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Non-Stueckelberg oscillatory behavior in ion-solid charge transfer

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Reionization of an ion colliding with a solid surface is possible when the atomic level of the ion crosses the Fermi level of the surface. We use a recently reported soluble model to show that a shift of the atomic level above the Fermi level comparable to the surface bandwidth leads to oscillations in the ion survival probability, even when Stueckelberg oscillations are absent. The competing mechanisms responsible for this interesting oscillatory behavior are elucidated and related to a previously unexpected interference between the initially full and empty metallic states.

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Understanding the processes of charge transfer in ion scattering from metallic surfaces is fundamental to investigations of chemical reactions, adsorption and desorption at surfaces, and ion based spectroscopies and the corresponding experimental and theoretical literature is sizeable. Theoretical studies, usually based on some version of the Newns-Anderson Hamiltonian [1,2] and usually requiring lengthy numerical computations, involve either single nondegenerate atomic levels (i.e., single-particle models) [3–8] or multiple and degenerate atomic orbitals (i.e., many-body theories) [9–11]; the influence of Auger processes on charge exchange with jellium metals has also been studied [12,13]. Generally speaking, for a normally incident positive ion, the behavior of the survival probability $P_a(t_f)$ at final time $t_f \rightarrow \infty$, either decreases monotonically or displays oscillations as a function of the inverse velocity. The former behavior was originally predicted by the infinitely wide band model (TWB) [3] and the latter by the two-level model (TNB) [14–20], both of which are soluble, single-particle models. Indeed, the oscillations arise from quasiresonant interactions between the atomic energy levels and a narrow band of the solid, in analogy to the quantum-mechanical phase interference between atomic levels and localized states responsible for the Stueckelberg oscillations (SOs) [15] in atomic collisions [14–20].

The predictive capacity of these soluble, single-particle models was important in achieving the results underlying much of our intuition regarding the problem. In recent work [21,22], we reported a soluble single-particle model which covers the range between the TNB and the TWB and readily permits an exhaustive investigation of the systematic influence of the parameters of the problem, in contrast to previous work involving numerical computations: among the results was the demonstration of the destruction of the analog SO when the atomic level is close enough to the Fermi energy [21,22].

In this paper, we report a type of oscillation, which we call a *reionization oscillation* for reasons discussed below, in $P_a(t_f)$ which can exist even when the usual Stueckelberg oscillations have been destroyed and whose magnitude rivals that of the SO. We attribute the origin of these reionization oscillations to an unexpected type of interference, that between metallic states caused by their strong interaction with an atomic level which rises above the Fermi level during its trajectory. Although many-body decoherence processes, such as those associated with electron-phonon and electron-electron interactions, might destroy these oscillations, the fact that their size rivals that of the Stueckelberg oscillations, which do survive the influence of electron-electron interactions in more elaborate treatments [10], indicates that the oscillations we report should be considered in future work.

In order to discuss this phenomenon more clearly, we rely on some model independent arguments before describing the results of our model in more detail. Consider a system which, at some initial time t_0 (i.e., $t_0 \rightarrow -\infty$), consists of a separated positive ion (described by one-electron state $|a,t_0\rangle$ corresponding to the isolated atomic orbital) and metal (one-electron states $|k,t_0\rangle$, assumed orthogonal to $|a,t_0\rangle$): at later times t the two subsystems interact and the initial states evolve, respectively, into $|a,t\rangle$ and $|k,t\rangle$. The many-body system Hamiltonian is given by $H(t)=\psi^{+}(t)h(t)\psi(t)$, where $\psi(t)$ is the field operator in the Heisenberg representation and $h(t)$ is the one-particle Hamiltonian, which includes the atom-solid interaction. $\psi(t)$ obeys the same Schrödinger equation as do $|a,t\rangle$ and $|k,t\rangle$: $(\hbar i\,\partial/\partial t)\psi(t) = h(t)\psi(t)$. We can therefore write $\psi(t)$ in two alternative ways [23]: $\psi(t)=c_a(t)|a,t_0\rangle+\Sigma_k c_k(t)|k,t_0\rangle=c_a(t_0)|a,t\rangle +\Sigma_k c_k(t_0)|k,t\rangle$, where $c_\lambda(t)$ is the annihilation operator for state $|\lambda,t\rangle$ (i.e., $\lambda=a$ or k) and the sum is over all metal states. Since the atomic occupation at time t, $n_a(t) =\langle\Phi|c_a^{+}(t)c_a(t)|\Phi\rangle$, where $|\Phi\rangle$ is the state of the isolated subsystems $[P_a(t)\equiv 1-n_a(t)]$, we derive immediately Eqs. (1)–(3) in terms of $n_\lambda(t_0)$ [i.e., $\lambda=a$ or k] the initial occupations

$$n_a(t) = n_a(t_0)|\langle a,t_0|a,t\rangle|^2 + \sum_k n_k(t_0)|\langle a,t_0|k,t\rangle|^2$$

$$= \sum_{kocc} |\langle a,t_0|k,t\rangle|^2 \equiv N_{occ}(t), \qquad (1)$$

$$P_a(t) = [1 - n_a(t_0)]|\langle a,t_0|a,t\rangle|^2$$

$$+ \sum_{kun} |\langle a,t_0|k,t\rangle|^2 = S_a(t) + N_{un}(t), \qquad (2)$$

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$$S_a(t) + N_{occ}(t) + N_{un}(t) = 1. \tag{3}$$

In Eqs. (1)–(3), the subscripts *occ* and *un* correspond, respectively, to the metallic states occupied and unoccupied initially. The symbols S_a and N_{un} represent, respectively, the first and second terms of Eq. (2) for $n_a(t_0)=0$, and Eq. (3) is merely a reflection of the orthonormality and completeness of the states. The second equality in Eq. (1) corresponds to $n_a(t_0)=0$, the initial positive ion.

Equations (1)–(3) express the occupations in terms of the properties of the one-electron states, which are determined by $h(t)$. When we consider a reversible trajectory and an initially empty atomic energy level below the Fermi energy in the limit of low velocities, $\nu \to 0$, that is, the adiabatic limit, we expect the system to relax eventually to its ground state, so that $n_a(t_f)=N_{occ}(t_f)=1$ and, from Eq. (3), $S_a(t_f)=N_{un}(t_f)=0$. This means that $|a,t_f\rangle$, the state which, initially, was the atomic state, $|a,t_0\rangle$, can be considered as consisting solely of metallic states, $|k,t_0\rangle$. From Eq. (1), we see that $|a,t_0\rangle$ is distributed among the metallic states $|k,t_f\rangle$ which were initially occupied; indeed, in the limit $\nu \to 0$, it is clear that, because of conservation of energy, the overall spectrum does not change and $|a,t_0\rangle$ is mixed completely into the metallic state which is degenerate with the atomic level at large distances [4]. This complete intermixing of the initial atomic state into the $|k,t_f\rangle$ and the initial metallic states into $|a,t_f\rangle$ corresponds to extremely strong ion-solid coupling. Since the opposite limit, $\nu \to \infty$, corresponds to small changes of the initial states, we may consider that the effective interaction parameters—the interaction potential, the energy differences and the bandwidth—should be renormalized by $1/\nu$ [21,22].

Such intermixing of the states occurs whenever $S_a(t_f)=0$. Since, from Eq. (1), $S_a(t_f)$ equals the difference in $n_a(t_f)$ between that for an initially occupied and that for an initially empty atomic level, or the memory of the initial occupation, we identify $S_a(t_f)=0$ with the absence of memory, which, in turn, is associated with such strong intermixing. We expect the condition $S_a(t_f)=0$ to hold throughout a range of velocities. Of course, for higher velocities, the metallic states which intermix with $|a,t_0\rangle$ are distributed throughout a range of energies, in agreement with the uncertainty principle [24]: this range is no longer restricted only to the initially occupied states because $n_a(t_f) \neq 1$, in general.

In order to understand the origin of these reionization oscillations for an initially empty atomic state, we resort to analogy with the SO [15], which are most evident when the narrow band is completely initially full. In this case, $N_{un}(t_f)=0$, $P_a(t_f)=S_a(t_f)$, $n_a(t_f)=N_{occ}(t_f)$, and $S_a(t_f)+N_{occ}(t_f)=1$. In the region of $1/\nu$ of oscillations, both $S_a(t_f)$ and $N_{occ}(t_f)$ oscillate because of the aforementioned interference between the atomic state and a narrow range of occupied metallic states [14–20]. When we consider a band no longer initially full, and $S_a(t_f) \approx 0$, we have $P_a(t_f) \approx N_{un}(t_f)$, $n_a(t_f)=N_{occ}(t_f)$, and $N_{occ}(t_f)+N_{un}(t_f) \approx 1$, which provides necessary conditions for an analogous interference between the initially occupied and unoccupied metallic states. Sufficient conditions can be reached if the ion couples strongly with a narrow range of occupied and unoccupied metallic states. One way of achieving these conditions is by allowing the atomic level to vary with time, rising above the Fermi energy at some point and interacting resonantly with unoccupied states: an ion neutralized in the incoming path of its trajectory when the atomic level is below the Fermi level E_F can be reionized in the region where the level is above E_F. This reionization reflects the resonant interaction with the unoccupied states [25].

The exact requirements for the reionization oscillations involve the extent to which the atomic level rises and the shape of the band. Below, we describe the appearance of these oscillations within the context of our model [21,22]. We concentrate here on the dynamics of the process of reionization and the influence of level shift and band shape. That the model utilized here is analytically soluble permits us to achieve unexpected physical insights into the problem not discovered in previous numerical computations. As we show below, a previously unexpected coherent process emerges from the combined influence of shift and band effects.

Although our model [21,22] is formulated in terms of the time evolution of the annihilation operators, in order to be able to treat many-body effects, such as Auger processes, our discussion above involves the one-electron state solutions, $|\lambda,t\rangle$, $\lambda=a$ or k, of the Schrödinger equation $(\hbar i\,\partial/\partial t)|\lambda,t\rangle=h(t)|\lambda,t\rangle$. Since the two treatments yield identical results for single-particle Hamiltonians, we retain the state description and the conventions stated previously. Abbreviating the initial states $|\lambda,t_0\rangle$ by $|\lambda\rangle$, $\lambda=a$ or k, $h(t)$ for the time-dependent, spinless, Newns-Anderson Hamiltonian is given in Eq. (4):

$$h(t) = \sum_k \varepsilon_k |k\rangle\langle k| + \varepsilon_a(t)|a\rangle\langle a| + \sum_k [V_k(t)|k\rangle\langle a| + V_k^*(t)|a\rangle\langle k|], \tag{4}$$

where the first term refers to a band of states, characterized by one-electron energies ε_k, the second to the localized level in the incident atom, with time-dependent energy $\varepsilon_a(t)$, and the last term represents the one-electron coupling between the atomic level and the band. In our model [21,22], we assume that $V_k(t)=V_k u(t)$, and that the level width has a Lorentzian form, as in Eq. (5):

$$\Lambda(\varepsilon) \equiv \pi \sum_k |V_k|^2 \delta(\varepsilon - \varepsilon_k) = V^2\Gamma/[(\varepsilon-\varepsilon_c)^2 + \Gamma^2]. \tag{5}$$

In our applications of the model here, the time dependences are given by $u(t)=\exp(-\gamma\nu|t|)$ and $\varepsilon_a(t)=\varepsilon_{ai}+\Delta\exp(-2\gamma\nu|t|)$, where ν is the constant atomic speed, ε_{ai} is the atomic energy at large distances, $\varepsilon_{ai}+\Delta$ is the energy at closest approach, $t_0=-\infty$, and $t_f=+\infty$. The atomic occupation takes the form of Eq. (1), where $N_{occ}(t)=\int_{-\infty}^{E_F} d\varepsilon\,|S(\varepsilon,t)|^2$ $[N_{un}(t)=\int_{E_F}^{\infty} d\varepsilon\,|S(\varepsilon,t)|^2]$. The amplitudes $S_a(t)$ and $S(\varepsilon,t)$ can be written in terms of confluent hypergeometric functions [21,22,24]. We should note that, for a full band, the SO occur for $\Gamma \leqslant 2V$ (i.e., the narrow band regime), otherwise the survival probability decreases monotonically (i.e., the wide band regime) [21,22], in agreement with independent calculations [10].

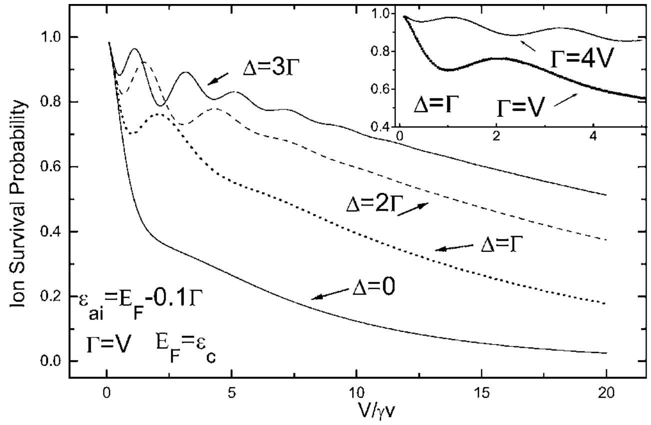

FIG. 1. The ion survival probability for an ion interacting with a half filled, narrow band ($\Gamma = V$) as a function of $V/\gamma\nu$ and Δ is varied from 0 to 3Γ. The atomic level is chosen to be slightly below the Fermi level (i.e., 0.1Γ below) initially and finally so that electron-hole pair production suppresses the SO for $\Delta = 0$ [22]. For the nonzero values of Δ, the level shifts above the Fermi level a distance comparable to the bandwidth, and an oscillatory behavior appears, which is the consequence of the process of oscillatory reionization of the ion. In the inset, we compare the wide band regime ($\Gamma = 4V$), where SO do not occur, with the narrow band regime ($\Gamma = V$), where SO can occur, indicating that the oscillations are not SO (all other parameters are the same as in the main part of the figure).

In Fig. 1, we present a graph of the ion survival probability, $P_a(\infty)$, as a function of the dimensionless parameter $V/\gamma\nu$ for a half full band. The main part of Fig. 1 corresponds to the narrow band regime ($\Gamma = V$). The atomic level is chosen to be slightly below the Fermi level (i.e., 0.1Γ below) initially and finally and Δ is varied from 0 to 3Γ. When there is no level shift (i.e., $\Delta = 0$), $P_a(\infty)$ decreases monotonically, despite the narrowness of the band and the consequent possibility of SO. This absence of SO occurs because of the production of electron-hole pairs [22]. When we vary the atomic level shift in time, we observe an increase in $P_a(\infty)$, which reflects the increased resonant interaction with the initially unoccupied states (i.e., reionization) and the decreased resonant interaction with the initially occupied band states, as well as the appearance of oscillations in $P_a(\infty)$. The decreased resonant interaction with the states at the band center is probably the reason that $S_a(\infty)$ (i.e., memory) is non-negligible for lower velocities than when the atomic level is constant.

In order to demonstrate explicitly that the new oscillations are not SO, we compare, in the inset, $P_a(\infty)$ for the narrow band regime (i.e., $\Gamma = V$) and for the wide band regime (i.e., $\Gamma = 4V$); the other parameters correspond to those in the main part of Fig. 1. We note that oscillations appear in both regimes, even though, for a constant atomic level resonant with the band center, SO in $P_a(\infty)$ are produced only in the narrow band regime [21].

In order to better understand these oscillations, we present the component quantities $S_a(\infty)$, $N_{occ}(\infty)$, and $N_{un}(\infty)$, as well as $P_a(\infty)$ for the case of $\Delta = 2\Gamma$ in Fig. 2. The atomic component, $S_a(\infty)$ is a monotonically decreasing function of $V/\gamma\nu$ and becomes smaller than both of the other components for $V/\gamma\nu > 2$ and rapidly becomes negligible thereafter. For $1 < V/\gamma\nu < 6$, both $N_{occ}(\infty)$ and $N_{un}(\infty)$ oscillate out of phase, in a manner reminiscent of that of the Stueckelberg oscillations illustrated in the inset (the inset was prepared for a full, narrow band, with $\Gamma = 0.1V$, and an atomic level resonant with the band center, but is meant to sketch the behavior qualitatively). Such behavior would suggest interference between the states initially occupied and unoccupied. Still lower velocities tend to destroy the interference as they do when there are SO. For $V/\gamma\nu \leq 1$, $N_{un}(\infty)$ rises monotonically with increasing $V/\gamma\nu$, whereas $N_{occ}(\infty)$ and $P_a(\infty)$ [which is the sum of $S_a(\infty)$ and $N_{un}(\infty)$] manifest out of phase oscillations. In view of the monotonic natures of $S_a(\infty)$ and $N_{un}(\infty)$ in this range, such oscillatory behavior would suggest a complicated interference in this range between the occupied band states, on the one hand, and the atomic and unoccupied band states on the other.

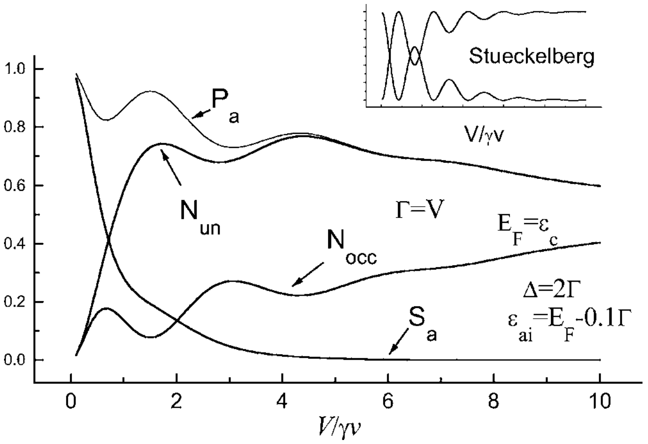

FIG. 2. $P_a(\infty)$ and the atomic $[S_a(\infty)]$, initially occupied $[N_{occ}(\infty)]$, and unoccupied $[N_{un}(\infty)]$ components as a function of $V/\gamma\nu$ for the $\Delta = 2$ curve in Fig. 1. The out-of-phase oscillations of $N_{occ}(\infty)$ and $N_{un}(\infty)$ for $S_a(\infty) \cong 0$ suggest the presence of interference between the occupied and unoccupied states, in analogy with the Stueckelberg oscillations in the inset which were calculated for a constant atomic level in resonance with the center of a full, narrow ($\Gamma = 0.1V$). We omit the argument "∞" in the figure.

In order to further elucidate the behavior in Figs. 1 and 2, we present, in Fig. 3(a), $P_a(t)$ as a function of the dimensionless time $\gamma\nu t$ for the nonzero values of Δ and the other parameters used in Fig. 1 and for $V/\gamma\nu = 4$. The larger the value of Δ, the earlier the level rises above the Fermi energy, the less the atomic level interacts with the occupied states at the band center and the more it interacts with the unoccupied states above E_F: the result of this process is an increase in the overall scale of $P_a(t)$ because of the increased reionization. The first minimum before the turning point in all three curves reflects this process: initially, the ion is increasingly neutralized in the incoming part of the trajectory; after it crosses E_F, it interacts more with the empty states and less with the full states, until the atom starts to get reionized. For $\Delta = \Gamma$, $P_a(t)$ is symmetric around $t = 0$, and the maximum corresponds to the inverse of the process we have described. For higher values of Δ, the maxima are no longer symmetrically

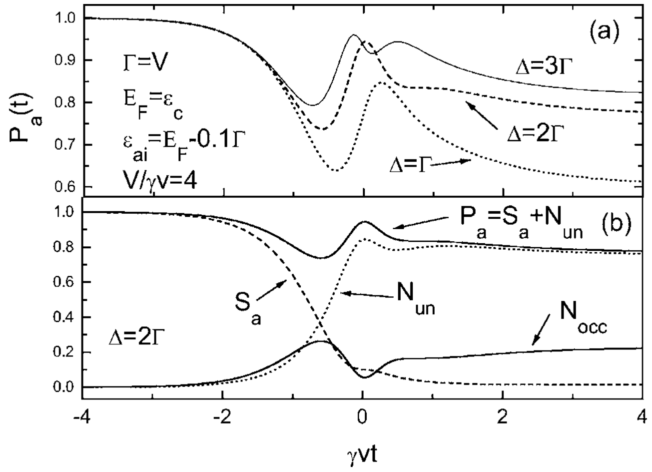

FIG. 3. (a) $P_a(t)$ as a function of $\gamma\nu t$ for the nonzero Δ values in Fig. 1 and $V/\gamma\nu=4$. The overall scale of $P_a(t)$ increases with Δ because of increased coupling with the unoccupied states and decreased coupling with the occupied. The oscillations which appear correspond to oscillatory reionization. (b) $P_a(t)$ and the atomic $[S_a(t)]$, initially occupied $[N_{occ}(t)]$ and unoccupied $[N_{un}(t)]$ components as a function of $\gamma\nu t$ for the $\Delta=2$ curve in Fig. 1. $S_a(t)$ decreases monotonically and the oscillations in $N_{un}(t)$ and $N_{occ}(t)$ when $S_a(t)\cong 0$ suggest interference between the occupied and unoccupied states.

placed and oscillations appear as the atomic level shifts up a distance comparable to the bandwidth.

In Fig. 3(b), we display the results in detail for $\Delta=2\Gamma$, where we present $P_a(t)$, as well as the components $S_a(t)$, $N_{occ}(t)$, and $N_{un}(t)$. Initially, $S_a(t)$ decreases monotonically, reflecting the strong ion-solid interaction discussed above, and $N_{un}(t)$ increases monotonically: these two contributions combine to produce the first minimum in $P_a(t)$. $N_{un}(t)$ has a maximum for $\gamma\nu t\cong 0$ and a small minimum for $\gamma\nu t\approx 0.5$, which corresponds to out of phase oscillation in $N_{occ}(t)$. Since $S_a(t)$ is very small here, these oscillations would appear to be caused by interference between the initially occupied and unoccupied band states. Two tendencies control the oscillations: first, the atomic level interacts with the unoccupied band states with which it is resonant; the other tendency is for the atomic level to interact with states near the Fermi level. The first tendency would dominate and there would not be oscillations if the band were flat, since, in that case the atom would couple equally with all band states it crosses. Since the band has a shape which varies with energy, the two tendencies can compete, producing oscillatory reionization of the atom. It is as though the atom could “feel” the band variable shape as its energy level shifts with time. Calculations of $|S(\varepsilon,t)|^2$, which represents the contribution of band states to charge transfer, indicate a strong change in the weights of the states near and above the center of the band as time is varied [26]. This seems to confirm that the atom “feels” the band variable shape. As we noted in our discussion of the inset in Fig. 1, the above-described behavior also occurs in the wide band case (for $\Gamma\geqslant 2V$), when there are no SOs [21], since the existence of oscillatory reionization requires only that the atomic level shift be comparable to the bandwidth.

Finally, we address the question of whether oscillatory reionization is observable. For this process to occur, it is necessary that the atomic level shift a distance comparable to the bandwidth (in order to interact resonantly with the initially empty band states) and that it cross the Fermi level sufficiently far from the target (in order to weaken the interaction with the states near the band center). From consideration of the energetics [27], this behavior of the atomic level shift occurs for a variety of alkali ions near free-electron metals. Such considerations have been applied to Na^+ scattering from copper surfaces [28]; our results would suggest that the oscillations we report may be observable for alkali ions scattering from transition-metal surfaces, especially where the density of states is high near the Fermi level (for example, Na^+ scattering from nickel surfaces). More experimental evidence is needed, however, for us to conclude that oscillatory reionization is important for understanding experimental results. Nevertheless, it is clear that this phenomenon should be taken into account in more elaborate theoretical treatments.

In conclusion, we present evidence of interesting oscillations in the ion survival probability whose magnitude is similar to that of the more usual Stueckelberg oscillations. The origin of these oscillations appears to be an unexpected type of quantum-mechanical interference among the metallic states. The use of a soluble model permits identification of these oscillations.

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