The Cauchy-Schwarz (CS) inequality is ubiquitous in mathematics and physics [1]. Its utility ranges from proofs of basic theorems in linear algebra to the derivation of the Heisenberg uncertainty principle. In its basic form, the CS inequality is satisfied, for example, by two classical currents emanating from a common source.

In quantum mechanics, correlations can, however, exceed classical bounds. Here we realize four-wave mixing of atomic matter waves using colliding Bose-Einstein condensates, and demonstrate the violation of a multimode CS inequality for atom number correlations in opposite zones of the collision halo. The correlated atoms have large spatial separations and therefore open new opportunities for extending fundamental quantum-nonlocality tests to ensembles of massive particles.

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than those that violate a CS inequality. Nevertheless, the importance of understanding the CS inequality in new physical regimes lies in the fact that: (i) they are the simplest possible tests of stronger-than-classical correlations, and (ii) they can be viewed as precursors, or necessary conditions, for the stricter tests of quantum mechanics.

The atom-atom correlations resulting from the collision and violating the CS inequality are measured after long time-of-flight expansion using time- and position-resolved atom detection techniques unique to metastable atoms [13]. The 307 ms long expansion time combined with a large collision and hence scattering velocity results in a ~6 cm spatial separation between the scattered, correlated atoms. This separation is quite large compared to what has been achieved in recent related BEC experiments based on double-well or two-component systems [14–16], trap modulation techniques [17], or spin-changing interactions [18, 19]. This makes the BEC collisions ideally suited to quantum-nonlocality tests using ultracold atomic gases and the intrinsic interatomic interactions.

In a simple two-mode quantum problem, described by boson creation and annihilation operators \( \hat{a}_i^\dagger \) and \( \hat{a}_i \) (\( i = 1, 2 \)), the Cauchy-Schwarz inequality of the form of Eq. (1) can be formulated in terms of the second-order correlation functions, 

\[
G_{12}^{(2)}(\Delta \mathbf{k}) = \langle \hat{n}_1 \hat{n}_2 \rangle = \langle \hat{a}_1^\dagger \hat{a}_1^\dagger \hat{a}_2 \hat{a}_2 \rangle,
\]

and reads [2–4]

\[
G_{12}^{(2)} \leq \left[ G_{11}^{(2)} G_{22}^{(2)} \right]^{1/2}, \tag{2}
\]

or simply \( G_{12}^{(2)} \leq G_{11}^{(2)} \) in the symmetric case of \( G_{11}^{(2)} = G_{22}^{(2)} \).

Here, \( G_{12}^{(2)} = G_{21}^{(2)} \), \( \hat{n}_i = \hat{a}_i^\dagger \hat{a}_i \) is the particle number operator, and the double colons indicate normal ordering of the creation and annihilation operators, which ensures the correct quantum-mechanical interpretation of the process of detection of pairs of particles that contribute to the measurement of the second-order correlation function [2]. Stronger-than-classical correlation violating this inequality would require \( G_{12}^{(2)} > \left[ G_{11}^{(2)} G_{22}^{(2)} \right]^{1/2} \), or \( G_{12}^{(2)} > G_{11}^{(2)} \) in the symmetric case.

The situation we analyze here is counterintuitive in that we observe a peak cross-correlation (for pairs of atoms scattered in opposite directions) that is smaller than the peak autocorrelation (for pairs of atoms propagating in the same direction). In a simple two-mode model such a ratio of the cross-correlation and auto-correlation satisfies the classical CS inequality. However, in order to adequately treat the atom-atom correlations in the BEC collision problem, one must generalize the CS inequality to a multimode situation, which takes into account the fact that the cross- and auto-correlations in matter-wave optics are usually functions (in our case of momentum). The various correlation functions can have different widths and peak heights, and one must define an appropriate integration domain over multiple momentum modes to recover an inequality that plays the same role as that in the two-mode case and is actually violated, as we show below.

The experimental setup was described in Refs. [11, 12]. Briefly, a cigar-shaped BEC of metastable helium, containing approximately ~10^5 atoms, trapped initially in a harmonic trapping potential with frequencies \( (\omega_x, \omega_y, \omega_z) / 2\pi = (1500, 1500, 7.5) \) Hz, was split by Bragg diffraction into two parts along the axial (z-) direction [see Fig. 1(a)], with velocities differing by twice the single photon recoil velocity \( v_{\text{rec}} = 9.2 \text{ cm/s} \). Atoms interact via binary, momentum conserving s-wave collisions and scatter onto a nearly spherical halo [see Fig. 1(b)] whose radius in velocity space is about the recoil velocity [11, 20]. The scattered atoms fall onto a detector that records the arrival times and positions of individual atoms [13] with a quantum efficiency of ~10%. The halo diameter in position space at the detector is ~6 cm. We use the arrival times and positions to reconstruct 3D velocity vectors \( \mathbf{v} \) for each atom. The scattered BECs locally saturate the detector. To quantify the strength of correlations corresponding only to spontaneously scattered atoms, we exclude from the analysis the data points containing the BECs and their immediate vicinity \( |v_x| < 0.5 v_{\text{rec}} \) and further restrict ourselves to a spherical shell of radial thickness 0.9 < \( v_y / v_{\text{rec}} < 1.1 \) (where the signal to noise is large enough), defining the total volume of the analyzed region as \( V_{\text{data}} \).

Using the atom arrival and position data, we can measure the second-order correlation functions between the atom number densities \( \hat{n}(\mathbf{k}) \) at two points in momentum space, \( G_{ij}^{(2)}(\mathbf{k}, \mathbf{k}') = \langle \hat{n}(\mathbf{k}) \hat{n}(\mathbf{k}') \rangle \) (see Supplementary Material [21]), with \( \mathbf{k} \) denoting the wave-vector \( \mathbf{v} = m \mathbf{v} / h \) and \( h \mathbf{k} \) the momentum. The correlation measurements are averaged over a certain counting zone (integration volume \( V \)) on the scattering sphere in order to get statistically significant results. By choosing \( \mathbf{k}' \) to be nearly opposite or nearly collinear to \( \mathbf{k} \), we can define the averaged back-to-back (BB) or collinear (CL) correlation functions,

\[
G_{BB}^{(2)}(\Delta \mathbf{k}) = \int_V d^3 k \ G^{(2)}(\mathbf{k}, -\mathbf{k} + \Delta \mathbf{k}), \quad (3)
\]

\[
G_{CL}^{(2)}(\Delta \mathbf{k}) = \int_V d^3 k \ G^{(2)}(\mathbf{k}, \mathbf{k} + \Delta \mathbf{k}), \quad (4)
\]

which play a role analogous to the cross- and auto-correlation functions, \( G_{12}^{(2)} \) and \( G_{ii}^{(2)} \), in the simple two-mode problem discussed above. The BB and CL correlations are defined as functions of the relative displacement \( \Delta \mathbf{k} \), while the dependence on \( \mathbf{k} \) is lost due to the averaging.

The normalized BB and CL correlations functions, \( g_{BB}^{(2)}(\Delta \mathbf{k}) \) and \( g_{CL}^{(2)}(\Delta \mathbf{k}) \), averaged over the unexcised part of the scattering sphere \( V_{\text{data}} \) are shown in Fig. 2. The BB correlation peak results from binary, elastic collisions between atoms, whereas the CL correlation peak is a variant of the Hanbury Brown and Twiss effect [22, 23]—a two-particle interference involving members of two different atom pairs [9, 10, 24, 25]. Both correlation functions are anisotropic because of the anisotropy of the initial colliding condensates.

An important difference with the experiment of Ref. [9] is that the geometry in the present experiment (with vertically elongated condensates) is such that the observed widths of the correlation functions are not limited by the detector resolution. Here we now observe that the BB and CL correlations have very different widths, with the BB width being significantly larger than the CL width. This broadening is largely due to the
size of the condensate in the vertical direction (∼1 mm). The elongated nature of the cloud and the estimated temperature of ∼200 nK also means that the condensates correspond in fact to quasicondensates [26] whose phase coherence length is smaller than the size of the atomic cloud. The broadening of the BB correlation due to the presence of quasicondensates will be discussed in another paper [27], but we emphasize that the CS inequality analyzed here is insensitive to the detailed broadening mechanism as it relies on integrals over correlation functions. This is one of the key points in considering the multimode CS inequality.

Since the peak of the CL correlation function corresponds to a situation in which the two atoms follow the same path, we can associate it with the auto-correlation of the momentum of the particles on the collision sphere. Similarly, the peak of the BB correlation function corresponds to two atoms following two distinct paths and therefore can be associated with the cross-correlation function between the respective momenta. Hence we realize a situation in which one is tempted to apply the CS inequality to the peak values of these correlation functions. As we see from Fig. 2, if one naively uses only the peak heights, the CS inequality is not violated since \(g_{BB}^{(2)}(0) < g_{CL}^{(2)}(0)\) and hence \(g_{BB}^{(2)}(0) < g_{CL}^{(2)}(0)\) due to the nearly identical normalization factors [21].

We can, however, construct a CS inequality that is violated if we use integrated correlation functions, \(\overline{g}_{ij}^{(2)}\), that correspond to atom numbers \(\hat{N}_i = \int d^3k \hat{\alpha}_i(k) \hat{\alpha}_j(k)\) in two distinct zones on the collision halo [21]:

\[
\overline{g}_{ij}^{(2)} = \langle \hat{N}_i \hat{N}_j \rangle = \int \frac{d^3k}{V_i} \int \frac{d^3k'}{V_j} \overline{g}^{(2)}(k, k').
\]

The choice of the two integration (zone) volumes \(V_i\) and \(V_j\) determines whether the \(\overline{g}_{ij}^{(2)}\) correlation function corresponds to the BB \((i \neq j)\) or CL \((i = j)\) correlation functions, Eqs. (3) and (4).

The CS inequality that we can now analyze for violation reads \(\overline{g}_{12}^{(2)} \leq \overline{g}_{11}^{(2)} \overline{g}_{22}^{(2)}\). To quantify the degree of violation, we introduce a correlation coefficient

\[
C = \frac{\overline{g}_{12}^{(2)}}{\overline{g}_{11}^{(2)} \overline{g}_{22}^{(2)}}^{1/2},
\]

which is smaller than unity classically, but can be larger than unity for states with stronger-than-classical correlations.

In Fig. 3 we plot the correlation coefficient \(C\) determined from the data for different integration zones \(V_1\) and \(V_2\), but always keeping the two volumes equal. When \(V_1\) and \(V_2\) correspond to diametrically opposed, correlated pairs of zones (red circles), \(C\) is greater than unity, violating the CS inequality, while for neighboring, uncorrelated pairs (blue squares) the CS inequality is not violated. The figure also shows the results of a quantum-mechanical calculation of \(C\) using a stochastic Bogoliubov approach (green solid curve) [20, 21, 28]. The calculation is for the initial total number of atoms \(N = 85\,000\) and is in good agreement with the observations. The choice of large integration volumes (small number of zones \(M\)) results in only weak violations, while using smaller volumes (large \(M\)) increases the violation. This behavior is to be expected [21] because large integration zones include many, uncorrelated events which dilute the computed correlation. The saturation of \(C\), in the current arrangement of integration zones – with a fixed number of polar cuts and hence a fixed zone size along \(z\) which always remains larger than the longitudinal correlation width – occurs when the tangential size of the zone begins to approach the transverse width of the CL correlation function. If the zone sizes were made smaller in all directions, we would recover the situation applicable to the
peak values of the correlation functions (and hence no CS violation) as soon as the sizes become smaller than the respective correlations widths (see Eq. (S11) in [21]).

We have shown the violation of the CS inequality using the experimental data of Ref. [11] in which a sub-Poissonian variance in the atom number difference between opposite zones was observed. Although the two effects are linked mathematically in simple cases, they are not equivalent in general [8, 21]. Because of the multimode nature of the four-wave mixing process, we observe stronger (weaker) suppression of the variance below the shot-noise level for the larger (smaller) zones (see Fig. 3 of [11]), whereas the degree of violation of the CS inequality follows the opposite trend. This difference can be of importance for other experimental tests of stronger-than-classical correlations in inherently multimode situations in matter-wave optics.

The nonclassical character of the observed correlations implies that the scattered atoms cannot be described by classical stochastic random variables [29]. Our experiment is an important step towards the demonstrations of increasingly restrictive types of nonlocal quantum correlations with matter waves, which we hope will one day culminate in the violation of a Bell inequality as well. In this case, the nonclassical character of correlations will also defy a description via a local hidden variable theory [4, 29]. Non-optical violations of Bell’s inequalities have so far only been demonstrated for pairs of massive particles (such as two trapped ions [30] or proton-proton pairs in the decay of $^2$He [31]), but never in the multi-particle regime. The BEC collision scheme used here is particularly well-suited for demonstrating a Bell inequality violation [32] using an atom optics analog of the Rarity-Tapster setup [33].

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