Matter-wave grating distinguishing conservative and dissipative interactions

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We propose an optical grating for matter waves that separates molecules depending on whether their interaction with the light is conservative or dissipative. Potential applications include fundamental tests of quantum mechanics, measurement of molecular properties, and the ability to selectively prepare matter waves with different internal temperatures.

I. INTRODUCTION

The manipulation of quantum-mechanical waves associated with center-of-mass motion, or matter waves, has been demonstrated for a variety of atoms and molecules in a wealth of experiments [1,2]. A key element in many setups is an optical grating [3–7]. Figure 1(a) shows the conventional arrangement, where the grating is formed by overlapping two counterpropagating light beams to produce a standing wave. The energy and hence the phase of matter waves propagating through the grating, in accord with the Schrödinger equation, is modulated due to the spatially varying strength of the electric-dipole interaction. This results in a spatially varying probability density downstream: an interferogram.

An atom or a molecule can undergo two types of interaction with an optical grating: conservative and dissipative. Conservative interactions, such as the coherent transfer of a photon from one beam to the other, do not change the internal state of the atom or molecule and dominate when the frequency of the light lies far from any internal resonance. Dissipative interactions, such as the absorption of a photon from one of the beams, do change the internal state and become important at or near resonance. Their effects are somewhat more subtle than those associated with coherent processes. In particular, atoms tend to decay spontaneously soon after absorbing a photon, releasing enough which-path information to suppress quantum interference [8]. In contrast, many complex molecules prefer to redistribute absorbed photon energy *internally* through non-radiative processes which reveal no which-path information, protecting the center-of-mass coherence [9].

In this paper we propose an optical grating for complex molecular matter waves that removes the collinearity between conservative and dissipative interactions. Figure 1(b) shows our modified arrangement: by tilting the light beams used to make the optical grating with respect to one another, these effects are projected onto orthogonal axes of the transverse-momentum distribution. This separates molecules that have undergone distinct conservative and dissipative interactions and so offers the potential to coherently probe molecular ensembles with significantly different internal temperatures.

The upper half of Fig. 2(a) depicts a conservative interaction in a conventional optical grating; the molecule coherently transfers a photon from beam 1 to beam 2 and in doing so absorbs a linear momentum of $\hbar\mathbf{k}_1 - \hbar\mathbf{k}_2$. In the lower half

a sequence of purely dissipative interactions is depicted; the molecule absorbs two photons in succession from beam 1 and in doing so absorbs a linear momentum of $\hbar\mathbf{k}_1 + \hbar\mathbf{k}_1$. The linear momentum transfer is the *same* in both cases; $\hbar\mathbf{k}_1 - \hbar\mathbf{k}_2 = \hbar\mathbf{k}_1 + \hbar\mathbf{k}_1$. Conservative and dissipative interactions are thus difficult to distinguish, a challenge compounded by the fact that many of the molecule-detection methods currently employed do not discern well between different internal molecular states [9]. Figure 2(b) depicts the same interactions for our modified optical grating. The linear momentum transfer is now *different* in the two cases. The interactions can thus be distinguished and so separately exploited.

II. MODEL OF FUNCTIONALITY

We consider a molecular sample (for example, C_{70}) sublimated or evaporated from an oven at temperature T_0. A flux of these molecules emerge in an effusive manner from an orifice to form a dilute beam that is collimated and velocity selected to a degree suitable for matter-wave diffraction at an optical grating. The molecule-light interaction imposes a spatially varying phase on the matter waves and hence a spatially varying probability density downstream. We focus our attention upon the far field and work in a laboratory frame of reference x, y, and z with time t and $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$ unit vectors in the $+x$, $+y$, and $+z$ directions. The matter waves propagate in the $+z$ or longitudinal direction so that x and y define the transverse plane and the vertical direction is $\hat{\mathbf{y}}$ so that $\mathbf{g} = -|\mathbf{g}|\hat{\mathbf{y}}$ is the acceleration due to gravity.

Let the grating be comprised of two quasimonochromatic beams of light linearly polarized parallel to the z axis, one with central wave vector $\mathbf{k}'_1 = |\mathbf{k}|(\cos\theta\hat{\mathbf{x}} + \sin\theta\hat{\mathbf{y}})$ and the other with central wave vector $\mathbf{k}'_2 = |\mathbf{k}|(-\cos\theta\hat{\mathbf{x}} + \sin\theta\hat{\mathbf{y}})$, where $0 \leqslant \theta \leqslant \pi/2$. We approximate the electric field of the grating accordingly as

$$\mathbf{E}(x,y,z,t) = E_0 \mathrm{Re}[\tilde{f}(x,y)\exp(-i\omega t)]g(z)\hat{\mathbf{z}}, \quad (1)$$

with

$$\tilde{f}(x,y) = \exp(i|\mathbf{k}|\sin\theta y)\Bigg\{ \exp(i|\mathbf{k}|\cos\theta x)$$
$$\times \exp\left[-\frac{(\cos\theta y - \sin\theta x)^2}{2u^2}\right] + \exp(-i|\mathbf{k}|\cos\theta x)$$
$$\times \exp\left[-\frac{(\cos\theta y + \sin\theta x)^2}{2u^2}\right]\Bigg\} \quad (2)$$
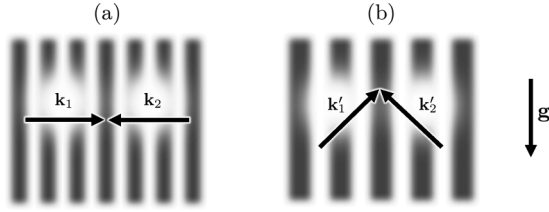
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FIG. 1. (a) A conventional optical grating formed by overlapping two counterpropagating beams of light. (b) Our modified optical grating comprised instead of tilted beams of light. Here, $\mathbf{k}_1$ and $\mathbf{k}_2$ are the central wave vectors of the beams, with primes distinguishing between conventional and modified gratings.

describing the transverse spatial variation and

$$g(z) = \exp\left(-\frac{z^2}{2w^2}\right) \tag{3}$$

describing the longitudinal variation. Each beam has a central angular frequency $\omega = c|\mathbf{k}|$, an amplitude E_0, a transverse width u, and a longitudinal width w.

A molecule introduced adiabatically to the light, while notionally being held at x, y, and z, exhibits an induced electric-dipole moment [10]

$$\boldsymbol{\mu}(x,y,z,t) = \mathrm{Re}[\tilde{\boldsymbol{\mu}}(x,y,z)\exp(-i\omega t)] \tag{4}$$

to leading order, with $\tilde{\boldsymbol{\mu}}(x,y,z) = \tilde{\alpha}\tilde{\mathbf{E}}(x,y,z)$ where $\tilde{\alpha}$ is the polarizability of the molecule at ω. For simplicity, we assume the polarizability to be identical for all internal molecular states of relevance. The use of a scalar rather than tensorial polarizability is justified for molecules like C_{70}. However, more careful treatment of orientational effects might be required for molecules of lower symmetry [11].

Consider, first, conservative interactions between the molecule and the light. The coherent transfer of photons from one beam to the other results in a potential-energy shift

$$U(x,y,z) = -\frac{1}{2}\overline{\boldsymbol{\mu}(x,y,z,t) \cdot \mathbf{E}(x,y,z,t)}, \tag{5}$$

with the overbar indicating an average over one optical cycle. In the thin grating, or Raman–Nath [3] regime a molecule traveling with moderate speed v through the grating therefore
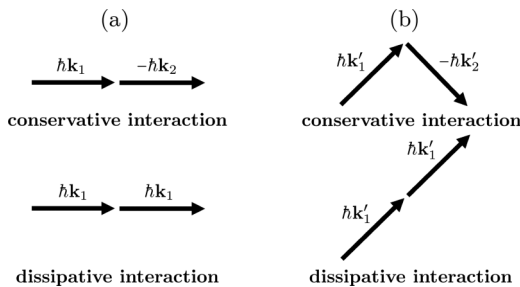

FIG. 2. (a) A conventional optical grating can transfer the same linear momentum to a molecule via conservative and dissipative interactions. (b) Our modified optical grating instead transfers different linear momenta and thus enables these interactions to be distinguished and separately exploited.

acquires an additional phase factor

$$\tilde{c}(x,y) = \exp\left[-\frac{i}{\hbar}\int U(x,y,vt)dt\right]. \tag{6}$$

Next, consider the concomitant dissipative interactions. On average the molecule at rest absorbs energy from the light at a rate of [10]

$$\Gamma(x,y,z) = \overline{\frac{d\boldsymbol{\mu}(x,y,z,t)}{dt} \cdot \mathbf{E}(x,y,z,t)}, \tag{7}$$

such that the mean number of absorbed photons for a molecule passing through the grating is

$$\overline{n}(x,y) = \frac{1}{\hbar\omega}\int \Gamma(x,y,vt)dt. \tag{8}$$

If we assume successive photon absorptions to be independent, then the probability of a particular molecule absorbing $n \in \{0,1,\dots\}$ photons is

$$p_n(x,y) = \frac{\overline{n}^n(x,y)\exp[-\overline{n}(x,y)]}{n!}. \tag{9}$$

This is justified for complex molecules like C_{70} which internally redistribute a photon's energy on a timescale short compared with successive photon absorptions [5,9]. We thus identify a combined amplitude and phase factor

$$\tilde{d}_n(x,y) = \sqrt{p_n(x,y)}\left[\frac{\tilde{f}(x,y)}{\left|\tilde{f}(x,y)\right|}\right]^n \tag{10}$$

for a molecule passing through the grating while undergoing a total of n successive photon absorptions. Each of these molecules occupies an internal state characterized by a temperature

$$T(\omega) = T_0 + \frac{n\hbar\omega}{C_V}, \tag{11}$$

where C_V is the molecule's heat capacity, which we take to be identical for all internal states of relevance. We assume that T_0 and n are sufficiently small that decoherence from the thermal emission of radiation is of negligible importance [12].

Finally, we combine the effects of conservative and dissipative interactions into one transmission function

$$\tilde{t}_n(x,y) = \tilde{c}(x,y)\tilde{d}_n(x,y). \tag{12}$$

The probability density for detecting molecules at a distance L downstream from the grating then follows from the usual far-field diffraction integral [13], the Born rule, and an incoherent sum over speeds and absorbed photon numbers [5,9],

$$P(x,y,L) \propto \int_0^\infty f(v)\sum_{n=0}^\infty \left| v\int_{-\infty}^\infty\int_{-\infty}^\infty \psi(x',y',0)\tilde{t}_n(x',y') \right.$$

$$\times \exp\left\{-\frac{iMv}{\hbar L}\left[xx' + \left(y + \frac{|\mathbf{g}|L^2}{2v^2}\right)y'\right]\right\}$$

$$\left. \times dx'dy' \right|^2 dv.$$

Here, $f(v)$ and $\psi(x,y,0)$ are the speed distribution and wave function of the matter waves incident upon the grating and M

is the mass of a single molecule. We have assumed an equal detection efficiency for all molecules and that the molecules fly horizontally in the plane of the grating. Let us emphasize here that the distinction between a conventional optical grating and our modified optical grating is essentially a geometrical one and that the molecules can be detected by using any of the myriad techniques already employed with the ability to resolve molecules spatially in two dimensions. One method is to have the diffracted molecules adsorb on a suitable surface which can then be examined by using scanning tunneling microscopy to recover the interferogram [2].

Suppose now that molecules are incident near the center of the grating, with

$$\psi(x,y,0) \propto \exp\left[-\frac{(x^2+y^2)}{2\sigma^2}\right]. \tag{13}$$

We assume the radial standard deviation σ to be small relative to u and so approximate

$$\mathbf{E}(x,y,z,t) = 2E_0\cos(|\mathbf{k}|\cos\theta x)\cos(|\mathbf{k}|\sin\theta y - \omega t)$$
$$\times \exp\left(-\frac{z^2}{2w^2}\right)\hat{\mathbf{z}}, \tag{14}$$

and

$$\tilde{t}_n(x,y) = \sqrt{n!}\exp(i\tilde{\Phi})[\mathrm{Im}(\tilde{\Phi})]^{\frac{n}{2}}\sum_{\ell=-\infty}^{\infty}i^\ell J_\ell(\tilde{\Phi}) \tag{15}$$
$$\times \sum_{q=0}^{n}\frac{\exp\{i|\mathbf{k}|[(2\ell+n-2q)\cos\theta x + n\sin\theta y]\}}{(n-q)!q!},$$

where $\tilde{\Phi} = \sqrt{\pi}\tilde{\alpha}wE_0^2/(2\hbar v)$ and $J_\ell(\tilde{\Phi})$ is a Bessel function of the first kind of order ℓ. Taking the speed distribution to be

$$f(v) \propto \exp\left[-\frac{(v-\overline{v})^2}{2(\Delta v)^2}\right], \tag{16}$$

with $\overline{v}$ being the average speed and Δv being the standard deviation of the distribution, we then obtain

$$P(x,y,L) \propto \int_0^\infty \exp\left[-\frac{(\mu-1)^2}{2(\Delta\mu)^2}\right]\mu^2\sum_{n=0}^{\infty}n![\mathrm{Im}(\tilde{\Phi})]^n$$
$$\times \sum_{\ell=-\infty}^{\infty}\left|i^\ell J_\ell(\tilde{\Phi})\sum_{q=0}^{n}\frac{1}{(n-q)!q!}\right.$$
$$\times \exp\left\{-\frac{1}{2\Sigma^2}[\xi\mu-(2\ell+n-2q)\cos\theta]^2\right\}$$
$$\times \left.\exp\left\{-\frac{1}{2\Sigma^2}(\Upsilon\mu - n\sin\theta + \Upsilon_0\mu^{-1})^2\right\}\right|^2 dv', \tag{17}$$

with $\mu = v/\overline{v}$, $\Delta\mu = \Delta v/\overline{v}$, $\Sigma = 1/\sigma|\mathbf{k}|$, $\xi = xM\overline{v}/\hbar|\mathbf{k}|L$, $\Upsilon = yM\overline{v}/\hbar|\mathbf{k}|L$, and $\Upsilon_0 = M|\mathbf{g}|L/(2\hbar|\mathbf{k}|\overline{v})$.

Figure 3(a) depicts a simulated interferogram produced by counterpropagating beams ($\theta = 0$), which is the configuration found in conventional optical gratings. Here, molecules that have undergone different dissipative interactions overlap and
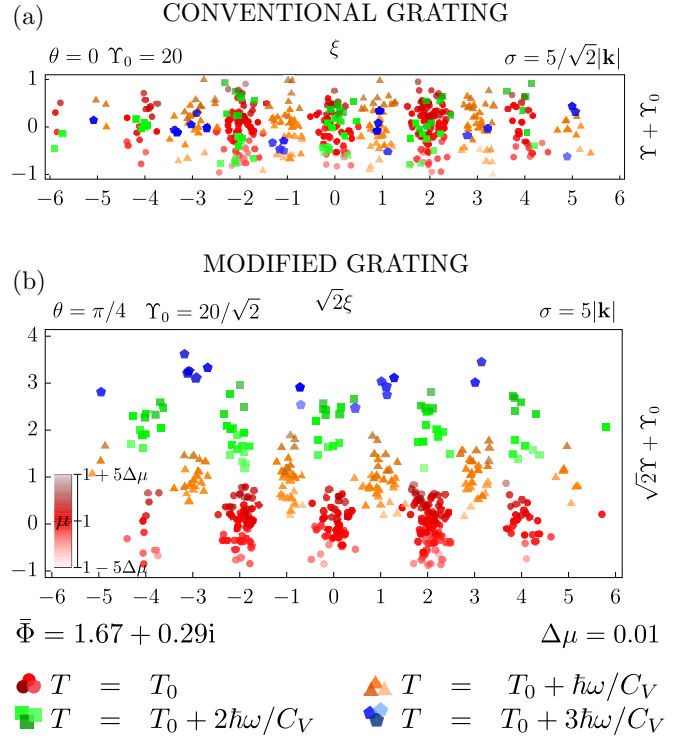

FIG. 3. (a) A simulated interferogram produced by a conventional optical grating. Molecules that have undergone different dissipative interactions with the light overlap spatially. (b) A simulated interferogram produced by our modified optical grating. Molecules that have undergone different dissipative interactions, and so are characterized by different internal temperatures, are separated. This has the added benefit of improving interference contrast. For each internal temperature the shading of the corresponding color refers to the velocity of the molecule within $5\Delta\mu$ from the mean $\mu = 1$ with faster molecules being darker and displaced upward, while slower molecules have a lighter color and are displaced downward.

cannot be distinguished spatially without ambiguity, reducing the interference contrast [5]. Thermally resolving detectors may, in principle, be able to alleviate this issue [14], because the number of photons absorbed determines the internal temperature of the molecules. This remains beyond current capabilities, however.

In contrast, Fig. 3(b) shows the interferogram produced when the light beams are tilted by an angle $\theta = \pi/4$: our modified optical grating. Here, molecules that have undergone different dissipative interactions, and therefore have different internal temperature distributions, are separated vertically. These can be identified independently, without the need for temperature-resolving detectors. In addition, the interference visibility for each particular thermal component is improved.

Note that, for the geometry presently under consideration, our modified grating demands a somewhat more stringent velocity selection than is usual (around $100\% \times \Delta v/\overline{v} \approx 1\%$, say), which may reduce the available flux of molecules and so necessitate longer measurement times. Although we have focused here on far-field diffraction, our grating can also be applied to near-field interferometry [6,7,15].

III. OUTLOOK

The ability to distinguish between molecules that have undergone different dissipative interactions with light could find use in studies of fundamental questions regarding measurement and coherence in quantum mechanics [9,12]. The production of interferograms with improved contrast [5] might facilitate better measurements of various molecular properties, such as the optical polarizability and absorption cross section [15,16]. The preparation of matter waves with different internal molecular temperatures, that can be fine tuned through ω, might find use in the study and exploitation of quantum thermodynamics, thermochemical reactions, and surface interactions, for example.

The next step is to pursue an experimental demonstration of our grating. One particularly convenient method, borrowed from the field of atom chips [17], would be to use an arrangement of reflection gratings to manipulate a single incoming laser beam into the two overlapping beams required here. The angle between the beams, and therefore the period of the optical grating thus formed, are controlled by the periods of the reflection gratings. The relative phases of the beams are automatically determined by the surfaces of the reflection gratings and ensure stable intensity fringes without active compensation. More sophisticated methods which control the polarization of the tilted beams are conceivable. We note in particular that rotating the polarization of one of the beams in our modified optical grating by $\pi/2$ yields light of uniform intensity sporting helicity fringes [18] which can be used to diffract chiral molecules [19,20]. We shall return to these and related ideas in more detail elsewhere.

ACKNOWLEDGMENTS

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

[2] T. Juffmann, H. Ulbricht, and M. Arndt, Rep. Prog. Phys. **76**, 086402 (2013).

[3] P. E. Moskowitz, P. L. Gould, S. R. Atlas, and D. E. Pritchard, Phys. Rev. Lett. **51**, 370 (1983).

[4] M. K. Oberthaler, R. Abfalterer, S. Bernet, C. Keller, J. Schmiedmayer, and A. Zeilinger, Phys. Rev. A **60**, 456 (1999).

[5] O. Nairz, B. Brezger, M. Arndt, and A. Zeilinger, Phys. Rev. Lett. **87**, 160401 (2001).

[6] S. Gerlich, L. Hackermüller, K. Hornberger, A. Stibor, H. Ulbricht, M. Gring, F. Goldfarb, T. Savas, M. Müri, M. Mayor, and M. Arndt, Nat. Phys. **3**, 711 (2007).

[7] P. Haslinger, N. Dörre, P. Geyer, J. Rodewald, S. Nimmrichter, and M. Arndt, Nat. Phys. **9**, 144 (2013).

[8] T. Pfau, S. Spälter, Ch. Kurtsiefer, C. R. Ekstrom, and J. Mlynek, Phys. Rev. Lett. **73**, 1223 (1994).

[9] J. P. Cotter, S. Eibenberger, L. Mairhofer, X. Cheng, P. Asenbaum, M. Arndt, K. Walter, S. Nimmrichter, and K. Hornberger, Nat. Commun. **6**, 7336 (2015).

[10] L. D. Barron, *Molecular Light Scattering and Optical Activity* (Cambridge University Press, Cambridge, 2004).

[11] B. A. Stickler and K. Hornberger, Phys. Rev. A **92**, 023619 (2015).

[12] L. Hackermüller, K. Hornberger, B. Brezger, A. Zeilinger, and M. Arndt, Nature (London) **427**, 711 (2004).

[13] J. W. Goodman, *Introduction to Fourier Optics* (McGraw-Hill, New York, 1996).

[14] M. Sclafani, M. Marksteiner, F. McLennan Keir, A. Divochiy, A. Korneev, A. Semenov, G. Gol'tsman, and M. Arndt, Nanotechnology **23**, 065501 (2012).

[15] L. Hackermüller, K. Hornberger, S. Gerlich, M. Gring, H. Ulbricht, and M. Arndt, Appl. Phys. B: Lasers Opt. **89**, 469 (2007).

[16] S. Eibenberger, X. Cheng, J. P. Cotter, and M. Arndt, Phys. Rev. Lett. **112**, 250402 (2014).

[17] C. C. Nshii, M. Vangeleyn, J. P. Cotter, P. F. Griffin, E. A. Hinds, C. N. Ironside, P. See, A. G. Sinclair, E. Riis, and A. S. Arnold, Nat. Nanotechnol. **8**, 321 (2013).

[18] R. P. Cameron, S. M. Barnett, and A. M. Yao, J. Mod. Opt. **61**, 25 (2014).

[19] R. P. Cameron, S. M. Barnett, and A. M. Yao, New J. Phys. **16**, 013020 (2014).

[20] R. P. Cameron, A. M. Yao, and S. M. Barnett, J. Phys. Chem. A **118**, 3472 (2014).