Observation of Atom-Wave Beats Using a Kerr Modulator for Atom Waves

B. Décamps, J. Gillot, J. Vigué, A. Gauguet, and M. Büchner[*]

Laboratoire Collisions Agrégats Réactivité-IRSAMC, Université de Toulouse-UPS and CNRS UMR 5589, Toulouse, France
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A phase modulation puts the atom in a coherent superposition of quantum states with different kinetic energies. We have detected the interference of such modulated waves at the output of our atom interferometer, and we have observed beats at the difference of the modulation frequencies and its harmonics, in good agreement with theory. The phase modulations were produced by a Kerr phase modulator, i.e., by the propagation of the atom wave in a time-dependent electric field. An extension of this technique to electron interferometry should open the way to very high temporal resolution in electron microscopy.

Wave beating is ubiquitous in physics [1]: It was discovered with acoustic waves and extended to electromagnetic waves, from radio frequencies to the laser domain. The extension to matter waves is feasible and requires the production of coherent superposition of quantum states of different kinetic energies. In the present Letter, we describe an atom-wave phase modulator, which is the perfect atom-optics analogue of the Kerr modulator for light waves [2], and we use it to modulate the waves propagating in the arms of an atom interferometer. We observe beats of atom waves, at the modulation frequencies and at its harmonics or, if we use two different frequencies, at the frequency difference and its harmonics. We thus achieve heterodyne experiments with matter waves.

The first use of a Kerr modulator for atom waves is due to Roberts *et al.* [3] in order to produce a tailored velocity dependence phase shift. This technique was extended to measure the mean velocity of an atomic beam [4]. However, in both experiments, no temporal modulation of the interferometer signal was detected.

A Kerr modulator can produce coherent sidebands to any atom wave, and this should open numerous possibilities. As the analogy with light waves is enlightening, let us briefly recall the history of beats with light waves: Around 1880, Righi produced phase modulations and beats thanks to rotating polarizing elements [5]; in 1955, Forrester, Gudmundsen, and Johnson observed beats of incoherent light sources [6]; in 1962, beats of independent laser beams were observed [7,8] and this technique is the basis of very accurate heterodyne measurements of laser frequencies, particularly since the development of the frequency comb [9]. Obviously, this analogy should not be pushed too far because of important differences between light and atom waves, discussed at the end of this Letter.

Coherent superpositions of quantum states of different kinetic energies have already been produced, either with neutrons or with atoms, using a large variety of techniques: reflection of the wave on a vibrating mirror [10–13],

transmission by a moving or intensity-modulated grating [14–17], Zeeman interaction with a time-dependent magnetic field [18–20], and atom-laser emitting on two or several modes [21,22]. If the modulation frequency ω is larger than $\delta E_{\rm kin}/\hbar$ ($\delta E_{\rm kin}$ is the width of the kinetic energy distribution), the modulation may be detected by observing resolved sidebands in the energy spectrum, while, in the opposite case, the only possible detection is the observation of temporal beats: Our experiment belongs to this last case.

Diffraction is the general technique to produce coherent superposition of quantum states [23]: An incident plane wave $|\mathbf{k}_i, \Omega_i\rangle$ ($\hbar\mathbf{k}_i$ is the momentum, $\hbar\Omega_i$ the energy) interacts with a perturbation periodic in space and time, of wave vector $\boldsymbol{\kappa}$ and angular frequency ω. This interaction can transfer p quanta of momentum $\hbar\boldsymbol{\kappa}$ and energy $\hbar\omega$, thus producing a superposition of plane waves $|\mathbf{k}_i + p\boldsymbol{\kappa}, \Omega_i + p\omega\rangle$. If the perturbation is periodic in time only, this is a diffraction-in-time process [24], and the transmitted wave is given by

$$|\Psi_t\rangle = \sum_p A_p |\mathbf{k}_p, \Omega_p\rangle, \tag{1}$$

with $\Omega_p = \Omega_i + p\omega$, p being the diffraction order. $|\mathbf{k}_p|$ is given by energy conservation, $|\mathbf{k}_p| \approx |\mathbf{k}_i| + pm\omega/(\hbar|\mathbf{k}_i|)$, valid if $p\omega \ll \Omega_i$, m being the particle mass.

The theory of a phase modulator is the same for light or matter waves, the main difference being due to the propagation delays which are very different, in their order of magnitude and dispersion. We simplify the calculation by considering a 1D geometry, with a plane wave propagating along the z axis through a time-dependent refractive index $n(z,t)$. Assuming that $|n(z,t) - 1| \ll 1$, the induced phase shift $\varphi_m(t_d)$ is given by

$$\varphi_m(t_d) = k_i \int [n(z, t_d - \tau) - 1] dz, \tag{2}$$

where t_d is the time at which the wave reaches the detector located in z_d, τ is the propagation delay, $\tau = (z_d - z)/v$, and v is the wave group velocity equal, for an atom, to its classical velocity. If the refractive index $n(z,t) - 1 = \varepsilon_m \cos(\omega t)$ extends over a length L centered at z_c, we get

$$\varphi_m(t_d) = \varphi_{\max} \cos[\omega(t_d - \tau_c)] \qquad (3)$$

with

$$\varphi_{\max} = \varepsilon_m k_i \left[\frac{2v}{\omega} \sin\left(\frac{\omega L}{2v} \right) \right] \quad \text{and} \quad \tau_c = \frac{z_d - z_c}{v}, \qquad (4)$$

where the expression between the square brackets takes into account the variation of the refractive index during the interaction time L/v. If this time is negligible, $\omega L/2v \ll 1$, then $\varphi_{\max} \to \varepsilon_m k_i L$. Using the Jacobi-Anger expansion [25] of $\exp[i\varphi_m(t_d)]$, the transmitted wave function at z_d is described by Eq. (1) with A_p equal to

$$A_p = J_p(\varphi_{\max}) \exp\left[ip\left(\frac{\pi}{2} - \psi_m(p) \right) \right], \qquad (5)$$

where J_p is the Bessel function of the first kind of order p and $\psi_m(p) = p\omega\tau_c$ is the phase shift due to the propagation delay.

The time-dependent refractive index $n(z,t)$ is produced by an electric field $\mathbf{E}(z,t)$ which polarizes the atom and induces a Stark energy shift [26,27]:

$$n(z,t) - 1 = \frac{2\pi\varepsilon_0\alpha}{mv^2} \mathbf{E}^2(z,t), \qquad (6)$$

where α is the static electric polarizability, provided that the modulation frequencies are negligible with respect to the first atom resonance frequency. This effect is the matter-wave analogue of the Kerr effect for light [2].

Nowadays, most phase modulators for light waves use the Pockels effect which is linear in an electric field [28]. Such a linear effect is negligibly small for matter waves, because parity and time reversal are almost exact symmetries [29]. Finally, for atoms in an S ground state, the polarizability α and, as a consequence, the index of refraction are almost independent of the Zeeman-hyperfine sublevel [30,31].

To detect phase modulations, we use our lithium Mach-Zehnder interferometer described in detail in Ref. [32] and schematically represented in Fig. 1. The lithium atomic beam produced by supersonic expansion of lithium seeded in argon has a Gaussian velocity distribution with a mean velocity $v_m \approx 1050$ m/s and a 24% FWHM, corresponding to a mean de Broglie wavelength $\lambda_{\mathrm{dB}} \approx 54$ pm and kinetic energy, expressed in frequency units, $\Omega_i/(2\pi) \approx 9$ THz. The signal is measured by a Langmuir-Taylor detector [33], with a response time due to the surface ionization process on the order of 0.12–0.3 ms (see [34] for more details): This
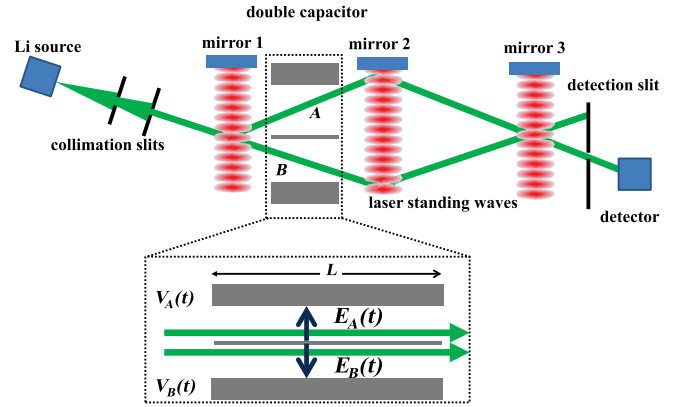

FIG. 1.　Schematic drawing of our atom interferometer (not to scale). The atomic beam collimated by two slits is diffracted by three quasiresonant laser standing waves produced by reflecting laser beams on three mirrors. Two output atomic beams carry complementary interference signals. A slit selects one of these beams whose intensity is measured by a Langmuir-Taylor surface ionization detector. The interferometer arms labeled A and B are represented by full (green) lines, and the diffraction angle, ~ 160 μrad, has been grossly exaggerated. Lower part: Detailed view of the interaction region, with the interferometer arms going through two capacitors which share a common grounded electrode. The length of the capacitor is $L \approx 48$ mm, and the distance from their center to the detector is $z_d - z_c = 1423 \pm 10$ mm.

response time is poorly known, in particular because it varies very rapidly with the surface temperature. The time resolution is further limited by the counting period equal to 1 ms. The interferometer signal is given by

$$I(t_d) = I_0\{1 + \mathcal{V}_0 \cos[\varphi_d + \Delta\varphi_m(t_d)]\}, \qquad (7)$$

where I_0 is the mean signal intensity and $\mathcal{V}_0$ the fringe visibility. φ_d is a phase due to atom diffraction, and it is used to scan the interference fringes. This phase is independent of the atom velocity.

We apply different phase modulations to the two interferometer arms (A,B), and the resulting phase shift is given by the difference: $\Delta\varphi_m(t_d) = \varphi_{m,A}(t_d) - \varphi_{m,B}(t_d)$. With the help of the double capacitor, shown in Fig. 1, we apply oscillating electric fields to both interferometer arms. Using Eqs. (2) and (6) and setting $V_A = V_0 + V_1 \cos(\omega t)$ and $V_B = -V_0 + V_1 \cos(\omega t)$, where V_0 is a voltage offset, we get

$$\Delta\varphi_m(t_d) = 4K(\omega)V_0 V_1 \cos[\omega(t_d - \tau_c)], \qquad (8)$$

with $K(\omega) \approx 4.79 \times 10^{-4}$ rad/V^2 for small ω. We achieve a maximum value $\Delta\varphi_{\max} \sim 15$ rad for $V_0 = 800$ V and $V_1 = 10$ V [34].

Figure 2 presents the interferometer signal as a function of time in the presence of a low modulation frequency, $\omega/(2\pi) = 21$ Hz, and the signal exhibits clearly the contributions of the first two harmonics. The Fourier transform
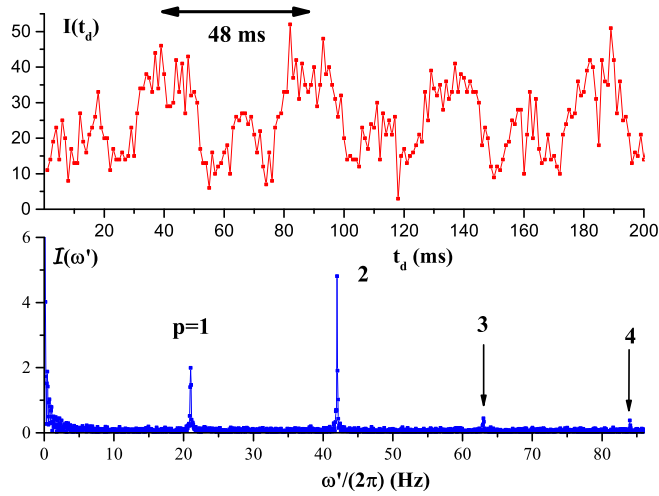
FIG. 2.   The interferometer signal in the presence of a phase $\Delta\varphi_{\max} \sim 2.7$ rad oscillating at a frequency $\omega/(2\pi) = 21$ Hz. The diffraction phase $\varphi_d \approx -0.5$ rad has been chosen to induce a strong non-linearity. Top panel: direct recording of the interferometer signal equal to the number of atoms detected per millisecond. Bottom panel: the modulus of the Fourier transform of a 16.4-s-long record reveals the presence of the first four harmonics.
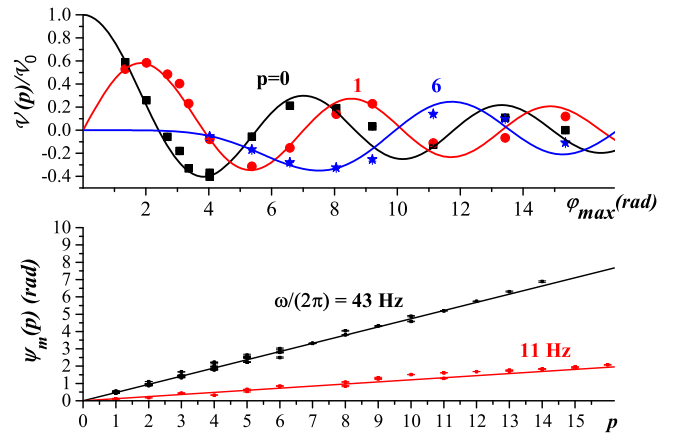

FIG. 3.   Measurement of the fringe visibility and the phase shift of the harmonics of the modulation frequency equal to 11 or 21 Hz. Upper panel: $\mathcal{V}(p)/\mathcal{V}_0$ as a function of $\Delta\varphi_{\max}$ for $p = 0$ (squares), 1 (bullets), and 6 (stars); the curves are our theoretical results, with no free parameters. Lower panel: Phase shift $\psi_m(p) = p\omega\tau_c$ as a function of the harmonic order p for two modulation frequencies $\omega/(2\pi) = 11$ and 43 Hz. The total delay τ_c deduced from these experiments is $\tau_c \approx 1.65 \pm 0.01$ ms: It is the sum of the atom propagation time, estimated near 1.36 ms, and of the delay due to the ionization process. From the measured τ_c value, we deduce an ionization delay near 0.29 ms.

of this signal also reveals the presence of the third and fourth harmonics.

We have collected many similar signals with different values of the modulation amplitude $\Delta\varphi_{\max}$. We record interference fringes by slowly scanning the phase φ_d. We apply half of the time the modulated phase $\Delta\varphi_m(t) = \Delta\varphi_{\max} \cos(\omega t)$, while no phase modulation is present for the other half.

To analyze the interferometer signal $I(t_d)$, we calculate its Fourier transform $\mathcal{I}(\omega') = \int_0^T I(t_d) \exp(-i\omega' t_d) dt_d/T$, where T is the record duration. We thus get the amplitude $\mathcal{I}(p\omega)$ of the modulation at the pth harmonics of the modulation frequency ω:

$$\frac{\mathcal{I}(p\omega)}{I_0} = \delta_{p,0} + \mathcal{V}(p) \exp[-i\psi_m(p)] \cos\left(\varphi_d + p\frac{\pi}{2}\right), \quad (9)$$

where $\delta_{p,0}$ is the Kronecker delta. [$\mathcal{I}(p\omega)$ is plotted as a function of the diffraction phase φ_d in Supplemental Material [34]]. From such plots, we extract the visibility $\mathcal{V}(p)$ and the phase $\psi_m(p)$ of the pth harmonic. Thanks to Eq. (5), these two quantities are given by $\mathcal{V}(p)/\mathcal{V}_0 = J_p(\Delta\varphi_{\max})$ and $\psi_m(p) = p\omega\tau_c$. Before comparison with experiments, the theoretical interferometer signal must be averaged over the velocity distribution, with $\Delta\varphi_{\max}$ and τ_c being both functions of the velocity: $\Delta\varphi_{\max} \propto v^{-1}$ if $\omega L/2v \ll 1$ and τ_c is the sum of the propagation time $\propto v^{-1}$ and of a smaller, velocity-independent, contribution due to the ionization process (more details in Supplemental Material [34]).

Figure 3 presents the results of a series of experiments with different values of the maximum modulation phase $\Delta\varphi_{\max}$ and two different modulation frequencies. The modulation amplitude $\mathcal{V}(p)/\mathcal{V}_0$ of harmonic p varies with the modulation phase amplitude $\Delta\varphi_{\max}$, with a Bessel-like behavior while the phase shift $\psi_m(p)$ is a linear function of the harmonic order p for a given frequency and it also increases linearly with the modulation frequency ω. All these results are in good agreement with our theoretical model (see [34], Chap. II).

We have also made experiments with the two arms modulated with different frequencies $\omega_A/(2\pi)$ and $\omega_B/(2\pi)$, and we detect the beat at the frequency difference $(\omega_A - \omega_B)/(2\pi)$ and its harmonics: This is a heterodyne experiment with matter waves. We choose $\omega_A/(2\pi)$ and $\omega_B/(2\pi)$ in the 5–30 kHz range, where no modulation can be detected because of the detector response time. We may note that, even with a fast detector, the dispersion of the delay around its mean value $\tau_c \approx 1.65$ ms would prevent the detection of these modulations. We have kept the frequency difference equal to 13 Hz while varying the two frequencies, and we have observed the beat and its first harmonics on the interferometer signal. The theory of heterodyne modulations is given in Supplemental Material [34]. We have analyzed the modulated signal as in the previous case. Figure 4 presents the modulation amplitude $\mathcal{V}(p)/\mathcal{V}_0$ of the interferometer signal for the first harmonics of the frequency difference as a function of $\omega_A/(2\pi)$. The experimental data agree reasonably well with
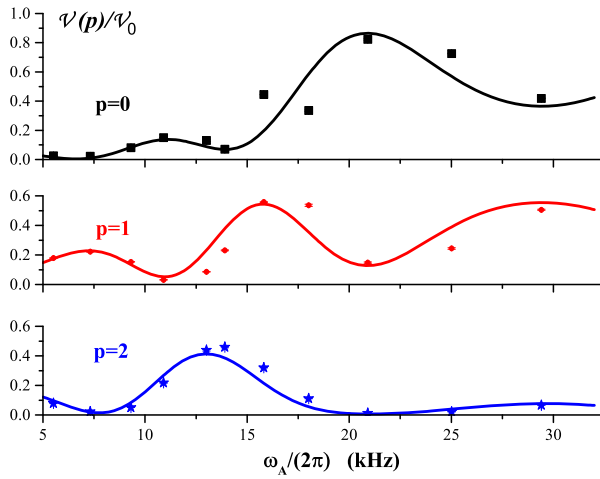
FIG. 4.   Beats of matter waves: The quantity $\mathcal{V}(p)/\mathcal{V}_0$ measured on the modulation at the pth harmonics of $(\omega_A - \omega_B)/(2\pi) = 13$ Hz is plotted as a function of the frequency $\omega_A/(2\pi)$ in kilohertz. The symbols represent our measurements, while the full lines represent the theoretical results, taking into account the velocity distribution. The value of φ_{max} is ≈ 6.7 rad.

the theoretical results (see Supplemental Material [34]) which include averaging over the velocity distribution.

Finally, we have also used our setup to apply phase modulation of atom waves to an educational purpose, transmitting sound and image. The experimental protocol and results are detailed in Supplemental Material [34].

In the present study, we have used a Kerr phase modulator for atom waves. The electric field varies periodically in time, which creates a diffraction-of-time process for atom waves. This phase modulation is detected with an atom interferometer. With only one modulation frequency, the atom interferometer signal presents oscillations at this frequency and its harmonics: The phase shift, due to the propagation delay from modulator to detector, agrees with the value calculated using the atom-wave group velocity. With two different modulation frequencies applied on the interferometer arms, the signal exhibits beats at the frequency difference and its harmonics. The observed modulations are due to the production of a coherent superposition of different kinetic energy states and this superposition is robust, although the modulation frequencies, up to 30 kHz, represent an extremely small fraction of the atom kinetic energy equal to 9 THz when expressed in frequency units. The modulations of the signal are due to the nonlinear character of quantum measurement (signal proportional to the modulus square of the wave function): This is particularly obvious for the beat frequency, as the atoms never interact with a perturbation oscillating at the beat frequency. The modulation frequencies were presently limited below ~ 50 kHz by the dispersion of the interaction time. A very short capacitor, with a length $L \sim 1$ mm, would reduce the interaction time by a factor of 50 and increase the frequency limit by the same factor. To further

reduce the interaction time, the best way is to use the dynamical Stark effect by a laser beam strongly focused on one interferometer arm: With a laser waist diameter ~ 50 μm and an atom velocity ~ 1000 m/s, the interaction time would be ~ 50 ns, allowing modulation frequencies up to 10 MHz.

In the introduction, we have recalled the impressive development of beats with light waves. A direct extension to atom waves is not straightforward because of two important differences between light and atom waves: (i) Vacuum being dispersive for atom waves, propagation will wash out fast modulations except if the source has a very narrow velocity distribution, and (ii) the response time of the atom detector, which must be smaller than the beat period, is limited by the time spent in the detection volume.

These two difficulties may be solved by using charged particles, in particular, electrons, for which extremely narrow velocity distributions can be achieved and very fast detectors are available. We propose to use the techniques described in the present Letter with an electron holographic microscope [43]: As such, a microscope is a separated arm interferometer; an electric potential applied on arm A and oscillating at the frequency ω_A will induce a modulated phase $\varphi_A(t)$, while arm B will be modulated by interaction with a sample excited by a perturbation $U(t) = U_0 \cos(\omega_B t)$. In the linear response approximation, the modification of refractive index δn_s of the sample for the electron wave also oscillates at the same frequency:

$$\delta n_s(t) = \alpha(\omega_B) \cos[\omega_B t + \psi(\omega_B)], \qquad (10)$$

where the amplitude $\alpha(\omega_B)$ and the phase shift $\psi(\omega_B)$ describe the response of the sample to the perturbation. $\delta n_s(t)$ induces a phase shift $\varphi_B(t) = k_i e \delta n_s(t)$, where k_i is the electron wave vector and e is the sample thickness. The interference signal, calculated in Supplemental Material [see Eqs. (7)–(10) [34]], will exhibit modulations at the difference frequency $(\omega_A - \omega_B)$: This heterodyne effect enables one to transfer the interesting signal at the difference frequency, which may be very low. The interferometer phase $\psi(\omega_B)$ is the response to a time-dependent perturbation of the sample in the frequency domain which is related to the time domain by Fourier transform. In fact, our proposal is analogous to the phase-shift technique [44,45] classically used to measure lifetimes of atomic or molecular excited states, and a great advantage of the phase-shift technique is that the measured lifetime can be considerably shorter than the modulation period. The extension of phase-shift techniques to electron holography should make it possible to measure the response of the sample with very high spatial and temporal resolutions.

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*matthias.buchner@irsamc.ups-tlse.fr

[1] R. Feynman, R. B. Leighton, and M. Sands, *The Feynman Lectures on Physics* (Addison-Welsley, Reading, MA, 1962), Vol. I, 2nd printing (1966).

[2] J. Kerr, Philos. Mag. **4**, 337 (1875).

[3] T. D. Roberts, A. D. Cronin, M. V. Tiberg, and D. E. Pritchard, Phys. Rev. Lett. **92**, 060405 (2004).

[4] W. F. Holmgren, I. Hromada, C. E. Klauss, and A. D. Cronin, New J. Phys. **13**, 115007 (2011).

[5] M. A. Righi, J. Phys. Theor. Appl **2**, 437 (1883).

[6] A. T. Forrester, R. A. Gudmundsen, and P. O. Johnson, Phys. Rev. **99**, 1691 (1955).

[7] A. Javan, E. A. Ballik, and W. L. Bond, J. Opt. Soc. Am. **52**, 96 (1962).

[8] T. S. Jaseja, A. Javan, and C. H. Townes, Phys. Rev. Lett. **10**, 165 (1963).

[9] S. T. Cundiff and J. Ye, Rev. Mod. Phys. **75**, 325 (2003).

[10] W. A. Hamilton, A. G. Klein, G. I. Opat, and P. A. Timmins, Phys. Rev. Lett. **58**, 2770 (1987).

[11] A. Steane, P. Szriftgiser, P. Desbiolles, and J. Dalibard, Phys. Rev. Lett. **74**, 4972 (1995).

[12] P. Szriftgiser, D. Guéry-Odelin, M. Arndt, and J. Dalibard, Phys. Rev. Lett. **77**, 4 (1996).

[13] J. Felber, R. Gähler, C. Rausch, and R. Golub, Phys. Rev. A **53**, 319 (1996).

[14] A. I. Frank, S. N. Balashov, I. V. Bondarenko, P. Geltenbort, P. Hoghoj, S. V. Masalovich, and V. G. Nosov, Phys. Lett. A **311**, 6 (2003).

[15] Th. Hils, J. Felber, R. Gähler, W. Gläser, R. Golub, K. Habicht, and P. Wille, Phys. Rev. A **58**, 4784 (1998).

[16] S. Bernet, M. K. Oberthaler, R. Abfalterer, J. Schmiedmayer, and A. Zeilinger, Phys. Rev. Lett. **77**, 5160 (1996).

[17] S. Bernet, R. Abfalterer, C. Keller, M. K. Oberthaler, J. Schmiedmayer, and A. Zeilinger, Phys. Rev. A **62**, 023606 (2000).

[18] J. Summhammer, K. A. Hamacher, H. Kaiser, H. Weinfurter, D. L. Jacobson, and S. A. Werner, Phys. Rev. Lett. **75**, 3206 (1995).

[19] E. T. Smith, Al-Amin Dhirani, D. A. Kokorowski, R. A. Rubenstein, T. D. Roberts, H. Yao, and D. E. Pritchard, Phys. Rev. Lett. **81**, 1996 (1998).

[20] R. A. Rubenstein, Al-Amin Dhirani, D. A. Kokorowski, T. D. Roberts, E. T. Smith, W. W. Smith, H. J. Bernstein, J. Lehner, S. Gupta, and D. E. Pritchard, Phys. Rev. Lett. **82**, 2018 (1999).

[21] I. Bloch, T. W. Hänsch, and T. Esslinger, Nature (London) **403**, 166 (2000).

[22] P. Bouyer, S. A. Rangwala, J. H. Thywissen, Y. Le Coq, F. Gerbier, S. Richard, G. Delannoy, and A. Aspect, J. Phys. IV France **12**, 115 (2002).

[23] Ch. J. Bordé, in *Atom Interferometry*, edited by P. R. Berman (Academic, San Diego, CA, 1997), pp. 257–292.

[24] M. Moshinsky, Phys. Rev. **88**, 625 (1952).

[25] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions, NBS*, Applied Mathematics Series–55 (U.S. GPO, Washington, DC, 1964).

[26] A. D. Cronin, J. Schmiedmayer, and D. E. Pritchard, Rev. Mod. Phys. **81**, 1051 (2009).

[27] Atom Interferometry, *Proceedings of the International School of Physics "Enrico Fermi,"* edited by G. M. Tino and M. A. Kasevich (Società Italian di Fisica, Bologne, Italy, 2014).

[28] F. Pockels, Abb. Goettingen Ges. D. Wiss. **39**, 169 (1894).

[29] M.-A. Bouchiat and C. Bouchiat, Rep. Prog. Phys. **60**, 1351 (1997).

[30] P. G. H. Sandars, Proc. Phys. Soc. **92**, 857 (1967).

[31] S. Ulzega, A. Hofer, P. Moroshkin, and A. Weis, Europhys. Lett. **76**, 1074 (2006).

[32] A. Miffre, M. Jacquey, M. Büchner, G. Trénec, and J. Vigué, Eur. Phys. J. D **33**, 99 (2005).

[33] R. Delhuille, A. Miffre, E. Lavalette, M. Büchner, C. Rizzo, G. Trénec, J. Vigué, H. J. Loesch, and J. P. Gauyacq, Rev. Sci. Instrum. **73**, 2249 (2002).

[34] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.116.053004 for more details, which includes Refs. [33, 35–42].

[35] C. R. Ekstrom, J. Schmiedmayer, M. S. Chapman, T. D. Hammond, and D. E. Pritchard, Phys. Rev. A **51**, 3883 (1995).

[36] S. Lepoutre, A. Gauguet, G. Trénec, M. Büchner, and J. Vigué, Phys. Rev. Lett. **109**, 120404 (2012).

[37] S. Lepoutre, J. Gillot, A. Gauguet, M. Büchner, and J. Vigué, Phys. Rev. A **88**, 043628 (2013).

[38] S. Lepoutre, V. P. A. Lonij, H. Jelassi, G. Trénec, M. Büchner, A. D. Cronin, and J. Vigué, Eur. Phys. J. D **62**, 309 (2011).

[39] J. P. Toennies and K. Winkelmann, J. Chem. Phys. **66**, 3965 (1977).

[40] D. R. Miller, in *Atomic and Molecular Beam Methods*, edited by G. Scoles (Oxford University, New York, 1988).

[41] Original sound file: http://www.lvbeethoven.fr/Oeuvres/HymneEuropeen-OdeALaJoie.html.

[42] Peter August Böckstiegel, self-portrait, 1917/1918, woodcut: http://www.paboeckstiegel.de/.

[43] F. Hasselbach, Rep. Prog. Phys. **73**, 016101 (2010).

[44] F. Duchinsky, Z. Phys. **81**, 7 (1933).

[45] L. Brewer, C. Geoffrey James, R. G. Brewer, F. E. Stafford, R. A. Berg, and G. M. Rosenblatt, Rev. Sci. Instrum. **33**, 1450 (1962).