Neutral dipolar molecules can be decelerated and trapped using time-varying inhomogeneous electric fields. This has been demonstrated only for molecules in low-field seeking states, but can, in principle, be performed on molecules in high-field seeking states as well. Transverse stability is then much more difficult to obtain, however, since molecules in high-field seeking states always experience a force towards the electrodes. Here we demonstrate that an array of dipole lenses in alternate gradient configuration can be used to maintain transverse stability. A pulsed beam of metastable CO in high-field seeking states is accelerated from 275 to 289 m/s as well as decelerated from 275 to 260 m/s.

The same situation is encountered in charged particle accelerators, where this problem has been resolved by applying the alternate gradient (AG) focusing method [10]. This method came out of the realization that a long-established fact of geometrical optics was applicable to ion optics as well; for a pair of lenses that have equal focal lengths but with one lens converging and the other diverging, the total focal length is always positive. This same principle can be applied to polar molecules when using electrostatic dipole lenses. These lenses focus the molecular beam in one direction but simultaneously defocus the beam in the orthogonal direction. By alternating the orientation of these lenses, an electric field geometry with a focusing effect in both directions can be created. By switching these lenses on and off at the appropriate times, AG focusing and deceleration of a molecular beam can be achieved simultaneously.

The application of AG focusing to polar molecules was first considered by Auerbach et al. [11] in a design study for a neutral particle accelerator at the University of Chicago [12]. AG focusing was experimentally demonstrated by Kakati and Lainé [13] and by Günther and co-workers [14]. Although the Chicago group demonstrated AG focusing of a molecular beam as well, their attempts to demonstrate acceleration remained without success [15]. More recently, AG focusing has been used to image an atomic beam [16]. In this Letter we demonstrate acceleration and deceleration of polar molecules in high-field seeking states. A pulsed beam of metastable CO is accelerated from 275 to 289 m/s as well as decelerated from 275 to 260 m/s using an array of dipole lenses in AG configuration.

A scheme of the experimental setup is shown in Fig. 1. The prototype AG decelerator consists of 12 dipole lenses, positioned symmetrically around the molecular beam axis. One of the lenses is shown enlarged in the inset at the left-hand side of Fig. 1, along with a ($\hat{x}, \hat{y}, \hat{z}$)-axis system. A lens is composed of two identical thin electrodes (2 mm thick) with the side facing the molecular beam rounded off with a radius of curvature of 1 mm. The distance between
the two electrodes is 2 mm. Typically, voltages of +10 and −10 kV are applied to these electrodes, yielding an electric field at the molecular beam axis close to 90 kV/cm. The electric fields along \( \hat{x} \) and \( \hat{y} \) are shown in the inset at the right-hand side of Fig. 1. It is seen that molecules in high-field seeking states experience a force that repels them from the molecular beam axis in the plane of the electrodes, and attracts them toward the molecular beam axis in the plane perpendicular to this. The force acting on a polar molecule is \( \mathbf{F} = -\nabla W(E) \), with \( W(E) \) being the Stark energy of the molecule in the electric field of magnitude \( E \). Based on symmetry arguments, \( \nabla \cdot \mathbf{F} = 0 \) on the molecular beam axis for a molecule with a first or second order Stark interaction [11]. Assuming the electrodes to be infinitely long, \( F_z \) is equal to zero, and it follows that \( \partial F_x/\partial x = -\partial F_y/\partial y \).

Close to the molecular beam axis the force is linear to the displacement. For molecules moving with a constant velocity \( \nu \) along the molecular beam axis, the equations of motion along \( \hat{x} \) and \( \hat{y} \) can therefore be written as

\[
\frac{\partial^2 z}{\partial t^2} + \Omega^2 z = 0 \quad \text{and} \quad \frac{\partial^2 y}{\partial t^2} - \Omega^2 y = 0
\]

with \( \Omega = \sqrt{|k_x|/\mu \nu^2} \) and \( k_x \) and \( k_y \) the force constants. Following standard procedures in ion optics [10,17] the general solution in \( \hat{x} \) and \( \hat{y} \) can be written in matrix form as

\[
\begin{pmatrix}
\frac{\partial x}{\partial t} \\
\frac{\partial y}{\partial t}
\end{pmatrix} = \begin{pmatrix}
\cosh \Omega L & \frac{1}{\nu \Omega} \sinh \Omega L \\
\sinh \Omega L & \frac{1}{\nu \Omega} \cosh \Omega L
\end{pmatrix} \begin{pmatrix}
\frac{\partial x_0}{\partial t} \\
\frac{\partial y_0}{\partial t}
\end{pmatrix} = \begin{pmatrix}
B \frac{\partial x_0}{\partial t} \\
B \frac{\partial y_0}{\partial t}
\end{pmatrix},
\]

where the \( 2 \times 2 \) matrices describe the transfer of the molecules through a focusing (\( F \)) and defocusing (\( D \)) lens of length \( L \). The path of a molecule through the array of lenses can now be calculated, realizing that the molecules pass alternatingly through focusing and defocusing lenses. The field-free regions of length \( s \) in between the lenses are described by a simple transfer matrix \( O \) of the form

\[
O = \begin{pmatrix}
1 & s/\nu_z \\
0 & 1
\end{pmatrix}.
\]

The transfer matrix of the complete AG decelerator with \( 2N \) lenses is given by \( (FODO)^N \). In order for the transverse motion to be stable it is necessary that

\[
\text{all the elements of this transfer matrix remain bounded when } N \text{ increases indefinitely, which is the case when } -1 < \frac{1}{2} \text{Tr} (FODO) < +1.
\]

In Fig. 2 typical trajectories through the decelerator are shown using experimental parameters. The gray-shaded area indicates the region of stable trajectories. It is seen that the beam envelope is larger at the positive lenses than at the negative lenses.

In order to decelerate or accelerate the molecules, time-varying electric fields are applied. A molecule in a high-field seeking state will gain kinetic energy as it enters the field of a lens, while it loses kinetic energy as it leaves the lens. When the electric field is switched on when the molecule is inside a lens, there will be no change to its kinetic energy but it will decelerate while leaving the lens. The moment the field is switched on determines the effective length \( L \) of the lens; the focusing properties

FIG. 1. Scheme of the experimental setup. In the inset at the left-hand side of the figure an enlarged view of a lens is shown together with a \( (\hat{x}, \hat{y}, \hat{z}) \)-axis system. The electric fields along \( \hat{x} \) and \( \hat{y} \) are shown in the inset at the right-hand side of the figure.

FIG. 2. A number of trajectories through the AG decelerator, calculated using the matrix formalism described in the text, are shown. The gray-shaded area shows the beam envelope.
can thus be optimized without affecting the deceleration properties. Phase stable operation [7] can be obtained by switching the electric fields off again when the molecules have not yet left the field of a lens completely. In that case, molecules which are at the head of the pulse will lose more kinetic energy while molecules which are at the pulse’s tail will lose less kinetic energy. Therefore, molecules in a narrow position and velocity interval will be confined to this area of phase space throughout the decelerator (see Fig. 3). It is convenient to define a phase angle \( \phi \) as a measure of the energy loss per stage \( \Delta W = W_0 \sin \phi \), with \( W_0 \) the maximum (negative) Stark shift on the molecular beam axis.

In order to demonstrate the performance of the alternate gradient decelerator, experiments have been carried out on CO molecules in the \( a^3\Pi \) state. The main reason for choosing metastable CO molecules is that (i) they can be prepared in a single quantum state at a well-defined position and time, and (ii) their velocity distribution can be readily recorded.

A pulsed beam of CO is produced by expanding a 5% CO in Xe mixture into vacuum, using a modified solenoid valve. Cooling the valve housing to 180 K (just above the boiling point of Xe) reduces the mean velocity of the CO molecules in the beam to 275 m/s, corresponding to an initial kinetic energy of \( E_{\text{kin}} = 89 \text{ cm}^{-1} \). The velocity spread is approximately 10%, corresponding to a temperature of \( \sim 0.5 \text{ K} \) in the moving frame. The CO molecules pass through a 1.0-mm-diam skimmer into a second, differentially pumped, vacuum chamber. Metastable CO molecules in a single quantum state are prepared by direct laser excitation of the ground state molecules on the spin-forbidden \( a^3\Pi(v' = 0) \leftrightarrow X^1\Sigma^+(v'' = 0) \) transition, using pulsed 206 nm (6.0 eV) radiation. For this experiment the molecules need to be prepared in states that experience a negative Stark shift which are the lower components of the \( \Lambda \) doublets in the \( a^3\Pi \) state [18]. In the experiments reported here, laser preparation of the \( J' = 1 \ a^3\Pi \) level via the \( R_2(0) \) transition is used. By setting the polarization of the laser perpendicular to the stray fields present in the decelerator only the \( M \Omega = 1 \) high-field seeking state is excited.

Laser preparation is performed in a 1-mm-diam spot, 5 mm in front of the 35-cm-long AG decelerator consisting of an array of 12 equidistant 27-mm-long dipole lenses. Successive lenses are separated by a 4-mm-long drift region. The two opposing electrodes of a lens are simultaneously switched by two independent high voltage switches to maximum voltages of +10 and −10 kV. The first and last lenses have just half the length in order to have a more symmetric acceptance along \( x \) and \( y \). The time-of-flight (TOF) distributions over the 54 cm distance from laser preparation to detection are recorded by measuring the number of electrons emitted from a flat gold surface when the metastable CO molecules impinge on it.

In Fig. 4 the measured TOF distributions are plotted for several values of \( \phi_0 \), as indicated. The lower curve is the TOF distribution of the original beam, when no voltages are applied, corresponding to a beam with a mean velocity of 275 m/s. Using the 12 stages, molecules are decelerated from 275 to 260 m/s, or accelerated from 275 up to 289 m/s, depending on the phase angle \( \phi_0 \) that is used. The thin curves show the results of a Monte Carlo simulation using a finite element method to calculate the electric fields. The calculations are seen to describe the TOF distributions for the decelerated bunch rather well; similarly, good agreement is obtained in the simulation for the accelerated beam (not shown). In order to match the measured TOF distributions we have scaled down the simulations by a factor of 20; upon applying the electric fields the integrated signal decreases by a factor of \( \sim 7 \), where we would have expected an increase in signal by at least a factor of 3. This discrepancy is most likely due to misalignments in the decelerator; with a random displacement of \( \pm 0.2 \text{ mm} \) of the lenses, the observed TOF distributions can be quantitatively reproduced.

The proof-of-principle experiment described in this Letter demonstrates the potential of an AG decelerator for producing beams of polar molecules with arbitrarily (low) velocities. In the deceleration process the phase-space density remains constant; bunches of molecules are kept together independent of the number of lenses used in the
parity violation induced by the weak interaction [25]. The
at testing the time reversal symmetry [24] and at testing
used to slow down molecules that are used in studies aimed
in particular, the AG decelerator can be
heavy molecules with large dipole moments and small ro-
erating not only small polar molecules but also relatively
feasible. The AG deceleration scheme is suited for decel-
or of phase-space density via evaporative cooling might be
via inelastic collisions [23]. Therefore, a further increase
est energy quantum state, which are stable against losses
scheme is applicable to samples of molecules in their low-
storage ring [11,22]. The AG deceleration and trapping
trapped in an electrodynamic trap [19,20] or in a ring ge-
expected to be less critical to misalignments. Once suf-
tor must be improved. Alternatively, a geometry where a
be required to bring the metastable CO molecules to a total
 With the present geometry, 90 lenses would
be to bring the metastable CO molecules to a total standstill.
the mechanical design of the decelerator must be improved. Alternatively, a geometry where a
single lens is composed of multiple deceleration stages can
be used, i.e., a geometry with a number of subsequent elec-
trode pairs oriented along the same plane. The latter design
is expected to be less critical to misalignments. Once suf-
ciently slow (below, say, 25 m/s) the molecules can be
trapped in an electrodynamic trap [19,20] or in a ring ge-
ometry, for instance, around a wire [18,21] or in an AG
storage ring [11,22]. The AG deceleration and trapping
scheme is applicable to samples of molecules in their low-
est energy quantum state, which are stable against losses
via inelastic collisions [23]. Therefore, a further increase
of phase-space density via evaporative cooling might be
feasible. The AG deceleration scheme is suited for decel-
erating not only small polar molecules but also relatively
heavy molecules with large dipole moments and small ro-
tational constants. In particular, the AG decelerator can be
used to slow down molecules that are used in studies aimed
at testing the time reversal symmetry [24] and at testing
parity violation induced by the weak interaction [25]. The
increased interaction time when using slow molecules will
be highly beneficial for the attainable spectral resolution
in these studies.

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