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Metastable rare gas atoms scattered by nano- and micro-slit transmission gratings

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Abstract. The transmission of metastable argon atoms through nano-slit or micro-slit gratings is studied by use of time of flight and angular analysis. This transmission departs from the simple geometric one essentially by two ways: (i) the elastic or diagonal part of the van der Waals (vW) interaction with the solid causes an angular narrowing of the emerging beam; (ii) the off-diagonal vW interaction induces the exothermal fine structure transition $^3\mathrm{P}_0 \rightarrow {}^3\mathrm{P}_2$ ($\Delta E = 175$ meV) leading to large scattering angles; the resulting angular distribution is very sensitive to the roughness of the surface in the direction of the depth. An extension of these experiments to transversally coherent beams is proposed. It should be considered as a first step towards a new type of interferometer in which the inelastic diffraction makes the gratings work as beam splitters or mirrors.

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The fast development of modern technology has given access to rapidly emerging fields, like scanning probe microscopies or nanophysics [1]. This has paved the way to new advances in single atom manipulation, atom interferometry with material gratings or atom lithography [2]. The interaction of atomic systems with (dielectric or metallic) nanostructures, along with the strong modifications of the (free-space) atomic properties, are a key feature in these fields [3]. Atom wave deflection and diffraction by nanoscopic objects are governed by both the atom-surface long-range forces, and the coherent character of atom diffraction for sufficiently large de Broglie wavelengths. For instance, particle interferometry and diffraction with transmission nanogratings has been demonstrated for atoms, molecules and clusters [4,5]. For ground-state rare gas atoms, the important role of atom-surface van der Waals (vW) attraction has been recently studied [6]. In this letter, we analyse *metastable* rare gas atoms transmission through dielectric nanogratings and metallic microgratings, and show the central role of vW surface interactions, which are responsible not only for strong modifications of the angular transmission of the metastable beam, due to a large atomic polarisability, but also for atom symmetry breaking and metastable level coupling observed by means of an angular and time-of-flight resolved detection (as previously demonstrated in single slit transmission [7]). Owing to the collision kinematics, this exothermal process is a sensitive probe of the surface roughness along the depth of the slits. Also observed at large angle with the nanograting are inelastic processes presumably related to dielectric surface excitation modes.

Metastable rare gas atoms (Ne*, Ar*, Kr*) are produced by electron bombardment of an effusive beam of ground state atoms. Metastable atoms are then velocity selected ($\delta v/v = 10\%$) by means of a double chopping technique using a pulsed source and a synchronised slotted disk. The distance from the source to the grating is 121 mm. The angular aperture of the incident beam is 1.4°, a value much too large to give any significant coherence in a direction perpendicular to the slits. As a consequence, in all experiments described further, no coherent atomic diffraction effect is observed: the intensity scattered by N slits simply behaves as N times that of a single one. Obviously the main interest of using gratings instead of a single slit is a strong enhancement of the signal (by a factor ranging from a few 10^3 up to a few 10^4). Atoms scattered by the grating are detected by secondary electron emission of a metallic plate followed by a channel electron multiplier. This detector can be rotated around the vertical grating axis, within the range $-50° \leq \theta \leq +73°$.

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The distance from the grating to the detecting plate is 164 mm and the angular resolution is about 0.4°. Various elastic and inelastic processes induced by the interaction with surfaces (the slit walls) are identified by their times of flight. Two types of gratings have been used: a silicon nitride *nano-slit* grating (slit width 50 nm, period 100 nm, thickness 50 nm) with which most of the experiments have been carried out, and a copper *micro-slit* grating (slit width 20 μm, period 45 μm, thickness 20 μm). The former grating being made of a dielectric material it is expected that the impact of metastable atoms accumulates positive charges on it. The electrostatic potential on the grating increases up to a value such that an electron ejected by the atom impact can no longer escape, *i.e.* a few volts. As the positive charge density may be expected to be roughly uniform on the surface, the resulting electrostatic field has a negligible effect on the atom-surface interaction.

The strongest and easiest to observe effect of the nano-slit grating is *a narrowing of the angular aperture* of the emerging beam. This is due to the attractive diagonal (elastic) part of the vW interaction which deviates the atom trajectories, making some of them hit the walls, in which case the metastable atom is quenched, *i.e.* excluded from our observation. The effective width of the slits is then smaller than the geometrical one. Similar effects have been already observed: (i) the transmission of Rydberg atoms through a micro-slit is affected by the van der Waals forces [8]; (ii) in the diffraction of ground state atoms by a grating it is responsible for anomalies in the intensities of odd and even diffraction orders [6]. As expected, in the present experiment, the narrowing is more marked than in the latter one because of higher polarisabilities, *i.e.* higher vW constants. The effect can be roughly and phenomenologically described by assuming that the emerging trajectories make with the incident beam axis an angle smaller than or equal to some characteristic angle α. Indeed one finds for Ar*, Ne* and Kr*, $\alpha = 0.48°, 0.31°, 0.28°$, the polarisabilities being respectively (in atomic unit (au)): 47.8, 186, 341 [9]. A more rigorous description implies the calculation of the elastic differential cross-sections (DCS). A semi-classical calculation has been carried out for Ar* atoms (velocity 500 m/s), with an adjusted vW constant of 4 au. As it is seen in Figure 1 the calculated DCS, once convoluted by the angular resolution, agrees rather well with the observed angular profile, except at very small angles ($\theta < 1°$) where semi-classical treatments are known to fail. The asymmetry observed in the experimental data around ±5° is probably due to a misalignment of the normal to the grating with respect to the incident axis.

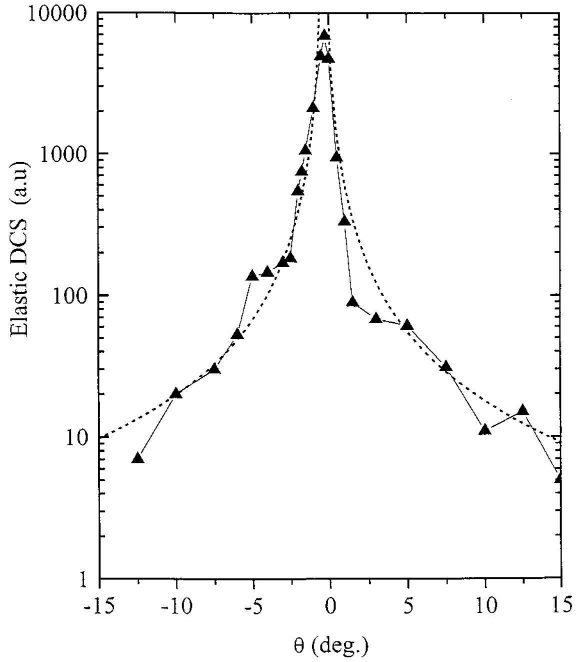

Fig. 1. Elastic differential cross-section (DCS) of metastable argon atoms (velocity $v_0 = 500$ m/s) by a nano-slit grating. Up triangles and full line: experiment (in arbitrary unit); dotted line: semi-classical calculation using the vW potential (in atomic units) $-4/z^3$. The calculated DCS is in a_0^2/rad.

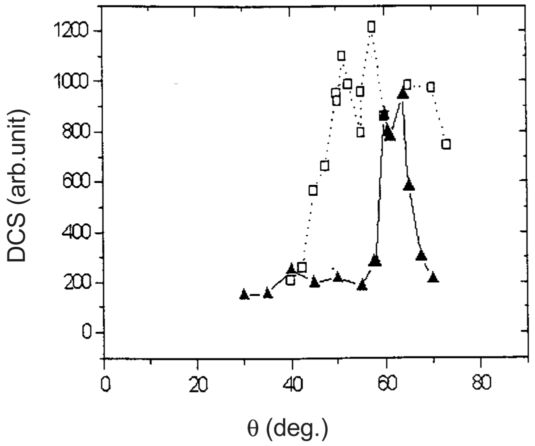

Fig. 2. Open squares, broken line: inelastic differential cross-section for the process $^3P_0$–$^3P_2$ induced by a silicon nitride nano-slit grating, at $v_0 = 500$ m/s. The predicted scattering angle is $\theta_f = 61°$; full triangles, full line: same measurement, with a micro-slit copper grating.

Because of the kinematics, exothermal processes with an energy gain ΔE comparable to the initial kinetic energy E_0 appear at large angles. For a grazing incidence at velocity v_0 on an ideal planar surface the final velocity for such a process is given by: $v_f = [v_0^2 + \Delta E/(2m)]^{1/2}$ where m is the atom mass. As the interaction depends only on the distance z to the surface, the component of the momentum parallel to the surface is conserved which results into a scattering angle $\theta_f = \cos^{-1}(v_0/v_f)$. Such an exothermal process has been clearly identified [10]: its energy gain, $\Delta E = 175$ meV for argon atoms, corresponds to the surface induced transition between metastable levels $^3P_0 \rightarrow {}^3P_2$, already observed with a single copper slit [7]. The DCS at a velocity of 500 m/s (see Fig. 2, broken line) has a main maximum at an angle close to the expected one: $\theta_f = 61°$. Nevertheless it is seen that this DCS has a rather large angular width, about 26° FWHM, *i.e.* even larger than that given by a single slit (22°), within which

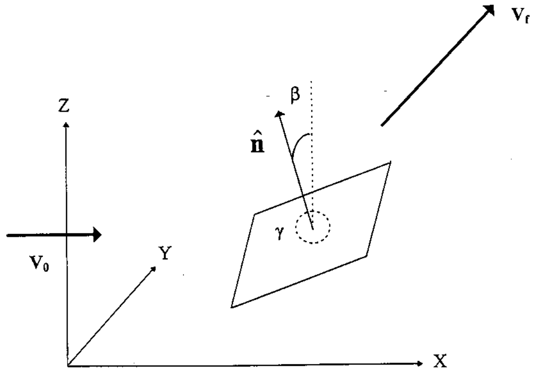

Fig. 3. 2D-model for a rough surface. The polar angles of the local normal $\hat{\mathbf{n}}$ to the surface are β and γ. Only the deflection θ in plane (x, z) is measured.

it exhibits some structures. In order to show that this spreading is due to the state of the surface, the experiment has been repeated using the micro-slit copper grating in place of the nano-slit grating. As it is seen in Figure 2, full line, the DCS is much narrower than the previous one (about 5° FWHM). At first sight this result might seem surprising since the amplitude of the surface defects (given by the manufacturers) is much smaller ($\sim$1–2 nm) in the former grating than it is in the latter one ($\sim$1 μm). However, it is easily understood in a 1D model where the deviation ζ of the surface with respect to an ideal plane is a function of a single coordinate (x) and neglecting any curvature effect in the vW interaction, that since θ_{f} has a well defined value, the dispersion of the slopes is directly responsible for the angular dispersion: an atom experiencing the transition in the vicinity of a given point on the surface where the derivative $\zeta' = \mathrm{d}\zeta/\mathrm{d}x$ is positive (otherwise it cannot approach the surface) is emitted at an angle: $\theta \approx \theta_{\mathrm{f}} + \zeta' + \zeta'^2/(4\sin^2\theta_{\mathrm{f}})$: this direction may be seen as a needle indicating the changes in the normal to the surface from point to point. It may be noticed that this exploration of the roughness is made in the direction of the depth (thickness) of the slits, a direction almost inaccessible to near-field microscopes. If the roughness is described as a statistical ensemble of sine profiles $\{a\cos\Omega x\}$, then the angular dispersion is directly related to the distribution $\rho(a\Omega)$ of the random variable $a\Omega$. This explains why a very small *mean value of a* (1 nm) gives rise to a larger effect than a much larger one (1 μm): it is just a matter of spatial frequency spectrum, as it has been already noticed in cold atom reflection by an evanescent wave mirror [11]. Thus the state of the crystalline silicon-nitride surface seems to be a lot more rough than that of copper surfaces. However this model is clearly too crude since it is unable to explain the dispersion of the scattering angles at values lower than θ_{f}. Actually the deviation ζ depends on the 2 coordinates $(x,\ y)$ in the reference plane of the surface (see Fig. 3). As the detector, preceded by a 5 mm-high vertical slit (parallel to those of the grating, *i.e.* to y-axis), is rotated in the horizontal plane, one observes the deviation θ in this plane (x, z). Let β, γ be the polar angles (around the z-axis) of the normal to the surface.

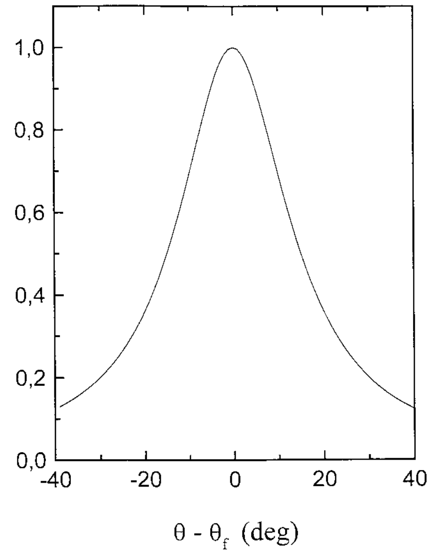

Fig. 4. Calculated angular distribution due to the 2D roughness of the surface. The azimuthal angle of the normal to the surface is assumed uniformly distributed. The distribution of the polar angle β has the form: *const* $(\beta/c)^2/[1 + (\beta/c)^2]^2$ with $c = 15°$.

The scattering angle is given by:

$$\tan\theta = \frac{A\cos\beta}{1 + A\sin\beta\cos\gamma}$$

where: $A = \sin\beta\cos\gamma + (\tan^2\theta_{\mathrm{f}} + \sin^2\beta\cos^2\gamma)^{1/2}$.

Assuming that the azimuthal angle γ is uniformly distributed, the resulting angular distribution is related to the distribution $\rho(\beta)$ of β. Figure 4 shows as an example the angular distribution centered at θ_{f} obtained with: $\rho(\beta) =$ *const* $(\beta/c)^2/[1 + (\beta/c)^2]^2$, where $c = 15°$. This value allows us to approximately reproduce the shape of the DCS given by the nano-slit grating (except for the structures in it). A similar but 5 times narrower distribution gives a DCS close to that of the micro-slit grating. Work is in progress to solve the "inverse problem", *i.e.* to extract $\rho(\beta)$ from the angular distribution, a difficult problem as neither β nor $(\theta - \theta_{\mathrm{f}})$ are in general small angles. This should be used for instance in the interpretation of the structures seen in the DCS of the nano-slit grating.

As we already mention, in the present experiment no coherence effect such as the atomic diffraction by the grating is observed. The diffraction of metastable helium atoms in the elastic channel has been recently observed [12]. An interesting question is the following: assuming a sufficiently well collimated incident beam, how will behave the diffraction in the *inelastic* exothermal channel? To get a diffraction peak at an angle θ implies that inelastic amplitudes produced at all points on the surfaces are in phase. Assuming perfect planar walls, one readily obtains 2 conditions. The first one involves 2 points separated by one period $(\Lambda = 2\pi/K)$ of the grating:

$$k_0\sin\theta_0 + k_{\mathrm{f}}\sin\theta(N) = N\,K$$

where k_0, k_f are the incident and final wave numbers, θ_0 is the (small) incident angle, N is an integer. The second condition is of the Bragg type. It involves 2 points on the same wall:

$$(\mathbf{k}_\mathrm{f} - \mathbf{k}_0) \times \hat{\mathbf{n}} = 0 \quad \text{or} \quad k_0 \cos\theta_0(N) = k_\mathrm{f} \cos\theta(N)$$

where $\mathbf{k}_{0,\mathrm{f}}$ are wave vectors and $\hat{\mathbf{n}}$ is the normal to the surface. It may be noticed that now θ_0 (noted $\theta_0(N)$) is also well defined at a given diffraction order. Actually the latter condition is automatically fulfilled since it expresses the conservation of the momentum parallel to the surface. For a grazing incidence the Bragg condition implies that $\theta(N)$ takes a stationary value (the detector is fixed), $\theta(N) \approx \cos^{-1}(k_0/k_\mathrm{f}) = \theta_\mathrm{f}$. Then the diffraction peaks are observable by varying θ_0 (rotation of the grating), provided that the angular spread of the incident beam is smaller than the separation between subsequent diffraction orders: $\delta\theta \approx K/k_0 = \lambda_0/L = 2 \times 10^{-4}$ rad in the present case. This condition is automatically fulfilled in as much as it coincides with that required to get the atomic diffraction. As each slit has two parallel edges, inelastically diffracted atoms appear (at any order) at angles $\pm\theta_\mathrm{f}$. Therefore the grating plays the role of a beam splitter providing a particularly large angular separation (about 120° in our case). It can also act as an "inelastic mirror" since atoms in state $^3\mathrm{P}_{2,M=0}$, the reference axis being the normal to the surface (*i.e.* the final state of the exothermal transition), impinging the grating at velocity v_f and angle θ_f, will partly emerge in state $^3\mathrm{P}_0$, perpendicularly to the grating with velocity v_0. Then using 2 gratings as beam splitters and in between them 2 others as mirrors (*cf.* Ref. [4] for an elastic version) one builds a Mach Zehnder interferometer including a large area (a few mm^2) between its 2 arms. This should provide us with a very high sensitivity ($\sim 10^{-14}$ rad/s) in angular velocity measurements.

The major obstacle in the realization of such a device is obviously the roughness of the surfaces. Here it is the *amplitude* a of the deviation that must be small compared to the final wave length λ_f. This condition is far from being realized under the present experimental conditions since $a \approx 1$ nm whereas $\lambda_\mathrm{f} \approx 10^{-2}$ nm. This very serious difficulty could be overcome by using falling cold atoms ($v_0 = 0.5$ m/s or less, $\lambda_0 = 20$ nm or more) together with an exothermal transition the energy gain of which is comparable to E_0 (54 neV or less) in order to get a sufficiently large angle θ_f. Such a process could be a transition of initially polarized atoms between two Zeeman states splitted by an adjustable magnetic field. For instance for $\Delta E = E_0$ ($\theta_\mathrm{f} = 45°$) one gets $\lambda_\mathrm{f} = 14$ nm (or more).

In conclusion we have shown that both diagonal and off-diagonal terms of the vW interaction induce dramatic effects on the transmission of metastable argon atoms through nano-slit gratings. The exothermal fine structure transition $^3\mathrm{P}_0 - {}^3\mathrm{P}_2$ leads to an angular distribution of the scattered atoms which is strongly altered by the roughness of the surfaces making the device a sensitive probe of this roughness in a direction perpendicular to the grating plane which is difficult to access by other means. The future extension of these experiments to transversally coherent beams is very promising and this work should be considered as a first step towards a new metastable atom interferometry and its important applications in gyrometry and atom lithography. In spite of the difficulty due to the surface roughness, the use of inelastic diffraction has the advantage to give an (adjustable) separation angle at the beam splitters much larger than that obtained by elastic diffraction. Therefore a compact device (*e.g.* 1 cm in length) would be able to give a wide separation of the two arms (allowing for instance a relatively easy manipulation of the atomic state in one arm not in the other one) and correlatively a large area in between them, *i.e.* a high sensitivity to rotation.

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