Tunnel ionization of highly excited atoms in a noncoherent laser radiation field

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A theory is developed of the ionization of highly excited atomic states by a low-frequency field of noncoherent laser radiation with a large number of modes. Analytic formulas are obtained for the probability of the tunnel ionization in such a field. An analysis is made of the case of the hydrogen atom when the parabolic quantum numbers are sufficiently good in the low-frequency limit, as well as of the case of highly excited states of complex atoms when these states are characterized by a definite orbital momentum and parity. It is concluded that the statistical factor representing the ratio of the probability in a stochastic field to the probability in a monochromatic field decreases, compared with the case of a short-range potential, if the "Coulomb tail" is included. It is shown that at a given field intensity the statistical factor decreases on increase in the principal quantum number of the state being ionized.

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We shall consider the process of ionization of highly excited atomic states by the field of low-frequency laser radiation. In a static electric field the ionization probability w_c for an atomic state with the principal quantum number n and parabolic quantum numbers n_1 and n_2 is determined using the hydrogen-like approximation[1,2] valid in the case of highly excited states, i.e., when $n \gg 1$:

$$w_c=\left(\frac{4}{n^3\mathscr{E}}\right)^{n-n_1+n_2}\frac{\exp[-2/3\mathscr{E}n^3+3(n_1-n_2)]}{n^3 n_2!\,(n-n_1-1)!}. \tag{1}$$

Here, $\mathscr{E}$ is the electric field intensity.

In the case of a monochromatic field of low frequency ω, such that the condition $(\omega/n\mathscr{E})^2 \ll 1$ is satisfied,[3] the tunneling probability per unit time w_m is obtained from Ref. 4:

$$w_m=(3\mathscr{E}n^3/4\pi)^{1/2}w_c, \tag{2}$$

where w_c is given by Eq. (1). The result given by Eq. (2) applies when $n_2 > n_1$. If $n_2 < n_1$ Eq. (2) should be modified by the transposition $n_1 \rightleftarrows n_2$. It should be noted that Eq. (2) is obtained ignoring exponentially small corrections in the $n \gg 1$ case. It is shown in Ref. 5 that, in reality, the result (2) is valid subject to a condition more stringent than that given above, namely $(\omega/n\mathscr{E})^2 n \ll 1$. If only our less stringent condition is obeyed, the splitting of the initial term into quasienergy levels becomes important; the formulas for the tunneling probability then become much more complex. They are given in Ref. 5.

We shall assume that the conditions are such that we are well within the tunneling region, so that the expression (2) is valid in a monochromatic field. However, in reality, a laser radiation field is frequently nonmonochromatic. Our aim will be to calculate the probability of tunnel ionization in a multimode laser radiation field. It is known that such a field is characterized by the following distribution between the amplitudes of the electric field of the wave:

$$W(\mathscr{E})=(2\mathscr{E}/\overline{\mathscr{E}}^2)\exp(-\mathscr{E}^2/\overline{\mathscr{E}}^2). \tag{3}$$

Here, $\overline{\mathscr{E}}$ is the rms amplitude of the electric field of the laser radiation wave.

We shall multiply Eq. (2) by the distribution (3), and integrate the product with respect to $\mathscr{E}$ between the limits 0 and ∞ using the steepest descent method. The justification for the use of this method is the smallness of the value of $\overline{\mathscr{E}}$ with the characteristic atomic field $1/n^4$. Omitting the intermediate steps, we give only the final result:

$$w=\exp\left[3(n_1-n_2)-\frac{3^{1/3}}{n^2\overline{\mathscr{E}}^{2/3}}\right]\times\left(\frac{4\cdot 3^{2/3}}{n^2\overline{\mathscr{E}}^{2/3}}\right)^{n-n_1+n_2}\frac{2}{3^{1/3}n^3 n_2!\,(n-n_1-1)!}. \tag{4}$$

Like Eq. (2), it applies to the case $n_2 > n_1$. If $n_2 < n_1$ then Eq. (4) should be modified by the transposition $n_1 \rightleftarrows n_2$.

We shall now calculate the statistical factor G which is defined as follows:

$$G=w(\overline{\mathscr{E}})/w_m(\overline{\mathscr{E}}).$$

Dividing Eq. (4) by Eq. (2), we obtain the result in the form which is valid when $n_1 > n_2$ or $n_1 < n_2$ (and also when $n_1 = n_2$):

$$G=\frac{2}{3}\left(\frac{\pi}{\overline{\mathscr{E}}n^3}\right)^{1/2}(3n^3\overline{\mathscr{E}})^{(n+|n_1-n_2|)/3}\exp\left[\frac{2}{3n^3\overline{\mathscr{E}}}-\frac{3^{1/3}}{n^2\overline{\mathscr{E}}^{2/3}}\right]. \tag{5}$$

The first multiplier in Eq. (5) represents the difference between the statistical factor in the Coulomb potential and the statistical factor in the short-range potential.[6] Since this multiplier is small compared with unity, we may draw the conclusion that in the Coulomb potential the statistical factor is much less than in the short-range potential and that on increase in the principal quantum number n the statistical factor decreases rapidly (for a fixed value of the field intensity $\overline{\mathscr{E}}$).

It is assumed in these calculations that on application of the field the parabolic quantum numbers n_1 and n_2 remain "good," in spite of the fact that the external field is alternating. The small degree of their mixing is due to the fact that it occurs only as a result of allowance for other principal atomic shells and the effect of these shells is small on condition[7]

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that $\alpha_n \mathscr{E}^2 \ll \omega_{nn'}$, where α_n is the atomic polarizability of the n-th level. An estimate based on $\alpha_n \propto n^6$, yields the condition $\mathscr{E} \ll n^{-4.5}$, when Eq. (5) is valid.

It should also be pointed out that under these validity conditions the frequency ω of the external field is known to be low compared with the classical frequency $1/n^3$ of the revolution of an electron along a highly excited orbit.

We shall now consider the case when the atomic states are characterized by an orbital momentum l and its projection m. In the case of highly excited (Rydberg) atomic states this situation is encountered when the energy separations between the levels of one multiplet with different values of l are large compared with the perturbation, i.e., compared with the value of $n^2\mathscr{E}$. According to Ref. 4, in a monochromatic field the probability w_m of tunnel ionization is of the form

$$w_m=|C_{nl}|^2 \frac{(2l+1)(l+|m|)!}{2^{|m|}2n^2(|m|)!(l-|m|)!} \left(\frac{3n^2\mathscr{E}}{\pi}\right)^{1/2} \left(\frac{2}{n^3\mathscr{E}}\right)^{2n-|m|-1} \times \exp\left(-\frac{2}{3n^3\mathscr{E}}\right). \quad (6)$$

Here, C_{nl} is the coefficient in the asymptotic expansion of an unperturbed atomic wave function $\Psi(\mathbf{r})$ at large distances $\mathbf{r}$ from an atom:

$$\Psi(\mathbf{r})_{r\to\infty}=C_{nl}n^{-3/2}\left(\frac{r}{n}\right)^{n-1} \exp\left(-\frac{r}{n}\right) Y_{lm}(\Omega). \quad (7)$$

In spite of the dependences of the level energies on l because of perturbation of the atomic core potential, the wave functions Ψ of highly excited states remain, to a good approximation, hydrogen-like. According to Ref. 8, the coefficient C_{nl} in Eq. (7) has the following value:

$$C_{nl}=2^n[n(n+l)!(n-l-1)!]^{-1/2}. \quad (8)$$

Equations (6) and (8) yield the expression for the probability w_m of tunnel ionization from a highly excited state (nlm) in a monochromatic electromagnetic field:

$$w_m=\left(\frac{3}{\pi}\right)^{1/2} \frac{2(2l+1)(l+|m|)!}{n^3(n+l)!(n-l-1)!(|m|)!(l-|m|)!} \times\left(\frac{4}{n^3\mathscr{E}}\right)^{2n-|m|-3/2} \exp\left(-\frac{2}{3n^3\mathscr{E}}\right). \quad (9)$$

Multiplying Eq. (9) by the distribution (3) and integrating with respect to $\mathscr{E}$, we find the probability w of tunnel ionization of a state (nlm) in a stochastic electromagnetic field of low frequency. We shall give only the relevant statistical factor G, where w_m is substituted in the form of Eq. (9):

$$G=\frac{2}{3}\left(\frac{\pi}{\mathscr{E}n^3}\right)^{1/2} (3n^3\mathscr{E})^{(2n-|m|-1)/3} \exp\left[\frac{2}{3n^3\mathscr{E}}-\frac{3^{1/3}}{n^2\mathscr{E}^{1/3}}\right]. \quad (10)$$

Naturally, it differs from Eq. (5) only by a different form of the preexponential multiplier.

It is worth noting that, in contrast to the probabilities w_m and w themselves, the statistical factor of Eq. (10) is independent of the orbital momentum l.

We may therefore conclude that the multimode nature of a laser radiation field increases the probability of tunnel ionization compared with the monochromatic field case, but the increase becomes smaller for larger principal quantum numbers of the state being ionized.

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