

Ionization of atoms by a low-frequency field and an optical-frequency field

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A new formalism generalizes the quasienergy method to the case of two monochromatic fields with different frequencies. Under certain conditions, ionization occurs by a mechanism in which an electron initially absorbs a fixed number of optical phonons and then undergoes a tunneling due to infrared light. Rigorous calculations are carried out in a model of a short-range potential. The possibility of observing this effect in atoms is discussed. Oscillations should appear in the frequency dependence of the photoelectric effect upon the imposition of an intense low-frequency field.

1. INTRODUCTION

The nonlinear ionization of atoms by laser beams has been studied thoroughly only in the many-photon limit, where the so-called adiabatic parameter¹ is much greater than unity:

$$\gamma = \omega (2Im)^{1/2} / ef \gg 1. \quad (1)$$

Here f and ω are the wave amplitude and frequency, and I is the binding energy of the atomic level of interest. In the region $\gamma \lesssim 1$ the ionization cannot be described as a process consisting of absorption of a fixed number of photons. In the limit $\gamma \ll 1$, the ionization is determined by the tunneling of an electron through an oscillating potential barrier; extremely little information is available on this case. Nevertheless, a study of ionization at small values of γ is extremely worthwhile both for determining the general behavior of the ionization in a low-frequency field, $\omega \ll I/\hbar$, and for studying the breakdown of a gas in an intense infrared field (see Ref. 2, for example). Although experimental work in this field has been carried out for a long time now (see Refs. 3–5, for example), it was only a recent study⁶ that established the very fact that ionization of an atom by an infrared field could be observed experimentally.

We see from (1) that by reducing γ (at fixed values of f and ω) we can also attain a decrease of the binding energy I . One possibility here is to use a target of excited atoms, which might be excited, for example, through resonant cascade filling of highly excited states.

In this paper we discuss another possibility: an ionization caused by two fields—an optical field (of frequency Ω) and an infrared field (of frequency ω)—which does not require a special frequency Ω for the actual filling of excited levels. The only necessary condition is that the energy of one or several optical phonons be near the boundary of the continuous spectrum at the atom. In this case, an electron which absorbs N optical phonons goes into a virtual state with an energy $|E_N| = I - N\hbar\omega \ll I$, from which tunneling ionization occurs in an infrared field with an effective adiabatic parameter

$$\gamma_N = \omega (2|E_N| m)^{1/2} / ef,$$

which satisfies $\gamma_N \ll \gamma$. The possibility of implementing a similar mechanism has been discussed previously⁷ for the case of a static field; we demonstrate the possibility of this mechanism in an infrared field in Sec. 3 below for the parti-

cular case of a model problem which can be solved exactly. In Sec. 4 we discuss the manifestation of this mechanism in the ionization of atoms and the implementation of tunneling ionization in fields of two frequencies.

2. EXTENSION OF THE QUASIENERGY APPROACH TO THE CASE OF A QUANTUM-MECHANICAL SYSTEM IN FIELDS OF TWO FREQUENCIES

We begin with a rather general discussion of the behavior of a quantum-mechanical system in two monochromatic fields of frequencies Ω and ω . We write the operator representing the interaction with these fields in the customary form (in the dipole approximation):

$$V(\mathbf{r}, t) = -\mathbf{d} \operatorname{Re} \{ f e^{-i\omega t} + F e^{-i\Omega t} \}, \quad (2)$$

where \mathbf{d} is the dipole moment of the system, and f and F are the complex field amplitudes.

In a field of a single frequency ω it is extremely convenient to use the quasienergy solutions of the Schrödinger equation

$$i\hbar \frac{\partial \psi(\mathbf{r}, t)}{\partial t} = [H_0 + V_\omega(\mathbf{r}, t)] \psi(\mathbf{r}, t); \quad (3)$$

these solutions are

$$\Phi_E(\mathbf{r}, t) = e^{-iEt/\hbar} \Phi_E(\mathbf{r}, t), \quad \Phi \left(t + \frac{2\pi}{\omega} \right) = \Phi(t),$$

$$\left[H_0 + V_\omega(\mathbf{r}, t) - i\hbar \frac{\partial}{\partial t} \right] \Phi_E(\mathbf{r}, t) = E \Phi_E(\mathbf{r}, t).$$

The reason for taking this approach is that the Φ_E form a natural orthonormal basis, and the quasienergies E determine the spectral characteristics of the quantum system in the external field.⁸ The same approach is effective in problems involving the decay of a system caused by an external field. In this case, the radiation condition at infinity is imposed on the functions Φ_E , so that the quasienergies are complex, and $\operatorname{Re} E$ and $\operatorname{Im} E$ determine the positions and widths (decay probabilities) of quasistationary states.⁹

We will show that in a field with two frequencies there are also states which are analogous to quasienergy states. To construct these solutions we consider the auxiliary eigenvalue problem

$$\left[H_0 + V(\mathbf{r}, t_1, t_2) - i\hbar \frac{\partial}{\partial t_1} - i\hbar \frac{\partial}{\partial t_2} \right] \Phi(\mathbf{r}, t_1, t_2) = E \Phi(\mathbf{r}, t_1, t_2), \quad (4)$$

$$V(\mathbf{r}, t_1, t_2) = -\mathbf{d} \operatorname{Re} \{ f e^{-i\omega t_1} + F e^{-i\Omega t_2} \},$$

where Φ satisfies the periodicity conditions

$$\Phi(t_1 + 2\pi k/\omega; t_2 + 2\pi n/\Omega) = \Phi(t_1, t_2),$$

and k and n are integers. It is not difficult to see that the functions

$$\Psi_E(\mathbf{r}, t) = e^{-iEt/\hbar} \Phi(\mathbf{r}, t_1=t, t_2=t)$$

satisfy Eq. (3) with perturbation operator (2). The function $\Phi(\mathbf{r}, t, t)$ does not satisfy any definite periodic conditions, as it does in the case of a single frequency; it is nevertheless a particular solution which describes some steady "stationary" state of the system in fields of two frequencies. From the mathematical standpoint, the function $\Phi(\mathbf{r}, t, t)$ considered as a function of the time, falls in the class of nearly periodic functions,¹⁰ which have some important applications in mathematical physics.

By analogy with the single-frequency case we can show that the Φ_E with different values of E (for brevity we will again call the quantities E "quasienergies") are orthogonal and can thus be used as a basis for analyzing the properties of the system in two fields. Working from the "stationary" equation (4), we can construct several general theorems for the "quasienergy" states Φ_E , and we can construct a systematic perturbation theory in f and F to calculate Φ_E and E in an approach analogous to that taken previously for a single frequency.^{11,12} In sufficiently weak fields the spectral frequencies of the system are determined by the quasienergies E_i :

$$\omega_{if, n, k} = (E_j - E_i)/\hbar + n\omega + k\Omega.$$

The solutions of Eq. (4) with radiation conditions on \mathbf{r} and with complex E give us quasistationary states which describe the decay of the system in two fields. For these solutions we can also construct a perturbation theory in complete analogy with the single-frequency case.¹³ In the absence of resonances, Φ_E and E can be written as power series in f and F ; in particular, if the perturbation theory uses only one of the fields (F , say), then in a nonresonant case Φ_E and E have the structures

$$\Phi_E(\mathbf{r}, t, t) = \Phi^{(0)}(\mathbf{r}, t) + \sum_{k=x, y, z} [\Phi_k^{(1)}(\mathbf{r}, t) F_k e^{-i\Omega t} + \Phi_k^{(-1)}(\mathbf{r}, t) F_k^* e^{i\Omega t}] + \dots, \quad (5a)$$

$$E = E^{(0)} + \sum_{k, m=x, y, z} \chi_{k, m} F_k F_m^* + \dots, \quad (5b)$$

where $\Phi^{(0)}$ and $E^{(0)}$ are quasienergy solutions in the field f , and the $\Phi^{(1)}(\mathbf{r}, t)$ are periodic in t with a period $2\pi/\omega$.

It is clear that a corresponding procedure could in principle be followed for an arbitrary number of external fields, and the result would be qualitatively the same. In practice, however, as the number of external fields becomes large the final expressions become too complicated for practical use.

3. DECAY OF A WEAKLY BOUND STATE IN INFRARED AND OPTICAL FIELDS

The probability for an electromagnetic field to cause a decay of the system can be calculated exactly, without resorting to perturbation theory, only for the extremely simple

model of a particle in a δ -function potential, in which case the unperturbed Hamiltonian H_0 has only a single bound state, with a binding energy I . For this model and in the quasistationary approach, the complex quasienergy is determined by the one-dimensional integral equation (cf. Ref. 7; we are using atomic units, with $e = m = \hbar = 1$)

$$[(2E)^{1/2} - (2I)^{1/2}] \varphi(t) = \frac{1}{(2\pi i)^{1/2}} \int_0^\infty \frac{d\tau}{\tau^{1/2}} e^{iE\tau} [e^{iS(t, t-\tau)} \varphi(t-\tau) - \varphi(t)] \quad (6)$$

for the function $\varphi(t)$, which is determined by the behavior of $\Phi_E(\mathbf{r}, t, t)$ at the origin of coordinates:

$$\varphi(t) = \lim_{\mathbf{r} \rightarrow 0} \mathbf{r} \Phi_E(\mathbf{r}, t, t).$$

Here $S(t, t') = S_{cl}(\mathbf{r} = 0; t; \mathbf{r}' = 0, t')$, where S_{cl} is the classical action for a free particle in an electromagnetic field. Corresponding to interaction (2) we have

$$S(t, t') = \frac{[\alpha(t) - \alpha(t')]^2}{t - t'} - \frac{1}{2} \int_{t'}^t \left[\text{Im} \left(\frac{\mathbf{f}}{\omega} e^{-i\omega t''} + \frac{\mathbf{F}}{\Omega} e^{-i\Omega t''} \right) \right]^2 dt'',$$

$$\alpha(t) = \text{Re} \left(\frac{\mathbf{f}}{\omega^2} e^{i\omega t} + \frac{\mathbf{F}}{\Omega^2} e^{-i\Omega t} \right).$$

To analyze the mechanism for the decay of the system we must examine the case

$$f, F \ll F_0 = (2I)^{1/2}, \quad \omega \ll I, \quad \Omega \sim I, \quad (7)$$

in which the optical field F can be taken into account by perturbation theory; in this case, we seek the solution $\varphi(t)$, E of Eq. (6) in the form of expansion (5). For simplicity we will take F into account in only the first nonvanishing order (the system absorbs only a single optical phonon), and we will replace E by $-I$ on the right side of (6). Now expanding e^{iS} in (6) in terms of F ,

$$e^{iS(t, t-\tau)} = e^{(0)}(t, \tau) + \sum_{k=x, y, z} [e_k^{(1)}(t, \tau) F_k e^{-i\Omega t} + e_k^{(-1)}(t, \tau) E_k^* e^{i\Omega t}] + \sum_{k, m=x, y, z} e_{k, m}^{(1, -1)}(t, \tau) F_k F_m^* + \dots,$$

we find equations for successively determining $\varphi^{(0, \pm 1)}$ and χ . Using the identity

$$(-2E_1)^{1/2} - (-2E_2)^{1/2} = \frac{1}{2\pi i} \int_0^\infty \frac{d\tau}{\tau^{1/2}} [e^{iE_2\tau} - e^{iE_1\tau}],$$

we can write these equations as

$$[(-2E^{(0)})^{1/2} - (2I)^{1/2}] \varphi^{(0)}(t) = \frac{1}{(2\pi i)^{1/2}} \int_0^\infty \frac{d\tau}{\tau^{1/2}} e^{-iE\tau} \times [e^{(0)}(t, \tau) \varphi^{(0)}(t-\tau) - \varphi^{(0)}(t)], \quad (8a)$$

$$[(-2(E^{(0)} \pm \Omega))^{1/2} - (2I)^{1/2}] \varphi_k^{(\pm 1)}(t) = \frac{1}{2\pi i} \int_0^\infty \frac{d\tau}{\tau^{1/2}} e^{-i(t \pm \Omega)\tau} [e^{(0)}(t, \tau) \varphi_k^{(\pm 1)}(t-\tau) - \varphi_k^{(\pm 1)}(t) + e_k^{(\pm 1)}(t, \tau) \varphi^{(0)}(t-\tau)]. \quad (8b)$$

Equation (8a) determines the behavior of the system under the influence of the infrared field alone. Under conditions (7), the right sides of (8) contain small factors (so that the difference $|E^{(0)} - I|$ is proportional to f^2); furthermore, $\varphi^{(0)}$ depends only weakly on the time.⁷ With an accuracy appropriate to the present calculations we can set $\varphi^{(0)}(t) = \text{const}$ and discard terms $\sim \varphi^{(\pm 1)}$ from the right side of (8b) (the difference between such terms also gives us a small quantity $\sim f^2$). As a result we find

$$\varphi^{(0)}(t) = 1,$$

$$\varphi_k^{(\pm 1)}(t) = \frac{1}{(2(I \pm \Omega))^{1/2} - (2I)^{1/2}} \times \frac{1}{(2\pi i)^{1/2}} \int_0^\infty \frac{d\tau}{\tau^{3/2}} e^{-i(I \pm \Omega)\tau} e_k^{(\pm 1)}(t, \tau).$$

Calculating the terms of second order in F , and taking an average over the period of the infrared field, we find the following expression for χ_{km} in (5b):

$$\chi_{km} = -(2I)^{1/2} \frac{\omega}{2\pi} \int_0^\infty dt \int_0^\infty \frac{d\tau}{\tau^{3/2}} e^{-iI\tau} [e_k^{(1)}(t, \tau) \varphi_m^{(-1)}(t-\tau) + e_m^{(-1)}(t, \tau) \varphi_k^{(1)}(t-\tau) + e_{k,m}^{(1,-1)}(t, \tau)]. \quad (9)$$

This expression holds under conditions (7).

The expression for $e^{(\pm 1)}$ and $e^{(1,-1)}$ for an arbitrary relative orientation of the vectors \mathbf{f} and \mathbf{F} are quite cumbersome, so to analyze the result we consider the simplest case, in which the infrared radiation is linearly polarized and the vectors \mathbf{f} and \mathbf{F} are orthogonal. In this case we can write

$$e_k^{(\pm 1)} = 0, \\ e_{k,m}^{(1,-1)} = \delta_{k,m} \frac{i}{\Omega^4 \tau} \left(\sin^2 \frac{\Omega \tau}{2} - \frac{\tau^2 \Omega^2}{4} \right) \times \exp \left\{ \frac{if^2}{\omega^4 \tau} \left[\sin^2 \frac{\omega \tau}{2} (1 - \cos \omega(\tau - 2t)) + \frac{1}{4} \omega \tau \sin \omega \tau \cos \omega(\tau - 2t) - \frac{\tau^2 \omega^2}{4} \right] \right\},$$

and for $\chi_{k,m} = \chi_{\delta k,m}$ we find

$$\chi = \frac{I^{1/2}}{2(i\pi)^{1/2} \Omega^4} \int_{-\pi}^\pi dt \int_0^\infty \frac{d\tau}{\tau^{3/2}} \left(\frac{1}{\tau} \sin \frac{\Omega \tau}{2} - \frac{1}{4} \tau \Omega^2 \right) \times \exp \left\{ -i \left(I + \frac{f^2}{4\omega^2} \right) \tau + \frac{if^2}{\omega^4 \tau} \times \left[\sin^2 \frac{\omega \tau}{2} (1 - \cos t) + \frac{1}{4} \omega \tau \sin \omega \tau \cos \omega t \right] \right\}. \quad (10)$$

The integrals in (10) can be evaluated easily in the case $f = 0$. The dynamic Stark effect of a weakly bound particle in an optical field (cf. Ref. 14) is given by

$$\chi(f=0) = \chi^{(0)} = \frac{1}{4\Omega^2} + \frac{I^{1/2}}{3\Omega^4} [2I^{3/2} - (I+\Omega)^{3/2} - (I-\Omega)^{3/2}]. \quad (11)$$

If $\Omega > I$, the imaginary part of $\chi^{(0)}$ is nonzero and gives the

familiar expression for the probability of the photoelectric effect in the model of a δ -function potential:

$$W = -2|\mathbf{F}|^2 \text{Im} \chi^{(0)} = \frac{|\mathbf{F}|^2}{2\Omega^4} (\Omega - I)^{3/2}. \quad (12)$$

In the case $f \neq 0$ it is convenient to evaluate the integral over τ by deforming the integration contour in the complex plane. The integral from 0 to τ_0 is real if $\Omega < I$ and does not give rise to a tunneling via quasienergy states. In the case $|\tau_0| \ll 0$, this integral gives us $\chi^{(0)}$ and corrections to $\chi^{(0)}$ of the form

$$a(\omega, \Omega) f^2 + b(\omega, \Omega) f^4 + \dots$$

There is no difficulty in evaluating the coefficients a, b, \dots , but we will not go through these calculations since we have already discarded terms of the same order of magnitude in deriving (9). We might note that the exact expressions for the terms $\sim f^2 F^2$, etc., in E (the nonlinear susceptibilities in fields of two frequencies) can be found by solving (4) by perturbation theory or by using an expansion in both fields, f and F , when solving Eq. (6) (the latter approach is technically simpler in the case of a δ -function potential; cf. Ref. 7).

The integral over the remainder of the contour is evaluated by the method of steepest descent (as is the integral over t). We should first write the expression with $\sin^2(\Omega\tau/2)$ in the form

$$\frac{1}{\tau} \sin^2 \frac{\Omega \tau}{2} - \frac{1}{4} \tau \Omega^2 = \sum_{l=0, \pm 1} C_l(\tau) e^{il\Omega\tau}. \quad (13)$$

The saddle point for each of the terms in (13) is determined by

$$t^{(l)} = 0, \quad \tau_0^{(l)} = -\frac{2i}{\omega} \text{Arsh} \gamma_l, \quad \gamma_l = \frac{\omega}{f} [2(I - l\Omega)]^{1/2}.$$

If $\Omega < I$, the result of the integration will be purely imaginary and will determine that width of the quasistationary level which is caused by the joint effects of the optical and infrared fields. The value of χ is dominated by the term in (13) with $l = 1$, for which γ_l is at a minimum. The expression for the ionization probability contains a characteristic tunneling exponential function, which was first derived by Keldysh¹:

$$W \sim F^2 \exp \left[-\frac{2(I - \Omega)}{\omega} \right] \times \left[\left(1 + \frac{1}{2\gamma_1^2} \right) \text{Arsh} \gamma_1 - \frac{(1 + \gamma_1^2)^{1/2}}{2\gamma_1} \right], \quad (14)$$

with the effective tunneling parameter

$$\gamma_1 = \gamma_0 (1 - \Omega/I)^{1/2}. \quad (15)$$

Expressions (14) and (15) justify interpreting the mechanism for the decay of the system in optical and infrared fields as tunneling induced by the infrared field from a virtual state (a harmonic of a quasienergy state) having an energy $E = -I + \Omega$ and populated with a weight $\sim F^2$. We will not reproduce here the extremely complicated coefficient of the exponential function in (14) (cf. Ref. 15). The expression for this coefficient simplifies in the limit $\omega \rightarrow 0$, in which case W becomes

$$W = F^2 \left(\frac{3If^5}{4\pi \cdot 2^{1/2} (I-\Omega)^{1/2}} \right)^{1/2} \exp \left\{ -\frac{2}{3} \frac{[2(I-\Omega)]^{3/2}}{f} \right. \\ \left. \times \left[1 - \frac{\omega^2 (I-\Omega)}{5f^2} + \dots \right] \right\}$$

and agrees in the case $\omega = 0$ with the result for a static field, averaged over the period of the infrared field.

We see that the "interference" of the two fields is most pronounced when the frequency Ω is near the ionization threshold,

$$I - \Omega \ll I, \quad (16)$$

so that the condition $\gamma_1 \ll \gamma_0$ holds. When the frequency Ω is small, and condition (16) does not hold, an analogous effect arises when the field F is taken into account in higher-order perturbation theory if

$$I - N\Omega \ll I.$$

In this case the population of the N th harmonic and also W are proportional to F^{2N} , and the tunneling exponential function contains a parameter

$$\gamma_N = \gamma_0 (1 - N\Omega/I)^{1/2}.$$

The condition for the applicability of the method of steepest descent imposes a condition on ω which is more stringent than that in (7), specifically,

$$\omega \ll I - N\Omega.$$

At $\Omega > I$ the parameter γ_1 becomes purely imaginary, and expression (14) and thus the imaginary part of the integral in (10) are oscillatory functions of f and Ω . The ionization probability in this case will thus contain a small oscillating correction in addition to the monotonic term in (12). This effect is analogous to the oscillations in the cross section for the photoelectric effect upon the imposition of a static electric field; these oscillations have been observed experimentally^{16,17} and have been studied in detail theoretically.^{18,19} It follows from this discussion that the imposition of an infrared field can also cause a substantial modification of the frequency dependence of the probability for the photoelectric effect.

4. TUNNELING IONIZATION OF ATOMS IN FIELDS OF TWO FREQUENCIES

Calculations dealing with the ionization of atoms by a monochromatic field under the condition $\gamma \lesssim 1$ are complicated considerably by the presence of the long-range Coulomb tail on the atomic potential. A semiclassical analysis of this problem²⁰ shows that the ionization probability in a low-frequency field is determined by the asymptotic form of the unperturbed wave function at large distances from the nucleus, as in the case of ionization by a static field. The exponential factor in W has the same form as for a short-range potential, but so far no correct calculations have been carried out for the coefficient of the exponential function for complex atoms, in contrast with the case of the static field.

The qualitative aspects of the ionization process in the presence of an additional field in the optical frequency range can be seen most simply in the limit $\omega = 0$. In this case, weak optical radiation sends the unperturbed state

$\psi_0(\mathbf{r}, t) = \Phi_0 \mathbf{r} e^{iHt}$ to the state $\psi(\mathbf{r}, t)$. In first-order perturbation theory we have

$$\psi(\mathbf{r}, t) = \Phi_0(\mathbf{r}) e^{iHt} + \Phi_1(\mathbf{r}) e^{i(I-\Omega)t} + \Phi_{-1}(\mathbf{r}) e^{i(I+\Omega)t}, \quad (17) \\ \Phi_{\pm 1} = -1/2 G_{-I \pm \Omega}(\mathbf{dF}) |\Phi_0\rangle,$$

where G_E is the Green's function of the atomic electron. We assume $\Omega < I$, so that the ordinary photoelectric effect cannot occur, while the probability for many-photon ionization is negligibly small because F is small. The imposition of a static field f on the atom in the state $\psi(\mathbf{r}, t)$ gives rise to a tunneling, whose probability is determined by the asymptotic form of ψ at large r . Under the condition $I - \Omega \ll I$, the tunneling from the harmonic Φ_1 , for which the binding energy is lowest, is obviously the most effective. In the general case in which the energy of the N th harmonic is near the threshold ($0 < I - N\Omega \ll I$) the decay probability is determined by tunneling from the state

$$\Phi_N = (-1/2)^N G_{N\Omega-I}(\mathbf{dF}) \dots G_{\Omega-I}(\mathbf{dF}) |\Phi_0\rangle. \quad (18)$$

Calculating the asymptotic behavior

$$\Phi_N(\mathbf{r}) \xrightarrow{r \rightarrow \infty} \sum_{LM} A_{LM}^{(N)} Y_{LM} \left(\frac{\mathbf{r}}{r} \right) r^{z\nu-1} e^{-r/\nu}, \quad (19)$$

where $\nu = [2(I - N\Omega)]^{-1/2}$, $A_{LM}^{(N)} \sim F^N$, and z is the charge of the atomic core, and using a method like that of Ref. 21 to join this asymptotic behavior to the semiclassical wave function of the electron at $r \gg \nu$, we find the following expression for the tunneling probability²²:

$$W = \frac{1}{2} \exp \left(-\frac{2}{3f\nu^3} \right) \sum_M \frac{(\nu/2)^{|M|}}{|M|!} \left(\frac{f\nu^2}{2} \right)^{|M|+1-2z\nu} \\ \times \left| \sum_L A_{LM}^{(N)} \left(\frac{(2L+1)(L+|M|)!}{(L-|M|)!} \right)^{1/2} \right|^2 \quad (20)$$

Since Φ_N is a superposition of states with different values of L , we see that W contains terms corresponding to an interference of the amplitudes $A_{LM}^{(N)}$. The quantum numbers L and M in (20) are determined by the selection rules in matrix element (18) and depend on the polarization of the field F and the relative orientation of the vectors \mathbf{F} and \mathbf{f} . In particular, in the case of a circular polarization of F we would have $L = M = N$, and W would contain a small factor $\sim (f\nu^2)^N$ not present in the case of a linear polarization of F .

It is clear from the physical standpoint that again in the case $\omega \neq 0$, $\omega \ll \Omega$, the tunneling induced by the infrared field will almost certainly proceed from the harmonic Φ_N and will be determined by the same coefficients A_{LM} as in (19). As we mentioned earlier, the exponential factor in W has the same form as for a weakly bound level with a binding energy $I - N\Omega$, so we can write

$$W = CF^{2N} \exp \left\{ -\frac{2(I - N\Omega)}{\omega} \right. \\ \left. \times \left[\left(1 + \frac{1}{2\gamma_N^2} \text{Arsh } \gamma_N - \frac{(1 + \gamma_N^2)^{1/2}}{\gamma_N} \right) \right] \right\}. \quad (21)$$

Although again in this case we are not able to derive an exact expression for the coefficient C of the exponential, we can

make some general comments about the dependence of C on the frequency Ω . The strongest frequency dependence $C(\Omega)$ is determined by the frequency dependence of the coefficients $A_{LM}^{(N)}$ in (19), which are expressed as combinations of composite matrix elements of the N th-order perturbation theory. At energies $E_N = -I + N\Omega$ near the ionization threshold we can use the well-known expression for the Green's function in the approximation of the quantum-defect method¹⁴ for the Green's function G_{E_N} in (18). For the case $\nu \gg 1$ in which we are interested here, the dependence of $A_{LM}^{(N)}$ on ν can then be written

$$A_{LM}^{(N)} \sim \frac{(2e)^{2\nu}}{\nu^{2\nu-1/2}} \frac{1}{\sin \pi(\nu + \mu_L)}. \quad (22)$$

Here μ_L is the quantum defect of states with angular momentum L . Those values of ν for which the sine in (22) vanishes correspond to N -photon resonances involving real atomic levels. Near a resonance the sum $\nu + \mu_L$ is approximately equal to an integer n , the principal quantum number of the state with the energy $E_{nL} = -1/2(n - \mu_L)^{-2}$. Carrying out an expansion of the sine in (22) for this case, we find

$$|A_{LM}^{(N)}|^2 \sim \frac{(2e)^{2\nu} \nu^{-(4\nu+5)}}{(N\Omega - I - E_{nL})^2}. \quad (23)$$

In the immediate vicinity of the resonance we should add a term $\Gamma_{nL}^2/4$ to the denominator of (23), where Γ_{nL} is the width of the $|n, L\rangle$ level. The interference of the $A_{LM}^{(N)}$ with different values of L becomes unimportant, since the resonant value $A_{LM}^{(N)}$ is much larger than the others, and W in (21) breaks up into the product of the probability for the N -photon population of the real level $|n, L\rangle$ and the probability for the ionization of this level by the infrared field.

In conclusion we note that this mechanism for ionization in fields of two frequencies is pertinent to an experimental study of the tunneling ionization of atoms. This method makes it a simple matter to vary γ (by a small change in the frequency Ω of the optical radiation) and to study the functional dependence of the probability on the amplitude of the infrared field. The saturation effects which would ordinarily complicate such measurements can easily be eliminated in this case, by changing the intensity of the optical field. It is of course necessary that the tunneling probability outweigh the probability for direct $(N + 1)$ -photon ionization in the field F . The best situations are thus those with $N = 1$ and 2, e.g., the case of alkali atoms with $N = 2$ in the field of a ruby laser.

In this case the tunneling ionization in the field of a CO₂ laser will be dominant at optical fields $F \lesssim 10^5$ V/cm. We should also point out that it is possible to extract further information about the ionization process by studying the dependence of W on the relative orientation of the polarization vectors of the infrared and optical fields. As in the case of a static field,⁷ W can be expected to reach a maximum in the case of parallel polarizations and a minimum in the case of orthogonal polarizations.

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