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Semiclassical theory of laser-assisted dissociative recombination

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Abstract

We study the process of laser-assisted dissociative recombination of an electron with a molecular cation using a semiclassical approach. In the region outside a reaction sphere the electron motion in the combined laser and Coulomb fields is treated classically. Within the sphere the laser-field effects are neglected, and the recombination probability is obtained from quantum-mechanical cross sections calculated for the laser-free process. Specific calculations are performed for dissociative recombination of H_2^+ in the field of the intensity 2.09 GW/cm² and the wavelength 22.8 μ m. In the energy region above 1 meV the cross section is significantly enhanced, as compared to the field-free case, due to the Coulomb focusing effect. The influence of the indirect process due to electron capture into Rydberg states is also investigated. Although the Rydberg resonances are washed out due to the field effects, they influence significantly the magnitude of the dissociative recombination cross section.

I. INTRODUCTION

Electron scattering processes can be controlled by external fields. In particular the lowenergy electron bremsstrahlung and radiative recombination processes can be strongly enhanced by placing electron beam in an infrared field of a moderate intensity (about $10^9 - 10^{12}$ W/cm²) [1, 2]. This effect occurs due to the Coulomb focusing [3]: a slow electron can perform many oscillations in the ac field during which it approaches closely to the Coulomb center. This makes bremsstrahlung and radiative recombination possible even at large impact parameters corresponding to large initial angular momenta, whereas ordinary these processes occur at low angular momentum of the incident electron. In the present paper we consider the dissociative recombination (DR) process

$$e + AB^+ \to A + B$$

which typically occurs through the formation of indermediate resonance states of the neutral molecule. Another mechanism involves capture into a Rydberg state of AB with the subsequent predissociation by the state A + B.

DR is the main mechanism of destruction of electrons and molecular cations in the Early Universe [4], in the interstellar molecular clouds [5, 6] and in the planetary ionospheres [7, 8]. Moreover, it is often present in various media of technological or industrial interest: the plasma formed at the boundary layer of a spacecraft entering the ionosphere of planets [9, 10], the edge plasma at the wall of the thermonuclear controlled fusion devices [11] and the plasma used for etching and implantation in semiconductor industry [12]. A possible influence of external fields on DR processes has been discussed in [13, 14].

Similar to the laser assisted processes discussed above, if the molecular ion AB^+ is placed in an ac field, the Coulomb focusing can bring the electron close to the molecular target, making the process possible even at large impact parameters and increasing the cross section substantially. Below we will develop a semiclassical theory of this process.

II. THEORY

Our theory of laser-assisted dissociative recombination (LADR) is based on the division of the electron configuration space into two regions separated by the reaction sphere of radius r_0 . Outside this sphere the electron-ion interaction is approximated by the Coulomb potential, and the electron motion in the combined Coulomb and ac fields is treated classically. A typical polarizability of a simple molecular ion is about a few a.u. For example, the polarizability of H_2^+ is 3.17 a.u. [15], and therefore r_0 should be greater than about 4 to 5 a.u. Inside the sphere we neglect the laser field as compared to the Coulomb interaction, and use quantum-mechanical approach to find the probability of DR within this region. After neutral fragments are formed, the laser field effects are negligible, and the fragments separate as in the laser-free case.

DR cross sections calculated this way will weakly depend o r_0 due to the conservation of the angular momentum within the reaction sphere as long as the influence of the laser field within the sphere can be neglected. The corresponding condition, $r_0^2 \ll 1/F$, can be satisfied by $r_0 < 30$ a.u. for the electric field stength lower than about 0.0005 a.u. (intensity lower than 9 GW/cm²). This is sufficient for description of the direct process. However, Rydberg states with the principal quantum numbers n > 5 will not fit within this radius, therefore the indirect process involving these states will not be adequately described by this model. We will initially assume that contribution of these states can be neglected, and will discuss their possible influence after presentations of the results.

We use the electric dipole approximation so that the force on the electron from the laser field writes

$$F = F_0 \cos(\omega t + \phi_0)$$

where F_0 is the force amplitude, ω is the frequency, and ϕ_0 is a constant phase. Like in our previous calculations on the laser-assisted bremsstrahlung [1] and the laser-assisted radiative recombination [2], we consider a geometry in which the incident electron velocity is parallel to the electric field (directed along the x axis). Then the classical electron motion is planar, and in the region where the field of the molecular ion can be neglected, the x-component of the electron velocity is

$$v_x = v_0 + \frac{F_0}{\omega} [\sin(\omega t + \phi_0) - \sin(\phi_0)].$$
(1)

The average velocity outside the Coulomb zone is

$$\bar{v}_x = v_0 - \frac{F_0}{\omega}\sin(\phi_0),$$

and the Coulomb focusing effect is most efficient when \bar{v}_x is close to 0. If $v_0 < F_0/\omega$ then

there are two values of the angle ϕ_0

$$\phi_1 = \arcsin \frac{v_0 \omega}{F_0}, \quad \phi_2 = \pi - \phi_1 \tag{2}$$

for which $\bar{v}_x = 0$.

The cross section should be averaged over ϕ_0 which is equivalent to averaging over the electron position at the instant when it enters the field region [16]. This is valid if this position is far enough from the molecular ion so that electron-ion interaction can be neglected as compared to the electron-field interaction.

The LADR cross section is calculated as the integral over the impact parameter b and average over ϕ_0

$$\sigma = \frac{1}{2\pi} \int \sigma(\phi_0) d\phi_0,$$

$$\sigma(\phi_0) = 2\pi \int P[l(b,\phi_0), E(b,\phi_0)] b db$$
(3)

where P(l, E) is the probability of DR for electron having angular momentum l and energy E when it approaches the separation sphere. Since the electric field within the sphere can be neglected, it cannot change l and E, and therefore for the function P(l, E) we use the field-free quantum-mechanical probability. Typically the resonance capture is dominated by one partial wave $l = l_r$. For example, low-energy DR of H_2^+ dominated by l = 2, although a contribution from l = 0 is not negligible. The quantum-mechanical DR cross section is

$$\sigma_{DR} = g \frac{\pi}{2E} \sum_{l} |S_{dl}(E)|^2 \tag{4}$$

where g is the statistical weight of the electron scattering channel, and $S_{dl}(E)$ is the matrix element for transition from the electron scattering channel to the dissociating channel. As was discussed above, the sum is dominated by one or two terms. For the transition probability we therefore have

$$P(l,E) = \frac{2E}{\pi}\sigma_l(E) \tag{5}$$

where $\sigma_l(E)$ is the partial cross section

$$\sigma_l(E) = g \frac{\pi}{2E} |S_{dl}(E)|^2.$$

If partial cross sections are known from quantum-mechanical calculations, the cross section in the presence of the laser field can be calculated by combining Eqs. (3) and (5). Note that the angular momentum l as a function of the impact parameter b and the phase ϕ_0 , $l(b, \phi_0)$, is a classical quantity whereas quantum P(l, E) is defined for the quantized angular momentum l_r where l_r is an integral value of angular momentum allowed by the symmetry of the resonance. Therefore we make P(l, E) nonzero if classically calculated angular momentum l satisfies the condition $l_r - 0.5 < l < l_r + 0.5$.

III. LADR OF H_2^+

A. Model Calculations

For calculations we have chosen DR of H_2^+ . The process occurs efficiently at low energies below 0.1 eV due to electron capture into the doubly excited resonant state $(2p\sigma_u)^2$ which is dominated by the *d* wave, although some admixture of the *s* wave is also present [17, 18]. At collision energies above 1 eV other resonant states contribute to the DR cross section. However, in this paper we are mostly interested in the region below 1 eV where the cross section is large (above about 10^{-16} cm²). The cross section for the direct process in this energy range is roughly inversely proportional to the incident electron energy, but it is also affected by the indirect mechanism producing window resonances [18]. To analyze the LADR due to the direct process, we have chosen calculations of Takagi *et al* [19] from which we conclude that on the average the DR cross section can be well described by the following probability as a function of energy for $l_r = 2$

$$P(E) = 0.0030 - 0.0022E \tag{6}$$

where E is the electron energy in eV. We assume for simplicity that the probability is 0 for any other angular momentum l and for E > 1.36 eV. The physical reason for the decrease of P with E is autoionization which leads to an additional factor in P called the survival probability [17]. An additional consideration is required for E < 0. Due to the energy transfer from the electric field the electron can gain energy, but also lose energy when it approaches the molecular ion. In the case E < 0 the resonant state becomes stable, and electron capture occurs with 100% survival probability. Although the capture probability can be still energy dependent, it is reasonable to assume P(E) = P(0) for E < 0 if |E| is small compared to the Coulomb interaction in the reaction region.

In what follows we will consider DR in an infrared field with intensity $I = 2.09 \text{ GW/cm}^2$ ($F_0 = 0.000244 \text{ a.u.}$) and the wavelength 22.8 μ m ($\omega = 0.002 \text{ a.u.}$). The critical phases



FIG. 1. Critical phases, Eq. (2) as functions of the electron velocity v_0 for $F_0 = 0.000244$ a.u., $\omega = 0.002$ a.u..

 ϕ_1 and ϕ_2 , Eq. (2), for which the Coulomb focusing is most efficient, are plotted in Fig. 1. In the vicinity of critical phases the Coulomb focusing can occur for an arbitrary large value of the impact parameter, therefore the LADR cross section becomes infinite. Like in the case of laser-assisted radiative recombination [2], we limit the cross section by a finite duration τ of the laser pulse. For our calculations we have chosen τ in the range 5-10 ps. For $v_0 > F_0/\omega = 0.122$ a.u. the Coulomb focusing effect becomes weaker, and the LADR cross section is expected to drop.

In Fig. 2 we present the DR probability as a function of the impact parameter b for the electron velocity $v_0 = 0.1$ a.u. and the phase $\phi_0 = 1.12$ rad. Note that in this case the x-component of the electron velocity outside the Coulomb zone varies according to Eq. (1) with the average velocity $\bar{v}_x = -0.00981$ a.u. and the oscillation amplitude 0.122 a.u. Since \bar{v}_x is small, the Coulomb focusing effect is strong, and the nonzero probability can be found at impact parameters b as large as 1400 a.u. There is no regular pattern in the dependence reflecting the chaotic nature of scattering in combined laser and Coulomb fields [20-22]. Moreover, the P(b) dependence exhibits a fractal structure which is demonstrated in lower panels with enlarged scales.

In Fig. 3 we present the impact parameter dependence of the closest approach r_{cl} . In this calculation the scattering center was modeled as a point Coulomb potential -1/r, although the actual potential in the $e-H_2^+$ problem at $r < r_0$ is different. The graph just serves to



FIG. 2. DR probability as a function of the impact parameter b for the model problem with the field-free probability given by eq. (6), v = 0.1 a.u., $F_0 = 0.000244$ a.u., $\omega = 0.002$ a.u., $\phi_0 = 1.12$. Lower panels are showing progressively enlarged scales in b.

show the range of impact parameters, determined by the condition $r_{cl} < r_0$, which contribute to the DR reaction. Further restriction comes from the condition 1.5 < |l| < 2.5 where lis the projection of the angular momentum on the z axis within the reaction sphere. (We choose xy plane as the plane of motion, therefore $l_x = l_y = 0$). To demonstrate this, in Fig. 4 we present l as a function of the impact parameter. Positions of peaks and dips of |l|correlate with those of r_{\min} meaning that low values of |l|, necessary for the DR process to occur, correspond to close collisions.

Fig. 5 demonstrates the chaotic and fractal dependence of the electron energy within the reaction zone on b. Irregularities presented in Figs. 3, 4, and 5 are responsible for the chaotic behavior of the DR probability shown in Fig. 2

Finally in Fig. 6 we present the LADR cross section in the energy range where the Coulomb focusing effect is important. The cross section remains high up to the critical



FIG. 3. The distance of the closest approach of electron to the Coulomb center as a function of b for the same field parameters as in Fig. 2

velocity $v_{cr} = F_0/\omega = 0.122$ a.u., and peaks at even higher velocity $v_0 = 0.126$ a.u., but then drops sharply. It remains substantially higher than the field-free cross section for energies up to 2 eV. Note, however, that we are talking about the energy (or velocity) of the electron before entering the field region. After entering the field, the electron gets the additional ponderomotive energy which amounts to 101.2 meV in our case.

A dip in the velocity region $v_0 = 0.20$ a.u. (E = 544 meV) can be explained by a pecularity of the electron motion in the combined field in this region: instead of the Coulomb



FIG. 4. z-projection of the electron angular momentum within the reaction sphere as a function of b for the same field parameters as in Fig. 2

focusing many trajectories exhibit a "defocusing effect". This is demonstrated in Fig. 7 where we present two trajectories for the same impact parameter but two different initial velocities corresponding to energies 0.306 and 0.544 eV. At higher energies the focusing effect is restored, and this effect shows up as a shoulder in the cross section at about 0.850 eV.



FIG. 5. The electron energy within the reaction sphere as a function of b for the same field parameters as in Fig. 2

B. Inclusion of indirect process in *ab initio* calculations

Generally DR can be affected by the indirect mechanism [17, 18], electron capture into a Rydberg state supported by a vibrationally excited molecular ion with subsequent predissociation into the valence state. Since the size of the Rydberg state is large, the radius of the separation sphere used in our LADR calculations should be increased. This decreases the range of the field intensities for which our theory is valid. However, for the field intensity chosen above (2.09 GW/cm²) we can still vary r_0 in a wide range. Our calculations have



FIG. 6. The DR cross section as a function of electron energy for the model problem with the field-free probability (6) and the same field parameters as in Fig. 2.

shown that the increase of r_0 from 5 to 20 a.u. affects very little LADR cross sections. Therefore capture into Rydberg states with the principal quantum number n up to 4 can be safely included in our model. For calculations of laser-free DR incorporating both direct and indirect processes we used updated techniques [23] as outlined in the next subsection.

1. Laser-free DR calculations

In the present study we restrict ourselves to the case where the rotational excitation and the rotational couplings during the collision are neglected, and the lowest dissociative state of capture of ${}^{1}\Sigma_{q}^{+}$ symmetry is available only.

The Multichannel Quantum Defect Theory (MQDT) approach starts with the building of the interaction matrix \mathcal{V} , performed in the so-called 'A-region' of the configuration space [17, 18, 24], where the Born-Oppenheimer approximation is appropriate for the description of the collision system. In this region, the electronic states belonging to an ionization channel associated with a partial wave l of the optical electron may be characterized with respect to



FIG. 7. Electron trajectories for two values of initial velocity, $v_0 = 0.15$ and 0.20 a.u. (energies 0.306 and 0.544 eV respectively), for impact parameter b = 199 a.u. Both trajectories start at (x, y) = (-300, 199). The reaction sphere is schematically labeled by the circle with the center at the origin. The second trajectory exhibits a "defocusing" effect.

hydrogenic states in terms of the quantum defect μ_l dependent on the internuclear distance R but assumed to be independent of energy. An ionization channel associated with the vibrational state of quantum number v is *electronically* coupled to the dissociation channel d through an R-dependent scaled 'Rydberg-valence' interaction term, $V_{dl}^{(el)}$, assumed to be, as the quantum defect μ_l , independent of the energy of the electronic state:

$$V_{dl}^{(el)} = \langle \Phi_d^{(el)} | H_{el} | \Phi_l^{(el)} \rangle , \qquad (7)$$

where H_{el} denotes the electronic Hamiltonian, $\Phi_d^{(el)}$ is the electronic wave function of the dissociative state, $\Phi_l^{(el)}$ is the electronic wave function describing the neutral molecular system "electron + ion", and the scalar product involves integration over the electron coordinates only. Eventually, for a given value of the total energy $\mathcal{E} = E + E_{v_0}$ of the system (*E* is the incident electron energy, and E_{v_0} is the energy of the initial vibrational state v_0), the convolution of the electronic coupling $V_{dl}^{(el)}$ with the local Franck-Condon factor provides the elements of the interaction matrix \mathcal{V} :

$$\mathcal{V}_{dlv}(E) = \langle F_d(E) | V_{dl}^{(el)} | \chi_v \rangle, \tag{8}$$

 $F_d(E)$ and χ_v being the internuclear wave-functions corresponding to the dissociative state d and to the ionization channel associated with the state v, respectively.

The interaction matrix \mathcal{V} , whose elements are given by (8), on one hand, and the zeroorder Hamiltonian, in which the Rydberg-valence interaction is neglected, on the other hand, allow the building of the reaction \mathcal{K} -matrix, corresponding to the total energy \mathcal{E} , which satisfies the Lippmann-Schwinger equation [17]. In order to express the effect of the short-range interaction in terms of phase-shifts, a unitary transformation of the initial basis into a new one, corresponding to eigenchannels is performed, *via* the diagonalization of this reaction matrix .

In the external 'B-region' [17, 18, 24], characterized by large electron-core distances, the Born-Oppenheimer model is no longer valid for the neutral molecule, Λ ceases to be a good quantum number, and a frame transformation [17–19] is performed, *via* the projection coefficients:

$$\mathcal{C}_{l\nu,\alpha} = \sum_{\nu'} U_{l\nu',\alpha} \langle \chi_{\nu} | \cos(\pi \mu_l + \eta_{\alpha}) | \chi_{\nu'} \rangle \quad \mathcal{C}_{d,\alpha} = U_{d\alpha} \cos \eta_{\alpha} , \qquad (9)$$

and, replacing cos by sin, $S_{lv,\alpha}$ and $S_{d,\alpha}$. Here α are indices for the eigenchannels, $U_{lv',\alpha}$ and $U_{d\alpha}$ the elements of the α -eigenvector, and η_{α} the phaseshift associated to the α -eigenvalue of the \mathcal{K} -matrix.

The matrices C, whose elements are given by (9), and S, are subsequently used in building the generalized scattering matrix X, in which all the channels, open ('o') and closed ('c'), are represented, and which consists in 4 sub-matrices:

$$\boldsymbol{X} = \frac{\boldsymbol{\mathcal{C}} + i\boldsymbol{\mathcal{S}}}{\boldsymbol{\mathcal{C}} - i\boldsymbol{\mathcal{S}}}, \qquad \boldsymbol{X} = \begin{pmatrix} X_{oo} & X_{oc} \\ X_{co} & X_{cc} \end{pmatrix}.$$
(10)

The boundary conditions - 'elimination' of the closed channels - results in the *physical* scattering matrix S [25]:

$$\boldsymbol{S} = \boldsymbol{X}_{oo} - \boldsymbol{X}_{oc} \frac{1}{\boldsymbol{X}_{cc} - \exp(-i2\pi\boldsymbol{\nu})} \boldsymbol{X}_{co}, \qquad (11)$$

Here the diagonal matrix ν is formed with the effective quantum numbers (using atomic units for energy)

$$\nu_v = [2(E_v - \mathcal{E})]^{-1/2}, \qquad (12)$$

associated with each *closed* channel, i.e. to each vibrational threshold E_v of the ion *larger* than the total energy \mathcal{E} .

The cross section of DR of an electron of energy E into the dissociative state d writes similar to Eq. (4) where we specify now the initial vibration state v of H_2^+ :

$$\sigma_{DR,v}(E) = \sum_{l} \sigma_{l,v}(E) = g \frac{\pi}{2E} \sum_{l} |S_{dl,v}(E)|^2 , \qquad (13)$$

where

$$\sigma_{l,v}(E) = g \frac{\pi}{2E} |S_{dl,v}(E)|^2 , \qquad (14)$$

is the partial cross section associated to the *l*-partial wave of the incident electron. In specific calculations we have chosen the initial state to be the ground vibrational state v = 0.

2. Results and discussion

As mentioned above we have chosen the version of *ab initio* DR calculations which includes indirect process of capture into vibrationally excited Rydberg states, but does not include the rotational structure of the target's energy spectrum. Inclusion of rotational structure creates many additional resonances which are smeared out by the field effects and do not affect the magnitude of LADR cross sections. In contrast, Rydberg resonances supported by vibrational states of H_2^+ , although also smeared out by the field effects, influence the magnitude of cross section substantially.

In Fig. 8 we present the field free and LADR cross sections. Because of the wiggling motion of electron in the laser field, the resonance structure is smeared out, but overall we observe a substantial increase of the cross section except in the region of ultralow energies below 1 meV where the laser-free cross section continues to grow towards lower energies whereas LARD cross section remains close to 800×10^{-16} cm². It is apparent that, due to the wiggling electron motion, the field-free 1/E singularity in the cross section no longer exists.

The defocusing effect discussed in the previous section does not depend on specific values of the field-free cross section, therefore it manifests itself in the form of a shoulder in the same energy region as in the model calculations shown in Fig. 6.



FIG. 8. The DR cross section as a function of electron energy for a field-free case and the laserasisted case for the same field parameters as in Fig. 2, with the indirect process included. Solid (purple) line: field-free results. LADR results are for the laser pulse duration 5 ps, $z_0 = 500$ a.u. and $r_0 = 10$ a.u. Dashed (green) line: calculations assuming that only d electrons are captured. Dotted (red) line: calculations including capture of both s and d electrons.

The first calculation (dashed line) was carried out with assumption that only d electrons are captured by H₂⁺. Calculation of the field-free direct process shows that s-wave capture contributes about 20% to the DR cross section. To estimate the s-wave effect, we assumed that the s-wave contribution and d-wave contribution do not interfere, and have calculated LADR cross section with the s-wave capture cross section being 1/4 of the d-capture cross section. The results (dotted line) change very insignificantly. In particular the peak value of the LADR cross section has been reduced by 11%. This indicates that LADR cross sections are not very sensitive to the partial-wave composition of the resonance. Therefore inclusion of the indirect process, even within the framework of the s + d model, should not affect significantly the accuracy of the results, although Rydberg states can possess angular momenta other than 0 or 2.

The dependence of the cross section on r_0 is negligible when we vary this parameter within

the limits discussed in sec. II. On the other hand, it grows with the laser pulse duration τ , but this growth is slow and almost unnoticeable on the scale of drawing when τ is increased from 5 to 10 ps.

IV. LIMITATIONS OF THE MODEL

The classical treatment of the electron in the outer region implies several approximations most important of which are the assumption that electron kinetic energy is a function of position and the classical treatment of the angular momentum as a continuous quantity. The first assumption is well justified for low-velocity motion in the Coulomb field [26]. The second assumption is the most questionable since only a few discrete angular momenta contribute to the quantum-mechanical cross section, whereas calculation of classical cross section involves integration over the continuous impact parameter. In addressing this problem we note first that, due to the Coulomb focusing effect, the inclusion of the laser field substantially broadens the range of impact parameters contributing to the cross section. Second, even in the case of zero field the approximation of continuous l turns out to be not so severe. To demonstrate this, in Fig. 9 we present the DR cross section calculated from the classical formula, Eq. (3), and compare it with the original quantal cross section. The agreement is perhaps is not as good as it looks because of the logarithmic scale, but typically the discrepancy does not exceed a factor of two which is substantially smaller than the field effect shown in Fig. 8. And again, the approximation of continuous l is much more justified for nonzero fields.

Another limitation of the model has to do with the finite radius of the reaction sphere, $r_0 < 30$ a.u. in the present calculations, which does not fit Rydberg states with *n* higher than 5. Physically this means that these states are influenced by the laser field in the form of the dynamical Stark shift and the laser-induced ionization. The latter phenomenon will suppress the DR process. On the other hand, direct and indirect processes in DR interfere destructively [18], therefore the neglect of Rydberg states with higher *n* should increase the DR cross section. We therefore expect that inclusion of the interaction of higher Rydberg states with the external field will not reduce, at least significantly, the enhancement factor obtained in the present calculations. However, a quantitative investigation of these effects is certainly warranted.



FIG. 9. The DR cross section as a function of electron energy for a field-free case, comparison of the original quantum calculation with the classical result, Eq. (3), obtained from the quantum probability.

V. CONCLUSION

Studies of influence of external fields on collision processes allows us to understand how the rates of field-free processes change in the presence of the field. They also allow us to develop tools for manipulation and control of collision processes. The important process of dissociative recombination has been studied in the present paper. We have shown that infrared laser fields of a moderate intensity can significantly enhance DR cross sections in the low-energy region due to the Coulomb focusing effect. The process has been studied for the parallel geometry whereby the velocity of the incident electron is parallel to the field polarization vector. It seems that the Coulomb focusing effect is most efficient for this geometry, although the other geometric configuration would be also of interest. Future studies of the dependence of the effect on the field intensity and the wavelength would allow us to find the optimal field parameters for controlling the DR process.

Another effect which requires further investigation is the dynamical Stark effect in in-

termediate Rydberg states. The Stark shift and autoionization can influence the indirect process. Although this effect can reduce the probability of indirect process through higher Rydberg states, we anticipate that the strong enhancement due to the Coulomb focusing will not be significantly affected, particularly because in the case of zero field direct and indirect processes interfere destructively.

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