Signature of the s-wave regime high above ultralow temperatures

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Physical processes involving charge transfer, spin exchange, or excitation exchange often occur in conditions of resonant scattering. We show that the s-wave contribution can be used to obtain a good approximation for the full cross section. This approximation is found to be valid for a wide range of scattering energies, including high above the Wigner regime, where many partial waves contribute. We derive an analytical expression for the exchange cross section and demonstrate its relationship to the Langevin cross section. We give examples for resonant charge transfer as well as spin-flip and excitation exchange. Our approximation can be used to gain information about the s-wave regime from data obtained at much higher temperatures, which would be advantageous for systems where the ultracold quantum regime is not easily reachable.

In recent years, rapid progress has been made to increase the number of systems which can be studied at ultralow temperatures, including atomic systems,[1] and also molecular[2–5] and ionic species.[6, 7] In many cases, the quantum regime where s-wave scattering dominates is still outside the reach of today’s experimental techniques, such as in atom-ion hybrid system[8–10]. However, a large class of physical systems is characterized by two states that are asymptotically degenerate, and for which an initial scattering state can be described by a superposition of those states; interference between the two possible interaction paths may lead to resonant exchange between the two states. Such processes have been studied in the scattering of neutral atoms, e.g., spin-flip in alkali atom collisions[11–13] with singlet and triplet potential curves, as well as in S–P excitation exchange for identical atoms[14], and charge transfer between an atomic ion and its neutral parent atom.[20, 21] Moreover, in cases involving quasi-resonant scattering, e.g., when considering different isotopes, the resonant approximation adequately describes the behavior of the system if the scattering energy is higher than the energy splitting between the asymptotic states.[22, 23]

In this Letter, we study the resonant exchange process

\[ X^α + X^α' \rightarrow X^α' + X^α, \]

where \( α \) and \( α' \) denote internal states. For example, in charge transfer \((X + X^- \rightarrow X^- + X)\) \( α \) denotes the charge, with \( α = 0 \) and \( α' = +1 \), while for excitation exchange \( α = S \) and \( α' = P \) are the electronic states. For such resonant exchange processes, the cross section reads[6, 20, 24]

\[ σ_{\text{exc}}(E) = \frac{π}{k^2} \sum_{\ell=0}^{∞} (2\ell + 1) \sin^2(η_\ell^{(a)} - η_\ell^{(b)}), \]

where \( k = \sqrt{2\mu E/\hbar^2} \) is the center of mass wave number for the scattering of a pair of particles of reduced mass \( \mu \) and collision energy \( E \). Here, \( η_\ell^{(a/b)} \) is the scattering phase shift of the \( \ell \)th partial wave along the interaction potential \( V_{a/b}(r) \), which correspond to the two asymptotically degenerate channels.

For energies high above the Wigner regime, where many partial waves are contributing, we can regard \( \ell \) as a continuous variable and use the semi-classical expression[24, 25]

\[ \frac{∂η_\ell}{∂\ell} \approx \frac{π}{2} + \int_{r_0(J)}^{∞} dr \frac{∂J}{∂J} \left[ 2μ(E - V(r)) - \frac{J^2}{r^2} \right]^{1/2}, \]

where \( J = (\ell + \frac{1}{2})h \), and \( r_0(J) \) is the inner classical turning point. Although \( \ell \) can be large, we assume that the centrifugal term \( J^2/r^2 \) is small perturbation on the potential \( V(r) \), i.e., \( J^2/r^2 \ll 2μ(E - V(r)) \). Under such conditions, the scattering wave function still probes the inner region, and \( r_0 \) depends weakly on \( J \); we thus take it to be independent of \( J \) and equal to the s-wave turning point, i.e., \( r_0(J) \approx r_0 \). We now take the partial derivative out of the integral (3) and expand the integrand in small powers of \( J^2/r^2 \) to obtain

\[ \frac{∂η_\ell}{∂\ell} \approx \frac{π}{2} - J \int_{r_0}^{∞} dr \frac{1}{\sqrt{2μ(E - V)}} \approx \frac{π}{2} - \frac{J^2}{4A}, \]

where \( A \) is an integral independent of \( J \). Using \( J = (\ell + \frac{1}{2})h \), we have \( \frac{∂η_\ell}{∂\ell} \approx π/2 + A(\ell + 1/2) + \mathcal{O}(\ell^2) \), which after integration over \( \ell \) gives

\[ η_\ell \approx η_0 + \frac{π}{2} \ell + \frac{A}{2}(\ell + 1) + \mathcal{O}(\ell^2). \]

Using the Levinson theorem[24, 25], we have \( η_0 = Nπ + δ_0 \), where \( δ_0 \) is the s-wave phase shift modulo \( π \) and \( N \) is the number of bound states. Therefore, the phase shift difference \( Δη_\ell \equiv η_\ell^{(a)} - η_\ell^{(b)} \) reads

\[ Δη_\ell \approx πN + Δδ_0 + η_0 + π(\ell + 1) + \mathcal{O}(\ell^2). \]

where \( ΔN = N_a - N_b \), \( Δδ_0 = δ_0^{a} - δ_0^{b} \), and \( ΔA = A_a - A_b \), and we obtain

\[ \sin^2(Δη_\ell) \approx \sin^2 \left[ \frac{1}{2} (Δδ_0 + η_0 + π(\ell + 1) + \frac{ΔA}{2}) \right]. \]

We now approximate the sum in Eq. (2) with an integral, \( σ_{\text{exc}} \approx \frac{π}{k^2} \int_0^{L} d\ell (2\ell + 1) \sin^2(η_\ell^{(a)} - η_\ell^{(b)}) \), which yields

\[ σ_{\text{exc}} \approx \frac{π}{k^2} \int_0^{L} d\ell (2\ell + 1) \sin^2 \left[ Δδ_0 + η_\ell^{(a)} + \ell(1 + \frac{ΔA}{2}) \right]. \]

In the integral above, the upper limit \( L \) is set to a sufficiently large value of \( \ell \) such that the two phase shifts become equal; thus, there will be no further contribution to the integral for \( \ell > L \). This occurs when the centrifugal barrier becomes dominant for both potential curves[20]. Changing variable to
\[
x = \Delta \delta_0 + \ell (\ell + 1) \frac{A_0}{\Delta A}, \text{ our integral simply becomes } \sigma_{\text{exc}} \simeq \frac{\pi}{2} \frac{1}{\Delta A} \int_{\mu}^{1} dx \sin^2 x = \frac{\pi}{2} \frac{1}{\Delta A} \left[ \frac{1}{2} - \frac{3}{4} \sin(2x) \right]_{\mu_0}, \text{ with } \mu_0 = \Delta \delta_0 \text{ and } \mu_L = \Delta \delta_0 + L(L + 1)\Delta A/2, \text{ giving } \\
\sigma_{\text{exc}} \simeq \frac{\pi}{2} \frac{1}{\Delta A} \left[ L(L + 1) \frac{\Delta A}{2} + \frac{1}{2} \sin(2\Delta \delta_0) - \frac{1}{2} \sin(2\Delta \delta_0 + L(L + 1)\Delta A) \right]. \tag{9}
\]

Finally, we can simplify our result if we employ the approximation \(L(L + 1)\Delta A \ll 1\), which can be justified if we examine the parameter \(A_i\) given by Eq. (11), i.e.
\[
A_i = \frac{\hbar}{\sqrt{2\mu}} \int_{r_i(E)}^{r_0} \frac{dr}{\sqrt{E - V_i(r)}}, \tag{10}
\]
where the inner turning point \(r_i\) for the potential \(V_i\) depends on the scattering energy \(E\). In resonant processes where the long-range tail of each \(V_i\) is the same, only their shorter range difference contribute to \(\Delta A\). Typically, \(\Delta A\) varies little with \(E\), and is of the order 0.01–0.001 for the physical systems considered in this Letter, with \(\Delta A\) smaller for heavier systems due the \(2\mu\) factor. We now return to Eq. (9) and use the approximation \(\sin(2\Delta \delta_0 + L(L + 1)\Delta A) \approx L(L + 1)\Delta A \cos(2\Delta \delta_0) + \sin(2\Delta \delta_0)\) to obtain
\[
\sigma_{\text{exc}}(E) \simeq \frac{\pi}{2} \frac{1}{\Delta A} \left[ L(L + 1) \frac{\Delta A}{2} \left[ 1 - \cos(2\Delta \delta_0) \right] \right], \tag{11}
\]
where we assume \(L(L + 1) \approx L^2\).

The expression above can be related to the Langevin cross section \(\sigma_{\text{L}}\). Indeed, we defined \(L\) as the maximum \(\ell\) for which the phase shift difference is non-negligible, which corresponds to the height of the centrifugal barrier slightly larger than \(E\). This critical value of \(\ell\) also defines \(\sigma_{\text{L}}\), which is determined by the impact parameter \(b_{\text{max}}\) still allowing penetration in the inner region where the exchange process occurs with unit probability \([6, 20, 24]\). With the impact parameter \(b \equiv (\ell + \frac{1}{2})/k\), we obtain
\[
\sigma_{\text{L}}(E) = \pi b_{\text{max}}^2 \simeq \frac{\pi}{k^2} L^2, \tag{12}
\]
where we assume \(L + \frac{1}{2} \approx L\) for large \(L\). We remark that \(L\) has the same value for both potentials \(V_a\) and \(V_b\), which is a valid assumption for the energy range dominated by the long range tail (which is the same for both potentials). For potentials with an asymptotic behavior \(V(r) \sim -C_n/r^n\), the location of the top of the barrier is \(r_{\text{top}} = \left(\frac{\mu C_n}{(\ell + 1)k^2}\right)^{\frac{1}{n+2}}\), and \(L(E)\) is obtained from \(E = V(r_{\text{top}})\), which yields
\[
L(L + 1) = \frac{1}{k^2} \left( n \frac{n - 2}{n - 2} \right)^{\frac{1}{2}} (\mu n C_n)^{\frac{1}{2}} (2\mu E)^{\frac{n - 2}{2}}. \tag{13}
\]
Again, assuming \(L(L + 1) \approx L^2\), we can write the Langevin cross section \(\sigma_{\text{L}} = \frac{\pi}{k^2} L^2\) as
\[
\sigma_{\text{L}}(E) = \pi \left( n \frac{n - 2}{n - 2} \right)^{\frac{1}{2}} (\mu n C_n)^{\frac{1}{2}} (2\mu E)^{-\frac{n - 2}{2}}. \tag{14}
\]

The expressions for the most common long range inverse power-law potentials are listed in Table I with \(n = 3\) appearing in dipole allowed excitation exchange, \(n = 4\) in polarization potentials between atoms and ions, and \(n = 6\) in van der Waals interactions between ground state atoms.

| \(n\) | \(3\) | \(4\) | \(6\) \\
|-----|-----|-----|-----|
| \(\sigma_{\text{L}}\) | \(3\pi (C_3 / 2E)^{2/3}\) | \(2\pi (C_4 / E)^{1/2}\) | \(3\pi / 2 \times 2C_6 / E^{1/3}\) |

Table I: Langevin cross section \(\sigma_{\text{L}}\) for various \(n\).

This equation explicitly shows how the \(s\)-wave regime modulates the Langevin cross section, leading to a signature of the \(s\)-wave regime at higher temperatures.

To illustrate the effect of the \(s\)-wave regime at higher energies, we first consider charge transfer between Yb and Yb\(^{+}\) for a variety of isotopes. As noted in a previous article on resonant charge transfer \([26]\), the cross section exhibits an intermediate “modified” Langevin regime where the \(\sigma_{\text{exc}}\) seems to be affected by the ultracold behavior, even if many partial waves contribute to its overall value. In fact, one could notice a “correlation” between the cross section at ultralow energy and at much higher energies. The expression above provides the explanation for the correlation noted in \([26]\): if the \(s\)-wave phase shifts corresponding to states \(a\) and \(b\) happen to have nearly equal values, their difference remains small even at higher scattering energy. This can be seen from the WKB approximation, or equivalently Eq. (3), as \(n_i\) varies slowly with \(\ell\). If \(\Delta \delta_0\) is small, the phase difference for higher \(\ell\) will also remain small for a wide range of partial waves, due to the phase shift “locking” described above, and will result in a reduced cross section. Naturally, the applicability of Eq. (15) depends on the details of the potentials and validity of the approximations involved in its derivation.

In Fig. 1 we compare the simple expression (15) to the full numerical results computed using the approach described in \([26]\). The potentials \(V_k\) and \(V_b\) corresponding to the \(2\Sigma_g^+\) and \(2\Sigma_u^+\) of Yb\(^{+}\) behave as \(-C_n/r^n\) with \(C_4 = 72.5\) a.u. at large separation. The cross section \(\sigma_{\text{exc}}(E)\) depends strongly on the atomic mass of the Yb isotopes. In each plot of Fig. 1 the “standard” Langevin cross section \(\sigma_{\text{L}}\) is included to emphasize the size of the \(s\)-wave phase shifts. In some cases, like plots (b), (c), and (d), corresponding to isotopes 170, 172, and 173, both \(\sigma_{\text{L}}\) and \(\sigma_{\text{exc}}\) give similar values, i.e. the \(s\)-wave has no sizable effect on the cross section. Actually, \(\sigma_{\text{exc}}\) is roughly \(\frac{1}{2} \sigma_{\text{L}}\), which is to be expected in general since \((\sin^2 \Delta \delta_0) = 1/2\) if the phase shift difference \(\Delta \delta_0\) is random. However, in other
cases, like for isotopes 168, 174, or 176 in (a), (e) and (f) respectively, the signature of the s-wave regime is noticeable, with a reduction of two orders of magnitude for (a) and (e), and one for (f). As mentioned above, this is due to the accidental proximity in values of the residual phase shifts $\delta^{r}_{0}$ corresponding to $V_{g(a,i)}$, and the phase shift locking as $E$ increases. We note that according to $E^{15}$, $L^2 < 1$ when $E$ becomes smaller than roughly $10^{-13}$ a.u. for the Yb systems above, at which point the s-wave contribution (negligible at higher $E$) satisfying the Wigner regime kicks in. It is worth noting that when the s-wave suppression of $\sigma_{exc}$ is significant, as in Figs. 1(a) and (e), the underlying shape resonances become more apparent as the background cross section diminishes. Naturally, these resonances are absent from our WBK-treatment in Eq. (15), which reproduces the general trend of the numerical results over a large range of $E$.

Atom-ion scattering can also lead to a resonant spin-flip process, such as in $^{27,28}$, where a ground state Na atom approaching a Ca$^+$ ion can interact via a singlet $A^1\Sigma^+$ or a triplet $a^3\Sigma^+$ electronic state described by the singlet (triplet) potential $V_{S(T)}$ with corresponding residual phase shift $\delta^{S(T)}$. In that work, $\sigma_{exc}$ was found to be roughly $\frac{1}{2}\sigma_{L}$. Recent experiments on Yb$^+$+$^{87}$Rb $^{12}$, Yb$^+$+$^{4}$Li $^{29}$, and $^{88}$Sr$^+$+Rb $^{30,31}$ have explored spin-flip dynamics. In Fig. 2 we explore the effect of the s-wave scattering on the spin-flip in Ca$^+$+Na, using $V_{S(T)}$ described in $^{27,32}$ (behaving as $r^{-4}$ at large $r$) for four isotopes of Ca, namely 40, 42, 43, and 44. Again, the simple expression $^{15}$ agrees with the numerical cross sections over a wide range of energy. Fig. 2 shows a variety of behavior of $\sigma_{exc}$. For example, in (a) and (d), $\sigma_{exc} \approx \sigma_{L}$ at higher energies, corresponding to $\Delta \delta_{0} = \delta^{s}_{0} - \delta^{t}_{0} \approx \pi/2$, while (c) depicts a small suppression by a factor of about 1.5. The case of $^{40}$Ca leads to a substantial reduction of about 200, again revealing the underlying shape resonances.

Spin-flip collisions have also been studied between neutral atoms, especially alkali atoms, such as in Li $^{17}$ or Na $^{18}$. Again, the ground state atoms approach each other in a superposition of singlet $X^{1}\Sigma^{+}$ and triplet $a^{3}\Sigma^{+}$ states, behaving asymptotically as $-C_{6}/r^{6}$. To illustrate the effect of the s-wave regime on $\sigma_{exc}$, we consider a system for which the scattering lengths are known to be close to each other, namely $^{87}$Rb. Using the potential curves described in $^{33}$, we computed $\sigma_{exc}$ for pure $^{87}$Rb, $^{85}$Rb, and their mixture. The results are shown in Fig. 3. $\sigma_{exc}$ for the mixture in (a) follows roughly $\sigma_{L}$ away from ultracold temperatures. As expected, for $^{87}$Rb in (b) with both singlet and triplet scattering lengths almost equal ($a_{S} \approx a_{T} \approx 100$ a.u.), the s-wave suppression is drastic, with shape resonances emerging from the suppressed background. Although not perfect, the simple expression $^{15}$ tracks the overall reduction of a factor of $10^{4}$ in $\sigma_{exc}$. Much more surprising is the result for $^{85}$Rb (c) with scattering lengths ($a_{S} \approx 2500$ a.u. and $a_{T} \approx -390$ a.u.) which are very different. In this case, one could have expected the system to follow the Langevin case. However, a closer look at the s-wave phase shifts explains the seemingly unusual cross section. The large positive and negative values of $\delta_{S}$ and $\delta_{T}$ imply rapid changes of $\delta_{S(T)}$ with energy in the Wigner regime, as shown in Fig. 3(d). The large initial value of the phase shift difference $\Delta \delta_{0} (\text{mod } \pi)$ quickly evolves into a much smaller

FIG. 1. (Color on line) Resonant charge transfer $\sigma_{exc}$ between various isotopes of Yb and Yb$^+$ vs. scattering energy $E$. The numerical results (black line) are compared to Eq. (15) (magenta line), together with its components; the s-wave contribution $\frac{\sigma_{L}}{2} \sin^{2} \Delta \delta_{0}$ alone (black dashed line) and the $\sigma_{L} \sin^{2} \Delta \delta_{0}$ alone (solid blue line). The standard $\sigma_{L}$ (blue dot-dashed line) is shown for comparison purposes. Isotopes 168 in (a), 174 in (e), and 176 in (f) show significant suppression when compared to $\sigma_{L}$, while 170 in (b), 172 in (c), and 173 in (d), $\sigma_{exc} \approx \frac{1}{2} \sigma_{L}$ over a wide range of energies.

FIG. 2. (Color on line). Same as Fig. 1 for spin-flip in collision of Na and various isotopes of Ca$^+$, 40 in (a), 42 in (b), 43 in (c), and 44 in (d). Significant suppression occurs in (b), while $\sigma_{exc}$ is close to $\sigma_{L}$ for the other isotopes.
σ (a.u.)

σ_{exc} (a.u.)

σ_{exc} = \frac{\pi}{3k^2} \sum_{\ell=0}^{\infty} (2\ell + 1) \left[ \sin^2 \Delta \delta_{\Sigma} + 2 \sin^2 \Delta \delta_{\Pi} \right], \quad (16)

\Delta \delta_{\Sigma} = \delta_{\Sigma,\ell}^s - \delta_{\Sigma,\ell}^u

\Delta \delta_{\Pi} = \delta_{\Pi,\ell}^s - \delta_{\Pi,\ell}^u.

Since the Σ and Π curves have different C_3 values, L for both sets is different. Using our approximations, \sigma_{exc} becomes

\sigma_{exc} = \frac{1}{3} \left[ \frac{\pi}{k^2 + \sigma_L^u} \right] \sin^2 \Delta \delta_{\Sigma} + \frac{2}{3} \left[ \frac{\pi}{k^2 + \sigma_L^u} \right] \sin^2 \Delta \delta_{\Pi}, \quad (17)

where \sigma_{L(\Sigma)} is obtained with the appropriate value of C_3.

The results shown in Fig. 3 were obtained with the Σ_{g(u)}^+ and Π_{g(u)}^+ from Jrai et al. [33]. The Π curves are repulsive at large separation behaving as +C_3^{\Pi} / r^3 with C_3^{\Pi} = -13.95 a.u.; the gerade and ungerade phase shifts are basically equal for all \ell, their cancellation leading to a negligible Π contribution. The two Σ curves are attractive, and were matched at large separation to −C_4 / r^3 − C_5 / r^5 with C_4 = 1082 a.u., and C_5 = 27.9 a.u. Since there is only one stable isotope of Cs, we rescaled its mass to model a different isotope. For the real mass of Cs, the cross section is roughly half the Langevin cross section, while choosing m_{Cs} = 132.75 u, the cross section is reduced by a factor of 20, again exposing the resonances as in previous examples.

In conclusion, we derived a simple expression for resonant scattering processes, relating the cross section to the Langevin cross section and the s-wave regime. By relying on the WKB approximation, we derived the expression for the exchange cross section, and showed that it has wide applicability, as long as the pairs of potential curves have the same long range tail. We illustrated the range of applicability using various resonant systems such as charge transfer, spin-flip, and excitation exchange, and for a variety of long range inverse power-law tail behaving as r^{−n} covering the most common powers. The expression points to the signature of the s-wave regime at higher temperatures, and how the s-wave phase shift “locking” actually modulates the cross section. The results presented here also provide a diagnostic tool particularly relevant to system for which ultracold temperatures are not easily achievable, such as atom-ion hybrid systems for which the nK regime remains a challenge. In fact, by measuring the cross section or rate for a resonant process, e.g., charge transfer or spin-flip, at higher temperatures more easily accessible, one can gain information about the s-wave regime. If a sizable suppression is observed as compared to \sigma_s, this implies that the s-wave phase shifts are close to each other. In addition, the suppression helps revealing shape resonances otherwise submerged which, together with the s-wave suppression, can help determining the potential curves more accurately down to the s-waves. Finally, the expression should be applicable to quasi-resonant processes as well [24], such as charge transfer in systems with mixed isotopes [22, 23], or in reactions involving isotope substitutions, as long as the scattering energy is larger than the energy gap between the asymptotes of the relevant potentials.

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FIG. 4. (Color on line). Same as Fig. 1 for excitation exchange in Cs_6+Cs(6p), for the real mass in (a), and a fictitious mass of 132.75 u in (b), to illustrate the s-wave suppression.
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