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Slow heavy dimers in a two-dimensional degenerate electron gas: Calculation of orientation-dependent retarding forces

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We present a theoretical study on the orientation-dependent retarding force experienced by slow dimers moving at arbitrary alignment with the direction of their velocity in a two dimensional degenerate electron gas. The influence of the individual constituents of a dimer on independent electrons is modeled by effective, *s*-type phase shifts η. Analytical results are derived by considering the effects of interference and multiple scattering. The orientation-dependent expressions reveal the dependence of stopping power on the classical geometry and these effects of quantum dynamics. A brief account on strongly related problems of resistivity caused by imperfections in the many-body system and pure dephasing of elastically scattered electrons is made, as well. The physically motivated case of $\eta(k_F)=\pm(\pi/2)$ for the leading phase shift is explicitly discussed.

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I. INTRODUCTION AND MOTIVATION

Electronic excitations play an important role for the kinetic energy losses of heavy particles moving through the three-dimensional electron gas of a metal, or making displacements in the two dimensional fermion systems of semiconductor heterostructures[1,2] of tunable densities, and on close packed noble metal surfaces.[3,4] The retarding forces experienced by *slow* heavy particles along their classical trajectories are due primarily to the creation of propagating elementary composite excitations, named as electron-hole pairs. A deep understanding of details contributing to an effective (frictionlike) retarding force is of utmost importance[5] to realistic estimations needed, for instance, in computer-aided simulations of dynamics of slow heavy particles in three- and two dimensional fermion systems. Furthermore, there is an inherent relation between the low-velocity retarding force (stopping power) and the embedded-impurity contribution to residual resistivity.

The expression for the magnitude of the energy-transfer per unit path length for a single scattering center moving slowly ($v<v_F$) with velocity v, is well known and is given by

$$\frac{1}{v}\frac{dE}{dt}=m_e v n_0 v_F \sigma_{tr}(v_F) \tag{1}$$

in three and two dimensions.[6,7] Here $n_0(D)$ and $v_F(D)$ are, respectively, the density and Fermi velocity of the degenerate electron gas and $\sigma_{tr}(D)$ is the transport cross section.[8–11] The friction coefficient is defined via $(dE/dt)/(Mv^2)$ for a slow particle with mass $M \gg m_e$. One can interpret the left-hand side (lhs) of Eq. (1) in terms of an averaged density of excited electrons $[n_0(v/v_F)]$, an averaged energy change $(m_e v_F^2)$ of these electrons, and an effective area or length (depending on D) for the two-body elastic scattering process. Alternatively, one can interpret the retarding force on the lhs in terms of an electron flux $(n_0 v_F)$ on the moving scattering center and the net momentum change $(m_e v)$ governed by the momentum transfer cross section $\sigma_{tr}(v_F)$. From now on we use atomic units, $\hbar=m_e=e^2=1$.

It is the method of the kinetic or transport theory on which the second interpretation for a moving single center is based. Such a method for the retarding force via the momentum change in a flux of electrons was used already for dimers moving in a three-dimensional electron gas.[12–14] Within this kinetic framework the role of *inhomogeneity* of the electron gas above metal surfaces of has been modeled[15] for atomic intruders, as well. As a natural extension for dimers, we investigate the case of a two dimensional electron gas in this work. Clearly, a proper understanding of various experimental data with dimers, for instance, at surfaces with molecules,[16–19] requires such a theoretical attempt on the details.

Section II is devoted to the quantum-mechanical theory of different cross sections for electron scattering off dimers in two dimension. The closed expressions derived are analyzed for the important limiting cases. Two illustrative figures are given in order to facilitate comparisons with the corresponding[14] three-dimensional results for retarding forces. Experimentally motivated, concrete possibilities for future *specific* applications are pointed out at the relevant places. In Sec. III, we give a short summary.

II. THEORY AND ANALYSIS

Following closely earlier detailed[12,14] works, the magnitude of the energy loss per unit path of a slow dimer moving in two dimensional electron gases can be written as a decomposition in terms of the parallel ($\|$) and perpendicular ($\perp$) transport cross sections

$$\left[\frac{1}{v}\frac{dE}{dt}\right](\alpha)=n_0 k_F v[\cos^2(\alpha)\sigma_{tr}^{\|}(k_F)+\sin^2(\alpha)\sigma_{tr}^{\perp}(k_F)]. \tag{2}$$

Here α denotes the polar angle between the dimer orientation and its velocity direction. In the plane of the many-body system two coordinate axes can be introduced via these vectors. To random situations the angle averages are $\langle\cos^2(\alpha)\rangle=\langle\sin^2(\alpha)\rangle=(1/2)$ in $D=2$. The directions of the scattering electron momenta, $\mathbf{k}$ and $\mathbf{k}'$, in the center-of-mass system, are in the above plane. This constraint is different from the $D=3$ case.[12,14]

In the knowledge of the (usually complex) single-particle scattering amplitude $F(\mathbf{k}_F,\mathbf{k}'_F)$, the differential cross section (length) is related to the $|F(\mathbf{k}_F,\mathbf{k}'_F)|^2$ quantity.[20] In our orientation-dependent problem with transfer cross sections, proper integrations over the $\theta_{\mathbf{K}} \in [0,\pi]$ interval with $d\theta_{\mathbf{K}}$ weighting are needed in both angles. The cross sections are

$$\sigma_{tr}^{\parallel}(k_F) = \frac{4}{\pi}\int d\theta_{\mathbf{k}_F}\int d\theta_{\mathbf{k}'_F}(\cos\,\theta_{\mathbf{k}_F} - \cos\,\theta_{\mathbf{k}'_F})\cos\,\theta_{\mathbf{k}_F}\frac{1}{k_F}|F(\mathbf{k}_F,\mathbf{k}'_F)|^2, \tag{3}$$

$$\sigma_{tr}^{\perp}(k_F) = \frac{4}{\pi}\int d\theta_{\mathbf{k}_F}\int d\theta_{\mathbf{k}'_F}\sin^2\,\theta_{\mathbf{k}_F}\frac{1}{k_F}|F(\mathbf{k}_F,\mathbf{k}'_F)|^2 \tag{4}$$

according to the above details on kinematics of elastic scattering in two dimension. In order to implement these expressions, one needs the complex scattering amplitude which encodes the effect of dimer geometry and potential field on the scattering of a one-electron wave.

A. Role of interference

In order to understand the *details* of two dimensional friction for dimers, and to provide an easily controllable background to future extensions, in this section we regard the two (not necessarily equivalent) centers, separated by a distance d, as scattering independently but coherently. Realistic[14] values of d are about $d \geq 2$. Thus, the effect of interference (without multiple scattering) on orientation-dependent friction will be analyzed. In order to achieve this goal, the dominating s-type partial waves are used for both constituents of a dimer. With these last, physically very reasonable and commonly applied[17,18,21] approximations in two dimensions, one gets

$$|F(\mathbf{k},\mathbf{k}')|^2 = \frac{2}{\pi}\{[\sin^2\,\eta^{(1)} + \sin^2\,\eta^{(2)}] + A_{12}\cos[(\mathbf{k}-\mathbf{k}')\cdot\mathbf{d}] + B_{12}\sin[(\mathbf{k}-\mathbf{k}')\cdot\mathbf{d}]\}, \tag{5}$$

where $A_{12} \equiv 2\,\sin\,\eta^{(1)}\,\sin\,\eta^{(2)}\,\cos(\eta^{(1)}-\eta^{(2)})$ and $B_{12} \equiv 2\,\sin\,\eta^{(1)}\,\sin\,\eta^{(2)}\,\sin(\eta^{(1)}-\eta^{(2)})$.

At this point we make a remark. In a first-order Born approximation, on which a dielectric description of energy loss of point-charge dimers under random condition[22,23] is based, one uses pure real scattering amplitudes, i.e., the Fourier transforms of screened interaction potentials. This perturbative approximation, which is perfectly reasonable for small phase shifts values, is recovered by the $\cos(\eta^{(1)} - \eta^{(2)}) \equiv 1$ and $\sin(\eta^{(1)}-\eta^{(2)}) \equiv 0$ necessary substitutions. Due to these, such a linear-response-based description cannot treat properly even a screened proton-antiproton ($Z=\pm 1$) dimer, the dipole composition of the smallest real charges. Earlier result[24] of a numerical calculation (performed in three dimensions with such a dipole) also signals the limitation of the dielectric description, i.e., the absence of a *sign change* via $\cos(\eta^{(1)}-\eta^{(2)})$ for not small $[|\eta^{(1)}-\eta^{(2)}| > (\pi/2)]$ phase shift differences.

The case of a conventional dimer is, fortunately, simpler. Due to parity consideration, the term proportional to B_{12} disappears after angle integrations ($\mathbf{K}\cdot\mathbf{d} = Kd\,\cos\,\theta_{\mathbf{K}}$) in the above Eqs. (3) and (4) with Eq. (5) for the scattering amplitude and we obtain the informative

$$\frac{k_F\sigma_{tr}^{\parallel}(k_F)}{4} = [\sin^2\,\eta^{(1)} + \sin^2\,\eta^{(2)}] + 2A_{12}\left[J_0^2(x) - J_1^2(x) - \frac{J_0(x)J_1(x)}{x}\right], \tag{6}$$

$$\frac{k_F\sigma_{tr}^{\perp}(k_F)}{4} = [\sin^2\,\eta^{(1)} + \sin^2\,\eta^{(2)}] + 2A_{12}\frac{J_0(x)J_1(x)}{x} \tag{7}$$

expressions, in which $x \equiv k_F d$ is a shorthand. These orientation-dependent, closed expressions encode the effects of dimer-geometry *and* wave-mechanical interferences in two dimensions. For the usual case of homonuclear dimers (molecules), it is illuminating to introduce dimensionless ratios (r), which are denoted now by $g_r^i(D,x)$, to the present approximation

$$g_r^{\parallel}(2,x) = 1 + 2\left[J_0^2(2,x) - J_1^2(x) - \frac{J_0(x)J_1(x)}{x}\right], \tag{8}$$

$$g_r^{\perp}(2,x) = 1 + 2\frac{J_0(x)J_1(x)}{x}. \tag{9}$$

For the asymptotic $x \gg 1$ limit, i.e., at high densities since d is bounded in a realistic dimer, one has $[g_r^{\parallel}(2,x)-1] \simeq (4\,\sin\,2x)/(\pi x)$ and $[g_r^{\perp}(2,x)-1] \simeq 2(1-2\,\cos^2\,x)/(\pi x^2)$. We can conclude that the important effect of wave mechanical interference results in a slowly decaying oscillating function for dimers aligned with the direction of their movement. Due to the special weighting, this remains the character of the proximity effect under random situations, as well. The minimal value of $g_r^{\parallel}(2,x)$ is at $x \approx 2$, and is roughly 0.3. Thus, the retarding force for a perfectly aligned dimer can be about 1/3 of the sum of forces for two independent atoms. By using earlier[14] results for $D=3$, in Fig. 1 we exhibit the D dependence of the $g_r^i(D,x)$ ratios. In the asymptotic $x \to 0$ limit, i.e., a very low densities since d is bounded from below, one gets simply the total *number* of dimer constituents.

We continue this subsection by pointing out that in a random situation with homonuclear dimers, and in D dimension, one gets after orientation averaging

$$\langle\cos(\mathbf{k}-\mathbf{k}')\cdot\mathbf{d}\rangle = \Gamma\left(\frac{D}{2}\right)\left(\frac{2}{qd}\right)^{(D-2)/2} J_{(D-2)/2}(qd), \tag{10}$$

where $q=2k\,\sin(\theta/2)$ is the momentum change in elastic scattering. In this case one can use directly[25] the averaged from

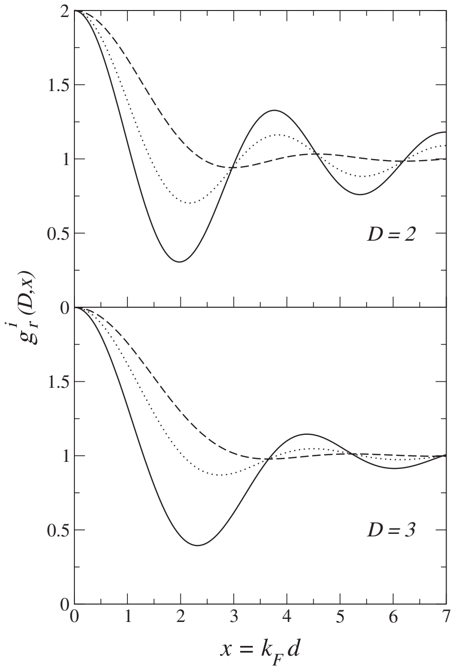

FIG. 1. Illustrative ratios, denoted as $g_r^i(D,x)$, showing the orientation dependence of renormalized stopping powers of D dimensional degenerate electron gases for in-moving slow homonuclear dimers. The parallel (‖) (solid curves), perpendicular (⊥) (dashed curves), and random (dotted curves) cases refer to fixed angles (see the text) between dimer orientation and its velocity direction. The curves are exhibited for the $x \in [0,7]$ range, where $x=k_F d$. The upper panel is devoted to the two dimensional ($D=2$) case, while the lower panel (based on Ref. 14) to the three dimensional ($D=3$) one. All curves are obtained in the perturbative impulse approximation.

$$\langle \sigma_{tr}(k) \rangle = \frac{4}{k_F} \int_0^{\pi} d\theta (1 - \cos\,\theta) \langle |F(\mathbf{k},\mathbf{k}')|^2 \rangle \tag{11}$$

to describe the average retarding force in two dimension. With the expansion of

$$J_0(qd) = \sum_{m=-\infty}^{\infty} J_m^2(k_F d) e^{im\theta} \tag{12}$$

one arrives at the above result obtained by proper weighting of force components. In the light of the remarkable oscillating character found in the retarding force, there is a quite natural question. What is the effect of proximity in an averaged total (t) cross section? Based on Eq. (11), but without the $(1-\cos\,\theta)$ factor, we obtain for the $D=2$ case

$$\langle \sigma_t(k_F) \rangle = \frac{4}{k_F} 2[1 + J_0^2(k_F d)] \sin^2\,\eta. \tag{13}$$

It is useful to compare this result with the three-dimensional ($D=3$) total cross section obtained earlier[26] by Brueckner under perfectly similar conditions

$$\langle \sigma_t(k_F) \rangle = \frac{4\pi}{k_F^2} 2\left[1 + \left(\frac{\sin(k_F d)}{k_F d}\right)^2\right] \sin^2\,\eta. \tag{14}$$

Both expressions show that there are oscillating *enhancements* with dimer impurities due to the proximity effect, in comparison with the case of well-separated atoms. The enhancement decay with separation is slow $\sim(1/d)^{(D-1)}$. The two dimensional result found here may be useful in interpretations of lifetime data measured[4,27] by excited electrons on close packed noble metal surfaces. Generally, a reasonable estimation of the electron mean-free path, in the presence of unavoidable or designed defects, requires the associated total cross sections. Remarkably, careful experiments[4,28] performed with molecules and well-separated atoms on Cu surfaces show notable differences in electron dephasing rates $\sim(k_F \sigma_t)$.

B. Influence of multiple scattering

Beyond the above-applied, so-called impulse approximation, now we outline the effect of multiple scattering on the vicinage functions by treating the case of homonuclear dimers with short-range potentials separated by d and characterized by a common s-type effective phase shift $\eta(k_F)$. We stress the point, once more, that such a modeling is very reasonable physically. The additional screening[17,29] by bulk electrons at close packed noble metal surfaces justifies the assumption that the scattering potentials of embedded pairs are nonoverlapping. Thus within our framework, one can get useful information on the nontrivial interplay of wave interference and multiple scattering in two dimensional quantum dynamics of electrons moving in the field of an in-plane dimer.

The approach applied in this subsection for two dimensions is strongly tied to standard methods established[30,31] for nonoverlapping (short-range) symmetric potentials in the more common three-dimensional case. In fact, the form of the solution for the key $F(\mathbf{k},\mathbf{k}')$ quantity is quite similar (see, below) in two and three dimensions. Technically, one has to solve two coupled algebraic equations[31] with single-particle inputs characterizing the individual scattering amplitude and the single-particle Green's function $G_D(\mathbf{x},\mathbf{x}')$. It is this well-known function of mathematical physics which describes the particle propagation with outgoing boundary condition, in free space

$$G_D(\mathbf{x},\mathbf{x}') = -\frac{i}{4}\left(\frac{k}{2\pi}\right)^{\nu} \frac{H_{\nu}^{(1)}(k|\mathbf{x}-\mathbf{x}'|)}{|\mathbf{x}-\mathbf{x}'|^{\nu}}, \tag{15}$$

where $\nu=(D-1)/2$, and $H_{\nu}^{(1)}(\rho)$ is the Hankel function of order ν. In the $D=2$ case one has $H_0^{(1)}(\rho)=J_0(\rho)+iN_0(\rho)$, in terms of Bessel ($J_0$) and Neumann ($N_0$) functions.

The amplitude has the following[14,31] common and convenient form:

$$F(\mathbf{k},\mathbf{k}') = \frac{2b}{b^2-a^2} \cos\left[(\mathbf{k}-\mathbf{k}') \cdot \frac{\mathbf{d}}{2}\right] - \frac{2a}{b^2-a^2} \cos\left[(\mathbf{k}+\mathbf{k}') \cdot \frac{\mathbf{d}}{2}\right]. \tag{16}$$

In the investigated two dimensional case one has $b=(i-\cot\,\eta)$ while in the $D=3$ case $b=k(i-\cot\,\eta)$. The asymptotic form of the scattered wave prescribes a

$=(i/2)H_0^{(1)}(kd)$ in two dimensions and not $a=e^{ikd}/d$ as in three dimensions. The illustrative representation in Eq. (16) shows that the "expansion parameter," which encodes deviations from the perturbative impulse approximation where $a\equiv 0$, is the important $|a|/|b|$ ratio. For fixed (and physical) d values and at vanishing ($k_F\to 0$) scattering momentum the Green's functions determine the characteristic behavior of the probability amplitude $|F(\mathbf{k},\mathbf{k}')|^2$. This fact results in marked deviations (see Fig. 2, below) of cross sections from the ones which include *solely* the role of wave-mechanical interference without coupling the centers.

The rest of the two dimensional calculation is tedious but straightforward, similarly to the one in three dimensions.[14] Instead of Eqs. (8) and (9), obtained without the modulating effect of multiple scattering, we derive

$$G_r^{\parallel}(2,x) = A(1+J_0)\left(\frac{1}{2}+J_0-\frac{J_1}{x}\right) + B(1-J_0)\left(\frac{1}{2}-J_0+\frac{J_1}{x}\right) - 2CJ_1^2, \tag{17}$$

$$G_r^{\perp}(2,x) = A(1+J_0)\left(\frac{1}{2}+\frac{J_1}{x}\right) + B(1-J_0)\left(\frac{1}{2}-\frac{J_1}{x}\right) \tag{18}$$

for the *new* ratio (r) functions $G_r^i(D,x)$. The argument of the Bessel functions is $x\equiv k_F d$. The averaged (av) energy loss per unit path length in $D=2$ has a quite simple form of

$$\left[\frac{1}{v}\frac{dE}{dt}\right]_{av} = n_0 k_F v[(4/k_F)\sin^2\eta][A(1+J_0)^2 + B(1-J_0)^2 - 2CJ_1^2]. \tag{19}$$

The expressions in Eqs. (17)–(19) result in the previous ones ($G_r^i\to g_r^i$) when $A=B=C=1$, i.e., when the so-called impulse approximation for electron-homonuclear-dimer scattering is applied. Deviations of $A(D,x,\eta)$, $B(D,x,\eta)$ and $C(D,x,\eta)$ factors from unity encode the modulating role of multiple scattering beyond the interference effect. The functions for these multiplying factors are (cf., Ref. 14, for the $D=3$ case) given by

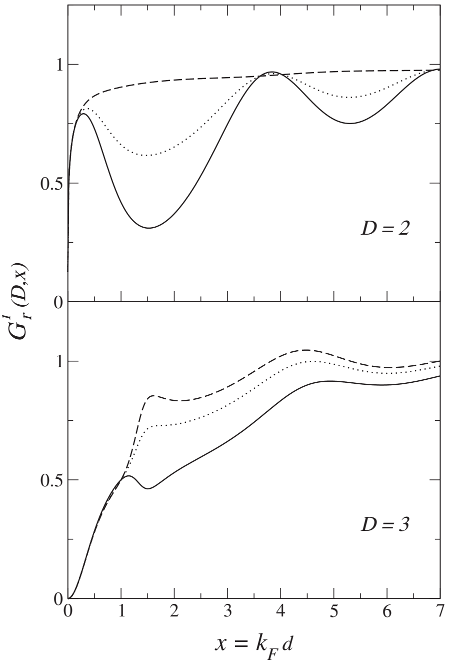

FIG. 2. Illustrative ratios, denoted as $G_r^i(D,x)$, showing the orientation dependence of renormalized stopping powers of D-dimensional degenerate electron gases for in-moving slow homonuclear dimers. The parallel (∥) (solid curves), perpendicular (⊥) (dashed curves), and random (dotted curves) cases refer to fixed angles (see the text) between dimer orientation and its velocity direction. The curves are exhibited for the $x\in[0,7]$ range, where $x=k_F d$. The upper panel is devoted to the two dimensional ($D=2$) case while the lower panel (taken from Ref. 14) to the three-dimensional ($D=3$) one. The curves are obtained beyond the perturbative (cf., Fig. 1) impulse approximation, by considering the modulating effect of multiple scattering with $\eta=\pi/2$.

$$A(2,x,\eta) = \frac{1}{1+\sin\eta[J_0(x)\sin\eta + N_0(x)\cos\eta] + \left(\frac{\sin\eta}{2}\right)^2[J_0^2(x)+N_0^2(x)]}, \tag{20}$$

$$B(2,x,\eta) = \frac{1}{1-\sin\eta[J_0(x)\sin\eta + N_0(x)\cos\eta] + \left(\frac{\sin\eta}{2}\right)^2[J_0^2(x)+N_0^2(x)]}, \tag{21}$$

$$C(2,x,\eta) = \frac{1-\left(\frac{\sin\eta}{2}\right)^2[J_0^2(x)+N_0^2(x)]}{\left[1-\left(\frac{\sin\eta}{2}\right)^2[J_0^2(x)+N_0^2(x)]\right]^2 + \sin^2\eta[J_0(x)\cos\eta - N_0(x)\sin\eta]^2}. \tag{22}$$

Now, we make an asymptotic evaluation, with $\eta(k_F) = \pm\pi/2$ for the phase shift,[17,21] of the ratio function $G_r^{\parallel}(D,x)$ of the parallel case. Leading-order expansions for high x values, i.e., $A(2,x)\simeq[1-J_0(x)]$, $B(2,x)\simeq[1+J_0(x)]$, and $C(2,x)\simeq 1$, result in

$$G_r^{\parallel}(2,x) \simeq [1 - J_0^2(x) - 2J_1^2(x)] \Rightarrow \left[1 - \frac{3}{\pi x}\left(1 - \frac{1}{3}\sin 2x\right)\right] \tag{23}$$

to a useful comparison with the perturbative expression

$$g_r^{\parallel}(2,x) \simeq [1 + 2J_0^2(x) - 2J_1^2(x)] \Rightarrow \left[1 + \frac{4}{\pi x}\sin 2x\right]. \tag{24}$$

Clearly, the role of multiple scattering can be very important, as the deviation between the above asymptotic forms heralds. $G_r^{\parallel}(2,x)$ tends to unity from *below*. Notice that a notable reduction (see, Fig. 2 for $D=3$) was vital to explain[14] measured data for stopping power of a carbon target for slow intact C_2 dimer in three dimension. These results of the quantum mechanical evaluations for complete alignment could fit to a classical picture where the second constituent of the dimer travels in an effective shadow of the first one.

Figure 2 is devoted, via the ratio functions $G_r^i(D,x)$ evaluated at $\eta=\pi/2$, to an illustration on the role of dimensionality. One can see that the $[G_r^{\parallel}(2,x)/G_r^{\perp}(2,x)]$ quantity (a measure of the ratio of residual resistivities, as well) is appreciably smaller than unity over a broad range of x: roughly in the $x\in[0.7,2.7]$ interval around a strong minimum. In harmony with our general analysis given above after Eq. (16), the effect of multiple scattering is a robust one in both dimensions at finite d values and very small k_F scattering momenta. In *this* mathematical limit for scattering momenta, and in three dimensions, we have a $\sigma_{tr}^i\propto d^2$ characteristic behavior for the integrated[14,26] cross sections due to $|1/a|^2\sim d^2$. In two dimensions the same (i.e., at finite d) limit for integrated scattering lengths is governed[20] by an $|1/a|^2 \sim[1/\ln(1/k_Fd)]^2$ dependence due to the dominance of the Neumann $[N_0(x)]$ function in the corresponding Green's function at small enough x values.

III. SUMMARY AND OUTLOOK

Motivated by the experimental relevance and importance of two dimensional electron gases in various condensed matter systems, and the possible role of embedded molecules or created dimers, the scattering aspects of independent electrons off such unavoidable or designed centers are investigated here. Special attention has been payed to the effects of interference and multiple scattering. The important influence of these combined effects is analyzed in a comparative manner. In particular, a remarkable reduction in friction was found for perfectly aligned dimers. The problem of pure dephasing of elastically scattered electrons is briefly discussed via the relevant total cross sections. The *detailed* results derived could be useful in various problems of important experimental relevance in the field of two dimensional degenerate electron gases.

The orientation-dependent $\sigma_{tr}^i(\alpha)$ transport cross sections might have applications in the theoretical understanding of stabilization of structures based on atom deposition[32] on close-packed noble metal surfaces under voltage-driven transport conditions, as well. As it is well known, the conductivity or the resistivity is determined by the $k_F\sigma_{tr}$ product. Recent experiments[33] with linear atomic chain on Ag(111) surface addressed this question of stability in the presence of an electric current. In this case one has a problem of electromigration in which wind forces are proportional[34] to the transport cross sections.

Different charge states of dimer constituents represent another realistic challenge, similarly to the atomic case[35,36] in three dimensions. In two dimensions this problem requires further careful considerations since a single not everywhere nonpositive or not everywhere nonnegative circular potential can easily bind electrons under very general constraining conditions according to an earlier analytical treatment of Simon[37] and its recent numerical *extension*.[38] Experiments[4,39] seem to support this fine result of mathematical physics. The preliminary short analysis given after Eq. (5) in the text was, in fact, motivated by this remarkable effect in two dimensions. Finally, we note that in the so-called two-impurity Kondo model[40] the input (bare) coupling parameters should depend heavily on the way of modeling the interimpurity range, beyond a simple plane-wave approximation.

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