoided crossing, but upon returning, with $\mu = \mu_4$, there is no branch cut to cross — the real parts of the eigenvalues undergo a true crossing. Hence, after one complete circuit, the original eigenray is mapped into another eigenray, the one corresponding to the other level with which the original level is resonant. After a second complete circuit, the branch cut is crossed again, at F_1, and the original eigenray is mapped into itself. This is the situation realized in case (a) of Fig. 1, where only one degeneracy is enclosed by the circuit. In cases (b) and (c) of Fig. 1, respectively, two and zero branch cuts in the F plane are crossed at different places on the circuit. However, in the case of Fig. 1(b) we do not know whether the branch cut in the F plane moves over the real axis as Φ_H varies, as in Fig. 6, so that there may be zero, not two, degeneracies enclosed by the circuit. The same is true in the case of Fig. 1(c).

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[2] *Geometric Phases in Physics*, edited by A. Shapere and F. Wilczek, Advanced Series in Mathematical Physics Vol. 5 (World Scientific, Singapore, 1989).

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[4] The occurrence of a complex geometric phase in the case of a two-level dissipative system driven by an intense periodic field has also been described by S.-I Chu, Z.-C. Wu, and E. Layton, Chem. Phys. Lett. **157**, 151 (1989), and by D.J. Moore and G.E. Stedman, Phys. Rev. A **45**, 513 (1992); see the latter paper for additional references to earlier works.

[5] Note, incidentally, that each harmonic component is multiplied by a different dynamical phase factor $\exp[-\frac{i}{\hbar}\int^t dt'\,(E + N\hbar\omega)]$.

[6] For experimental confirmation of this, see, e.g., C. Chen, Y-Y Yin, and D.S. Elliott, Phys. Rev. Lett. **64**, 507 (1990); H. Muller, P. H. Bucksbaum, D. W. Shuhmacher, and A. Zavriyev, J. Phys. B **24**, L641 (1991); and S.M. Park, S.-P. Lu, and R.J. Gordon, J. Chem. Phys. **94**, 8622 (1991).

[7] R. M. Potvliege and P. H. G. Smith, J. Phys. B **24**, L641 (1991); J. Phys. B (to be published).

[8] See, e.g., T. S. Ho, S.-I. Chu, and J. V. Tietz, Chem. Phys. Lett. **96**, 464 (1983); and M. Dörr, R. M. Potvliege, D. Proulx, and R. Shakeshaft, Phys. Rev. A **44**, 574 (1991).

[9] A. Messiah, *Quantum Mechanics* (Wiley, New York, 1962), Vol. 2.

[10] In applying Green's theorem to the analytically continued volume integral, the surface integral vanishes, and so the Hamiltonian is Hermitian.

[11] At this point it may be instructive to verify that Eq. (2) is in accord with what is obtained by calculating B_Φ from Eq. (38), now keeping the frequency fixed. To this aim, first note that in the case of a monochromatic field the Φ dependence of $|\,\bar{\mathcal{F}}_N(\Phi)\rangle$ can be written explicitly as $e^{-iN\Phi}\,|\,\bar{\mathcal{F}}_N\rangle$, hence Eq. (38) yields

$$B_\Phi = i\oint d\Phi \frac{\langle\langle \bar{\mathcal{F}}^{(*)}(-\Phi)\,|\,\partial/\partial\Phi\,|\,\bar{\mathcal{F}}(\Phi)\rangle\rangle}{\langle\langle \bar{\mathcal{F}}^{(*)}(-\Phi)\,|\,\bar{\mathcal{F}}(\Phi)\rangle\rangle} = \oint d\Phi \frac{\sum_N N\langle \bar{\mathcal{F}}_N^{(*)}\,|\,\bar{\mathcal{F}}_N\rangle}{\sum_N \langle \bar{\mathcal{F}}_N^{(*)}\,|\,\bar{\mathcal{F}}_N\rangle}.$$

This can be cast with Eq. (17) (for the case of a monochromatic field) in the alternative form (where $|\,\bar{\mathcal{F}}\rangle\rangle$ denotes the vector with components $|\,\bar{\mathcal{F}}_N\rangle$)

$$B_\Phi = -\oint d\Phi \frac{\langle\langle \bar{\mathcal{F}}^{(*)}\,|\,\partial H/\partial\omega\,|\,\bar{\mathcal{F}}\rangle\rangle}{\langle\langle \bar{\mathcal{F}}^{(*)}\,|\,\bar{\mathcal{F}}\rangle\rangle}.$$

Equation (2) then follows immediately from the Feynman-Hellmann theorem.

[12] We can, of course, take into account any variation in Φ through a variation in ω, as explained in the Introduction.

[13] M. Pont and R. Shakeshaft, Phys. Rev. A **43**, 3764 (1991).

[14] Garrison and Wright [3] found a complex geometric phase in the monochromatic case, when the "circuit" is simply the line followed by increasing the phase of the field from 0 to 2π at constant intensity; we obtained similar results in the two-color case. For example, $R = -0.57 - 0.62i$ for $I_L = 1.3 \times 10^{13}$ W/cm^2 and $I_H = 0.04 I_L$ and for Φ_H varying from 0 to 2π.

[15] Actually, there are *four* branch points associated with a resonance. To see this, note first that the quasienergy must be independent of a shift in the origin of time. Now, if we turn off the low-frequency field, and shift the origin of time by either π/ω or $\pi/(2\omega)$, depending on whether p is even or odd, respectively, the effect on the interaction is the same as a change of F to $-F$. Hence if there is a branch point at $F_{\rm br}$, there is also one at $-F_{\rm br}$, at least in the limit of vanishing low-frequency field; these two "inverse" branch points must survive the turning on of the low-frequency field even though they may not remain inverses of each other.

[16] In the complex F plane the two branch cuts are so close together that the second cut is crossed almost immediately after the first, and there is only a momentary change in the character of a state.