Eur. Phys. J. D **47**, 427–431 (2008)
DOI: 10.1140/epjd/e2008-00051-1

THE EUROPEAN
PHYSICAL JOURNAL D

Schlieren imaging of nano-scale atom-surface inelastic transition using a Fresnel biprism atom interferometer

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Received 1st November 2007
Published online 19 March 2008 –

Abstract. Surface-induced exo-energetic inelastic transitions among atomic Zeeman states in a magnetic field ("van der Waals – Zeeman" transitions) are useable as tuneable beam splitters. A transversally coherent atom beam impinging a pair of opposite surfaces (e.g. 2 edges of a slit or of an ensemble of periodic slits) gives rise to two coherently diffracted wave packets. Within the wave packet overlap, non-localised interference fringes of the Young-slit type are predicted. From the diffraction pattern observed in the Fraunhofer regime (Schlieren image), detailed information about the transition amplitude on a scale of a few nanometers should be derived.

PACS. 03.75.Be Atom and neutron optics – 37.25.+k Atom interferometry techniques – 34.-35.+a Interactions and atom-molecule potentials and forces

1 Introduction

The interaction between a solid and an atom located at a mean distance d from the solid surface ($0.5 \leq d \leq 100$ nm) is of the van der Waals (vdW) type (for reviews, see e.g. [1,2] and references therein). In general, for atoms having an internal angular momentum $\boldsymbol{J}$, this interaction is not spherically symmetric because, in addition to the well known scalar vdW potential $V_s = -C_3/d^3$, there is a quadrupolar contribution $V_q = -\eta(J_z^2 - \boldsymbol{J}^2)/(16d^3)$, where η is a constant, z being the normal to the surface. This part of the interaction breaks the atomic state symmetry and is able to induce, for instance, fine structure transitions in metastable argon and krypton atoms (Ar*, Kr*, $^3P_0 \rightarrow ^3P_2$) [3]. In the presence of an external magnetic field, this interaction induces transitions among atomic Zeeman states ($M \rightarrow M' = M + \Delta M$), as it has been shown for metastable Ne*($^3P_2$) atoms [4]. These transitions have been called "van der Waals – Zeeman (vdW-Z) transitions". In the case of an exo-energetic transition ($\Delta M < 0$), energy and linear momentum conservation rules imply that the atom trajectory is deflected outwards by an angle $\gamma \approx (\Delta E/E_0)^{1/2}$, where E_0 is the initial kinetic energy and $\Delta E = g\mu_B B|\Delta M|$ is the Zeeman energy splitting in magnetic field B, g being the Landé factor and μ_B the Bohr magneton. For B of a few hundred Gauss and $E_0 = 65$ meV, this angle γ is of a few mrad. Calculations, based upon either the sudden approximation [4] or the Landau-Zener formula [5], show that, at thermal energy E_0, the complex transition amplitude $A(\rho)$ rapidly oscillates in the vicinity of the surface as a function of the impact parameter ρ (i.e. the distance of closest approach) and then falls down to zero for $\rho > 2$ to 3 nm. These calculations also show that the transition probability $P(\rho) = |A(\rho)|^2$ increases at decreasing collision energies. Owing to the fact that the effective width ρ_{ef} of the transition zone (a few nm) is much smaller than the transverse coherence radius of the beams commonly used in such experiments (hundreds of nm), the inelastic process is fully coherent in the sense that it produces a linear superposition of wave packets in the various outgoing channels ($\Delta M = -1, -2, \ldots$), each of them propagating along its own direction (angle γ) [4]. As a consequence the vdW-Z transition zone behaves as a beam splitter usable in atom interferometers. This beam splitter is tuneable since γ depends on the applied magnetic field B. In the present paper, a novel phenomenon in the domain of coherent atom optics is predicted and analysed, namely the atomic counterpart of the Fresnel biprism interferometer using vdW-Z transitions as beam splitters. The principle of this device is described in Section 2. Then a discussion based upon a realistic example will show how detailed information about the transition amplitude can be extracted

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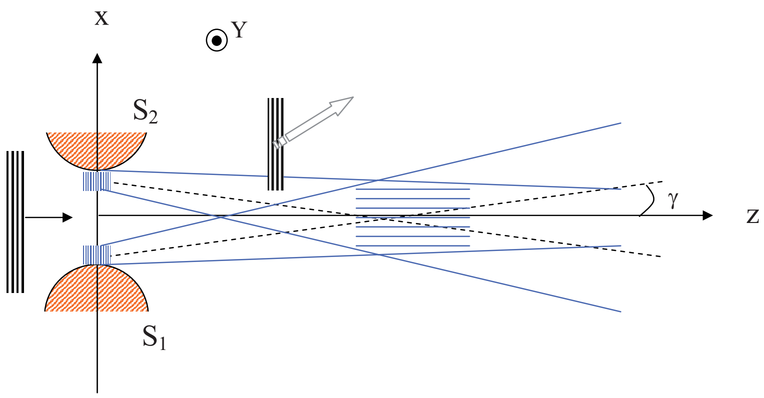

Fig. 1. (Color online) Principle of the biprism interferometer: S_1, S_2 are two opposite surfaces separated by a distance w along the x axis, able to induce the vdW-Z transition $\Delta M = -1$ in a magnetic field B. A single incident atomic wave packet in Zeeman state $M = +2$ (in black), wider than w in the x direction, generates in the vicinity of $S_{1,2}$, narrow wave packets in Zeeman state $M = +1$ (in blue) which are deflected out of the surface and strongly spread as they propagate towards $z > 0$. Meanwhile the $M = +2$ wave packet is simply elastically diffracted by the slit and then eliminated by an appropriate filter. In the overlapping zone of $M = +1$ wave packets interference fringes appear (horizontal blue lines).

from the interference pattern (Sect. 3). Finally conclusions and perspectives will be given in Section 4.

2 Principle of the atomic Fresnel biprism

Let us consider two identical opposite solid surfaces (e.g. the 2 edges of a metallic slit), separated by a distance w of a few μm (see Fig. 1). These surfaces can be modelled by parallel cylinders the radius of which is typically 1 μm, but the exact value of the curvature radius is not important for our purpose. Atoms with non-zero angular momentum, initially polarised in a Zeeman state $|M\rangle$, are assumed to be normally incident on the slit at a velocity v_i. In the following calculations and simulations, we will consider metastable argon atoms $Ar^*(^3P_2)$, at velocity 56 m/s, polarised in Zeeman state $M = +2$. In previous experiments using a special nozzle beam of Ar^* atoms at a velocity of 560 m/s [6], it has been shown that the transverse coherence radius was $R_c = 0.62$ μm. At lower velocity, this radius will be larger, certainly larger than $w/2$, even for w of a few μm. Under such conditions, a single incident atomic wave packet can touch both surfaces at the same time, generating, via the vdW-Z transition ($\Delta M = -1$), two symmetric wave packets in Zeeman state $M = +1$, the width of which along the x direction is $\rho_{ef} \approx 2-3$ nm. These packets are deflected by opposite angles $\pm\gamma$. In the present case, for argon atoms with $v_i = 56$ m/s and $B = 100$ G, $\gamma \approx 36$ mrad. Because of their small width, the packets strongly spread as they freely propagate towards $z > 0$. The related diffraction regime depends on the Fresnel number $F = a^2/(\lambda z_0)$ where a is the size of the diffracting object, λ the de Broglie wavelength and z_0 the distance of observation. If the size of the diffracting object under consideration is $\rho_{ef} \approx 2$ nm, then at $v_i = 56$ m/s, i.e. $\lambda = 0.18$ nm, one gets the Fraunhofer regime, i.e. $F \ll 1$, as soon as $z_0 \gg 22$ nm. This leads to an angular width of the corresponding diffraction pattern $\Delta\theta \approx 60-90$ mrad. It may be noted that a similar diffraction effect can be made responsible for the rather large angular width (several mrad) of the vdW-Z peaks previously observed with Ne^* atoms at 780 m/s [4,5]. Beyond some distance from the slit ($z_0 > w/[2(\gamma+\Delta\theta)]$), the two wave packets overlap, giving rise to non-localised interference fringes [7,8] of the Young-slit or Fresnel-biprism type, along the transverse (x) direction. Actually for a pair of surfaces, the diffraction regime needs to be re-examined in so far as the related Fresnel number is $F' = w^2/(\lambda z_0)$. This leads to the Fraunhofer regime provided that $z_0 \gg w^2\lambda$ For example, with $w = 5$ μm, one gets the more severe constraint $z_0 \gg 0.137$ m. In the following discussion we shall assume that the condition of validity of the Fraunhofer regime always holds. Under such conditions the diffracted amplitude at a transverse abscissa X and a distance z_0 from the slit, has a purely angular dependence, on the angle (assumed to be small) $\theta = X/z_0$. It is simply the Fourier transform of the inelastic amplitude $A_{12}(x)$ generated by the two surfaces in plane $z = 0$. Up to a constant factor it is given by:

$$u(\theta) = \int_{-\infty}^{+\infty} A_{12}(x)\, \exp[i\,k\,\theta\,x]\, dx. \tag{1}$$

Therefore, once one has eliminated the transmitted wave packet remained in the initial Zeeman state $M = +2$, one gets a purely diffractive image, an *atomic Schlieren photography*, of the vdW-Z transition amplitudes, the range of which is of a few nanometers.

The complex amplitude $A_{12}(x)$ is symmetric in x within the interval $[-w/2, +w/2]$. It is the sum:

$$A_{12}(x) = A(w/2 + x) + A(w/2 - x) \tag{2}$$

where $A(\rho)$ is the vdW-Z transition amplitude as a function of the impact parameter ρ. The two amplitudes in (2) do not overlap since $\rho_{ef} \ll w$. The Fourier transform (1) takes the form:

$$u(\theta) = e^{-ikw\theta/2} \int_0^{\varepsilon} A(\rho)\, e^{ik\theta\rho}\, d\rho + e^{+ikw\theta/2} \times \int_0^{\varepsilon} A(\rho)\, e^{-ik\theta\rho}\, d\rho \tag{3}$$

where ε is a value of ρ such that $w/2 > \varepsilon > \rho_{ef}$, with $A(\rho) \approx 0$ for $\rho > \varepsilon$. As $A(\rho) = 0$ for $\rho < 0$, the first integral in (3) is the Fourier transform $B(\theta)$ of the transition amplitude since:

$$B(\theta) = \int_{-\infty}^{+\infty} A(\rho)\, e^{ik\theta\rho}\, d\rho. \tag{4}$$

Finally one gets a diffraction amplitude $u(\theta)$ symmetric in θ:

$$u(\theta) = e^{-ikw\theta/2}B(\theta) + e^{+ikw\theta/2}B(-\theta). \tag{5}$$

From (5) one readily derives the intensity:

$$\begin{aligned} I(\theta) =& |u(\theta)|^2 \\ =& |B(\theta)|^2 + |B(-\theta)|^2 + 2|B(\theta)\,B(-\theta)| \\ & \times \cos[kw\theta - \varphi(\theta) + \varphi(-\theta)] \end{aligned} \tag{6}$$

where $\varphi(\theta) = \mathrm{Arg}[B(\theta)]$.

Young-slit-, or biprism-interference fringes are governed by the phase term $kw\theta$ which gives rise to a fast oscillation. In general the interference pattern has its central bright fringe at $\theta = 0$, but the location of lateral fringes is modified by the term $\varphi(\theta) - \varphi(-\theta)$. This effect (as well as the angular width of $|B(\theta)|$), is related to the finite spatial width of $A(\rho)$. It is especially marked when the ratio ρ_{ef}/w is not too small.

The contrast of the fringes is limited since it is given by the expression:

$$\Gamma = \frac{2|B(\theta)\,B(-\theta)|}{|B(\theta)|^2 + |B(-\theta)|^2}. \tag{7}$$

It should be noted that, if $A(\rho)$ is real, then $B(\theta) = B(-\theta)^*$ and $\Gamma \equiv 1$, but this is not the case for inelastic transition amplitudes which are generally complex. The asymmetry of $|B(\theta)|$, in other words the limited contrast, i.e. the angular behaviour of upper and lower envelopes of the fringes, reflects the oscillatory character of $A(\rho)$. This point is readily seen in assuming for instance that $A(\rho) = e^{iK\rho}$ for $0 \leq \rho \leq \varepsilon$ and $A = 0$ elsewhere, K being a real constant. Then $B(\theta) = \varepsilon \exp[\frac{i}{2}(K + k\theta)\varepsilon]$ Sinc $[\frac{1}{2}(K + k\theta)\varepsilon]$, where $\mathrm{Sinc}(u) = \sin u/u$. Oscillations in $A(\rho)$ and correlatively the shift of the maximum of $|B(\theta)|$ with respect to $\theta = 0$, are the fingerprint of the phase shift induced by the interaction potential accumulated along a rectilinear trajectory of impact parameter ρ. From a semi-classical viewpoint, the stationary-phase point corresponds to the classical deflection angle γ. In a general case, the examination of the lower and upper envelopes of the fringes allows us, in principle, to derive – up to a constant factor – the modulus $|B(\theta)|$, whereas that of the fringe spacing leads to the argument of $B(\theta)$. Nevertheless, from an experimental point of view, this evaluation is not as obvious as one could expect because of the finite velocity spread $\delta v/v$ of the atom beam (typically 1%). This velocity dispersion strongly limits the number of visible fringes, to about $v/\delta v$ (here 100, whereas, for w of a few μm, thousands of fringes are expected within the envelope). As a consequence, except in the central part of the pattern, only the mean value of the intensity, i.e. $\langle I\rangle = |B(\theta)|^2 + |B(-\theta)|^2$ can be measured. The extension of this central region is a matter of compromise.

3 Experimental constraints

We have assumed so far two diffracting surfaces strictly symmetric with respect to the z axis. Actually these surfaces depart from this ideal geometry by two types of defects: (i) some roughness adding a random contribution ξ to the width w, (ii) a misalignment relatively to the x axis, i.e. a tilt of the slit by some angle α in the $x-z$ -planes whereas the incident direction remains along z. The first defect is expected to cause an effect similar to that of the velocity dispersion, namely a loss of contrast limiting the number of visible fringes to about $w/\langle\xi\rangle$, where $\langle\xi\rangle$ is the rms value of ξ. Owing to usually accessible mechanical accuracy, this effect should be negligible compared to that of the velocity spread. The misalignment (α) would have strictly no effect in the case of an elastic process. Here however the wave number k is changed by δk, which shifts the central fringe to an angle $\theta_c = (\delta k/k)\,\alpha = (\gamma^2/2)\,\alpha \sim 10^{-3}\,\alpha$. Then this is a quite negligible effect. Another effect of the tilt, present for both elastic and inelastic processes, is a change in the open part of the grating slits which results into a change of intensity at different orders, but the diffraction peak positions are not modified. Such tilted gratings have been previously used in the analysis of the effect of the scalar vdW potential on the elastic diffraction [9]. The insensitivity of the pattern to α, i.e. to the z coordinates of the active zones of the surfaces, is clearly an advantage when the slit is deformed along the y direction, i.e. neither strictly rectilinear nor parallel to the y axis, making α randomly vary as a function of y (see Fig. 1). It is also an important advantage when a transmission grating is used. However, and precisely for this reason, tilting the slit is not a mean to significantly move the central fringe – accompanied by the narrow zone of fringe visibility – along the pattern. It is possible to use, instead of a strictly homogeneous magnetic field, a tiny longitudinal gradient along the x axis. For large distances of observation (several mm along z), such a field gradient is able to phase-shift one diffracted amplitude with respect to the other by a large number of 2π, i.e. to move the central fringe by a large number of inter-fringe spacing.

When a transmission grating is used instead of a single slit and provided that a single incident wave packet traverses N slits, only the figure of the Young-slit fringes is modified. It is simply multiplied by the standard grating factor $|\sin[(N+1)\varphi/2]/\sin(\varphi/2)|^2$ where $\varphi = k\,\Lambda\theta$ is the phase shift between the amplitudes diffracted by two successive slits, Λ being the grating period. On the other hand the envelopes, which are entirely determined by the transition amplitude profile, remain the same. A geometry of special interest is that in which $\Lambda = 2w$. In such a case, the grating diffraction peaks alternately coincide with bright and dark biprism fringes. The whole information about the transition amplitude is preserved, with, theoretically, a substantial gain on the signal since the bright-fringe intensity is multiplied by N^2. However this gain is limited by the angular resolution because the peak width is divided by N. When diffraction peaks are no longer resolved, the gain is N rather than N^2. Such a gain, N^2 or N, appears to be important from an experimental point of view because of the smallness of the expected signal. As mentioned before [4,5], van der Waals – Zeeman transitions have been

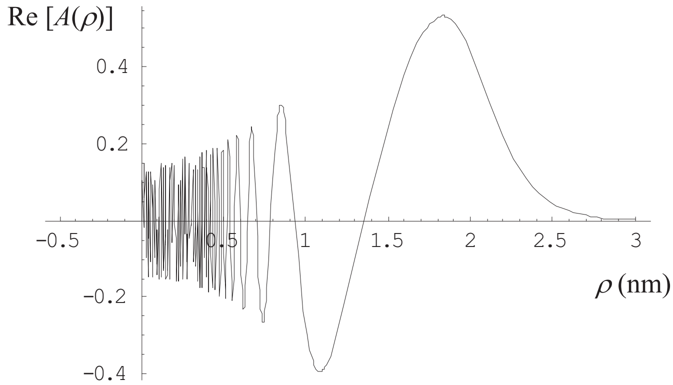

Fig. 2. Behaviour of the real part of the vdW-Z transition amplitude $A(\rho)$ generated by a surface immersed in a magnetic field (see text, Eq. (8)), as a function of the impact parameter ρ (in nm).

previously observed with fast metastable neon atoms (Ne* $^3P_2$, velocity 780 m/s) traversing a copper grating (effective slit width 5.8 μm) over 7 slits, within a magnetic field ranging from 150 to 600 Gauss. The inelastic signal for $\Delta M = -1$ produced by one series of slit upper edges has been estimated to be about 2×10^{-4} times the flux directly transmitted through the grating (about 10^5 atoms per second), which leads to 20 atoms/s in the inelastic channel. Under the present conditions (Ar* atoms slowed down at 56 m/s) one expects a transition probability about 5 times larger than the previous one, which gives the relatively reasonable value of 15 atoms/s for a single slit.

According to previous computer calculations (see Ref. [4]), a typical vdW-Z transition amplitude $A(\rho)$ can be reasonably modelled for $\rho > 0$ by the following analytical function:

$$A(\rho) = \frac{B + C\rho^2}{1 + \exp[D(\rho - \rho_{ef})]} \exp[-2\pi i E(\rho + F)^{-3}] \quad (8)$$

where (all distances are in nm) $B = 0.15$, $C = 0.2125$ nm^{-2}, $D = 7.0$ nm^{-1}, $E = 0.625$ nm^3; $F = 0.01$ nm is a cut-off parameter. The real part of $A(\rho)$, with $\rho_{ef} = 2$ nm, is shown in Figure 2. This model will essentially serve as a test for the "partial inversion" used to extract as much information as possible about $A(\rho)$ from the overall diffraction pattern, i.e. from the angular dependence of the diffracted *intensity* $I(\theta)$ given by equation (6). The Fourier transform $B(\theta)$ of $A(\rho)$ (squared modulus and argument) is shown in Figure 3a. It is seen that, as expected, $|B(\theta)|^2$ is neither symmetric nor centred. Its maximum is located at $\theta \approx -65$ mrad. This value is comparable to typical deflection angles γ, which just shows that our model is reasonable. It is interesting to compare these results to those obtained when the oscillating factor in equation (8) is suppressed (see Fig. 3b). As expected the Fourier transform $B'(\theta)$ (squared modulus and argument) becomes centred and symmetric. Actually $B(\theta)$ is simply the convolution product of $B'(\theta)$ by the frequency spectrum of the oscillations present in $A(\rho)$.

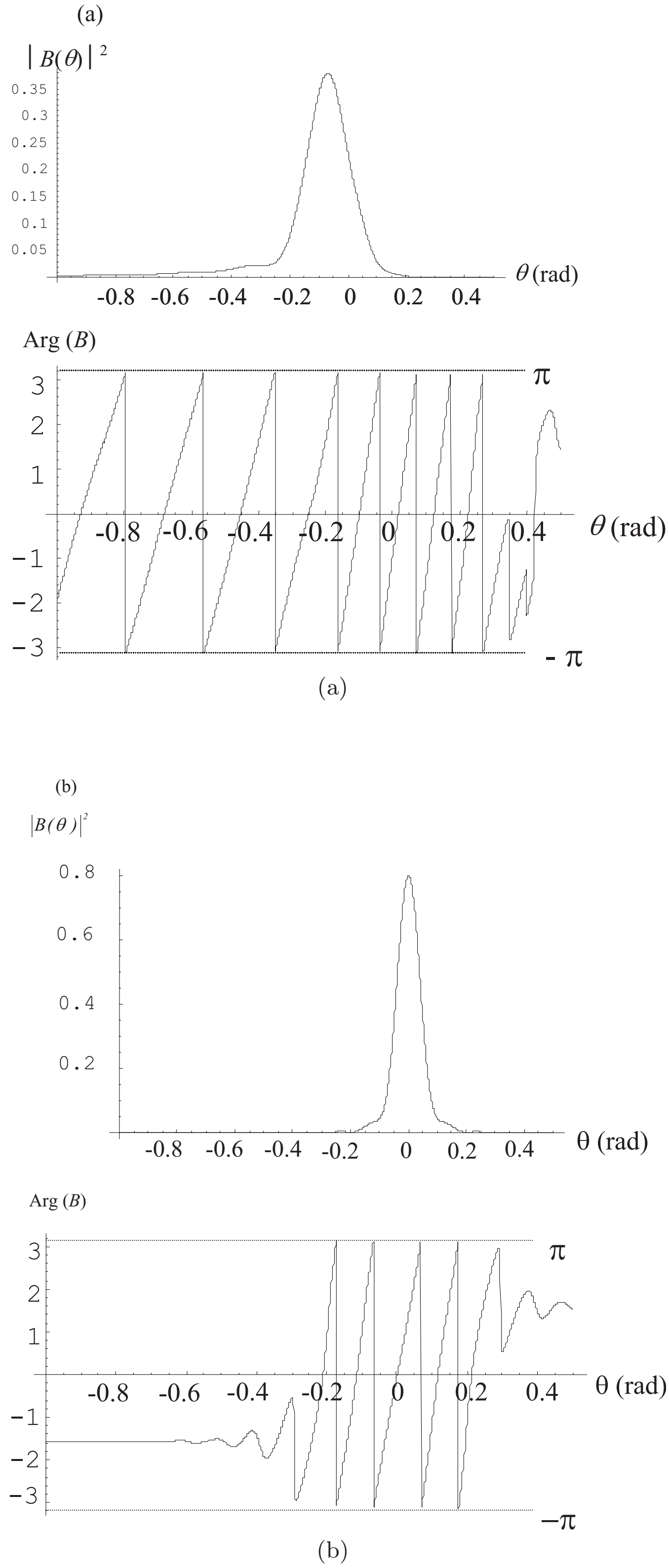

Fig. 3. (a) Fourier transform $B(\theta)$ of $A(\rho)$: upper part, squared modulus; lower part, argument (modulo [π]). (b) Same as (a) but the oscillating factor in $A(\rho)$ is ruled out. As expected the Fourier transform $B'(\theta)$ is now centred and symmetric.

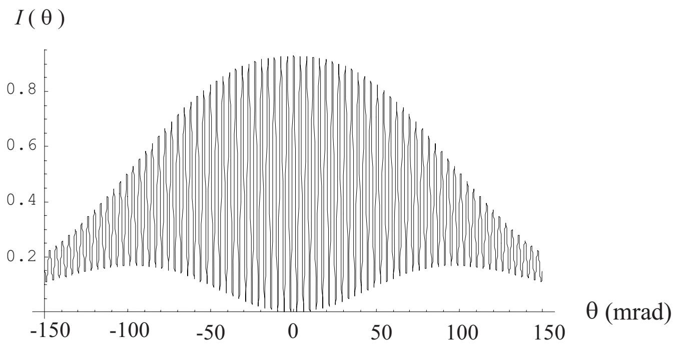

Fig. 4. Calculated interference pattern of a Fresnel atomic biprism operating with Ar* metastable atoms at a velocity of 56 m/s (de Broglie wavelength $\lambda_{dB} = 0.18$ nm), through a slit of width $w = 50$ nm. The real part of the transition amplitude $A(\rho)$ is shown in Figure 2. The fringe spacing is $\Delta\theta = 3.8$ mrad.

Therefore in principle, provided that a realistic model is chosen for the transition probability, a de-convolution procedure is able to extract the oscillatory factor present in the transition amplitude. Figure 4 shows the complete diffraction pattern for $w = 50$ nm. This rather small (but realisable) distance has been chosen in order to make visible the fringes (separated by 3.8 mrad) together with their upper and lower envelopes, which extends over about ±200 mrad.

4 Conclusion and perspectives

The examination of short-range vdW-Z transition amplitudes is made possible owing to the combination of atomic diffraction and atomic interference, at moderately low velocities. There is no doubt that such an experiment is difficult but it is worth to be undertaken because of the large amount of information it is expected to provide. Indeed not only transition probabilities are measurable but also some information about the oscillating part (the phase) of transition amplitudes can be obtained, providing us with a detailed information on the quadrupolar part of the vdW-surface interaction. The present Fresnel biprism configuration produces non-localised interference fringes. Such a device is then an *open* interferometer, in so far as its structure does not contain any well-defined single loop, contrarily to, for instance, Mach Zehnder or polarisation interferometers. As a consequence this interferometer is not sensitive to inertial effects, e.g. rotation. Various methods can be used to close the interferometer. One of the simplest one is to insert a transmission grating within the overlap region. By superposing order 0 from one side with order 1 from the other side, co-propagating phase-shifted wave packets are obtained, the wave vector matching being realized e.g. by choosing the convenient velocity. Note that in its simple open configuration, the Fresnel biprism is an instrument to directly measure the degree of transverse coherence in an atomic beam, e.g. by using a slit of an adjustable width.

The authors are members of the *Institut Francilien de Recherche sur les Atomes Froids* (IFRAF).

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