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Two-Source Double-Slit Interference in Angle-Resolved High-Energy Above-Threshold Ionization Spectra of Diatoms

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When an electron from a diatomic molecule undergoes tunneling-rescattering ionization, a novel form of destructive interference can be realized that involves all four geometric orbits that are available to the electron when it is freed, because both ionization and rescattering may take place at the same or at different centers. We find experimentally and confirm theoretically that in orientation-averaged angle-resolved high-order above-threshold ionization spectra the corresponding destructive interference is visible for O_2 but not for N_2. This effect is different from the suppression of ionization that is well known to occur for O_2.

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Interference of matter waves is at the heart of quantum mechanics. Its simplest manifestation may be the famous double-slit experiment, as realized comparatively recently by Jönsson [1]. Electron emission from diatomic molecules is conceptually similar, but instead of the electron passing through two slits in a screen it is ejected from a molecular orbital that is described by a linear combination of two atomic orbitals localized at the position of either atom. The electron waves emitted coherently from the two atoms exhibit an intrinsic interference effect equivalent to the double-slit interference. Cohen and Fano [2] predicted this interference effect more than 40 years ago. However, only recently was the double-slit interference in electron emission from diatomic molecules such as H_2 and N_2 observed in ion-impact [3,4] and photon-impact [5–8] experiments.

Electron emission from diatomic molecules in strong laser fields should also exhibit double-slit interference. Without the interference, within the conventional Ammosov-Delone-Krainov tunneling model, the ionization rates of molecules are supposed to be identical to those of their companion atoms with (approximately) the same ionization energies. Indeed, nearly identical strong-field ionization rates were measured for N_2 and its companion Ar [9–11]. However, similar experiments revealed that the ionization rate of O_2 is an order of magnitude lower than the rate for its companion Xe [10–12]. According to the theoretical studies [11,13], tunnel ionization is suppressed in those homonuclear molecules that possess a valence orbital with antisymmetric character (with respect to the reflection symmetry of homonuclear molecules), like the outermost π_g orbital in O_2, owing to destructive rather than constructive interference of the contributions of emission from the two atoms.

Half of the electrons released by tunnel ionization (those set free between an extremum of the field and its subsequent zero crossing) will be driven back by the oscillating electric field into a recollision with their parent ion [14]. If these electrons are elastically backscattered, they may subsequently gain additional energy from the laser field to form the plateau of the high-energy above-threshold ionization (HATI) spectrum. HATI electrons that undergo close collisions with their parent ion and scatter by a large angle provide an excellent probe of its structure [15]. Indeed, Okunishi *et al.* [16] and Ray *et al.* [17] have proven that it is possible to extract electron-ion elastic differential cross sections from the angular distributions of the HATI electrons of rare-gas atoms.

Two-source double-slit interference effects occur if the returning electrons are rescattered by the diatomic molecular ion. There are at least four geometrical paths available to the electron on its way from its initial state into a final state with specified momentum, as its orbit in the continuum may originate at the position of one or the other center of the diatom. Milošević *et al.* discussed how this two-source double-slit interference manifests itself in the HATI spectra of N_2 and O_2 with fixed orientation [18,19]. However, without special alignment techniques, molecules are randomly oriented. For conventional electron emission from diatomic molecules, even though the molecules are randomly oriented, double-slit-interference effects survive after averaging over the orientations, as predicted by Cohen and Fano [2] and experimentally observed [3,4,6]. Destructive double-slit interference also survives for tunneling ionization from the antisymmetric orbital (such as π_g) of randomly oriented molecules in the case of O_2.

In the present Letter, we demonstrate experimentally that this two-source double-slit interference remains vis-

ible in the angle-resolved electron energy spectra for randomly oriented O_2 molecules but not so for N_2. We will show by simulations that, in contrast to the double-slit interference in "direct ionization" where rescattering is not involved, the manifestation of the two-source double-slit interference in *fixed-orientation* spectra actually is very similar for N_2 and O_2. However, in the *orientation-averaged* spectra, molecular symmetry effects mask the two-source double-slit pattern in N_2, while it remains visible in O_2.

The experimental setup is almost the same as that used previously [16,20,21]. The fundamental output (800 nm) from an amplified Ti:sapphire laser system (pulse width: 100 fs, repetition rate: 1 kHz) was used as the ionizing radiation. The laser beam was introduced into the vacuum chamber and focused by a $f = 60$ mm lens to ionize the sample molecules. The sample gas was effusively introduced into the vacuum chamber. Electrons were detected by using a 264 mm-long linear time-of-flight (TOF) spectrometer with a limited detection angle ($\sim 0.0014 \times 4\pi$ sr). The time difference between the laser pulse and the arrival time of the electron was recorded by a time-to-digital converter (TDC). The overall electron energy resolution achieved was $E/\Delta E > 50$. We avoided dead time and saturation effects by adjusting the gas pressure to a level where less than one electron was detected per laser shot. The working pressures in the vacuum chamber were 6×10^{-6} mb for N_2 and 3×10^{-7} mb for O_2, while the base pressure was less than 10^{-9} mb. The resulting electron count rates were typically 400 counts/sec, but most of the counts came from direct ionization. Only a few percent of the counts correspond to the rescattering process.

The polarization of the linearly polarized laser pulse is initially horizontal, i.e., in the direction of the electron TOF, but the polarization direction can be rotated in any direction perpendicular to the beam by using a $\lambda/2$ plate. For angular-distribution measurements, the $\lambda/2$ plate was mounted on a rotator with a constant rotation period of 60 000 laser shots. One of the additional channels of the TDC module is used to record the absolute position of the rotator. In this way, the spectra for all angles can be recorded at virtually the same time. Averaging over several tens of hours of rotation, the angular distributions are free of the effects due to laser power variations.

In Fig. 1, we compare measured and calculated angle-resolved electron energy spectra for N_2 and O_2. The simulations were done with the molecular strong-field approximation (MSFA) [22] extended to include rescattering as introduced and applied in Refs. [18,19]. Averaging over the laser pulse (with a Gaussian intensity distribution) and the orientation of the molecule were done as shown in Ref. [21]: We first calculated the HATI spectrum as a function of the alignment angle θ_L and then averaged over θ_L assuming a planar geometry. Clearly, in the experimental HATI spectra, high-energy electron emission

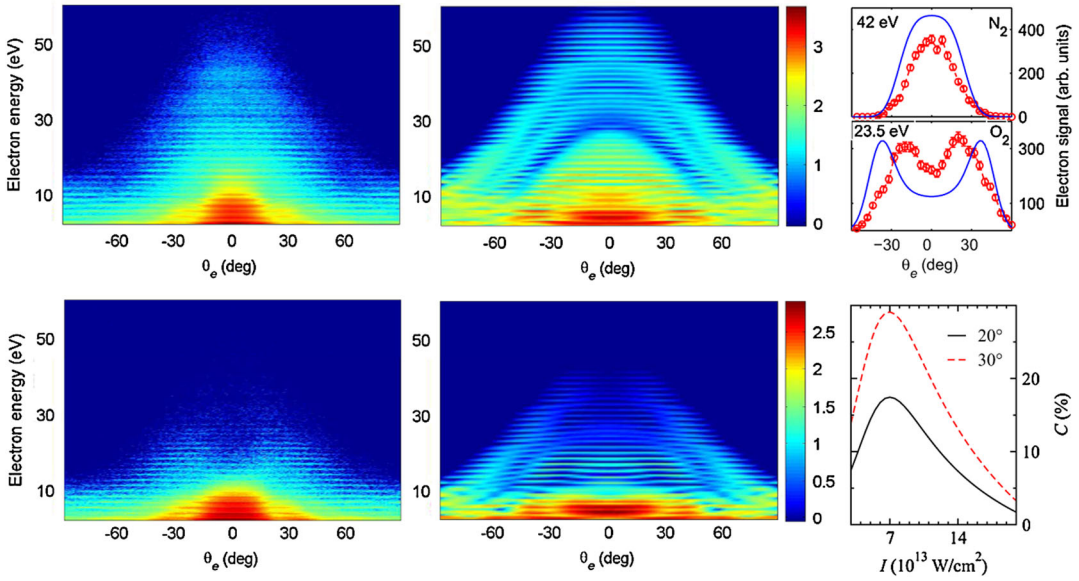

FIG. 1 (color online). False-color angle-resolved electron energy spectra for ionization of N_2 and O_2 by a 100 fs 800 nm laser pulse with an estimated peak intensity I. Left (right) panels: Experimental (theoretical) results. Upper panels: N_2 at $I = 9 \times 10^{13}$ W/cm^2. Lower panels: O_2 at $I = 7 \times 10^{13}$ W/cm^2. The lower right panel presents the contrast $C = [w(\theta_e) - w(0)]/[w(\theta_e) + w(0)]$ of the suppression for O_2 as a function of intensity for two different values of $\theta_e = 20°$ and 30°, evaluated by three-dimensional orientation averaging of Eq. (2) for π symmetry at the energy corresponding to the cutoff. This panel shows that we have chosen the optimal intensity (7×10^{13} W/cm^2) for the observation of the destructive two-source double-slit interference. The two panels on the upper right present cuts through the experimental (red curves with error bars) and the theoretical (solid blue curves) at 42 (for N_2) and 23.5 eV (for O_2). They quantitatively assess the quality of the MSFA and clearly demonstrate the suppression for O_2 and its absence for N_2.

from O_2 in the direction parallel to the laser polarization is strongly suppressed, in comparison with that from N_2. This suppression is well reproduced in the calculated spectra. The suppression is related to destructive double-slit interference. However, the exact mechanism also involves the presence of two sources as well as the symmetry of the molecular orbitals. This will be analyzed in the remainder of the Letter.

Figure 2 exhibits the four different paths from which the ionized electron can choose for its travel from its initial atom to the detector (assumed at infinity so that the final state is a plane wave with momentum $\mathbf{p}_f$), such that exactly one act of rescattering occurs. The two upper panels depict orbits such that the electron is born at and rescatters off the same atom. Their contribution is $T^{++} + T^{--} = A(e^{i\alpha} + s_{a\lambda}e^{-i\alpha})$, where $\alpha = \mathbf{p}_f \cdot \mathbf{R}_0/2$ and $s_{a\lambda}$ specifies the symmetry of the molecular orbital, $s_{a\lambda} = \pm 1$ for σ and π orbitals, respectively. Hence, whether the interference of these two contributions is constructive or destructive depends on the orbital in question. In the two lower panels, ionization and rescattering take place at different atoms. Assuming that the amplitude of both terms is again A, their contribution is $T^{+-} + T^{-+} = A(e^{-i\beta} + s_{a\lambda}e^{i\beta})$, with $\beta = (\mathbf{p}_f - 2\mathbf{k}_s) \cdot \mathbf{R}_0/2$, where $\mathbf{k}_s$ denotes the drift momentum in between ionization and rescattering. Again, whether these two orbits interfere constructively or destructively depends on the symmetry. However, the total contribution of all four rescattering orbits can be factorized in the form

$$2A\cos\left(\frac{\alpha+\beta}{2}\right)(e^{i(\alpha-\beta)/2} + s_{a\lambda}e^{-i(\alpha-\beta)/2}). \qquad (1)$$

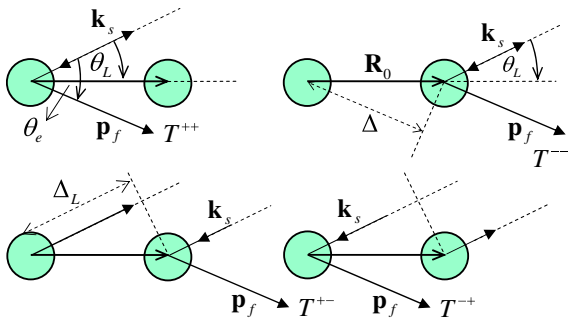

FIG. 2 (color online). Schematic diagram of the four rescattering T-matrix contributions to strong-field ionization of a diatomic molecule. The intermediate (between ionization and rescattering) electron drift momentum $\mathbf{k}_s$ is along the laser polarization axis. The final momentum $\mathbf{p}_f$ and the relative nuclear coordinate $\mathbf{R}_0$ include the angles θ_e and θ_L with respect to the former. In the upper panels the contributions T^{++} and T^{--} are depicted, where the electron rescatters at the same center at which it was born, and the corresponding electron path-length difference $\Delta = R_0\cos(\theta_e - \theta_L)$ is indicated. The two lower panels show the contributions of the processes where ionization and rescattering occur at different centers. The path-length difference of these two contributions, before rescattering, is $2\Delta_L = 2R_0\cos\theta_L$.

The surprising implication is that the first factor does not depend on the molecular symmetry. It reflects a novel interference, which involves all four of the rescattering orbits and yields complete destructive interference when $\cos\gamma = 0$, where $2\gamma = \alpha + \beta = (\mathbf{p}_f - \mathbf{k}_s) \cdot \mathbf{R}_0$, regardless of the symmetry $s_{a\lambda}$.

To be more specific, the total rescattering T-matrix element $T^{++} + T^{--} + T^{+-} + T^{-+}$ for the ath constituent of the molecular orbital having the symmetry σ (for N_2) or π (for O_2) is found to be proportional to [18,19]

$$\begin{aligned} &\cos\delta\cos\gamma \quad \text{for } a = s \text{ and } \sigma, \\ &\cos\theta_L\sin\delta\cos\gamma \quad \text{for } a = p \text{ and } \sigma, \\ &\sin\theta_L\sin\delta\cos\gamma \quad \text{for } a = 2p \text{ and } \pi. \end{aligned} \qquad (2)$$

Here $2\delta = \alpha - \beta = \mathbf{k}_s \cdot \mathbf{R}_0$, and we employed spherical coordinates for the internuclear coordinate vector $\mathbf{R}_0 = (R_0, \theta_L, \varphi_L)$ with the z axis along the laser polarization vector. Note that the contribution of each orbital is proportional to $\cos\gamma$. The condition that $\cos\gamma \approx 0$ defines an area of suppression in the $(E_{\mathbf{p}_f}, \theta_e)$ plane. The angle θ_e is enclosed by the outgoing electron and the laser polarization. Furthermore, it was shown [18,19] that this area survives the averaging over the laser intensity distribution. Now the question is whether or not it also survives the averaging over the molecular orientation with respect to the laser polarization.

Figure 3 exhibits angle-resolved spectra at fixed molecular orientation θ_L for N_2 and O_2. For intermediate angles $30° < \theta_L < 60°$, for both N_2 and O_2 there is a conspicuous suppression for emission in directions near the laser polarization axis. For larger and smaller angles θ_L, the suppression moves to larger angles θ_e. The two molecules significantly differ in the spectra for angles θ_L close to 0° and 90°. Namely, around these orientation angles the rescattering plateau is strongly suppressed for O_2, while for N_2 it is very intense, especially for $\theta_L = 90°$. This behavior can be traced to the symmetry of the molecular orbitals, as expressed in Eq. (2). Indeed, for O_2, due to the factor $\sin\theta_L\sin\delta$ the rescattering rate is negligible near $\theta_L = 0°$ and $\theta_L = 90°$. For N_2, on the other hand, the factors $\cos\delta$ for s orbitals and $\cos\theta_L\sin\delta$ for p orbitals can be dominant for $\theta_L = 0°$ or $\theta_L = 90°$. Therefore, for emission at angles around $\theta_e = 0°$, for N_2 the contributions to the orientation-averaged ionization rate from $\theta_L \approx 0°$ and especially from $\theta_L \approx 90°$ swamp those from the intermediate angles where the suppression is pronounced. This is in contrast to O_2, where the suppression survives owing to the absence of the rescattering plateau for $\theta_L = 0°$ and 90°. This explains the different appearance of the orientation-averaged angle-resolved spectra for N_2 and O_2. Whether or not the area near $\theta_e = 0°$ remains suppressed in the orientation-averaged angle-resolved spectrum is a subtle question whose answer depends on the relative magnitude of the contributions from the regions suppressed and not sup-

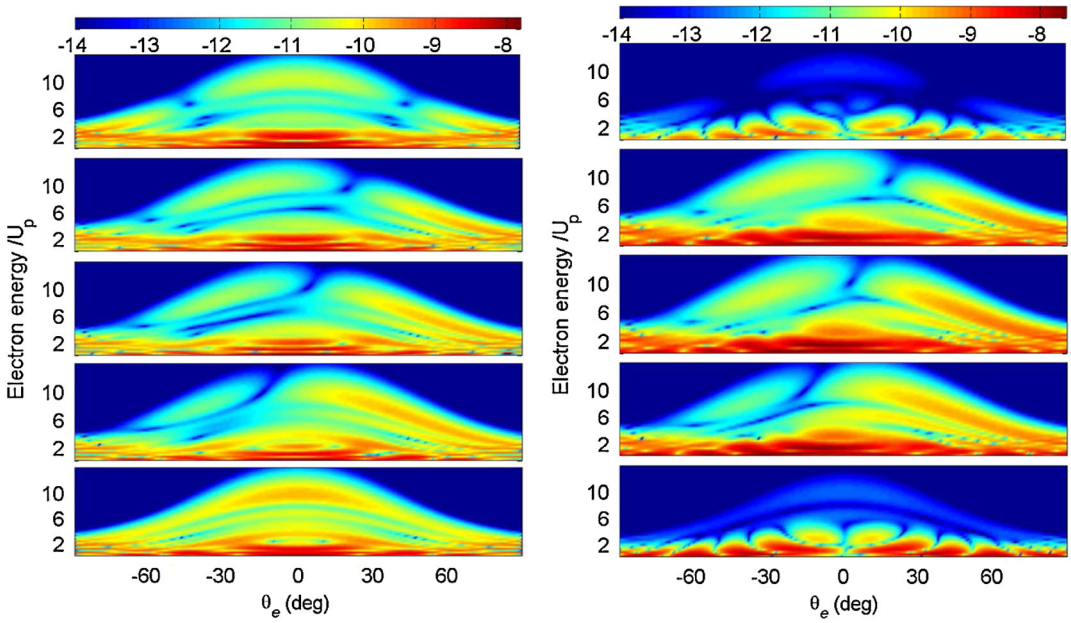

FIG. 3 (color online). Calculated angle-resolved ATI spectra for N_2 (left) and O_2 (right) for fixed orientation of the molecular axis for the intensities of Fig. 1. The orientation is at angles of $\theta_L = 0°$, 30°, 45°, 60°, and 90° from top to bottom. For O_2, we used $\theta_L = 1°$ and 89° in place of 0° and 90°. For the latter angles, the rescattering plateau is entirely absent.

pressed. The lower right panel in Fig. 1 displays the contrast of the suppression as a function of intensity. For the low intensity of the current Letter, the contrast is higher by approximately a factor of 2 than for the higher intensity of 1.6×10^{14} W/cm^2, which underlies Ref. [21]. Hence, in the experimental angle-resolved spectra of Ref. [21], the suppression for O_2 is all but invisible.

In conclusion, we have experimentally observed the manifestation of a novel destructive interference in laser-irradiated diatoms, which involves the superposition of the contributions of four geometric orbits. This is closely related to the double-slit interference but takes into account that there are two spatially separate sources of the electron wave packet. Our theoretical MSFA-based calculations show that for some range of fixed molecular orientations the resulting two-source double-slit destructive interference manifests itself almost identically in the angle-resolved electron energy spectra of N_2 and O_2, in marked contrast to the double-slit interference of direct ionization. The fact that the suppression remains visible in the orientation-averaged spectrum of O_2 but not of N_2 is due to the fact that for N_2 it is filled in by other contributions, which are absent for O_2 due to the π symmetry of its highest occupied molecular orbital.

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