Helium atom in metallic electron gases: A comparative study based on screened Schrödinger Hamiltonians

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Abstract

In the presence of an environment of mobile charges, the bound-state Schrödinger Hamiltonian for an embedded He atom differs from its vacuum form. The central problem of incorporating screening in the nucleus-bound-electron and bound-electron-bound-electron terms of this Hamiltonian is investigated here for the He ground-state in a comparative manner by using two models, and the same product form of $1s$ -type parametric hydrogenic functions to perform exploratory variational calculations. Both models employ induced charge densities in the corresponding Poisson's equations with a fixed point-like nucleus, but the underlying charge-density response of the host system is generated by differently chosen perturbations. These are the point-charge nucleus and the nucleus-bound-electron charge distribution as external perturbations. The repulsive bound-electron-bound-electron interaction in the Hamiltonian is modeled by a parametric Yukawa-type potential. Using the consistent variational results for the binding energies and wave functions, the charge-state dependent stopping power of a metallic target for slowly moving He is briefly discussed.

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I. MOTIVATIONS

Understanding two-electron atoms was crucial in showing [1] that Schrödinger's wave mechanics was not just an *ansatz*, which works fortuitously for the one-electron hydrogen atom. The structure of such electronic systems within a given nuclear framework depends not only on the balance between the kinetic energy of bound electrons and their attraction to the nucleus, but also [2] on the mutual electric repulsion. The last effect can cause difficulties (self-interaction, and its interplay with local exchange-correlation) in modeling based on an effective [3, 4] Hartree-type *one-electron* Hamiltonian for He. It is well-known, however, that the variational method applied to the Schrödinger Hamiltonian

$$H = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{r_{12}}, \quad (1)$$

with a simple parametric (Z_v) form for the two-electron trial wave function

$$\psi_{gr}(r_1, r_2) = \sqrt{\frac{Z_v^3}{\pi}} e^{-Z_v r_1} \sqrt{\frac{Z_v^3}{\pi}} e^{-Z_v r_2}, \quad (2)$$

can give a very reasonable result for the ground-state energy in vacuum. Hartree atomic units (a.u.), $e^2 = \hbar = m_e = 1$, are used throughout this paper. This energy is the sum of the expectation values of kinetic and potential energies of $\psi_{gr}(r_1, r_2)$, and it is

$$E_{gr}(Z_v) = 2 \left(\frac{Z_v^2}{2} - Z Z_v \right) + \frac{5}{8} Z_v. \quad (3)$$

The variational constraint results in $Z_v^{(0)} = 27/16$ at $Z = 2$, and thus gives $E_{gr} \simeq -2.85$. There is an important reduction in the ground-state energy due to the last term of Eq.(1).

Since the characterization of plasma effects on a simple atomic system, He, embedded in the degenerate electron gases of free-electron-like metals is one of the major concerns of current experimental research [5] on the interaction of slow He^+ intruders with condensed matter, consistent modifications of the interaction terms in Eq.(1) is of considerable interest. For instance, proper calculations of matrix elements for single and double re-ionizations [5, 6] in distance dependent collisions with lattice ions during penetration requires reasonably accurate knowledge of binding energies of electrons and wave functions around the slowly moving projectile. The present paper is devoted, on the theoretical side, to this important basic aspect of a more detailed understanding of slowing down processes where, in target-dependent [7] re-ionization, the kinetic energy of the projectile is decreased [5] due to promotion of the electronic bound state above the Fermi energy of a metal.

Notice finally that by using more sophisticated [8] wave functions in variational calculations with the bare Hamiltonian one can get values closer to the experimental ground-state energy $E_{gr} \simeq -2.90$. However, since our paper is devoted to a comparative study of an embedded He atom problem, we use the simple one-parametric form given by Eq.(2) in this exploratory variational attempt. At embedding, as was clearly emphasized recently [9], the really important physical problem is a reasonable modeling of modifications in the potential energy terms by considering the response of an electronic many-body host. Furthermore, in modern information-theoretic [10] terminology, the quantum entanglement in the He ground state is far less than in its triplet or excited states.

The rest of this paper is organized as follows. The next Section is devoted to constructing of a model and discussing the results. Section III contains a summary and comments.

II. EMBEDDING HAMILTONIANS AND RESULTS

The electrostatic modeling [11, 12] considers first the induced charge generated by a point-like attractive nucleus and a point-like repulsive electron as classical external perturbations. This induced density can be calculated by using the screened [13] density-density response function, which is a system-dependent quantity, in its Thomas-Fermi (TF) limit:

$$\chi(q) = \frac{k_F}{\pi^2} \frac{1}{1 + \lambda_{TF}^2/q^2}, \quad (4)$$

where, in atomic units, $\lambda_{TF}^2 = 4k_F/\pi$ in terms of the Fermi wave number $k_F = (3\pi^2 n_0)^{1/3}$, determined by the density n_0 of the gas. One can use $\lambda_{TF} = 1.563/\sqrt{r_s}$ as well, in terms of the Wigner-Seitz radius; $r_s \in [1, 6]$ for metals. With a point-like test-charge Q , one has

$$n_{sc}^{(1)}(q) = \frac{4\pi Q}{q^2} \chi(q), \quad (5)$$

for the induced charge density in momentum space. From the Poisson equation

$$V^{(1)}(q) = \frac{4\pi}{q^2} [Q - n_{sc}^{(1)}(q)], \quad (6)$$

and by Fourier transformation, we can get a TF (also named as Yukawa- or Debye-type) screened potential $V^{(1)}(r) = (Q/r) \exp(-\lambda_{TF} r)$ in real space.

In such a manner one gets the conventional [11, 12] embedded (*em*) model Hamiltonian

$$H_{em}^{(1)} = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z}{r_1} e^{-\lambda_{TF} r_1} - \frac{Z}{r_2} e^{-\lambda_{TF} r_2} + \frac{1}{r_{12}} e^{-\lambda_e r_{12}}, \quad (7)$$

where, in order to adjust (at $Z = 2$) the electron-electron interaction, we use a different parameter (λ_e) to characterize screening in the inter-electronic repulsive term. The parametric ground-state energy ($Z = 2$) of $\psi_{gr}(r_1, r_2)$ with $H_{em}^{(1)}$ becomes

$$E_{gr}^{(1)}(Z_v, \lambda_{TF}, \lambda_e) = 2 \left[\frac{Z_v^2}{2} - \frac{4Z Z_v^3}{(2Z_v + \lambda_{TF})^2} \right] + \frac{Z_v^3}{2(2Z_v + \lambda_e)^4} (20Z_v^2 + 8Z_v\lambda_e + \lambda_e^2). \quad (8)$$

The last term in Eq.(8), which relates to the expectation value of electron-electron repulsion, can be obtained by integration in real space using elliptic coordinates or, alternatively, by using the Faltung (convolution) theorem and performing a single-integration in wave-vector space. We will employ such a momentum-space integration below, for simplicity.

Before turning to a more consistent model of the nucleus-bound-electron screened interaction, we outline those arguments on which our choice for λ_e to model a bound-electron-bound-electron screened repulsion at embedding of He into a cold plasma is based. In a classical picture one can use $\lambda_e = \lambda_{TF}(k_F)$ to describe a hole-like distribution of unit norm in the surrounding average charge density induced by a static electron [14] (without statistical properties) or an antiproton [15]. This Coulombically produced hole disappears at vanishing coupling. However, in quantum mechanics, the state function of the many-body system is a Slater determinant due to Pauli's exclusion principle. Therefore, even without Coulomb interaction between the system's mobile electrons, there is a co-moving statistical spherical hole [16, 17] of unit norm around each electron. It was noticed [18, 19] that this statistics-based hole efficiently quenches the direct (polarization) action of electrons.

Clearly, it is just the *interrelation* between direct and exchange interaction on which the effect of a reduced screening in cold quantum plasmas is based. Quantitatively, λ_e becomes essentially smaller [20] than $\lambda_{TF}(k_F)$. A very recent theoretical [21] analysis of data on two-electron emission from iron results in $\lambda_e \simeq 0.1$ instead of the much higher $\lambda_{TF} = 0.9$. Furthermore, in the case of atom embedding [22], the polarization action of the free electrons on the interaction of atomic electrons is expected to be even less [23] effective, since electrons in atoms cannot move around as readily [24] as free electrons. In harmony with this, Anderson used a completely unscreened ($\lambda_e = 0$) Coulomb integral in his problem with well-localized atomic orbitals in metals [25]. Our modeling in this work via a reduced λ_e screening is based [see, below Eq. (11)] on the above-outlined qualitative and quantitative arguments. Now we turn to the other, equally important problem of He embedding.

So, how to change Eq.(5) and thus the associated Eq.(6) with $Q = Z = 2$ consistently? We follow here an earlier idea applied [26] to He^+ in an electron gas. Thus, we take the charge density of a sub-system composed of the nucleus and a bound electron as external generator of the induced charge polarization to be used in Poisson's equation to arrive at an effective central potential in the bound-state Hamiltonian for the other bound electron. In such a manner all electrons, except the one which is singled out, are considered solely via electrostatics. We stress at this point that our construction is free from self-screening. The proper elimination of such a spurious effect is a key question [27] in recently applied many-body approximations that are based on Hartree-type one-electron potentials.

In the light of the above, the induced screening density is now as follows

$$n_{sc}^{(2)}(q) = \frac{4\pi}{q^2} \left[Z - \frac{16Z_v^4}{(q^2 + 4Z_v^2)^2} \right] \chi(q), \quad (9)$$

where the second term in the brackets is the Fourier transform of the properly normalized hydrogenic charge density $(Z_v^3/\pi) \exp(-2Z_v r)$ of a $1s$ -type state. The corresponding Poisson equation takes the form

$$V^{(2)}(q) = \frac{4\pi}{q^2} [Z - n_{sc}^{(2)}(q)], \quad (10)$$

instead of Eq.(6), which refers to a different model. By applying (in momentum space, as we discussed above) the modified embedded $[H_{em}^{(2)}]$ Schrödinger Hamiltonian for bound states we arrive, after straightforward analytic calculations, at the parametrized energy

$$E_{gr}^{(2)}(Z_v, \lambda_{TF}, \lambda_e) = E_{gr}^{(1)} - 2 \left[\frac{5Z_v}{8} - \frac{Z_v^3}{2(2Z_v + \lambda_{TF})^4} (20Z_v^2 + 8Z_v\lambda_{TF} + \lambda_{TF}^2) \right], \quad (11)$$

where $E_{gr}^{(1)}$, the conventional estimation, is given by Eq.(8) above.

As a comparison of Eq.(9) and Eq.(5) already shows, the consideration of a repulsive bound-state charge distribution together with the field of the attractive nucleus weakens the perturbation on the rest of the system somewhat, resulting in less effective nucleus-bound-electron screening in the bound-state Schrödinger Hamiltonian and finally leads to a negative energy contribution in Eq.(11). Straightforward Taylor expansions show that the asymptotic behaviors of these energies for small enough screenings are

$$E_{gr}^{(1)} \simeq 2 \left(\frac{Z_v^2}{2} - ZZ_v + \frac{5Z_v}{16} \right) + (2Z\lambda_{TF} - \lambda_e) - \frac{3\lambda_{TF}^2}{2Z_v} \left[Z - \frac{35}{48} \left(\frac{\lambda_e}{\lambda_{TF}} \right)^2 \right],$$

$$E_{gr}^{(2)} \simeq 2 \left(\frac{Z_v^2}{2} - ZZ_v + \frac{5Z_v}{16} \right) + [2(Z-1)\lambda_{TF} - \lambda_e] - \frac{3\lambda_{TF}^2}{2Z_v} \left[\left(Z - \frac{35}{24} \right) - \frac{35}{48} \left(\frac{\lambda_e}{\lambda_{TF}} \right)^2 \right].$$

FIG. 1: The variationally determined parameters $Z_v^{(2)}$ (solid curve) and $Z_v^{(1)}$ (dashed curve) as a function of k_F . The dotted curve is based on Eq.(12) for $Z_v^{(2)}$. See the text for further details.

Our consistent model, which considers the reaction of free electrons to the presence of bound electrons as well, leaves the inert atom more intact at embedding. Both expressions give stronger binding with growing λ_e at fixed λ_{TF} . On the other hand, for the $\lambda_e = 0$ choice in our $E_{gr}^{(2)}(Z_v, \lambda_{TF}, \lambda_e)$, a high value of $\lambda_e = 2\lambda_{TF}$ would be needed in $E_{gr}^{(1)}(Z_v, \lambda_{TF}, \lambda_e)$ to get the same value of total energy in a leading-order expansion. The role of such an un-physical over-screening was investigated recently [28] in a high-precision conventional study.

The above expressions are based on second-order Taylor's expansions. Remarkably, the variational solution for $Z_v^{(2)}$ based on the above $E_{gr}^{(2)}$ turns out to be accurate when we compare it with the numerically exact solution. Thus, one can safely apply (see, Fig. 1) the following analytic approximation for the important metallic ($0.6 \leq k_F \leq 1.3$) range

$$Z_v^{(i)} \simeq Z_v^{(0)} - \frac{A^{(i)}}{2} \left[\frac{1}{Z_v^{(0)}} \right]^2, \quad (12)$$

where $A^{(i)}[Z, \lambda_{TF}, \lambda_e]$ are the multiplying factors of $(1/Z_v)$ in the last terms of the parametric ground-state energies $E_{gr}^{(i)}$. Thus, we get $Z_v^{(2)} \simeq [(27/16) - 0.14\lambda_{TF}^2]$ with $\lambda_e = 0$. As the structure of $A^{(2)}(\lambda_{TF}, \lambda_e)$ signals, the agreement with numerically determined variational results for $Z_v^{(2)}$ would be, at least mathematically, even better for the $\lambda_e > 0$ choices in $E_{gr}^{(2)}$, since in such cases one could get smaller pre-factor than the above 0.14 valid at $\lambda_e = 0$.

Now we turn to the presentation and discussion of our variationally determined numerical results. The variational values for Z_v are shown in Fig. 1, as a function of k_F , in order to characterize the wave-function extension. The solid curve [$Z_v^{(2)}$] is based on $E_{gr}^{(2)}(Z_v, \lambda_{TF}, \lambda_e = 0)$, while the dashed curve on the conventional $E_{gr}^{(1)}(Z_v, \lambda_{TF}, \lambda_e = \lambda_{TF})$.

FIG. 2: The negative of the variationally determined ground-state energies, $E_{gr}^{(i)}$, as a function of k_F . The solid curve is based on Eq.(11) with the $\lambda_e = 0$ choice, while the dashed curve on Eq.(8) with the $\lambda_e = \lambda_{TF}$ choice, respectively. See the text for further details.

The dotted curve is based on the analytic result in Eq.(12), for $Z_v^{(2)}$ with $\lambda_e = 0$. In our consistent modeling (solid curve) the wave-function extension remains more intact, than in the conventional modeling (dashed curve).

In Fig. 2 we exhibit the negative of the ground-state binding energies as a function of the system variable $k_F \in [0.3, 2]$. The simple metals are in this range. For instance, in a free-electron-like Al host used [5] in experiment, $k_F \simeq 0.9$ roughly. The solid curve is based on Eq.(11) with the theoretically [25], and experimentally [21] as well, motivated choice for electronic screening: $\lambda_e = 0$. The dashed curve shows Eq.(8) with the conventional [11, 12] classical choice $\lambda_e = \lambda_{TF}$. At around $k_F \simeq 1$, there is a factor of about two between the two, solid and dashed, results for ground-state binding energies.

Fig. 3 is devoted to the ionization potentials $I^{(i)}$ at embedding. They are defined via

$$I^{(i)} = -E_{gr}^{(i)}(Z_v, \lambda_{TF}, \lambda_e) + (1/2) E_{gr}^{(1)}(Z_v, \lambda_{TF}, \lambda_e = \infty),$$

by using the corresponding, variationally determined energies. As the curves of Fig. 2 already heralds, the ionization potential is considerable higher in our consistent ($i = 2$) modeling than the one ($i = 1$) based on the conventional approximation. The stars in Fig. 3 refer to the negative of Kohn-Sham 1s energy eigenvalues [4] in He at $r_s = 2$ and $r_s = 4$ for the host. These eigenvalues are 0.35 and 0.45, respectively. The $I^{(2)}$ values are, roughly, two-times higher than these auxiliary eigenvalues. Of course, it is well-known that even in vacuum the Kohn-Sham eigenvalue (-0.58) lies above the Hartree-Fock value (-0.92). This

FIG. 3: The ionization potentials, $I^{(i)}$ as a function of k_F . To the solid ($i = 2$) curve we applied Eq.(11) with $\lambda_e = 0$, while to the dashed ($i = 1$) curve Eq.(8) with $\lambda_e = \lambda_{TF}$, respectively. The dotted and dash-dotted curves show the Fermi energy (E_F) and the chemical potential (μ), respectively. Stars represent the negative of the auxiliary Kohn-Sham $1s$ single particle energies at $r_s = 4$ and $r_s = 2$, obtained within LDA. See the text for further details.

figure shows by dotted and dash-dotted curves, respectively, the Fermi energy, $k_F^2/2$, and the chemical potential, $\mu(k_F)$, of the interacting (paramagnetic) degenerate electron gas

$$\mu(k_F, T = 0) \simeq 0.5 k_F^2 - 0.3 k_F [1 + (0.1/k_F) \ln(1 + 6 k_F)].$$

This thermodynamical quantity, the basic constant [29] to a grand-canonical description in the presence of an external field, is plotted only for its positive range as a function of k_F .

It was pointed out earlier in a careful analysis [26] that energies calculated relative to their vacuum values must be altered by adding the magnitude of the work function, so the binding becomes weaker. We can see (c.f., Ref.[29]) that the chemical potential becomes equal to our $I^{(2)}$ only at the relatively high density ($r_s \simeq 1.25$) of the homogeneous (interacting) model system. The crossing of $I^{(2)}$ with E_F is at about $r_s \simeq 1.6$, the density parameter of a carbon target. For the most important practical range ($k_F \leq 1$) we can avoid, in our consistent modeling, the many-body problems associated with auto-ionization and line-broadening.

From the variationally determined numerical results, see Figs.(1-3), we can draw the following important conclusions. The ground-state energy $E_{gr}^{(i)}$, and the optimized $Z_v^{(i)}$ parameter, depend quite strongly on the form of the underlying screened Schrödinger Hamiltonian for bound states. Our consistent modeling gives a more rigid embedded atom, i.e., the net effect of the charge response of the surrounding metallic ($k_F \leq 1$) electron gas on an

embedded chemically inert atom is weaker. This behavior is expected physically.

On the other hand, there is a reduction and variation of energies over the metallic density range, in harmony with the earlier forecast [4] based on the Hartree-like one-electron Kohn-Sham treatment of He embedding. Even for a low density electron gas there is a shift of about $(2 - 3) eV$ in our ionization potential. Remarkably, this value is similar to the one derived for the level shift in He close enough [30] to an aluminum surface. Clearly, the assumption of an energetically rigid core, fixed for vacuum conditions, is not particularly good for the chemically inert He at embedding. Thus pre-fixed pseudo-potentials, applied for instance to drive the time-evolution of auxiliary one-electron states in non-adiabatic situations by assuming a rigid-core He, bear some re-examination.

In order to discuss briefly the atomic collision of slow He and Al or Mg lattice atoms in free-electron-like ($k_F \simeq 0.9$ and $k_F \simeq 0.7$, respectively) metals [5] at small impact parameters, we apply quantum mechanic adiabatic perturbation theory for charge-exchange processes. We note, parenthetically, that the energy difference $[I^{(2)} - \mu]$ is higher at $k_F = 0.7$ than at $k_F = 0.9$. Now, if the relevant electronic states of the colliding atoms are similar, in their extensions and energies, then there is [31] an average 50% probability for charge exchange for one active electron. In a highly simplified independent-electron picture this number results, in our case, in a probability of 25% for the double-exchange.

So, under the above solid-state condition for atom-atom collisions, a neutral-projectile assumption is valid only with a probability of 25% or 50%. Therefore, a discussion of the stopping power of metals for slow He intruders requires, beyond an energy change in promotions [5, 6], the consideration of charge-state-dependent channels [32, 33] for energy transfers between the projectile and scattering free electrons as well. In these channels the independent electron-hole pair excitations at the Fermi level are responsible [34–36] for inelastic energy losses. This constraint is prescribed by the Pauli exclusion principle.

Since a charge-state-dependent stopping $[S^{(i)}]$ power is higher for He^+ [26] and (see below, Sec. III) for He^{++} [36] ions than for a neutral He quasi-atom [34], the weighted ($\bar{S}$) sums

$$\bar{S} = \alpha_1 S^{(0)} + \beta_1 S^{(+)}$$

$$\bar{S} = \alpha_2 S^{(0)} + \beta_2 S^{(+)} + \gamma_2 S^{(++)}$$

as conservative $[(\alpha_1 + \beta_1) = 1 \text{ or } (\alpha_2 + \beta_2 + \gamma_2) = 1]$ estimations, would give $\bar{S} > S^{(0)}$ under charge-exchange condition in harmony with data [5]. We expect, however, a rapid approach

to the $\bar{S} \simeq S^{(0)}$ limit for a situation where the distance-sensitive (impact parameter, and velocity-dependent closest approach [37]) processes become ineffective as, for instance, under channeling conditions [38] in bombarding experiments on metals.

III. SUMMARY AND COMMENTS

Motivated by the recent experimental activity [5] on the problem of He embedding into metallic electron gases, we used two screened Schrödinger Hamiltonians in order to characterize bound states around an intruder in a variational comparative manner. The special role of different screening constants in nucleus-bound-electron and bound-electron-bound-electron interactions has been investigated in detail. The results obtained for binding energies and wave function characters could contribute to the general understanding of the problem relative to the charge states of slow atoms in a screening environment, and thus to promotion processes and the associated average total stopping power, the only directly observable quantity in heavy-particle slowing down.

A more quantitative estimation of the total inelastic stopping power needs separate and detailed investigations into the precise experimental conditions for the ion-bombardment of different metals. We make here, however, a general comment following Lindhard's earlier enlightening work [39]. Statistically, one can consider the slowing down of heavy intruders as a result of a stationary energy transfer between free electrons and moving intruders. In other words, in the stationary flow there is a steady current of conduction electrons towards the slow projectile, and a steady current of scattered electrons away from it. One has to satisfy charge and current conservation laws simultaneously [13], via the amplitude of a backflow, for the case of slowly moving intruders, i.e., close enough to the pure reactive limit.

If one includes, as we indicated above in the main text, higher-order (charge-exchange and promotion) effects, electrons can by mutual collisions jump between various excited states. The total balance in the abovementioned stationary flow *can replace* [39] the often used concept of competing capture and loss processes. For instance, the modeling in [36] for He^{++} stopping rests on such a conserving approximation and gave an about $S^{(++)} \simeq 2S^{(0)}$ at $r_s \simeq 3$. Notice finally that since a proton intruder [5] does not have [40] bound states in metals, we do not expect similar charge-exchange and promotion effects as in the case of He. Further research on the stopping problem for light atomic intruders is in progress.

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