Scattering of Slow Metastable Argon Atoms by Dielectric Nanospheres[†]

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The elastic scattering at low energy of metastable argon atoms with internal angular momentum $J = 0$ and 2 by dielectric nanospheres is investigated. The differential cross sections are calculated for both isotropic and anisotropic interactions. A polarization effect is clearly evidenced. The possible use of a metastable atom beam as a probe of an ensemble of nanospheres deposited on a passive substrate is examined.

1. Introduction

The interaction between an atom and a solid surface at mean distance ($d < 200$ nm), i.e., in absence of any retardation effect, is of the van der Waals (vdW) type. For atoms possessing an internal angular momentum **J**, as metastable argon atoms Ar*($^3P_2$), this interaction combines a scalar term and a quadrupolar term.[1] The former (dominant) term is responsible for elastic collisions with the surface, whereas various inelastic processes are induced by the latter one.[2] In the low atomic velocity domain considered here (a few tens of meters per second), the internal degrees of freedom of the solid are not involved and excitation processes such as surface phonons can be ignored. Similarly nonadiabatic atomic transitions can be neglected. Under such conditions, the expression of the interaction is readily obtained using the electrostatic image method: it is simply the interaction between the instantaneous atomic dipole **D** and its instantaneous image in the solid surface. When the solid surface is planar or more generally when its radius of curvature a is much larger than the range of the interaction (a few nanometers), one gets the well-known vdW interaction in $1/d^3$, for both scalar and quadrupolar terms. This is no longer the case when the solid is a sphere of a nanometric size. Such titanium oxide nanospheres, of radius $a \approx 2.5$ nm, deposited on a planar solid substrate, have been recently produced.[3] The elastic scattering of metastable atoms by such spheres, which mainly involves the scalar part of the vdW potential, is expected to provide us with information about the interaction itself, the size of the spheres, and the dispersion of their diameters as well as their 1D or 2D ordering on the substrate plane. Actually, the elastic problem is relatively simple only in the case of atoms having a zero angular momentum, such as Ar*($^3P_0$) (section 2). For atoms with $J \neq 0$, an anisotropic diagonal interaction must be considered. Under such conditions, the treatment of the collision becomes rather similar to that of atom—molecule collisions (section 3). The most intricate problem arises when inelastic processes are involved. Indeed it has been shown[4] that, in the presence of a static magnetic field **B**, the quadrupolar part of the vdW interaction is responsible for so-called van der Waals—Zeeman transitions, i.e., transitions from a given Zeeman sublevel M to another one M′. Theoretical treatments of such transitions have been already proposed in the case of a solid target of a large curvature radius (a few micrometers or more). They use either the sudden[5] or the Landau—Zener[6] approximation. In both approaches, the normal to the surface was considered as fixed for rectilinear atomic trajectories grazing the surface at small distances. This was justified because of the very short range of distances ($d < d_{max} = 2-3$ nm) within which inelastic processes are expected to occur. This is no longer the case for target spheres the radius of which is comparable to d_{max} and the whole treatment must be reconsidered.

Another important difficulty comes from the fact that the nanosphere is deposited on a planar solid. Even if this solid is assumed to be totally passive, the geometry of the scattering is fundamentally modified. Moreover, in the presence of several deposited spheres, the mutual distances of which are small, multiple scattering effects are likely to occur. A considerable extension should be given to the model to include these facts. The scope of the present paper will not be to treat these questions but rather to solve the simple scattering of a planar atomic wave by a single nanosphere in free space.

2. Elastic Scattering by an Isotropic Potential

For atoms having a zero internal angular momentum, such as metastable Ar*($^3P_0$) atoms, the vdW interaction with a dielectric nanosphere, the permittivity ε of which is much larger than 1, is readily derived from the electrostatic image method[7]

$$V_{vdW}(r) = -8C_3\left(\frac{a}{r^2 - a^2}\right)^3 \qquad (1)$$

where r is the distance to the center of the sphere and C_3 is the standard vdW constant obtained with a planar solid. Indeed, setting $r = a + d$, one recovers, for $a \gg d$, the well-known form $V \approx -C_3/d^3$. On the other hand, for $r \gg a$, one gets $V_{vdW}(r) \approx -8\ C_3\ a^3/r^6$, as expected for the interaction between metastable and ground-state atoms. Actually at short distances ($d \ll a$), atom and solid- electronic orbitals overlap, which results into a repulsive interaction in d^{-9}. At the same time, this overlap induces a strong quenching of metastable atoms. This quenching will be ignored since it has a negligible effect on the scattering, except at angles close to π (front collisions). Then the total potential takes the form

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$$V(r) = -8C_3\left(\frac{a}{r^2 - a^2}\right)^3 + C_9(r - a)^{-9} \qquad (2)$$

For metastable argon atoms, $C_3 = 3.456 \times 10^5$ atomic units (au), $C_9 = 4.345 \times 10^{10}$ au.[8] Calculations will be carried out for an initial atomic velocity $v_0 = 56$ m/s, i.e., a de Broglie wavelength $\lambda_0 = 3.4037$ au $= 0.18$ nm, and a sphere of radius $a = 48.26$ au $= 2.5$ nm. Such a low velocity is obtained using a Zeeman slower operating on Ar*($^3P_2$) atoms with a laser beam at $\lambda = 811.5$ nm,[9] these atoms being then transferred to the $^3P_0$ state by optical pumping. Assuming an incident atomic plane wave, the standard scattering amplitude at angle θ is given by[10]

$$f_0(\theta) = \frac{1}{2ik_0}\sum_{l=0}^{\infty}(2l + 1)[\exp(2i\delta_l) - 1]P_l(\cos\theta) \qquad (3)$$

where $k_0 = 2\pi/\lambda_0$ and P_l is the Legendre polynomial. Owing to the relative shortness of the wavelength, phase shifts, δ_l are calculated using the semiclassical expression

$$\delta_l = \int_{r_0(l)}^{\infty}\left(\left[k_0^2 - U(r) - \left(l + \frac{1}{2}\right)^2/r^2\right]^{1/2} - k_0\right)\mathrm{d}r + \frac{\pi}{2}\left(l + \frac{1}{2}\right) - k_0 r_0(l) \qquad (4)$$

where $U(r) = (2m/\hbar^2)\,V(r)$, m being the atom mass and $r_0(l)$ the distance of closest approach for angular momentum l. The resulting differential cross section $\sigma_0(\theta) = |f_0(\theta)|^2$ is shown in Figure 1. It corresponds to the scattering by an isolated sphere. In fact the sphere stands over a planar substrate and the incident atomic plane wave propagates in a direction making some angle θ_i with this plane. If the substrate is assumed to be passive, then its effect is simply to cancel the scattered intensity at some geometrically defined directions, e.g., in the incidence plane, the intensity cancels for $\theta < \theta_i$. This explains why a rainbow effect expected around $\theta = 1.6$ rad is not observed. In Figure 1, in order to get a preliminary account of the effect of the substrate on which the sphere is deposited, rapid oscillations and supernumerary rainbow have been artificially canceled by suppressing the interference between positive and negative angle contributions. Presumably for this reason, cross sections calculated with different sphere radii ranging from 2 to 3.3 nm show a weak dependence on a.

For N identical spheres randomly distributed on the plane, the scattered intensity is simply multiplied by N, provided that their mutual distances are large enough to make negligible multiscattering phenomena. On another hand, when $N \times N$ identical spheres are distributed over a regular 2D grating of period b along both axes η and ζ (the third ξ-axis being normal to the plane), the scattering amplitude is the single-sphere one multiplied by a "grating factor" G. Let (θ, φ) be the angular coordinates (around the ζ axis) of the final momentum direction, those of the initial one being $(\theta_i, 0)$, and $\phi_1 = k_0\, b \sin\theta \sin\varphi$, $\phi_2 = k_0\, b\,(\cos\theta - \cos\theta_i)$. One easily gets: $G = G_1G_2$ with $G_{1,2} = \exp[i\phi_{1,2}/2]\,\sin[(N + 1)\phi_{1,2}/2]/\sin(\phi_{1,2}/2)$. The visibility of the G factor readily gives b and an estimate of the ordering of the nanospheres over the substrate.

3. Elastic Scattering by an Anisotropic Potential

Coupled Equations in the General Case. Let us consider a plane wave, in $\exp(ik_0z)$, of metastable atoms (Ar* ($^3P_2$), polarized in Zeeman state $M_0 = +2$ with respect to some fixed axis (in fact a magnetic field **B**), incident on a nanosphere of radius a. In the magnetic field, at an infinite distance from the sphere, the internal atomic energies depend on M through the term $-g\mu_B BM$, where g is the Landé factor and μ_B the Bohr magneton. Consequently, only those collisions in which the final value M' of the magnetic number remains equal to M_0 are elastic. Inelastic collisions with $M' \neq M_0$ involve so-called "van der Waals−Zeeman" (vdW-Z) transitions.[4] In fact the treatment of the collision problem includes both elastic and inelastic channels. For the sake of simplicity we shall assume that (i) the incident z axis is collinear with **B**, (ii) the sphere material is either a perfect metal, or a dielectric with a permittivity $\varepsilon \gg 1$. Point (i) could be rather simply generalized: it is a matter of decomposition of the plane wave. To extend point (ii) is a more intricate matter, but it is not absolutely necessary in our case because of the rather high permittivity of the material used here (for titanium oxide, $\varepsilon \approx 7$).

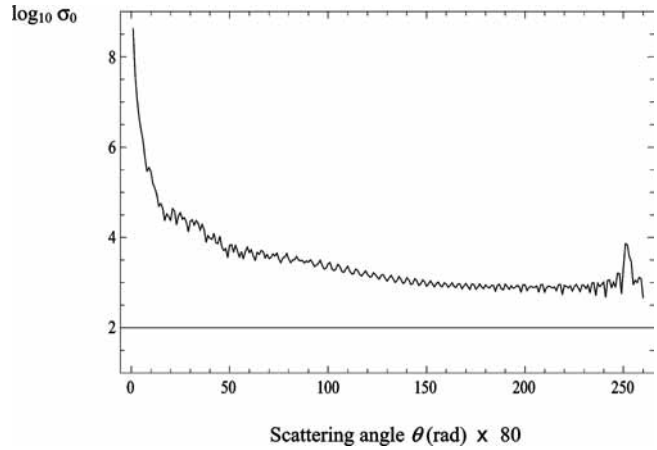

Figure 1. Differential elastic cross section σ_0 (in au, log scale) for the isotropic potential (see text), as a function of the scattering angle θ. The index number in abscissa corresponds to 80 times θ (in rad) (index number 251 corresponds to $\theta = \pi$).

The atomic motion being described by a wave function expanded over Zeeman states $|M\rangle$, namely $|\Psi\rangle = \sum_{M'}\Phi_{M'}(\mathbf{r})\,|M'\rangle$, the stationary Schrödinger equation for total energy E is

$$\left(-\frac{\hbar^2}{2m}\Delta_r + \underline{\mathbf{V}}\right)|\Psi\rangle = \mathrm{E}|\Psi\rangle \qquad (5)$$

where $\underline{\mathbf{V}}$ is the interaction operator. According to the electrostatic image method applied to a perfect metallic sphere, the nonretarded van der Waals interaction takes the form[5]

$$\underline{\mathbf{V}} = -f(r)\left[C_3 + \frac{\eta}{16}(J_r^2 - J^2/3)\right] + g\mu_B\mathbf{B}\cdot\mathbf{J} \qquad (6)$$

where, as before

$$f(r) = 8a^3(r^2 - a^2)^{-3} \qquad (7)$$

The novelty is the presence of a quadrupolar term, proportional to the constant η, in the vdW interaction. For Ar*(3P2) atoms this constant is $\eta \approx \sim C_3/10$. Here J_r is the radial component of $\mathbf{J}$, $\mathbf{J}^2/3 = 2$ and $\mathbf{B}$. $\mathbf{J} = BJ_z$. The matrix elements of $\underline{\mathbf{V}}$ in the $|M\rangle$ basis set (referred to the z axis) are

$$\langle M|\underline{\mathbf{V}}|M'\rangle = [g\mu_B BM - f(r)(C_3 - \eta/8)]\delta_{MM'} - f(r)(\eta/16)\langle M|J_r^2|M'\rangle \tag{8}$$

The matrix elements of J_r^2 are given in ref 5 as functions of the angle α between the direction of **B** and that of the vector position **r** of the point under consideration. Note that **r** plays the role of the normal to the surface of the sphere. Here it can no longer be considered as a fixed direction. Hence α becomes a *variable*—an ordinary spatial coordinate—of the problem. Consequently the matrix elements of $\underline{\mathbf{V}}$, even the diagonal ones, behave as *anisotropic* potentials. The matrix elements of J_r^2 can be written (M, M' being ordered as −2, −1, 0, +1, +2)

$$\begin{array}{ccccc}
2 + 2P_2 & -3\sin\alpha\cos\alpha & \sqrt{\frac{2}{3}(1 - P_2)} & 0 & 0 \\
-3\sin\alpha\cos\alpha & 2 - P_2 & -\sqrt{\frac{3}{2}\sin\alpha\cos\alpha} & 1 - P_2 & 0 \\
\sqrt{\frac{2}{3}(1 - P_2)} & -\sqrt{\frac{3}{2}\sin\alpha\cos\alpha} & 2 - 2P_2 & \sqrt{\frac{3}{2}\sin\alpha\cos\alpha} & \sqrt{\frac{2}{3}(1 - P_2)} \\
0 & 1 - P_2 & \sqrt{\frac{3}{2}\sin\alpha\cos\alpha} & 2 - P_2 & -3\sin\alpha\cos\alpha \\
0 & 0 & \sqrt{\frac{2}{3}(1 - P_2)} & 3\sin\alpha\cos\alpha & 2 + 2P_2
\end{array} \tag{9}$$

where P_2 is the Legendre polynomial $P_2(\cos\alpha)$. It should be noted that remaining nonzero terms in $\sin\alpha\cos\alpha$ can be expressed as well with Legendre polynomials, using

$$P_2^{\pm 1}(\cos\alpha) = \sqrt{\frac{15}{4\pi}}\sin\alpha\cos\alpha$$

With (8), eq 5 can be written

$$\left[-\frac{h^2}{2m}\Delta_r - f(r)(C_3 - \eta/8 + (\eta/16)\langle M|J_r^2|M\rangle)\right]\Phi_M - f(r)(\eta/16)\sum_{M'\neq M}\langle M|J_r^2|M'\rangle\Phi_{M'} = (E - g\mu_B BM)\Phi_M \tag{10}$$

In the following, all incident atoms will be assumed to be initially polarized in Zeeman state $|M_0 = +2\rangle$. In this special case, eq 9 becomes[1]

$$\left[-\frac{h^2}{2m}\Delta_r - f(r)(C_3 + (\eta/8)P_2(\cos\alpha))\right]\Phi_2 - f(r)(\eta/16)\sum_{M'\neq M}\langle 2|J_r^2|M'\rangle\Phi_{M'} = (E - 2g\mu_B B)\Phi_2 \tag{11}$$

(This corresponds to the incident wave. One has to be aware that, in the presence of couplings with some other states M', one will have also to consider a "master equation" for $\Phi_{M'}$ similar to (10), but containing a diagonal potential term proportional to $\langle M'|J_r^2|M'\rangle$ and a total energy $E - g\mu_B BM'$.) Note that the maximum magnitude of the anisotropic part of the diagonal potential, $(\eta/8)f(r)$, is relatively small compared to the isotropic one.

Radial Equations in the *Elastic* Case.

General Expression. When all couplings to sublevels $M' \neq +2$ are ignored, the collision is purely elastic. Nevertheless it is governed by a diagonal *anisotropic* potential containing a term in $P_2(\cos\alpha)$, able to induce couplings among the partial waves associated with various values of the angular momentum l. This point is easily understood when considering an atom with its spin along a fixed direction (bold arrows in Figure 2), passing in the vicinity of a spherical target: the interaction is expected to depend not solely on the distance r but also on the angle α between **r** and the z axis. When the interaction is able to move the spin out of the z direction (dotted arrows in Figure 2), this means that $M-M'$ transitions (vdW-Z inelastic transitions) occur and that the collision is partly inelastic.[11]

Having in view a separation of variables r and α (the azimuthal angle around z is not involved), one expands $\Phi_2(\mathbf{r})$ over Legendre polynomials $P_l(\cos\alpha)$

$$\Phi_2(r) = \sum_l r^{-1}\varphi_l(r)P_l(\cos\alpha)$$

where radial wave functions are $r^{-1}\varphi_l(r)$. In the purely elastic case, eq 11 gives

$$\varphi_l'' + [k_0^2 - l(l+1)/r^2 - U_0(r)]\varphi_l - 2U_2(r)\sum_{l'} c[l,l',2;0,0,0]^2\varphi_{l'}' = 0 \quad (12a)$$

where $k_0^2 = 2mE/\hbar^2$ (squared incident wavenumber), $U_0 = (2m/\hbar^2)[-f(r)\ C_3]$ (isotropic part of the potential), $U_2(r) = -(2m/\hbar^2)(\eta/8)f(r)$ (anisotropic part of the potential) and $c[l,l',2;\ 0,0,0]^2$ is a squared 3*j*-coefficient. This coefficient differs from zero only when $l' = l$ or $l \pm 2$ with

$$c[l,l,2;0,0,0]^2 = l(l+1)/[(2l-1)(2l+1)(2l+3)] = b(l)$$

$$c[l,l,\pm 2,2;0,0,0]^2 = 9(l+1)(l+2)/[(2l+1)(2l+3)(2l+5)] = 9b(l+1)$$

This (slightly) simplifies the expression of diagonal and off-diagonal parts in (12a) into

$$\varphi_l'' + [k_0^2 - l(l+1)/r^2 - U_0(r) - 2b(l)U_2(r)]\varphi_l - 18U_2(r)b(l+1)(\varphi_{l-2} + \varphi_{l+2}) = 0 \quad (12b)$$

From (9) it is seen that for $l \geq 2$, φ_l is coupled to $\varphi_{l-2} + \varphi_{l+2}$. Introducing a column matrix $\underline{\mathbf{\Phi}}$ the elements of which are the φ_l-s, eq 11 can be written in a more compact form

$$\overline{\Phi''} + [k_0^2\underline{\mathbf{1}} - \overline{\mathbf{U}}_e(r)]\underline{\Phi} = 0 \quad (13)$$

where $\underline{\mathbf{1}}$ is the unitary $L \times L$ matrix (L being some arbitrary high upper value of l) and $\underline{\mathbf{U}}_e(r)$ the effective potential matrix. The main remaining problem is that $\underline{\mathbf{U}}_e(r)$ contains off-diagonal elements which mutually couple the radial equations.

Elimination of Static Couplings. Fixing the value of L (it turns out that $L \sim 100$ is sufficient), one can diagonalize $\underline{\mathbf{U}}_e(r)$ into $\underline{\mathbf{E}}(r) = \underline{\mathbf{C}}^{-1}(r)\underline{\mathbf{U}}_e(r)\underline{\mathbf{C}}(r)$, where $\underline{\mathbf{C}}$ is an appropriate orthogonal $L \times L$ matrix. This operation gives at the same time eigen-values $e_l(r)$ and eigen-vectors. This is equivalent to change the radial functions φ_l into

$$\Psi_l = \sum_{l'} d_{ll'}(r)\varphi_{l'}$$

or inversely

$$\varphi_l = \sum_{l'} c_{ll'}(r)\Psi_{l'}$$

Note that both $d_{ll'}(r)$ and $c_{ll'}(r)$ coefficients tend to $\delta_{ll'}$ when $r \to \infty$. Owing to this diagonalization, couplings due to potential terms cancel. Unfortunately, as usual, because of the r dependence of coefficients $c_{ll'}$, dynamical couplings of the type $\langle\psi_l|(c_{ll'})'^{(\prime\prime)}|\psi_{l'}\rangle$, where $'(\prime\prime)$ indicate r derivatives, appear. Coefficients c_{ll}' depend on r *grosso modo* as the potentials do. In the JWKB approximation where the potentials are assumed to vary slowly at the wavelength scale ($k_0|c'|$ and $k_0^2|c''| \ll 1$), the latter couplings can be ignored, which leads to simplified "ordinary" radial equations

$$\psi_l'' + [k_0^2 - e_l(r)]\psi_l = 0 \quad (14)$$

which can be solved as before (section 2, eqs 3 and 4). In the present case, it turns out that, for $r < 80$ au, the e_l values differ from the diagonal terms of $\underline{\mathbf{U}}_e(r)$ only for values of l smaller than 70. At larger values of r, the e_l values are almost identical to the diagonal terms. Finally the scattering amplitude $f_1(\theta)$ and the related differential cross section $\sigma_1(\theta)$ are derived. This differential cross section is shown in Figure 3 as a function of the scattering angle. It is seen that it significantly differs (by a factor of about 30) from that related to the isotropic potential, σ_0, essentially at angles $\theta > 1.5$ rad (Figure 4). As above (section 2), interference terms between positive and negative angle contributions have been artificially canceled. Correlatively a rather weak dependence of both σ_1 and the ratio σ_0/σ_1 on the sphere radius is obtained.

4. Conclusion

A beam of moderately slow metastable atoms (velocity of a few tens of meters per second) appears to be an efficient probe usable in the characterization of nanospheres deposited on a passive planar substrate. From the angular width of diffraction patterns, size and size dispersion can be derived. Similarly, it may be expected that constants (C_3, C_9) of the interaction can be determined from the differential cross sections. The ordering of nanospheres, when they are more-or-less well organized over a 2D grating, can be derived from the contrast of the corresponding "grating factor", at least if their mutual distances is

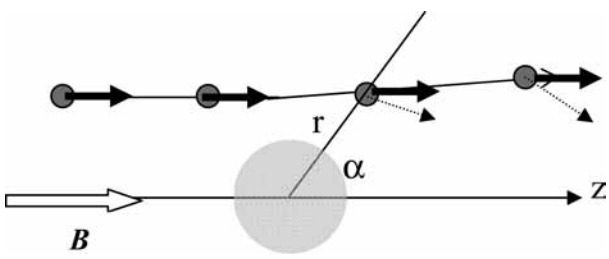

Figure 2. Scheme of a classical collision of atoms polarized along the incident axis (z or **B**) with a nanosphere. The atomic spin is represented by a black arrow (elastic collision) or by a dotted arrow (transitions among magnetic Zeeman states M).

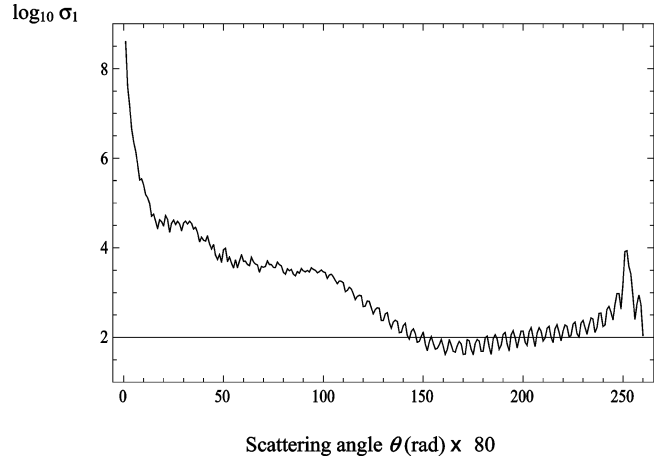

Figure 3. Differential elastic cross section σ_1 (in au, log scale) for the anisotropic potential (see text), as a function of the scattering angle θ (same scale as in Figure 1). The effect of anisotropy is seen at angles larger than 1.5 rad.

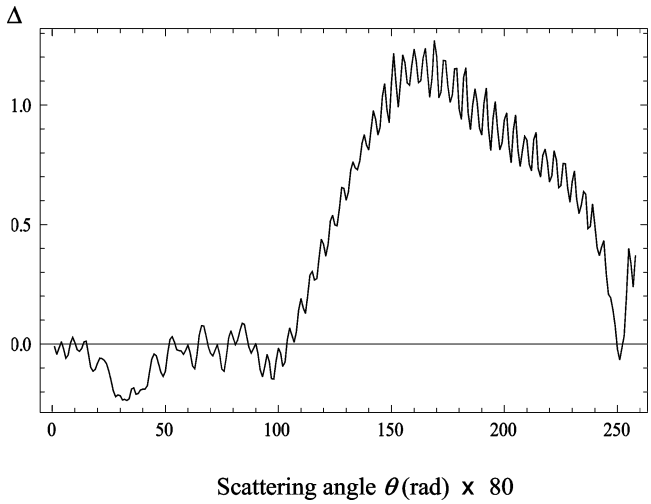

Figure 4. Ratio $\Delta = \sigma_0 / \sigma_1$ in log-scale, exhibiting the anisotropy effect (the θ scale is the same as in Figures 1 and 3).

large enough to avoid any multiscattering phenomena, these latter phenomena being observable by comparing more or less compact deposits. Other possible targets are helium nanodroplets—eventually seeded with extra molecules—developed by Toennies and co-workers.[12] Polarization effects due to the anisotropic part of the interaction appear to be observable despite their smallness. The presence of a static magnetic field (typically a few tens of gauss), needed as a reference axis, is expected to induce cocalled van der Waals−Zeeman transitions among magnetic sublevels with a significant probability. Such transitions, which have not been investigated in the present paper, are nevertheless worth analyzing insofar as (i) they appear to be an extension of previously observed effects[4] to the case of surfaces of very small curvature radii, (ii) they are the analogue of polarization or spin effects occurring in thermal collisions between metastable atoms with spin (namely, $Ne^*(^3P_2)$ atoms) and ground-state atoms.[13] To treat this problem, it will be needed to solve the full system of coupled equations given by (11). It should be also noted that polarization effects, even in the sole case of elastic scattering, should provide us with some information about the shape of imperfect "nanospheres".

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