Criterion for the yield of micro-object ionization driven by few- and subcycle radiation pulses with nonzero electric area

Nikolay Rosanov,[1,2,*] Dmitry Tumakov,[1,2] Mikhail Arkhipov,[2] and Rostislav Arkhipov[1,2]
[1]*Ioffe Institute, Politekhnicheskaya str. 26, St. Petersburg 194021, Russia*
[2]*St. Petersburg State University, 7/9 Universitetskaya nab., St. Petersburg 199034, Russia*

(Received 12 October 2021; accepted 18 November 2021; published 2 December 2021)

The photoionization yield is analyzed for three-dimensional quantum systems with finite number of discrete spectrum states driven by unipolar subcycle and few-cycle electromagnetic pulse with a duration much less than "the Kepler period," electron oscillation period in the ground state. The yield for such objects—symmetric quantum dots and those described by the zero-radius potential—is compared with the yield for hydrogen atom. In all these cases, the standard Keldysh ionization theory is inapplicable. It is shown that ionization probability is determined by the ratio of the electric pulse area (integral of the electric field strength over time) and its characteristic value inversely proportional to the size of the electron localization, and not by the pulse energy or its maximum intensity.

DOI: 10.1103/PhysRevA.104.063101

I. INTRODUCTION

An important trend in modern laser physics and technology is the shortening of laser pulses, which makes it possible to achieve extremely strong fields [1] and control extremely fast processes [2,3]. Today, radiation pulses with a duration of 42 as have been experimentally obtained [4,5], which is significantly less than the time of an electron's revolution in the first Bohr orbit. Various methods of obtaining unipolar radiation pulses are described in Ref. [6], and in Ref. [7] a cascade scheme is proposed in which a sub-10-as pulse duration is attainable. Transportation of subcycle and unipolar pulses is possible in coaxial waveguides, for which there is no cutoff frequency [8]. Half-cycle and unipolar pulses were demonstrated experimentally and studied theoretically in Refs. [9–13].

Subcycle and unipolar pulses with a significant content of the zero-frequency component of the field serve as the limit of pulse shortening. Many concepts of optics and interaction of radiation with matter require revision for such pulses.

The first publication on unipolar radiation pulses, apparently, belongs to Bessonov [14], who called them "strange waves." Further studies, reviewed in Refs. [6,15], revealed the essential role of such a characteristic of extremely short pulses as their electric area $\mathbf{S}_E = \int \mathbf{E}\, dt$, where $\mathbf{E}$ is the electric field strength and t is the time. In macroscopic electrodynamics, this quantity has nontrivial conservation properties that are valid for almost any media and are essential in the analysis of a number of effects [6,15].

At the microlevel, it turns out it is the electrical area of the pulses, and not their energy, that determines the effectiveness of the action of extremely short pulses on both free [16] and bound charges [6,17–24]. It is associated with the unidirectionality of the action of unipolar pulses, as opposed to bipolar ones. The magnitude of their impact can be estimated from the following considerations. For the short pulses under consideration, their effect on an electron with a charge e is reduced to imparting a momentum $\delta\mathbf{p} = e\mathbf{S}_E$ to it, while the electron does not have time to move a significant distance during the radiation pulse. The impact is significant if this momentum is comparable to the characteristic momentum of the micro-object $\mathbf{p}_0$.

From dimensional considerations, $p_0 = mc$ for a free electron, where m is the electron mass and c is the speed of light in vacuum. Then we obtain the scale of the electric area for a free electron $S_f = mc/e$, in accordance with Ref. [16].

The characteristic momentum of a bound electron localized in a region with size a, according to the uncertainty relation, is equal to $p_0 = \hbar/a$. Hence $S_b = \hbar/(ea)$, that is, the scale of the pulse electric area is determined by the size of the electron localization region. This conclusion is confirmed for atomic systems [21,24]: After replacement of a by the radius of the first Bohr orbit a_0, S_b coincides with the atomic measure of the pulse electric area S_{at} [24]. Below we will show that the conclusion is also valid for a wider range of quantum systems.

One of the important phenomena of nonlinear optics is laser-induced ionization of micro-objects. For multicycle laser pulses, the character of ionization is determined by the Keldysh parameter [25], which describes in a unified manner the mechanisms of multiphoton ionization and electron tunneling through a potential barrier. For extremely short pulses, for which the concept of a carrier frequency loses its meaning, it is already inappropriate to speak of multiphoton processes, and the meaning of the Keldysh parameter requires clarification; another condition of the Keldysh theory applicability is that the pulse duration should exceed the Kepler period, electron oscillation period in the ground state. According to a recent paper [26], the criterion for the minimum time of photoionization is the Keldysh time, the ratio of the Keldysh parameter to the carrier frequency; for extremely short pulses, both the numerator and the denominator of the ratio are

*nnrosanov@mail.ru

undefined. The importance of the carrier-envelope phase of the pulse in the quantum tunneling processes is underlined, e.g., in Ref. [27].

In this paper, we consider radiation pulses with duration shorter than the Kepler period. Using examples of a 3D-potential well, the zero-range potential model, and a hydrogen atom, we show that the scale of the pulse electric area, determining the probability of ionization, is inversely proportional to the size of the electron localization region. In the two first cases, the analytical consideration is based on the approximation of sudden perturbations [28], while in the third problem, the direct numerical solution of the time-dependent Schrödinger equation is used.

II. 3D-POTENTIAL WELL

A model of *a centrally symmetric potential well* can describe an isolated quantum dot [29]; for relatively long, femtosecond pulses, quantum dots' photoionization was considered in review (Ref. [30]) and references therein. In the absence of the radiation pulse, the solution of the unperturbed Schrödinger equation shows a finite number of discrete energy levels, depending on the depth of the well width a and depth (ionization potential) U_0 [31]. The potential is written as

$$U(r) = -U_0 \quad (r < a), \quad U(r) = 0 \quad (r > a),$$
$$r = (x^2 + y^2 + z^2)^{1/2}. \tag{1}$$

The radial part of the wave function of an electron with angular momentum $l = 0$ for the ground state (with the lowest energy $E_{w,0}$) inside ($r < a$) and outside ($r > a$) the well has the form

$$\begin{aligned} \psi(r) &= A_0 \sin(kr)/r \quad (r < a),\\ \psi(r) &= A_\infty \exp(-\kappa r)/r \quad (r > a),\\ k &= (1/\hbar)[2m(U_0 + E_{w,0})]^{1/2},\\ \kappa &= (1/\hbar)(-2mE_{w,0})^{1/2}. \end{aligned} \tag{2}$$

Normalization and continuity conditions for the wave function and its derivative at $r = a$ carry out to the relations

$$\begin{aligned} A_\infty &= A_0 \exp(\kappa a)\sin(ka),\\ A_0^{-2} &= 2\pi a\left[1 - \frac{\sin(2ka)}{2ka} + \frac{1}{\kappa a}\sin^2(ka)\right]. \end{aligned} \tag{3}$$

The transcendental equation, which determines the energy levels, also follows from them. The minimum well depth at which a discrete level appears is $U_{0,\min} = \pi^2\hbar^2/(8ma^2)$ [31]. The discrete level will be the only one up to the well depth $U_{0,1} \approx 10.1\hbar^2/(ma^2)$.

The approximation of sudden perturbations [28] is valid only if the duration of the perturbation (in the case under consideration, a radiation pulse) is much shorter than the characteristic period of variation of the unperturbed system (here $2\pi\hbar/E_{w,0}$). If this condition is not met, the intrinsic evolution of the unperturbed system during the time of the perturbation is essential. Within the approximation of sudden perturbation, an extremely short radiation pulse leads to an instantaneous change in the electron momentum, so that the wave functions before, $\psi^{(-)}$, and after, $\psi^{(+)}$, the pulse action are related by the relation $\psi^{(+)} = \psi^{(-)}\exp(-ieS_E z/\hbar)$ (in the electric dipole approximation). Therefore, the amplitude of the probability of conservation of the ground state with the wave function ψ_0 after the action of the pulse is

$$\begin{aligned} a_{00} &= \iiint |\psi_0(r)|^2 \exp(-iqz) r^2 \sin\theta\, dr d\theta d\varphi\\ &= 4\pi\int_0^\infty |\psi_0(r)|^2 \frac{\sin(qr)}{qr} r^2 dr\\ &= \frac{4\pi}{qa}A_0^2 a\left[\frac{1}{2}\mathrm{si}(qa) - \frac{1}{4}\mathrm{si}(qa - 2ka) - \frac{1}{4}\mathrm{si}(qa + 2ka)\right.\\ &\quad \left. + \exp(2\kappa a)\sin^2(ka)\mathrm{Im}\mathrm{E1}(2\kappa a - iqa)\right]. \end{aligned} \tag{4}$$

Here θ and φ are the polar and azimuthal angles, $q = eS_E/\hbar$, and si and E1 are integral sine and exponential integrals [32]. The probability of maintaining the ground state $w_0 = |a_{00}|^2$. The probability of ionization in the above range of well depths, in which the discrete level remains the only one, $w_{\text{ion}} = 1 - w_0 = 1 - |a_{00}|^2$. For large values of the parameter $qa = S_E/S_w$ ($qa \gg 1,\ qa \gg ka,\ qa \gg \kappa a$), the form of Eq. (4) is simplified:

$$\begin{aligned} a_{00} &\approx 4\pi A_0^2 a[ka\sin(2ka) + 2\sin^2(ka)(1 + \kappa a)]\\ &\quad \times \frac{\sin(S_E/S_w)}{(S_E/S_w)^3}, \quad S_w = \hbar/(ea). \end{aligned} \tag{5}$$

The dependence of the probability of excitation and ionization on the pulse electrical area is shown in Fig. 1. The characteristic scale of the change in the electrical area is the value S_w, in accordance with the estimation. It can be seen that with increase in the electrical area, the excitation probability increases and rapidly tends to 1. Note that this conclusion is also valid for a greater depth of the well, when there are several discrete levels in it, due to fast oscillations of the factor $\exp(-iqz)$ in integrals of type (4) at large qa. Equation (5) and Fig. 1 show also the presence of oscillations in the given dependences, and the population zeros correspond to the complete depletion of the ground state and full ionization.

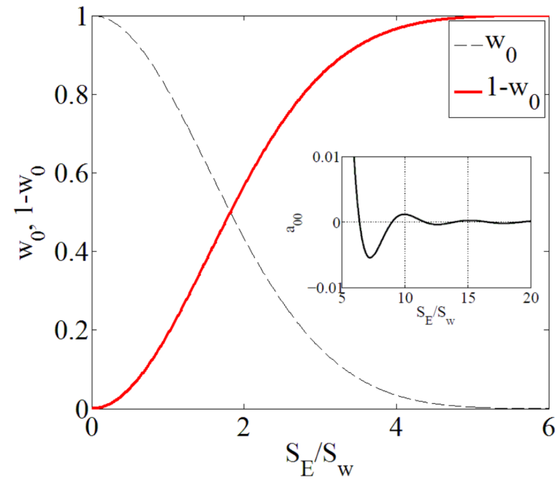

FIG. 1. Dependence of probability of nonexcitation (dashed line) and ionization (solid line) of a spherical wall on the pulse electric area. Parameters: $a = 1.25 \times 10^{-7}$ cm, $U_0 = 3.95 \times 10^{-13}$ erg, $S_w = \hbar/(ea) = 1.76 \times 10^{-11}$ ESU.

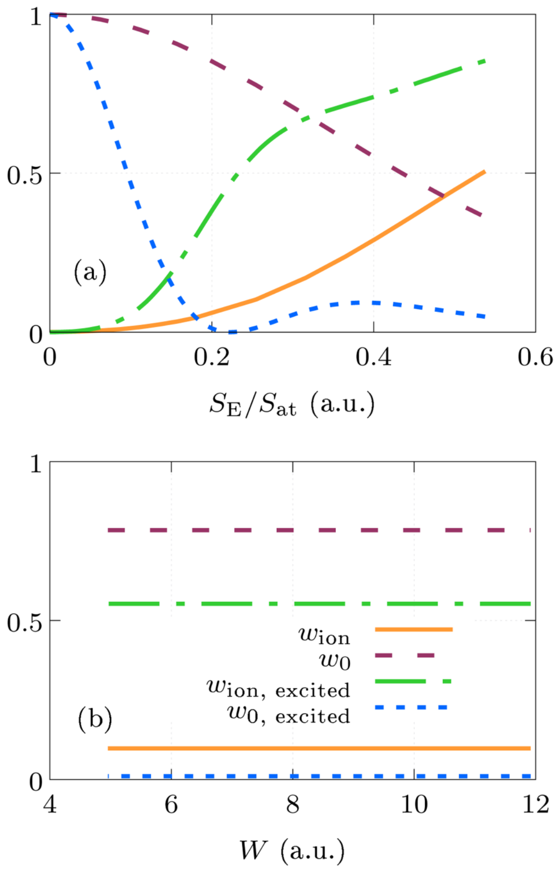

FIG. 2. Nonexcitation and ionization probabilities for ground (w_0, w_{ion}) and excited ($2p$, $w_{0,excited}$, $w_{ion,excited}$) states of atom H after the interaction with a radiation pulse with (a) fixed energy density $W = 12$ a.u. (atomic units) and (b) fixed electric area $S_E = 0.25\ S_{at}$. Pulse duration $\tau = 7.4$ as (0.3055 a.u.), its "frequency" $\omega = 18.225$ a.u.

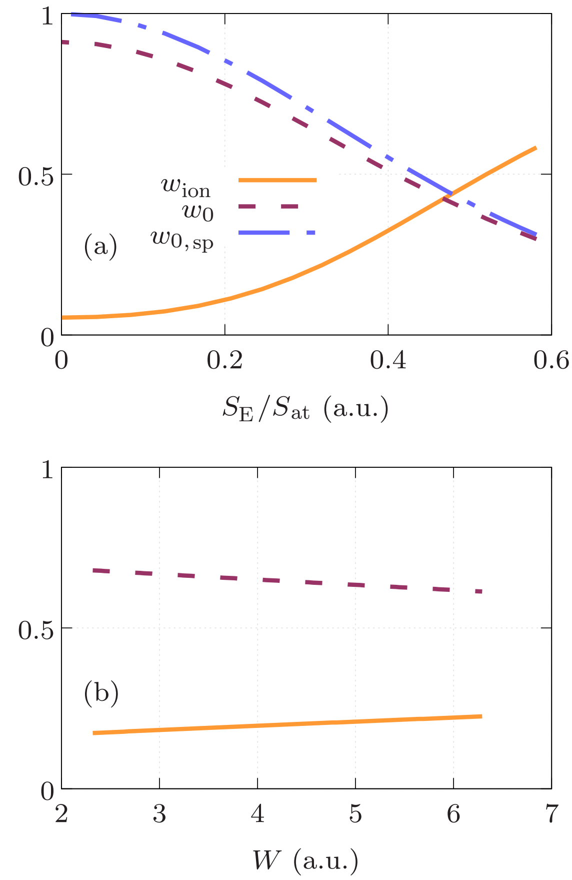

FIG. 3. The same as in Fig. 2 for the initial $1s$ state, pulse duration $\tau = 50$ as (2.067 a.u.), and $\omega = 0.09$ a.u. (a) $W = 4$ a.u.; (b) $S_E = 0.3\ S_{at}$; $w_{0,sp}$ is the nonexcitation probability according to Eq. (10).

III. ZERO-RADIUS POTENTIAL MODEL

The *zero-radius potential model*, which consists of replacing the electron-confining potential with a delta function of coordinates, was initially used in nuclear physics [33], and then in a wide range of problems in atomic physics [34]. In this case, there is only one level of the discrete spectrum with the wave function

$$\psi_0 = (\alpha/2\pi)^{1/2} \exp(-\alpha r)/r. \tag{6}$$

The parameter α can be expressed in terms of the electron affinity. The probabilities of nonexcitation and ionization by a subcycle radiation pulse [35] can be written in the following form:

$$\begin{aligned} w_0 &= a_{00}^2, \quad w_{ion} = 1 - w_0, \\ a_{00} &= (S_0/S_E)\arctan(S_E/S_0). \end{aligned} \tag{7}$$

In atomic units, the characteristic value of the electric area $S_0 = 2\alpha$. Since the size of the electron localization region is inversely proportional to the parameter α, this confirms the above conclusion about the scale of electric area of subcycle pulses.

IV. ATOM H

Let us present next the results of a direct numerical solution of the time-dependent Schrödinger equation (without using the approximation of sudden disturbances) for a pulse with an electric strength profile

$$E(t) = E_0 \exp(-t^2/\tau^2)\sin(\omega t + \varphi_0). \tag{8}$$

The corresponding electric area S_E and the quantity $W = \int E^2(t)\,dt$ proportional to the pulse energy density have the form

$$\begin{aligned} S_E &= \sqrt{\pi} E_0 \tau \exp[-(\omega\tau)^2/4] \sin\varphi_0, \\ W &= \sqrt{\pi/8} E_0^2 \tau \{1 - \exp[-(\omega\tau)^2/4] \cos(2\varphi_0)\}. \end{aligned} \tag{9}$$

The generalized pseudospectral method [36,37] was used for the solution. Figure 2 shows the dependence of the probabilities of maintaining the ground state $1s$ and ionization for the ground and excited state $2p$ on the electric area S_E and energy density W for extremely short pulses for two scenarios. In the first scenario [Fig. 2(a)], the energy density is constant, and in the second one [Fig. 2(b)] the pulse electric area S_E is constant, which was achieved by simultaneous variation of the peak electric strength E_0 and the initial phase φ_0. The pulse duration τ was fixed in both cases: $\tau = 7.4$ as; the maximum peak field amplitude corresponds to intensity about

10^{19} W/cm^2. The figures indicate that it is the scaled electric area of extremely short pulses S_E/S_{at} that determines the probability of their ionization. Note also that the dependence of $w_0(S_E)$ in Fig. 2(a) found numerically in the case of ionization from the ground state coincides with graphic accuracy with that given by the formula [21]

$$w_{0,\text{sp}} = [1 + (S_E/S_{\text{at}})^2]^{-4}. \quad (10)$$

This confirms the applicability of the sudden perturbation approximation; see also Ref. [20]. The probability of ionization complements the probabilities of excitation of all levels of the discrete spectrum to unity.

In Fig. 3 we present similar results for pulses, the duration of which, $\tau = 50$ as, is still shorter, but already comparable to the time of an electron's revolution in the Bohr orbit. Figure 3(a) shows that in this case also the approximation of sudden perturbation turns out to be quite accurate: the discrepancy between the analytical and numerical results when determining the probability of atom nonexcitation is less than 10%. However, in this case, a weak dependence of the probabilities of excitation and ionization on the pulse energy appears; see Fig. 3(b).

The above examples confirm the decisive role of the subcycle pulse electric area with the scale determined by the size of electron localization region in the photoionization yield of quantum objects. The Keldysh parameter of theory of ionization $\gamma_K = \omega\sqrt{2mI_0}/(eE_0)$ [25], where ω is the frequency of an electromagnetic wave with amplitude E_0, and I_0 is the ionization potential, is not applicable for purely unipolar pulses with the central frequency $\omega = 0$. In Ref. [26], the value of the Keldysh time $\tau_K = \gamma_K/\omega$ is introduced. For a hydrogen atom $I_0 = me^4/(2\hbar^2)$ and for a pulse with duration τ_K, we obtain $S_E = S_{\text{at}}$. Thus, the efficiency of not only excitation, but also ionization of atoms by extremely short pulses is determined by the ratio S_E/S_{at}.

V. CONCLUSION

Thus, we have studied ionization of very different types of three-dimensional quantum objects with number of discrete spectrum states from one to infinity, driven by few- and subcycle pulses with duration smaller than quantum wave-packet oscillation period of the object. We show that ionization yield is determined mainly by ratio of the pulse electric pulse area S_E to its characteristic value $S_{\text{quant}} = \hbar/(ea)$ inversely proportional to the size of electron localization region a. In most previous studies, interaction of pulses with duration of hundreds of attoseconds or longer was studied; see, e.g., Refs. [2,3,9,11,38]. Their duration is longer than the Kepler period of an electron in ground state (150 as for hydrogen atom). Since methods of generation of attosecond and unipolar half-cycle pulses with much shorter duration have been proposed recently [4–7], the results presented above can establish theoretical background for the study of interaction of such pulses with various quantum objects.

ACKNOWLEDGMENTS

The research was supported by the Russian Science Foundation, Project No. 21-72-10028 (quantum systems' control by unipolar subcycle pulses), and the Russian Foundation for Basic Research, Project No. 19-02-00312 (ionization of quantum wells and atom H).

[1] https://eli-laser.eu/.
[2] F. Krausz and M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).
[3] J. Biegert, F. Calegari, N. Dudovich, F. Quéré, and M. Vrakking, J. Phys. B: At. Mol. Opt. Phys. **54**, 070201 (2021).
[4] Zhiyi Wei, J. Hebling, and K. Varjú, J. Opt. Soc. Am. B **35**, AST1 (2018).
[5] T. Gaumnitz, A. Jain, Y. Pertot, M. Huppert, I. Jordan, F. Ardana-Lamas, and H. J. Wörner, Opt. Express **25**, 27506 (2017).
[6] R. M. Arkhipov, M. V. Arkhipov, and N. N. Rosanov, Quantum Electron. **50**, 801 (2020).
[7] Y. Shou, R. Hu, Z. Gong, J. Yu, Jia erh Chen, G. Mourou, X. Yan, and W. Ma, New J. Phys. **23**, 053003 (2021).
[8] N. N. Rosanov, Opt. Spectrosc. **127**, 1050 (2019).
[9] M. T. Hassan, T. T. Luu, A. Moulet, O. Raskazovskaya, P. Zhokhov,M. Garg, N. Karpowicz, A. M. Zheltikov, V. Pervak, F. Krausz, and E. Goulielmakis, Nature (London) **530**, 66 (2016).
[10] J. Xu, B. Shen, X. Zhang, Y. Shi, L. Ji, L. Zhang, T. Xu, W. Wang, X. Zhao, and Z. Xu, Sci. Rep. **8**, 2669 (2018).
[11] H. C. Wu and J. Meyer-ter-Vehn, Nat. Photonics **6**, 304 (2012).
[12] M. V. Tsarev and M. I. Bakunov, Opt. Express **27**, 5154 (2019).
[13] A. V. Bogatskaya, E. A. Volkova, and A. M. Popov, Phys. Rev. E **104**, 025202 (2021).
[14] E. G. Bessonov, Sov. Phys. JETP **53**, 433 (1981).
[15] N. N. Rosanov, R. M. Arkhipov, and M. V. Arkhipov, Phys.-Usp. **61**, 1227 (2018).
[16] N. N. Rosanov and N. V. Vysotina, JETP **130**, 52 (2020).
[17] P. H. Bucksbaum, in *Fourteenth International Conference on Atomic Physics 14*, edited by D. J. Wineland, C. E. Wieman, and S. J. Smith, AIP Conf. Proc. No. 323 (AIP, New York, 1994), p. 416.
[18] D. Dimitrovski, E. A. Solov'ev, and J. S. Briggs, Phys. Rev. Lett. **93**, 083003 (2004).
[19] D. Dimitrovski, E. A. Solov'ev, and J. S. Briggs, Phys. Rev. A **72**, 043411 (2005).
[20] A. Lugovskoy and I. Bray, Eur. Phys. J. D **69**, 271 (2015).
[21] N. N. Rosanov, Opt. Spectrosc. **124**, 72 (2018).
[22] R. M. Arkhipov, A. V. Pakhomov, M. V. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, and N. N. Rosanov, Opt. Lett. **44**, 1202 (2019).
[23] R. Arkhipov, A. Pakhomov, M. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, and N. Rosanov, Sci. Rep. **11**, 1961 (2021).
[24] R. M. Arkhipov, M. V. Arkhipov, A. V. Pakhomov, and N. N. Rosanov, JETP Lett. **114**, 129 (2021).
[25] L. V. Keldysh, Sov. Phys. JETP **20**, 1307 (1965).
[26] A. M. Zheltikov, Opt. Lett. **46**, 989 (2021).
[27] S. Kim, T. Schmude, G. Burkard, and A. S. Moskalenko, New J. Phys. **23**, 083006 (2021).

[28] A. B. Migdal, Zh. Éksp. Teor. Fiz. **9**, 1163 (1939).

[29] V. Bonacic-Koutecky, P. Fantucci, and J. Koutecky, Chem. Rev. **91**, 1035 (1991).

[30] L. Seiffert, S. Zherebtsov, M. F. Kling, and T. Fennel, arXiv:2109.02367 [physics.optics].

[31] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics. Nonrelativistic Theory* (Pergamon Press, Oxford, 1965).

[32] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1965).

[33] E. Fermi, Ric. Sci. **2**, 13 (1936).

[34] Yu. N. Demkov and V. N. Ostrovsky, *Zero-Range Potentials and Their Application in Atomic Physics* (Plenum Press, New York, 1988).

[35] T. P. Grozdanov and J. Jaćimović, Phys. Rev. A **79**, 013413 (2009).

[36] G. Yao and S.-I. Chu, Chem. Phys. Lett. **204**, 381 (1993).

[37] D. A. Telnov and Shih-I. Chu, Phys. Rev. A **59**, 2864 (1999).

[38] J. T. Karpel and D. D. Yavuz, Opt. Lett. **43**, 2583 (2018).