

## Dependence of Rydberg-state field-ionization thresholds on $|m_l|$

W. E. Cooke and T. F. Gallagher

*Molecular Physics Laboratory, SRI International, Menlo Park, California 94025*

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The variation of field-ionization thresholds due to the  $m_l$  quantum number is calculated using a classical model. Results are compared to the  $|m_l|$ -dependent field-ionization thresholds of sodium Rydberg states. An approximate solution and scaling laws are presented.

There has been much recent interest in the field ionization of Rydberg states because of new laser techniques developed to produce them and because the Stark fields necessary to ionize them may be easily produced in the laboratory.<sup>1</sup> The simplest model of ionization is the classical model which predicts ionization when the the field is large enough to reduce the potential barrier below the electron's energy. This model predicts a threshold ionizing field of

$$F_{\text{th}} = \frac{1}{4}(W^*)^2 = 1/(2n_s)^4 \quad (\text{a.u.}), \quad (1)$$

where  $W^* = 1/2n_s^2$  is the energy of the electron in the Stark field  $F_{\text{th}}$ , and  $n_s$  is defined to be the effective quantum number.<sup>2</sup> Attempts to treat the problem of field ionization of hydrogen quantum mechanically by calculating the tunneling probability of the electron through the potential barrier<sup>3,4</sup> or by using a variational procedure to determine bound states<sup>5</sup> have met with success for lower  $n$  states. In particular, calculations predict that those states which are Stark shifted to greater binding energies (red shifted) will ionize at lower fields than those which are shifted to lower binding energies (blue shifted). For states near  $n=5$ , this behavior and the threshold field values agree with experiment.<sup>6</sup> However, experiments in Na excited in a Stark field have now demonstrated that the effects of other manifold states can add considerable further complications. Some states, in the  $n=15$  manifold, were found to have ionization rates which did not monotonically increase with electric field, but rather exhibited local maxima.<sup>7</sup> In these cases, even the concept of a threshold field may become nebulous.

In contrast to this complexity, other experiments with Na, using pulsed electric fields to detect Rydberg states of  $n \geq 16$ , have shown well-behaved ionization thresholds, which can be simply characterized by the classical model.<sup>8</sup> Presumably, the strong couplings to other manifolds induce the classical behavior (e.g., blue-shifted states are found to have lower thresholds than red-shifted states), while the dynamic detection technique effectively obscures any subtleties in the

ionization thresholds. In these studies, the general tendencies for  $m_l=0$  states were modeled by assuming adiabatic passage as the electric field increased in time (with a slew rate of up to  $2 \times 10^{10}$  V/cm sec). These studies have shown that the classical ionization threshold field of Eq. (1) is valid for  $m_l=0$  states. States of higher  $|m_l|$  value were found to ionize at progressively higher thresholds, and this was attributed qualitatively to an energy conservation effect. For an electron with a nonzero projection of angular momentum along the field ( $z$ ) axis, a portion of its kinetic energy is used in momentum perpendicular to the field. This energy is not useful for escape, and therefore a higher field is necessary to ionize the atom. In this comment we calculate the shift in ionization thresholds due to nonzero  $m_l$ , using this classical model. Although such variation is implicit in the previously mentioned quantum-mechanical models, the calculation presented here results in a simple variation which scales with the energy of the state and avoids a complicated dependence upon the parabolic quantum numbers. Thus, this calculation extends the usefulness of the classical model for characterizing field-ionization detection schemes.

The effective potential energy of an electron, at position  $\vec{r}$ , including a centrifugal term resulting from a projection of angular momentum along the  $z$  axis ( $m\hbar$ ), can be written in atomic units:

$$V_{\text{eff}} = m^2/2\rho^2 - z/r_1^2 - 1/r, \quad (2)$$

where  $\rho^2 = r^2 - z^2$ , and  $r_1$  is defined so that  $(r_1)^{-2}$  is the electric field. The extrema of the effective potential may be found by setting the derivatives to zero:

$$\frac{\partial V_{\text{eff}}}{\partial z} = \frac{z}{r^3} - \frac{1}{r_1^2} = 0, \quad (3a)$$

$$\frac{\partial V_{\text{eff}}}{\partial \rho} = -\frac{m^2}{\rho^3} + \frac{\rho}{r^3} = 0. \quad (3b)$$

Using Eq. (3),  $\rho$  and  $z$  may be eliminated to obtain an equation for the value of  $r$  at which the extrema occur. This equation may be set into a dimension-

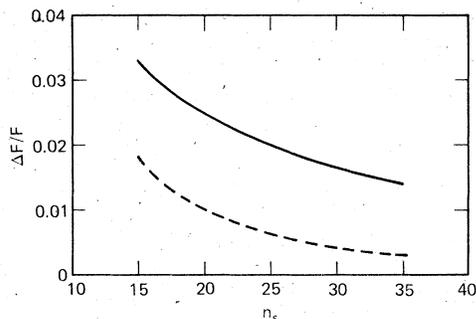


FIG. 1. Solid line is the difference between  $m=0$  and  $|m|=1$  ionization threshold fields, as a function of the effective quantum number. The dashed line is the difference between the thresholds for adjacent manifold states for  $m_l=0$ .

less form by defining  $x=r/r_1$ , which results in

$$x(1+x)^2(1-x)^2(1+x^2)^2 = m^2/r_1. \quad (4)$$

For  $m=0$ , Eqs. (3a) and (3b) may be easily solved to find the saddle point of the potential, which occurs at  $r_0$ , given by

$$\rho_0=0, \quad r_0=z_0=r_1, \quad x_0=+1. \quad (5)$$

In this case, the electron escapes over the saddle point at a distance of  $r_1$  along the  $z$  axis. For small, but nonzero values of  $m^2/r_1$ , this escape site splits into a line of saddle points which encircle the  $z$  axis near the old value  $x=1$ , but with a small nonzero value of  $\rho$ . To determine the position of the saddle ring, Eq. (4) may be solved for the roots near  $x=1$ .

For the electron to pass over this ring, its energy must be greater than or equal to  $V_{\text{eff}}$  at the escape site, leading to a threshold energy of

$$E_{\text{th}} = (1+3x^4)/(2r_1x) = 1/(2n_s^2), \quad (6)$$

which gives a threshold field

$$F_{\text{th}} = \frac{4x^2}{(1+3x^4)^2} E_{\text{th}}^2 = \frac{x^2}{(1+3x^4)^2 n_s^4}. \quad (7)$$

When Eq. (6) is used to eliminate  $1/r_1$  from Eq. (4), the resulting equation for the position of the saddle point is

$$(1+x)(1+3x^4)^{1/2}(1-x)(1+x^2) = |m|/n_s, \quad (8)$$

which now determines  $x$  from the energy of the

state when it ionizes. The  $x$ , then determined, may be used with Eq. (6) to obtain the threshold field for ionization of a state as a function of the energy of that state.

A solution of these equations near  $n_s=17$  predicts that the  $|m|=1$  ionization thresholds should be 3.0% higher than the  $m=0$  states. Gallagher *et al.*<sup>8</sup> report an increase of 3%, in excellent agreement. For the  $|m|=2$  states, however, the agreement is not as good. The model predicts an increase of 6.3% over the  $m=0$  thresholds, while an increase of 23% was actually observed. This discrepancy could be due to the breakdown of the adiabatic passage approximation, which was already suggested by the observation of multiple  $|m|=2$  thresholds for some  $d$  states. A 4% decrease in the  $d$ ,  $|m|=2$  state energy (relative to the  $m=0$  state) due to nonadiabatic passage would explain the difference.

An approximate solution of Eqs. (7) and (8) leads to an expression for the fractional change of threshold field from the  $m=0$  value:

$$\Delta F/F = m/2n_s. \quad (9)$$

This should be compared to the typical difference between ionization thresholds for  $m=0$  states of angular momentum  $l$  and  $l+1$ , obtained using the adiabatic model of Gallagher *et al.*<sup>8</sup>:

$$\Delta F/F = 4/n_s^2. \quad (10)$$

Figure 1 illustrates the dependence of the two fractional changes on the effective quantum number  $n_s$ . Since the variation due to a change in  $m$  is much greater than the variation from one manifold state to an adjacent state for large  $n_s$ , this implies that field ionization will be better suited for differentiating between  $m$  states rather than  $l$  states. This may be of use in resonance experiments at higher  $n$  values, for example, where the detection system could monitor the depolarization of a prepared  $|m_l|$  state, rather than the population increase in a state with only a marginally different threshold.

#### ACKNOWLEDGMENT

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<sup>8</sup>T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein, *Phys. Rev. A* 16, 1098 (1977).