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Intense laser field effects on the binding energy of impurities in semiconductors

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Nonperturbative, accurate variational results for the binding energy of a shallow hydrogenic impurity in a bulk semiconductor irradiated by a high-frequency, intense laser field are presented. It is pointed out that variational results found with $1s$ and $2s$ atomic trial wave functions presented in previous works on this subject are incorrect because these wave functions do not allow the stretching of the electronic cloud along the polarization direction. This is corrected here by choosing appropriate trial wave functions for the ground and first excited states, which resemble those for the $(1s)\sigma$ states of the $H_2{}^+$ molecular ion. Special attention is paid to the limit of large values for the laser-dressing parameter α_0, where our model furnishes almost exact results. In this limit, we found that the binding energy tends to $-1/4\ \mathrm{Ry}^*$ with the increase of α_0. Then, impurity stability against ionization is expected, which is in contrast to previous works.

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With the advent of high-power, tunable, linearly polarized laser sources such as CO_2 and free-electron lasers (FELs), new possibilities have arisen in the study of the interaction of intense laser fields (ILFs) with electrons in semiconductors.[1,2] As a consequence, some important and distinctive phenomena associated with laser-driven systems have been theoretically anticipated and observed, such as the reduction of energy gaps,[3] distortion of optical absorption edges,[4] strong modulation of electron densities of states via the dynamical Franz-Keldysh effect (DFKE),[5] and tuning of the plasmon spectrum.[6] Progress has also been made in investigating the physical properties of impurity states in semiconductors and their behavior when irradiated by ILFs. Since the first work on this subject in the mid-1990s,[7] this topic has gained importance because the laser-driven impurity ionization process is the main source of carriers in modern optoelectronic devices. Moreover, ionized donors and acceptors compose the main carrier scattering mechanism in bulk semiconductors, and this determines their transport properties at low temperatures.[8] Hence, the investigation of the changes in the impurity binding energy as a function of the laser intensity based upon accurate models is relevant for understanding the physically measurable properties of semiconductor systems.

Recently, Nie and co-workers argued that the binding energy of a hydrogenic impurity irradiated by a linearly polarized ILF should decrease monotonically (in absolute values) with the increase of the laser intensity, vanishing for extremely high intensities. This means that it would be possible to provoke impurities ionization by increasing just the laser intensity.[9] However, the variational wave functions they have used are not suitable to describe the impurity states under a terahertz laser in the limit of high laser intensities since they do not allow the electronic cloud to stretch along the polarization direction, a phenomenon that is known by atomic physicists as the *dichotomy* of the hydrogen atom under ILF conditions.[10,11] Therefore, it is expected that the hydrogenic impurities stretch under the action of an ILF and a better variational treatment is clearly needed for elucidating whether it will ionize or become stable against ionization in the high-intensity limit.

In this Brief Report we investigate the effect of a monochromatic, linearly polarized ILF on the binding energy of a hydrogenic impurity in a bulk semiconductor by employing a suitable variational treatment. Our approach takes into account trial wave functions similar to that for the ground and first excited states of the $H_2{}^+$ molecular ion. These wave functions allow the dichotomy of the hydrogenic impurity under an ILF, differently from the $1s$ hydrogen-atom-like wave functions used previously.[7,9]

A nonrelativistic electron subjected to a time-independent potential $V(\mathbf{r})$ and moving under the action of a laser radiation field can be described by the following time-dependent Schrödinger (SCD) equation:

$$\left(\frac{(\mathbf{p}-q\mathbf{A})^2}{2m}+V(\mathbf{r})\right)\psi(\mathbf{r},t)=i\hbar\,\frac{\partial\psi(\mathbf{r},t)}{\partial t}, \tag{1}$$

where m is the electron mass, $q=-e$ is its charge, $\mathbf{p}=-i\hbar\boldsymbol{\nabla}$ is the momentum operator, and $\mathbf{A}$ is the vector potential associated with the radiation field. We choose the Coulomb gauge to treat this problem ($\boldsymbol{\nabla}\cdot\mathbf{A}=0$ and $\phi=0$). In this gauge, one has $\mathbf{F}=-\partial\mathbf{A}/\partial t$.

The task of computing the solution $\psi(\mathbf{r},t)$ of Eq. (1) is difficult because this differential equation cannot, in general, be solved analytically and an accurate numerical solution for the full (3+1)-dimensional [(3+1)D] problem is hard to develop computationally for a Coulombian potential.[11,12] Therefore, nonperturbative approaches are sought for solving Eq. (1).[13] Within the nonrelativistic dipole approximation for $\mathbf{A}=\mathbf{A}(t)$ we can make use of a well-known nonperturbative method for treating this radiation-matter interaction problem, namely, the Kramers-Henneberger (KH) unitary translation transformation.[14,15] By applying this transformation on Eq. (1), one has

$$-\frac{\hbar^2}{2m}\nabla^2\tilde{\psi}+V[\mathbf{r}-\boldsymbol{\alpha}(t)]\tilde{\psi}=i\hbar\,\frac{\partial\tilde{\psi}}{\partial t}, \tag{2}$$

where

$$\boldsymbol{\alpha}(t) \equiv \frac{e}{m}\int^{t} \mathbf{A}(t')dt'. \tag{3}$$

This SCD equation is suitable to describe electron states under the action of a laser field because the time dependence has been transferred from the momentum to the Coulombian potential, being equivalent to Eq. (1).

Let us now apply this nonperturbative method in the calculation of the binding energy of a substitutional impurity in a bulk semiconductor under an ILF. We will focus our analysis on shallow traps since the wavefunction for deep levels presents intrinsic mathematical complexities.[16] For simplicity, we consider a hydrogenic impurity under a monochromatic, linearly polarized laser field in the dipole approximation. Within the effective mass approximation, and assuming that the dielectric constant ϵ_r and the electron effective mass m^* are both isotropic,[17] the SCD equation reads

$$\left\{\frac{[\mathbf{p}+e\mathbf{A}(t)]^2}{2m^*} - \frac{k}{\epsilon_r}\frac{e^2}{|\mathbf{r}|}\right\}\psi(\mathbf{r},t) = i\hbar\frac{\partial\psi(\mathbf{r},t)}{\partial t}, \tag{4}$$

where $k \equiv 1/(4\pi\epsilon_0)$ is the Coulomb constant and ϵ_0 is the vacuum permittivity. The origin of the laboratory reference frame was chosen at the center of the impurity site. Proceeding with the KH transformation, one has

$$\left(-\frac{\hbar^2}{2m^*}\nabla^2 - \frac{k}{\epsilon_r}\frac{e^2}{|\mathbf{r}-\boldsymbol{\alpha}(t)|}\right)\tilde{\psi} = i\hbar\frac{\partial\tilde{\psi}}{\partial t}. \tag{5}$$

By considering a laser field polarized along the z-axis direction, one has $\mathbf{F}(t)=F_0\sin(\omega t)\mathbf{k}$, where F_0 is the field amplitude (constant), ω is the laser angular frequency, and $\mathbf{k}$ is the unit vector for the z axis. For this field, the vector potential can be written as $\mathbf{A}(t)=A_0\cos(\omega t)\mathbf{k}$, where $A_0=F_0/\omega$. Then, from Eq. (3), one has

$$\boldsymbol{\alpha}(t) = \boldsymbol{\alpha}_0\sin(\omega t), \tag{6}$$

where $\boldsymbol{\alpha}_0=\alpha_0\mathbf{k}$, with $\alpha_0=eF_0/(m^*\omega^2)$ being the laser-dressing parameter.[18] This parameter has a classical counterpart, namely, the amplitude of the electron oscillations along the polarization direction.

Since the time-dependent potential in Eq. (5) oscillates periodically, the Floquet method can be applied in seeking for a quasiperiodic solution.[11,15] This yields a system of coupled differential equations in coordinate space relating the coefficients of the series expansions for the potential and the wave function. For solving this system, an iteration scheme has been worked out which proceeds essentially in powers of $1/\omega$.[11] To lowest order in iteration, the system reduces to only one equation, namely,

$$-\frac{\hbar^2}{2m^*}\nabla^2\tilde{\psi}_0 + V_0(\mathbf{r};\alpha_0)\tilde{\psi}_0 = E\tilde{\psi}_0. \tag{7}$$

In this order, the laser field is incorporated into the potential, which becomes

$$V_0(\mathbf{r};\alpha_0) = \frac{1}{\pi}\int_{-1}^{+1} V(\mathbf{r}-\boldsymbol{\alpha}_0 u)\frac{du}{\sqrt{1-u^2}}, \tag{8}$$

the so-called "dressed" potential. It has been shown that this lowest-order approximation is valid when the ponderomotive (here, the DFKE) energy is much larger than the photon energy,[19] i.e., $e^2F_0^2/(4m^*\omega^2) \gg \hbar\omega$. This simplifies to $\alpha_0 \gg 2/\sqrt{\omega}$ (in effective units). For a terahertz laser, e.g., one has $\alpha_0 \gg 2.7a_B^*$, where $a_B^* \equiv \hbar^2\epsilon_r/(m^*ke^2)$ is the effective Bohr radius of the hosting material (in GaAs, e.g., $a_B^*=86$ Å in bulk[20] GaAs). For sufficiently high frequencies, it was shown by Ehlotzky that $V_0(\mathbf{r};\alpha_0) \approx 1/2[V(\mathbf{r}+\boldsymbol{\alpha}_0) + V(\mathbf{r}-\boldsymbol{\alpha}_0)]$ is a good approximation in seeking for low-lying states solutions of Eq. (7).[21] By taking this approximation into account, a 3D SCD equation arises, namely, $H_0\tilde{\psi}_0 = E\tilde{\psi}_0$, where

$$H_0 = -\frac{\hbar^2}{2m^*}\nabla^2 - \frac{1}{2}\frac{ke^2}{\epsilon_r}\left(\frac{1}{|\mathbf{r}+\boldsymbol{\alpha}_0|} + \frac{1}{|\mathbf{r}-\boldsymbol{\alpha}_0|}\right) \tag{9}$$

is the Hamiltonian operator, whose energy eigenvalues are E. This Hamiltonian is analogous to that for the electronic motion in the H_2^+ molecular ion (assuming the Born-Oppenheimer approximation) with an internuclear distance $R=2\alpha_0$, except for a factor of 1/2 multiplying the potential.[21] In view of this analogy we found it appropriate to follow the theory of molecular orbitals, which has been so successful in molecular structure calculations.[22] Then we choose the variational version of the Linear combination of atomic orbitals (LCAO) method (minimal basis set),[23] with trial wave functions similar to those for the $(1s)\sigma$ states (gerade and ungerade) of the H_2^+ molecule,[24], as given by

$$\tilde{\psi}_0(\mathbf{r};\boldsymbol{\alpha}_0,\beta) = B[\varphi_+(\mathbf{r};\boldsymbol{\alpha}_0,\beta) \pm \varphi_-(\mathbf{r};\boldsymbol{\alpha}_0,\beta)]. \tag{10}$$

The +(−) signal is for the σ_g (σ_u) state. In Eq. (10), $\varphi_{\pm}=\tilde{B}(\beta)\times\exp(-\beta|\mathbf{r}\pm\boldsymbol{\alpha}_0|/a)$ are prenormalized 1s atomic orbitals (AOs)—i.e., $\tilde{B}^2=\beta^3/(\pi a^3)$ – and β is the only variational parameter (dimensionless).[25] For details on the accuracy of LCAO variational solutions for the H_2^+ molecule, see Ref. 24. The Rayleigh-Ritz variational method follows with the calculation, for a given α_0, of the expectation value $\langle H_0\rangle(\beta)=\langle\tilde{\psi}_0|H_0|\tilde{\psi}_0\rangle(\beta)=\int\int\int\tilde{\psi}_0^* H_0\tilde{\psi}_0 d^3r$, which is to be minimized with respect to β. The integrand may be separated in kinetic and potential terms, some of them being identical (by symmetry). By grouping such terms, one has

$$\langle H_0\rangle(\beta) = -2B^2\left[\frac{\hbar^2}{2m^*}(T_{11}\pm T_{12}) + \frac{ke^2}{2\epsilon_r}(C_{11}\pm C_{12})\right], \tag{11}$$

where $T_{11}(\beta)\equiv\langle\varphi_+|\nabla^2|\varphi_+\rangle$, $T_{12}(\beta)\equiv\langle\varphi_+|\nabla^2|\varphi_-\rangle$, $C_{11}(\beta)\equiv\langle\varphi_+|(1/|\mathbf{r}+\boldsymbol{\alpha}_0|+1/|\mathbf{r}-\boldsymbol{\alpha}_0|)|\varphi_+\rangle$, and $C_{12}(\beta)\equiv\langle\varphi_+|(1/|\mathbf{r}+\boldsymbol{\alpha}_0|+1/|\mathbf{r}-\boldsymbol{\alpha}_0|)|\varphi_-\rangle$. The normalization condition for $\tilde{\psi}_0$, namely, $\langle\tilde{\psi}_0|\tilde{\psi}_0\rangle=1$, yields $B^2=1/(2\pm 2S)$, where $S(\beta)\equiv\langle\varphi_+|\varphi_-\rangle$ is the overlap integral. All these integrals can be solved analytically by using confocal elliptic coordinates.[24] We obtain

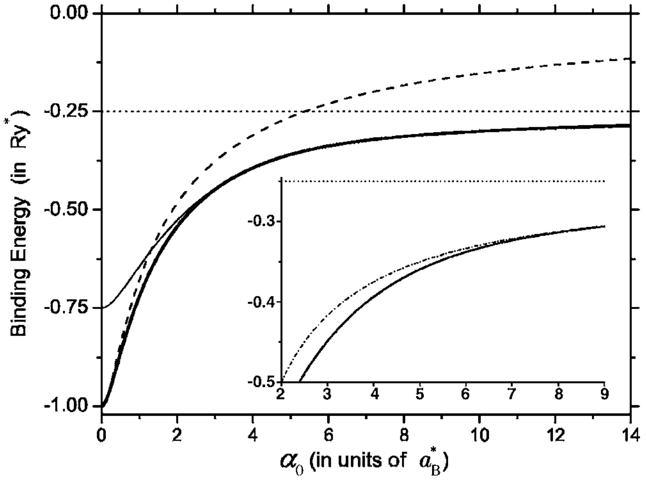

FIG. 1. Ground-state impurity binding energy as a function of α_0, the laser-dressing parameter. The solid curves are our results, which were found via H_2^+-like usual LCAO (thin line) and LCAO variational models (thick line). The result obtained by Nie *et al.* with a 1*s* trial wave function (dashed line) is shown for comparison. The horizontal dotted line marks the $-1/4$ Ry* level. In the inset, the solid line is our LCAO variational result and the dash-dotted line is the approximation we are proposing in Eq. (13).

$$T_{11}(\beta) = -\frac{\beta^2}{a^2}, \quad T_{12}(\beta) = -\frac{\beta^2}{a^2} g(\rho\beta)e^{-\rho\beta}, \tag{12a}$$

$$C_{11} = \frac{2}{a\rho} f(\rho\beta)\sinh(\rho\beta), \quad C_{12} = \frac{2\beta}{a} f(\rho\beta), \tag{12b}$$

$$S(\beta) = \left(1 + \rho\beta + \frac{\rho^2}{3}\beta^2\right)e^{-\rho\beta}, \tag{12c}$$

where $f(x) \equiv (1+x)\exp(-x)$, $g(x) \equiv 1+x-x^2/3$, and $\rho \equiv 2\alpha_0/a$ is a useful dimensionless parameter. The root β_0 of $d\langle H_0\rangle/d\beta=0$ marks the minimum of $\langle H_0\rangle(\beta)$. We found that it decreases from 2 to 1 for the ground state and increases from 0.4 to 1 for the first-excited state. This was expected since our system scales with the H_2^+ molecule, in which the variational parameter presents the same behavior as R goes from 0 (a He$^+$ ion) to ∞ (two separated 1*s* hydrogen AOs, just one being occupied). Once β_0 is found, we compute $E^{var}=\langle H_0\rangle(\beta_0)$, which is a very good approximation for E, the actual energy eigenvalue of H_0.[22,24]

Our results for the impurity binding energy as a function of α_0 are represented by the solid curves in Fig. 1 (σ_g ground-state) and Fig. 2 (σ_u first-excited state). For comparison purposes, the result obtained by Nie *et al.* using atomiclike trial wave functions (dashed curves) are also shown.[9] As may be seen in Fig. 1, our accurate variational model (thick solid curves) yields the correct values for the binding energy in both limits $\alpha_0 \to 0$ (no laser field) and $\alpha_0 \to \infty$ (i.e., in the high-intensity limit), which are -1 Ry* and $-1/4$ Ry*, respectively, for the ground state. For the first excited state, as seen in Fig. 2, we found -0.2 Ry* as $\alpha_0 \to 0$ and $-1/4$ Ry* as $\alpha_0 \to \infty$.[26]Here, 1 Ry$^* \equiv m^*k^2e^4/(2\hbar^2\epsilon_r^2)$ is the effective Rydberg energy (in GaAs under ILF, e.g., one has[20] 1 Ry*=7.7 meV). Note that the results obtained with the usual LCAO wave functions (thin solid lines)—i.e., by fixing β=1 in Eq. (10)—and LCAO variational wave functions (thick solid lines) approach each other asymptotically with the increase of α_0, being almost the same for α_0 greater than, say, $3a_B^*$. Note that the solid lines in both figures tend to $-1/4$ Ry* with the increase of α_0; thus the impurity becomes stable against ionization in the high-intensity limit, for a given frequency. This is in contrast to the results presented by Nie *et al.* (dashed curves), in which the binding energies tend to 0 when $\alpha_0 \to \infty$. This means that, for a given frequency, the impurity should ionize with increase of the intensity, which conflicts with our results just in the limit in which our LCAO models are known to be more accurate.

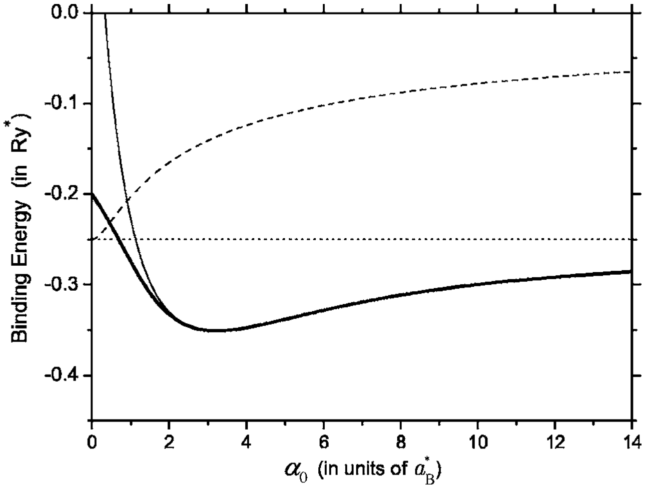

FIG. 2. Same as Fig. 1 for the first excited state. The result obtained by Nie *et al.* with a 2*s* trial wave function (dashed line) is shown for comparison.

In treating the high-intensity limit, for a frequency high enough,[19] we found a simple analytical approximation for the impurity binding energy as a function of α_0, contrarily to one of the conclusions of Nie *et al.*[9] By noting that in the limit $\alpha_0 \gg 1a_B^*$ the overlap integral tends to zero and $\tilde{\psi}_0$ becomes two isolated 1*s* AOs, then $\beta \to 1$. Hence, by making β=1 in Eqs. (12) and then neglecting the terms with $\exp(-\rho)$, we found

$$E^{var}(\alpha_0 \gg 1) \approx \left(-\frac{1}{4} - \frac{1}{2\alpha_0}\right) \text{Ry}^*, \tag{13}$$

with α_0 in units of a_B^* (this will be assumed hereafter). As may be seen in the inset of Fig. 1 for the ground state, this formula approximates asymptotically the binding energy obtained with our LCAO variational model, yielding almost the same numerical values for $\alpha_0 > 7$. In terms of the laser output power, the approximation above is valid for intensities above I_{min}, the intensity for which α_0=7. We found that I_{min} depends on the laser frequency ν (in THz) according to $I_{min} \approx 0.92\epsilon_r^{5/2}\nu^4$ (kW/cm^2).[18] For instance, in GaAs one has

$I_{min} \approx 360$ kW/cm^2 for $\nu = 1$ THz, which corresponds to a field amplitude of 9.1 kV/cm.[20] Note that the current generation of FELs can provide ILFs in thefrequency range $\sim 0.1 - 10$ THz, with amplitudes up to ~ 100 kV/cm ($\alpha_0 \approx 77$ for a THz laser),[2] which makes our results testable. Hence, experimental results for the impurity binding energy as a function of laser intensity, mainly for intense THz laser fields and low-temperature conditions,[27] are awaited.

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[17] This guarantees that the impurity states in this semiconductor are strictly scaled-down versions of those in the **H** atom. This occurs, e.g., in GaAs, in which the minimum of the conduction band (at $\mathbf{K}=0$) has Γ_6 symmetry.

[18] For a given laser source, whose frequency is ν and output power is I, the following practical formulae are useful: F_0 (in kV/cm) $\approx 0.87\sqrt{I}/\sqrt[4]{\epsilon_r}$ and α_0 (in units of a_B^*) $\approx 7.31\epsilon_r^{-5/4}\sqrt{I}/\nu^2$, for I in kW/cm^2 and ν in THz.

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[22] J. P. Lowe and K. A. Peterson, *Quantum Chemistry*, 3rd ed. (Elsevier/North-Holland, Amsterdam, 2006), Chap. 7.

[23] The usual H_2^+ LCAO wave function is simply $\psi(\mathbf{r},\mathbf{R}) = C[\phi_+(\mathbf{r},\mathbf{R}) \pm \phi_-(\mathbf{r},\mathbf{R})]$, where $\phi_\pm(\mathbf{r},\mathbf{R})$ are 1s AOs.

[24] J. C. Slater, *Quantum Theory of Matter*, 2nd ed. (McGraw-Hill, New York, 1968), pp. 368–400.

[25] Note that the 1s AO that composes our trial wave function should correspond to a system composed by an electron attracted by a center of charge $+e/2$. The Bohr radius for such an AO is $a = 2a_B^*$.

[26] The LCAO variational result for the $(1s)\sigma_u$ state of the H_2^+ molecule in the limit $R \to 0$ is -0.8 Ry, somewhat above the exact value of -1 Ry (the energy of the 2p AO for the He$^+$ ion). Thus the exact value projected for the impurity in the absence of a laser field is -0.25 Ry*, instead of -0.20 Ry*. This small difference is not relevant here because we are interested in the limit of large α_0.

[27] The low-temperature condition is crucial since thermal ionization can mask the laser ionization process. Thermal ionization is expected to be efficient at temperatures above T_{max}, given by $3/2\ k_B T_{max} = 1/4$ Ry*. In GaAs, one has $T_{max} = 14.8$ K (easily attainable in the laboratory).