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SINGLE-ELECTRON DESCRIPTION OF THE STRONG-FIELD ELECTRON DETACHMENT OF HYDROGEN NEGATIVE ION[†]

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Abstract. The applicability of the single-electron model in describing the electron detachment of the hydrogen negative ion in strong (static or laser) fields is examined. By comparing the values for the lowest state energies and detachment rates obtained using two different short-range model-potentials with the results of recent ab initio calculations using the full two-electron description (Milošević and Simonović, 2016), it is found that the single-electron description is applicable for the field intensities up to few hundred GW/cm². This description, therefore, can be used for studying multiphoton processes or the electron detachment via tunneling at these field strengths, but fails in over-the-barrier regime.

Key words: hydrogen negative ion, electron detachment, strong field, short-range potential

1. INTRODUCTION

Negative ions generally represent a special class of atomic systems (see overview in Andersen, 2004) due to the properties which are significantly different from those of neutral atoms and positively charged ions. While in the former the electrons are bound in the long-range Coulomb potential, the excess electron in negative ions is due to the neutral atomic residue bound in a short-range potential. In addition, the corresponding binding energies are significantly lower than the atomic ionization potentials. The hydrogen negative ion (H⁻), as the simplest among them, has also some specific features like the absence of singly excited states. In other words this two-electron system has only one bound state, that is the ground state, and a series of autoionizing doubly-excited states. The binding energy of H⁻ is $E_B = 0.7542$ eV (0.0277 a.u.) (see e.g. Andersen et al.,

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1999). A very weak binding and the absence of a long-range Coulomb attraction for the separated electron (the atomic residue is the neutral hydrogen atom) leads to a specific radial correlation between the electrons in the ground state such that one electron is bound much closer to the nucleus than the other which is weakly held at a distance of 4-5 Bohr radii from the nucleus (Chandrasekhar, 1944, see also Rau, 1996). Such a configuration suggests a very useful single-electron picture where the outer electron is weakly (loosely) bound in a short-range attractive potential. To a good approximation the potential acting on the outer electron due to the neutral atom is a sum of a short-range potential and the polarization potential falling off as $1/r^4$.

Collisions between photons and H⁻ leading to one-electron ejection (electron photodetachment) have played an important role in the study of this ion (see Andersen, 2004 and references therein). At the threshold of the single-photon detachment the residual hydrogen atom is in principle left in the ground state and no long-range forces act on the outgoing electron (Bryant and Halka, 1996). The experimental cross section is found to be in a good agreement with the Wigner low (that is a feature of short-range potentials), confirming also the threshold energy $E_{\rm th} = E_{\rm B} = 0.7542$ eV (Lykke *at al*, 1991). During the last two decades, intense lasers have made it possible to observe the effects of multiphoton detachment processes (Rau, 1996, Andersen, 2004). In contrast to the single-photon case, the multiphoton detachment may occur at the photon energies $\hbar \omega < E_{\rm B}$, but since the detachment rates in this case are significantly lower, in order to get a measurable effect one needs much larger intensities.

At stronger fields, however, another mechanism for the electron detachment arises – the quantum-mechanical tunneling. A strong field distorts the potential of atomic residue forming a potential barrier (Stark saddle) through which the electron can tunnel. At even stronger fields the barrier may be suppressed below the energy of the bound state and over-the-barrier detachment (OBD) occurs. The transition from the multiphoton to the tunnelling regime is governed by the Keldysh parameter $\gamma = \omega (2m_e E_B)^{1/2}/eF$ (Keldysh, 1965), where ω is the frequency of the electromagnetic field and *F* is the peak value of its electric component. This parameter characterizes the degree of adiabaticity of the motion through or over the barrier. If $\gamma >> 1$ (low-intensity/short-wavelength limit) multiphoton processes dominate, whereas for $\gamma \ll 1$ (high-intensity/long-wavelength limit) the tunneling or OBD mechanism does. In the latter case the quasistatic description is a good approximation. It assumes that the electric field changes slowly enough that a static detachment rate can be calculated for each instantaneous value of the field. Then the detachment rate for the alternating field can be obtained by averaging the static rates over the field period.

In a recent study we compared the values for the lowest state Stark shift and detachment rates for H⁻ at different strengths of the applied (quasi)static field, determined by using the single-electron and the full two-electron description (Milošević and Simonović, 2016). It is shown that the single-electron description fails in OBD and partially in the tunneling regime. Here we focus on the range of field strengths where the single-electron approach may be valid. In the next section we introduce the single-electron model for the hydrogen negative ion in the external (quasi)static field and consider three different short-range model-potentials. The numerical (complex-rotation) method is described in Section 3. Finally, in Section 4, we present numerical results (the lowest-state energies and detachment rates) and compare them with the results obtained using the full two-electron description.

2. MODELS

As mentioned in the introductory part, the configuration of the ground state of H⁻ suggests a one-electron description where the outer (loosely bound) electron moves in a short-range potential V(r) describing the attraction by the neutral atomic residue. Then, in the presence of a (quasi)static electric field *F* the dynamics of the outer electron may be described by the Hamiltonian (in atomic units)

$$H = \frac{\mathbf{p}^2}{2} - V(r) - F_Z, \qquad (1)$$

where $\mathbf{r} = (x, y, z)$ and \mathbf{p} are the electron's position and momentum, respectively. The potential V(r) is usually calibrated such that the lowest eigenenergy $\varepsilon(F)$ of this Hamiltonian when F = 0 has the value $-E_{\rm B}$. When $F \neq 0$ the total potential $V_{\rm tot} = V(r) - Fz$ has the potential barrier which the electron can tunnel through or escape over it. Thus, the lowest state, which is an exact bound state when F = 0, has the resonant character when the field is present. The width Γ of this resonant state, which increases with *F*, determines the electron detachment rate $w(F) = \Gamma(F)/\hbar$ (hereafter we set $\hbar = 1$).

Another effect of the field is the Stark shift of energy levels. For $F \ll 1$ the Stark shift can be expanded in a Maclaurin series, where in the ground state case the lowest order term is quadratic, i.e. $\Delta E(F) = E(F) - E(0) = -\alpha F^2/2! - \gamma F^4/4! - \ldots$ The first two coefficients α and γ are known as the dipole polarizability and the second dipole hyper-polarizability, respectively. For the hydrogen negative ion these values are $\alpha = 206$ and $\gamma = 8.03 \cdot 10^7$ (Radzig and Smirnov, 1985, Pipin and Bishop, 1992). Therefore, for weak fields the energy of the outer electron of H⁻ in the lowest state is determined approximately by the fourth-order formula

$$\varepsilon(F) = \varepsilon(0) + \Delta E = -E_{\rm B} - \frac{1}{2}\alpha F^2 - \frac{1}{24}\gamma F^4.$$
⁽²⁾

In theoretical studies it is convenient to treat the energies and widths of resonant states simultaneously using the scattering matrix formalism which deals with complex energies. Within this formalism a resonant state is an eigensolution of the Schrödinger equation that is not square integrable (it asymptotically behaves as a purely outgoing wave) and corresponds to a complex eigenenergy E_{res} . This energy is related to a pole of the scattering matrix and its real and imaginary parts determine the energy (position) and the width of the resonance, $E = \text{Re}(E_{\text{res}})$ and $\Gamma = -2\text{Im}(E_{\text{res}})$.

2.1. The zero-range potential

The simplest short-range potential that can be used to describe the dynamics of a weakly bound electron in negative ions is the zero-range potential (ZRP, see e.g. Demkov and Ostrovskii, 1988)

$$V(r) = -a\delta(r), \quad a > 0. \tag{3}$$

This potential supports only one bound state with the binding energy $E_{\rm B} = a^2/2$. The corresponding wave function has the form $\psi = (\kappa/2\pi)^{1/2} \exp(-\kappa r)/r$, where $\kappa = (2E_{\rm B})^{1/2} = a$. In the case of H⁻ it is a = 0.2354.

The eigenvalue problem of the single-electron Hamiltonian (1) with the ZRP admits for weak fields a solution in a closed analytical form. The position and width of the lowest energy level are determined from the real and imaginary parts of the associated complex eigenenergy (Demkov and Drukarev, 1964, Demkov and Ostrovskii, 1988)

$$\varepsilon = -E_{\rm B} - \frac{F^2}{32E_{\rm B}^2},\tag{4}$$

$$\Gamma = c \frac{F}{\kappa} \exp\left(-\frac{2\kappa^3}{3F}\right),\tag{5}$$

where c = 1/2. The polarizability of H⁻ resulting from Eq. (4), $\alpha = 9/(8E_B^2) \approx 81.5$, is much lower than the experimental value or that obtained from *ab initio* calculations using the full twoelectron description (Milošević and Simonović, 2016). On the other hand, the formula (5) for the width (detachment rate) agrees approximately with the *ab initio* calculations that give $c \approx$ 0.6. Here we note that both the polarizability and the decay rate (for weak fields) given by the ZRP approximation are minimal among all systems with potentials V(r) < 0 at a fixed value of the binding energy E_B (Demkov and Ostrovskii, 1988).

2.2. Polarization potentials

More realistic single-electron models for H⁻ can be constructed by including the polarizability of the atomic residue. In such a model the loosely bound electron moves in an effective potential that is the sum of a short-range potential and the polarization term which asymptotically behaves as $1/r^4$. The parameters of the effective potential must be tuned to provide the correct value of the binding energy $E_{\rm B}$. An alternative is the model-potential consisting of a single term which provides both the correct binding energy and the correct asymptotics. Several model-potentials of these two types, used by different authors, are given in a short overview by Grujić and Simonović (1998).

One of the most widely used model-potentials of the first type is that proposed by Cohen and Fiorentini (1986)

$$V_{\rm CF} = -\left(1 + \frac{1}{r}\right)e^{-2r} - \frac{\alpha_{\rm H}}{2r^4}e^{-r_0^2/r^2},$$
(6)

where $\alpha_{\rm H} = 9/2$ is the polarizability of the hydrogen atom. The exponential factor in the polarization term removes the unphysical singularity of this term as $r \rightarrow 0$. The parameter $r_0 = 1.6$ is chosen by the condition that the potential (6) has a single bound state with the correct binding energy.

Another, more approximate but simpler model-potential which belongs to the secondtype is the so-called Buckingham polarization potential (Buckingham, 1938, see also Mittleman and Watson, 1960, Grujić and Simonović, 1998)

$$V_{\rm B} = -\frac{\alpha_{\rm H}}{2(r^2 + d^2)^2} \,. \tag{7}$$

For d = 1.033906 this potential has a single bound state with the correct value of binding energy for H⁻.

An external (quasi)static field distorts the potential of atomic residue forming a potential barrier (Stark saddle), the saddle point of which is located at the z (field) axis. Its position $\mathbf{r}_{sp} = (0, 0, z_{sp})$ and hight $V_{sp} = V_{tot}(\mathbf{r}_{sp}; F)$ depend on the field strength *F* and can be determined from the rule $(\partial V_{tot}/\partial z)_{x=y=0} = 0$. The field strength F_S that separates the tunneling and OBD regimes is defined by the condition $\varepsilon(F_S) = V_{sp}(F_S)$. Note that these values may vary by changing the model-potential V(r). (Note that in the case of ZRP one has $z_{sp} = V_{sp} = 0$ for any field strength and OBD never occurs.) Using the dependence $\varepsilon(F)$ determined by solving the eigenvalue problem of Hamiltonian (1) for the potentials (6) and (7) numerically (see Table 1) we obtain $F_S = 0.0055$ a.u. (in both cases) that corresponds to the laser field intensity of about 10^{12} W/cm².



Fig. 1 (a) The x = y = 0 cut of the Cohen-Fiorentini (full line) and Buckingham (dashed line) potential. (b) The cut of the total potential $V_{tot} = V(r) - Fz$ for F = 0.004 a.u. The horizontal lines mark the positions of the lowest energy level ε .

3. NUMERICAL METHOD

For a given model-potential $V(\mathbf{r})$ and the field strength the energy and width of the lowest state of H⁻ in electric field can be determined numerically by the use of the complex rotation method (see e.g. Reinhardt, 1976, Buchleitner, et al. 1994). The basic idea of this method is to make the resonance wave function $\psi(\mathbf{r})$ square integrable by a complex rotation of the coordinate, $\psi(\mathbf{r}) \rightarrow \psi_{\theta}(\mathbf{r}) = \psi(e^{i\theta} \mathbf{r})$, where θ is a real parameter called the 'rotation angle'. Such a 'rotated' state $\psi_{\theta}(\mathbf{r})$ is an eigenfunction of the so-called complex rotated Hamiltonian H_{θ} , obtained from the original Hamiltonian H by the transformations $\mathbf{r} \rightarrow e^{i\theta} \mathbf{r}$, $\mathbf{p} \rightarrow e^{-i\theta} \mathbf{p}$. H_{θ} is a non-Hermitian operator, whose spectrum is in general complex, depends on the rotation angle θ and has the following properties: (i) The bound (discrete) spectra of H_{θ} and H coincide; (ii) The continua are rotated by the angle 2θ into the lower complex energy halfplane; (iii) The resonances of H coincide with the complex eigenvalues of H_{θ} .

The spectrum of Hamiltonian (1), therefore, can be computed by diagonalizing the corresponding rotated Hamiltonian

$$H_{\theta} = e^{-2i\theta} \frac{\mathbf{p}^2}{2} - V(e^{i\theta}r) - e^{i\theta}Fz, \qquad (8)$$

with a properly tuned parameter θ in a square-integrable basis which must be complete in a sense that it covers the continuous part of the spectrum, too. For this purpose, we choose the Sturmian basis (see e.g. Avery and Avery, 2006)

$$\chi_{nlm}^{(k)}(\mathbf{r}) = 2k^{3/2} \sqrt{\frac{(n-l-1)!}{n(n+l)!}} (2kr)^l e^{-kr} \mathcal{L}_{n-l-1}^{2l+1}(2kr) Y_{lm}(\vartheta, \varphi), \qquad (9)$$

where L denotes the generalized Laguerre polynomials. The functions (9), the so-called Coulomb Sturmians, are solutions of the equation $(-\nabla^2 + k^2 - 2nk/r)\chi_{nlm}^{(k)}(\mathbf{r}) = 0$ and obey the potential-weighted orthonormality relation

$$\int d^{3}\mathbf{r}\chi_{nlm}^{(k)*}(\mathbf{r})\frac{1}{r}\chi_{n'l'm'}^{(k)}(\mathbf{r}) = \frac{k}{n}\delta_{nn'}\delta_{ll'}\delta_{mm'} \cdot$$
(10)

Although the main purpose of this basis is to study the systems with dominant Coulomb interaction, we demonstrated that it can be used in the problems with short-range potentials, too. Here it is particularly convenient that the matrix elements of the rotated Hamiltonian (8) in this basis, using either the potential (6) or (7), can be expressed in analytical forms. The Buckingham potential (7) essentially has the same form as the dipole term in the Bardsley pseudopotential used recently to study alkali-metal atoms in electric field (Milošević and Simonović, 2015). Thus, for the H⁻ model with the model-potential (7) all required matrix elements can be found in Appendices A and B in this reference.



Fig. 2 Complex trajectories showing the evolution of complex energy of the lowest eigenstate of rotated Hamiltonian (8) (with the Cohen-Fiorentini potential) for F = 0.004 a.u. by varying the rotation angle θ (with a constant step) for a fixed value of the parameter k. Diagonalization of Hamiltonian (8) is performed in the Sturmian basis with $n_{\text{max}} = 12$. The arrows are directed towards the growth of angle θ . The position of the true value of complex eigenenergy (in this case $E_{\text{res}} = -0.5289 - i0.546 \cdot 10^{-3}$) is located in a close vicinity of the stationary points (which can be identified by the maximum density of points) of complex trajectories.

It should be mentioned that, due to non-orthogonality of the functions (9), the Schrödinger equation $(H_{\theta} - E_{res}) \psi_{\theta} = 0$ represented in the Sturmian basis reduces to the generalized eigenvalue problem of the form $(\mathbf{H} - E_{res}\mathbf{S}) \mathbf{x} = 0$. The matrices \mathbf{H} and \mathbf{S} are given by the matrix elements $\langle nlm; k | H_{\theta} | n'l'm'; k \rangle$ and $\langle nlm; k | n'l'm'; k \rangle$, respectively (see Appendices A and B in Milošević and Simonović, 2015), whereas the components of the eigenvectors \mathbf{x} are $\langle nlm; k | \psi_{\theta} \rangle$. Convergence of the results was assured by optimizing the Sturmian parameter k and the rotation angle θ . For a large basis (few tens of states (9)) the resonances only weakly depend on the rotation angle θ . Here, with $k \sim 1$, θ takes the values between 0 and 1 rad (depending on the field strength). When the parameters are adequately adjusted the computed resonance energies are approximately stationary with respect to variations of these parameters (see Fig. 2).

4. RESULTS AND CONCLUSIONS

The lowest-state energies $E(F) = \varepsilon(F) - 0.5$ a.u. and widths $\Gamma(F)$ (detachment rates w(F)) at different strengths of the applied electric field *F*, obtained using the single-electron approach with the Cohen-Fiorentini and the Buckingham polarization potential (Eqs. (6) and (7), respectively), are shown in Table 1 and Fig. 3 together with the results of *ab initio* calculations determined using the full two-electron description (Milošević and Simonović, 2016). The results obtained by applying the polarization potentials are very close to each other for all field strengths. In addition, at weak fields (approximately for $F < F_S/2$) the corresponding values for energies E(F) agree well with the result of *ab initio* calculations, as well as with those estimated from the Stark shift expansion (2) (see Fig. 3(a)). At stronger fields, however, the difference between the results obtained using the single- and twoelectron models increases. In the OBD area ($F > F_S$) the Stark shift ΔE determined numerically using the full two-electron approach is about two times larger than that obtained from the single-electron models.

The detachment rates obtained using the polarization potentials (6) and (7), as well as the ZRP, agree well with the results of *ab initio* calculations at weak fields ($F < F_S/2$, see Fig. 3(b)). One can see that the rates generally agree mutually better than the energies. This can be explained by the fact that the form of total potential V_{tot} is less sensitive to the form of the model-potential V in the barrier area than in the atomic area. The numerical results for rates obtained using the models (6) and (7), as well as the results of *ab initio* calculations, fit approximately to the form (5) determined using the ZRP model, but for different values of the constant c (for the ZRP model it is c = 0.5). In order to estimate this constant from numerical data we expressed the rate w in terms of the variable $\xi = (F/\kappa) \exp(-2\kappa^3/3F)$. Then Eq. (5) reduces to the linear dependence $w = c\xi$. The linear fit for the results of *ab initio* calculations gives $c \approx 0.62$ in the tunneling domain and $c \approx 0.58$ in OBD area (Milošević and Simonović, 2016). As mentioned above, the results obtained using the single-electron models (6) and (7) are at weak fields ($F < F_S/2$) close to the results of *ab initio* calculations (see the inset in Fig. 1(b)). (Note that in this area the ZRP values are indeed the minimal.) At stronger fields, however, these results are closer to the values given by the ZRP model.



Fig. 3 (a) The lowest state energy E and (b) width Γ (the electron detachment rate w) as functions of the electric field strength F. The open and gray circles denote the results obtained numerically using the single-electron models with the Buckingham and the Cohen-Fiorentini potential, respectively. The dashed and full lines in part (a) show the lowest state energy E(F) estimated using the second and the fourth order Stark shift expansion formula, respectively. The full lines in part (b) show the rate w(F) given by the ZRP model (Eq. (5)). For comparison, the corresponding results obtained using the full two-electron description (Milošević and Simonović, 2016) are shown (+ symbols). The inset shows the same rates as functions of the variable ζ (see text).

In conclusion, the single-electron description of the strong-field electron detachment of hydrogen negative ion using polarization short-range model-potentials is applicable for the field strengths $F < F_S/2$ that correspond to the laser field intensities up to 0.2-0.3 TW/cm². Consequently, these models can be used for studying multiphoton processes or

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the electron detachment via tunneling at these field strengths, but fail in over-the-barrier regime. In addition, the single-electron approach using the ZRP model gives approximately good values for the detachment rates, however it fails for energies. Finally, in order to get accurate results at stronger fields ($F > F_S/2$) the full two-electron approach is necessary.

Table 1 The lowest state energy E and width Γ of the hydrogen negative ion at different strengths of the applied electric field F obtained using the single-electron picture with the Cohen-Fiorentini (6) and the Buckingham potential (7). For comparison, the results obtained using the full two-electron model are shown (Milošević and Simonović, 2016).

	Cohen-Fiorentini potential		Buckingham potential		Two-electron model	
F	E	Г	Ε	Г	Ε	Г
0	-0.52775	0	-0.52770	0	-0.52763	0
0.001	-0.52782	_	-0.52777	_	-0.52773	_
0.002	-0.52806	7.330E-5	-0.52802	7.312E-5	-0.52814	_
0.003	-0.52846	4.511E-4	-0.52840	4.301E-4	-0.52867	4.310E-4
0.004	-0.52887	1.091E-3	-0.52882	1.040E-3	-0.52928	1.247E-3
0.005	-0.52931	1.913E-3	-0.52924	1.885E-3	-0.52997	2.475E-3
0.006	-0.52974	2.933E-3	-0.52967	2.860E-3	-0.53057	3.845E-3
0.007	-0.53014	4.122E-3	-0.53004	3.968E-3	-0.53118	5.369E-3
0.008	-0.53053	5.335E-3	-0.53040	5.151E-3	-0.53177	7.022E-3
0.009	-0.53088	7.034E-3	-0.53074	6.430E-3	-0.53236	8.789E-3
0.010	-0.53121	8.440E-3	-0.53107	7.752E-3	-0.53293	0.01066
0.011	-0.53153	9.872E-3	-0.53137	9.095E-3	-0.53347	0.01258
0.012	-0.53182	0.01132	-0.53166	0.01049	-0.53397	0.01451
0.013	-0.53214	0.01284	-0.53192	0.01191	-0.53451	0.01654
0.014	-0.53240	0.01443	-0.53219	0.01337	-0.53503	0.01861
0.015	-0.53265	0.01595	-0.53243	0.01484	-0.53559	0.02078
0.016	-0.53289	0.01750	-0.53266	0.01634	-0.53609	0.02291
0.017	-0.53313	0.01894	-0.53289	0.01785	-0.53660	0.02505
0.018	-0.53336	0.02065	-0.53311	0.01937	-0.53708	0.02730
0.019	-0.53360	0.02227	-0.53332	0.02090	-0.53762	0.02956
0.020	-0.53376	0.02394	-0.53352	0.02244	-0.53817	0.03186
0.021	-0.53399	0.02562	-0.53371	0.02399	-0.53864	0.03414
0.022	-0.53422	0.02720	-0.53390	0.02556	-0.53915	0.03647
0.023	-0.53441	0.02883	-0.53408	0.02712	-0.53965	0.03883
0.024	-0.53454	0.03066	-0.53426	0.02870	-0.54016	0.04122
0.025	-0.53473	0.03229	-0.53443	0.03028	-0.54071	0.04362
0.026	-0.53492	0.03395	-0.53460	0.03187	-0.54120	0.04606
0.027	-0.53512	0.03563	-0.53476	0.03346	-0.54170	0.04848
0.028	-0.53525	0.03733	-0.53492	0.03506	-0.54222	0.05097
0.029	-0.53538	0.03900	-0.53507	0.03667	-0.54274	0.05349
0.030	-0.53556	0.04062	-0.53522	0.03827	-0.54320	0.05599

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JEDNOELEKTRONSKI OPIS OTKIDANJA ELEKTRONA OD NEGATIVNOG VODONIKOVOG JONA U JAKIM POLJIMA

Ispitivana je primenljivost jednelektronskog modela u opisu otkidanja elektrona od negativnog jona vodonika u jakim (statičkim ili laserskim) poljima. Poređenjem vrednosti za energije najnižeg stanja i stope otkidanja dobijenih pomoću dva različita kratkodometna model-potencijala sa rezultatima nedavnih ab initio izračunavanja koristeći puni dvoelektronski opis (Milošević i Simonović, 2016) utvrđeno je da je jedinoelektronski opis primenljiv u oblasti intenziteta do nekoliko stotina GW/cm². Ovakav opis se, prema tome, može koristiti kod proučavanja multi-fotonskih procesa ili otkidanja elektrona tuneliranjem pri ovim vrednostima polja, ali ne i u prekobarijernom režimu.

Ključne reči: negativni vodonikov jon, elektronsko otkidanje, jako polje, kratkodometni potencijal